

ABSTRACT

Simulation of Micro-Void Development within Large Scale Additively Manufactured Polymer Composite Deposition Beads

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Despite the growing level of technology advancement that characterizes extrusion-deposition additive manufacturing technology, there remains significant knowledge gap in fully understanding the process-structure-property relationship involved in this technology. The ability to effectively and efficiently control the manufacturing process provides an opportunity for optimizing the complex microstructure of the polymer composite prints which can lead to significant improvement of the associated print properties and in-service part performance. Modeling the polymer melt flow extrusion-deposition process is important in understanding the development of the inherent microstructural formation. The current research presents a computational-based approach for investigating process-induced micro-voids and their impact on print properties. We develop a multiscale FEA simulation tool to predict the global and local flow-fields during the polymer-melt flow process to investigate underlying mechanisms that may promote the micro-void development within the bead microstructure and determine potential process related factors that can be controlled to mitigate micro-void formation and growth. The research also

presents a computational based method for assessing the impact of the micro-voids on the effective print properties. The direct implication of a reduced part voidage levels is the development of high-quality functional components for specialized applications such as light weight & high strength integrity composites widely used in a variety of industries particularly the automobile, aerospace, renewable energy and defense industries.

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DEDICATION

To “God Almighty” and to my beloved parents, “Evelyn and Monday Awenlimobor”.

1 CHAPTER ONE

1.1 Research Motivation and Objective

Extrusion-deposition additive manufacturing (EDAM) technology offers numerous advantages compared to other subtractive technologies such as increased time and energy efficiency, high design flexibility and cost effectiveness. Despite the growing level of technology advancement that characterizes the technology, there remains significant knowledge gap in understanding the complex microstructural development of the parts printed from this technology especially the micro-void formation within the prints which directly affects the material behavior and in-service part performance. The ability to control the microstructure of the polymer composite prints during processing presents an advantage for significantly improving desired part performance especially by minimizing the inherent micro-voids formation. The objective of the proposed research is to utilize computational finite element analysis (FEA) modelling technique to simulate flow processes in extrusion-deposition based additive manufacturing (EDAM) of particulate polymer composites to better understand the evolution of the inherent bead microstructure including the porosities and fibrous structure. Micro-voids within printed beads are well known manufacturing defects that significantly impair the quality of fabricated components and could lead to component failure. The research investigates key mechanisms that are responsible for the development of micro-voids within the bead microstructure during polymer composite processing to understand the various factors that may influence the void formation process providing a means for leveraging our knowledge of suitable control parameters that would be tailored at mitigating the final part voidage levels. To the best of our knowledge, no known computational efforts currently exist in literature that are aimed at understanding the development of ‘process-induced’ voids within the microstructure of short fiber polymer composites including

EDAM beads. Previous studies have utilized experimental methods to investigate potential sources and factors that influence final part voidage which does not address the actual dynamic mechanisms involved in the evolution of the complex microstructure during the polymer melt flow process in the extruder-nozzle that results in the development of micro-voids.

Presently, the widespread perception on the main source of micro-voids within the prints is the mechanically entrapped air preexisting in the raw pellets prior to processing, however literature has revealed significant increase in the micro-void content from the raw pelletized feedstock to the final processed print beads without providing solid basis for the experimentally observed rise in void levels. By establishing a valid process-structure-property space map, the research is aimed at enhancing the in-service performance of additively manufactured (AM) short fiber reinforced polymer composite parts by optimizing the inherent microstructural formations particularly the intra-bead voids or micro-voids content within the composite bead. Our simulation is a first attempt in predicting the development of the local flow-field and the microstructural dynamics during EDAM polymer composite processing that helps provide new insight into the possible mechanisms that are potentially responsible for the observed rise in void levels. For improved model accuracy, our investigation further explores various effects that approximates conditions found in the actual polymer melt flow extrusion-deposition process such as the shear-thinning fluid rheology, the inter and intra-fiber forces, confinement effects etc. Additionally, we carry out parametric studies on the sensitivity of the primary flow variable that determines the void formation and characteristics to various process parameters that could potentially be fine-tuned to minimize the final part voidage levels.

1.2 Introduction

AM technology is a minimal wastage, cost saving technology with high manufacturing throughput, capable of producing large scale complicated geometries. For instance, the Thermwood LAAM system is reported to have a printing capacity of 30m x 3m x 1.5m and a material output rate of 226kg/hr [1]. Extrusion-Deposition based AM (EDAM) technology finds increasing application in various industries including automotive, aerospace, marine, space technology, renewable energy, housing, etc. particularly, in the fabrication of tooling and load-bearing components with complex intricate geometric structure and functionally graded materials (FGM) due to the inherently high design flexibility, speed, cost-effectiveness and large-scale capabilities. In the EDAM system, pelletized polymer composite feedstock material is melted as it is conveyed through the extruder screw before flowing through the contraction zone to the nozzle capillary and subsequently ejected into a moving substrate where solidification takes place under atmospheric conditions. The shear thinning fluid rheology of the polymer melt usually results in shear rate dependent viscosity and modified viscoelastic local stiffness within the flow. Usually shear rates in excess of 5000s^{-1} can exist in the narrow annular clearance within the extruder due to the high rotational velocity of the screw [2] while shear rates typically below 300s^{-1} can be found in regions of the nozzle resulting from flow acceleration. The shear rate variability across the extruder further complicates the final bead microstructure. The pelletized polymer feedstock is often reinforced with chopped fibers for numerous advantages such as increased part dimensional stability resulting from reduced coefficient of thermal expansion (CTE), improved stiffness, strength, flexure, and thermal conductivity of the part, and higher corrosion resistance. Despite these known advantages, studies have shown that the fiber inclusion in the polymer matrix introduces voids within the bead microstructure [3]. The resulting microstructural constituents thus

consist of reinforcing fibers, the polymer matrix, and microstructural voids. Voids within EDAM polymer parts typically exist in two different scales which include 1) The meso-scale voids or inter-layer voids and 2) the micro-scale or intra-layer voids. Meso-voids exist as narrow gaps between adjoining bead layers that align along the beads loading direction and prevalent in low fiber content composites with less deleterious effect to the mechanical properties of the part however the micro-voids that often segregate at the interface of individual fiber surface are predominant in high fiber content composites and pose as sites of stress concentration that effectively reduces the load bearing capacity of the composite part [3], [4]. For instance, interlaminar shear strength of composites have been reported to decrease by about 7% for each 1 % void up to a total void content of about 4% [5]. Similarly, the toughness of a polymer composite has been shown to reduce by as much as 15% for about 1.5- 3.5% shear rate regions in a LAAM extruder-nozzle head void contents [6]. The characterization of voids within the composite prints can provide useful information on the originating cause and type of the voids. The inter-bead voids are usually prismatic shaped and caused by weak inter layer adhesion, however the intra-bead voids have rather irregular shapes typically resembling a spheroid and involves a more complex microstructural formation process [7]. Most inter-layer voids can be controlled somewhat with lateral bead space and post-deposition compaction (i.e., with a tamper or roller. Intra-layer voids within the micro-structure of polymer composites are predominantly heterogenous in nature, existing between the interface of the fiber and matrix phase and are seldom homogenous in nature when nucleated in isolation within the matrix [3], [5]. Prior to processing, the raw pellets are found to contain inherent voids content due to air entrapment during the compounding stage usually below 6% which can be controlled by adequate venting measures [5], [8]. However, post extrusion, void contents up to 8, 20, and 35% void content for the 10, 20, and 30 wt.% glass fiber content in

Noryl extrudate have been reported [5]. The primary mechanism responsible for the micro-void development within the polymer melt during EDAM processing is that of bubble nucleation and growth which could result from a combination of any of the following subcategories:

1. Moisture/volatile absorption-desorption induced void nucleation mechanism which is based on the classical theory of nucleation [9], [10], [11], [12], [13].
2. Constrained volume contraction micro-void nucleation mechanism which results from uneven cooling across the extrudate due to thermal stratification from the core regions to the outer surface [5], [14], [15].

In the theoretical development of both nucleation mechanisms, a requirement for the nucleation of voids is the occurrence of sufficiently low localized pressure regions in the polymer melt below some reference value such as the vapor pressure of the gaseous phase of the volatile content in the case of the former mechanism or simply the atmospheric pressure in the latter mechanism. In either case, knowledge of the local fluid pressure distribution is important in determining the propensity for void nucleation in sites where they occur. Despite the challenges posed by voids ranging from reduced expectancy in the intrinsic material behavior to increased property anisotropy, the research efforts geared towards investigating the defects in short fiber reinforced polymer composite parts pales in comparison to the attention given to the studies of defects in long fiber consolidated polymer composite counterparts. Moreover, existing research on voids in the AM printing of chopped fiber polymer composites has placed more emphasis on the inter-bead voids or meso-voids that form between layers of deposited beads while studies on the more deleterious inter-bead voids that form within the complex fibrous microstructure has received very little focus.

Various factors have been experimentally found in literature to influence the micro-void distribution within the composite beads such as the rheological properties of the polymer suspension, the extrusion operating conditions, the local fluid visco-elastic stiffness defined by the local resin richness or lack thereof, fiber orientation distribution, and the fiber's aspect ratio [5]. During solidification of the extrudate, the cooling rate and the fiber-matrix CTE mismatch were also observed to promote higher levels of void crystallization [5], [8]. At the terminal portion of the extrusion process where the polymer melts leave the nozzle and where die expansion occurs, the micro-void content have been reported to increase significantly however in the filament feed and in the heating/extrusion zones of the extruder and nozzle insignificant micro-void content were observed [16]. Although there exists overwhelming experimental evidence supporting the existence and dependence of micro-voids within polymer composites beads on the various factors discussed above, there has been significant knowledge gap in understanding the actual micro-voids formation and evolution process within the bead microstructures during EDAM polymer processing and also in establishing concrete relationship that may exists between void development and the prevailing process conditions and other relevant parameters. On the other hand, there has been significant progress and continuous improvement in modelling the evolution of other microstructural descriptor counterparts, mainly the fibers orientation and distribution within the bead. One such analytical model used to predict the average fiber orientation states is the Advani-Tuckers 2nd order fiber orientation tensor equation of state [17], [18] developed from the well-known Jeffery's equation [19] which overtime has undergone various model improvement that more accurately simulate the momentum diffusion term and the appropriate 4th order orientation tensor closure approximation used in the model [20]. Alternatively, coupled, or uncoupled flow-fiber orientation numerical simulation models have also been developed by

various researchers to predict the flow field within the extruder-nozzle and the associated fiber orientation state. For instance, Finite Element Analysis (FEA) simulations was used independently by Heller et al. [21], Wang et al. [22] and Russell et al. [23] to simulate the flow of fiber filled polymer melt in a LAAM extruder nozzle to evaluate the orientation state of suspended short carbon fibers and the resulting thermo-mechanical properties. Likewise, coupled Smoothed Particle Hydrodynamic (SPH) and Discrete Element Method (DEM) numerical techniques were used by Yang et al. [24] to simulate the polymer deposition process of fiber filled polymer composite. While numerical models that accurately predict the fiber orientation and distribution during polymer composite processing are important in estimating and controlling the average thermo-mechanical properties of prints, models that also describe the micro-void formation within the microstructure of the prints and predict their characteristics are likewise important owing to the significant impact they have on the microstructural properties and behavior of the printed parts. Till date, modern day cutting edge computational tools that simulates and characterizes the development of the inherent bead microstructure during polymer composites processing are either inadequate or completely lacking especially in their ability to relate the evolution of the inherent bead microstructure with the relevant process variables and flow parameters among other factors that may influence their development. In addressing the fundamental problem of the formation and growth process of the inherent microstructural voids within the printed beads, we develop a set of hypotheses which are validated through series of computational investigations presented in subsequent chapters.

Firstly, we stipulate that besides the pre-existing voids present in the raw-pelletized feedstock that may subsist in the microstructure till the final stage of processing, two major mechanisms would potentially dictate the pore formation within the microstructure of the polymer

composite during processing at different regions of the EDAM extrusion-deposition process [5]. Within the extruder and nozzle, where the polymer melt temperature is relatively high, the moisture/volatile induced void nucleation mechanism would be the driving mechanism. It is assumed that the polymer material has some degree of moisture or dissolved additives/volatile content which may be pre-existing in the raw pellets or absorbed during processing. By reason of the multiphase constitution of the polymer melt flow, the mode of micro-void nucleation is predominantly of a heterogenous nature with the crystallization of a third phase at the fiber-matrix interface. Pores are predicted to nucleate in regions of the polymer melt with sufficiently low localized pressure below the moisture vapor pressure and at process temperature above the glass transition temperature and melt temperature. More details on their model development can be found in [9], [10], [11], [12], [13]. Post-extrusion, when the visco-elastic polymer melt is ejected from the nozzle exit into the atmosphere, where die swell/expansion occurs and at the onset of solidification, the restrained volumetric shrinkage mechanism developed by Titomanlio et. al [14], [15] is expected to be the dominant mechanism responsible for the formation of voids. Constrained contraction of the inner core region of the extrudate due to uneven solidification resulting from temperature stratification across transverse sections of the extrudate may create a solidification front. Once there is insufficient compensatory flow of polymer melt in the cavity in response to the pressure drop caused by densification, voids are likely to nucleate at regions where the cavity pressure P drops below zero. The voids are likely to segregate at the interface of the fiber and matrix due to weak interfacial adhesion and owing to mismatch in the CTE between the microstructural constituents [5], [8]. Irrespective of the dominant void formation mechanism, it is evident that the occurrence of very low local fluid pressure is a necessary requirement for void formation hence the fluid pressure is a primary variable. The low pressure most likely occurs at

sharp transitions in the flow path geometry such as at the edge of a screw flight, at nozzle exit or cavitation at the fiber ends during flow acceleration [2]. Most voids would nucleate at the fiber-matrix interface specifically close to the fiber's ends where the hydrostatic stress reaches a maximum on the fiber's surface. In essence, the voids distribution is expected to follow the local fiber concentration. Additionally, we can exclude bubble formation resulting from phase transformation since the polymer melt process temperature is well below boiling point. Likewise, we can exclude the void nucleation mechanisms based on cavitation since the Reynolds number of polymer melt flow is negligible in the absence of inertia forces resulting in cavitation number much greater than unity.

Secondly, we postulate that the void growth is governed by the difference in the internal pressure between the nucleated void and the surrounding pressure of the viscous polymer melt and by coalescence of smaller bubbles with larger ones driven by their pressure difference [2], [5]. The void growth rate is expected to depend on the concentration of dissolved moisture/volatile, the molecular diffusivity, the visco-elastic non-Newtonian polymer melt rheology and the gradient profiles of the flow temperature and pressure [5]. The void size inevitably depends on the magnitude of local pressure drop in the gas bubble, the instant and streamline location in the viscous flow where the void nucleated during processing. For instance, voids that are nucleated early in the flow on streamline location with relatively low velocities will have sufficient time to grow and allow for diffusion of smaller gas bubbles along its travel path. Moreover, since the experimentally observed pore sizes are relatively small, voids likely form late in the extrusion process near the nozzle [2]. Additionally, the average fiber aspect ratio, its geometry and elastic/plastic properties may likely influence the void formation and growth process [5], [8].

Lastly, following from our first hypothesis, we stipulate that micro-void formation primarily depends on the local fluid pressure especially near sharp transition regions within the flow. The fluid pressure depends on the level model accuracy that simulates the actual flow conditions in the extruder-nozzle such as the shear-thinning fluid behavior that may be important in high shear regions of the flow such as the lubrication flow region near the screw edge or flow acceleration region near the nozzle [2], fiber-fiber interaction forces, fiber-wall interaction forces, the intra fiber forces (bending, buckling, & breakage), etc., we can more accurately predict the likelihood of micro-void formation and growth. Likewise, by parametric studies involving the pressure response to possible flow parameters that may influence the void formation process identified in the second hypothesis, we can leverage established pressure relationships to determine possible control variables that may possibly mitigate the development of voids during the extrusion-deposition process.

1.3 Order of Dissertation

The second chapter presents summarily literature on the trend in extrusion-deposition based additive manufacturing (AM) of short fiber polymer composites; a detailed background on process induced microstructural voids formation, known sources and causes, their impact on properties and performance of printed parts and existing theoretical models for predicting their formation and growths. The literature also provides a review on the current trend in analytical and numerical based methods for evaluating the effective thermo-mechanical properties of composite materials. Lastly, the literature provides an extensive review on multiscale simulation of extrusion-deposition based AM process and detailed review on the physics involved in modelling transport phenomena associated with the process.

In the third chapter, we carry out detailed microstructural characterization of a 13% carbon fiber filled ABS LSAM polymer composite bead specimen using 3D X-ray micro computed tomography image acquisition and analysis to investigate the phenomenon of micro-void nucleation at the fiber/matrix interface, especially those that form at fiber tips. Since microstructural voids within short fiber polymer composites beads produced by additive manufacturing (AM) technology are known to alter the expected material behavior and significantly impair print performance, in-depth microstructural analysis and characterization can help improve our understanding on the formation mechanisms of these micro-voids during the polymer extrusion/deposition processing. The bead microstructure is characterized using various metrics including the micro-constituents phase fractions and volume fractions of interest features, distribution of average void size, average sphericity and average fiber orientation. To understand the impact of the final bead microstructural configuration on effective composite properties, the development of efficient and accurate material property predictive tool is very crucial which could provide a reliable means for assessing the effectiveness of control measures in fine-tuning the microstructure of SFRP composite to meet desired performance requirement.

Chapter four presents a finite element analysis (FEA) based numerical homogenization approach for evaluating the effective thermo-mechanical properties of LSAM particulate-filled composite using realistic periodic representative volume elements (RVEs) generated from reconstructed X-ray μ -CT image scans of the 3D printed bead. The chapter goes into details on the process of determining a suitable RVE size from a single region of interest (ROI) extracted from the bead's volume based on some dispersion tolerance metric and presents a method for validation of the numerical procedure by benchmarking results of predicted effective quantities with the well-known Mori-Tanaka-Benveniste's analytical estimate. Ultimately, chapter four aims to study the

impact of the inherent micro-porosities on the resulting composite material behavior and investigate the effect of anisotropy due to spatial variation in the microstructure across the bead specimen on the computed composite effective properties. We expect a priori what is otherwise known from literature that the inherent micro-voids would negatively affect the computed effective properties. It thus remains for us to present a detailed computational methodology which would be used to better understand the formation mechanism of these process-induced micro-void within the microstructure of polymer composite beads which makes up the bulk of the remainder of the dissertation.

Simulating the flow behavior of fiber suspension during SFRP EDAM composite processing is a typical Fluid-Structure Interaction (FSI) problem that can provide useful insight into potential mechanisms responsible for the resulting microstructure of the SFRP composites particularly the deleterious micro-voids known to impair print quality. A common starting point for modelling dilute fiber suspension has been to utilize the well-known Jeffery's analytical equations which has been in existence since 1922. Although Jeffery's model has gained popularity overtime in simulating the orientation dynamics of suspended particles in dilute viscous homogenous suspension, the model is seldom used in understanding the development of other microstructural formations such as the process induced micro-voids that form within polymer print beads. Chapter five extends Jeffery's model to simulate particle behavior in a special class of homogenous Newtonian flows with combined extension and shear rate components typically found in axisymmetric EDAM nozzle flow contractions. The chapter also presents a method for optimization of Jeffery's pressure equation using exact gradients and hessian to obtain the location within the flow-fluid and the particle orientation at which the fiber surface pressure extremes reach maximum. The chapter further addresses some limitations of the Jeffery's model, for instance the

model is confined to simulating rigid ellipsoidal shaped particles in viscous Newtonian homogenous flow and cannot model other phenomenon found in the actual extrusion-deposition polymer melt flow such as shear-thinning behavior of the polymer melt that may be important in high shear lubrication flow regions such as near the screw edge or flow acceleration region near the nozzle, cylindrical particle shapes with sharp geometrical transitions at the ends which better characterizes the geometry of chopped fiber in polymer melt suspension. Moreover, other effects such the inter-fiber forces in concentrated or confined flows and the intra-fiber forces, Brownian effects etc. cannot be modeled with Jeffery's equation. The chapter thus presents detailed methodology for the development of a custom-built numerical FEA model to simulate single particle motion in viscous suspension with GNF fluid rheology that can account for the various effects neglected by Jeffery's model and presents the results of various sensitivity analysis conducted considering other factors that may influences the fiber surface pressure distribution like the particle aspect ratio, the initial particle orientation and the shear-extensional rate ratio. The FEA model is validated by comparing results of the Newtonian case with results obtained from the well-known Jeffery's analytical model. Preliminary findings from this study conducted in this chapter provides an improved understanding of key transport phenomenon related to physical processes involving FSI such as that which occurs within the flow-field developed during EDAM processing of SFRP composites, and these results could provide insight into important microstructural formations within the print beads.

Highly loaded fiber polymer suspension flows usually involves long and short-range hydrodynamic interaction forces between suspended particles. Unfortunately, Jeffery's equation is limited to simulating particle motion in dilute regime and does not account for momentum diffusion due to inter particle interaction in concentrated polymer suspension flow. Overtime, more

advanced macroscopic fiber orientation models have been developed that account for rotary diffusion due to fiber-fiber and fiber-matrix interaction such as the well-known Advani-Tucker's even order moment-tensor fiber orientation evolution model. Unfortunately, these advanced models can only provide information about the overall transient fiber orientation state and cannot predict other field state information such as the local pressure distribution around suspended particles which may be useful in understanding the formation mechanism of other microstructural features like the inherent porosities. While it is unrealistic to simulate the motion of every individual particle in the fiber suspension and their interaction with each other, we present a simpler and novel approach for accounting for rotary diffusion in our single particle FEA model which we explain in detail in chapter seven. The method ultimately relates the Folgar-Tucker's phenomenological interaction coefficient with the effective fluid domain radius of influence utilized in the single fiber FEA model. A crucial step in the methodology involves relating the interaction coefficient with the steady state fiber orientation tensor using one of the available advection-diffusion fiber orientation tensor evolution models. Traditionally explicit numerical IVP-ODE transient solvers like the fourth order Runge-Kutta method have been used to determine the steady-state fiber orientation. Chapter six presents a computationally efficient and faster numerical method for determining the steady state fiber orientation for a range of diffusion interaction coefficients based on a Newton-Raphson iterative technique using exact derivatives of the second order fiber orientation tensor equation of state with respect to the independent components of the orientation tensor. The chapter considers various existing macroscopic-fiber orientation models and different closure approximations to ensure the robustness and reliability of the method and evaluates the performance and stability of the numerical scheme in determining physical solutions in various complex flow fields. Validation of the derived exact partial

derivatives of the fiber orientation tensor material derivative is carried out by benchmarking with results of finite difference techniques. Fiber orientation is an important descriptor of the intrinsic microstructure of polymer composite materials and the ability to predict the orientation state accurately and efficiently is crucial in evaluating the bulk thermo-mechanical behavior and consequently performance of printed part.

As we previously established, simulating polymer melt flow during EDAM processing is crucial in understanding the underlying mechanisms responsible for the microstructural formation and associated properties of the print. The final chapter of this dissertation presents a multi-scale computational FEA method that computes the global and local flow-field development of a typical EDAM polymer melt flow process particularly the fiber-induced local pressure fluctuations and orientation distribution across sections of the EDAM nozzle which may promote void formation within the bead's microstructure. On a macro-scale, the global flow field of a purely viscous, Newtonian planar polymer deposition flow through an EDAM nozzle is computed which are utilized on a micro-scale model that simulates the evolution of a single ellipsoidal fiber along streamlines obtained from the macro-model. The model development of the micro-scale single fiber evolution FEA model is presented in detail in chapter six. Chapter eight also presents a technique to account for rotary diffusivity due to short-range fiber-fiber interaction in the FEA simulation by determining an effective fluid domain size representative of the interaction coefficient of the melt flow through a correlation analysis that yields an equivalent steady state orientation based on the Advani-Tuckers equation. For robustness of the solution, different possible motions of the fiber along individual EDAM flow paths from a given set of random initial fiber conditions are considered to determine pressure bounds on the fiber surface along each streamline. The chapter concludes by assessing the effect of shear thinning on the computed local

flow-field responses. The simulation results of the pressure distribution around the surface of suspended fibers along streamlines of the EDAM flow-field are used to interpret the experimentally observed microstructural formations and characteristics particularly the voids that form within typical EDAM printed bead using data from the microstructural analysis carried out in chapter three.

2 CHAPTER TWO

2.1 Literature Review

2.1.1 Additive Manufacturing of SFRP Composites

Additive Manufacturing (AM) finds increasing applications in the fabrication of tooling and end-use load-bearing components with complicated geometry and functionally graded materials due to the inherently high design flexibility and cost-effectiveness that characterize the technology. Moreover, the technology is characterized by high material throughput due to the relatively high manufacturing speed associated with other technologies such as those requiring extensive cure cycles with complex cure chemistries [25]. AM has been classified based on the processing state of matter into solid extrusion based, powder based or liquid based systems [26]. These systems are further subcategorized based on the various fabrication techniques employed. For instance, fused deposition modelling (FDM) technique is used in solid extrusion-based systems; selective laser sintering (SLS), electron beam melting (EBM), selective laser melting (SLM) and direct metal laser sintering (DMLS) are techniques used in powder-based systems, while stereolithography (SLA) is mainly used in liquid-based system [27]. FDM terminology and fused filament fabrication (FFF) are often used interchangeably, however both techniques differ slightly in the processing environmental conditions. While FDM takes place in a thermally controlled enclosure with limited envelope, FFF is conducted under atmospheric conditions [28]. Extensive literature review on AM technology can be found in [1], [25], [27], [29]. Additionally, AM of polymer composites can be classified based on the aspect ratio of the reinforcing agents broadly into discontinuous (short) or continuous (long) fiber composites [25]. Polymer composites have been manufactured using most of the available AM technologies listed above including FDM, SLS, and SLA. Of particularly interest to the current research is the FDM or extrusion-deposition

AM (EDAM) technique. Extrusion-deposition AM (EDAM) uses a thermoplastic as feedstock materials such as acrylonitrile butadiene styrene (ABS), polycarbonate (PC), polylactide (PLA), polyamide (PA), thermoplastic polyurethane (TPU), polyetherimide (PEI), polyethylene terephthalate (PET) and polyetheretherketone (PEEK). The feedstock material is heated into a viscoelastic polymeric melt state within a heating extruder chamber and the melted material is ejected through a nozzle/die under pressure where the strands are deposited as beads onto a build surface to form the desired 3D geometry [30], [31], [32], [33]. Cooling and solidification of the polymer melt material follows immediately upon exposure of the processed material to the atmosphere at the nozzle exit. EDAM technology can be classified based on extrusion mechanism into three (3) broad categories namely the filament based, plunger or syringe based and the screw-based mechanism [28]. Overtime, EDAM technology has experienced significant scale-up in manufacturing capacity from desktop or small-scale AM (SSAM) to commercial medium- scale AM (MSAM) and large-scale AM (LSAM) systems which have higher material throughput and printing speed necessitated by industrial need in vast economic sectors including automotive, aerospace, renewable energy, defence etc and made possible by the utilization of pelletized feedstock for polymer processing. MSAM systems have build volumes ranging from 1 to 7m³, extrusion head diameters ranging from 0.1 to 4.0mm and deposition rates between 0.5 to 4.0 kg/h while LSAM systems have build volumes greater than 7m³, extrusion head diameters ranging from 4.0 to 7.6mm and deposition rates between 4.0 to 50kg/h [34]. For instance, the Oak Ridge National Laboratory (ORNL) Big Area Additive Manufacturing (BAAM) system with a build capacity of 6×2.5×1.8m and maximum material output of 45kg/h. Likewise, the Thermwood Corporation LSAM system has a larger build capacity reaching 30×3×1.5m and material output rates of up 227kg/h [35]. LSAM systems have less energy requirements than expected due to

various factors considered in its design such as the elimination of the heating chamber, although the accompanying cost and energy savings are a trade off with the lowering of the print parts quality and introduction of defects such as warpage, delamination and cross-sectional tapering [34]. To combat these associated prints problems without necessarily modifying the manufacturing method or increasing production cost, the polymer feedstock materials in LSAM systems are usually reinforced with short glass fibers (GF) or carbon fibres (CF) to yield enhanced thermal-mechanical properties as compared with the neat polymers [33], [36], [37]. For example, Love et al. [37] showed that the addition of CF to ABS thermoplastic, significantly reduced the warping in manufactured parts by lowering the coefficient of thermal expansion. Tekinalp et al. [3] also showed an improvement in stiffness and strength along the print direction within the carbon fiber ABS (CF/ABS) composite compared to neat ABS due to the alignment of fibers along the print direction. Somireddy et al. [38] showed a significant improvement in flexural properties of the CF/ABS compared with the neat ABS. Tensile properties and Young Modulus of CF/ABS SFRP composites were observed to increase in magnitude with increasing carbon fiber content up to a saturation limit of about 7.5%wt CF content and with accompanying decrease in toughness, yield strength and ductility [39]. Typical length scale of short fibres used in AM may range from that of milled fibres with length up to hundreds of microns to those of chopped fibres usually few millimetres long [25]. Maximum saturation limit of the fiber content and packing efficiency in a polymer composite material is limited by the average length and degree of alignment of the fiber reinforcement. High fiber loads up to 30% in polymer composites are achievable with highly aligned fibres or milled fibres with very small aspect ratio. Consequently, the fiber length is an important factor that affects the attendant SFRP composite properties. Unfortunately, the average fiber length used in SFRP composites (<150um) is well below the critical fiber length of 640um

which has been deemed necessary for effective transfer of the fiber micro-constituent strength to the overall composite strength [25]. Moreover, long length discontinuous fibers are prone to excessive bending or breakage during polymer composite processing due to fiber attrition and may lead to clogging of the nozzle orifice. Although most fiber breakage occurs in regions of the screw extruder of a modular composite mixer that are partially filled with polymers compared to starved regions, the fiber length in both cases have been predicted to decrease exponentially with polymer processing time based on a kinetic model [40]. Although, there are obvious benefits resulting from the reinforcement of polymers with fibres, optimal material behaviour of manufactured composites is limited by the inherent complexities of the uncontrolled microstructure particularly the unwanted micro-porosities and unpredictable distribution of the fiber orientation [1], [31], [41], [42]. The section following provides summarily literature on the voids in polymer composites manufactured from LSAM technology.

2.1.2 Voids in Large Area Additively Manufactured Polymer Composites

Voids in short carbon fiber-reinforced composite AM parts can be categorized into two distinct types: inter-layer voids and intra-layer (microstructural) voids. Inter-layer voids form gaps between the deposited beads that occur due to the rounded shape of the bead corners and insufficient bonding between beads of material during the 3D printing process [3], [43]. Conversely, intra-bead or microstructural voids (referred to here as micro-voids) develop within individual beads where the micro-void size is typically much less than that of inter-layer voids. Yu et al. [44] found that the addition of carbon fibers into the polymer matrix increases the composite's viscosity, leading to microstructural voids forming within the AM part. Sayah et al. [31], [32] also identified the presence of micro-voids within the pellet's microstructure prior to the extrusion-deposition process as well as within the beads following deposition.

Although short fiber reinforcement of polymers offers various benefits such as enhanced thermal, mechanical, and anti-corrosion properties of the component parts with improved dimensional stability [3], however, the development of intralayer micro-voids within the bead microstructure due to fiber reinforcement are known to alter the material behaviour that may impair the part performance and possibly result in compromised structural integrity. Intralayer micro-voids also affect the capacitance and electrical permittivity of the short fiber polymer composite. Higher levels of voids within polymer composites can also increase moisture absorption during polymer composite processing and has been shown to degrade mechanical properties [45]. Interlayer voids that form at the interstices of AM extrudate strands tend to orient along the axis of the bead and are less detrimental to the mechanical behaviour of the polymer composite compared to intralayer micro-voids, especially those formed at the fiber-matrix interface which acts as sites of stress concentration that reduces the load bearing capacity of the polymer composite material [4], [25]. Voids are unwanted defects that arise within the structural fabric of the composite material due to unsuitable process conditions which should not be confused with micro-damage which is internal microstructural failures that occurs during composite loading [46]. EDAM process-induced voids may originate during the flow of polymer melt through the extruder nozzle or during solidification when beads are deposited on the moving substrate. It has been experimentally observed that the micro-void content is highest when the polymer melt exits the nozzle during die-swell/expansion and subsequently decreases when the beads are deposited on the bed [16].

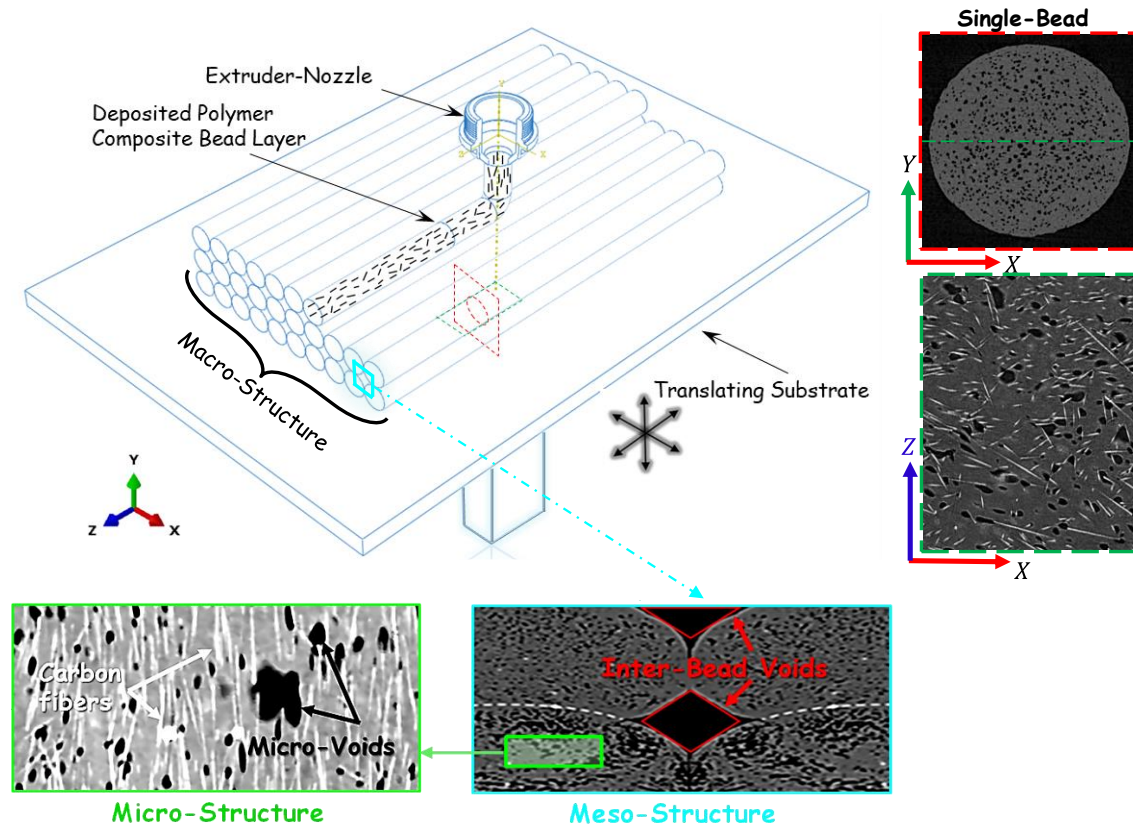


Figure 2.1: Schematic of the Multi-Scale Structure of EDAM SFRP Composite Processing

Various sources that may induce micro-void formation within EDAM beads during polymer composite extrusion/deposition processing have been identified and include bubble encapsulation within the pellets during the compounding process which can be reduced by adequate venting measures, absorption of moisture and gases including dissolution of chemical volatiles within the polymer melt, and development of internal stresses in excess of the intrinsic visco-elastic strength of extrudate during cooling due to uneven contraction during solidification [5], [8], [47]. Knowledge of the morphology of voids present within an EDAM printed sample can provide insight into the cause and type of voids present in the composite structure [7]. For instance, interlayer voids formed between adjoining strands having a flat-bar prismatic shape suggest weak interlayer adhesion as the responsible agent, while intralayer micro-voids formed at the interface between the fiber and matrix constituents suggests a possible weak adhesive strength of the sizing

agent. In the latter, ellipsoidal shaped voids and gas pockets have been shown to result from excessive temperature in the melt [7]. Narkis et al. [8] found that the contribution of the interfacial micro-void content due to fiber-matrix debonding compared to the overall void content within the extrudate was minimal. Most interlayer voids can be controlled somewhat with lateral bead space and post-deposition compaction (i.e., with a tamper or roller). Of the two types of voids, interlayer voids aligned in the loading direction are less detrimental to the mechanical properties of the AM printed composite than intralayer micro-voids within the bead microstructure. Additional factors that may influence the nucleation of micro-voids within polymers have been identified as well. For example, Vaxman et al. [5] and Sayah et al. [31] independently found that the print processing conditions like temperature, pressure, and flow rate affected the final void content of an extruded short fiber composite. During the process of extrudate solidification, factors like non-uniform and faster cooling rate, mismatch between the fiber and matrix thermal expansion coefficient and the die-swell at the nozzle exit of the free extrudate due to pressure difference upon atmospheric exposure were observed to promote voids [5], [8], [16].

Of particular interest in our work are contributing factors for void formation that relate to the reinforcing fiber constituent. Micro-void volume fraction has been experimentally observed to increase with increasing fiber concentration and fiber aspect ratio due to an increase in the effective viscosity of the polymer composite melt [5], [16], [39], [48]. At lower fiber volume fractions, interlayer voids between beads were experimentally observed to be more prevalent in the printed composite, however, when the fiber content is high, intralayer micro-voids were found to be more dominant where interlayer void content decreased due to reduced extrudate diameter resulting from lower die expansion and higher thermal conductivity [3], [4]. Additionally, Yang et al. [16] found that during polymer processing, the volume fraction of micro-voids is negligible within the

extruder/nozzle, however the void content peaks when the polymer melt just exits the nozzle during die-swell and drops to a stable value upon bead deposition. The findings also revealed that an increase in the void content in the polymer composite at the nozzle exit where die swell occurs is a direct consequence of a decrease in the fiber volume fraction due to an overall decrease in the effective viscosity of the suspension. Sayah et al. [31] also showed that the degree of fiber misalignment in various regions of the extrudate correlate directly with the measured void volume fraction in these regions. Under favourable operating conditions (temperature, pressure and extrusion rate), micro-bubbles may form within the pure polymer matrix phase or at the interface of the fiber and matrix phase. Micro-voids that form at the fiber-matrix interface may be due in part to failure of the adhesive/sizing agent that results in fiber-matrix debonding. Of significance to our study is the observed higher likelihood of micro-voids occurring at the ends or tips of suspended fibers within the polymer composite beads with high fibre volume fractions [5], [8]. Although high fibre packing is found to reduce the potential of micro-voids nucleation at the interstices between fibres, the increased number of fibre ends are observed to provide favourable sites for void nucleation to occur. The air pockets or micro-voids that solidify at fiber terminations are typically characterized by melt pressure variations and micro-void growth/collapse or re-dissolution in the polymer matrix which is influenced by the difference between the internal micro-voids pressure and external pressure within the surrounding melt [8].

A relevant analogy can be drawn from micro-damage initiation as it relates to flow-induced micro-void formation where micro-damage nucleation sites tend to occur at stress concentrations occurring close to the ends of fibers [49], [50]. Known micro-damage nucleation mechanisms have been related to excessive interfacial shear stress at the fiber-matrix interface [49] and the maximum hydrostatic stress within the matrix [50], both of which occur at the fiber tips. In these cases, micro-

crack initiation is shown to occur at the point where the maximum stress exceeds a critical value related to the intrinsic strength of the composite material such as the interfacial fiber-matrix bond strength or matrix fracture strength. Hu et al. [49] showed that fiber aspect ratio and orientation were significant microstructural parameters that influence the maximum stress at the fiber tips and consequently micro-crack initiation. Separate investigations conducted by Agyei et al. [51] and Hu et al. [49] showed that the local stiffness of the ductile fracture region within the matrix where micro-crack initiation and progression mainly occur depend on the stress concentration at fiber tips, the degree of fiber misalignment and the average tip distance between fibers within this region. In addition, computational studies performed by Awenlimobor et al. [52] indicated that the hydrostatic pressure within the fluid surrounding the fiber surface reaches an extreme value at the fiber tips where micro-voids typically occur, and the tip pressure depends on the fiber aspect ratio and orientation angle.

2.1.3 Micro-Void Nucleation Mechanisms

Various mechanisms potentially responsible for void formation within the microstructure of short fiber polymer composite print beads have been investigated in numerous literatures. Of the known mechanisms, the encapsulation of low molecular weight substances within the beads during compounding of the pelletized material and subsequent extrusion-deposition of the polymer melt in the EDAM nozzle extruder has been identified as one major cause of void formation [5]. Other process induced mechanisms include the uneven volumetric shrinkage mechanisms due to temperature stratification during solidification [5], [14], [15], volatile/moisture absorption and desorption induced void nucleation mechanism [9], [10], [11], [12], [13], and stress-instigated cracking mechanism [47]. All the listed mechanisms require a critical criterion to be satisfied for the onset of void initiation. For instance, volatile induced void formation requires the surrounding

fluid pressure to drop below the polymer melts vapor and operating pressure. Likewise, volumetric shrinkage that leads to void nucleation results from insufficient compensatory flow of polymer melt once the cavity pressure drops below zero [14], [15]. These void formation mechanisms suggest that void nucleation within the microstructure of short fiber reinforced polymer composites during EDAM processing is to some extent dependent on the melt pressure field. These mechanisms do not act independently but often involves an interplay to achieve stable void development. Each could, however, be classified as a homogenous or heterogenous mechanism. In the former, micro-voids form within a single phase under critical conditions while in the latter class, micro-void formation occurs at the interface between two existing phases such as the interface between fiber and matrix [13]. The characteristics of a void may suggest the dominant mechanism responsible for their formation. For instance, most micro-voids have been observed to be positioned at the ends of fibers, which suggests a heterogenous void formation process. Alternatively, the presence of isolated micro-voids within the matrix suggests the responsible mechanism is one of a homogenous nature. In subsection below, we present a summary of critical criterions of some known mechanisms necessary for void formation.

2.1.3.1 Moisture/Volatile Induced Mechanism

The motivation for evaluating pressure on the fiber surface stems from classical nucleation theory that addresses void initiation and growth within a polymer melt investigated by Han and Han [10], Stewart [11], and Han [12], who also investigated the dynamics of void initiation in polymer melts under shear flow. Colton and Suh [13] distinguished between two mechanisms of nucleation which includes 1) homogenous classification involving the formation of a new stable phase in a primary phase with dissolved secondary components under critical conditions due to thermal fluctuations and molecular interaction and 2) a heterogenous classification involving the

crystallization of a third phase at the interface of two other phases, usually a liquid and a solid. Both forms of nucleation can coexist and occur concurrently under a mixed classification. However, in a system such as a colloidal solution, depending on the volume fraction of the suspension, a heterogenous nucleation is more likely to be dominant due to smaller activation energy barrier. The polymer melt material considered in the simulations above is composed of 13% filled carbon fiber ABS (Acrylonitrile Butadiene Styrene) such that a heterogenous dominant mode of nucleation is expected to occur at the interface of the carbon fiber and polymer. Also, it is expected that the polymer material has some degree of absorbed moisture or dissolved additives/volatile. In the model development by Roychowdhury et al. [9], a necessary requirement for potential homogenous void nucleation is the occurrence of very low localized fluid pressure P_L below the moisture vapor pressure P_V i.e., $P_L < P_V$ at process temperature $\mathcal{T}_p$. The nucleation rate J_n (i.e., $J_n \geq 1$ for void nucleation) as modified by Colton and Suh [13] in heterogenous systems is

$$J_n = N_v \sqrt{\frac{2\gamma_t}{\pi\tilde{m}}} \exp \left[-\frac{16\pi\gamma_t^3}{3k_B\mathcal{T}(P_V - P_L)^2} S(\varphi) \right] \quad (2.1)$$

where N_v is the number of molecules per unit volume of the volatile phase, $\tilde{m}$ is the molecular mass of the volatile phase, γ_t is the surface tension at characteristics temperature $\mathcal{T}$, and k_B is the Boltzmann constant. In the above,

$$S(\varphi) = (1/4)(2 + \cos \varphi)(1 - \cos \varphi)^2 \quad (2.2)$$

where φ is the wetting angle of the interface. Usually, the characteristics temperature of nucleation $\mathcal{T}_n$ stays well above the glass transition/melt temperature $\mathcal{T}_g/\mathcal{T}_m$ (i.e., $\mathcal{T}_n \sim \mathcal{T}_p \geq \mathcal{T}_g/\mathcal{T}_m$) and the phenomenon takes place almost instantaneously. Colton and Suh [13] determined the moisture vapor pressure from the moisture concentration distribution in the polymer using Henry's Law,

$P_V = c/H_V$ where c is the concentration and H_V is Henry's constant for moisture in a polymer.

Based on classical nucleation theory, the characteristics nucleation time t_n is given by

$$t_n = r_c^2/D \quad (2.3)$$

where D is the moisture diffusivity defined by $D = D_o e^{-A_E/\mathcal{T}}$; and D_o is the moisture diffusion constant within the polymer, A_E is an activation energy related material constant, and $\mathcal{T}$ is the temperature. r_c is the critical radius on nucleation given by

$$r_c = 2\gamma_t/(P_V - P_L) \quad (2.4)$$

The single fiber pressure solutions appearing above show that the calculated localized fluid pressure P_L (i.e., the minimum pressure extremes calculated in the micro-model) may fall below processing pressure P_ψ which increases propensity for void nucleation at these sites. An additional requirement for void nucleation is that the void nucleation time t_n must be less than the streamline deposition time t_d . i.e., $t_n < t_d$. Han and Han [10] showed that the classical nucleation theory under predicts the propensity for void nucleation in polymer solutions with significant proportion of dissolved volatile components. They observed nucleation at critical pressures P_L above the vapor pressure P_V and developed a more applicable model incorporating the Flory Huggins theorem to account for reduced entropies due to restrictions posed by macromolecules in the solvent yielding a nucleation rate of

$$J_n = [N_v][B_F]e^{(-\Delta F_p^*/nk_B\mathcal{T})} \quad (2.5)$$

where B_F is the frequency factor given by

$$B_F = B_1[D(\mathcal{T})/4\pi r_c^2] \exp(-B_2/\mathcal{T}) \quad (2.6)$$

and $D(\mathcal{T})$ is the molecular diffusivity of the volatile phase which Han and Han [10] obtained using free volume theory of Vrentas and Duda given by

$$D(\mathcal{T}) = D_o(1 - 2\chi_F\vartheta_1)(1 - \vartheta_1)^2 \exp(-E/R_G\mathcal{T}) \exp(\zeta(w_1\hat{V}_1^* + w_2\hat{V}_2^*q)/\hat{V}_{HF}^*) \quad (2.7)$$

The free energy for critical void nucleation in polymer solutions ΔF_p^* given by

$$\Delta F_p^* = (16/3)\pi\gamma_t^3(P_V - P_L)^2 - nk_B\mathcal{T} \left\{ \ln\left(\vartheta_1 \frac{P_G}{P_V}\right) + \vartheta_2 \left(1 - \frac{V_1}{V_2}\right) + \chi_F\vartheta_2^2 \right\} \quad (2.8)$$

In eqns. (2.6) through, (2.8) B_1 & B_2 are empirical constants dependent on the polymer solution, w_i , ϑ_i and V_i are the weight fraction, volume fraction and molar volume of constituent i respectively, subscript $i = 1$ for solvent and $i = 2$ for solute. In our material systems, the proportion of molar volume of the volatile phase in the polymer is much less than unity, i.e., $V_1/V_2 \ll 1$, ζ is the free volume overlap factor, q is the critical molar volume ratio of jumping units of solvent to jumping units of polymer solution, and $\hat{V}_{HF}^*$ is the average hole free volume per gram of mixture. χ_F is the Flory Huggins interaction parameter and P_G/P_V defines the degree of saturation of the gas phase, P_G being the pressure inside the critical bubble given as

$$P_G = (3/2)\rho_L\dot{r}_c^2 + 2\eta/r_c + 4\mu_0(\dot{r}_c/r_c) + P_L \quad (2.9)$$

where $\dot{r}_c$ is the growth rate at the onset of nucleation, ρ_L is the liquid density and μ_0 is the viscosity at zero shear. The surface tension γ_t at the elevated temperature at which the polymers are processed is estimated using expression by Sugden [53] thus

$$\gamma_t(\mathcal{T}) = (P_a/\bar{V}(\mathcal{T}))^4 \quad (2.10)$$

where P_a is the Parachor and $\bar{V}(\mathcal{T})$ is the molar volume of the liquid. The consequence of this is that the surface tension at an elevated temperature can be estimated with knowledge of the surface tension at a reference temperature through

$$\gamma_t(\mathcal{T}_2) = \gamma_t(\mathcal{T}_1) \left[\frac{\rho(\mathcal{T}_2)}{\rho(\mathcal{T}_1)} \right]^4 \quad (2.11)$$

and the contact angle can be obtained from equation by Girifalco and Good [53]

$$\cos \varphi = 2\hat{\phi}\sqrt{\left(\gamma_{t,s}/\gamma_{t,l}\right)} - 1, \quad \hat{\phi} = 4\left[V_s^{-1/3} + V_l^{-1/3}\right]^{-1}, \quad V = \frac{N_v}{\rho} \quad (2.12)$$

The details here provide a possible basis for estimating the potential for void nucleation within a polymer melt given a known amount of volatile content in future research.

2.1.3.2 Restrained Volumetric Shrinkage Mechanism

The theory behind the constrained volume contraction void initiation mechanism has been developed by various researchers, especially by Titomanlio et. al. [14], [15]. The basic principle of void formation based on this mechanism is restricted contraction of the inner core region of the extrudate due to temperature stratification across transverse section of the cavity during solidification process which results in the creation of a tapered solidification front. Insufficient compensatory flow of polymer-melt from the extruder-nozzle in response to pressure drop in the enclosure due to densification would then lead to void formation. The melt viscosity and front geometry dictates the pressure drop within the cavity which is exacerbated with tip advancement leading to void growth. Assuming a rigid, non-deformable solidification front an approximate expression that describes its geometry given as [14], [15]

$$\frac{s(z)}{R} = \left(1 - \frac{z}{L}\right)^a \quad (2.13)$$

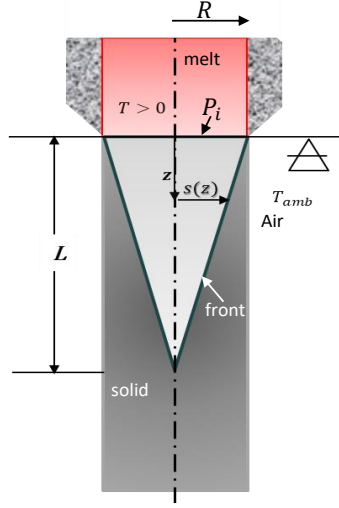


Figure 2.2: Schematic of the Polymer Extrusion at the Nozzle Exit showing Solidification Front where $s(z)$ is the radial position of the front at any arbitrary distance z from the nozzle of exit radius R , a is a shape power index, and L is the enclosure length which is proportional to the vertical velocity $\dot{z}$ of the solid extrudate given as

$$L = l \frac{\dot{z} R^2}{\alpha} \quad (2.14)$$

l is the dimensionless enclosure length, α is the thermal diffusion coefficient. Void nucleation would take place when the pressure $p(\zeta)$ at some arbitrary dimensionless axial distance $\zeta = (1 - z/L)$ within cavity drops below zero. i.e., $p(\zeta) \leq 0$. Assuming the polymer melt rheology has a power law fluid definition and considering isothermal condition, the pressure $p(\zeta)$ within the cavity is given as

$$p(\zeta) = \left[p - C \frac{m \dot{z}^{n+1} R^{1-n}}{\alpha} \right] (\zeta^{1-b} - 1) \quad (2.15)$$

where constants C and b are respectively given as

$$C = \left(\frac{3n+1}{n} \right)^n \frac{2\beta^n l}{b-1}, \quad b = a(1+n) \quad (2.16)$$

m is the power law viscosity coefficient and n is the power law viscosity exponent and β is the fractional volume contraction on solidification given as $\beta = (\rho_s - \rho_l)/\rho_l$. ρ_l & ρ_s are the densities of the polymer melt and solid extrudate phase respectively. The foregoing mechanisms presented above show that void nucleation within the microstructure of short fiber reinforced polymer composite during processing is to some extent dependent on the pressure field which is the hypothesis of the current study.

2.1.3.3 Stress Induced Mechanisms

Various studies that investigate stress induced micro-void nucleation in polymers currently exists. These stresses may arise from internal sources such as development of residual stresses within polymer undergoing restrained expansion or contraction or may arise from external sources when the polymeric articles are subject to an active load or displacement. Eom et al. [47] studied the voids that form in a thermosetting polymer due to chemical shrinkage during cure that results in the buildup of internal tensile stresses which exceed the material intrinsic strength. He proposed a critical value for the internal stress that determines the onset of void nucleation prior to gelation. In typical carbon fiber reinforced polymer deposition process, solidification occurs immediately following bead exposure to the atmosphere because of temperature differences. Depending on the thermal expansion coefficient which determines the cooling rate and viscoelastic transformation process, volumetric shrinkage may occur giving rise to residual stresses within the substrate [4], [45], [54]. In thermosetting polymers, residual stress may also arise excessive cure temperatures or poor heat transfer during curing typical in thick composites with associated thermal degradation [45]. Analogously, void would result within the bead microstructure when these residual stresses exceed the limit strength of the material. Micro-void nucleation due to complex microstructural behaviour of chopped fibre reinforced polymer composite materials under an external load have

also been investigated by various researchers. Hu et al. [49], conducted a study on micro-voids that form at the ends of fiber due to shear stress concentration based on a shear-lag model that depended on the fiber's length and orientation. They found that shear stress reached a maximum at the fibers end which may exceed the interfacial bonding strength between the fiber and matrix and likely result in micro-void nucleation at the interface. According to the shear lag model, the shear stress along the fiber length is given by:

$$\tau_m = \tau_Q \sinh \eta z - \sigma \sin \phi \cos \phi \sin \theta, \quad \tau_Q = \tau_Q(G_m, E_m, E_f, l_f, r_f, v_f) \quad (2.17)$$

where, τ_Q depends on the material properties and fiber geometry, z is the transverse distance from the fiber center, σ is the applied stress, ϕ is the fiber orientation, θ is the polar angle measured from the plane normal to the fiber direction, G_m is the matrix shear modulus, E_m & E_f are the Young modulus of the matrix and carbon fiber respectively, r_f is the fibers radius, l_f is the fiber's half length, and v_f is the fibers volume fraction [26].

Additionally, they found that micro-voids potentially formed at regions with high agglomeration of fiber ends due to high stress concentration. Hanhan et al. [50] showed that the hydrostatic stress distribution in the matrix can be used to predict the probability and possible locations of void initiation within the composite microstructure. Experimentally, they showed that the location of fiber ends played an important role in determining where micro-voids formed. By superposing the spatial locations of hydrostatic tensile stress hotspots obtained from FEA simulations with the actual microstructural locations where the voids were observed to nucleate based on in-situ experimental data, they showed both locations correlated with each other. The hydrostatic stress was calculated based on eqn. (2.18) below.

$$\sigma_{hyd} = \frac{\sigma_{xx} + \sigma_{yy} + \sigma_{zz}}{3} \quad (2.18)$$

The various stress-induced micro-damage mechanisms presented here suggests that knowledge of the stress distribution of the composite polymer EDAM flow field can potentially provide information of where voids are likely to nucleate. Based on Jeffery's assumption [19], the active stress on the surface of fibre in viscous suspension is simply hydrostatic fluid pressure $\sigma_{hyd} = -p$. Accordingly, focus is placed on the pressure peak response on the surface of fiber particularly at the ends during its motion in homogenous viscous flow which may serve as pointers to potential mechanism responsible for micro-void segregation at the fiber terminations.

2.1.4 Computational assessment of the effective thermo-mechanical properties of SFRP

Composites

The performance of randomly dispersed short fiber reinforced polymer (SFRP) composites readily depends on its inherent microstructural characteristics such as the concentration, orientation and length distribution of the fiber reinforcement, the content, distribution and morphology of the inherent micro-voids, the fiber-matrix inter-layer adhesion, etc. [55]. Spatial variations in the heterogenous microstructure results in intrinsic anisotropic macroscopic behavior of the composite material. Property prediction of randomly distributed misaligned SFRP composite becomes more complicated with increased non-uniformity and anisotropy across the heterogenous composite microstructure.

Several computational micro-mechanics techniques have been developed by various researchers for estimating the effective material properties of SFRP as an alternative to experimental characterization which includes the analytical mean-field homogenization techniques, numerical modelling methods and the statistical continuum mechanics methods [56]. Analytical methods usually involves a two (2) step homogenization approach for property prediction of randomly misaligned SFRP composite which includes (a) determination of effective

properties for the pseudo-grains of the decomposed RVE structure based on available analytical mean-field models for unidirectional aligned SFRP composite of uniform fiber length with isotropic microconstituent properties and (b) averaging of the predicted properties over the fiber orientation and length distribution amongst the pseudo-grains of the misaligned SFRP composite to account for spatial variations in the microstructural configuration using either the Voight or Reuss models [57], [58]. Numerous classical analytical micromechanics models for predicting approximate fourth-order elastic tensor of unidirectional SFRP composite with uniformly distributed fibers currently exists with varying degree of accuracy which are well documented in literature [55], [59], [60], [61], [62]. Analytical models based on either variational or energy principle that provide solutions to the lower and upper bounds on the composite stiffness have been developed by various researchers. These includes first order bounds of Reuss [59] and Voight [63], second order bounds of Hashin and Shtrikman [64], Walpole [65], [66], [67], Willis [68] and Wu et al. [63], and third order bounds of Miller [69], Milton [70] and Beran et al. [71]. A family of models known as Eshelby's equivalent inclusion models have gained popularity over time. The original Eshelby's model was an exact solution to a single homogenous ellipsoidal inclusion problem. With time, Eshelby's model has been extended to incorporate inhomogeneous inclusions with non-zero far-field strain that considers the effect of interactions between neighboring inclusions. The Mori-Tanaka approach [72] which modifies the dilute strain concentration tensor (Eshelby's tensor) to account for inter-particle interaction was extended to modelling short fiber composites by Taya et al. [73] and Taya M. [74]. Traditionally, the Halpin-Tsai empirical relations [75], [76] originally derived from the work of Hermans [75] and Hill [76] were pioneering solutions to directly derive the complete set of engineering constants for SFRP composites. More recently, Tandon and Weng [77] exploited the Mori-Tanaka's model to derive explicit solutions

to the complete set of engineering constants particularly applicable to SFRP composites. Mori-Tanaka's assumption is only valid for low concentration particulate volume fractions and is a lower-bound solution to the composite stiffness. Lielen's model (double-inclusion model) [78], was developed for wide-range particulate volume fraction application, by interpolating between the Hashin-Shtrikman-Willis composite stiffness bounds [64], [65], [66], [67], [68] using the inverse rule of mixture principle.

Without claim to completeness, other analytical micromechanics models developed to predict elastic properties of SFRP composite includes the Cox-shear lag model [79], self-consistent method [80], the laminate analogy approach [55] etc. Bibliography on existing theoretical models for predicting other intrinsic quantities of unidirectional SFRP composites like the effective coefficient of thermal expansion (ECTE) and the effective thermal conductivity (ETC) are well documented in numerous literature [81], [82], [83]. Like the elasticity tensor, various solutions to the upper and lower bounds on the ECTE with differing level of accuracy have been developed such as upper bound models of Voight [84] and Kerner [85], and lower bound models of Reuss [86] and Turner [87]. Other solutions to the limit bounds on the ECTE of transversely isotropic composites includes models of Van Fo Fy [88], Schapery [89], Chamberlain [90] and Schneider [81], and Rosen and Hashin [91]. Analogically, the Mori-Tanaka's principle for predicting elasticity tensor of unidirectional SFRP composite has been extended by various researchers to predict the ECTE tensor [92], [93], [94], [95], [96], [97]. Other existing models for estimating the ECTE of SFRP composites includes but not limited to the models of Chamis [98], Thomas [99] and Cribb [100] etc. Existing analytical models for predicting the ETC tensor of unidirectional SFRP composite includes the equations of Halpin-Tsai [101], Nielsen [102], [103], [104], Nomura and Chou [105], Thornburg and Pears [106], Springer and Tsai [107] etc. The Giordano's approach

for predicting the permittivity of unidirectional SFRP composites based on dielectric theory of inclusions has also been extended to estimate the thermal conductivity tensor [108]. Elasticity models based on Eshelby's theory of inclusion such as the Mori-Tanaka and Lielens' double inclusion models have likewise been extended to predict thermal conductivity tensor of unidirectional SFRP composites which are known to yield more accurate results [97], [108], [109]. Traditionally, most mean-field theories used in the first homogenization step, such as the Mori-Tanaka-Benveniste formulations are limited to only two-phase composites. For multiphase heterogeneous composites, such as one having inherent pore inclusions or inhomogeneities, various studies [110], [111] have revealed higher levels of accuracy with multi-level homogenization schemes involving a lower-level pre-homogenization of the matrix with embedded pore phases or other inhomogeneities prior to an upper-level homogenization of the equivalent matrix with the filler reinforcements as compared to direct Voight averaging of the pseudo-grains resulting from decomposition of the RVE based on the different inclusion phases and characteristics. Although the Mori-Tanaka model can be generalized for multiphase composites, Norris [112] has shown that the method does not always satisfy Hashin - Shtrikman and Hill - Hashin effective stiffness bounds as would the two-phase composites. While theoretical asymptotic formulations based on multi-step and (or) multi-level mean-field homogenization approach are orders of magnitude faster and less computationally intensive in predicting effective properties of composites, they however fall short in terms of accuracy when considering the interaction between inclusions or estimating the microscopic stresses associated with the particulates especially when analyzing composites with sharp phase property contrast or high inclusion aspect ratio and volume fraction [56], [113], [114]. Moreover, they lack the capability to accurately model geometric peculiarities of inclusions such as irregularities in particle's morphology and characteristics and the spatial variations in the

distribution of microstructural features across the RVE which are typically found in actual SFRP composites [115].

With growing computational power and quantum processing speed, numerical boundary value problem (BVP) full-field methods, mainly finite element analysis (FEA) based homogenization methods have become more convenient for estimating effective properties of SFRP composites considering their high level of prediction accuracy and ability to model complex intricate microstructural geometric details associated with inclusions. Existing studies on numerical based homogenization method for property prediction of random SFRP composites are predominantly based on computer-generated deterministic RVE volumes stochastically filled with particulates based on a statistical technique [116], [117], [118], [119], [120], [121]. Examples of numerical homogenization studies involving periodic deterministic RVEs generated from statistical based techniques such as modified random sequential adsorption (RSA) algorithm for property prediction of SFRP composites which showed good agreement with results obtained from analytical based methods includes the studies of Berger et al. [122], Moussaddy et al. [123], Qi et al. [124], Mortazavi et al. [56], etc. These studies have shown that numerical based methods prevail in terms of accuracy over the analytical based methods when predicting properties of composites having inclusions with high aspect ratio and high-volume fraction [123], [125]. The continuum mechanics technique based on statistical correlation methods are known to perform poorly for property prediction of composites with non-spherical shaped inclusions. Series of publications [116], [118], [125], [126] have revealed that the RVE size, the required number of realizations and desired precision in predicting properties of heterogeneous SFRP composite depends on several factors including the microstructural composition and concentrations, the microconstituents phase property contrasts, the morphology, characteristics and dispersion of inclusion phases and the

evaluated quantity of interest. Given a desired level of accuracy and a reasonable RVE size, a method for determining the required number of deterministic RVE instances irrespective of the type of boundary conditions to predict the mean effective property of a random two-phase three-dimensional (3D) Voronoi mosaic SFRP composite with minimal dispersion in quantities was developed by Kanit et al. [116]. The method particularly applies to predicting effective properties of large volumes with few realizations of reasonable sized RVEs.

Over time, accurate microstructural characterization has become possible with advancement in modern imaging techniques. Reconstruction of 3D voxelated grayscale radiographs obtained from X-ray micro-computed tomography (μ -CT) imaging technique has gained popularity for characterizing the microstructure of SFRP composites [50], [108], [115], [127] and has been used to generate realistic RVEs for more accurate micromechanical analysis. For instance, Guven et al. [115] generated realistic RVEs of various sizes from 3D X-ray μ -CT voxelated images to numerically evaluate the effective material properties of two-phase particulate filled polymer composite which was found to have close agreement with experimentally measured properties. Although their study was based on two-phase SFRP composite, the method has been successfully extended to study the impact of micro-porosities or particulate inhomogeneities on predicted effective properties of multiphase particulate composites [110], [128]. While extensive studies have been previously carried out that numerically assessed the impact of porosities on the effective properties of SFRP composites using deterministic RVEs [110], [113], to the best of the authors knowledge, no known studies currently exist, that conducted impact assessment of porosities on the effective properties of SFRP composites utilizing realistic RVEs. The closest study that utilized realistic RVEs was carried out on nickel-reinforced alumina composites with roughly spherical shaped nickel particle reinforcement [128]. However, fillers typically found in

AM manufactured SFRP composites for strength related functionality are cylindrical shaped with high aspect ratio.

2.1.5 Overview of EDAM Process Simulation

With computational advancement and sophistication, simulation of manufacturing flow processes has gained traction overtime to gain in-depth understanding of the underlying physics responsible for process states to control and optimize the actual process and fine-tune the final print microstructure and effective properties to improve quality. Moreover, iterative design of manufacturing processes such as nozzle design, via experimental based approach could be very expensive compared to computational based methods. Additionally, in-process monitoring which may be intrusive and sensitive to disturbance is often limited by ready accessibility which complicates the measuring process [129]. Modelling of the flow process usually begins with identification of the manufacturing process and process variables including the precursor material parameters (type and characteristics of reinforcement, matrix, and other additives). Three (3) major manufacturing methods are widely used to fabricate composites depending on the precursor material properties which include: (1) short fiber suspension methods (2) squeeze flow methods (or advanced thermoplastic composites methods) and (3) porous media methods (or advanced thermoset composite methods). The current investigation focuses on the short fiber suspension methods which involves transport of fiber filled polymer suspension into mold cavity or through a die to form the composite and can be further classified into (a) injection molding, (b) compression molding and (c) extrusion processes. The reader is directed to [130] for further details on the process description, transport phenomena and applications. The extrusion and injection molding process have similar characteristics however while the processed polymer composite melt is injected into a closed cavity to solidify into shape in injection molding, the molten polymer is

ejected through and shaped by a die into an open environment in extrusion process. Although injection molding is a well-established and widely used method to batch process large quantities of thermoplastic composites part, the extrusion manufacturing method is a more promising technology due to the relatively lower material wastage, lower energy requirement and cost savings associated with the technology compared to the former. Process modelling of extrusion-deposition AM (EDAM) method is more pertinent to the current scope of work. Traditionally, polymer composite flow processes modelling is usually approached on a multi-scale level. The macro-scale level is usually on the order of the smallest part dimension (e.g. bead diameter) while the microscale level is usually on the order of the reinforcing particle's diameter. Coupling of macroscale and microscale physics is required to capture localized phenomena during the flow process. On the macroscale level, the primary aim is to relate the process/printing parameters to the flow or global deformation of the polymeric material. The key elements of a typical EDAM process (cf. Figure 2.3) include the feeding mechanism, heating and transport mechanism within the extruder-nozzle, bead deposition and road spreading/wetting process, inter-bead bond formation mechanism and bead cooling/solidification mechanism [131]. The choice of modelling aspect depends on the phenomena being investigated and the associated process parameters that can be controlled. For instance, various transport phenomena that influences the process/printing parameters includes the combined drag and pressure driven flow due to the turning action of the screw within the barrel that determines power and flowrate requirement, the contribution of the viscous dissipation in momentum transport to the overall energy transport that determines the systems heating or cooling requirements and the phenomena of extrudate swelling and melt fracture at the nozzle exit that affects the bead shape and stability [130].

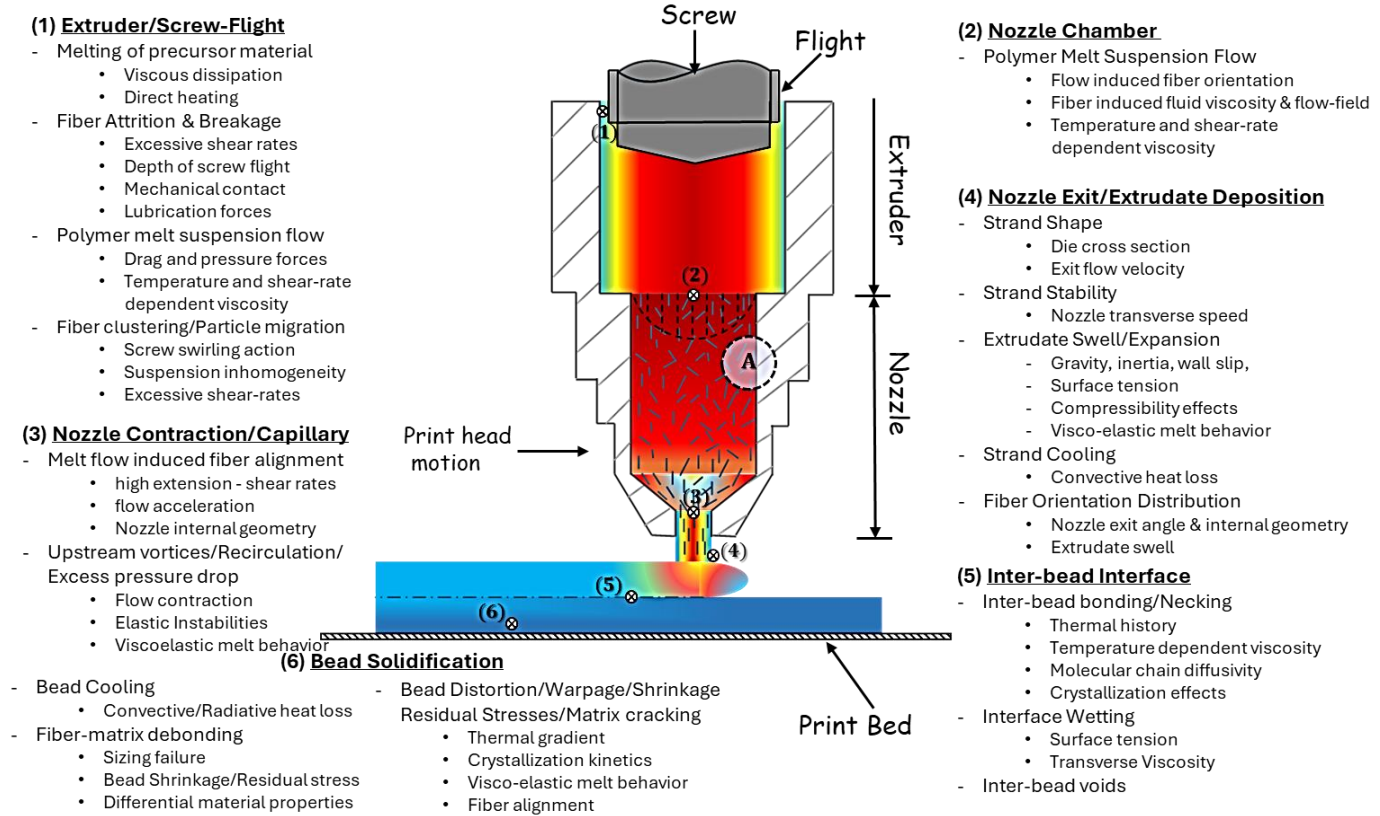


Figure 2.3: Aspects of EDAM Polymer Composite Processing Macro-Scale Level Modelling

Modelling the flow of polymer melt during EDAM composite processing can provide useful information about underlying mechanisms such as the velocity, pressure and temperature flow-fields. The macroscale simulation of the melt flow process can be divided into two categories namely the extrusion flow through the extruder-nozzle and the deposition flow onto the substrate. As the heterogeneous phase polymer melt flows through the liquefier, the orientation of the suspended fibres is determined by the flow-field. For highly loaded fibre suspension, the melt suspension viscosity and the flow-field are simultaneously influenced by the distribution of the fibre orientation. The mutually dependent phenomena necessitate a coupling of the flow-fibre orientation physics for more accurate process simulation. The rotational motion of the extruder screw induces swirling flow at the lubrication zone between the screw flight and the barrel walls

that results in high shear stresses. Figure 2.4a shows typical swirling streamlines of non-Newtonian viscoelastic polymer melt flow within the liquefier obtained from a one-way coupled flow-fiber orientation FEA simulation [132]. The flow swirling is known to induce stratum of fiber clusters with preferred orientation. Fiber attrition and breakage have been observed to occur mostly at the feed and compression zone of the liquefier [133]. Fiber breakage is known to relate to the shear stresses that develop at the screw flight in the compression zone, as such the flight depth is a major design parameter to control fiber breakage. Deeper screw flight-depth characteristics of variable pitch screws can reduce shear stresses and resulting fiber breakage and vice versa for narrower flight-depth in standard screws [133]. On the contrary, higher shear stresses was found to improve fiber dispersion. Figure 2.4b shows typical distribution of shear stresses in the screw flights in the metering zone for the standard and variable-pitch screws. Euler buckling has been identified as the primary failure mode responsible for fiber breakage due to hydrodynamic forces acting on the fibers [40]. Assuming constant fiber diameter and a kinetic model, [40] derived analytical expression for the residual fiber length. Three relevant interactions were identified by [134] to contribute significantly to the fiber breakage mechanisms including the fiber–fiber interaction, fiber–wall interaction, and fiber–matrix interaction. Optimum screw design through simulation can be used to reduce fiber breakage and at the same time improve dispersion.

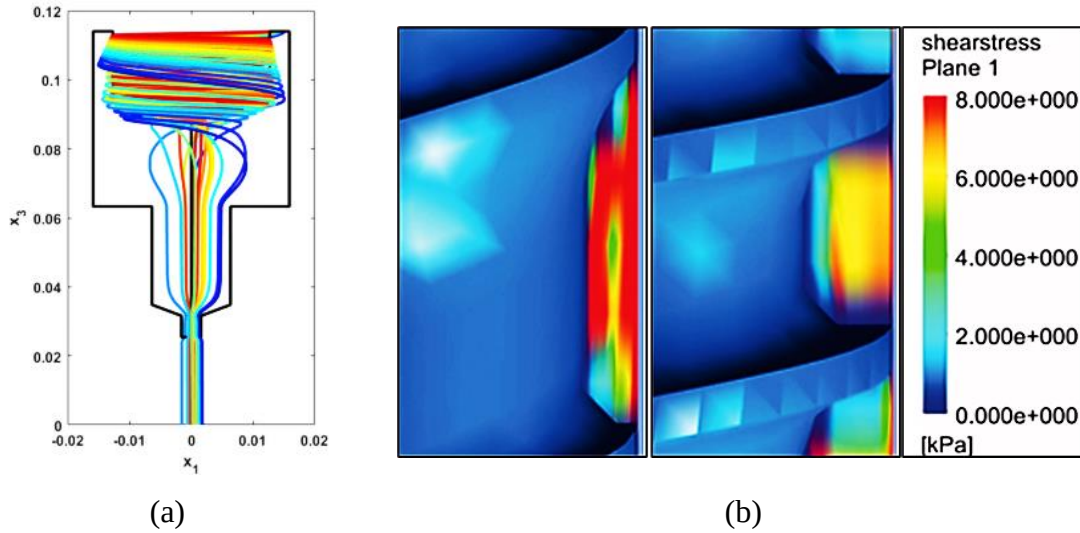


Figure 2.4: (a) Streamlines of non-Newtonian viscoelastic polymer melt swirling flow within the extruder-nozzle [132] (b) Distribution of shear stress in the screw flights in the metering zone for the standard and variable-pitch screws [133].

Majority of suspended fibers in the polymer melt within the nozzle are primarily aligned in the flow direction due to high shear stresses developed at the nozzle walls and high extension rate at the nozzle centre. Prior investigation based on simulation revealed higher levels of fiber alignment along the flow direction occur in the nozzle convergent contraction and nozzle capillary zones (cf. Figure 2.5a []). Flow vortices or recirculation are a result of abrupt nozzle contraction that are found at sharp corners and are found to be dependent on the visco-elastic properties of the polymer melt (cf. Figure 2.5b). Mezi et al. [135] found that increased fluid shear thinning reduced the upstream vortices. These vortices influence the fiber orientation field and results in significant pressure drop. Moreover, the dominant shear induced normal stress difference at the nozzle contraction and die exit were found from simulation to be majorly responsible for stable vortices and excess pressure drop [136]. The nozzle internal geometry and flow-field are thus key elements that determine the flow induced fiber orientation field. The flow aligned fiber orientation field in turn results in excessively high elongational viscosity within the nozzle due to the flow-fiber orientation coupling effect [137].

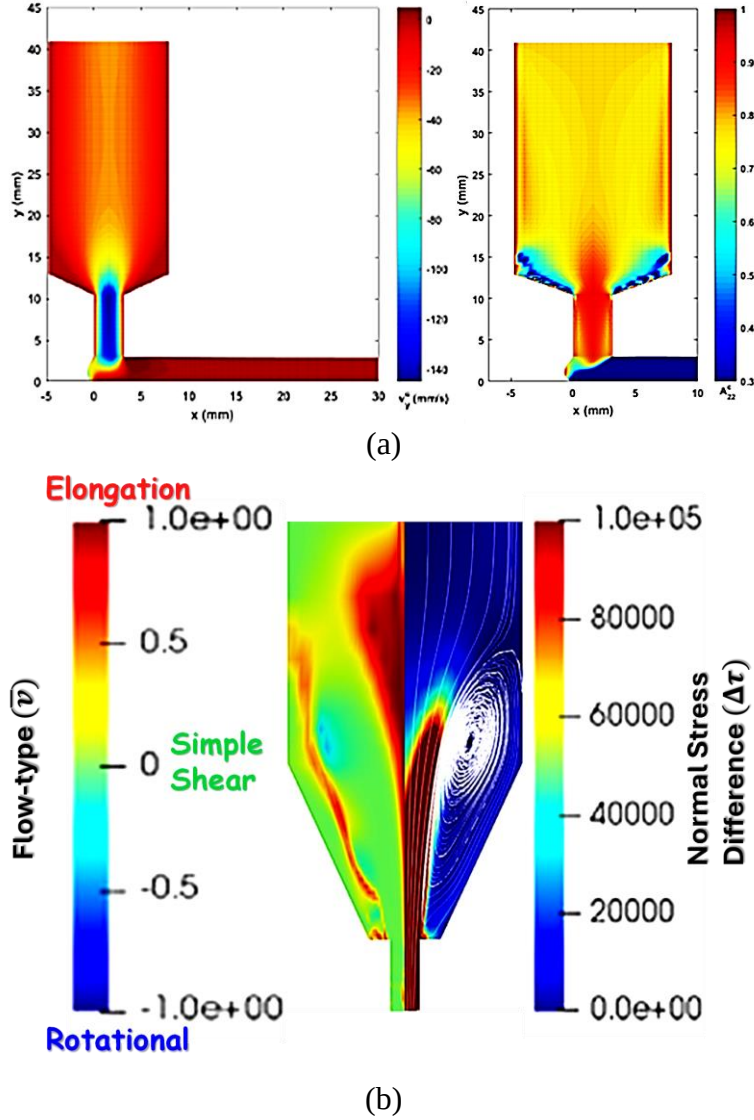


Figure 2.5: (a) Simulated results of the vertical velocity flow field - v_y (left), vertical fiber orientation distribution - A_{yy} (right) of typical polymer composite processing [22]. (b) Flow type $\bar{v}$ and normal stress difference $\Delta\tau$ from viscoelastic model with high extrusion velocity [136]:

Cooling of the extrudate via convection begins once it exits the nozzle and exposed to the open environment. Unexpectedly, melt flow simulations revealed that the rate of convective heat loss from the extrudate was declined with increased melt conductivity within the nozzle [138]. As the bounding surfaces of the extrudate becomes unconfined and exposed to the open environment, the parabolic melt flow velocity profile is transformed to a plug-flow velocity profile and due to stress relaxation and release of elastically stored deformation energy, sudden expansion of the

extrudate in the radial direction occurs, a phenomenon commonly referred to as extrudate swell/expansion [131], [138]. Extrudate swell ratios of polymeric composites have been predicted by various researchers using coupled flow-fibre orientation simulation [21], [22], [138], [139] and are typically found to range between 1.05 to 1.3 (cf. Figure 2.6a) [131]. The swell ratio is influenced by the viscoelastic properties of the polymer melt. Analytic approximations of the swell ratio developed by [140] is found to depend on the normal stress difference and shear stress at the nozzle wall. The effects of various factors on the extrudate swell have also been investigated by various researchers using simulation. For instance, [141] found that inertia and gravity effects which depends on the extrudate length significantly decreased swell ratio. Additionally, the effects of surface tension, wall slip, and pressure dependent-viscoelastic melt rheology were independently found to monotonically decrease swell ratios while compressibility effects resulted in an overall increase in the swell ratio. Flow-fibre orientation simulations also revealed that short fibres reinforcement reduced the extrudate swell ratio of polymer melt. However, the resulting fibre orientation distribution is seen not to be significantly affected by extrudate swell phenomena (cf. Figure 2.6b).

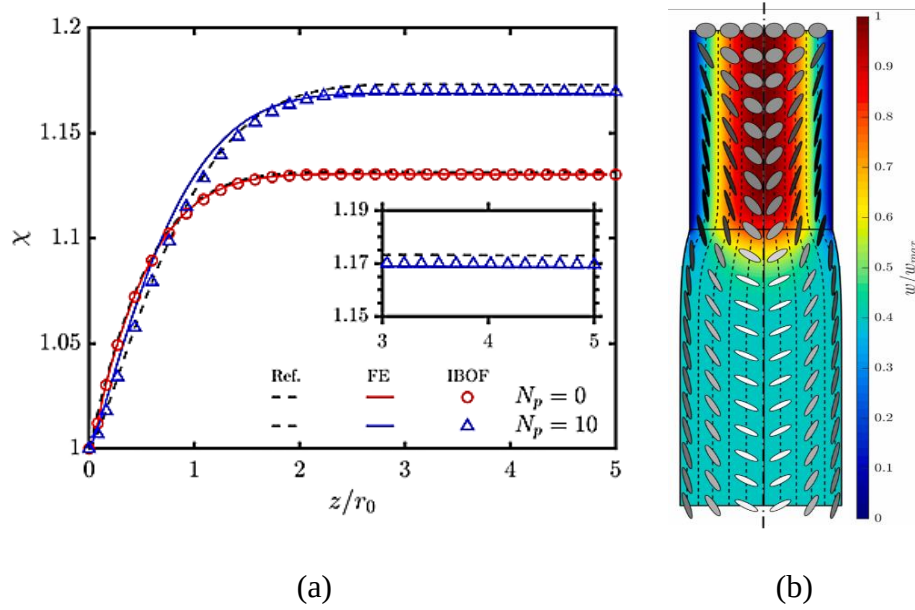


Figure 2.6: (a) Typical axisymmetric profile of the extrudate swell ratio starting from the nozzle exit for coupled (blue) and uncoupled (red) flow-fibre orientation solutions, (b) 2D axisymmetric view of the melt state in the nozzle and die-swell region showing velocity distribution (colormap), flow streamlines (dashed lines) and the average fibre orientation state (represented by ellipses) [142].

As the bead is deposited onto the substrate or onto a previous bead layer, the melt bends into a 90° shape. As a result of increasing radii from the bottom to the top across the bend, varying shear-rates develop across the bead which influences the fiber orientation state [1]. Various techniques have been employed to obtain the shape of the free surface of the deposited bead such the shape optimization technique [139], the finite volume/front-tracking method [143], or the algebraic coupled level-set/volume-of-fluid method [144]. The shape minimization techniques can be subdivided into the zero-surface tension method and the zero-penetration method depending on the choice of boundary condition and solution variables [139]. Various process conditions including gravity, inertia, wall slip, surface tension, pressure dependent viscosity and compressibility were found to significantly affect the extrudate swell ratio of Newtonian fluid up to -90% / +50% [141]. Moreover, viscoelastic non-Newtonian polymeric melts have been found to have higher swell ratios compared to Newtonian fluids due to additional elastic effects [141]

Numerical simulation carried out by [144] showed that the velocity ratio of the relative transverse velocity to melt flow velocity, and gap distance between the nozzle exit and substrate are parameters that influenced the bead shape. Low velocity ratio and small gap height resulted in better bead spreading and less circular and elongated cross section and vice versa (Figure 2.7a [145]). In a different numerical study, the bead morphology, inter-bead distance and layer height were found to be important parameters in minimizing inter-bead void volumes (Figure 2.7b [145]).

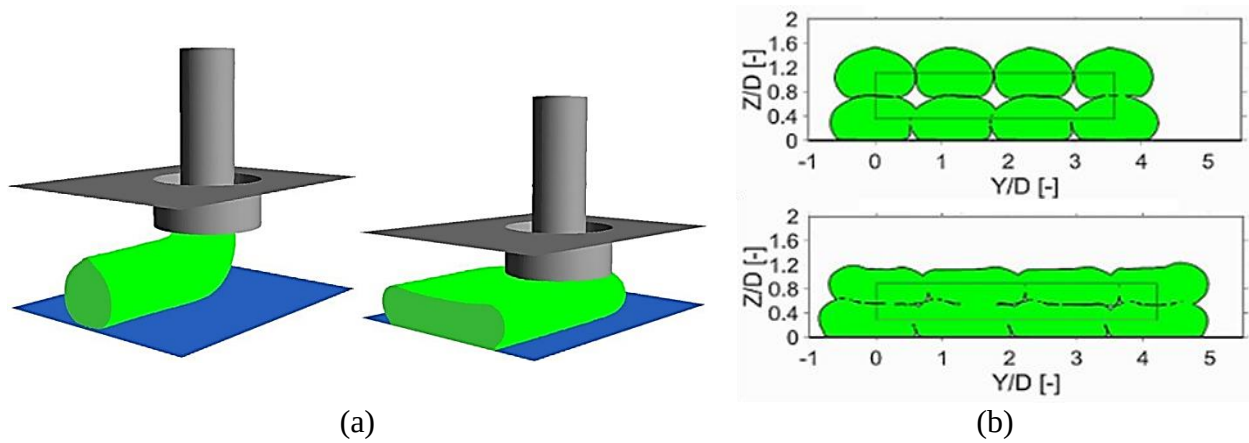


Figure 2.7: (a) simulated single bead deposition, for a large gap height and high velocity ratio (left), and a small gap height and low velocity ratio (right). (b) simulated beads cross sections showing the inter-bead voids for different layer height and inter-bead distances [145].

Bead stability is another modeling aspect investigated by various researchers. For instance, Balani et al. studied the effect of process parameters including the nozzle diameter, feed-rate and layer height which controlled flowrate, shear-rate and viscosity field, on extrudate deformation and inter-bead adhesion. Higher melt flow rates and higher shear rates were found to excessively reduce the viscosity and cause low precision that resulted in excessive extrudate deformation due to ‘sharkskin’ effect [146], [147]. High melt flow rates and small deposition times without provision for proper cooling of previous deposited bead layers can induce sagging due to gravity effects [1]. Consequently, excess deformation limits control of the resulting bead shape, surface roughness and reliability of print [148], [149]. As fresh and hot extrudates are deposited on previous bead layer, surface wetting, reheating/remelting ensues at the beads interface which are

two major factors responsible for effective interlayer bead adhesion [1]. The contact area between adjacent beads is determined by the wetting process while the dwell time ensures sufficient heating of the interface to allow adequate inter-molecular chain diffusion between adjacent beads through the interface thus ensuring proper fusion and inter-bead bond formation. A requirement for inter-diffusion bond formation is that the temperature of the polymer melt is above the glass transition temperature to reduce shape deformation and cracking. Surface wetting depends on the melt viscosity transverse to the lay direction and the relative surface energies between the bead and the adjacent surface (i.e. surface tension) and these properties in turn depend on the fiber reinforcement [1], [131]. Analytical models developed to simulate the bead wetting process includes the Crockett model [150], [151], and the Frenkel-type energy-based model [152]. Bellini et al. [138] utilized CFD to simulate the road spreading process as part of a complete EDAM process simulation. The bond formation or polymer sintering mechanism is popularly described using the reptation theory [1], [139] (Figure 2.8a). The process begins with the establishment of initial contact between adjacent beads, followed by the formation and growth of a neck at the beads' interface. Once a neck is formed, inter-diffusion of polymer chains across the neck takes place followed by a randomization of the polymer chains between the adjacent beads. Simultaneous cooling and phase transformation of the polymeric extrudate takes place due to convective heat loss during the wetting process which increases the melt viscosity. Adequate chain diffusion needed for effective bond formation depends on the thermal history at the adjoining bead interface. Rapid crystallization of the viscoelastic polymeric melt may also retard the bond formation process due to excessive increase in the polymer viscosity. Various techniques for predicting the thermal history of the bead have been developed such as the 2D model of Thomas and Rodriguez [153], and the lumped capacity model of Belleheumer et al. [154]. A bead's thermal history depends on

convective and radiative heat transfer across its surface and consequently the build environmental conditions that may or may not be controlled such as the air flow rate, and temperature. Heat dissipation from the print bead is facilitated by thermal conduction across the bead interfaces and through the conductive surface of the print bed. Phase transformation of the polymer melt from a viscous fluid to a viscoelastic solid during cooling results in evolution of transient relaxation moduli of the polymer. The solidification process results in material shrinkage and the development of internal stresses due to uneven cooling from temperature stratification in the radial direction coupled with the restraint posed by neighbouring beads. Additional stresses result from the anisotropic material behaviour due to the preferential alignment of the fiber reinforcement in the print direction and crystallization effects in semi-crystalline polymers. Moreover, disparity in the coefficient of thermal expansion between the fiber and matrix microconstituents contributes to the internal stress development during cooling. Realistic simulation of the solidification process would account for the various factors involved. 1D steady state and 2D quasi steady state heat transfer analyses have been found in literature to be insufficient in accurately simulating the heat transfer process and predicting the thermal history of the beads [155]. A comprehensive 3D analysis is necessary for a more accurate study of the bead cooling/solidification process. Figure 2.8b shows a typical temperature distribution across sections of the bead during the deposition process of fiber reinforced PEEK composite obtained from FEA simulation [156]. The heat transfer model was coupled with a non-isothermal dual crystallization kinetics model to predict the thermal history as well as the crystallization kinetics of the polymer composite and the influence on bond formation.

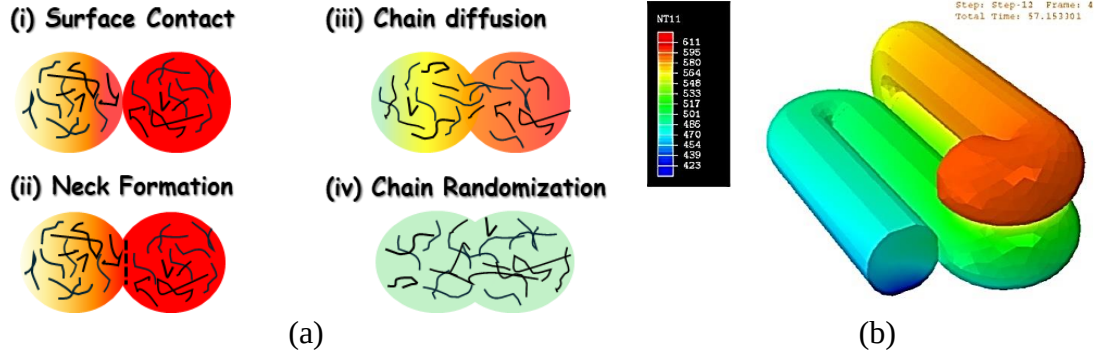


Figure 2.8: (a) Depiction of typical polymer sintering process (b) Simulated transient-state temperature distribution across bead sections during a typical deposition process of fiber reinforced PEEK composite [156].

Numerous efforts have been made in literature to simulate the solidification of prints in order to predict the resulting residual stresses and part deformations including warpage and sagging [157], [158], [159], [160]. For instance, Watanabe et al. [157] developed EDAM FEA process simulation models to predict the temperature distributions, deposited filament shapes, and warp deformation of a two (2) layer deposited polypropylene copolymer bead material, and the simulation results were found that agree well with experiments (cf. Figure 2.9a&b)

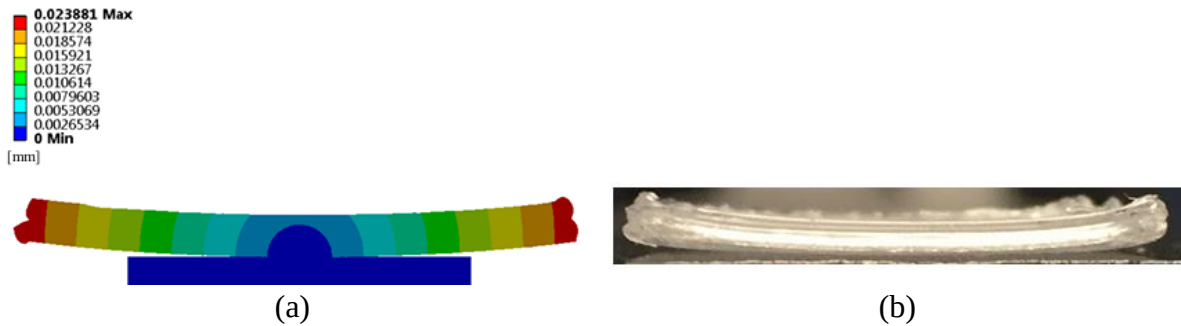


Figure 2.9: Steady-state warp deformation of two (2) layer polypropylene copolymer bead obtained from (a) FEA simulation (b) Experiment [157].

Most deposition flow simulation models are however based on numerous assumptions that oversimplify the actual solidification behavior. Currently, model improvement efforts are being made by various researchers such as the development of realistic 3D models that accurately capture the necessary physics involved in the process such as the thermo-viscoelastic behavior of the

polymer melt, the crystallization effects in semi-crystalline polymers and other non-linear effects [1], [156].

Evidently, much effort has been made to predict transport phenomena in EDAM polymer processing on a global level using macroscale simulation, however microscale simulations are important to more accurately predict certain phenomena on a local level such as the local flow-field around suspended particles, the motion of suspended particles, the deformation of suspended particles and the rheology of suspension. Theoretical analysis of single particle behavior in a viscous homogenous suspension is a well-known Fluid Structure Interaction (FSI) problem which has a variety of applications in key transport phenomena observed in physical rheological systems such as the movement of cells and platelets in blood plasma [161], the motion of reinforcing particles in fiber-filled polymer melt suspensions during polymer composite processing [1], proppants transport in fracturing fluids [162], migration of gaseous bubbles in quiescent viscous flows [163] etc. The rheology of particle suspensions is inherently complex due to a host of factors, including the presence of inter and intra particle forces arising from hydrodynamic interaction, contact collision between particles, confinement effect and particle deformability, Brownian disturbance, non-Newtonian viscoelastic fluid rheology, anisotropic particle geometry and concentration, and existence of various flow regimes within the system, etc. [20], [164], [165]. Various aspects of a typical microscale level simulation are depicted in Figure 2.10 below. The study of particle suspension dynamics often starts with the evaluation of single rigid spherical particle suspension under Newtonian simple shear flow which also provides insight into the rheology of dilute suspension [166], [167]. As an example, the dynamics of a single rigid ellipsoidal axisymmetric particle has been used extensively to investigate particle dynamics and flow-field structure of polymer composite melt flows during processing to assess their

microstructure [135], [165], [168]. Theoretical studies on particle migration in homogeneous viscous flow are commonly based on the assumptions of negligible inertia effects, Newtonian fluid rheology and non-deformable particle shape, conventionally referred to as “standard conditions” [169].

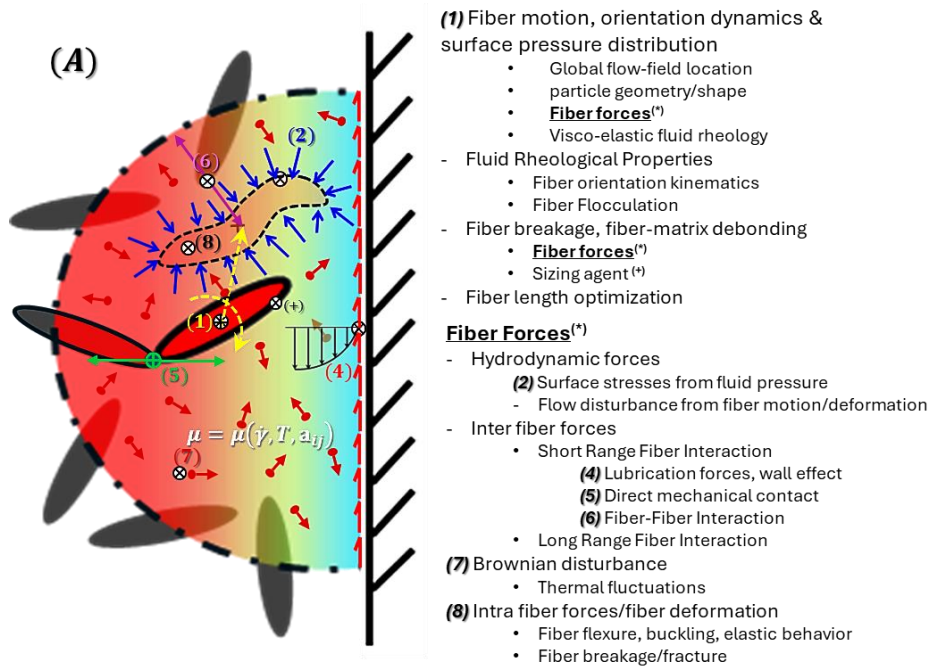


Figure 2.10: Aspects of EDAM Polymer Composite Processing Micro-Scale Level Modelling

Pioneering works of Oberbeck [170], Edwardes [171] and Jeffery [19] evaluated the orbit of an ellipsoidal rigid particle suspended in a homogenous shear viscous flow, where particle motion was determined to be a function of initial condition which has been validated experimentally [172]. In other work, Bretherton showed that lateral positioning of spherical isotropic particles remains unchanged relative to their initial position in quiescent sedimentation or unidirectional shear viscous flow [173]. Additionally, Cox [174] found that the orientation of transversely isotropic rigid particles in unconfined quiescent sedimentation would remain fixed at its initial value throughout its motion. These studies showed that under ‘standard conditions’, the motion and trajectory of a body of revolution depends on its initial conditions. For instance, the so

called ‘degeneracy’ of Jeffery’s orbits is used to describe the indeterminacy of particle’s motion in sheared viscous suspension whereby an axisymmetric particle may assume any of the infinitely possible metastable periodic orbits depending on its initial position. Experimental observations have revealed a tendency for suspended particles to eventually acquiesce to an equilibrium configuration within a finite timescale or equilibrium rate of approach irrespective of its initial configuration contrary to theoretical predictions based on “standard conditions” [169]. Jeffery [19] first suggested the possibility that spheroidal particles in a sheared viscous suspension with a theoretically indeterminate nature based on first order approximations, may eventually assume special configurations which are the path of least energy dissipation. Taylor [175] was one of the earlier researchers to provide experimental basis for Jeffery’s hypothesis and proposed that the higher order terms neglected in Jeffery’s approximate equations were responsible for the observed departure in the actual particle’s behavior from theoretical predictions. In a separate experimental study Saffmann et al. [176] showed that suspended particle’s do not always settle in preferred configuration states, however when they do, the contributions of the non-Newtonian fluid viscosity neglected in Jeffery’s approximate equations accounted mainly for the observed discrepancy between theoretical predictions and actual particle’s behavior. Other non-linear effects such as fluid and particle inertia, confinement and end effect were found to be infinitesimal as to significantly alter the particle’s motion within a finite timescale. Jeffery’s equations are generally found to be sufficient in predicting particle’s kinematics in a dilute and semi-dilute viscous shear-thinning particle suspension yielding only minor deviations from experimentally observed response [177], [178].

However, in the concentrated regime, the Jeffery’s model are no longer valid in predicting particle’s motion as the departures of theoretical prediction from experimental observations

becomes significant due to the combined effect of short range fiber interactions and shear-thinning fluid rheology neglected in Jeffery's model assumptions [177]. The effect of other rheological properties on the dynamics of a suspended particle such as higher order viscoelasticity fluid behavior that may be found in actual FSI physical systems have also been investigated by several researchers. An increase in the fluid elasticity results in a slow drift of prolate spheroids in sheared viscous suspension across spectrum of degenerate Jeffery orbits from a tumbling orbit to a log-rolling state and at drift rates proportional to the shear rate [178], [179]. Moreover, an excessive shear rate was found to result in particle realignment with the prevailing flow direction and the critical shear rate for flow realignment depended on the particles aspect ratio and Ericksen's number.

Computational models that account for particle inertia, non-Newtonian fluid rheology and/or shape deformability have more recently emerged. These more advanced models are often used to assess the departure of each from related theoretical predictions of fiber kinematics based on standard conditions. These advanced models are either developed from analytically formulations based on variational principles or asymptotic series expansion about the limits of standard theoretical model assumptions [169] or developed from numerical based simulations [180]. Analytical models are relatively faster, and computationally more efficient compared to numerical models, however these models are non-flexible, often restricted to predicting unique quantities and are less accurate due to oversimplification [129]. Methods based on variational principle are used to define limit bounds on the hydrodynamic drag coefficient of a spherical particle in GNF fluid subject to creeping flow [180]. The method has been successfully applied to obtain limit bounds solutions on the drag for spheres in GNF fluids for different viscosity models including the Newtonian model [181], power-law model [182], the Carreau model [183] and the

Ellis model [184]. The approach is more accurate for predicting hydrodynamic bounds in just Newtonian and power-law fluid models and the limit bounds diverges with increasing shear-thinning [180]. Perturbation based methods are generally used to compute solutions of fluid flow at relatively low Weissenberg number [185]. For instance, asymptotic perturbation about the leading order Newtonian fluid model has been used to evaluate the motion of transversely isotropic rigid particles in second-order viscoelastic fluid suspension [185], [186]. Consistent with experimental observations, at low shear rates, the viscoelastic fluids cause the suspended particle to slowly drift through various Jeffery's orbit until the attainment of an equilibrium orientation state in the flow vorticity direction. At higher shear rates, particles re-orient with the flow direction and its rotation suppressed. Extension of the theory to particle shapes revealed that while prolate spheroids tend towards a log-rolling position in the vorticity direction, oblate spheroids had an affinity for tumbling in the flow plane [187]. Deviations in particle shape from Jeffery's assumption of geometric asymmetry are found to result in significant changes in the particles motion. For general non-axisymmetric ellipsoids, Hinch and Leal [188] found the particles motion to be doubly periodic, consisting of a fast-tumbling motion around Jeffery's orbit and a slower drift representing a periodic change in Jeffery's orbit. On the contrary, application of the perturbation technique to investigate the effect of weakly shear-thinning fluid rheology on particles motion in unconfined sheared viscous suspension revealed that the degeneracy of Jeffery's orbit where unaffected by the non-Newtonian fluid rheology [189]. However, Jeffery's orbit and period were found to be instantaneously modified by the shear-thinning fluid behavior, and the quantitative modifications depended on the particle's initial conditions. Analytical based perturbation method have also been used to study the effect of other 'non-Standard' Jeffery conditions on the configurational determinacy of suspended particles and effective fluid rheology

of viscous flow suspension, such as particle and fluid inertia effects [169], [190], [191], contribution of Brownian disturbance [192], [193], [194], [195] and the effect of deformable particle shape [169], [196], [197]etc. As expected, the various investigated phenomena led to a modification of the dynamics and orbital configuration & drift of Jeffery-like particle.

Prior research that utilized numerical simulation techniques are summarized in various review literature [20], [198], [199], [200]. The method is tenable to increased model complexity and improved idealization of actual physical systems with increased accuracy. However, the method is computationally intensive and suffers from high computational cost. Numerical based models are classified into mesh-free or particle-based methods (PBM) and the traditional gridded continuum or element-based method (EBM) [198], [199]. PBM may be categorized based on physical or computational modelling. To avoid detraction from the primary focus of this dissertation, the reader is referred to existing review literature for more details [198], [199]. In PBM, the governing equations are discretized with moving sets of free particles retaining field-state information. PBM is a meshless, fully Lagrangian-based highly adaptive technique that allows for instantaneous tracking of individual particle response within a heterogeneous multiphase system and capable of modeling flow fronts, free surfaces and accurately solving large deformation problems [201], [202], [203], [204], [205]. Examples of PBM include the explicit Smoothed Particle Hydrodynamic (SPH) and the Moving Particle Semi-Implicit (MPS) method and Discrete Element Method (DEM). The SPH method utilizes an explicit FSI coupling algorithm, while the MPS technique uses an implicit fully coupled FSI algorithm for improved prediction accuracy and material properties [205]. Although PBM has been applied to evaluate the development of complex single-phase flows with non-linear fluid rheology [206], [207], [208], [209], the behavior of suspended particles in non-linear suspension flow are seldom evaluated with this method. In DEM,

the fiber particles are represented as chains of discrete particles (either hard spheres, rods or ellipsoids connected by joints/linkages with predefined mechanical behavior) that interact through hydrodynamic forces, inter-particle forces (short and long range hydrodynamic forces, Brownian and colloidal force), and intra-particle forces (elastic, flexural forces etc.) and the particles motion are computed by equilibrating the net force and torque on individual particles according to Newtons third Law [20], [200]. However, the fluid media is modelled as a continuum governed by the Navier-Stokes flow equation. In DEM one-way FSI coupling are often used for reduced computational cost, however for more accurate representation of the FSI interactions, back coupling that captures the effect of the fluids hydrodynamics forces on the particle's dynamics and the resulting disturbance on the surrounding flow due to the fiber's motion is required. Typical DEM solution techniques include the Dynamic Numerical Simulation (DNS), Lattice Boltzmann Method (LBM), and particle Finite Element Analysis (pFEA). The representation of fiber particles as interconnected chains of discrete particles interlinked with joints having directional stiffness and failure property definition makes it possible to capture the effect of fiber deformation and possible breakage at the joints and investigate the complex dynamics of the suspended particle in viscous flow field. Applications of DEM to FSI problems are summarized in various literature[20], [200]. DEM has been extensively used to study the behavior of single particles in Newtonian viscous suspension [210], [211], [212], [213], [214] and also in non-linear viscous suspension [215], [216]. Detailed bibliography on DEM based microscopic fiber suspension simulation can be found in Kugler et al. [20] considering different types of particle discretization method, flow-field types, FSI coupling types and active fiber forces with regards to various quantities being investigated such as particle motion and deformation, suspension rheology, fiber breakage, optimum fiber length etc. Two PBM methods may be combined to simulate EDAM process to

exploit their advantages. For instance, the SPH method is usually combined with the DEM method to exploit their advantage of SPH in simulating flows with moving boundaries/free surface, and the advantages of DEM in capturing inter-particle interactions and simulating large particle deformations [20], [24], [217].

EBM usually involves discretization of a continuum domain into sub-domain units. EBM types includes the Finite Element Method (FEM), the Finite Difference Method (FDM), the Finite Volume Method (FVM) and the Boundary Element Method (BEM) [198], [200]. In EBM, individual domain units are interconnected via topological maps. EBM involves transformation of a complex Partial Differential Equation (PDE) into a system of repetitive but simplified algebraic equations with solutions computed at the unit nodes, cells or elements level and collated to yield an approximate general solution. EBM is a well-established and highly evolved numerical technique extensively utilized in solving Computational Fluid Dynamics (CFD) and FSI transport problems. However, because FSI problems often involves free surfaces, moving and/or deformable boundaries, and very large deformation, the inherent complexities involved in remeshing, updating state variables and the errors introduced with excessive mesh distortion in EBM even with the Arbitrary Lagrangian-Eulerian (ALE) technique often makes EBM less attractive [198], [200]. For single particle suspension, an extra physical modelling that involves balancing the net hydrodynamic forces and couples on the surface of the particle is required to compute the particles motion.

In complex FSI systems with heterogenous distribution of suspended particles, it is customary to homogenize the multiphase continuum into a single uniform suspension phase having equivalent characteristics as the actual suspension using an averaging, smoothing or stochastic diffusing algorithm. Coupling of the characteristic aggregate particles' state (orientation and

spatial distribution) with the properties of the homogenized suspension is achieved using one of the available structure-based stress tensor rheological constitutive models [129], [130]. The evolution of the aggregate particles' average orientation dynamics can be computed using any of the available advection-diffusion moment tensor analytical models such as the Advani-Tucker's second order orientation equation of state [17], [20]. In BEM, solutions of state variables are computed only at the physical boundaries of the flow domain, hence reducing the problems dimensionality order by an order less compared to other EBM. BEM are thus faster, less computational expensive and more accurate than other EBM. FDM and FVM has been used to compute flow field and fiber orientation dynamics in mold filling process [218], [219], [220]. BEM has been successfully implemented to study flow-field development of particulate suspension in viscous shear flow [221], [222], [223] and FEM has been used to study single particle behavior in linear viscous shear flow [52], [224], [225], [226]. Relevant to this study are the applications of EBM in non-linear single particle suspension. For instance, 2D FEM has been used to simulate single rigid spheroidal particle behavior in dilute non-linear viscous shear flow [180], [226], [227], [228]. The findings revealed that shear-thinning effect only slightly affected the particle's kinematic, and this impact diminishes with increasing fiber slenderness. Moreover, increased shear-thinning was found to significantly reduced the magnitude of the pressure distribution surrounding the particle surface although had negligible effect on the shape pressure profile itself [180], [227]. Using a coupled FEM - Brownian dynamic simulation (BDS) based Langevin approach, Zhang et al. [229] investigated the effect of Brownian disturbance from the surrounding fluid molecules on a suspended single fiber's motion, which was found to be directly related to the magnitude of the Peclet number.

Macroscale level physics may vary depending on the manufacturing process and phenomena being investigated. Macroscale simulations are usually carried out to investigate global phenomena and predict processing conditions and global flow state such as velocity, pressure and temperature distribution field state, flowrates, rate of heat transfer, etc. The heterogenous nature of fiber suspension involving two phase mixture often necessitates multiscale level simulation to more investigate localized phenomena such as the development of micro-voids, fiber orientation, fiber breakage, etc. Moreover, localized phenomena can in turn influence macroscopic behavior such as the suspension rheology. Multiscale simulation involves coupling physics on two scales (i.e. macroscale and microscale) using constitutive equations [130]. Usually, macroscale level physics is based on the principle of continuum mechanics. For instance, using a one-way coupled multiscale FEA based modelling approach to simulate particle motion along streamlines of EDAM polymer melt flow process, Awenlimobor et al. [52], [227], [230], [231] investigated flow induced mechanisms that may be responsible for micro-void formation on the surface of suspended particles by studying the localized pressure distribution on the particles surface, which is a subject of the current research that would be explained in further details in succeeding chapters. Their findings showed that the surface pressure peaked at the tip of the suspended fibers during their evolution where most voids were observed to occur (cf. Figure 2.11).

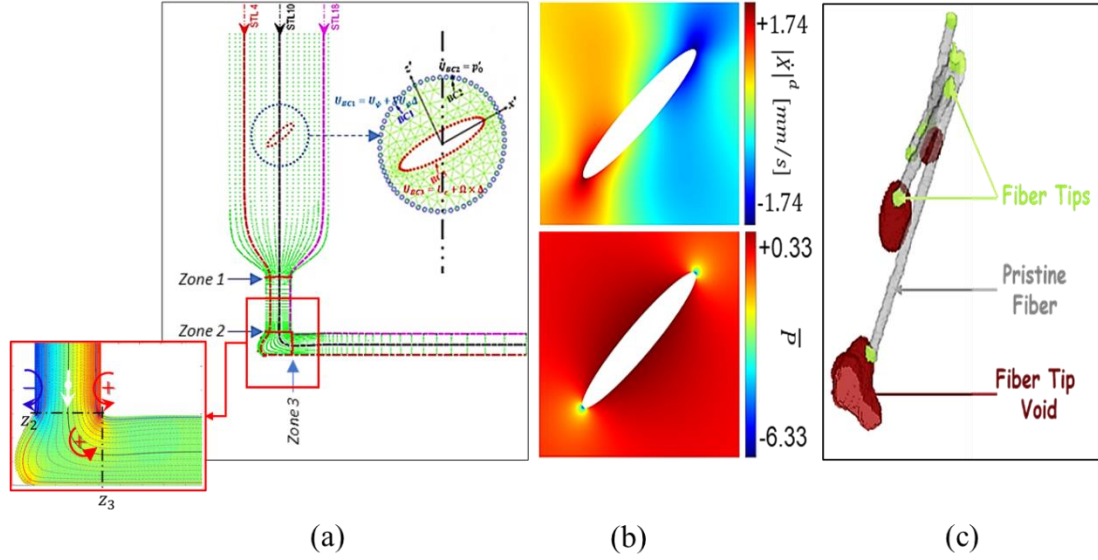


Figure 2.11: (a) Streamlines of typical macroscale EDAM polymer melt flow simulation showing single particle evolution micromodel analysis along a streamline location. (b) distribution of the velocity disturbance and pressure surrounding the particle at a peak minimum pressure location (c) Typical micro-void segregation at the tips of suspended fibers in 13% CF/ABS bead obtained X-Ray μ CT image analysis.

Two phase short fiber suspension flow simulation can be classified into three (3) based on the method of reckoning the suspended solid fiber phase in the polymer melt mixture [129] which includes:

- (a) Mathematical abstraction using analytical models such as the Folgar-Tucker's fiber orientation tensor model that predicts the transient fiber orientation tensor state based on the flow field velocity gradient, the particle's geometrical parameter and the fiber concentration accounted for in the phenomenological interaction coefficient used in the equation. Usually, the matrix fluid phase could be simulated using either any method such as EBM-FEM method [21], [22], [132], [232], or PBM-SPH method [202], [203]. Depending on the FSI coupling technique, either weak one-way or complete back-coupling model, the influence of the fibers on the polymer melt flow could be accounted for through the constitutive model used in the conservation equations that depends on the fiber orientation tensor state. This method is often used to study short fiber orientation evolution during EDAM processing.

(b) Discretization of the solid particle phase using either the PBM and/or EBM numerical approach. Here fiber motion and deformation such as fiber bending and breakage, etc. and its influence on the fluid's rheological properties and flow-field could be simulated and visualized. The method can be used to study fiber orientation evolution and nozzle clogging in EDAM processing. [24], [129]. Nozzle clogging is known to result from high degree of misaligned, long-length and cross-linked fibers in the nozzle contraction that results in large pressure drop. Two discretization numerical approaches could be coupled together to simulate the matrix fluid phase and the fiber solid phase separately to exploit their unique advantages. For instance [24] utilized discrete SPH method to model the Newtonian incompressible polymer matrix phase and used bonded DEM particles to model suspended fiber particles that allows the ability to capture fiber motion, deformation and breakage in typical EDAM polymer composite melt flow processing (cf. Figure 2.12).

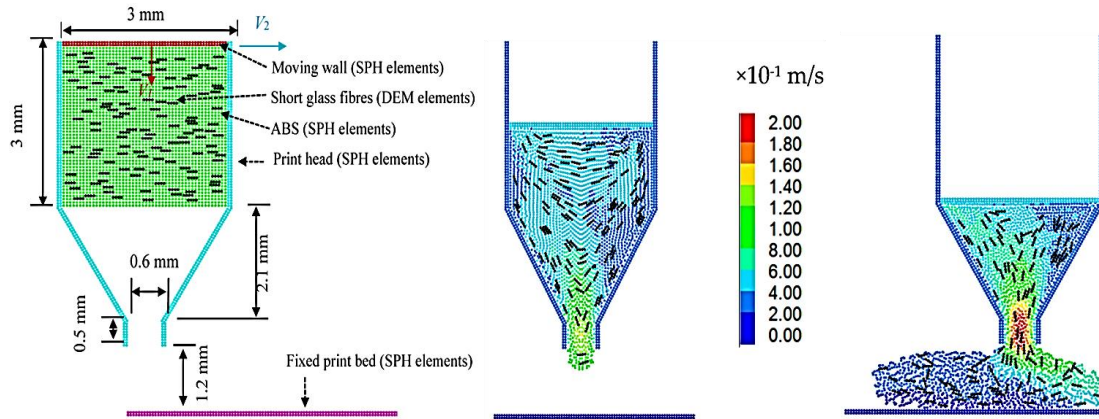


Figure 2.12: Coupled SPH-DEM EDAM SFRP polymer composite melt flow simulation showing initial configuration (left), velocities of polymer matrix at printing time of $t = 0.05$ sec (middle) and $t = 0.1$ sec (right). [24]

(c) Phase homogenization method using an equivalent fluid phase mixture that combines properties of the pure polymer resin and the fiber inclusions. The homogenized fluid phase can be simulated using the MPS particle method where each particle is a composite material having

equivalent physical properties of the resin and fiber phase present based on their weight fractions computed using the rule of mixture. The method has been used to simulate bead cooling during deposition and predict the evolution of deposited bead cross-section during solidification [233].

The choice of simulation method depends on the transport phenomena of the EDAM SFRP composite process being investigated, the desired degree of accuracy and the level of sophistication involved. More details on the physics involved in multiscale short fiber suspension flow simulation is provided in subsequent sections.

2.1.6 Physics of EDAM Process Simulation

The assumptions, system domain boundary, physical laws, constitutive equations, and boundary conditions necessary for development of an EDAM polymer composite melt flow process model are briefly discussed in the sections following. According to [130], the process of building a model is iterative and begins with identification of the physical process, followed by defining an objective, process simplification through assumptions, development of theoretical models to define the process, selection of a suitable solution technique, generating results/solutions and validating model predictions with experimental findings. If the solutions agree with process physics the model design is accepted, otherwise the model assumptions are revised, and the model development process is repeated until valid solutions are obtained.

1.1.1.1. Transport equations

The fundamental governing equations used to model transport phenomena include the conservation equations that describe the physical laws of the system, the constitutive relations that describe the material and their phenomenological behaviour and the boundary conditions that specifies constraints at the surfaces and interfaces of the specified process system domain to define

its interaction with its surrounding. Additionally, mathematical assumptions may be introduced to simplify the process for ease of computation of the desired solution variables.

1.1.1.1.1. Conservation equations

The primary equations governing the polymer melt flow are the conservation equations of mass, momentum and energy summarized below. For future reference, we define the material derivative operator used for Lagrangian to Eulerian frame transformation given as

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \dot{X}_j \nabla_{X_j} \quad (2.19)$$

where the gradient operator $\nabla_{X_j} = \partial/\partial X_j$. The conservation of mass or the continuity equation is given as

$$\frac{\partial \rho}{\partial t} + \nabla_{X_j}(\rho \dot{X}_j) = \frac{d\rho}{dt} + \rho \nabla_{X_j} \dot{X}_j = -s(t, \underline{X}) \quad (2.20)$$

where ρ is the fluid density, t is the time, X_j are component directions of the position vector $\underline{X}$, $\dot{X}_j$ are the scalar components of the velocity vector $\underline{\dot{X}}$ and s is the rate at which mass change per unit volume per unit time through the system. Since most polymer composites melt are incompressible and there is no mass change, eqn. (2.20) reduces to

$$\nabla_{X_j} \dot{X}_j = 0 \quad (2.21)$$

For fibrous suspension, assuming fiber are incompressible, with negligible velocity and stress-strain change and negligible body forces, the density ρ in eqn. (2.20) can be replaced with the partial density of the polymer matrix ρ_m such that $\rho_m = \rho \vartheta_m$, where ϑ_m is the volume fraction of the polymer matrix phase [130]. The equation for the conservation of momentum is given as

$$\rho \frac{d\dot{X}_i}{dt} = \nabla_{X_j} \sigma_{ji} + \rho f_i \quad (2.22)$$

where the Cauchy stress tensor σ_{ij} is given as $\sigma_{ij} = \tau_{ij} - p\delta_{ij}$, τ_{ij} is the deviatoric (viscous) stress tensor and f_i is the body force. In combination with the material derivative, the resulting (2.22) becomes

$$\rho \left(\frac{\partial \dot{X}_i}{\partial t} + \dot{X}_j \nabla_{X_j} \dot{X}_i \right) = \nabla_{X_j} \sigma_{ji} + \rho f_i \quad (2.23)$$

For homogenous fluids, the constitutive equation that relates the viscous stress tensor τ_{ij} to the strain rate tensor $\dot{\epsilon}_{ij}$ is given as

$$\tau_{ij} = C_{ijkl} \Gamma_{kl} \quad (2.24)$$

where the stress-strain rate constant C_{ijkl} is a fourth order viscosity tensor and the strain rate tensor Γ_{ij} is the symmetric part of the decomposed velocity gradient $L_{ij} = \nabla_{X_j} \dot{X}_i$ given as

$$\Gamma_{ij} = \frac{1}{2} (L_{ij} + L_{ji}) \quad (2.25)$$

The scalar magnitude of strain rate or deformation tensor $\dot{\gamma}$, which is independent of the coordinate system is given as $\dot{\gamma} = \sqrt{2\Gamma_{ij}\Gamma_{ji}}$. In continuum mechanics, the velocity vector, $\dot{X}_i$ describes the translation of continuum, while the strain rate tensor Γ_{ij} describes deformation of continuum and the rotation rate tensor Ξ_{ij} describes the rotation of continuum, where Ξ_{ij} is the anti-symmetric part of the decomposed velocity gradient L_{ij} , such that

$$\Xi_{ij} = \frac{1}{2} (L_{ij} - L_{ji}) \quad (2.26)$$

The vorticity tensor $\omega_{ij} = 2\Xi_{ij}$. Simplification of eqn. (2.24) for Newtonian fluids considering material symmetry and isotropy is given as

$$\tau_{ij} = \lambda_1 (\nabla_{X_k} \dot{X}_k) \delta_{ij} + 2\mu_1 \Gamma_{ij} \quad (2.27)$$

where λ_1 and μ_1 are the Lamé's constants. Based on the Stokes assumption of equal mechanical and thermodynamic pressures such that $\lambda_1 = -2/3 \mu_1$, (2.27) reduces to

$$\tau_{ij} = \mu_1 \left[-\frac{2}{3} (\nabla_{x_k} \dot{X}_k) \delta_{ij} + 2\Gamma_{ij} \right] \quad (2.28)$$

Because most polymer melts suspension are essentially incompressible, i.e. $\nabla_{x_k} \dot{X}_k = 0$, then $\tau_{ij} = 2\mu_1 \Gamma_{ij}$ and the conservation of momentum equation in (2.28) reduces to

$$\rho \left(\frac{\partial \dot{X}_i}{\partial t} + \dot{X}_j \nabla_{x_j} \dot{X}_i \right) = -\nabla_{x_i} p + \mu_1 \nabla_{x_j}^2 \dot{X}_i + \rho f_i \quad (2.29)$$

For non-Newtonian polymer melts rheological behavior, such as shear-thinning, shear-thickening and Bingham plastics, the viscosity may be expressed a function of the shear rate magnitude, temperature and pressure, i.e. $\mu_1 = \mu_1(\dot{\gamma}, p, \mathcal{T})$. Likewise, the conservation of energy equation is given as [130].

$$\rho \frac{de}{dt} = -\nabla_{x_k} q_k - p \nabla_{x_k} \dot{X}_k + \tau_{ij} \nabla_{x_i} \dot{X}_j + s \left(e + \frac{1}{2} \dot{X}_k \dot{X}_k \right) + \dot{r} \quad (2.30)$$

where e is the internal energy of fluid particles per unit mass, q_k is the heat flux vector, $\dot{r}$ is the rate of internal energy generation per unit volume such as from chemical reactions of the polymer chains and/or microwave induction heating of the polymer [130]. After many substitutions and simplification, utilizing the thermodynamic relations and ignoring higher order terms of small magnitude, the energy equation reduces to

$$\rho c_p \left(\frac{\partial \mathcal{T}}{\partial t} + \dot{X}_k \nabla_{x_k} \mathcal{T} \right) = -\nabla_{x_k} q_k + \tau_{ij} \nabla_{x_i} \dot{X}_j + \dot{r} \quad (2.31)$$

where c_p is the specific heat at constant pressure, and $\mathcal{T}$ is the temperature. The conductive heat flux q_i through the polymer composite melt is defined by Fourier's law of steady heat conduction given as

$$q_i = -k_{ij} \nabla_{x_j} \mathcal{T} \quad (2.32)$$

If we assume isotropic thermal conductivity, k for the polymeric material, then the conduction term in (2.31) is simplified to $-\nabla_{x_i} q_i = \nabla_{x_i} k_{ij} \nabla_{x_j} \mathcal{T} = k \nabla_{x_i}^2 \mathcal{T}$.

The phenomena being investigated, the type of analysis and process assumptions considered in the study determine the transport equations used and further simplifications of the equations for a particular simulation. For instance, isothermal processes that investigate melt flow and fiber orientation dynamics within the liquefier and extrudate deposition and swelling due to pressure drop post extrusion etc. involve computation of the velocity and pressure flow-field and fluid viscosity and may require only the mass and momentum conservation equations [21], [22], [132], [234], [235], [236]. Conversely, non-isothermal processes involving melting and flow dynamics within the liquefier which depend on the thermal properties of the material and heat capacities of the extruder-nozzle [237] or processes involving heat transfer such as bead cooling, solidification and crystallization [156], bond formation [154], residual stresses and warpage [157], [238] etc. may involve calculation of temperature distribution in addition to the velocity and pressure flow-field and thus require the energy conservation equation. Polymer melt flow simulations often assume steady state, viscous, incompressible fluid and low Reynolds number (creeping/Stokes) flow with negligible inertia. In such instance, the momentum equation is simply a balance between the viscous and body forces given as

$$\nabla_{x_j} \sigma_{ji} + \rho f_i = 0 \quad (2.33)$$

And the energy equation is a balance between the convection, conduction and viscous dissipation terms given as

$$\rho c_p \dot{X}_k \nabla_{x_k} \mathcal{T} - \nabla_{x_i} k_{ij} \nabla_{x_j} \mathcal{T} - \tau_{ij} \nabla_{x_i} \dot{X}_j = 0 \quad (2.34)$$

For very small Peclet number, the convection term in (2.34) vanishes, and for small Brinkman number, the viscous dissipation term likewise becomes negligible [239].

1.1.1.1.2. Constitutive equations

In addition to the fundamental governing laws of conservation, constitutive equations are objective empirical expressions that relate process parameters and define localized phenomenological material behavior on a global scale to completely describe the overall transport phenomena such as defining nonlinear material relations, fiber-matrix and fiber-fiber interactions, chemical kinetics etc. [130]. Depending on the transport phenomena, model assumptions and level of sophistication, various types of constitutive equations may exist, and we describe a few below that are relevant to EDAM polymer composite process simulation.

2.1.6.1.1.1 Homogenous pure solvent model

The relationship between the material stress tensor and strain rate tensor for isotropic and incompressible fluid and is typically expressed as

$$\tau_{ij}^s = 2\mu \Gamma_{ij} \quad (2.35)$$

Various rheological models for μ exists that define the material behavior which may depend on the process state variables. The process state dependency depends on the type of analysis and level of model sophistication. The most basic and simplest of these models is the linear Newtonian model where the viscosity μ is simply a constant (i.e. $\mu = \mu_1$). Other common models used to describe nonlinear behavior of thermoplastics material are given in Table 2.1 below [129], [130], [131].

Table 2.1: Typical thermoplastic viscosity models used in EDAM simulation

Viscosity Model	Expression	Parameters definition
Power Law	$\mu = \alpha_T m \dot{\gamma}^{n-1}$	m – consistency index n – power law index α_T – temperature dependence factor
Carreau-Yasuda	$\frac{\mu - \alpha_T \mu_\infty}{\mu_0 - \mu_\infty} = \alpha_T \{1 + (\lambda_t \alpha_T \dot{\gamma})^a\}^{\left(\frac{n-1}{a}\right)}$	μ_0 – zero shear viscosity μ_∞ – infinite shear viscosity λ_t – time constant a – transition parameter

Cross Law	$\frac{\mu}{\mu_0} = \alpha_T \{1 + (\mu_0 \alpha_T \dot{\gamma} / \tau_c)^{1-n}\}^{-1}$	τ_c – critical shear stress
Sprigg Law	$\frac{\mu}{\mu_0} = \alpha_T \begin{cases} 1 & \dot{\gamma} < \dot{\gamma}_0 \\ (\dot{\gamma} / \dot{\gamma}_0)^{n-1} & \dot{\gamma} \geq \dot{\gamma}_0 \end{cases}$	$\dot{\gamma}_0$ – zero shear rate

The Carreau-Yasuda model best describes the actual behavior of most thermoplastics materials since at low shear rates, the viscosity is basically Newtonian and viscosity plateaus at high shear rates. Although the power law model is more popular and simpler than other nonlinear models and it accurately captures shear-thinning behavior at moderate shear rates, however the model yields physically unrealistic values at low and high shear rate extremes while the Spriggs model yields erroneous results at high shear rates and does not transition smoothly from the Newtonian to the shear thinning behavior [130]. Various models have been used to represent the temperature shift factor in Table 2.1 above, a few of which are given in Table 2.2 below. Although the Arrhenius model is mostly used, the WLF model is more accurate for amorphous polymers. Other models include the Coffin Manson model and the modified Coffin Manson - Norris Landzberg model, the Enns and Gillham model, model of Stolin et al., Lee and Han model, Tajima and Crozier model and Hou's model etc. detailed in [129], [130], [131].

Table 2.2: Typical models for temperature shift factor (α_T) used in EDAM simulation

Shift Factor Model	Expression	Parameters definition
Arrhenius Law	$\alpha_T = \exp \left\{ \frac{E_a}{R} \left(\frac{1}{\mathcal{T}} - \frac{1}{\mathcal{T}_{ref}} \right) \right\}$	E_a – activation Energy R – ideal gas constant $\mathcal{T}_{ref}$ – reference temperature
Williams-Landel-Ferry (WLF)	$\alpha_T = \exp \left\{ - \frac{C_1 (\mathcal{T} - \mathcal{T}_{ref})}{C_2 + (\mathcal{T} - \mathcal{T}_{ref})} \right\}$	C_1 & C_2 – Fitting Constants
Coffin Manson	$\left(\frac{\Delta \mathcal{T}_{ref}}{\Delta \mathcal{T}} \right)^M$	M - acceleration rate $\Delta \mathcal{T}_{ref}$ – reference temperature difference

2.1.6.1.1.2 Heterogenous fiber suspension model

The effective viscosity of fiber suspension is known to be higher than the pure polymer material due to the influence of the suspended particles. Fiber suspension can be classified into dilute, semi-dilute and concentrated regime depending on the average number of fiber particles per unit volume, n_f (number density) or the fiber volume fraction $\vartheta_f = \pi n_f l_f^3 / 4 r_e^2$ as given in eqn. (2.36) below.

$$\left\{ \begin{array}{ll} n_f < \frac{1}{l_f^3} & \text{or} \quad \vartheta_f < \frac{1}{r_e^2} \quad \text{dilute} \\ \frac{1}{l_f^3} \leq n_f < \frac{1}{l_f^2 d_f} & \text{or} \quad \frac{1}{r_e^2} \leq \vartheta_f < \frac{1}{r_e} \quad \text{semidilute} \\ n_f \geq \frac{1}{d_f l_f^2} & \text{or} \quad \vartheta_f \geq \frac{1}{r_e} \quad \text{concentrated} \end{array} \right. \quad (2.36)$$

where l_f , d_f and r_e are the average fiber length, fiber diameter and aspect ratio respectively [57], [130], [240]. In the dilute regime, there is no restriction on the fibers motion due to hydrodynamic forces or mechanical contact. In the semi-dilute regime, the hydrodynamic forces influence the fiber's motion due to the suspension rheology however there are yet no physical constraints on particles motion due to mechanical contact. The average interparticle spacing h_m is small usually on the order of the fiber diameter i.e. $h_m \gg d_f$. h_m and consequently, the upper limit of n_f becomes dependent on orientation state i.e.

$$\left\{ \begin{array}{ll} h_m \cong \frac{1}{n_f l_f^2} & \text{or} \quad n_f \ll \frac{1}{d_f l_f^2} \quad \text{random} \\ h_m \cong \frac{1}{\sqrt{n_f l_f}} & \text{or} \quad n_f \ll \frac{1}{d_f^2 l_f} \quad \text{aligned} \end{array} \right. \quad (2.37)$$

In the concentrated regime, the average interparticle spacing h_m is very small such that fiber motion is affected by mechanical interactions between particles and physical boundaries.

Most commercial SFRP composites suspension fall within the concentrated class of fiber suspension.

A general expression for the composite stress tensor for fiber suspension [130], [241] is given as

$$\tau_{ij}^{s+f} = 2\mu \Gamma_{ij} + \vartheta_f \mu A_\tau \Gamma_{kl} a_{ijkl} + B_\tau [\Gamma_{ik} a_{kj} + a_{ik} \Gamma_{kj}] + C_\tau \Gamma_{ij} + F_\tau a_{ij} D_r \quad (2.38)$$

where A_τ, B_τ, C_τ and F_τ are material constants, D_r is the rotary diffusivity due to Brownian motion, and a_{ij} and a_{ijkl} are second and fourth order orientation tensors. Alternately, the above expression can be rewritten as follows [130]

$$\tau_{ij}^{s+f} = n_I \{ \Gamma_{ij} + n_P \Gamma_{kl} a_{ijkl} + n_S [\Gamma_{ik} a_{kj} + a_{ik} \Gamma_{kj}] \} \quad (2.39)$$

where n_I, n_P , and n_S are functions of $\mu, \vartheta_f, A_\tau, B_\tau$ and C_τ . n_I captures all isotropic contributions from the suspension microconstituents to the overall viscosity, while the particle number, n_P and the shear number n_S capture anisotropic contributions of the microconstituents. For dilute and semi-dilute high aspect ratio fiber suspension in the absence of Brownian motion, Lipscomb derived the following expression for the composite stress often called the Transversely Isotropic Fluid equation [200], given as [240]

$$\tau_{ij}^{s+f} = 2\mu \Gamma_{ij} + 2c_1 \vartheta_f \mu \Gamma_{ij} + 2\vartheta_f \mu N_p \Gamma_{kl} a_{ijkl} \quad (2.40)$$

where c_1 is a constant and N_p is a dimensionless FSI coupling parameter and is given by Lipscomb et al. [242] to be a function of r_e as

$$N_p = \frac{r_e^2}{2 \log r_e} \quad (2.41)$$

An alternative expression developed by Batchelor [243] for N_p likewise in terms of r_e is given as

$$N_p = \frac{1}{3} 4r_e^2 \varepsilon f(\varepsilon), \quad f(\varepsilon) = \frac{1 + 0.64\varepsilon}{1 - 1.50\varepsilon} + 1.659\varepsilon^2, \quad \varepsilon = \frac{1}{\log(2r_e)} \quad (2.42)$$

In the semi-dilute regime, expression for N_p was developed by Dinh and Armstrong [244] based on the slender body theory and given as

$$N_p = \frac{r_e^2}{3 \log(2 h_m/d_f)} \quad (2.43)$$

An alternative expression developed by Shaqfeh and Fredrickson [245] for dilute and semi-dilute fiber suspension with isotropic fiber orientation distribution is given as

$$N_p = \frac{4}{3} r_e^2 \left\{ \frac{1}{\log(\vartheta_f^{-1}) + \log \log(\vartheta_f^{-1}) + c''} \right\}, \quad c'' = \begin{cases} -0.66 & \text{random} \\ +0.16 & \text{aligned} \end{cases} \quad (2.44)$$

Likewise, Phan-Thien and Graham [246] proposed the following expression for N_p and for fiber aspect ratio in the range $5 < r_e < 30$ given as

$$N_p = \frac{r_e^2 (2 - \vartheta_f/g_v)}{2(\log(2r_e) - 1.5)(1 - \vartheta_f/g_v)^2}, \quad g_v = 0.53 - 0.013r_e \quad (2.45)$$

Azaiez [247] summarizes the above expressions for N_p dilute and semi-dilute suspension thus

$$N_p = \begin{cases} \frac{r_e^2}{2(\log(2r_e) - 1.5)} & \vartheta_f \leq \frac{1}{r_e^2} \quad \text{dilute} \\ \frac{r_e^2}{3 \log \left[\frac{\pi}{2\vartheta_f r_e} + g \left(\sqrt{\frac{\pi}{\vartheta_f}} - \frac{\pi}{2\vartheta_f r_e} \right) \right]} & \frac{1}{r_e^2} \leq \vartheta_f < \frac{1}{r_e} \quad \text{semi-dilute} \end{cases} \quad (2.46)$$

where $g_v = 1 - \sqrt[5]{4\epsilon_{ijk}a_{i1}a_{j2}a_{k3}}$. The foregoing expressions ignore interparticle interaction and are applicable only to dilute and semi dilute - high aspect ratio fiber suspension and cannot accurately model concentrated fiber suspension where fiber interaction involving mechanical contact may occur. However, the dilute and semi-dilute models can still be extended to some extent to model concentrated particle suspension typically by modifying the FSI coupling constant N_p where N_p is obtained from regression fitting operations to the rheological material functions of the suspension [240]. One way is to utilize direct simulations to obtain the aggregate hydrodynamic

torque Q_H^f and stresslet S_H^f acting on all fiber particles and computed from the forces and torques acting on the fibers suspended in a sufficiently large representative volume V . In such case, the ensemble average stress contributed by the particles to the composite stress is given as [200]

$$\tau_{ij}^f = \frac{1}{V} \sum_{\forall p} \left(S_{Hij}^f + \frac{1}{2} \epsilon_{ijk} Q_{Hk}^f \right) \quad (2.47)$$

The stresslet S_{Hij}^f acting on a particle f can be obtained by integrating the symmetric part of the first moment of the stress σ_{ij} over all possible fiber orientation $\underline{\rho}$ and over the particle surface S using the fiber orientation distribution weighting function $\psi(r, \underline{\rho}, t)$ according to

$$S_{Hij}^f = \frac{1}{2} \iint_S (X_i \sigma_{ji} \hat{n}_i + X_j \sigma_{ij} \hat{n}_j) \psi(r, \underline{\rho}, t) d\rho dS \quad (2.48)$$

The composite stress tensor is thus simply the superposition of the homogenous polymer solvent stress and average fiber stress tensors given as

$$\tau_{ij}^{s+f} = \tau_{ij}^s + \tau_{ij}^f \quad (2.49)$$

2.1.6.1.1.3 Viscoelastic fiber suspension model

A more comprehensive viscoelastic constitutive model would also include the contribution of the polymer matrix behavior to the overall suspension material behavior which is useful where elastic effects in transport phenomena becomes important during phase transformation such as extrudate swell/expansion during deposition or bead shrinkage during cooling/solidification. Viscoelastic models capture the memory effects (i.e. the cumulative effects of the polymer deformation history on the fluids internal stresses). The composite stress tensor will include contribution of the polymer matrix stress tensor in addition to either homogenous solvent stress tensor (in the absence of fibers) or to the heterogenous fiber suspension stress tensor (when fiber particles are present) i.e.

$$\begin{cases} \tau_{ij}^{s+p} = \tau_{ij}^s + \tau_{ij}^p & \text{viscoelastic solvent} \\ \tau_{ij}^{s+f+p} = \tau_{ij}^{s+f} + \tau_{ij}^p & \text{viscoelastic suspension} \end{cases} \quad (2.50)$$

The polymer stress contribution τ_{ij}^p can be model by any of the viscoelastic models usually given in differential or integral form. Perhaps the simplest of these models is the Oldroyd-B model given as

$$\lambda_r \overset{\nabla}{\tau}_{ij} + \tau_{ij}^p = 2\mu_p \Gamma_{ij} \quad (2.51)$$

where λ_r is the relaxation time, μ_p is the polymeric viscosity and $\overset{\nabla}{\tau}_{ij}$ is the upper-convected time derivative of τ_{ij}^p defined as

$$\overset{\nabla}{\tau}_{ij} = \frac{\partial}{\partial t} \tau_{ij}^p + \dot{X}_k \nabla_k \tau_{ij}^p - [\nabla_i \dot{X}_k \tau_{kj}^p + \tau_{ik}^p \nabla_j \dot{X}_k] \quad (2.52)$$

Another model is the Phan-Thien-Tanner (PTT) model given by the ODE as [129], [248]

$$\exp\left[\frac{\varepsilon_s \lambda_r}{\mu_p} \tau_{kk}^p\right] \tau_{ij}^p + \lambda_r \overset{o}{\tau}_{ij} = 2\mu_p \Gamma_{ij} \quad (2.53)$$

where ε_s is the extensibility parameter and $\overset{o}{\tau}_{ij}$ is the Gordon-Schowalter derivative of τ_{ij}^p defined as

$$\overset{o}{\tau}_{ij} = \frac{\partial}{\partial t} \tau_{ij}^p + \dot{X}_k \nabla_k \tau_{ij}^p - \nabla_j \dot{X}_i - [\nabla_k \dot{X}_i \tau_{kj}^p + \tau_{ik}^p \nabla_k \dot{X}_j] + \xi_s [\tau_{ik}^p \Gamma_{kj} + \Gamma_{ik} \tau_{kj}^p] \quad (2.54)$$

In eqn. (2.54) above, ξ_s is a slip parameter. The Giesekus model is given as [247]

$$\lambda_r \overset{\nabla}{\tau}_{ij} + c_d \tau_{ij}^p - \frac{\alpha_m \lambda_r}{\mu_p} \tau_{ik}^p \tau_{kj}^p + \frac{m_s (1 - c_d)}{2} [\tau_{ik}^p a_{kj} + a_{ik} \tau_{kj}^p] = -2\mu_p \Gamma_{ij} \quad (2.55)$$

where α_m is the mobility factor, m_s is the dimension of the space, c_d fiber orientation dependent drag coefficient. The FENE-P model is given as [247]

$$\tau_{ij}^p = -\mu_p \left(\frac{Z^p B_{ij}^p - \delta_{ij}}{\lambda_r} \right), \quad Z^p = \left(1 - \frac{B_{kk}^p}{b_p} \right)^{-1} \quad (2.56)$$

where b_p is a spring extensibility constant and B_{ij}^p is the solution to the ODE given as

$$\lambda_r \overset{\nabla}{B}_{ij} + Z^p \left\{ c_d B_{ij}^p + \frac{m_s(1-c_d)}{2} [B_{ik}^p a_{kj} + a_{ik} B_{kj}^p] \right\} = c_d \delta_{ij} + m_s(1-c_d) a_{ij} \quad (2.57)$$

Likewise, the FENE-CR model is given as

$$\tau_{ij}^p = -\mu_p \frac{Z^p B_{ij}^p}{\lambda_r} \quad (2.58)$$

where B_{ij}^p is the solution to a slight modification of ODE in eqn. (2.58) above by multiplying the

RHS by Z^p . Lastly, the K-BKZ time-integral model [129] is given as

$$\begin{aligned} \tau_{ij}^p = \frac{1}{1-y_p} \int_{-\infty}^t \sum_{k=1}^{N_k} \frac{a_k}{\lambda_k} \exp \left[-\frac{t-t'}{\lambda_k} \right] & \left[\frac{\alpha_p}{(\alpha_p - 3) + \beta_p I_{C^{-1}} + (1 - \beta_p) I_C} \right] [C_t^{-1}(t') \\ & + y_p C_t(t')] dt' \end{aligned} \quad (2.59)$$

where a_k is the relaxation modulus and λ_k is the relaxation time for mode k , N_k is the number of relaxation modes, α_p and β_p are nonlinear material constants, y_p is a normal stress difference control factor, I_C and $I_{C^{-1}}$ are first invariants of the Cauchy-Green strain tensor C_t and its inverse C_t^{-1} (also known as the Finger strain tensor) and t is the current time.

Review literature on various approaches in modelling other thermo-physical fluid parameters used in developing the transport equations such as temperature and pressure dependent density, temperature-dependent enthalpy, thermal conductivity and specific heat capacity, etc. including various models for the thermal radiation intensity can be found in [129].

1.1.1.1.3. Particle Motion/Migration

The fundamental equations governing the motion of particles suspended in the viscous polymer suspension are the Newton's second law for translational motion and the Euler's equation for rotational motion [179] given respectively in equations below

$$m^j \frac{d\dot{\underline{X}}^j}{dt} = \underline{F}_{\text{ext}}^j + \int_{S_p^j(t)} \underline{\sigma} \cdot \underline{\hat{n}} dS \quad (2.60)$$

$$\frac{d}{dt} \{ \underline{I}^j \underline{\dot{\Theta}}^j \} = \underline{Q}_{\text{ext}}^j + \int_{S_p^j(t)} \underline{\Delta}^j \times (\underline{\sigma} \cdot \underline{\hat{n}}) dS \quad (2.61)$$

where m^j , and $\underline{I}^j$ are the mass and moment of inertia of the j^{th} particle, $\dot{\underline{X}}^j$ and $\dot{\underline{\Theta}}^j$ are the translational and rotational velocities of the j^{th} particle, $\underline{\Delta}^j$ is the position vector of a point on the j^{th} particle's boundary reckoned from the particle's centroid, $\underline{F}_{\text{ext}}^j$ and $\underline{Q}_{\text{ext}}^j$ are the external force and couple acting on the j^{th} particle, $\underline{\hat{n}}$ is the outwardly directed unit normal vector on S_p^j and dS is the local surface area. Because it is impractical to simulate all the particles in a system during processing, particle migration phenomena in fiber suspension is often modelled using the diffusive flux model (DFM) defined in terms of the fiber concentration ϑ_f by the constitutive equation given as [249], [250]

$$\frac{\partial \vartheta_f}{\partial t} + \dot{\underline{X}} \cdot \underline{\nabla} \vartheta_f = -\underline{\nabla} \cdot (\underline{N}_c + \underline{N}_\mu + \underline{N}_b) \quad (2.62)$$

where, $\underline{N}_c$ is the flux due to interparticle hydrodynamic interaction, $\underline{N}_\mu$ is the contribution due to spatial variations in viscosity and $\underline{N}_b$ is the contribution due to Brownian diffusion of particles.

The flux terms $\underline{N}_c$, $\underline{N}_\mu$ and $\underline{N}_b$ are respectively given as

$$\underline{N}_c = -K_c l_f^2 (\vartheta_f^2 \underline{\nabla} \dot{\gamma} + \vartheta_f \dot{\gamma} \underline{\nabla} \vartheta_f), \quad \underline{N}_\mu = -K_\mu \dot{\gamma} \vartheta_f^2 \left(\frac{l_f^2}{\mu} \right) \frac{d\mu}{d\vartheta_f} \underline{\nabla} \vartheta_f, \quad \underline{N}_b = -D_b \underline{\nabla} \vartheta_f \quad (2.63)$$

where K_c and K_μ are proportionality and rate constants respectively of unity order, l_f is a characteristic particle dimension and D_b is the Brownian diffusivity. Although particle motion is a microscale level phenomenon, the overall dynamic behavior of suspended particles is often predicted on a global level using a macroscale dynamic model such as the macroscopic orientation tensor models given in details in [20], [251] and in Chapter 7 of this dissertation.

2.1.6.2 Boundary Conditions

Boundary conditions may be defined at the interface of adjoining phases such as at liquid/solid interface, liquid/liquid interface, liquid/vapor interface or at free surfaces. They are also specified at regions of domain continuity such as a control volume's inlet and outlet. There are generally two types of boundary conditions in continuum mechanics, namely (a) Essential or Dirichlet boundary condition where velocity or temperature field is imposed and (b) Free or Neumann boundary condition where traction/stress field or external heat flux is specified. In heat transfer analysis, a third boundary condition known as the Newton boundary condition that specifies convective heat transfer at the interface of two phases may also be prescribed. At the liquid/solid interface, the fluid is assumed to come to rest or move with the solid wall, a condition known as the 'no-slip' condition. For impermeable surface such as the liquefier wall, no mass flux through the normal surface can be assumed. Typical boundary conditions used in an axisymmetric polymer melt flow macro-model process simulation are shown in Figure 2.13 below.

Similarly, typical boundary conditions used in a microscale level process simulation such as in a single fiber motion and deformation FSI analysis. Example kinematic and stress-based boundary conditions for the fluid mechanics analysis appear in the schematic in Figure 2.14a while typical force and displacement constraints for the solid mechanics analysis appear in Figure 2.14b.

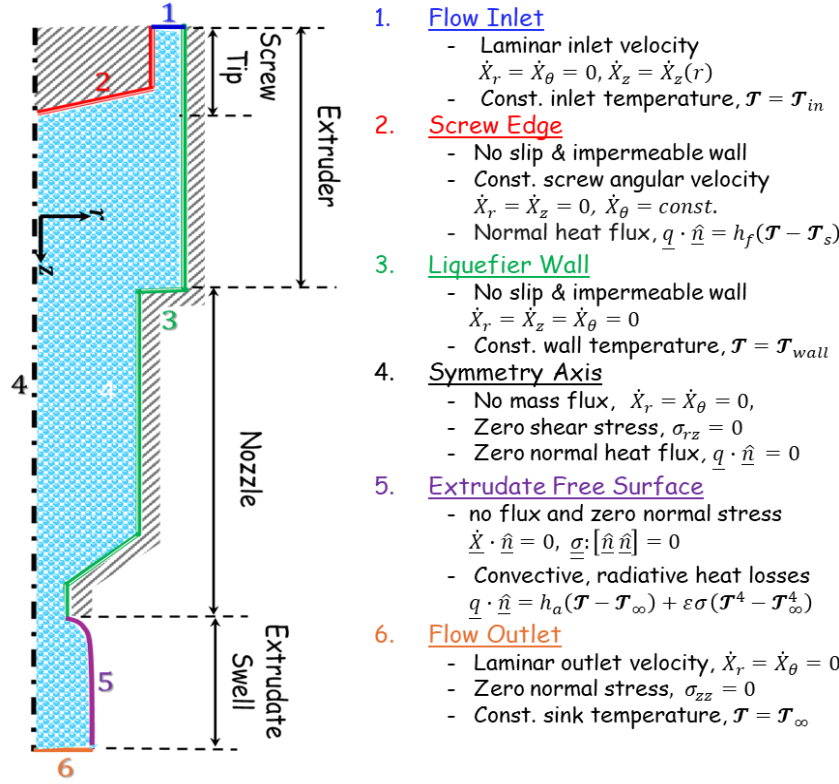


Figure 2.13: Typical boundary conditions prescribed in EDAM polymer melt flow macro-scale process simulation.

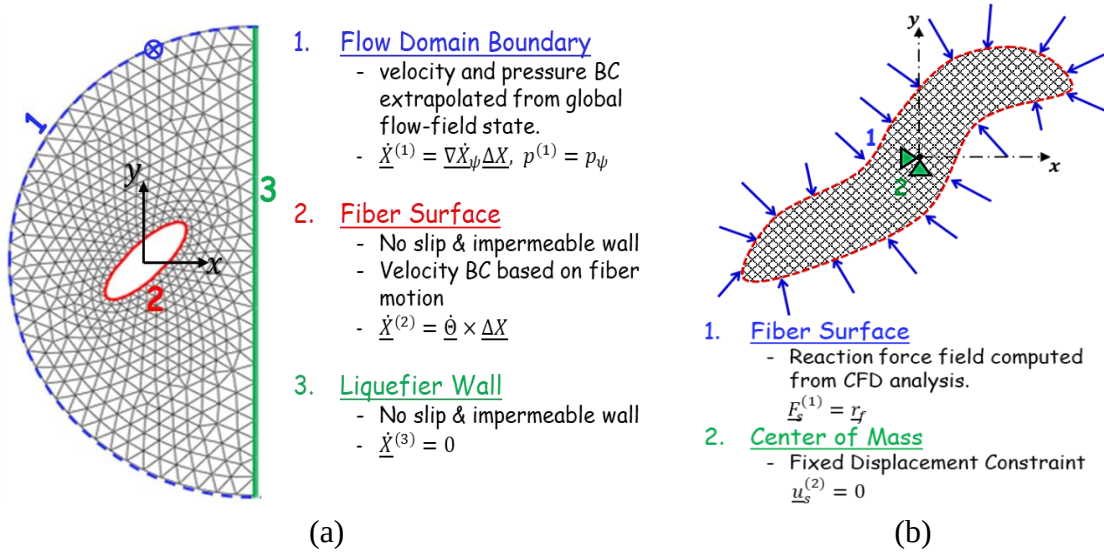


Figure 2.14: Typical boundary conditions prescribed in single fiber micro-scale coupled FSI process simulation for (a) fluid mechanics analysis (b) solid mechanics analysis.

Evidently, the scale and type of analysis, level of sophistication and model assumptions adopted dictate the boundary conditions to be applied to the system.

2.1.6.3 Macroscale modelling aspects of EDAM SFRP Process

Macro-Scale process simulation is often used to predict polymer melt flow-fields (including temperature, pressure and velocity fields), determine flowrate and power requirements, predict polymer melt flow behaviour such as swirling flow behaviour at screw flight to understand shear rate variability within the nozzle or solidification behaviour during deposition including viscoelastic effects. They are also used for printing process parameter optimization, printing path planning, real-time process monitoring and control, printing head design, nozzle design, operating limits setting, and performance optimization [208]. Macro-scale models could either be analytical based or numerical based. Analytical based solutions are relatively simpler than numerical solutions due to numerous assumptions considered in their development. They are also time and computationally more efficient than numerical based models. However, oversimplification and idealization in their development makes them inherently less accurate than numerical solutions due to approximations, they are usually non-flexible and often used for specific quantity prediction [208]. Commercial AM software packages are continuously being updated and improved for extended capabilities such as Dassault-Systemes ©, Digimat ©, Dieplast ©, EFD Lab, ANSYS©, STARCCM+, etc. [1] The various macro scale modelling aspects are discussed briefly in subsequent sections.

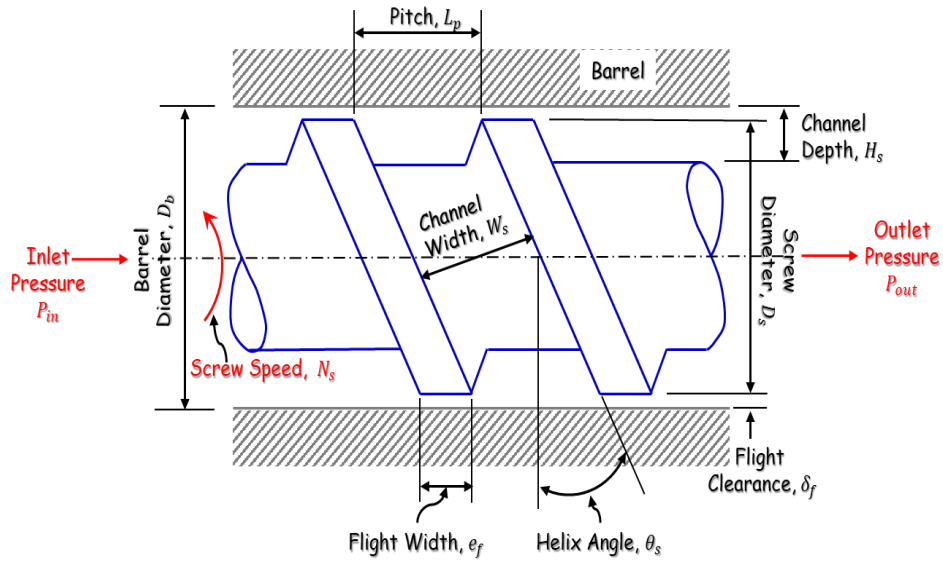
1.1.1.1.4. Flow modelling near the Extruder-Screw Zone

The transport of polymer composite material through the barrel is made possible by the turning action of the screw. A typical annotated schematic of a single flight extruder - screw section geometry is shown in Figure 2.15a. The pitch angle θ_s and channel width W_s calculated at the barrel wall are respectively given as

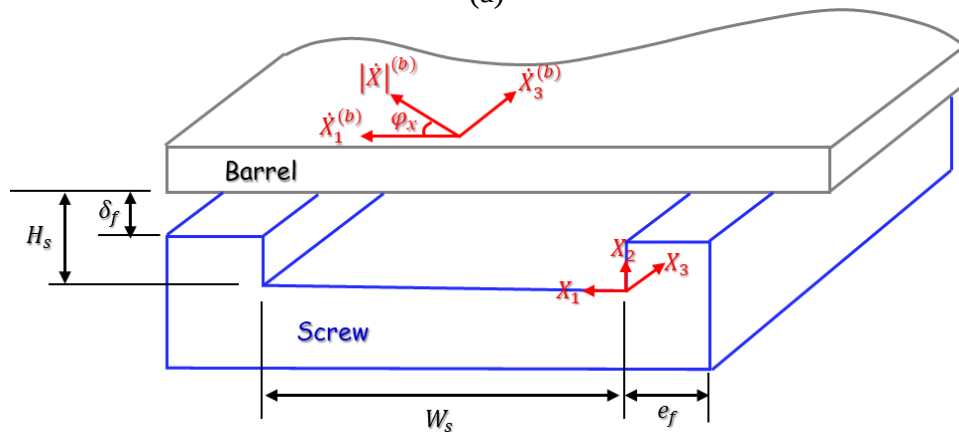
$$\tan \theta_s = \theta_s / \pi D_b, \quad W_s = L_p \cos \theta_s - e_f \quad (2.64)$$

To avoid analytical complications associated with spiral reference frame, flow calculations are often calculated using a coordinate reference frame that hypothetically assumes a straight channel flow obtained by unwinding the screws channel. The unwound channel length, L_c can be obtained in terms of the screws length L_s as $L_c = L_s / \sin \theta_s$. The reference is fixed at the screw, and the barrel is allowed to rotate relative to the fixed screws shown in Figure 2.15b. The relative velocities of the barrel w.r.t. the screw in terms of the screw's angular velocity N_s is given as [130], [252]

$$\dot{X}_1^b = \pi D_b N_s \sin \theta_s, \quad \dot{X}_3^b = \pi D_b N_s \cos \theta_s \quad (2.65)$$



(a)



(b)

Figure 2.15: Detailed annotated schematic of (a) a typical extruder-screw geometry section (b) an unwound screw channel.

The model is simplified assuming Newtonian, isothermal, inertia-less, low Reynolds number flow condition, and a fully developed flow along the channel length with no gravity effects. As such, the momentum conservation equations reduce to

$$-\frac{\partial p}{\partial X_1} + \mu \left[\frac{\partial^2 \dot{X}_1}{\partial X_1^2} + \frac{\partial^2 \dot{X}_1}{\partial X_2^2} \right]^T = 0, \quad -\frac{\partial p}{\partial X_3} + \mu \left[\frac{\partial^2 \dot{X}_3}{\partial X_1^2} + \frac{\partial^2 \dot{X}_3}{\partial X_2^2} \right]^T = 0 \quad (2.66)$$

The boundary conditions consider no slip at the channel walls, i.e. $\dot{X}_3 = 0$ at $X_1 = 0, W_s$ & $X_2 = 0, H_s$. The exact solution to the down-channel velocity distribution, $\dot{X}_3$ is given by an infinite Fourier series given by

$$\dot{X}_3(X_1, X_2) = \dot{X}_3^d - \dot{X}_3^p \quad (2.67)$$

where $\dot{X}_3^d$ and $\dot{X}_3^p$ are the velocity profiles due to the drag and pressure and are respectively given as [253]

$$\dot{X}_3^d = \dot{X}_3^b \frac{4}{\pi} \sum_{n=1,3,5}^{\infty} \frac{1}{n} \frac{\sinh\left(\frac{n\pi X_2}{W_s}\right)}{\sinh\left(\frac{n\pi H_s}{W_s}\right)} \sin\left(\frac{n\pi X_1}{W_s}\right)$$

$$\dot{X}_3^p = \dot{X}^m \left[\frac{X_2^2}{H_s} - \frac{X_2}{H_s} + \frac{8}{\pi^3} \sum_{n=1,3,5}^{\infty} \frac{1}{n^3} \frac{\cosh\left(\frac{n\pi W_s}{H_s} \left(\frac{X_1}{W_s} - \frac{1}{2}\right)\right)}{\cosh\left(\frac{n\pi W_s}{2H_s}\right)} \sin\left(\frac{n\pi X_2}{H_s}\right) \right]$$

where $\dot{X}^m = -\frac{H_s^2}{2\mu} \left(\frac{\partial p}{\partial X_3} \right)$, The flowrate $\dot{\vartheta}_s$ can be obtained by integrating the velocity over

the free cross-sectional area given as [130], [252]

$$\dot{\vartheta}_s = \dot{\vartheta}_s^d - \dot{\vartheta}_s^p = j \frac{W_s}{2} H_s \left[f_d \dot{X}_3^b + \frac{f_p}{3} \dot{X}^m \right] \quad (2.68)$$

where, f_d and f_p are drag and pressure shape factors respectively given as

$$f_d = \frac{16 W_s}{\pi^3 H_s} \sum_{n=1,3,5}^{\infty} \frac{1}{n^3} \tanh\left(\frac{n\pi H_s}{2W_s}\right), \quad f_p = 1 - \frac{192 H_s}{\pi^5 W_s} \sum_{n=1,3,5}^{\infty} \frac{1}{n^5} \tanh\left(\frac{n\pi W_s}{2W_s}\right) \quad (2.69)$$

Likewise, the power required to drive the screw $\dot{e}_s$ is given as

$$\dot{e}_s = \left[\frac{4\mu}{H_s} (\dot{X}_1^b)^2 \tan^2 \theta_s + \frac{\mu}{H_s} (\dot{X}_3^b)^2 + \frac{H_s}{2} \frac{\partial p}{\partial X_3} \dot{X}_3^b \right] W_s L_c \quad (2.70)$$

If the channel width is large compared to the channel depth, i.e. $H_s/W_s \ll 1$, then it is safe to assume $\partial^2 \dot{X}_3 / \partial X_1^2 \ll \partial^2 \dot{X}_3 / \partial X_2^2$ and $\partial \dot{X}_2 / \partial X_3 \ll \partial \dot{X}_3 / \partial X_2$ within the channel away from the channel edges, and assuming $f_d = f_p = 1$. With these assumptions, one can approximate for the transverse and down-channel velocity components $\dot{X}_1, \dot{X}_3$ to obtain the following

$$\dot{X}_1 = \dot{X}_1^b \frac{X_2}{H_s} \left[2 - 3 \frac{X_2}{H_s} \right], \quad \dot{X}_3 = \dot{X}_3^b \frac{X_2}{H_s} + \dot{X}^m \frac{X_2}{H_s} \left[1 - \frac{X_2}{H_s} \right] \quad (2.71)$$

and likewise, the flowrate $\dot{\vartheta}_s$ reduces to

$$\dot{\vartheta}_s = \frac{W_s}{2} H_s \left[\dot{X}_3^b + \frac{1}{3} \dot{X}^m \right] \quad (2.72)$$

For variable channel height $H_s = H_s(X_3)$, the flow rate can be derived as

$$\dot{\vartheta}_s = \frac{W_s}{2} \bar{H}_1 \left[\dot{X}_3^b + \frac{1}{3} \bar{X}^m \right], \quad \bar{X}^m = - \frac{\bar{H}_2^2}{2\mu} \frac{\partial p}{\partial X_3} \quad (2.73)$$

where $\bar{H}_1$ and $\bar{H}_2$ are harmonic and geometric mean values of the variable channel height minimum, H_s^- and maximum, H_s^+ values given as

$$\bar{H}_1 = 2 \left[\frac{1}{H_s^+} + \frac{1}{H_s^-} \right]^{-1}, \quad \bar{H}_2 = \sqrt{H_s^+ H_s^-} \quad (2.74)$$

Given that the pressure gradient in the X_3 direction is not a function of X_1 or X_2 and considering the flow is fully developed flow in the X_3 direction, then $\partial p / \partial X_3$ is a constant and can be given as

$$\partial p / \partial X_3 = \Delta P / L_c \text{ where } \Delta P \text{ is the pressure drop given as } \Delta P = P_{out} - \Delta P_{in}.$$

1.1.1.1.5. *Fiber Suspension modelling within the Nozzle*

The polymer composite melt flow-field pressure and velocity distribution within the EDAM nozzle can be analytically approximated or numerically determined depending on the level of sophistication and degree of accuracy desired. The melt flow-field is used to compute the orientation distribution of the suspended particles which in turn influences the fluid rheology and flow-field distribution hence necessitating a back-coupling algorithm. For simplification, most studies assume a steady state, viscous, incompressible fluid and low Reynolds number (creeping/Stokes) flow with negligible inertia and adopt a one-way flow-fiber orientation tensor weak FSI coupling approach. The subsequent sections discuss briefly previous efforts made to approximate the flow-field and fiber orientation within the nozzle.

Fiber orientation modelling

The orientation state of any fiber can be described by a probability distribution function (PDF) $\psi(\underline{\rho})$ of all the possible directions of $\underline{\rho}$ where $\underline{\rho}$ is the unit vector associated with the fiber given as [17]

$$\underline{\rho} = \begin{bmatrix} \sin \theta \cos \phi \\ \sin \theta \sin \phi \\ \cos \theta \end{bmatrix} \quad (2.75)$$

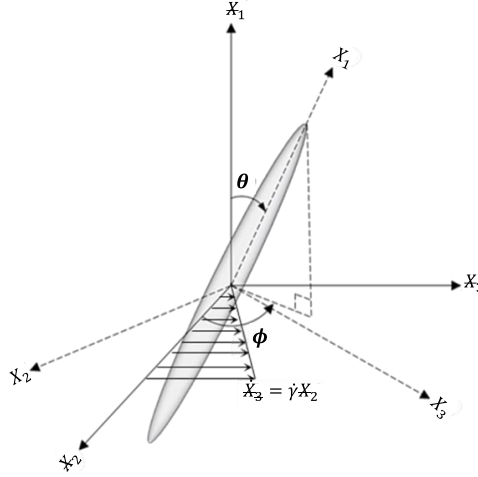


Figure 2.16: Single ‘rigid’ ellipsoidal fiber orientation

$\psi(\underline{\rho})$ is periodic i.e., $\psi(\underline{\rho}) = \psi(-\underline{\rho})$ and satisfies the normalization condition.

$$\oint \psi(\underline{\rho}) d\underline{\rho} = \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \psi(\theta, \phi) \sin \theta d\theta d\phi = 1 \quad (2.76)$$

The PDF satisfies the continuity condition [17]

$$\frac{D\psi}{Dt} = -\frac{\partial}{\partial \underline{\rho}}(\psi \underline{\dot{\rho}}) \quad (2.77)$$

Analytical modelling of the orientation of particles in suspension usually depends on a host of factors ranging from the adjacent flow-field, the particle geometry, the fluid’s material rheology, the force-field surrounding the particle, and the particle’s material behavior, etc. For simplification, only a few of the factors are usually accounted for in the mathematical model. The earliest analytical model that formed the basis for fiber orientation modelling in dilute suspension was developed by Jeffery in 1922 [19]. Jeffery’s model was based on the motion of a single rigid ellipsoidal particle suspended in incompressible, Newtonian viscous homogenous flow. Jeffery assumed that the particle moves in response to the surrounding fluid motion and based his model on the assumption of a very small or a very slow-moving particle. The equation describing

Jeffery's motion is provided in detail in Chapter 6. Jeffery's equation defining the orientation evolution of a single rigid axisymmetric particle is usually given in vector form as [19], [20], [254].

$$\dot{\rho}_i^{JF} = \Xi_{ij}\rho_j + \kappa(\Gamma_{ij}\rho_j - \Gamma_{kl}\rho_k\rho_l\rho_i) \quad (2.78)$$

where, Ξ_{ij} and Γ_{ij} are the anti-symmetric and symmetric decomposition of the deformation rate tensor $L_{ij} = \partial\dot{X}_i/\partial X_j$ and can be given respectively as

$$\Xi_{ij} = \frac{1}{2}(L_{ij} - L_{ji}), \quad \Gamma_{ij} = \frac{1}{2}(L_{ij} + L_{ji}) \quad (2.79)$$

Such that $L_{ij} = \Gamma_{ij} + \Xi_{ij}$, κ is a particle shape parameter given as $\kappa = (r_e^2 - 1)/(r_e^2 + 1)$, r_e is the geometric aspect ratio of the particle. Numerous enhancements have been made to Jeffery's single fiber model to more accurately represent the bulk behavior of fibers in semi-dilute and concentrated suspensions. While it is theoretically possible, simulating the behavior of each individual particle in fiber suspension flow is computationally costly and impractical. Batchelor's utilized the '*Slender Body Theory*' to determine the bulk stress for Newtonian particle suspension based on average contribution of individual arbitrary shaped particles and developed general constitutive equations for the particle suspension using distribution of *Stokelets* to represent each particle [255]. In a series of publication, Hinch & Leal [196], [197] extended the '*Slender Body Theory*' to develop constitutive equations for dilute particle suspension with deformable particle considering the effect of Brownian Motion [200] and studied the effect of small deviations from axisymmetric geometry on particle motion in homogenous flows [193]. Dinh & Armstrong [244], extended the "*cell model*" approach previously used by Batchelor's in determining extensional viscosity of concentrated slender particle suspension to develop general constitutive relations for semi-concentrated suspension of rigid fibers in suspended in Newtonian fluid. To account for the effect of rotary diffusion due to hydrodynamic interactions for concentrated fiber suspension, Folgar-Tucker model [256], [257] incorporated an isotropic rotary diffusion term having a linear

dependence on the scalar magnitude of the rate of deformation tensor and based on an orientation probability distribution function (ODF) in addition to the hydrodynamic contribution from Jeffery's model, given thus.

$$\dot{\rho}_i^{FT} = \dot{\rho}_i^{JF} - D_r \frac{1}{\psi} \frac{\partial \psi}{\partial \rho_i} \quad (2.80)$$

where D_r is the rotary diffusivity term and a constant value account for the Brownian effect of very fine particles. The PDF ψ defines the probability of a given fiber in a particular orientation state and the rate of change of ψ is given by the Fokker-Planck's continuity equation describing its time evolution.

$$\frac{d\psi}{dt} = - \frac{\partial}{\partial \rho_i} (\psi \dot{\rho}_i) \quad (2.81)$$

The PDF form of Folger-Tuckers model presented itself as a complicated and computationally intensive problem which made it difficult to use. Conventionally a numerical method such as finite volume [258] and more recently a computationally efficient exact spherical harmonics method [259] has been used to solve the Folgar-Tuckers (FT) equation of change for fiber orientation, however, the widely utilized method was developed by Advani and Tucker [17] who presented a simplified moment-tensor form to Folger-Tuckers model by defining a set of even order orientation tensors as integral products of the orientation vector $\underline{\rho}$ with the PDF ψ over the surface of a unit sphere. For the 2nd and 4th order tensor, these is respectively given as

$$a_{ij} = \oint \rho_i \rho_j \psi(\underline{\rho}) d\underline{\rho}, \quad a_{ijkl} = \oint \rho_i \rho_j \rho_k \rho_l \psi(\underline{\rho}) d\underline{\rho} \quad (2.82)$$

The tensors defined in this form are completely symmetric i.e.

$$a_{ij} = a_{ji}$$

$$a_{ijkl} = a_{jikl} = a_{kijl} = a_{likj} = a_{ikjl} = a_{iljk} = \dots, \quad 24 \text{ permutations}$$

and based on the normalization condition of eqn. (2.76), the following tensor properties were obtained.

$$a_{ii} = 1, \quad a_{ijkk} = a_{ij}$$

Consequently, there are only 5 independent components of the 9 components of the 2nd order tensor and 9 independent components of the 81 components of the 4th order tensor. The rest can be derived based on the above tensor properties. With this definition, Advani and Tucker developed an equation of change for the 2nd order orientation tensors in terms of the 2nd and 4th order tensors thus.

$$\frac{da_{ij}}{dt} = \dot{a}_{ij}^{FT} = \{\dot{a}_{ij}^{HD} + \dot{a}_{ij}^{IRD}\} \quad (2.83)$$

$\dot{a}_{ij}^{HD}$ is the hydrodynamic tensor component of the Folger-Tuckers that represents Jeffery's equation and given as

$$\dot{a}_{ij}^{HD} = -(\Xi_{ik}a_{kj} - a_{ik}\Xi_{kj}) + \kappa(\Gamma_{ik}a_{kj} + a_{ik}\Gamma_{kj} - 2\Gamma_{kl}a_{ijkl}) \quad (2.84)$$

And $\dot{a}_{mn}^{IRD}$ is the isotropic rotary diffusion term modelling fiber interaction and is given as

$$\dot{a}_{ij}^{IRD} = 2D_r(\delta_{ij} - \alpha a_{ij}) \quad (2.85)$$

α is a dimension factor, $\alpha = 3$ for 3D orientation and $\alpha = 2$ for 2D planar orientation. For slender long particles $\kappa \approx 1$, Folgar and Tucker suggested a relation for D_r i.e., $D_r = C_I \dot{\gamma}$, where C_I is a phenomenological interaction coefficient and $\dot{\gamma}$ is the scalar magnitude of the strain rate tensor Γ_{ij} given as $\dot{\gamma} = \sqrt{2\Gamma_{ij}\Gamma_{ji}}$. Folgar and Tucker [256] suggested that C_I depends on the fiber volume fraction v_f and aspect ratio r_e and Bay (1991) proposed

$$C_I = 0.0184e^{-0.7148\vartheta_f r_e} \quad (2.86)$$

where r_e is the fiber aspect ratio and ϑ_f is the fiber volume fraction. Phan-Thien et al. [260] developed a general correlation of C_I for wider range fiber volume fraction ϑ_f given as

$$C_I = 0.03(1 - e^{-0.224\vartheta_f r_e}) \quad (2.87)$$

A similar equation of change can be formulated for the 4th-order tensor using both 4th and 6th-order tensors and can be extended to even higher orders. Therefore, a closure approximation is necessary to achieve a closed set of equations. Various closure approximations for the 4th-order tensor and their derivatives are explored and discussed in Chapter 7. Due to the experimentally observed differences in fiber orientation kinetics based on the Advani-Tucker's equation compared to those predicted by traditional orientation models, various model corrections have been proposed to slow down the orientation kinetics which are discussed in detail in Chapter 7.

1.1.1.1.5.1. Analytical based flow-field solutions

Various researchers developed analytical estimates of the flow kinematics, and pressure drop within a nozzle contraction. For instance, Lubanzky et al. [261] developed analytical equations for the flow of fluid with high Trouton ratio through an abrupt nozzle axisymmetric contraction which typifies the flow of dilute Newtonian polymer through a nozzle. From the continuity equation, the axial and radial velocity field for the fully developed flow condition in a typical extrusion nozzle are given as follows [261].

$$\dot{X}_z = 2\bar{u} \left[1 - \left(\frac{X_r}{\mathbb{R}} \right)^2 \right], \quad \dot{X}_r = X_r \dot{X}_z \frac{\mathbb{R}'}{\mathbb{R}}, \quad \bar{u} = \frac{\mathbb{Q}}{\pi \mathbb{R}^2} \quad (2.88)$$

where $\bar{u}$ is the average axial velocity, $\dot{X}_z$ and $\dot{X}_r$ are the axial and radial velocities, $\mathbb{R}(X_z)$ is the nozzle radius at axial distance X_z reckoned from the nozzle exit, and $\mathbb{Q}$ is the volume flow rate.

The velocity gradient based on eqn. (2.88) above is thus obtained as

$$\begin{aligned} L_{11} = L_{rr} &= 2\bar{u} \frac{\mathbb{R}'}{\mathbb{R}} \left[1 - 3 \left(\frac{X_r}{\mathbb{R}} \right)^2 \right], \quad L_{33} = L_{zz} = 4\bar{u} \frac{\mathbb{R}'}{\mathbb{R}} \left[-1 + 2 \left(\frac{X_r}{\mathbb{R}} \right)^2 \right] \\ L_{22} = L_{\theta\theta} &= \dot{X}_z \frac{\mathbb{R}'}{\mathbb{R}}, \quad L_{13} = \frac{1}{\mathbb{R}} \frac{X_r}{\mathbb{R}} \left[-2\bar{u} \left(1 - \mathbb{R}'^2 \left(\frac{X_r}{\mathbb{R}} \right)^2 \right) + \frac{\dot{X}_z}{2} (\mathbb{R}\mathbb{R}'' - 3\mathbb{R}'^2) \right] \end{aligned} \quad (2.89)$$

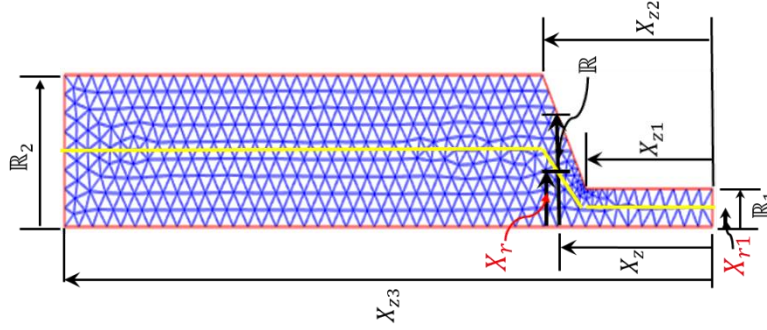


Figure 2.17: Annotated schematic of a typical axisymmetric nozzle contraction geometry in radial coordinates.

The equation of any streamline $\psi(X_r, X_z)$ in the axisymmetric flow domain is given as

$$\frac{X_r}{R} = \frac{X_{r1}}{R_1} \quad (2.90)$$

In the contraction zone, transition from uniaxial extension to biaxial extensional gradient occurs at $X_r = R/\sqrt{2}$ [261]. Considering the geometry of the nozzle contraction given in Figure 2.17, the nozzle internal radius R can be mathematically represented as a function of axial distance X_z according to

$$R = \begin{cases} R_1, & X_z \leq X_{z1} \\ k_1 + k_2 X_z, & X_{z1} < X_z < X_{z2} \\ R_2, & X_z \geq X_{z2} \end{cases}, \quad R' = \begin{cases} 0, & X_z \leq X_{z1} \\ k_2, & X_{z1} < X_z < X_{z2} \\ 0, & X_z \geq X_{z2} \end{cases} \quad (2.91)$$

where $k_1 = \left[\frac{X_{z2}R_1 - X_{z1}R_2}{X_{z2} - X_{z1}} \right]$, $k_2 = \left[\frac{R_2 - R_1}{X_{z2} - X_{z1}} \right]$. From the solution of the momentum equation, the pressure distribution across the nozzle contraction can be obtained as:

$$p(X_z) = \begin{cases} p_0 - \frac{8\mu\bar{u}}{R_1^2} X_z, & X_z \leq X_{z1} \\ p(X_{z1}) - \frac{8\mu\bar{u}}{R} \left[\left(\frac{R}{R_1} \right)^3 - 1 \right], & X_{z1} < X_z < X_{z2} \\ p(X_{z2}) - \frac{8\mu\bar{u}}{R_2^2} [X_z - X_{z2}], & X_z \geq X_{z2} \end{cases} \quad (2.92)$$

Numerous other works listed in literature that develop arithmetic solutions for creeping flow through axisymmetric sections of arbitrary geometry such as the work of Sisavath et al. [262]

can be extended to approximate solutions of the flow-field and pressure drop within a nozzle. The computed analytical flow-field can be used to determine the distribution of the fiber orientation within the nozzle using of the analytical models discussed above and in Chapter 7. Most analytical solutions are based on simple linear Newtonian creeping homogenous fluid flow. However, it becomes almost impossible to develop analytical solutions for complex non-linear heterogenous particle suspension flows, whereby numerical methods become attractive.

1.1.1.1.5.2. Numerical based flow-field solutions

Discretization approaches such as the particle-based methods (PBM) and the element-based methods (EBM) can be used to solve the governing equations and compute the flow-field of the polymer melt flow through the EDAM nozzle such as the EBM based FEM method (e.g. [21], [22], [132], [234]) or PBM based SPH or DEM method (e.g. [24], [202], [203]). The model development of these methods amongst other discretization methods is briefly discussed in the following subsections.

1. EDM-FEM Simulation Algorithm

As earlier stated, the FEM method discretizes a complex PDE domain into subdomain units to form a system of algebraic equations with solutions computed at the unit nodes or elements level and assembled to yield an approximate general solution. The process usually begins by simplification of the strong form governing equations based on valid assumptions and transformation of the simplified equations into weak integral forms. We consider the conservation equations defining the fluid flow through the 2D axisymmetric nozzle section (cf. Figure 2.17) in cylindrical coordinates. Under the assumption of steady state, viscous, low Reynolds number (creeping/Stokes), incompressible axisymmetric fluid flow, such that the time derivatives are zero

and spatial velocity and its derivatives in the θ component direction are zero, the fluid density is constant, and the inertia term is negligible. With these assumptions and in the absence of temperature dependent fields, the conservation equation for mass is reduced to

$$\frac{1}{X_r} \frac{\partial}{\partial X_r} (X_r \dot{X}_r) + \frac{\partial \dot{X}_z}{\partial X_z} = 0 \quad (2.93)$$

The momentum equations are given as

$$\begin{aligned} \frac{1}{X_r} \frac{\partial}{\partial X_r} (X_r \sigma_{rr}) + \frac{\partial \sigma_{zr}}{\partial X_z} - \frac{\sigma_{\theta\theta}}{X_r} + \rho f_r &= 0 \\ \frac{1}{X_r} \frac{\partial}{\partial X_r} (X_r \sigma_{rz}) + \frac{\partial \sigma_{zz}}{\partial X_z} + \rho f_z &= 0 \end{aligned} \quad (2.94)$$

where, f_r and f_z are the body forces in the X_r and X_z directions and $\underline{\underline{\sigma}} = \begin{bmatrix} \sigma_{rr} & - & \sigma_{zr} \\ - & \sigma_{\theta\theta} & - \\ \sigma_{rz} & - & \sigma_{zz} \end{bmatrix}$ is the

Cauchy stress tensor which can be given in terms of the deviatoric stresses as $\sigma_{ij} = \tau_{ij} - p\delta_{ij}$.

Wang et al. [263] assumed Tucker's model [264] for short rigid fiber suspension constitutive relation given as

$$\tau_{ij} = 2C_{\mu_{ijkl}} \Gamma_{kl} \quad (2.95)$$

where the deformation rate tensor is given as $\Gamma_{ij} = L_{ij} + L_{ji}$, and the velocity gradient tensor is given as

$$\underline{\underline{L}} = \begin{bmatrix} \partial \dot{X}_r / \partial X_r & - & \partial \dot{X}_r / \partial X_z \\ - & \dot{X}_r / X_r & - \\ \partial \dot{X}_z / \partial X_r & - & \partial \dot{X}_z / \partial X_z \end{bmatrix} \quad (2.96)$$

The 4th order anisotropic viscosity tensor is given as $C_{\mu_{ijkl}} = \mu(\delta_{ijkl} + N_p a_{ijkl})$, and the particle number N_p given in terms of shape factors f_f , g_f and fiber volume fraction ϑ_f as

$$N_p = \frac{f_f \vartheta_f}{(1 + g_f \vartheta_f)}, \quad f_f = \frac{r_e^2}{3 \log \sqrt{\pi/\vartheta_f}} \quad (2.97)$$

The above strong form governing transport equations is transformed to weak form integral equations considering weighting functions $\underline{\omega}_p$ & $\underline{\omega}_v$ for the continuity and momentum equations respectively and making necessary substitutions to derive

$$\begin{aligned} \int_{\vartheta^e} \underline{\omega}_p (\underline{\nabla} \cdot \underline{v}) d\vartheta &= 0 \\ \underline{\Sigma}^e = \int_{\vartheta^e} (\underline{\nabla}_s \underline{\omega}_v)^T \underline{\tilde{C}}_\mu \underline{\nabla}_s \underline{v} d\vartheta - \int_{\vartheta^e} \rho \underline{\omega}_v^T \underline{f} d\vartheta - \int_{S_\tau^e} \underline{\omega}_v^T \underline{\bar{t}} dS - \int_{\vartheta^e} p (\underline{\nabla}^T \underline{\omega}_v) d\vartheta &= 0 \end{aligned} \quad (2.98)$$

Among the various FEM solution techniques [265], [266], [267] the penalty method assumes for the pressure, the given form $p = -\gamma_e (\underline{\nabla} \cdot \underline{v})$ where γ_e is a penalty parameter. In eqn. (2.98) above, ϑ is the computational flow domain, S is the boundary surface where velocity and traction boundary conditions are imposed, $\underline{f} = [f_r \quad f_z]^T$ is the body force vector, $\underline{\bar{t}} = [t_r \quad t_z]^T$ is the prescribed traction on S_τ^e , $\underline{v} = [\dot{X}_r \quad \dot{X}_z]^T$ is the velocity vector and the strain displacement matrix, $\underline{\nabla}_s$ and gradients operator $\underline{\nabla}$ are respectively given as

$$\underline{\underline{\nabla}}_s = \begin{bmatrix} \frac{\partial}{\partial X_r} & 0 & \frac{\partial}{\partial X_z} & \frac{1}{X_r} \\ 0 & \frac{\partial}{\partial X_z} & \frac{\partial}{\partial X_r} & 0 \end{bmatrix}^T, \quad \underline{\nabla} = \left[\frac{1}{X_r} \frac{\partial}{\partial X_r} X_r \quad \frac{\partial}{\partial X_z} \right]^T \quad (2.99)$$

$\underline{\tilde{C}}_\mu$ is a 4 x 4 matrix of the 4th order anisotropic viscosity tensor in reduced form given as $\tilde{C}_{\mu ij} = \mu \tilde{C}_{o ij}$, where $\tilde{C}_{o ij} = (\overset{o}{C}_{ij} + 2N_p \overset{o}{A}_{ij})$, $\overset{o}{C}_{ij} = \delta_{ij} [2 - .25(1 - (-1)^i)(i - 1)]$ and $\overset{o}{A}_{ij}$ is given as a function of the components of the 6 x 6 4th order orientation tensor in contracted notation A_{ij} [263]

$$\underline{\underline{A}}^o = \begin{bmatrix} A_{11} & A_{13} & A_{15} & A_{12} \\ A_{31} & A_{33} & A_{35} & A_{32} \\ A_{51} & A_{53} & A_{55} & A_{52} \\ A_{21} & A_{23} & A_{45} & A_{22} \end{bmatrix}^T \quad (2.100)$$

The FEA Galerkin formulations of the weak form momentum equation (cf. eqn. (2.98)), after substituting the penalty-based pressure expression is obtained as

$$\underline{\underline{\Sigma}}^e = \underline{\underline{K}}^e \underline{\underline{v}}^e - \underline{\underline{f}}^e \quad (2.101)$$

where

$$\underline{\underline{K}}^e = \left[\int_{\vartheta^e} \underline{\underline{B}}_s^{eT} \underline{\underline{\tilde{C}}}_\mu \underline{\underline{B}}_s^e d\vartheta + \gamma \int_{\vartheta^e} \underline{\underline{B}}_s^{eT} \underline{\underline{I}} \underline{\underline{I}}^T \underline{\underline{B}}_s^e d\vartheta \right], \quad \underline{\underline{f}}^e = \left[\int_{\vartheta^e} \rho \underline{\underline{N}}_v^{eT} \underline{\underline{f}} d\vartheta - \int_{S_r^e} \underline{\underline{N}}_v^{eT} \underline{\underline{t}} dS \right] \quad (2.102)$$

$\underline{\underline{v}}^e$ is the nodal velocity vector, $\underline{\underline{N}}_v^e$ is the elemental interpolation function matrix, $\underline{\underline{B}}_s^e = \underline{\underline{\nabla}}_s \underline{\underline{N}}_v^e$ is the strain displacement matrix which depends on the choice and order of element type selection [268] and $\underline{\underline{I}} = [1 \ 1 \ 0 \ 1]^T$. The individual element residual vectors $\underline{\underline{\Sigma}}^e$ are collated and assembled into a global system of algebraic equations written in terms of the solution variable vector $\underline{\underline{v}}$ and the global system residual vector $\underline{\underline{\Sigma}}$ as

$$\underline{\underline{\Sigma}} = \underline{\underline{K}}(\underline{\underline{v}}) \underline{\underline{v}} - \underline{\underline{f}} \quad (2.103)$$

A nonlinear iterative algorithm is required to obtain solution $\underline{\underline{v}}$ in the above equation such as the Newton Raphson or Picard iteration scheme. In the Newton Raphson the solution variable $\underline{\underline{v}}$ is iteratively updated via a gradient based algorithm until it approaches the actual solution according to

$$\underline{\underline{v}}^+ = \underline{\underline{v}}^- - \underline{\underline{J}}^{-1} \underline{\underline{\Sigma}} \quad (2.104)$$

The Tangent Stiffness Matrix (TSM) $\underline{\underline{J}}$ is obtained by differentiating the free residual vector $\underline{\underline{\Sigma}}$ with respect to the solution variable $\underline{\underline{v}}$, i.e. $\underline{\underline{J}} = \partial \underline{\underline{\Sigma}} / \partial \underline{\underline{v}}$. Similar to the global residual vector, $\underline{\underline{\Sigma}}$, the

system TSM $\underline{\underline{J}}$ is assembled from the element TSM $\underline{\underline{J}}^e$, where $\underline{\underline{J}}^e$ is obtained by differentiating the residual $\underline{\underline{\Sigma}}^e$ with respect to $\underline{\underline{v}}^e$ to obtain

$$\underline{\underline{J}}^e = \frac{\partial \underline{\underline{\Sigma}}^e}{\partial \underline{\underline{v}}^e} = \int_{\vartheta^e} \underline{\underline{B}}_s^{eT} \underline{\underline{\tilde{C}}}_\mu \underline{\underline{B}}_s^e d\vartheta + \int_{\vartheta^e} \alpha \underline{\underline{B}}_s^{eT} \underline{\underline{\tilde{C}}}_\mu \underline{\underline{B}}_s^e \underline{\underline{v}}^e \underline{\underline{v}}^{eT} \underline{\underline{B}}_s^{eT} \underline{\underline{\tilde{C}}}_\mu^T \underline{\underline{B}}_s^e d\vartheta + \gamma \int_{\vartheta^e}^1 \underline{\underline{B}}_s^{eT} \underline{\underline{I}} \underline{\underline{I}}^T \underline{\underline{B}}_s^e d\vartheta \quad (2.105)$$

Where $\alpha = (1/\mu^2 \dot{\gamma}) (d\mu/d\dot{\gamma})$ and depends on the viscosity model, and $\dot{\gamma}(\xi, \eta) = \sqrt{\underline{\underline{v}}^{eT} \underline{\underline{B}}_s^{eT} \underline{\underline{\tilde{C}}}_\rho \underline{\underline{B}}_s^e \underline{\underline{v}}^e}$. The fiber orientation tensor can be computed using the same FEA procedure outlined above. Recasting the Advani-Tuckers 2nd order orientation evolution equation into a weak form integral equation given as [269]

$$\int_{\vartheta^e} \underline{\underline{\omega}}_a \left[\frac{\partial \underline{\underline{a}}}{\partial t} + \underline{\underline{v}} \cdot \underline{\underline{\nabla}} \underline{\underline{a}} - \underline{\underline{\dot{a}}} \right] d\vartheta \quad (2.106)$$

where the orientation vector, $\underline{\underline{a}}_k$ contains the 5 independent components of the second order tensor $\underline{\underline{a}}_{ij}$, i.e. $\underline{\underline{a}}_k = \underline{\underline{a}}_{ij}$ according to the index transformation $k = j + 2(i - 1) \mid i = 1, 2; j = i \dots 3$; $\underline{\underline{\omega}}_a$ is an arbitrary weight vector and $\underline{\underline{\dot{a}}}_k = \underline{\underline{\dot{a}}}_{ij}$ contains the same five independent components of $\underline{\underline{\dot{a}}}_{ij}$ using the same index transformation where

$$\underline{\underline{\dot{a}}}_{ij} = -(\underline{\underline{\mathcal{E}}}_{ik} \underline{\underline{a}}_{kj} - \underline{\underline{a}}_{ik} \underline{\underline{\mathcal{E}}}_{kj}) + \kappa (\underline{\underline{\Gamma}}_{ik} \underline{\underline{a}}_{kj} + \underline{\underline{a}}_{ik} \underline{\underline{\Gamma}}_{kj} - 2\underline{\underline{\Gamma}}_{kl} \underline{\underline{a}}_{ijkl}) + 2D_r (\delta_{ij} - \alpha \underline{\underline{a}}_{ij}) \quad (2.107)$$

Transformation of the weak form equation to the FEA Galerkin formulation, and adopting a backward finite difference algorithm in time yields the element algebraic equation given as

$$\underline{\underline{\Sigma}}_a^e = \underline{\underline{K}}_a^e \underline{\underline{a}}^e - \underline{\underline{f}}_a^e \quad (2.108)$$

where

$$\underline{\underline{K}}_a^e = \int_{\vartheta^e} \left[\frac{\underline{\underline{N}}_a^{eT} \underline{\underline{N}}_a^e}{\Delta t} + \underline{\underline{N}}_a^{eT} \underline{\underline{v}} \cdot \underline{\underline{\nabla}} \underline{\underline{N}}_a^e \right] d\vartheta, \quad \underline{\underline{f}}_a^e = \int_{\vartheta^e} \left[\frac{\underline{\underline{N}}_a^{eT} \underline{\underline{N}}_a^e}{\Delta t} \underline{\underline{a}}^{e-} + \underline{\underline{N}}_a^{eT} \underline{\underline{\dot{a}}} \right] d\vartheta \quad (2.109)$$

$\underline{a}^e$ and $\underline{a}^{e-}$ are the orientation tensor component solution at the element nodes at the current and previous time step, $\underline{N}_a^e$ is the orientation solution variable interpolation function. Again, an iterative method is required to solve the assembled systems residual $\underline{\Sigma}_a$, such as the Newton-Raphson algorithm. As such, the element Jacobian $\underline{J}_a^e$ is required and computed as

$$\underline{J}_a^e = \frac{\partial \underline{\Sigma}_a^e}{\partial \underline{a}^e} = \underline{K}_a^e - \int_{\vartheta^e} \left[\underline{N}_a^{eT} \frac{\partial \dot{\underline{a}}}{\partial \underline{a}} \underline{N}_a^e \right] d\vartheta \quad (2.110)$$

Method for obtaining the exact derivative $\frac{\partial \dot{\underline{a}}}{\partial \underline{a}}$ is explained in detail in [Chapter 7](#). The post computation output includes pressure, stresses and orientation components. It should be noted that the pressure nodes are an order less than the velocity nodes.

2. PBM-SPH/DEM Algorithm

The coupled SPH/DEM method was used by Yang et al. [24] to simulate EDAM SFRP composite isothermal flow process in 2D. The fluid matrix while assumed to be Newtonian and incompressible, is represented by set of discrete SPH particles whose motion are defined by the fundamental laws of continuum mechanics neglecting the conservation energy equation while the suspended solid fibre particles are modelled as deformable particles using interlinked DEM particles. In the SPH method, the governing PDEs are transformed to ODEs through kernel approximation and particle approximation [217]. The kernel function W is used as a weighting function to obtain physical quantity of any particle by taking weighted sum of the relevant properties of all the particles within the kernel. The integral of an arbitrary function $f(x)$ and its derivative based on the kernel weighting function W are respectively given as

$$f(x) = \int_{\vartheta} f(x') W(|x - x'|, h) dx' \quad (2.111)$$

$$\underline{\nabla} \cdot f(x) = \int_S f(x') W(|x - x'|, h) \cdot \underline{\hat{n}} dx' - \int_{\vartheta} f(x') \nabla W(|x - x'|, h) dx' \quad (2.112)$$

where h is the *smoothing length*. When modelling the fluid phase, each SPH particle is assigned a mass and density, and its motion is influenced by interactions with surrounding particles within the support domain (cf. Figure 2.18)

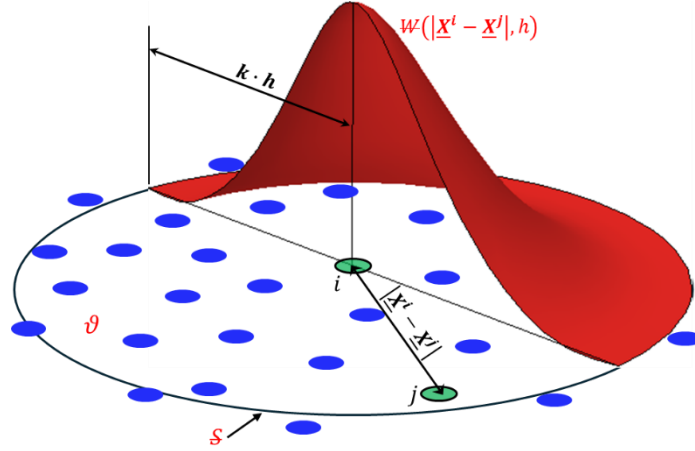


Figure 2.18: Schematic representation of the support domain $k \cdot h$ of the kernel for SPH particle i and its interactions with neighboring SPH particle j .

The i^{th} particle's density can be approximated using the *continuity density* equation given as

$$\frac{\partial \rho_m^i}{\partial t} + \underline{\dot{x}}_m^i \cdot \underline{\nabla} \rho_m^i = - \sum_{j=1}^N m_m^j \underline{\dot{x}}_m^{ij} \underline{\nabla} W_m^{ij} \quad (2.113)$$

where ρ_m^i , $\underline{\dot{x}}_m^i$ are the density and velocities of the i^{th} particle of the matrix phase denoted by subscript (m), m_m^j is the mass of the j^{th} neighbor particle, $\underline{\dot{x}}_m^{ij}$ is the relative velocities between the i^{th} & j^{th} particle given as $\underline{\dot{x}}_m^{ij} = \underline{\dot{x}}_m^i - \underline{\dot{x}}_m^j$, and W_m^{ij} is the kernel function whose gradient determines the contribution due to the relative velocities between the ij particle pairs. Likewise, the momentum equation in the SPH method is given as

$$\frac{\partial \underline{\dot{x}}_m^i}{\partial t} + \underline{\dot{x}}_m^i \cdot \underline{\nabla} \underline{\dot{x}}_m^i = \sum_{j=1}^N m_m^j \left[\frac{p_m^i}{(\rho_m^i)^2} + \frac{p_m^j}{(\rho_m^j)^2} + \Omega_m^{ij} + \Upsilon_m^{ij} \right] \underline{\nabla} W_m^{ij} + \frac{F_{ext}}{m_m^i} \quad (2.114)$$

where p_m^i is the particle pressure of the matrix phase given as $p_m^i = B[(\rho_m^i/\rho_m)^\gamma - 1]$, ρ_m^i is the matrix density associated with the i^{th} particle, ρ_m is the reference density of the matrix, γ is an exponent usually assumed to be $\gamma = 7$, B is the pressure constant. F_{ext}^i is the external force acting on the i^{th} SPH particle which is subject to reaction forces from the solid fiber phase DEM particles. Ω_m^{ij} is the viscosity term, Υ_m^{ij} is the anti-clump term for tensile instability respectively given as

$$\Omega_m^{ij} = m_m^j \frac{(\mu_m^i + \mu_m^j)}{\rho_m^i \rho_m^j} \frac{\Delta_m^{ij} \dot{\underline{X}}_m^{ij}}{(\Delta_m^{ij})^2 + 0.01h^2}, \quad \Upsilon_m^{ij} = \frac{v_{max}^2}{c_s^2} \left| \frac{p_m^i}{(\rho_m^i)^2} + \frac{p_m^j}{(\rho_m^j)^2} \right| \left[\frac{W_m^{ij}}{W(\Delta P)} \right]^4 \quad (2.115)$$

μ_m^i is the matrix viscosity associated with the i^{th} particle, Δ_m^{ij} is the distance between the ij particle pairs given as $\Delta_m^{ij} = |\underline{X}_m^j - \underline{X}_m^i|$, v_{max} is the maximum velocity of the fluid volume given as $v_{max} = c_s/10$, and ΔP is the initial particle spacing. The equation governing each solid fiber DEM particle considering the various forces acting on the particle is given as

$$m_f \left[\frac{d\dot{\underline{X}}_f}{dt} - \underline{g} \right] = \sum \underline{F}_{f,mech} + \sum \underline{F}_{f,lube} + \sum \underline{F}_{f,bond} + \underline{F}_{f,drag} + \underline{F}_{f,buoy} + \underline{F}_{f,FSI} \quad (2.116)$$

In eqn. (2.116) above, m_f and $\dot{\underline{X}}_f$ are the mass and velocities of a solid fiber phase particle denoted by subscript (f), $\underline{F}_{f,mech}$ is the net inter-particle direct contact forces, $\underline{F}_{f,lube}$ is the net lubrication forces between fiber particles, $\underline{F}_{f,bond}$ is the net force transfer across bonds between DEM particle elements, $\underline{F}_{f,drag}$ is the drag force acting on a fiber due to hydrodynamic resistance from the surrounding SPH fluid particles, $\underline{F}_{f,buoy}$ is the buoyancy force, $\underline{F}_{f,FSI}$ is the fluid-particle interaction force. The contact interaction between two DEM particle elements (cf. Figure 2.19) can be modeled by a spring and a dashpot in both the normal and tangential directions, along with a frictional element, as illustrated in Figure 2.19b. The mechanical contact force $\underline{F}_{f,mech}$ acting on a DEM particle element due to its interaction with other particle elements is dependent on the material behavior at the contact region and can be derived from the laws of motion. $\underline{F}_{f,mech}$ can be

decomposed into the normal $F_{f,mech}^n$ and shear $F_{f,mech}^s$ components. The normal force component can be computed as

$$F_{f,mech}^n = K_f^n U_f^n \quad (2.117)$$

where K_f^n is the normal stiffness and U_f^n is the overlap. The shear force components depend on the contact history and can be given as an integral over time

$$F_{f,mech}^s = - \int_0^t K_f^s v_f^s(\tau) d\tau, \quad v_f^s = \frac{\partial U_f^s}{\partial t} \quad (2.118)$$

where K_f^s is the shear stiffness at the contact, v_f^s is the shear component of the contact velocity at time t , and U_f^s is the shear component of the contact displacement. The maximum allowable shear contact force is limited by the slip condition (i.e. $F_{f,mech}^{s,max} = \mu_s |F_{f,mech}^n|$, μ_s is the slip friction coefficient at the contact).

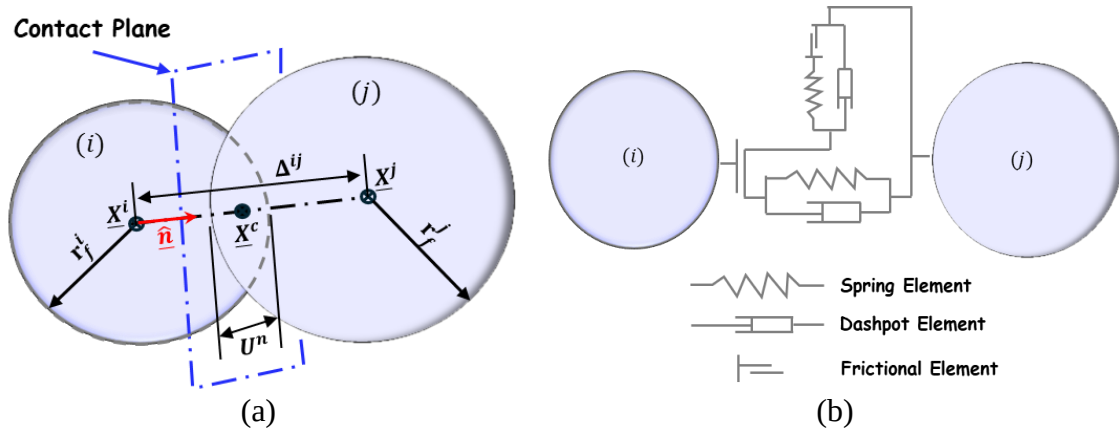


Figure 2.19: (a) Two DEM particle elements in direct contact with an overlap, (b) Representation of contact interaction between two DEM particle elements [217].

The contact stiffness k_c^{ij} between particle's i and j is a combination of the particle' element stiffness k_f^{ij} and parallel bond stiffness k_b^{ij} given as

$$k_c^{ij} = S_b k_b^{ij} + k_f^{ij} \quad (2.119)$$

where $k_f^{ij} = \left[k_f^{i-1} + k_f^{j-1} \right]^{-1}$, S_b is the bond cross-sectional area given as $S_b = 2r_b\delta$, r_b is the bond radius. The lubrication forces between two solid fiber DEM particles i and j is given as [270]

$$\underline{F}_{f,lube} = \begin{cases} -\frac{3\pi\mu_m d_f^{ij2}}{8(\Delta_f^{ij} - d_f^{ij})} \frac{\dot{\underline{X}}_f^{ij} \cdot \underline{X}_f^{ij}}{\underline{X}_f^{ij} \cdot \underline{X}_f^{ij}} \underline{X}_f^{ij} & \Delta_f^{ij} \leq 2d_f^{ij} \\ 0 & \Delta_f^{ij} > 2d_f^{ij} \end{cases} \quad (2.120)$$

where $\underline{X}_f^{ij} = \underline{X}_f^i - \underline{X}_f^j$, $\Delta_f^{ij} = |\underline{X}_f^{ij}|$, $d_f^{ij} = (d_f^i + d_f^j)/2$, d_f is the diameter of the DEM fiber particle and $2d_f^{ij}$ is the cutoff distance. The drag force $\underline{F}_{f,drag}$ can be mathematically modelled for a single DEM particle as

$$\underline{F}_{f,drag} = \frac{\mathfrak{P}_f}{1 - \mathfrak{d}_f} [\bar{\underline{X}}_m - \dot{\underline{X}}_f] \vartheta_f \quad (2.121)$$

where $\mathfrak{d}_f$ is the local mean voidage of fiber particle element, and $\bar{\underline{X}}_m$ is the average surrounding matrix flow velocity around a fiber particle which are respectively evaluated using Shepard filter given as

$$\mathfrak{d}_f = \frac{\sum \mathfrak{d}_m \vartheta_m W^{fm}}{\sum \vartheta_m W^{fm}}, \quad |\bar{\underline{X}}_m| = \frac{\sum \dot{\underline{X}}_m \vartheta_m W^{fm}}{\sum \vartheta_m W^{fm}} \quad (2.122)$$

where ϑ_f and ϑ_m are the volumes associated with the solid fiber and fluid matrix particles, $W^{fm} = W(|\underline{X}_f - \underline{X}_m|, h)$. In (2.122) above $\mathfrak{P}_f$ is the interphase momentum transfer coefficient, which can be expressed as a function the threshold value $\mathfrak{d}_f$ according to [271], [272]

$$\mathfrak{P}_f = \begin{cases} 150 \frac{(1 - \mathfrak{d}_f)^2}{\mathfrak{d}_f} \frac{\mu_m}{d_f^2} + 1.75 \frac{\rho_m}{d_m} (1 - \mathfrak{d}_f) |\bar{\underline{X}}_m - \dot{\underline{X}}_f| & \mathfrak{d}_f \leq 0.8 \\ 7.5 C_d \frac{\mathfrak{d}_f (1 - \mathfrak{d}_f)}{d_f} \rho_m |\bar{\underline{X}}_m - \dot{\underline{X}}_f| \mathfrak{d}_f^{-2.65} & \mathfrak{d}_f > 0.8 \end{cases} \quad (2.123)$$

In (2.123) above, and C_d is the drag coefficient on a single DEM particle given in terms of the Reynolds number Re_f as

$$C_d = \begin{cases} \frac{24}{Re_f} (1 + 0.15 Re_f^{0.687}) & Re_f \leq 10^3 \\ 0.44 Re_f & Re_f > 10^3 \end{cases}, \quad Re_f = \frac{|\bar{\dot{X}}_m - \dot{X}_f| d_f \rho_m d_f}{\mu_m} \quad (2.124)$$

The buoyancy force which results from density difference is given by

$$\underline{F}_{f,buoy} = d_f \rho_m \vartheta_f \cdot \underline{\hat{u}} \quad (2.125)$$

where $\underline{\hat{u}}$ is the unit vector parallel to the direction of the gravitational force acting on the solid particle. The kernel function is used to determine the apportioning of the reactions on each SPH particle by a weighted partitioning of the drag force acting on a DEM particle according to

$$F_{ext} = -\frac{m_m}{\rho_m} \sum \frac{1}{S_f^i} W^{fm} \underline{F}_{f,drag}, \quad S_f^i = \sum \frac{m_f^j}{\rho_f^j} W_f^{ij} \quad (2.126)$$

The process begins with particle element search of neighboring particle elements through a linked list algorithm and computation of the associated interaction forces acting on individual particle elements [270]. A finite difference scheme can be used to compute the SPH and DEM particles position and velocity from its acceleration at any instant. Subsequently the particles position and density is updated at the end of each time step, and the iteration process is repeated until the end of the computational cycle.

1.1.1.1.6. Deposition Flow Modelling

In deposition flow modelling, the energy conservation equation becomes important due to the associated convection/radiation heat transfer at the extrudate/bead surfaces and conduction heat transfer at bead-bead and bead-bed contact points during extrudate deposition/bead spreading. The different physical phenomenon involved in the deposition process includes the melt flow/melt front evolution, bead solidification, heat transfer, bead bonding/interlayer adhesion, polymer crystallization and viscoelastic stresses. Deposition flow models are often used to predict extrudate

shape/die swell phenomena, temperature distribution, warpage/deformation, residual stresses, bond area and integrity between adjacent beads and the reheat regions of the deposited beads [4]. The algorithm presented here are based on the work of [143], [273], [274]. The complete set of conservation equations for mass, momentum, and energy govern the transport phenomena during bead deposition and are given as.

$$\begin{aligned} \nabla_{\underline{X}_j} \dot{\underline{X}}_j &= \dot{\vartheta}^s \delta^s \\ \frac{\partial}{\partial t} (\rho \dot{\underline{X}}_j) + \nabla_{\underline{X}_i} (\rho \dot{\underline{X}}_i \dot{\underline{X}}_j) &= -\nabla_{\underline{X}_j} p + \tilde{\rho} f_j + \nabla_{\underline{X}_i} \sigma_{ij} + \gamma_t^f \int_{S_t^f} \kappa^f \hat{n}_j^f \delta^f dS^f \\ \frac{\partial}{\partial t} (\rho c_p \mathcal{T}) + \nabla_{\underline{X}_k} (\rho \dot{\underline{X}}_k \mathcal{T}) &= \nabla_{\underline{X}_k} (\kappa \nabla_{\underline{X}_k} \mathcal{T}) + \rho c_p \mathcal{T}^s \dot{\vartheta}^s \delta^s + \dot{q}_c \end{aligned} \quad (2.127)$$

where $\underline{\dot{X}}$ and $\mathcal{T}$ are the velocities and temperature at the material point ($\underline{X}$); $\dot{\vartheta}^s$, $\mathcal{T}^s$ are the volume flow rate and temperature at the source, $\underline{X}^s$ (cf. Figure 2.20), δ is a 3D delta function located at the flow front $\underline{X}^f$ or at the source $\underline{X}^s$, i.e. $\delta^f = \delta(\underline{X} - \underline{X}^f)$ and $\delta^s = \delta(\underline{X} - \underline{X}^s)$, γ_t is the surface tension at the polymer/air interface, ρ , c_p and κ are the density, heat capacity and thermal conductivity, respectively, $\tilde{\rho}$ is the variable density subject to the thermal expansion of air, and κ^f and $\hat{n}^f$ are the interface curvature and unit normal vector, respectively.

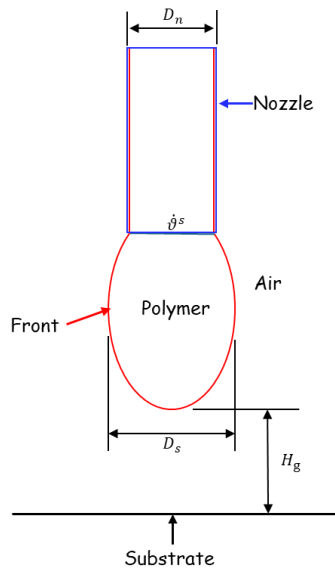


Figure 2.20: Typical Single strand deposition flow model computational domain

The effects of crystallization kinetics can be accounted for in the deposition process by coupling an external sink/source term $\dot{q}_c$ to the energy equation where $\dot{q}_c$ is given as

$$\dot{q}_c = \rho h_f \dot{\vartheta}_c \quad (2.128)$$

where h_f is the latent heat of fusion and $\dot{\vartheta}_c$ is the rate of crystallization [156] used an Avrami-type crystallization kinetic model that accounts for a dual crystallization mode and that captures the effect of trans-crystallinity around the suspended fiber particles. The model is given as [275]

$$\frac{\vartheta_c}{\vartheta_c^\infty} = \sum_{k=1,2} w_k \mathcal{F}_{c_k} \quad (2.129)$$

where ϑ_c is the volume fraction crystallinity, ϑ_c^∞ is the equilibrium volume fraction crystallinity, w_k is a weight factor describing relative occurrence of the dual crystallization process, and $\mathcal{F}_{c_k}$ represent the models for both crystallization process given as

$$\mathcal{F}_{c_k} = 1 - \exp \left[-\mathfrak{C}_{1k} \int_0^t \mathcal{T} \exp \left\{ \frac{-\mathfrak{C}_{2k}}{\mathcal{T} - \mathcal{T}_g + 51.6} - \frac{\mathfrak{C}_{3k}}{\mathcal{T}(\mathcal{T}_{m,k} - \mathcal{T})^2} \right\} n_k t^{n_k-1} dt \right] \quad (2.130)$$

where n_k and $\mathcal{T}_{m,k}$ are Advrami exponents and melt temperatures for both processes, $\mathcal{T}_g$ is the glass transition temperature of the polymer matrix and $\mathfrak{C}_{ij}$ are model constants. Mode details on crystallization kinetics during polymer processing can be found in [130].

A given material property ϕ at an arbitrary position $\underline{X}$ within the computational domain are evaluated from weighted averages of the polymer property ϕ^p and air property ϕ^a using an index function I_1 that differentiates the polymer phase from the air phase, i.e.

$$\phi(\underline{X}) = \phi^p(\underline{X}) + [\phi^a(\underline{X}) - \phi^p(\underline{X})] I_1(\underline{X}), \quad I_1 = \begin{cases} 0 & \text{polymer} \\ 1 & \text{air} \end{cases} \quad (2.131)$$

where $\phi = \rho, c_p, \kappa, \tilde{\rho}, \& \mu$. The effect of buoyancy due to temperature difference is accounted for in the air density and consequently the variable density $\tilde{\rho}$, using the Boussinesq approximation

i.e. $\tilde{\rho} = \rho^p + [\tilde{\rho}^a - \rho^p]I_1$, where, $\tilde{\rho}^a = \rho^a(1 - \alpha\Delta\mathcal{T})$ and α is the thermal expansion coefficient given as $\alpha = 1/\mathcal{T}$ for an ideal gas. In the usual manner, the Cauchy stress tensor is given as $\sigma_{ij} = \tau_{ij} - p\delta_{ij}$ where the deviatoric stress tensor is a composite stress made up of the fluid and viscoelastic stress contributions, i.e.

$$\tau_{ij} = (1 - I_s)[\tau_{ij}^m + \tau_{ij}^f] + I_s\tau_{ij}^s, \quad I_s = \begin{cases} 0 & \mathcal{T} > \mathcal{T}_m \\ 1 & \mathcal{T} \leq \mathcal{T}_m \end{cases} \quad (2.132)$$

where the polymer matrix solvent stress, $\tau_{ij}^m = 2\mu\Gamma_{ij}$, where $\mu = \mu(\dot{\gamma}, \mathcal{T})$, while τ_{ij}^s is the viscoelastic stress tensor due to solidification of the polymer at temperatures below the solidus point $\mathcal{T}_m$ and can be decomposed into the elastic, $\tau_{ij}^{s,e}$ and viscous damping, $\tau_{ij}^{s,v}$ terms, i.e.

$$\tau_{ij}^s = \tau_{ij}^{s,e} + \tau_{ij}^{s,v} \quad (2.133)$$

The viscous term has similar nature as the solvent stress given as $\tau_{ij}^{s,v} = 2\mu_s\Gamma_{ij}$, where μ_s is a damping coefficient. The elastic term is given as $\tau_{ij}^{s,e} = \mathbb{G}\mathcal{J}^{-\frac{5}{3}}\tilde{\mathbb{B}}_{ij}$, where $\mathbb{G}$ is the material's shear modulus $\mathcal{J} = \epsilon_{ijk}\Gamma_{i1}\Gamma_{j2}\Gamma_{k3}$, and $\tilde{\mathbb{B}}_{ij} = \mathbb{B}_{ij} - \mathbb{B}_{kk}/3$. The term $\mathbb{B}_{ij}$ is obtained from the deformation gradients, i.e., $\mathbb{B}_{ij} = \mathbb{F}_{ik}\mathbb{F}_{jk}$, where $\mathbb{F}_{ik}\mathbb{f}_{kj} = \delta_{ij}$. $\mathbb{F}_{ij}$ and $\mathbb{f}_{ij}$ are the deformation gradients and its inverse respectively and $\mathbb{f}_{ij}$ can be obtained from the solution to the differential equation given as

$$\frac{\partial \mathbb{f}_{ji}}{\partial t} + \dot{X}_k \frac{\partial \mathbb{f}_{ji}}{\partial X_k} + \mathbb{f}_{jk} \frac{\partial \dot{X}_k}{\partial X_i} = 0 \quad (2.134)$$

The contribution of the viscoelastic stress τ_{ij}^f due to the fiber orientation in the polymer, was assumed in [143] to be given as $\tau_{ij}^f = -\mu_0(\delta_{ij} - a_{ij})/\lambda_r$, where the zero-shear-rate viscosity $\mu_0 = \mu^*\alpha_T(\mathcal{T})$ and the relaxation time $\lambda_r = \lambda_r^*\alpha_T(\mathcal{T})$. The evolution of the fiber orientation tensor a_{ij} is defined by

$$\frac{\partial a_{ij}}{\partial t} + \dot{X}_k \cdot \nabla_{X_k} a_{ij} = \dot{a}_{ij} \quad (2.135)$$

where for the orientation tensor equation of change $\dot{a}_{ij}$, Xia et al. [143] assumed the following form

$$\dot{a}_{ij} = (\mathcal{E}_{ik} a_{kj} - a_{ik} \mathcal{E}_{jk}) + (\Gamma_{ik} a_{kj} + a_{ik} \Gamma_{jk}) - (\delta_{ij} - a_{ij})/\lambda_r \quad (2.136)$$

Following the work of Fattell et al. [276], for numerical stability, and to reduce the growth rate of the orientation tensor, Xia et al. [143] used a logarithmic form of the orientation tensor given as $Y_{ij} = \log[a_{ij}] = -\Phi_{im} \log[\mathbb{A}_{mn}] \Phi_{jn}$. With this transformation, the evolution equation can be rewritten in terms of Y_{ij} as

$$\frac{\partial Y_{ij}}{\partial t} + \dot{X}_k \cdot \nabla_{X_k} Y_{ij} = \dot{Y}_{ij} \quad (2.137)$$

where

$$\dot{Y}_{ij} = \mathfrak{U}_{ik} Y_{kj} - Y_{ik} \mathfrak{U}_{kj} + 2\mathfrak{U}_{ij} - (\delta_{ij} - e^{-Y_{ij}})/\lambda_r \quad (2.138)$$

$\mathfrak{U}_{ij}$ and $\mathfrak{L}_{ij}$ are obtained from decomposition of the velocity gradient according to $L_{ij} = \mathfrak{U}_{ij} + \mathfrak{L}_{ij} + \mathfrak{N}_{ik} \check{\mathfrak{A}}_{kj}$ and $a_{ik} \check{\mathfrak{A}}_{kj} = \delta_{ij}$. Moreover, $\mathfrak{U}_{ij} = \Phi_{im} \tilde{\mathfrak{U}}_{mn} \Phi_{jn}$, $\mathfrak{L}_{ij} = \Phi_{im} \tilde{\mathfrak{L}}_{mn} \Phi_{jn}$ and $\mathfrak{N}_{ij} = \Phi_{im} \tilde{\mathfrak{N}}_{mn} \Phi_{jn}$. Given, $\tilde{\mathfrak{U}}_{ij} = \Phi_{mi} \mathbf{L}_{mn} \Phi_{nj}$, then $\tilde{\mathfrak{L}}_{ij} = \delta_{ij} \tilde{\mathfrak{U}}_{ij}$ (no summation implied with repeated indices), $\tilde{\mathfrak{N}}_{ik} = (\tilde{\mathfrak{U}}_{ij} + \tilde{\mathfrak{U}}_{ji})/(\check{\mathfrak{A}}_{jj} - \check{\mathfrak{A}}_{ii}), i \neq j$, $\mathbb{A}_{ik} \check{\mathfrak{A}}_{kj} = \delta_{ij}$, and $\tilde{\mathfrak{U}}_{ij} = \tilde{\mathfrak{U}}_{ij} - \tilde{\mathfrak{L}}_{ij} - \tilde{\mathfrak{N}}_{ik} \check{\mathfrak{A}}_{kj}$. At the inlet and solid boundaries, the boundary condition $\hat{n}_k \cdot \nabla_{X_k} Y_{ij} = 0$ is imposed.

Solutions to the orientation tensor evolution equation can be obtained via explicit time integration, using a first order upwind approximation for the advection terms and the field-state variables including the position, velocities, pressure and temperature fields can be obtained by solving the Navier-Stokes equations via a finite volume approximation/front tracking scheme and

integration of the derivatives is achieved using a numerical ODE solution technique such as a high order Runge-Kutta or predictor-corrector method [143], [273], [274].

Extrudate swell of polymer melts during deposition is an important transport phenomena and modelling aspect of EDAM deposition flow process widely studied by various researchers. The swell phenomena of the compressed polymer as it is exposed to the environment is usually modelled by free surface minimization approach making several assumptions. The physics describing the die swell phenomena including the governing equations and applicable boundary conditions for a Newtonian polymer in a typical computational domain is shown in Figure 2.21 below for a straight flow model. Georgiou and Boudouvis [277] developed a Singular FEM (SFEM) method for solving the Newtonian extrudate-swell viscous flow problem with boundary stress singularities to obtain the position of the free-surface and the extrudate-swell ratio with improved convergence especially for low Reynold's number flow with high surface tension. Tanner [140] developed analytic solutions for the simplified extrudate swell problem assuming isothermal, incompressible flow, considering very high nozzle length to diameter ratio and ignoring body forces, surface tension, fluid inertia and small flow recovery far from the nozzle. He obtained for the extrudate swell ratio the following expression

$$w_f = \left[1 + \left(\frac{4-m}{m+2} \right) \left(\frac{\Delta\tau_1}{2\tau_w} \right)^2 \right]^{1/6} + 0.13 \quad (2.139)$$

where w_f is the extrudate swell ratio defined as the ratio of the extruded bead diameter to nozzle diameter, $\Delta\tau_1$ is the first normal stress difference given as $\Delta\tau_1 = \tau_{zz} - \tau_{rr}$, τ_w is the wall shear stress, i.e., $\tau_w = \tau_{rz}|_w$, m is a stress exponent, and the factor 0.13 accounts for small inelastic swelling in Newtonian creeping fluid flow. Heller [139] numerically studied the final extrudate shape for three (3) different types of deposition flow models including the (a) level flow, (b) bull

nose flow, and (c) falling flow models using free surface minimization technique. The initial geometry of the different flow models are determined by the gap height of the nozzle exit from the substrate and the leading edge of the flow-front upstream the deposited bead.

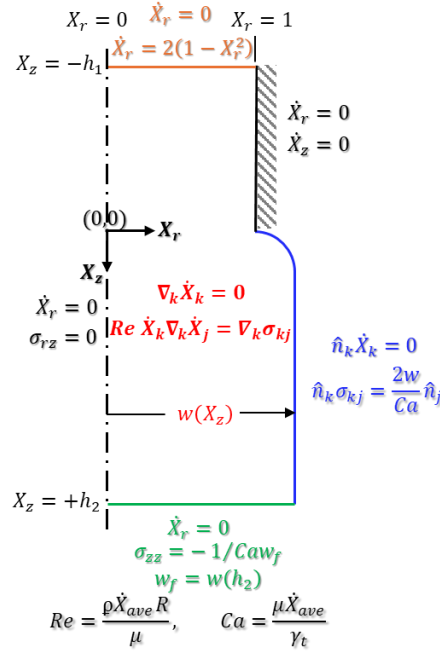


Figure 2.21: Computational domain and physics describing the extrudate-swell phenomena for a straight flow model

Bond formation mechanism between adjacent polymer beads was initially predicted by Bellehumeur et al. [154] using a simplified 1D lumped heat transfer model to simulate the cooling process of a single polymer bead road according to the energy ODE given by

$$\rho c_p S \dot{X} \frac{\partial \mathcal{T}}{\partial X} = S \frac{\partial}{\partial X} \left(\kappa \frac{\partial \mathcal{T}}{\partial X} \right) - h_T P (\mathcal{T} - \mathcal{T}_\infty) \quad (2.140)$$

where the deposited bead is assumed to be ellipse shaped (cf. Figure 2.22a) with cross sectional area $S = \pi r_1 r_2$ and perimeter $P = \pi(r_1 + r_2) [(64 - 3\mathfrak{d}^4)/(64 - 16\mathfrak{d}^2)]$ where $\mathfrak{d} = (r_1 - r_2)/(r_1 + r_2)$. He derived analytical solution to temperature field along the bead laying direction from the above ODE given as

$$\mathcal{T} = \mathcal{T}_\infty + (\mathcal{T}_0 - \mathcal{T}_\infty) e^{-m \dot{X} t} \quad (2.141)$$

where, $m = [-1 + \sqrt{1 + 4\alpha \cdot \beta}]/2\alpha$, $\alpha = \kappa/\rho c_p \dot{X}$, and $\beta = h_T P/\rho c_p S \dot{X}$. T_0 is the temperature at the source, T_∞ is the build environment temperature, h_T is a heat transfer coefficient that accounts for the effects of heat convection with air and conduction with substrate. Similarly, Thomas and Rodriguez [153] derived analytical solution for 2D transient heat transfer model for four stacked rectangular EDAM printed beads according to the Poisson's equation given as

$$\frac{\partial^2 \tilde{T}}{\partial X_1^2} + \frac{\partial^2 \tilde{T}}{\partial X_2^2} = \frac{1}{\alpha_k} \frac{\partial \tilde{T}}{\partial t} \quad (2.142)$$

where the normalized temperature $\tilde{T} = (T - T_\infty)/T_\infty$. Given the prescribed boundary conditions shown in Figure 2.22b, the solution of the temperature field averaged over the width of the bead is an eigenfunction series expansion given as

$$T_{ave}(X, t) = T_\infty + \frac{2T_\infty}{w} \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \left[\frac{n_{mn}}{\delta_n} \sin(\mathfrak{Q}_m X_2) \cos\left(\frac{\delta_n w}{2}\right) \right] e^{-\alpha_k^2 (\mathfrak{Q}_m^2 + \delta_n^2) t} \quad (2.143)$$

where $\alpha_k = \sqrt{\kappa/\rho c_p}$ and n_{mn} is given as

$$n_{mn} = \frac{4\tilde{T}_0}{\Omega_m^2 U_n^2 \mathfrak{Q}_m \delta_n} \sin\left(\frac{9\mathfrak{Q}_m h}{2}\right) \sin\left(\frac{\mathfrak{Q}_m h}{2}\right) \sin\left(\frac{\delta_n w}{2}\right) \quad (2.144)$$

where $\Omega_m^2 = 0.5(5h - \sin(10\mathfrak{Q}_m h)/2\mathfrak{Q}_m)$, $U_n^2 = 0.5(w + \sin(\delta_n w)/\delta_n)$, $\mathfrak{Q}_m$ and δ_n are solutions to the transcendental equations $\mathfrak{Q}_m \cot(5\mathfrak{Q}_m h) = -h_T/\kappa$, and $\delta_n \tan(0.5\delta_n w) = h_T/\kappa$. The 1D model is found to be more accurate for predicting bead temperature just after deposition while the 2D model more accurately predicts bead temperature after longer cooling times [139].

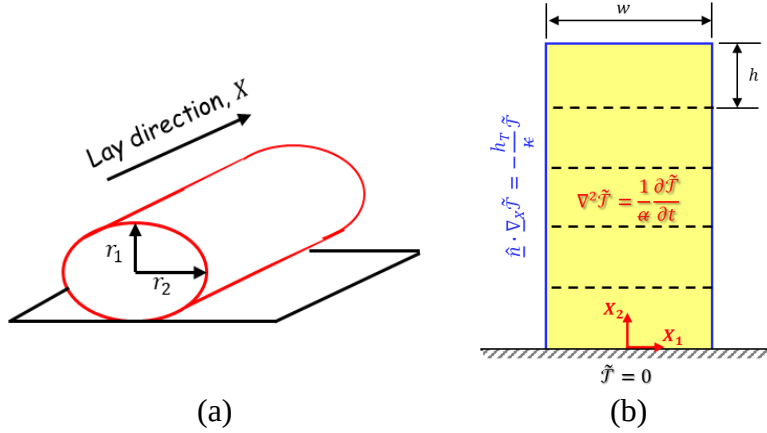


Figure 2.22: Schematic of (a) 1D single elliptical bead laying heat transfer process (b) 2D rectangular single road bead stack heat transfer model.

Bond formation usually begins with thermal excitation of polymer chains when a freshly deposited bead contacts a previously laid bead, followed by a wetting process that allows sufficient interface contact surface area between adjacent beads to form a well-defined bondline. The process is completed with the diffusion and randomization of polymer chains across the bondline according to the reptation theory. The bead wetting/spreading process determines the final shape of the deposited bead which is usually oblong shaped depending on the spreading rate, melt viscosity, relative surface energies of the bead and substrate surface and the interaction of the bead with the nozzle edges [131]. The bead spreading is usually accompanied by cooling and the final bead shape after solidification determines the contact surface area between adjacent beads, and the inter-bead void size and shape. An early theoretical model was proposed by Crockett et al. [150], [151] for approximating the bead spreading based on liquid droplet spreading model assuming laminar axisymmetric flow, constant bead cross section, Bingham fluid viscosity and ignoring nozzle tip interactions with the bead. The contact angles and active surface tension forces involved in the spreading process are shown in Figure 2.23a. He derived analytical solution for the change in contact angle θ_0 with time t given as

$$\left(\frac{\Delta\theta_0}{\Delta t}\right)_{y=R_s\theta_0} = \frac{S\gamma_{t,LV}}{8\phi\mu R_s^3} \left(\frac{\cos\theta_0 \cos(\phi - \theta_0)}{\phi} - \frac{2\sigma_y(t)R_s}{\gamma_{t,LV}} \right) \quad (2.145)$$

where S is the beads cross sectional area, θ_0 and ϕ are the bead contact angles, σ_y is the yield strength of the liquid, R_s is the spread radius, μ is the fluid viscosity, $\gamma_{t,LV}$, $\gamma_{t,SL}$, $\gamma_{t,SV}$ are the surface tensions at the liquid – vapor (air), solid – liquid and solid – vapor (air) interfaces respectively (cf. Figure 2.23a). The equation for the rate of change in contact angle θ_0 above was based on a free surface boundary condition. For a constrained surface boundary condition, the RHS of eqn. (2.145) is multiplied by a factor of 1/4. The Crockett's model does not account for the effect of cooling, temperature dependence of viscosity and actual polymer melt properties and thus yields inaccurate results when validated with experiment. The model, however, provides useful insight for understanding the bead spreading phenomena.

The bonding of adjacent polymer bead roads is interpreted in terms of the neck growth rate with respect to time which determines the rate of inter-molecular diffusion of polymer chains across the given neck area. Different models for the neck growth rate are presented in [139] and summarized in Table 2.3 below.

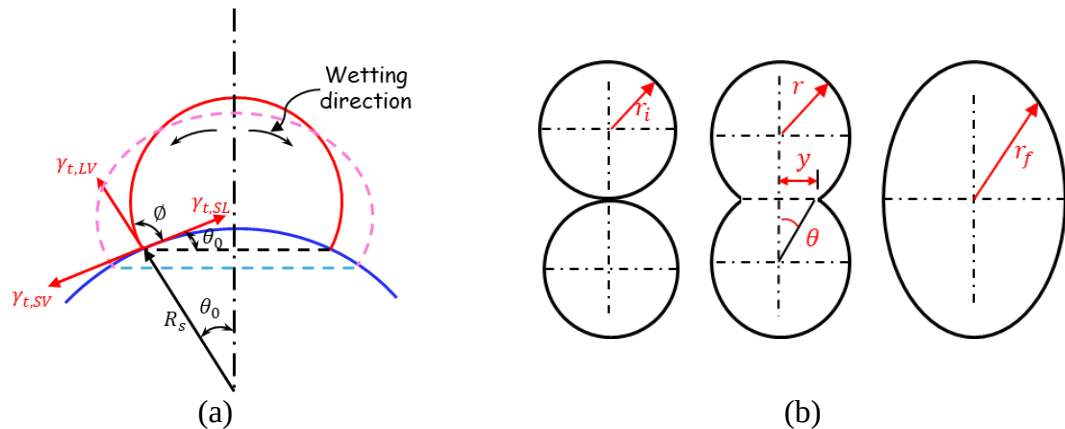


Figure 2.23: (a) Schematic representation of bead wetting and force balance based on Crockett's model (b) polymer bead bonding & neck growth process.

Table 2.3: Typical neck growth rate models

Models	Neck Growth Rate
Frenkel-Elshelby	$\theta = \sin^{-1} \sqrt{\frac{t\gamma_t}{\mu r_i}}$
Pokluda et al.	$\dot{\theta} = \frac{\gamma_t}{\mu r_i} \frac{2^{-5/3} \cos \theta \sin \theta (2 - \cos \theta)^{1/3}}{(1 - \cos \theta)(1 + \cos \theta)^{1/3}}$
Bellehumeur et al.	$8(A\lambda_r \kappa_1 \dot{\theta})^2 + \left(2A\lambda_r \kappa_1 + \frac{\mu r_i \kappa_2^2}{\gamma_t \kappa_1}\right) \dot{\theta} - 1 = 0$
Gurralla et al.	$\dot{\theta} = \frac{\gamma_t}{3\sqrt{\pi}\mu r_i} \left[\frac{[(\pi - \theta) \cos \theta + \sin \theta][(\pi - \theta) + \cos \theta \sin \theta]^{1/2}}{(\pi - \theta)^2 \sin^2 \theta} \right]$

In Table 2.3, r is the bead's radius, r_i is the initial beads radius, y is the sintering neck radius, θ is the angle between the bead's centroid and edge of the neck (cf. Figure 2.23b), t is the sintering time, γ_t is the surface tension and μ is the viscosity. In Table 2.3 above, $y/r = \sin \theta$, and $0 \leq y/r \leq 1$. λ_r is the relaxation time, A is a constant and $A = +1, -1, 0$ corresponds to the upper, lower and corotational derivatives of the viscoelastic extra stress tensor [278] and κ_1 and κ_2 are functions of θ given as

$$\kappa_1 = \frac{\sin \theta}{(1 + \cos \theta)(2 - \cos \theta)}, \quad \kappa_2 = \frac{2^{-5/3} \cos \theta \sin \theta}{(1 + \cos \theta)^{4/3} (2 - \cos \theta)^{5/3}} \quad (2.146)$$

Various models with differing levels of accuracy that predict the degree of healing d_h along the bondline during polymer bond formation have been developed by various researchers and summarized in [139]. One such model was developed by [Missing check Heller] and given as

$$d_h(t) = \left(\frac{l_p}{l_w}\right)^{1/2} = \left[\int_0^t \frac{dt}{t_w(\mathcal{T})} \right]^{1/4} \quad (2.147)$$

where t_w is the bondline weld time and is dependent on the temperature $\mathcal{T}$ as a function of time t , l_p is the minor polymer chain length defined in the reptation model, l_w is the minor chain length at reptation time.

The residual stresses and warpage that develop within the print during bead deposition and solidification impact the resulting strength properties and dimensional stability of the part. A simple analytical model based on beam bending theory was developed by Wang et al. [279] to predict the print deformation due to warpage δ_n given by

$$\delta_n = r_k \left(1 - \cos \frac{L}{2r_k} \right), \quad r_k = \frac{n^3 h}{6\alpha(\mathcal{T}_g - \mathcal{T}_\infty)(n - 1)} \quad (2.148)$$

where r_k is the radius of curvature, L is the section length of the stacking layers, n is the number of deposited layers, h is the layer height (cf. Figure 2.24), α is the linear shrinkage coefficient, $\mathcal{T}_g$ is the glass-transition temperature of the deposited materials, and $\mathcal{T}_\infty$ is the temperature of the build environment.

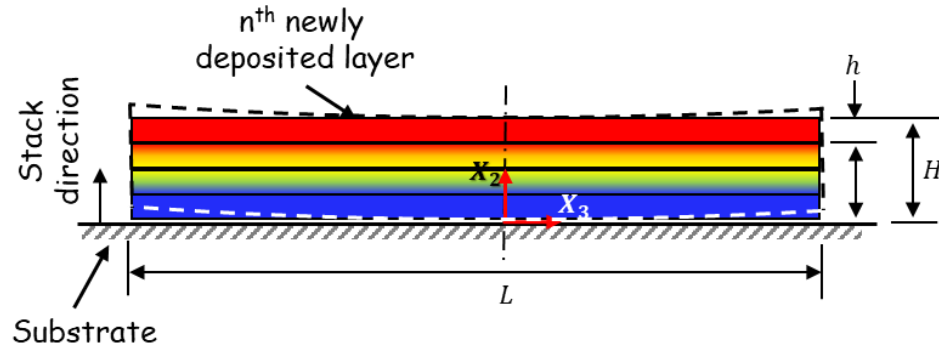


Figure 2.24: Annotated schematic of undeformed print bead stack

Likewise, Armillotta et al. [280] developed analytical expressions for the residual warp deformation at midspan based on experimental observations which is a combination of the elastic and plastic deflections and is given as

$$\delta_R = \frac{3}{4} \frac{\alpha L^2}{H} (\mathcal{T}_g - \mathcal{T}_\infty) \frac{mh}{H} \left(1 - \frac{mh}{H} \right) f_R \quad (2.149)$$

where $\mathcal{T}_m$ is the melt temperature and f_R is given as

$$f_R = \begin{cases} 1 & \text{if } H \geq h_R \\ 1 - .25(2 + c_R)(1 - c_R)^2 & \text{if } a_R < 4/3, H < h_R \end{cases} \quad (2.150)$$

where $m \approx (\mathcal{T}_m - \mathcal{T}_\infty)/(\mathcal{T}_g - \mathcal{T}_\infty)$, $c_R = [a_R - mh/H]/[3mh/H(a_R - mh/H)]$, $a_R = \sigma_Y/E_M\alpha(\mathcal{T}_g - \mathcal{T}_\infty)$, $h_R = 3mh/(2 - \sqrt{4 - 3a_R})$, σ_Y is the yield stress and E_M is the elastic modulus.

2.1.6.4 Microscale modelling of fiber suspension

Microscale level simulations are used to predict localized transport phenomena on the scale of the fiber particle smallest dimensions during EDAM SFRP polymer composite processing such as fiber motion/collision, fiber deformation/breakage, fiber clustering, micro-void formation, suspension rheology etc. Microscale simulations could likewise be sub-divided into analytical or numerical based simulations. The advantages and disadvantages of each modelling technique have been previously discussed in earlier sections. The subsequent sections present some discussion on analytical and numerical based micro-scale physics

1.1.1.1.7. Analytical-based simulation models

1. Particle Motion/Deformation

A common starting point for micro-scale modelling of dilute particle suspension is the utilization of the well-known Jeffery's analytical equation [19]. Jeffery's equations are often used to simulate single rigid particle's motion and more recently, the velocity and pressure flow-field around suspended particle in Newtonian viscous homogenous flows. Jeffery's model is used to predict the orientation dynamics of suspended particles and the rheology of dilute fiber suspensions. The Jeffery's model development is presented in Chapter 6 of this dissertation. Jeffery's assumptions of fixed particle shape, Newtonian fluid rheology and zero-Reynolds number flow are often termed as 'Standard Conditions' [169]. Deviations from the so called 'Standards Conditions' on the particle motion such as the presence of small degree of inertia, weakly non-

Newtonian fluid rheology and deformable particle shape are known to alter the deterministic behaviour of the particle's motion. Configurational indeterminacy thus depends on the characteristics of the undisturbed flow-field, the particle's geometry and shape deformability [169]. Traditionally, only studies based on small deviations from the Standard Conditions are theoretically feasible and are typically derived from asymptotic expansion about the leading order solution of creeping flow, Newtonian fluid rheology and fixed particle shape [169] which is described briefly in subsequent sections.

A. *Effect of weak non-Newtonian fluid rheology on particle motion*

Leal et al. [169] investigated the effect of weakly non-Newtonian fluid rheology on the motion of rigid particles. Given the Stokes approximation for creeping viscous fluid motion in terms of the disturbance quantities as

$$\nabla_{x_i} \dot{X}_i^d = 0, \quad \nabla_{x_i} \sigma_{ij}^d = 0 \quad (2.151)$$

and the boundary conditions of the undisturbed/infinite far-field flow and on the particle's surface, S_p respectively given as

$$\dot{X}_i^d \big|_{|\underline{X}| \rightarrow \infty} \rightarrow 0, \quad \dot{X}_j^{d,Sp} = \left[\dot{X}_{p_j} - \Gamma_{jk}^\infty X_k^{Sp} \right] + \epsilon_{jkm} [\dot{\theta}_{p_k} - \Xi_k^\infty] X_m^{Sp} \quad (2.152)$$

where $\dot{X}_i^d = \dot{X}_i - \dot{X}_i^\infty$ and the usual quantity $\dot{X}_i^\infty$ defined as $\dot{X}_i^\infty = L_{ij}^\infty \dot{X}_j$, where $L_{ij} = \nabla_{x_i} \dot{X}_j = \Gamma_{ij} + \epsilon_{imn} \Xi_m \delta_{nj}$. Leal et al. [169] incorporated the nonlinear effect into the constitutive equation σ_{ij}^∞ given as

$$\sigma_{ij}^d = -p^d \delta_{ij} + 2\Gamma_{ij}^d + \lambda [\Sigma_{ij}(\dot{\underline{X}}^d + \dot{\underline{X}}^\infty) - \Sigma_{ij}(\dot{\underline{X}}^\infty)] \quad (2.153)$$

where $\Gamma_{ij}^d = 1/2 [\nabla_{x_i} \dot{x}_j^d + \nabla_{x_j} \dot{x}_i^d]$, and $p^d = p - p_0$. The term $\Sigma_{ij}(\underline{\dot{x}})$ is a nonlinear function of $\underline{\dot{x}}$, and $\hat{\lambda} \ll 1$ is a small parameter that measures the magnitude of the non-linear contribution relative to the Newtonian stress. The equations for $\underline{\dot{x}}^\infty$ can be written as

$$\nabla_{x_k} \underline{\dot{x}}_k^\infty = 0, \quad -\nabla_{x_j} p_0 + \nabla_{x_k} \nabla_{x_k} \dot{x}_j^\infty + \hat{\lambda} \nabla_{x_k} \Sigma_{kj}(\underline{\dot{x}}^\infty) = 0 \quad (2.154)$$

Leal et al. [169] used an asymptotic expansion about the leading order solution method for obtaining solutions of $\underline{\dot{x}}_p$ and $\underline{\dot{\theta}}_p$ from the equations of the fluid motion and particle motion such that

$$u = u^{(0)} + \hat{\lambda} u^{(1)} + O(\hat{\lambda}^2) \quad (2.155)$$

where $u = \underline{\dot{x}}, p, \underline{\dot{x}}_p$, and $\underline{\dot{\theta}}_p$. The zeroth order solutions to the fundamental Stokes problem i.e., $u^{(0)} = \underline{\dot{x}}^{(0)}, p^{(0)}, \underline{\dot{x}}_p^{(0)}, \underline{\dot{\theta}}_p^{(0)}$, when $\hat{\lambda} = 0$ is assumed to be known and can be obtained from Jeffery's equations. The leading order solution of the particles motion $u^{(1)} = \underline{\dot{x}}_p^{(1)}, \underline{\dot{\theta}}_p^{(1)}$ can be obtained from the solution to the simultaneous equation given as [169].

$$\begin{aligned} -F_j^{(1)} + K_{T_{jk}} \dot{x}_{p_k}^{(1)} + K_{C_{jk}}^T \dot{\theta}_{p_k}^{(1)} &= \int_{\vartheta_f} [\Sigma_{mn}(\underline{\dot{x}}^d + \underline{\dot{x}}^\infty) - \Sigma_{mn}(\underline{\dot{x}}^\infty)] \nabla_{x_k} U_{T_{mn}} d\vartheta \\ -Q_j^{(1)} + K_{C_{jk}} \dot{x}_{p_k}^{(1)} + K_{R_{jk}} \dot{\theta}_{p_k}^{(1)} &= \int_{\vartheta_f} [\Sigma_{mn}(\underline{\dot{x}}^d + \underline{\dot{x}}^\infty) - \Sigma_{mn}(\underline{\dot{x}}^\infty)] \nabla_{x_k} U_{R_{mn}} d\vartheta \end{aligned} \quad (2.156)$$

where $[\Sigma_{mn}(\underline{\dot{x}}^d + \underline{\dot{x}}^\infty) - \Sigma_{mn}(\underline{\dot{x}}^\infty)]$ can be obtained from the solution to the disturbance-flow Stokes problem given by eqns. (2.153) - (2.154). The second order tensors $K_{T_{ij}}, K_{C_{ij}}^T, K_{C_{ij}}$, and $K_{R_{ij}}$ are given as integrals over the surface of the particle S_p , i.e.

$$\begin{aligned} K_{T_{ij}} &\equiv \int_{S_p} \hat{T}_{T_{ijk}} \hat{n}_k dS, & K_{C_{ij}}^T &\equiv \int_{S_p} \epsilon_{imn} X_m^{S_p} \hat{T}_{T_{njk}} \hat{n}_k dS \\ K_{C_{ij}} &\equiv \int_{S_p} \hat{T}_{R_{ijk}} \hat{n}_k dS, & K_{R_{ij}} &\equiv \int_{S_p} \epsilon_{imn} X_m^{S_p} \hat{T}_{R_{njk}} \hat{n}_k dS \end{aligned} \quad (2.157)$$

The second order tensors U_{Tij} , U_{Rij} and third order tensors, $\hat{T}_{Tijk}$ and $\hat{T}_{Rijk}$ can be obtained from solution to the complimentary Stoke's problem defined by the following sets of equations

$$\nabla_{X_i} \dot{X}'_i = 0, \quad \nabla_{X_i} \sigma'_{ij} = 0, \quad \sigma'_{ij} = -p' \delta_{ij} + 2\Gamma'_{ij} \quad (2.158)$$

and subject to the given sets of boundary conditions defined as

$$\dot{X}'_i \big|_{|\underline{X}| \rightarrow \infty} \rightarrow 0, \quad \dot{X}'_j{}^{Sp} = \hat{e}_{R_T} + \epsilon_{jkm} \hat{e}_{R_k} X_m^{Sp} \quad (2.159)$$

For the translation problem, the quantities $\dot{X}'_i$, p' and σ'_{ij} , in eqns. (2.158) - (2.159) are replaced by $\dot{X}'_{T_i}$, p'_T , & $\sigma'_{T_{ij}}$ which are respectively given as

$$\dot{X}'_{T_i} = U_{T_{ij}} \hat{e}_{T_j}, \quad p'_T = P_{T_j} \hat{e}_{T_j}, \quad \sigma'_{T_{ij}} = \hat{T}_{T_{ijk}} \hat{e}_{T_k} \quad (2.160)$$

Likewise, for the rotation problem, the quantities $\dot{X}'_i$, p' and σ'_{ij} , in eqns. (2.158) - (2.159) are replaced by $\dot{X}'_{R_i}$, p'_R , & $\sigma'_{R_{ij}}$ which are respectively given as

$$\dot{X}'_{R_i} = U_{R_{ij}} \hat{e}_{R_j}, \quad p'_R = P_{R_j} \hat{e}_{R_j}, \quad \sigma'_{R_{ij}} = \hat{T}_{R_{ijk}} \hat{e}_{R_k} \quad (2.161)$$

where $\hat{e}_T$ and $\hat{e}_R$ are the orientations of the translation and rotation axes of the particle. The complete solution to $\dot{X}'_i$, p' and σ'_{ij} are a combination of the individual solutions from the translation and rotation problems solved independently and given as

$$\dot{X}'_i = \dot{X}'_{T_i} + \dot{X}'_{R_i}, \quad p' = p'_T + p'_R, \quad \sigma'_{ij} = \sigma'_{T_{ij}} + \sigma'_{R_{ij}} \quad (2.162)$$

Complete asymptotic solution for a single particle motion in weakly non-Newtonian Carreau fluid was developed by Abtahi et al. [189] using similar methodology.

B. Effect of weak particle and fluid inertia on particle motion

Similar solution for single particle motion in Newtonian viscous flow with weak fluid inertia was developed by [169] using similar transport equations as with the weakly non-

Newtonian fluid solution (cf. eqn. (2.151) - (2.154)), however the non-linear stress contribution $\lambda \Sigma_{ij}(\underline{\dot{X}})$ is replaced by an inertial term $Re \mathcal{F}_j(\underline{\dot{X}})$ where

$$\mathcal{F}_k(\underline{\dot{X}}) = \frac{\partial \dot{X}_k}{\partial t} + \dot{X}_j \nabla_{X_j} \dot{X}_k \quad (2.163)$$

The leading order solutions of the particles motion $\underline{\dot{X}}_p^{(1)}$, $\underline{\dot{\theta}}_p^{(1)}$ for an unconfined domain based on $O(Re^2)$ asymptotic expansion is singular and requires a full matched asymptotic solution of the transport equations to obtain valid solution. The simple reciprocal theorem approach is however valid for confined flow problems that satisfies the condition $Re \ll (l_p/D_h)^m$, where l_p is the major particles length, D_h is a characteristic boundary dimension of the flow confinement and exponent m depends on the flow type (e.g. $m = 1$ for translation, $m = 2$, for shear flow, etc.). The leading order solutions of the particles motion $\underline{\dot{X}}_p^{(1)}$, $\underline{\dot{\theta}}_p^{(1)}$ under this consideration may thus be obtained from the solution to the simultaneous equation given as [169].

$$\begin{aligned} -F_j^{(1)} + K_{T_{jk}} \dot{X}_{p_k}^{(1)} + K_{C_{jk}}^T \dot{\theta}_{p_k}^{(1)} &= \int_{\vartheta_f} [\mathcal{F}_k(\underline{\dot{X}}^d + \underline{\dot{X}}^\infty) - \mathcal{F}_k(\underline{\dot{X}}^\infty)] U_{T_{kj}} d\vartheta \\ -Q_j^{(1)} + K_{C_{jk}} \dot{X}_{p_k}^{(1)} + K_{R_{jk}} \dot{\theta}_{p_k}^{(1)} &= \int_{\vartheta_f} [\mathcal{F}_k(\underline{\dot{X}}^d + \underline{\dot{X}}^\infty) - \mathcal{F}_k(\underline{\dot{X}}^\infty)] U_{R_{kj}} d\vartheta \end{aligned} \quad (2.164)$$

Recently, Einarsson et al. [190], [191], asymptotically obtained solutions for the motion of a small inertia ellipsoidal particle with dimensions $\mathcal{H}_1, \mathcal{H}_2$ ($\mathcal{H}_1 > \mathcal{H}_2$), suspended in Newtonian viscous shear flow with weak fluid inertia based on the perturbation theory. The solution is based on the dimensionless equation governing the particle's motion as

$$\dot{\rho}_j = \epsilon_{jkm} \dot{\theta}_{p_k} \rho_m, \quad St \left[I_{p_{jk}} \frac{\partial}{\partial t} \{ \dot{\theta}_{p_k} \} + \dot{\theta}_{p_k} \frac{\partial}{\partial t} \{ I_{p_{jk}} \} \right] = Q_j \quad (2.165)$$

in conjunction with the dimensionless transport equations governing the fluid motion given as

$$\nabla_{X_k} \dot{X}_k = 0, \quad Re_s \left[\frac{\partial \dot{X}_j}{\partial t} + \dot{X}_k \nabla_{X_k} \dot{X}_j \right] = -\nabla_{X_j} p + \nabla_{X_k} \nabla_{X_k} \dot{X}_j \quad (2.166)$$

subject to boundary conditions for the undisturbed flow at infinity and on the particle's surface, S_p respectively given as

$$\dot{X}_i|_{|\underline{X}| \rightarrow \infty} \rightarrow \dot{X}_i^\infty, \quad \dot{X}_j^{Sp} = \dot{X}_{pj} + \epsilon_{jkm} \dot{\theta}_{pk} X_m^{Sp} \quad (2.167)$$

where Re_s and St are the Reynold's and Stoke's number that quantifies the contribution of the fluid and particle's inertia to the particle's motion and respectively defined as $Re_s = \rho_p (\dot{\gamma} \mathcal{H}_1) \mathcal{H}_1 / \mu_f$, and $St = (\rho_p / \rho_f)$; ρ_p , and ρ_f are the particle's and fluid density respectively; $\dot{\gamma}$ and μ_f are the flow shear rate and fluids viscosity, $I_{p_{jk}}$ is the second order moment of inertia tensor given as

$$I_{p_{jk}} = C_1^I \rho_j \rho_k + C_2^I (\delta_{jk} - \rho_j \rho_k) \quad (2.168)$$

where the constants C_1^I and C_2^I are given as $C_1^I = 2/5 m_p \mathcal{H}_1^2$ and $C_2^I = 1/5 m_p (\mathcal{H}_1^2 + \mathcal{H}_2^2)$, m_p is the particles mass. He assumed for the solution of the particle's angular velocity the asymptotic series of the form

$$\dot{\theta}_{pk} = \dot{\theta}_{pk}^{(0)} + St \dot{\theta}_{pk}^{(1),St} + Re_s \dot{\theta}_{pk}^{(1),Re_s} + O(St^2) + O(Re_s^2) \quad (2.169)$$

where the zeroth order solution (i.e. $Re_s = St = 0$) for the particle's angular velocity is given as

$$\dot{\theta}_{pj}^{(0)} = \frac{1}{2} \epsilon_{jkm} \nabla_{X_k} \dot{X}_m^\infty + \kappa \epsilon_{jkm} \rho_k \Gamma_{mn}^\infty \rho_n \quad (2.170)$$

and the zeroth order solution for the particle's orientation evolution equation is given as

$$\dot{\rho}_j^{(0)} = \epsilon_{jrs} \dot{\theta}_{pr}^{(0)} \rho_s = \frac{1}{2} \epsilon_{jrs} \epsilon_{rkm} L_{km}^\infty \rho_s + \kappa \epsilon_{jrs} \epsilon_{rkm} \rho_k \Gamma_{mn}^\infty \rho_n \rho_s \quad (2.171)$$

which can be simplified further to yield the well-known Jeffery's equation thus

$$\dot{\rho}_j^{(0)} = \Xi_{jn}^\infty \rho_n + \kappa [\Gamma_{jn}^\infty \rho_n - (\rho_m \Gamma_{mn}^\infty \rho_n) \rho_j] \quad (2.172)$$

Einarsson et al. [190], [191] derived for small $-St$ and $-Re_s$ corrections to the Jeffery's equation of motion the following expression

$$\begin{aligned}\dot{\rho}_j = \dot{\rho}_j^{(0)} &+ \hbar_1(\rho_m \Gamma_{mn}^\infty \rho_n) \mathbb{P}_{jr} \Gamma_{rk}^\infty \rho_k + \hbar_2(\rho_m \Gamma_{mn}^\infty \rho_n) \Xi_{jk}^\infty \rho_k + \hbar_3 \mathbb{P}_{jn} \Xi_{nm}^\infty \Gamma_{mk}^\infty \rho_k \\ &+ \cdots \hbar_4 \mathbb{P}_{jn} \Gamma_{nm}^\infty \Gamma_{mk}^\infty \rho_k\end{aligned}\quad (2.173)$$

Or in spherical coordinates, eqn. (2.173) can be approximately written as

$$\begin{aligned}\dot{\phi} &\equiv \frac{1}{2}(\kappa \cos 2\phi - 1) + \frac{1}{8}\hbar_1 \sin^2 \theta \sin 4\phi - \frac{1}{4}(\hbar_2 \sin^2 \theta + \hbar_3) \sin 2\phi \\ \dot{\theta} &= \frac{1}{4}\kappa \sin 2\phi \sin 2\theta + \frac{1}{8}(\hbar_1 \sin^2 \theta \sin^2 2\phi + \hbar_3 \cos 2\phi + \hbar_4) \sin 2\theta\end{aligned}\quad (2.174)$$

The tensor $\mathbb{P}_{ij}$ projects components of a tensor T_{ij} in direction ρ_j such that $\mathbb{P}_{ik} T_{kj} = T_{ij} - \rho_k T_{ki} \rho_j$ and the four scalar coefficients $\hbar_\alpha$ are linear functions of Re_s and St . For particles with large aspect ratios $r_e = U_1/U_2 \gg 1$, the contributions of particles inertia were found to be negligible, and $\hbar_\alpha$ coefficients are only functions of Re_s and given as

$$\hbar_1 = \frac{7Re_s}{30 \log 2r_e - 45}, \quad \hbar_2 = \frac{3}{7}\hbar_1, \quad \hbar_3 = \hbar_4 = 0 \quad (2.175)$$

For nearly spherical particles the particles inertia becomes important, and $\hbar_\alpha$ coefficients are obtained to order $O(\epsilon_\lambda)$ as

$$\begin{aligned}\hbar_1 &= 0, \quad \hbar_2 = \epsilon_\lambda(St/15 + Re_s/35), \quad \hbar_3 = \epsilon_\lambda(St/15 - 37Re_s/105), \\ \hbar_4 &= \epsilon_\lambda(St/15 + 11Re_s/35)\end{aligned}\quad (2.176)$$

where $\epsilon_\lambda(r_e) = (r_e - 1)/r_e \rightarrow 0$ for nearly spherical particles.

C. *Effect of deformability on particle's motion*

Microscale simulation of fiber suspension that accounts for the fiber's flexibility is still at the nascent stage of research. Fiber's flexibility can typically be characterized using an effective stiffness dimensionless quantity [281] given as

$$S^{eff} \equiv \frac{E_p \pi}{4\mu_s \dot{\gamma} r_e^4} \quad (2.177)$$

where E_p is the modulus of elasticity of the particle, and μ_s is the suspension viscosity. Fiber's flexibilities are known to influence the suspension rheology. Existing theoretical models for simulating flexible fiber kinematics in dilute suspension regime can be divided into semi-flexible and flexible models. The semi-flexible bead-rod model was developed by Strautins and Latz [282] as an extension to the Jeffery's model consisting of two inter-connected rods each of length l'_p and having respective orientations $\rho^{(i)}$ and $\rho^{(j)}$ with attached beads at the ends and pivoted with a third bead and a spring of stiffness k_s at the joint that allows for flexibility and torsional resistance (cf. Figure 2.25). The beads provided surface area for hydrodynamic drag effects. The theory is limited to fibers with small bending angles.

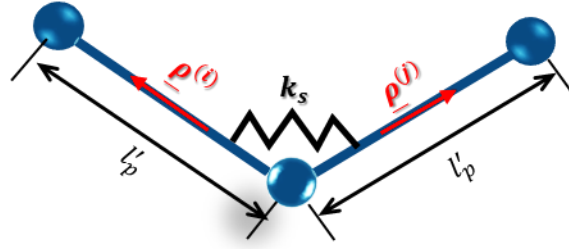


Figure 2.25: The semi-flexible fiber “bead-rod” model

The characteristics orientation tensors describing the semi-flexible fiber bead-rod model includes 1) a tensor $\underline{\underline{a}}^{(a)}$ that describes second moments of orientation vector of a single rod (i) with respect to the probability distribution function ψ , 2) a tensor $\underline{\underline{a}}^{(b)}$ that describes mixed products of the orientation vectors of both rods (i) and (j) with respect to the probability distribution function ψ and 3) a vector $\underline{\rho}^{(c)}$ that describes first moments of orientation vector of a single rod (i) with respect to the probability distribution function ψ . Mathematically $\underline{\underline{a}}^{(a)}$, $\underline{\underline{a}}^{(b)}$, and $\underline{\rho}^{(c)}$ are given as [240]

$$\begin{aligned}
\underline{\underline{a}}^{(a)} &= \iint \underline{\rho}^{(i)} \underline{\rho}^{(j)} \psi(\underline{\rho}^{(i)}, \underline{\rho}^{(j)}, t) d\underline{\rho}^{(i)} d\underline{\rho}^{(j)} \\
\underline{\underline{a}}^{(b)} &= \iint \underline{\rho}^{(i)} \underline{\rho}^{(j)} \psi(\underline{\rho}^{(i)}, \underline{\rho}^{(j)}, t) d\underline{\rho}^{(i)} d\underline{\rho}^{(j)} \\
\underline{\rho}^{(c)} &= \iint \underline{\rho}^{(i)} \psi(\underline{\rho}^{(i)}, \underline{\rho}^{(j)}, t) d\underline{\rho}^{(i)}
\end{aligned} \tag{2.178}$$

The equations of motion describing the evolution of these characteristic tensors of the semi-flexible particle in the presence of a flow-field are given as follows

$$\begin{aligned}
\dot{a}_{ij}^{(a)} &= [\Xi_{ik} a_{kj}^{(a)} - a_{ik}^{(a)} \Xi_{kj}] + [\Gamma_{lk} a_{kj}^{(a)} + a_{ik}^{(a)} \Gamma_{kj} - 2(\Gamma_{kl} a_{kl}^{(a)}) a_{ij}^{(a)}] \\
&\quad + \dots \frac{l'_p}{2} [\rho_i^{(c)} \mathbb{A}_j + \mathbb{A}_i \rho_j^{(c)} - 2(\mathbb{A}_k \rho_k^{(c)}) a_{ij}^{(a)}] - 2k_s [a_{ij}^{(b)} - a_{ij}^{(a)} a_{kk}^{(b)}] \\
\dot{a}_{ij}^{(b)} &= [\Xi_{ik} a_{kj}^{(b)} - a_{ik}^{(b)} \Xi_{kj}] + [\Gamma_{lk} a_{kj}^{(b)} + a_{ik}^{(b)} \Gamma_{kj} - 2(\Gamma_{kl} a_{kl}^{(a)}) a_{ij}^{(b)}] \\
&\quad + \dots \frac{l'_p}{2} [\rho_i^{(c)} \mathbb{A}_j + \mathbb{A}_i \rho_j^{(c)} - 2(\mathbb{A}_k \rho_k^{(c)}) a_{ij}^{(b)}] - 2k_s [a_{ij}^{(a)} - a_{ij}^{(b)} a_{kk}^{(b)}] \\
\dot{\rho}_j^{(c)} &= L_{kj} \rho_k^{(c)} - (a_{kl}^{(a)} L_{lk}) \rho_j^{(c)} + \frac{l'_p}{2} [\mathbb{A}_j - 2(\mathbb{A}_k \rho_k^{(c)}) \rho_j^{(c)}] - k_s \rho_j^{(c)} [1 - a_{kk}^{(b)}]
\end{aligned} \tag{2.179}$$

where $\mathbb{A}_k = [\nabla_{X_i} \nabla_{X_k} \dot{X}_i] a_{jk}^{(a)} \hat{e}_i$. The above model is based on Stokes flow, the flow induced bending of the particle would only occur if $\nabla_{X_i} \nabla_{X_k} \dot{X}_i$ exists. Hinch [196], [197] developed equations of motion for inextensible but fully flexible thread-like fiber particles, one governing the evolution of the particles motion, X_i and the other governing the tensile force, F_T in the threadlike particle which are respectively given as

$$\dot{X}_i = L_{ik} X_k + F'_T X'_i + \frac{1}{2} F_T X''_i, \quad F''_T - \frac{1}{2} F_T \|X''_i\|^2 = -\dot{X}_k \Gamma_{ki} \dot{X}_i \tag{2.180}$$

where the n th order partial derivative with respect to particle arc length s of a quantity f i.e. $\partial f^n / \partial^n s$ is represented by n superscripted apostrophes ('), and the deformation rate tensor Γ_{ij} is the symmetric part of the velocity gradient L_{ij} . Solution to X_i and F_T can be obtained from the above governing equations given an initial fiber orientation and boundary condition of zero initial tension on the particle (i.e. $F_T = 0$, @ $s = \pm l_p$). For nearly straight threadlike fibers, the asymptotic solution was given in the form

$$X_i(s, t) = s\rho_i(t) + \epsilon Y_i(s, t) + O(\epsilon^2) \quad (2.181)$$

where $\rho_i(t)$ is the solution to the fundamental Jeffery's orientation evolution equation, $\dot{\rho}_i$, and $Y_i(s, t)$ is given as

$$Y_i(s, t) = d_i(t) + q_i(t)\tilde{q}(s, t) + r_i(t)\tilde{r}(s, t) \quad (2.182)$$

where q_i and r_i are unit orthonormal directions to the orientation direction ρ_i and $\tilde{q}$ & $\tilde{r}$ are the respective shape orthonormal amplitudes which depending on the eigenmode shape (m) can be obtained from solution to the equation given as

$$\frac{d}{dt} \begin{pmatrix} \tilde{q}^{(m)} \\ \tilde{r}^{(m)} \end{pmatrix} = \begin{bmatrix} q_i \Gamma_{ij} q_j & q_i L_{ij} r_j \\ r_i L_{ij} q_j & r_i \Gamma_{ij} r_j \end{bmatrix} \begin{pmatrix} \tilde{q}^{(m)} \\ \tilde{r}^{(m)} \end{pmatrix} - \rho_i \Gamma_{ij} \rho_j \left(\frac{m^2 + m - 2}{4} \right) \begin{pmatrix} \tilde{q}^{(m)} \\ \tilde{r}^{(m)} \end{pmatrix} \quad (2.183)$$

The drift d_i is obtained from solution to the equation given as

$$\dot{d}_i = L_{ik} d_k + \rho_i \rho_m \Gamma_{mn} \left[\frac{1}{l_p} \int_{-l_p}^{l_p} (q_n \tilde{q} + r_n \tilde{r}) ds \right] \quad (2.184)$$

And the tension in the fiber can be obtained from

$$F_T = \rho_i \Gamma_{ij} \rho_j \frac{1}{2} (l_p^2 - s^2) - 2\epsilon + \rho_m \Gamma_{mn} \left[\int_{-l_p}^s Y_i ds - \frac{s + l_p}{2l_p} \int_{-l_p}^{l_p} Y_i ds \right] \quad (2.185)$$

Usually, an orthogonal unit direction to ρ_i is assumed e.g. r_i and the other, q_i can be found by vector algebra. Simulation results reveal a tendency for the particle to orient itself with the prevailing flow direction. Goddard and Huang [283] extended the dilute flexible particle model of Hinch to non-dilute systems by the introduction of a viscous drag transverse mobility tensor, K_{ij}^T (the hydrodynamic compliance per unit length) into the governing equations given as

$$\begin{aligned} \dot{X}_i &= X_k L_{ki} + K^L (\nabla_s F_T) (\nabla_s X_i) + \frac{1}{2} F_T K_{ij}^T (\nabla_s^2 X_j) \\ K^L \nabla_s^2 F_T - \frac{1}{2} K^N F_T \|\nabla_s^2 X_i\|^2 &= -\dot{X}_k \Gamma_{ki} \dot{X}_i \end{aligned} \quad (2.186)$$

where K^L and K^N are the lateral and normal components of the K_{ij}^T respectively given as

$$K^L = (\nabla_s X_i) K_{ij}^T (\nabla_s X_j), \quad \text{and}, \quad K^N = (\nabla_s^2 X_i) K_{ij}^T (\nabla_s^2 X_j) \|\nabla_s^2 X_i\|^{-2} \quad (2.187)$$

D. *Effect of Brownian disturbance on particle's motion*

Without Brownian couple, the motion of a rigid particle is typically described by the Jeffery's equation of fiber motion, $\dot{\underline{\rho}}$ however in the presence of Brownian couple, the particles orientation dynamics is best described statistically by a differential probability distribution function $\psi(\underline{\rho})$ based on the Fokker-Planck's continuity equation describing its time evolution given as

$$\dot{\psi} + \nabla_{X_i}(\psi \dot{\rho}_i - D_r \nabla_{X_i} \psi) = 0 \quad (2.188)$$

where D_r is the Stokes-Einstein diffusion coefficient. The bulk suspensions stress is obtained by volume average of the stress at the microscale of the suspended particles described by ψ to yield the expression given in eqn. (2.38). Additionally, by multiplying eqn. (2.188) above with $\rho_i \rho_j - 1/3 \delta_{ij}$ and integrating over ρ_i space, Prager [284] derived a direct orientation evolution equation for $a_{ij} = \langle \rho_i \rho_j \rangle$ commonly referred to as the Advani-Tucker's equation of change (cf. eqn. (2.83)).

2. *Forces acting on suspended particles*

The various forces and couple acting on a particle in viscous suspension are classified into three (3) including (a) hydrodynamic force contributions from the surrounding fluid medium (b) inter-particle hydrodynamic forces and (c) intra-particle fibre forces. Mathematically this can be written as

$$\underline{F}^T = \left(\sum \underline{F}^h \right)_{viscous} + \left(\sum \underline{F}^f \right)_{intraparticle} + \left(\sum \underline{F}^h \right)_{interparticle} \quad (2.189)$$

$$\underline{Q}^T = \left(\sum \underline{Q}^h \right)_{viscous} + \left(\sum \underline{Q}^f \right)_{intraparticle} + \left(\sum \underline{Q}^h \right)_{interparticle} \quad (2.190)$$

The various individual forces and torque contributions that influence the particle's motion, deformation and suspension rheology are briefly discussed below.

A. *Hydrodynamic viscous forces*

The hydrodynamic forces acting on a particle from its interaction with the surrounding fluid, F_i^H includes contribution from the (a) viscous drag forces, $\underline{F}^d$ due to the fluid resistance to the particles motion, (b) the force due to the acceleration of the suspending fluid medium, F_i^f and (c) the acceleration reaction on the particle, F_i^l mathematically given as

$$F_i^H = F_i^d + F_i^f + F_i^l \quad (2.191)$$

For single rod-like dilute particle suspension with negligible hydrodynamic interaction, the viscous drag force $\underline{F}^d$ and torque $\underline{Q}^d$ acting on a rigid cylindrical particle is respectively given as [285]

$$F_k^d \equiv D_{kl} \left(\bar{L}_{lm}^{\infty} X_{p_m} - \dot{X}_{p_l} \right), \quad Q_k^d \equiv \zeta_r \left[\dot{\theta}_k^{\infty} (\underline{\rho}) - \dot{\theta}_{p_k} \right] \quad (2.192)$$

where D_{kl} is the friction tensor given by $D_{kl} = \zeta_{\parallel} \rho_k \rho_l + \zeta_{\perp} (\delta_{kl} - \rho_k \rho_l)$, $\zeta_{\parallel}$, $\zeta_{\perp}$, are the parallel and perpendicular components of the friction tensor, and ζ_r is the rotational friction constant which are respectively given as

$$\zeta_{\parallel} = 2\pi\mu_0 l_p / \log(r_p), \quad \zeta_{\perp} = 2\zeta_{\parallel}, \quad \zeta_r = \pi\mu_0 l_p^3 / 3 \log(r_p) \quad (2.193)$$

For Newtonian viscous rod-like particle suspension system with negligible Brownian force, the average velocity gradient, $\bar{L}_{ij}$ and a macroscopic velocity field $\dot{X}_i^{\infty}$ are respectively given as [285]

$$\bar{L}_{ij} = \frac{1}{\vartheta} \int_{\vartheta} \frac{\partial \dot{X}_i}{\partial X_j} d\vartheta, \quad \dot{X}_i^{\infty} = \bar{L}_{ij}^{\infty} X_j \quad (2.194)$$

The eqn. (2.193) above is valid for $r_p \ll 1$ where r_p is the particle's aspect ratio given as $r_p \equiv l_p / 2a_p$ (cf. Figure 2.26). To avoid confusion, subscript (p) is used here to mean particle and is

not a tensor index. X_p , and $\underline{\rho}$ are the position vector of the center of mass and the unit vector along the symmetry axis of the particle, and $\dot{\theta}_j^\infty$ is the “torque free” angular velocity given as

$$\dot{\theta}_j^\infty = \epsilon_{jkm} \rho_k [(\kappa + 1) \bar{L}_{mn}^\infty \rho_n + (\kappa - 1) \bar{L}_{nm}^\infty \rho_n] \quad (2.195)$$

where $\kappa = (r_e^2 - 1)/(r_e^2 + 1)$, $r_e = r_e(r_p)$. Also, $\underline{\dot{X}}_p$ and $\underline{\dot{\theta}}_p$ are the velocity of the center of mass and the angular velocity of the particle, where $\dot{\theta}_{p_k} = \epsilon_{kmn} \rho_m \dot{\rho}_n$. The forces $\underline{F}^f$ and $\underline{F}^I$ in eqn. (2.191) are respectively given as [286]:

$$F_i^f = m_p \frac{\partial \dot{X}_i^\infty}{\partial t}, \quad F_i^I = -m_p \int_{\vartheta_f} \left[\frac{\partial \dot{X}_i}{\partial t} + \{ \dot{X}_k \nabla_{X_k} \dot{X}_j - \dot{\Delta}_{p_k} \nabla_{X_k} \dot{X}_j \} \right] \dot{M}_{ji}^+ d\vartheta \quad (2.196)$$

where $\dot{\Delta}_{p_k} = \dot{X}_{p_k} - \dot{X}_k^\infty$ and tensor $\dot{M}_{mn}^+$ is obtained from $\dot{M}_{mn}^+ \dot{\Delta}_{p_n} = \dot{X}_m^0$, and $\dot{X}_m^0$ is the Stokes velocity field obtained from the stokes equations $\nabla_{X_k} \dot{X}_k^0 = 0$, $-\nabla_{X_j} p_0 + \mu \nabla_{X_k} \nabla_{X_k} \dot{X}_j^0 = 0$. Using asymptotic expansion, Lovalenti et al. [286] derived for $\underline{F}^H$ the following expression

$$F_i^H = F_i^d + F_i^f + \tilde{F}_i^I + F_i^{OS} + F_i^{V\perp} + O(Re) + O(ReSl) \quad (2.197)$$

$$\tilde{F}_i^I = -\lambda_I \left\{ 6\pi \delta_{im} \delta_{mn} \delta_{nk} + \lim_{R_p \rightarrow \infty} \left(\int_{\vartheta_f(R_p)} \dot{M}_{ji}^+ \dot{M}_{jk}^+ d\vartheta - \int_{\vartheta_f(R_p) + \vartheta_p} \dot{M}_{ji}^p \dot{M}_{jk}^p d\vartheta \right) \right\} \frac{\partial}{\partial t} \dot{\Delta}_{p_k} \quad (2.198)$$

where λ_I is a constant, R_p is obtained from the expression $9/2 \pi [\delta_{ij} \delta_{jk}] R_p = \int_{\vartheta_f(R_p) + \vartheta_p} [\dot{M}_{ji}^p \dot{M}_{jk}^p] d\vartheta$ and tensor $\underline{\delta}$ is obtained from the viscous drag force such that $F_j^d = -6\pi \delta_{jk} \dot{\Delta}_{p_k}$. Also, $\dot{M}_{ij}^p$ is obtained from solution to $\nabla_{X_k} \dot{M}_{kj}^+ = 0$, $-\nabla_{X_i} p_j + \nabla_{X_k} \nabla_{X_k} \dot{M}_{ij}^+ = -6\pi \delta_{ik} \delta_{kj}(\underline{X})$. The term F_i^{OS} is the unsteady Oseen correction to the hydrodynamic force given as [286]

$$F_j^{OS} = \frac{3\lambda_{OS}}{8\sqrt{\pi}} \left\{ \int_{-\infty}^t \frac{2}{(t-\tau)^{1.5}} \left[\frac{2}{3} F_i^{d\parallel}(\tau) - f_{\bar{\lambda}} F_i^{d\parallel}(\tau) + \frac{2}{3} F_i^{d\perp}(\tau) - \left(\exp(-|\bar{\lambda}|^2) - \frac{1}{2} f_{\bar{\lambda}} \right) F_i^{d\perp}(\tau) \right] d\tau \right\} \delta_{ij} \quad (2.199)$$

where $F_j^{d\parallel}(t) = F_i^d(t)p'_ip'_j$, $F_j^{d\perp}(t) = F_i^d(t)(\delta_{ij} - p'_ip'_j)$, $f_{\bar{\lambda}}(t, \tau) = |\bar{\lambda}|^{-2}(.5\sqrt{\pi}|\bar{\lambda}|^{-1} \text{erf}|\bar{\lambda}| - \exp(-|\bar{\lambda}|^2))$, $\bar{\underline{\lambda}} = \bar{\underline{\lambda}}(t, \tau) = .5Re^{0.5}Sl^{-0.5}(t - \tau)^{-0.5}\underline{Y}(\tau)$, and λ_{OS} is a constant. Also, $\underline{Y}(\tau) = \int_{\tau}^t \underline{\dot{X}}_p d\tau$ and $\underline{p}'(\tau) = \underline{Y}(\tau)/|\underline{Y}(\tau)|$. The last term $F_i^{V\perp}$ which affects only the component of the force perpendicular to the slip velocity is given as

$$F_i^{V\perp} = -\lambda_{V\perp} \lim_{R_p \rightarrow \infty} \int_{\vartheta_f(R_p)} (\dot{X}_k^0 \nabla_{X_k} \dot{X}_j^0 - \dot{\Delta}_{p_k} \nabla_{X_k} \dot{X}_j^0) \dot{M}_{ji} d\vartheta \quad (2.200)$$

where $\lambda_{V\perp}$ is a constant.

B. Inter-particle hydrodynamic interaction forces

Interparticle hydrodynamic forces are split into long-range forces $\underline{F}^L$ and short-range forces $\underline{F}^S$. Analytical approximations of $\underline{F}^L$ based on asymptotic series expansion in existing literature are somewhat cumbersome and can be found in [287], [288]. The contribution of short-range lubrication forces to particles motion and overall suspension viscosity was analytically estimated by Yamane et al. [285] considering a simple shear flow system with velocity gradient given as $\bar{L}_{12} = \dot{\gamma}$, $\bar{L}_{ij} = 0$, $ij \neq 12$. Typical configuration for two-rod like particles hydrodynamically interacting with each other is shown in Figure 2.26

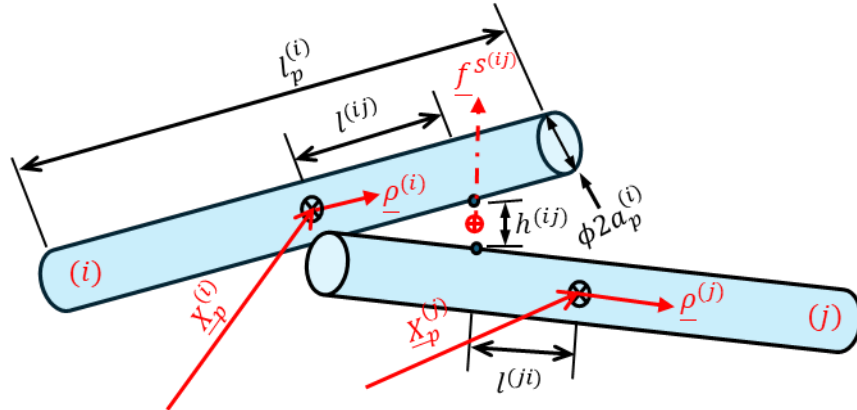


Figure 2.26: Two hydrodynamically interacting rod like particles at proximity

For a multi particle suspension system, the total hydrodynamic force, $F_k^{(i)}$ and torque, $Q_k^{(i)}$ acting on a particle (i) is given respectively as [285]

$$\begin{aligned} F_k^{(i)} &= F_k^H + \sum_{i \neq j} f_k^{s(ij)} + F_k^L + F_k^{ext} \\ Q_k^{(i)} &= Q_k^H + \sum_{i \neq j} l^{(ij)} \epsilon_{kmn} \rho_m^{(i)} f_n^{s(ij)} + Q_k^{ext} \end{aligned} \quad (2.201)$$

where $\underline{f}^{s(ij)}$ is the short-range lubrication force acting on particle (i) in a narrow gap $h^{(ij)} \ll a_p$, and due to its interaction with another particle (j). Considering a suspension system where $\underline{F}^f = \underline{F}^l = \underline{F}^{ext} = 0$, and $\underline{Q}^{ext} = 0$, Yamane et al. [285] obtained for $\underline{f}^{s(ij)}$ using lubrication theory the following expression:

$$f_k^{s(ij)} = \tilde{f}^{s(ij)} \hat{n}_k^{(ij)}, \quad \tilde{f}^{s(ij)} = K \dot{h}^{(ij)} \quad (2.202)$$

where, $K = 12\pi\mu_0 / \|\underline{\bar{\rho}}^{(ij)}\| \left(a_p^2 / h^{(ij)} \right)$, $\bar{\rho}_k^{(ij)} = \epsilon_{kmn} \rho_m^{(i)} \rho_n^{(j)}$, and $\hat{n}_k^{(ij)} = \bar{\rho}_k^{(ij)} / \|\underline{\bar{\rho}}^{(ij)}\|$. Also,

$h^{(ij)}$ and $\dot{h}^{(ij)}$ are respectively given as

$$\begin{aligned} h^{(ij)} &= \|\Delta_k^{(ij)} \hat{n}_k^{(ij)}\| - 2a_p \\ h^{(ij)} &= dh^{(ij)} / dt \\ &= \left[\left(\dot{X}_{p_k}^{(i)} + l^{(ij)} \epsilon_{kmn} \dot{\theta}_{p_m}^{(i)} \rho_n^{(i)} \right) \right. \\ &\quad \left. - \left(\dot{X}_{p_k}^{(j)} + l^{(ji)} \epsilon_{kmn} \dot{\theta}_{p_m}^{(j)} \rho_n^{(j)} \right) \right] \hat{n}_k^{(ij)} \end{aligned} \quad (2.203)$$

where $\Delta_k^{(ij)} = (X_{p_k}^{(i)} - X_{p_k}^{(j)})$, $l^{(ij)}$ is the distance between the center $\dot{X}_p^{(i)}$, of the particle (i) and the point on its axis that is nearest to particle (j), and is given as

$$l^{(ij)} = \frac{-\Delta_k^{(ij)} \rho_k^{(i)} + [\Delta_m^{(ij)} \rho_m^{(j)}] [\rho_n^{(i)} \rho_n^{(j)}]}{1 - [\rho_k^{(i)} \rho_k^{(j)}]^2} \quad (2.204)$$

By equilibrating the total drag, $F_k^{(i)}$ and torque $Q_k^{(i)}$, and making necessary substitutions, an expression was derived for $\tilde{f}^{s(ij)}$ and consequently $\underline{f}^{s(ij)}$, $\underline{\dot{X}}_p^{(i)}$ and $\underline{\dot{\theta}}_p^{(i)}$ for a given particle configuration given as

$$\begin{aligned} & \frac{\|\underline{\bar{\rho}}^{(ij)}\|}{12\pi\mu} \frac{h^{(ij)}}{a_p^2} \hat{n}_k^{(ij)} \hat{n}_k^{(ij)} \tilde{f}^{s(ij)} + \frac{1}{\zeta_\perp} \left[\sum_{\forall k} \hat{n}_m^{(ij)} \hat{n}_m^{(ik)} \tilde{f}^{s(ik)} + \sum_{\forall k} \hat{n}_m^{(ji)} \hat{n}_m^{(jk)} \tilde{f}^{s(jk)} \right] \\ & + \frac{1}{\zeta_r} \left[\sum_{\forall k} l^{(ij)} l^{(ik)} \hat{n}_m^{(ij)} \hat{n}_m^{(ik)} \tilde{f}^{s(ik)} + \sum_{\forall k} l^{(ji)} l^{(jk)} \hat{n}_m^{(ji)} \hat{n}_m^{(jk)} \tilde{f}^{s(jk)} \right] \quad (2.205) \\ & = -\hat{n}_m^{(ij)} L_{mn} \Delta_n^{(ij)} - \left[(\kappa + 1) l^{(ij)} \hat{n}_m^{(ij)} L_{mn} \rho_n^{(i)} + (\kappa - 1) l^{(ij)} \hat{n}_m^{(ij)} L_{nm} \rho_n^{(i)} \right] \\ & \quad - \left[(\kappa + 1) l^{(ji)} \hat{n}_m^{(ji)} L_{mn} \rho_n^{(j)} + (\kappa - 1) l^{(ji)} \hat{n}_m^{(ji)} L_{nm} \rho_n^{(j)} \right] \end{aligned}$$

Overall Yamane et al. [285] found the short-range hydrodynamic effects due to fiber interaction to be negligible, of the order $C_I \sim 10^{-7} - 10^{-4}$ in terms of the Folgar-Tuckers interaction coefficient.

C. Intra-fiber deformation forces

Forgacs and Mason [289] developed approximate analytical equations to estimate the forces causing deformation on a rigid thin rod particle in viscous suspension under simple shear, and neglecting Brownian motion based on *Burgers'* theory. The theory is used to investigate shear-induced fiber buckling phenomena under axial compression and possibly fiber breakage. Based on the theory, in the absence of inertia and assuming no slip at the rod-fluid interface, the total axial force F_a on the central cross section of the rod (cf. Figure 2.27) is approximately given as

$$F_a = - \int_{l_p/2}^0 f_a(l) dl \approx \frac{\pi \dot{\gamma} \mu l_p^2 M_\theta}{4 \log(2r_p) - 7} \quad (2.206)$$

where M_θ is an orientation factor given as $M_\theta = \sin^2 \theta \sin \phi \cos \phi$, θ and ϕ are the Euler orientation angles (cf. Figure 2.27). Given the total compressive force at an arbitrary point on the particle $r(\zeta, l)$ can be expressed as

$$F_c(l) = - \int_l^{l_p/2} p_a l \, dl = \frac{1}{2} p_a (l_p^2 - 4l^2), \quad p_a = \frac{\pi \dot{\gamma} \mu}{4 \log(2r_p) - 7} \quad (2.207)$$

Then based on the classical Euler's buckling theory, the critical condition under which rodlike particles with aspect ratio r_p and bending modulus E_b may be expected to buckle under shear-induced compression is approximately given as

$$(\dot{\gamma} \mu)_{crit} \approx \frac{E_b [\log(2r_p) - 1.75]}{2r_p^4} \quad (2.208)$$

Alternatively, for any given values of $\dot{\gamma}$, μ and E_b a critical aspect ratio $r_p = r_{p,crit}$ for which a particle may buckle under shear-induced compression which can be obtained from eqn. (2.208). By simulating the shear-rate variability within the liquefier using any of the viscosity models, flow regions where fiber breakage may occur can be approximated using the crude expression of eqn. (2.208) during preliminary studies.

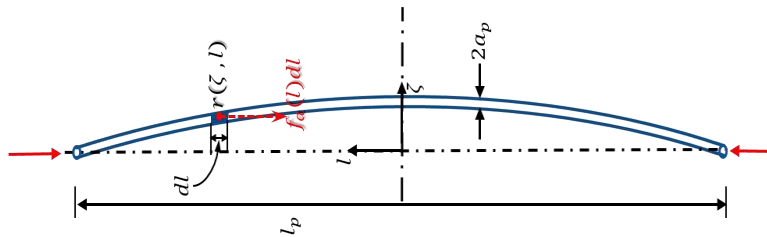


Figure 2.27: Typical fiber rod under axial compression used to investigate shear-induced buckling phenomena

3. Rheology of fiber suspension

The average suspension stress tensor can be derived using the principle of minimum energy dissipation given as [285]

$$\bar{\sigma}_{ij} = \frac{1}{2\vartheta} \frac{\partial \bar{E}_{min}}{\partial L_{ij}}, \quad \left| \min_{\underline{\dot{X}}, \underline{\dot{\Theta}}} \bar{E}(\underline{\dot{X}}, \underline{\dot{\Theta}}) \xrightarrow{\underline{\dot{X}}_{min}, \underline{\dot{\Theta}}_{min}} \bar{E}_{min}, \quad \bar{E} = \frac{\mu_0}{2} \int_{\vartheta_f} [L_{ij}(\underline{\dot{X}}, \underline{\dot{\Theta}})]^2 d\vartheta \quad (2.209)$$

The bulk stress of a dilute suspension with a force and torque “free” rod-like particle is given as [290].

$$\bar{\sigma}_{mn}^0 = \mu_0 \bar{f}_{mn} + N\pi a_p^2 l_p \mu_0 [2\bar{f}_{mn}] + \frac{\zeta_r}{2\vartheta} \sum_{i=1}^N \bar{L}_{iq} \rho_m^{(i)} \rho_n^{(i)} \rho_l^{(i)} \rho_q^{(i)} \quad (2.210)$$

The total bulk stress tensor for the concentrated suspension of many rod-like was particles consists of average stress resulting from the energy dissipation in dilute suspension $\bar{\sigma}_{mn}^0$, and dissipation due to inter-particle hydrodynamic interaction, $\bar{\sigma}_{mn}^{int}$ in concentrated regime given as [285]

$$\bar{\sigma}_{mn} = \bar{\sigma}_{mn}^0 + \bar{\sigma}_{mn}^{int} \quad (2.211)$$

where

$$\begin{aligned} \bar{\sigma}_{mn}^{int} = & -\frac{1}{\vartheta} \sum_{i < j} \left\{ f_{hm}^{(ij)} \left(\Delta_n^{(ij)} + l^{(ij)} \rho_n^{(i)} - l^{(ji)} \rho_n^{(j)} \right) \right. \\ & \left. + (\kappa - 1) [l^{(ij)} (\rho_n^{(i)} f_{hm}^{(ij)} + \rho_m^{(i)} f_{hn}^{(ij)}) - l^{(ji)} (\rho_n^{(j)} f_{hm}^{(ji)} + \rho_m^{(j)} f_{hn}^{(ji)})] \right\} \end{aligned} \quad (2.212)$$

Given a simple shear flow system with velocity gradient given as $\bar{L}_{12} = \dot{\gamma}$, $\bar{L}_{ij} = 0$, $ij \neq 12$, the excess viscosity can be computed from $\Delta\mu = (\bar{\sigma}_{12}/\dot{\gamma} - \mu_0)$.

1.1.1.1.8. Numerical-based simulations

Microscale level numerical simulations are either developed on the basis of the element or particle-based methods (EBM or PBM) as earlier discussed. Numerical methods popularly adopted in literature for microscale modelling of transport phenomena in EDAM polymer composite processing are the EBM based - finite element methods (FEM) [52], [52], [225], [227], [229], [230], [231], [291] the PBM based SPH & MPS methods [201], [202], [203], [207], [209], [233] and the PBM based - discrete element methods (DEM) also known as the particle simulation

method (PSM) or the *Stokesian Dynamics* method [200], [210], [212], [213], [214], [292]. Details on the microscale FEM model development can be found in [Chapter 7](#) of this dissertation. Literature on EBM based numerical simulations can be found in Kugler et al. [20] while the physics and details on the PSM model development can be found in [200] which we summarize in subsequent sections. In PSM, the fibers are modelled as a framework of rigid spheres inter-linked with extensible connector members having joints with axial, bending and torsional stiffness properties that allows for elastic and flexible motion of the bead-chain structure (cf. Figure 2.28a). Each rigid spheres or particle element are independently modelled, and their motion is governed by Newton's laws of motion given as

$$m \frac{d\dot{\underline{X}}}{dt} = \underline{F}^T, \quad I \frac{d\dot{\underline{\Theta}}}{dt} = \underline{Q}^T \quad (2.213)$$

where m is the particle element's mass, $I = \frac{2}{5} m a^2$ is the angular moment of inertia of the particle element of diameter a and $\underline{F}$ and $\underline{Q}$ are the forces and couples acting on the particle element respectively which consist of contributions from hydrodynamic viscous drag, intraparticle and interparticle interaction effects which are briefly discussed in subsequent sections. An arbitrary fiber structure (n) has a center of mass (CoM), ${}^n\underline{X}^c$ defined as the weighted average of the position vectors of the individual particle elements ($i = 1, 2 \dots N_e$) within the structure given as

$${}^n\underline{X}_k^c = \frac{\sum_{i=1}^{N_e} m_i {}^n\underline{X}_{k,i}}{\sum_{i=1}^{N_e} m_i} \quad (2.214)$$

Different strategies have been utilized by various researchers for modelling the joints with slightly different constitutive relations. For instance, [212], [292] places each joint between two particle elements (cf. Figure 2.28b) while [200] locates a joint at the center of each particle element (cf. Figure 2.28c). The physics presented here are for the latter case. The j th connector unit

orientation vector is given as ${}^n_j\hat{q} = {}^n_j\Delta / \|{}^n_j\Delta\|$ where ${}^n_j\Delta$ relates to the particle element global and local position vector n_iX and ${}^{nc}_i\Delta = {}^n_iX - {}^nX^c$ according to the expressions given respectively as

$${}^n_j\Delta_k = \sum_{i=1}^{N_e} b_{ji} {}^n_iX_k, \quad \text{and}, \quad {}^{nc}_i\Delta_k = \sum_{j=1}^{N_e} \tilde{b}_{ij} {}^n_j\Delta_k \quad (2.215)$$

where $b_{ji} = \delta_{j+1,i} - \delta_{j,i}$ and $\tilde{b}_{ij} = j/N_e - \mathfrak{d}_{ij}$, $\mathfrak{d}_{ij} = \begin{cases} 0 & j < i \\ 1 & j \geq i \end{cases}$. The inextensible connector

length $\tilde{\Delta} = \|{}^n_j\Delta\|$ is the same for all rigid connectors, i.e. ${}^n_j\Delta_k = \tilde{\Delta} {}^n_j\hat{q}_k$.

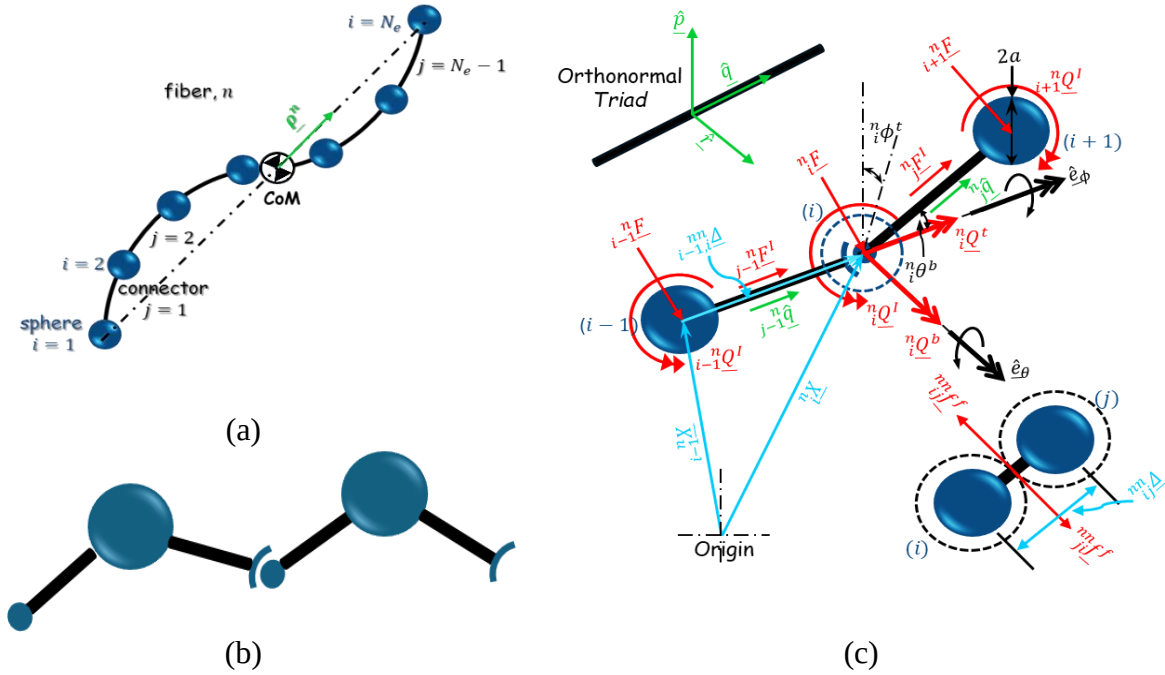


Figure 2.28: (a) Chain of spheres representing a flexible fiber model and showing the average orientation axis and fiber's center of mass (b) PSM joint linking two connectors (c), Free body diagram showing the various forces and moment acting on three (3) sphere PSM chain.

The viscous Stokesian drag forces and couple acting on a particle element (i) of a fiber structure (n) is given by [200], [210]

$${}^n_iF_k^D = -6\pi\mu_s a [{}^n_i\dot{X}_k - {}^n_i\dot{X}_k^\infty], \quad {}^n_iQ_k^D = -8\pi\mu_s a^3 [{}^n_i\dot{\theta}_k - {}^n_i\dot{\theta}_k^\infty] \quad (2.216)$$

where ${}^n_i\dot{X}_k^\infty = L_{mn}^\infty {}^n_iX_k$ and ${}^n_i\dot{\theta}_j^\infty \approx \epsilon_{jkm}\rho_k\dot{\rho}_m = \epsilon_{jkm}\rho_k[(\kappa+1)L_{mn}^\infty\rho_n + (\kappa-1)L_{nm}^\infty\rho_n]$. The hydrodynamic effect for a fiber (n) with N_e particle elements is calculated by [210]

$$\begin{bmatrix} {}^n\mathbf{F}^D \\ {}^n\mathbf{Q}^D \end{bmatrix} = -\underline{\underline{\mathcal{M}}}_{PSM}^{-1} \begin{bmatrix} {}^n\dot{\mathbf{X}} - {}^n\dot{\mathbf{X}}^\infty \\ {}^n\dot{\underline{\Delta}} - {}^n\dot{\underline{\Delta}}^\infty \end{bmatrix} \quad (2.217)$$

where $\underline{\underline{\mathcal{M}}}_{PSM}$ is a $6N_e \times 6N_e$ mobility matrix and all other quantities in eqn. (2.217) are $6N_e \times 1$ vectors. The contribution of the total disturbance forces acting on particle element (i) of fiber (n) due to long-range hydrodynamic interaction with other particle elements ($j = 1 \dots N_e$) from all fiber structures ($m = 1 \dots N_f$) is given as [200]

$${}^n\mathbf{F}_k^L = 6\pi\mu_s a \sum_{m=1}^{N_f} \sum_{j=1}^{N_e} \frac{(1 - \delta_{mn}\delta_{ij})}{8\pi\mu_s \|\mathbf{mn}_{ji}\underline{\Delta}\|} \{ \delta_{pq} + \mathbf{mn}_{ji}\hat{\underline{\Delta}}_p \mathbf{mn}_{ji}\hat{\underline{\Delta}}_q \} {}^m\mathbf{F}_q \quad (2.218)$$

where $\mathbf{mn}_{ji}\hat{\underline{\Delta}} = \mathbf{mn}_{ji}\underline{\Delta} / \|\mathbf{mn}_{ji}\underline{\Delta}\|$, $\mathbf{mn}_{ji}\underline{\Delta}_k = {}^m\mathbf{X}_k - {}^n\mathbf{X}_k$. Likewise, the contribution from short range hydrodynamic lubrication forces on particle element (i) of fiber (n) due to interaction with particle elements (j) of fiber structures (m) is given as [200], [210]

$${}^n\mathbf{F}_k^S = -3\pi\mu_s \sum_{\forall j} \left[\frac{a^2}{\|\mathbf{mn}_{ji}\underline{\Delta}\| - 2a} \mathbf{mn}_{ji}\hat{\underline{\Delta}}_k \mathbf{mn}_{ji}\hat{\underline{\Delta}}_q [{}^m\dot{\mathbf{X}}_q - {}^n\dot{\mathbf{X}}_q] \right] \quad (2.219)$$

The expression is valid for $\tilde{\epsilon}_{gap} \leq \tilde{\Delta} - 2a \leq 0.2a$, where $\tilde{\epsilon}_{gap}$ is a positive perturbation that ensures numerical stability. The force on particle element (i) of fiber (n) due to collision with another particle elements (j) of fiber structures (m) is given as [210]

$${}^m\mathbf{f}_{jk}^C = \begin{cases} 0 & \|\mathbf{mn}_{ij}\underline{\Delta}\| > 3a \\ -\frac{3}{2}\pi\mu_s \left[\frac{a^2}{\|\mathbf{mn}_{ji}\underline{\Delta}\| - 2a} \mathbf{mn}_{ji}\hat{\underline{\Delta}}_k \mathbf{mn}_{ji}\hat{\underline{\Delta}}_q [{}^m\dot{\mathbf{X}}_q - {}^n\dot{\mathbf{X}}_q] \right] & 2.001a \leq \|\mathbf{mn}_{ij}\underline{\Delta}\| < 3a \\ -D_c\pi\mu_s a^2 \dot{\gamma} \left[\mathbf{mn}_{ji}\hat{\underline{\Delta}}_k \exp \left[G_c \left(1 - \frac{\|\mathbf{mn}_{ji}\underline{\Delta}\|}{2a} \right) \right] \right] & \|\mathbf{mn}_{ij}\underline{\Delta}\| < 2.001a \end{cases} \quad (2.220)$$

where D_c and G_c are constants. The internal tension in the inextensible rigid connector (j) of a fiber structure (n) due to forces acting on particle elements ($j = 1 \dots N_e$) is given as

$${}^nF_k^I = -{}^n\hat{q}_k \sum_{i=1}^{N_e} \text{sgn}(i, j) {}^n\hat{q}_k {}^nF_k, \quad \text{sgn}(i, j) = \begin{cases} -1 & j \geq i \\ +1 & j < i \end{cases} \quad (2.221)$$

The internal couple acting on a joint (i) of fiber (n) arising from the moments due to the forces acting on the particle elements ($j = 1 \dots N_e$) of the same fiber structure is given as [200]

$${}^nQ_k^I = \sum_{j=1}^{N_e} \epsilon_{kpq} {}^{nn}\Delta_p {}^nF_q \quad (2.222)$$

The internal moment and torsion at a joint (i) due to flexural bending and twisting are dependent on the joint flexural properties and are respectively given as

$${}^nQ_k^b = k^b [{}^n\theta_k^b - {}^n\theta_k^{b0}], \quad {}^nQ_k^t = k^t [{}^n\phi_k^t - {}^n\phi_k^{t0}] \quad (2.223)$$

Where ${}^n\theta_k^b$ and ${}^n\phi_k^t$ are the bend and twist angles at the joint; and ${}^n\theta_k^{b0}$ and ${}^n\phi_k^{t0}$ are the bend and twist angles at the joint and at equilibrium position. k^b and k^t are the torsional stiffnesses respectively given as $k^b = \pi E_b a^3 / 8$ and $k^t = \pi G_t a^3 / 4$, E_b and G_t is the Young and shear modulus of the joint. If a connector is extensible, then the stretching force on the connector is simply given as ${}^nF_k^e = k^e [\tilde{\Delta} - \tilde{\Delta}^0] {}^n\hat{q}$, where $\tilde{\Delta}^0$ is the equilibrium connector length. The final equation of motion for a particle element (i) of fiber (n) is which is solved implicitly given as

$${}^n m \frac{d}{dt} \{ {}^n \dot{X}_k \} = {}^n F_k^D + {}^n F_k^I + {}^n F_k^L + {}^n F_k^S + {}^n F_k^e + \sum_{\forall j} {}^{mn} f_k^c + \sum_{\forall j} {}^{nn} f_k^f \quad (2.224)$$

$${}^n I \frac{d}{dt} \{ {}^n \dot{\theta}_k \} = {}^n Q_k^D + {}^n Q_k^I + {}^n Q_k^b + {}^n Q_k^t + \frac{1}{2} \sum_{\forall j} (\| {}^{nn} \underline{\Delta} \| - a) \epsilon_{krs} {}^{nn} f_r^f {}^{nn} \hat{\Delta}_s \quad (2.225)$$

$$[{}^n \dot{X}_k - {}^j \dot{X}_k] + (\| {}^{nn} \underline{\Delta} \| - a) \epsilon_{krs} [{}^n \dot{\theta}_r {}^{nn} \hat{\Delta}_s - {}^j \dot{\theta}_r {}^{nn} \hat{\Delta}_s] = 0$$

where ${}^{nn} f_k^f$ and $\| {}^{nn} \underline{\Delta} \|$ is the frictional force and interparticle distance between neighbouring particle elements (j) adjacent to particle element (i) of the same fiber structure (n) that ensures the no-slip constraint is satisfied such that $\| {}^{nn} \underline{\Delta} \| > 2a$.

The suspension viscosity can be obtained from the average normal tensor $\bar{\sigma}_{xx}$ and shear stress tensor $\bar{\sigma}_{xy}$ given by [293]

$$\bar{\sigma}_{xy} = \left(1 + \frac{5}{2}\vartheta_f\right)\mu\Gamma_{xy}^\infty + \Delta\bar{\sigma}_{xy}, \quad \bar{\sigma}_{xx} = p_0 + \Delta\bar{\sigma}_{xx} \quad (2.226)$$

where p_0 is the suspension pressure, and the fiber volume fraction $\vartheta_f = 4/3\vartheta \pi a^3 N_e N_f$, ϑ is the system volume and $\Delta\bar{\sigma}_{xy}$, and $\Delta\bar{\sigma}_{xx}$ are given as

$$\Delta\bar{\sigma}_{xy} = \frac{1}{\vartheta} \left[\sum_{i=1}^{N_e N_f} {}_i F_x^D {}_i X_y - \frac{1}{2} \sum_{i=1}^{N_e N_f} T_x^D \right], \quad \Delta\bar{\sigma}_{xx} = \frac{1}{\vartheta} \sum_{i=1}^{N_e N_f} {}_i F_x^D {}_i X_x \quad (2.227)$$

${}_i \underline{F}^D$ and ${}_i \underline{T}^D$ can be approximated from eqn. (2.224)-(2.225) assuming $m = l \approx 0$ and considering negligible Brownian disturbance.

Although DNS method is often used in microscale modelling of short fiber suspension due to its ability to incorporate flexural characteristics to the fiber particle, it lacks the ability to accurately model well-defined FSI boundaries such as the fiber-fluid interface. FEM allows for easier modeling of complex geometry and irregular shapes and can be used to simulate a wide variety of microscale level physics in EDAM polymer composite processing. FEM has been adopted for the numerical investigation of mechanisms responsible for micro void formation in current research and would be discussed in later chapters.

Microstructural Characterization of Large Area Additively Manufactured Polymer Composite Bead

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Microstructural characterization of SFRP composites beads is crucial in understanding how the inherent beads microstructure relates to the thermo-mechanical properties and part performance. The study can help us better understand and control manufacturing process conditions to optimum resulting bead microstructure and improve the associated microstructural properties and part performance. Traditional techniques that have been used to analyze the microstructure of polymer composites print beads includes Optical Microscopy, Transmission Electron Microscopy (TEM), Raman Spectroscopy, and Scanning Electron Microscopy (SEM), Optical Microscopy is utilized to observe the overall fiber orientation and distribution within the composite. It is frequently paired with image analysis software to quantify these characteristics. SEM technique provides highly magnified quality 2D images of the composite bead surface, allowing for the detailed examination of fiber-matrix interactions and fracture surfaces. The TEM technique can provide detailed nanoscale images, which are valuable for examining the fiber-matrix interface and identifying any nanoscale defects while Raman Spectroscopy technique is used to investigate the chemical composition and stress distribution within the fibers and matrix. The advent of X-ray micro-computed tomography (μ CT) imaging non-destructive technique has led to higher resolution three-dimensional (3D) visualization and more accurate characterization of the microstructure of polymer composites at the micron scale compared to 2D imaging techniques such as scanning electron microscopy (SEM) [31], [41], [294]. μ CT has been widely used to identify and characterize the microstructures of polymer composites including inherent

micro-constituents' phases and contents and defects. Additionally, μ CT can be used for in-situ real-time monitoring of processes at the micron-scale.

In this chapter, we aim to quantitatively characterize the micro-void content within an EDAM polymer composite bead microstructure with a focus on the relationship between micro-voids and fiber tips within the printed bead. In existing literature, the phenomenon of micro-voids nucleation at fibre tips have only previously been addressed from a qualitative perspective [5], [8]. To this end, the following experimentally examines microstructural formations of 13% carbon fiber filled ABS polymer composite EDAM beads using high resolution 3D X-Ray μ -CT imaging and computational methods for extracting quantifiable details from the μ -CT data.

3.1 Methodology

3.1.1 3D Printing Process

The Baylor University Large Area EDAM system (LAAM) with a print volume of 48" $\times$ 48" $\times$ 6" high was used to produce short fiber polymer composite beads for characterizing microstructural voids in this study. The LAAM system is composed of a Strangpresse Model 19 single-screw extruder (Strangpresse, Youngstown, OH, USA) with three temperature control regions along its length and a nozzle diameter of 3.172 mm. PolyOne CF/ABS (Avient Corporation, Avon Lake, OH, USA) with 13% carbon fiber weight fraction was used as the LAAM feedstock. Pellets were dried in a convection oven at 80°C for twelve hours before the 3D extrusion/deposition processing was performed. Figure 1 (a-d) is a flow-chart that illustrates a typical polymer composite deposition of a single bead on a print bed and post-3D image acquisition and analysis of a cut section taken from a straight printed bead sample which used for our study. The bead sample was sufficiently long to ensure that a quasi-steady extrusion/deposition process

was achieved. The EDAM internal nozzle geometry and printing process parameters appear in Figure 1(a) and Table 3.1.

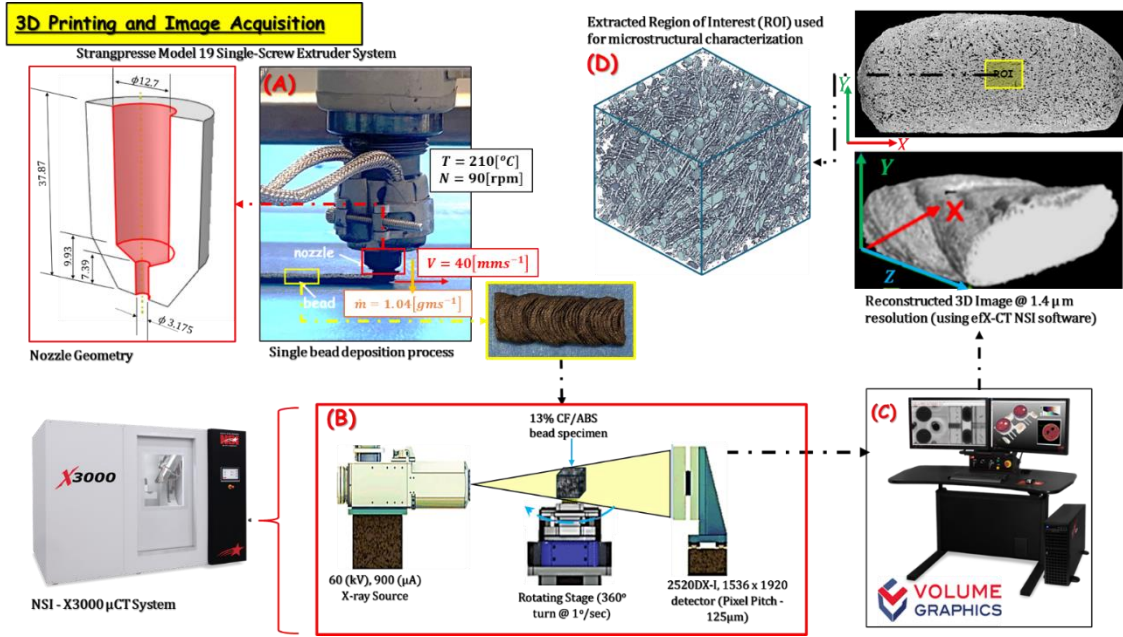


Figure 3.1: Flow-chart illustrating typical LAAM bead printing and 3D μ-CT image acquisition and analysis of a section from the printed bead specimen.

Table 3.1: LAAM bead print process parameters

Printing Process Parameters	Units	Value
Temperature	[°C]	210
Screw speed	[rpm]	90
Extruder mass flow rate	[gm/s]	1.04
Nozzle translation speed	[cm/min]	240
Nozzle diameter	[mm]	3.17
Nozzle height	[mm]	1.20

3.1.2 μ-CT Image Acquisition Technique

The North Star Imaging X3000, X-RAY μCT system (North Star Imaging, Rogers, MN, USA) was used to scan the CF/ABS deposited bead sample. μCT scans were performed at a resolution (voxel size) of 1.7 microns using an X-ray source at 60 kV and 900 μA to provide adequate contrast between the various constituent phases that compose the bead specimen. The sample was rotated 360 degrees in 1-degree increments, resulting in 2400 projections. The detector

captured the transmitted X-ray signals, obtaining 2D attenuation distribution data. The acquired μ CT scan data was then reconstructed using efX-CT software (North Star Imaging, Minnesota, USA). During reconstruction, an outlier median filter preprocessing technique was used to reduce noise and improve the detection of boundaries between microstructural features such as voids and fibers within the ABS matrix.

3.1.3 μ -CT Image Data Post Processing

μ -CT X-ray imaging techniques were used to generate 3D grayscale voxelated data based on density for a cube-shaped specimen with a side length of 1.2mm obtained from the CF/ABS bead where each voxel has a side length of 1.4 μ m. Unless stated otherwise, all post processing operations presented here are performed using built-in functions from MATLAB's (Mathworks, Natick, MA, USA) 3D image processing toolbox. For each μ -CT dataset, grayscale data is classified into three (3) groups using the '*imsegkmeans3*' statistical function to obtain binary data for each segment representing the different constituent phases that include ABS matrix, micro-void (air), and fiber inclusions as shown in the typical sample illustrated by Seq. #1 & #2 (cf. Figure 3.2). These images show a typical region of material from our LAAM bead appearing here for illustration of the imaging post processing analysis. The '*bwlabeln*' function is used to identify individual fiber or fiber clusters and void features by determining connected voxels having the same phase within each segment (Figure 3.2, Seq. #3). Separation of the fiber clusters into individual pristine fibers is achieved by slightly adjusting the cluster's intensity value and filtering the data using the Hessian-based Frangi-Vesselness '*fibermetric*' function. The transformation of a typical fiber cluster into separate fiber vessels after grouping and filtering operation is depicted by Seq. #4a (Figure 3.3). Subsequently, a skeletonization operation is performed to extract the individual vessel ribs using the '*bwskel*' function. Unfortunately, the filtering operation erodes the

cluster data which results in the splitting of some ideally pristine fiber skeletons into broken fragments as can be seen in the resulting fragmented skeletal framework after Seq. #4b (cf. Figure 3.3). To resolve this, a custom algorithm is implemented that identifies and stitches together line fragments belonging to unique pristine fibers by matching orientation data of fragment pairs along their centroidal axes within proximity to each other and connecting missing voxels of nearby ends in the predetermined direction.

The resulting skeletal framework of pristine fibers after stitching end extension operation is shown in the image after Seq. #4c (cf. Figure 3.3). The stitching algorithm is limited by the efficacy of the built-in skeletonization function in obtaining a sufficiently smooth and central skeletal framework. Region property information including the centroid, orientation, and geometry data for individual line segments is obtained using the “*regionprops3*” function. After a successful stitching operation is complete, the endpoints of pristine fiber skeletons are obtained using the “*bwmorph3*” morphological operation function which are depicted by blue markers in the image after Seq. #4c (cf. Figure 3.3). We found that grayscale data erosion due to the filtering operation often results in shorter pristine fiber skeleton ribs that necessitated the development of an algorithm that extends the skeleton terminals along its principal direction to the edge of the fiber feature. Fiber end regions for fibers with clearly defined tips are defined by a 5-voxel unit radius around the end points of individual fiber skeleton within the fiber grayscale dataset. The fiber tip regions are depicted by the green regions in the superposed volumetric plot of the fiber cluster (gray) overlaid on its skeleton (red) after Seq. #4d (cf. Figure 3.3). For irregular fiber features having no unique tips and having a low aspect ratio (typically less than 3), the entire fragment is considered a tip.

By juxtaposing individual void features with fiber features and individual void features with fiber tip voxels through appropriate indexing operation, the fraction of voids by volume isolated within the ABS matrix and those in contact with fiber tips are, respectively, determined. The probability of a pristine fiber feature extending beyond the volumetric bounds of the cut-specimen is accounted for by excluding fiber tip regions within 5-voxel units of the volume bounding surface. Figure 3.3 shows the typical fiber tip regions (green) in a ROI with random pristine fiber samples (gray) after fiber tip exclusion zone definition (Seq. #5), and a volumetric superposed plot highlighting all relevant interest features with unique colormaps after the feature identification through indexing operation (Seq. #6) including pristine fiber regions (gray) with their associated fiber tips (green) and interacting tip voids (blue) together with a few samples of voids isolated within the matrix (red). The regional mean fiber orientation is obtained by averaging orientation data of fragmented segments of pristine fiber skeletons within the region and the splitting operation is achieved using the built-in ‘*spectralcluster*’ function.

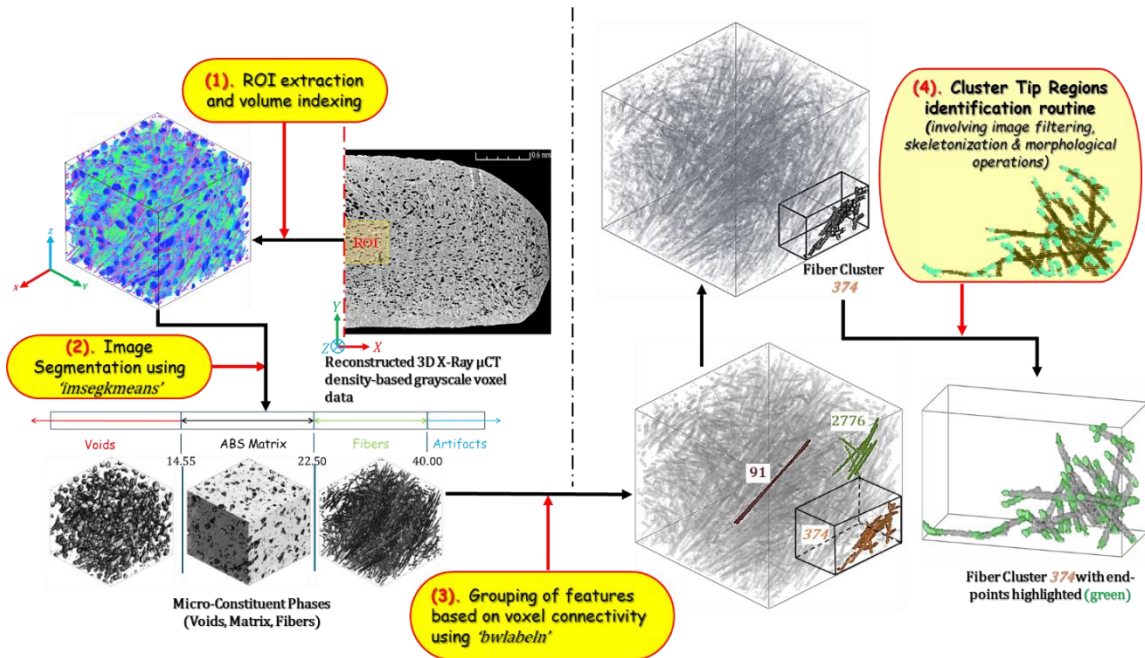


Figure 3.2: Flow-chart illustrating ROI extraction, grayscale thresholding based binary image segmentation of the bead sample, feature identification through grouping and filtering operation, and fiber cluster separation and tip region identification operation.

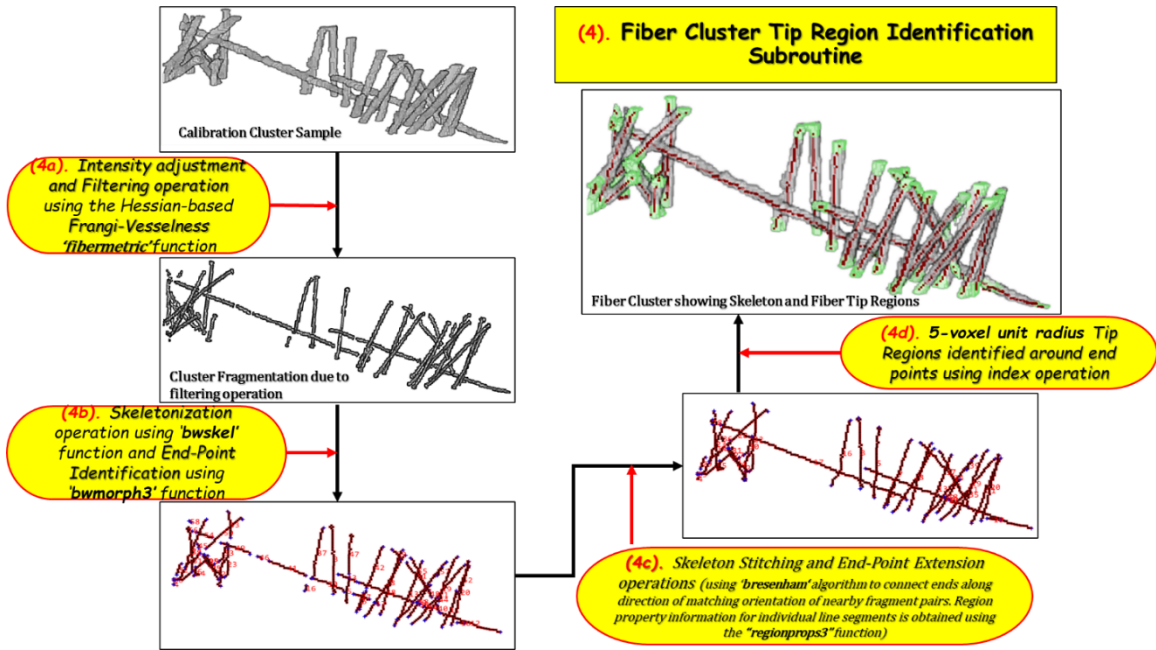


Figure 3.3: Flow-chart detailing the fiber cluster separation subroutine operation (cf. Figure 3.2, Seq. #4) including filtering, skeletonization, stitching and end-point extension and fiber tip region identification operations.

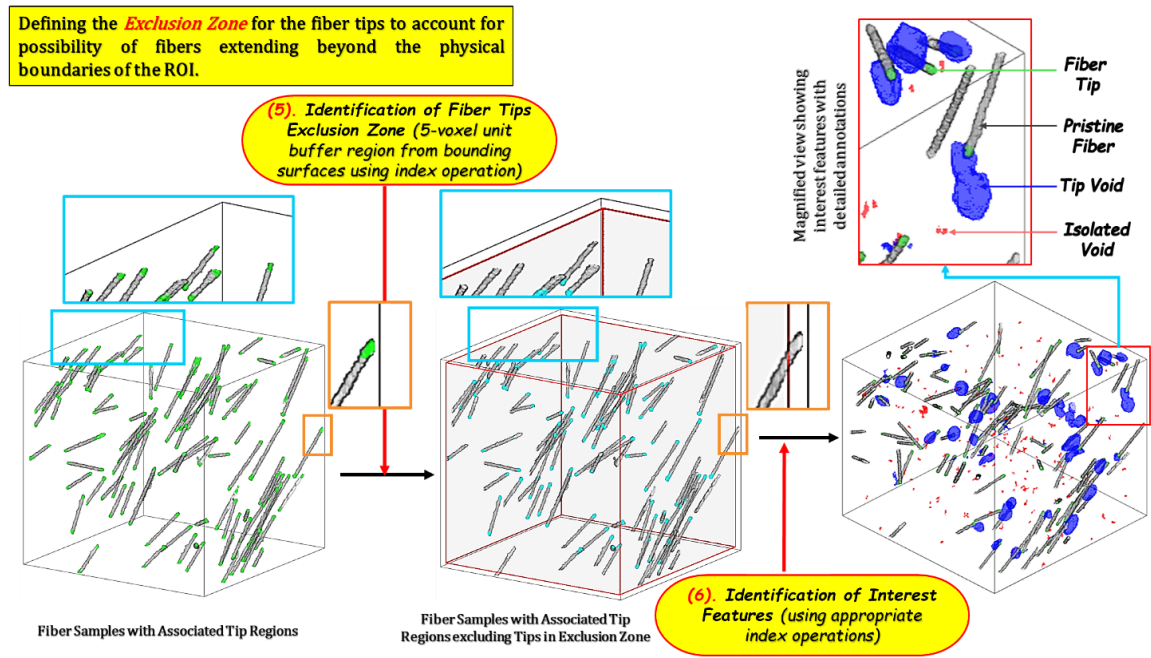


Figure 3.4: Flow-chart showing the fiber tip exclusion zone identification operation and the identification of interest features through indexing operation.

Figure 3.5a shows the resulting ROI highlighting relevant interest features contained within the volume after completion of the image post-processing process including the pristine fiber samples (gray), the voids interacting with fiber tips (blue) and the voids isolated within the matrix (red). For better visualization, Figure 3.5c shows a magnified cut section view extracted from the central region of the ROI (cf. Figure 3.5b) showing the relevant interest features within the volume.

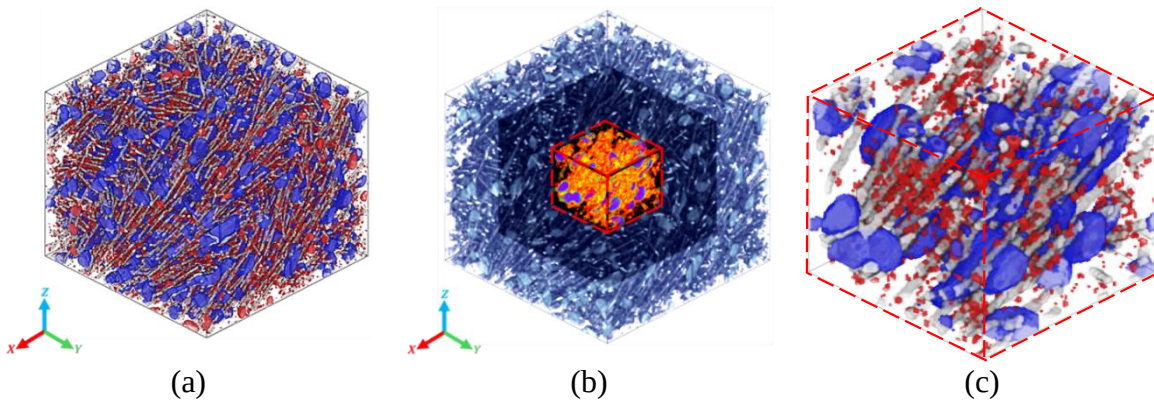


Figure 3.5: (a) ROI volume showing relevant interest features including pristine fiber samples (gray), micro-voids interacting with fiber tips (blue) and the micro-voids isolated within the matrix (red) (b) ROI volume highlighting a central region (c) magnified view of the central region extracted from the ROI volume (ROI Cubic Envelope Size: 0.35mm x 0.35mm x 0.35mm).

Of particular interest are the different classifications of the micro-void contained within the ROI volume determined from the image post-processing analysis. Figure 3.6a shows the volume content of homogenous micro-voids isolated within the matrix, while Figure 3.6b shows the content of heterogenous micro-voids touching fiber tips and Figure 3.6c shows the heterogenous micro-void content touching fiber but not fiber tips.

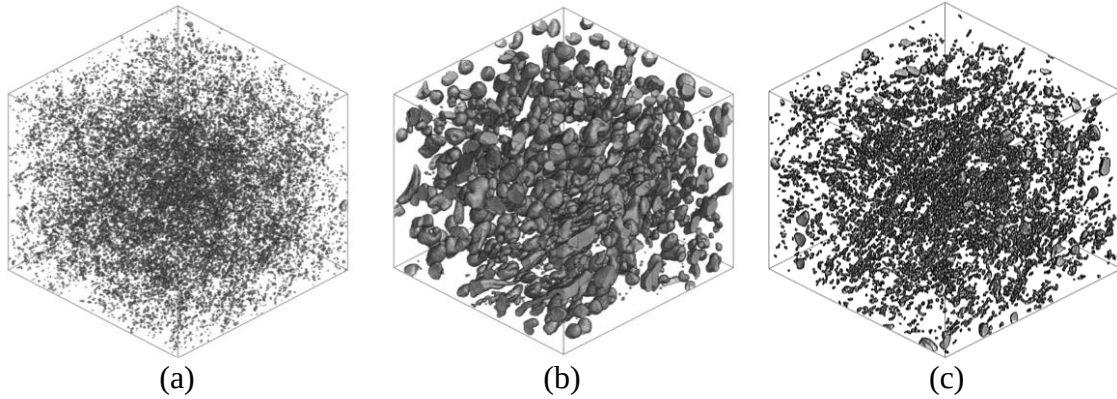


Figure 3.6: ROI volume showing (a) homogenous micro-voids isolated within the polymer matrix (b) heterogenous micro-voids with fiber tip interaction (b) heterogenous micro-voids without fiber tip interaction (Cubic Envelope Size:0.35mm x 0.35mm x 0.35mm)

Figure 3.7 presents tomography section slices of a typical microstructural region along the primary mid-planes of the 3D grayscale voxelated volume that shows the segmented microstructural features of interest including the micro-void regions in contact with a fiber tip (orange), the fiber regions (dark red), transition zones between two phases (bright greenish yellow) and the micro-void regions without fiber tip interaction (light blue spots).

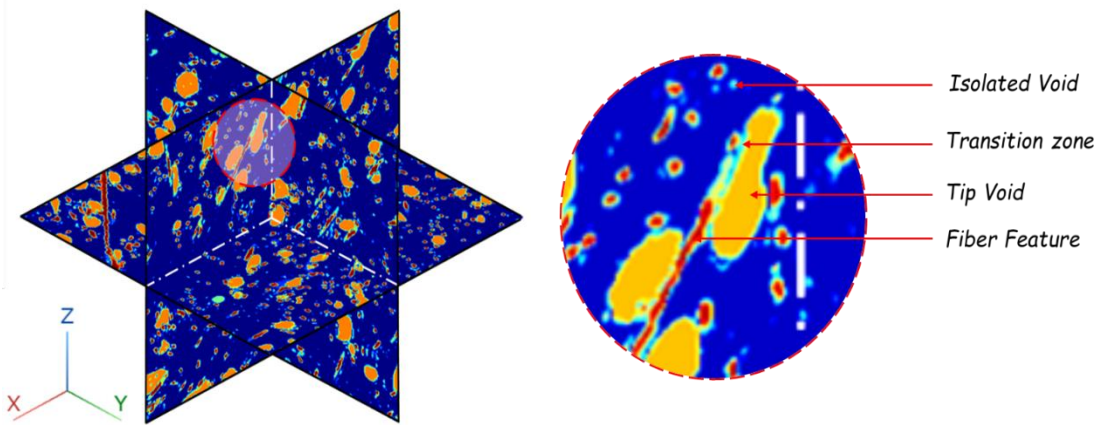


Figure 3.7: Volumetric tomography slice at the mid-planes of the 3D voxelated grayscale data.

3.1.4 μ -CT Data Analysis

The microstructural features of the 13% CF/ABS polymer composite bead specimen obtained from the 3D μ -CT voxelated data post processing technique described above were analyzed for four (4) regions of interest (ROIs) within the LAAM printed bead as shown in Figure

3.8. The ROIs include ROI-I near the base of the bead near the build surface, ROI-II at the beads center, ROI-III near the free surface at the edge of the bead, and ROI-IV near the upper surface of the bead. These ROIs were chosen to provide a representative sampling of the bead cross section in regions that appear to have variations in microstructure. Each cubic ROI volume has a side length of 0.35mm and consists of 250 equal sized voxel cubes per side with each voxel unit having side length of 1.4 μ m, yielding a total of 15,625,000 voxels per ROI.

The microstructure within each of the four (4) ROIs is characterized by nine (9) metrics with regards to micro-void formation which includes (1) the volume fraction of each constituent phase (i.e., micro-void phase, ϑ_v , fiber inclusions, ϑ_f , and polymer matrix, ϑ_m) (2) the fraction of micro-voids isolated within the polymer matrix ϑ_{vm} (or conversely, the fraction of those in contact with fibers $\vartheta_{vf} = 1 - \vartheta_{vm}$) (3) the fraction of micro-voids in contact with fiber tips ϑ_{vt} (4) the fraction of fibers having a tip in contact with a micro-void ϑ_{ft} (5) the average diameter of the micro-voids in contact with a fiber d_{vf} (6) the average diameter of the micro-voids isolated within the matrix d_{vm} (7) the average sphericity of micro-voids in contact with a fiber, Φ_{vf} (8) the average sphericity of the micro-voids isolated within the matrix Φ_{vm} and (9) the principal components of the region-averaged fiber orientation tensor a_{ij} .

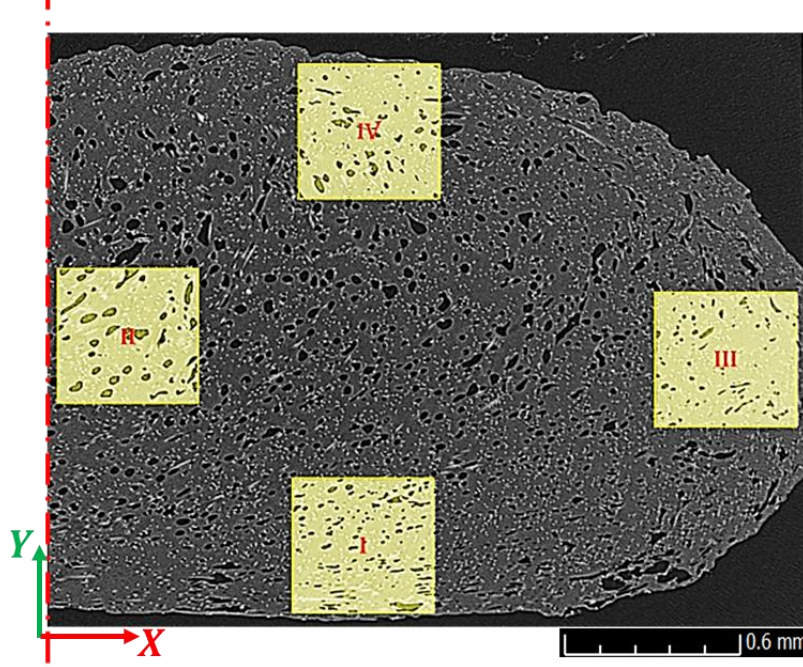


Figure 3.8: Regions of interest (ROIs) within the CF/ABS specimen from a polymer composite bead manufactured from Baylor's LAAM system.

3.1.5 Microstructure Assessment Metrics

The volume fraction of the p -th constituent phase ϑ_p is simply the ratio of the volume of the p -th phase V_p to the overall ROI volume V written as

$$\vartheta_p = V_p/V \quad (3.1)$$

where subscript (p) is either the void (v), fiber (f), or matrix (m) phase. The volume fractions of the constituent phases should satisfy the conservation requirement (i.e. $\vartheta_m + \vartheta_v + \vartheta_f = 1$). The fraction ϑ_{vf} is defined as the ratio of the volume of micro-voids in contact with fiber V_{vf} to the total micro-void volume within the ROI volume V_v . Likewise, the fraction ϑ_{ft} is defined as the ratio of fiber content with one or both tips in contact with one or more micro-voids V_{ft} to the total fiber content in the ROI V_f . The fractions ϑ_{vf} and ϑ_{ft} are, respectively given as

$$\vartheta_{vf} = V_{vf}/V_v \quad \text{and} \quad \vartheta_{ft} = V_{ft}/V_f \quad (3.2)$$

It follows that the fraction of micro-voids isolated within the polymer matrix is given as $\vartheta_{vm} = 1 - \vartheta_{vf}$. In addition to quantifying the micro-void content, we also consider micro-void characteristics within the ROI volume including the micro-void average diameter d_v and average sphericity Φ . The micro-void equivalent diameter d_v is from the diameter of a sphere with equal volume as the irregular shaped void as illustrated in Figure 3.9 and mathematically given as:

$$d_v = \sqrt[3]{6/\pi V_v} \quad (3.3)$$

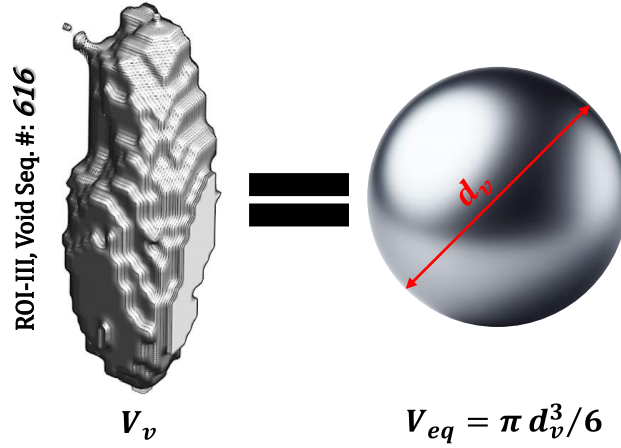


Figure 3.9: Visualization of a representative sphere element (right) with equal volume as an irregular shaped micro-void (left) used to determine equivalent void diameter and sphericity.

The void sphericity Φ is a measure of the irregularity of the void shape and is computed based on the Wadell definition [295] as

$$\Phi = \frac{\sqrt[3]{36\pi V_v^2}}{A_v} \quad (3.4)$$

where A_v and V_v are the 3D voxelated boundary area and volume of each connected void region. The 3D boundary area is computed by isolating individual void features using a binary assignment and summing the total number of facially connected non-unity neighbor voxels to each bounding voxels of the individual void region. Figure 3.10 shows three (3) different representative micro-

void features with different shapes and their respective sphericity values computed using eqn. (3.4).

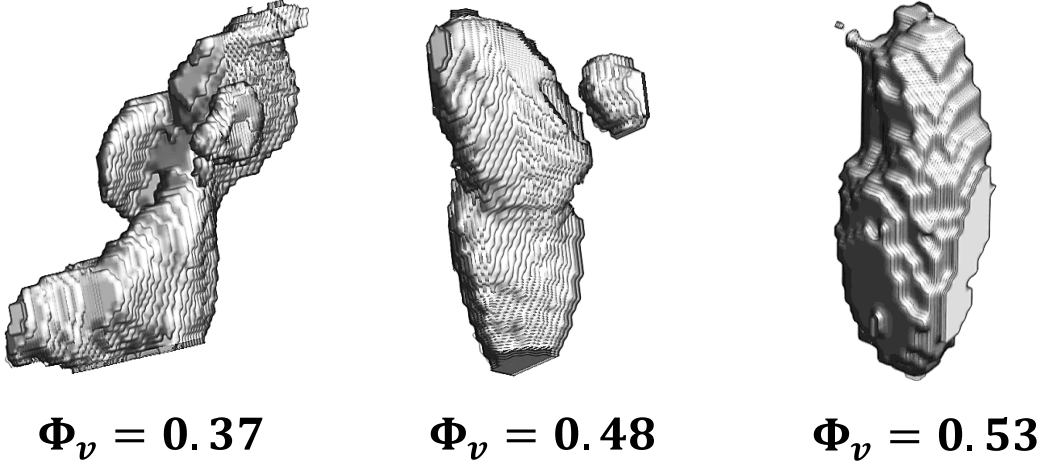


Figure 3.10: Showing different micro-void features and their respective computed sphericity values.

The fiber orientation tensor a_{ij} for an ROI volume is determined from length weighted ensemble average of the dyadic products of individual fiber orientation vector $\underline{p}$ contained within the region volume written as

$$a_{ij} = \frac{1}{n\bar{l}} \sum_{k=1}^n l_k p_i^k p_j^k, \quad \bar{l} = \frac{1}{n} \sum_{k=1}^n l_k \quad (3.5)$$

where l_k is the length of the k^{th} fiber skeleton, p_j^k is the j^{th} orientation vector component of the k^{th} fiber skeleton, and n is the total number of fiber skeletons contained within the ROI volume.

The orientation vector $\underline{p}$ is given as

$$\underline{p} = [\cos \phi \sin \theta \quad \sin \phi \sin \theta \quad \cos \theta]^T \quad (3.6)$$

where the Euler angles ϕ and θ are shown in Figure 3.11. The normalization condition which relates the diagonal components of the orientation tensor requires that $a_{ii} = 1$ where the repeated indices imply summation. The diagonal components of a_{ij} are used to describe the degree of fiber

alignment with any of the orthogonal reference axis. In the current study, the z -axis indicates the print direction, and the y -axis is perpendicular to the print bed. The magnitude of the diagonal tensor components a_{kk} (no summation implied) ranges from 0 to 1, i.e. $(0 \leq a_{kk} \leq 1)$, where a value of 1 indicates complete fiber alignment with the k -th coordinate direction and 0 indicates all fibers lie in a plane normal to the k -th coordinate direction.

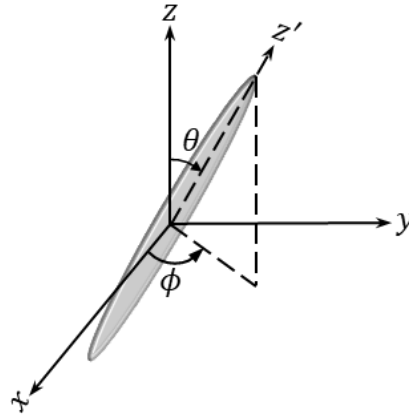


Figure 3.11: Showing the orientation of a single fiber with respect to the reference coordinate directions.

3.2 Results & Discussion

Figure 3.12 shows microstructural features for the four (4) ROIs that result from the segmentation procedure described above in the methodology section which includes fibers (gray), voids touching fibers (blue), and voids not contacting a fiber tip (red). By visual inspection, it is apparent in Figure 3.12 there is a relatively high content of micro-voids touching fibers (blue) as compared to micro-voids not in contact with fiber tips (red). The fibers in each ROI can also be seen to be more aligned with the z -direction (print direction) and to a greater degree in ROI-III (cf. Figure 3.12c) with more densely packed and highly aligned pristine fiber striations as compared to other component directions. Moreover, the sizes of voids touching fiber tips (blue) can be seen

to be relatively larger than that of other voids. We likewise observe more irregular and elongated shaped voids in ROI-III as compared to the other ROIs which have more spherical void shapes.

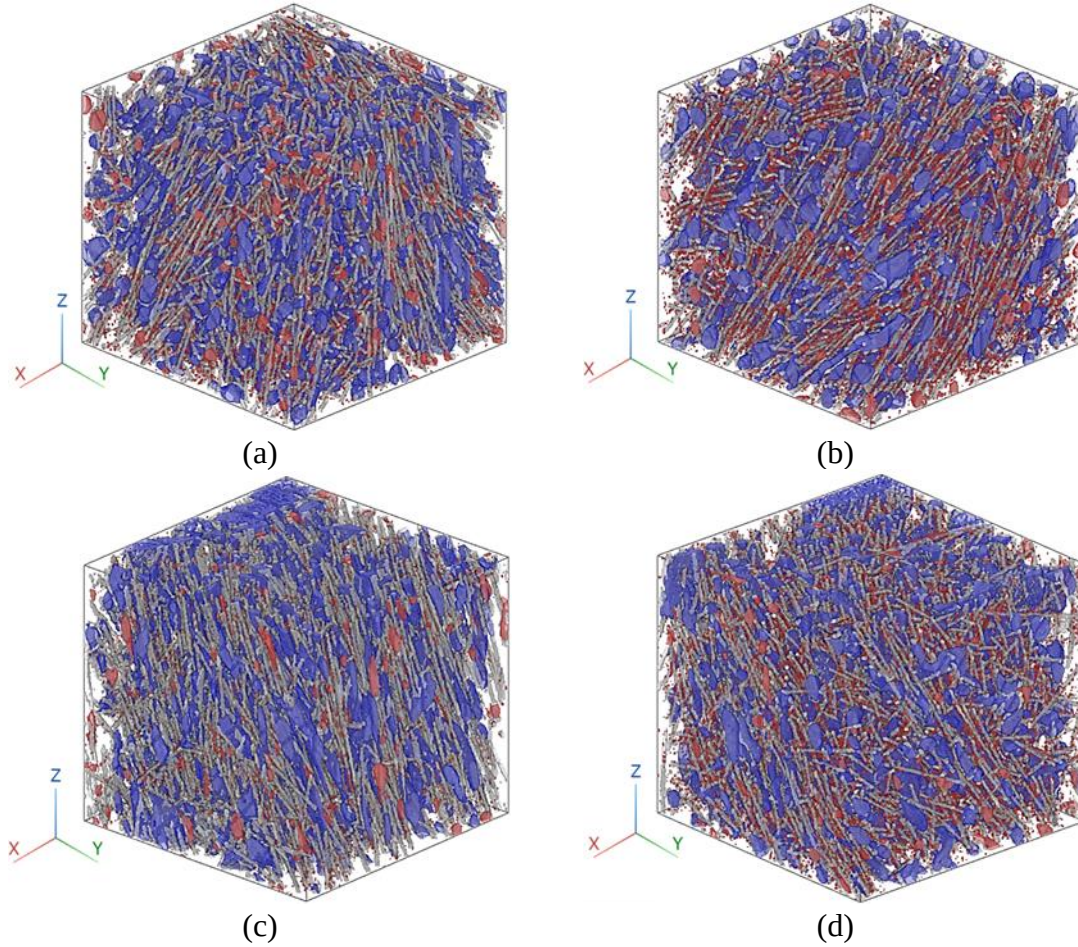


Figure 3.12: Segmented microstructural 3D image highlighting fiber features (gray), void with fiber tip interaction (blue), and voids without fiber tip interaction (red) for the different regions of interest of the CF/ABS bead (a) ROI-I (b) ROI-II (c) ROI-III (d) ROI-IV. (Cubic Envelope Size: 0.35mm x 0.35mm x 0.35mm).

Table 3.2 contains values of the metrics defined above for assessing the microstructural features in each ROI. Calculated results reveal an average micro-void volume fraction ϑ_v near 11% with a standard deviation for ϑ_v less than 1% across all four ROIs. Within these four ROIs, the highest ϑ_v recorded is in ROI-II near the bead center while the lowest ϑ_v is in ROI-III near the edge of the bead. Most notable is that more than 89% of the micro-void volume (see e.g., ϑ_{vt} in Table 3.2) represents micro-voids that are in contact with a fiber tip in all four ROIs with

as high as $\vartheta_{vt} = 95.7\%$ in ROI-III near the bead edge. In addition, the percentage of micro-void isolated within the matrix phase ϑ_{vm} is seen to increase with the overall void content in each ROI. The fraction of fiber skeletons having one or both tips in contact with one or more micro-voids (designated as ϑ_{ft}) were on average observed to be higher in ROI-II and IV, nearing $\sim 50\%$.

Table 3.2: Volume fractions of the microstructural features for the various regions of interest (ROIs) within the 13% CF/ABS EDAM printed bead.

Symbol	Definition	ROI-I	ROI-II	ROI-III	ROI-IV
ϑ_v (%)	Void volume fraction	10.70	12.27	10.06	11.17
ϑ_f (%)	Fiber volume fraction	6.96	6.65	7.25	7.53
ϑ_{vm} (%)	Fraction of voids isolated in matrix	2.67	3.92	1.34	3.39
ϑ_{vt} (%)	Fraction of voids touching fiber tip(s)	90.17	89.68	95.70	91.39
ϑ_{ft} (%)	Fraction of fibers skeleton with tip voids	37.33	48.00	29.27	51.62

As may be expected, the average equivalent diameter of the voids contacting fiber(s) d_{vf} , were seen to be higher than the average equivalent diameter of micro-voids isolated within the matrix, d_{vm} (cf. Table 3.3). The isolated micro-voids within the matrix on average had an equivalent diameter of $d_{vm} = 3.4 \mu m$ with a standard deviation less than $0.4 \mu m$ across all four ROIs. Alternatively, the equivalent diameter of the micro-voids in contact with fibers was on average seen to be higher in ROI-III - near the edge of the bead ($d_{vf} = 39.3 \mu m$) followed by ROI-IV near the top surface of the bead ($d_{vf} = 35 \mu m$) compared to the average equivalent diameter in other ROIs ($d_{vf} \approx 30 \mu m$). Figure 3.13a-d shows the post-processed result of the heterogenous micro-voids in contact with fiber tips for the various ROI volumes. Evidently, ROI-III (cf. Figure 3.13c) is seen to have larger and more elongated micro-voids than other ROI's.

The distribution of equivalent diameter of micro-voids in contact with fibers in the various ROIs appear in Figure 3.14 along with fitted parameters for various 2-parameter probability distribution functions (pdf). In ROI-I (cf. Figure 3.14a) we found that the probability distribution of d_{vf} is best represented by the Gamma pdf with a shape parameter $\alpha = 4.41$, and a scale

parameter $\beta = 6.22$. Alternatively, the distributions in ROI-II & ROI-IV for d_{vf} can best be fitted to a Weibull pdf (cf. Figure 3.14b & d) having a scale parameter of $\alpha = 32.27$ and a shape parameter $\beta = 2.71$ for ROI-II, and a scale parameter of $\alpha = 35.91$ with a shape parameter of $\beta = 2.07$ for ROI-IV. However, in ROI-III with larger sized voids, the distribution is best represented by a Lognormal pdf (cf. Figure 3.14c) having a location parameter $\alpha = 3.47$ and a scale parameter $\beta = 0.52$. The distributions of d_{vf} in ROI-I, II, & IV are seen to peak near the mean value from either extremity of the histogram, however, the histogram of d_{vf} in ROI-III is observed to skew to the right with larger void sizes.

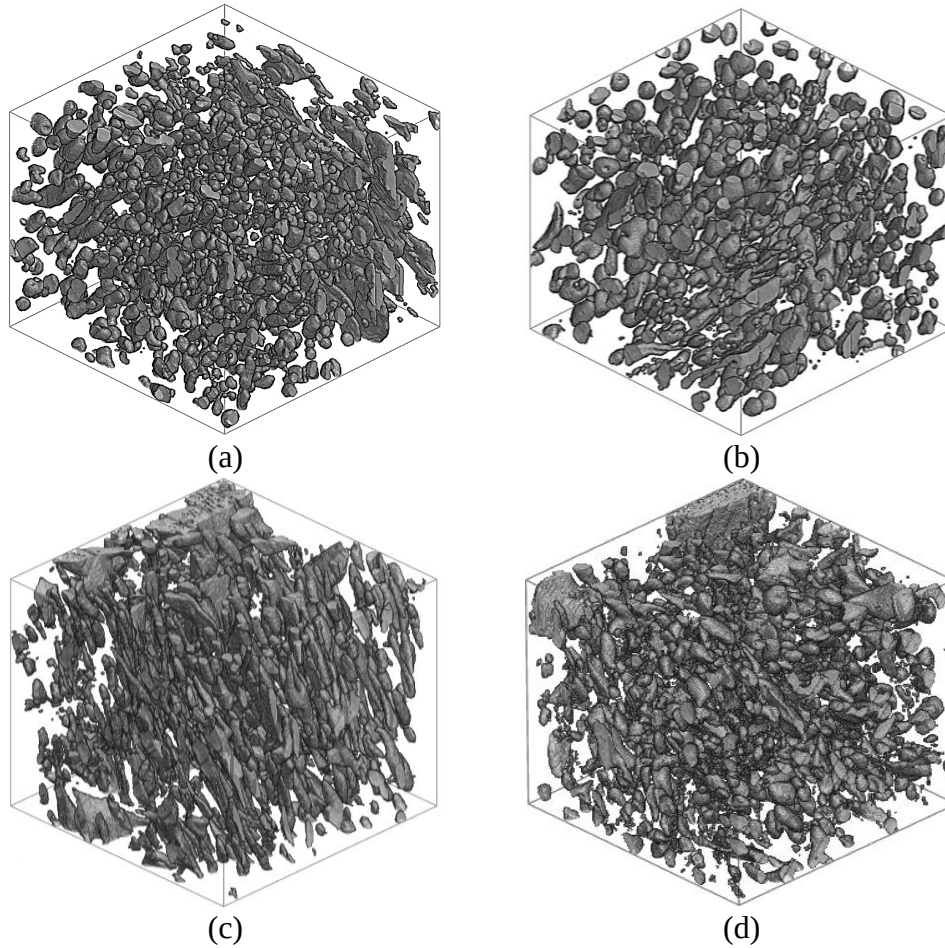


Figure 3.13: Micro-voids with fiber tip interaction for the different regions of interest of the CF/ABS bead (a) ROI-I (b) ROI-II (c) ROI-III (d) ROI-IV. (Cubic Envelope Size: 0.35mm x 0.35mm).

Table 3.3: Average diameter of the microstructural voids features with and without fiber interaction across all four (4) ROIs.

Symbol	Definition	ROI-I	ROI-II	ROI-III	ROI-IV
$d_{vm} (\mu m)$	Average diameter of voids isolated in matrix	3.83	3.24	3.59	2.95
$d_{vf} (\mu m)$	Average diameter of voids touching fiber (s)	30.96	29.35	39.26	34.85
pdf	Probability Distribution Function Type	Gamma	Weibull	Lognormal	Weibull
α	Shape Parameter (Location for Lognormal)	4.41	32.27	3.47	35.91
β	Scale Parameter	6.22	2.71	0.52	2.07

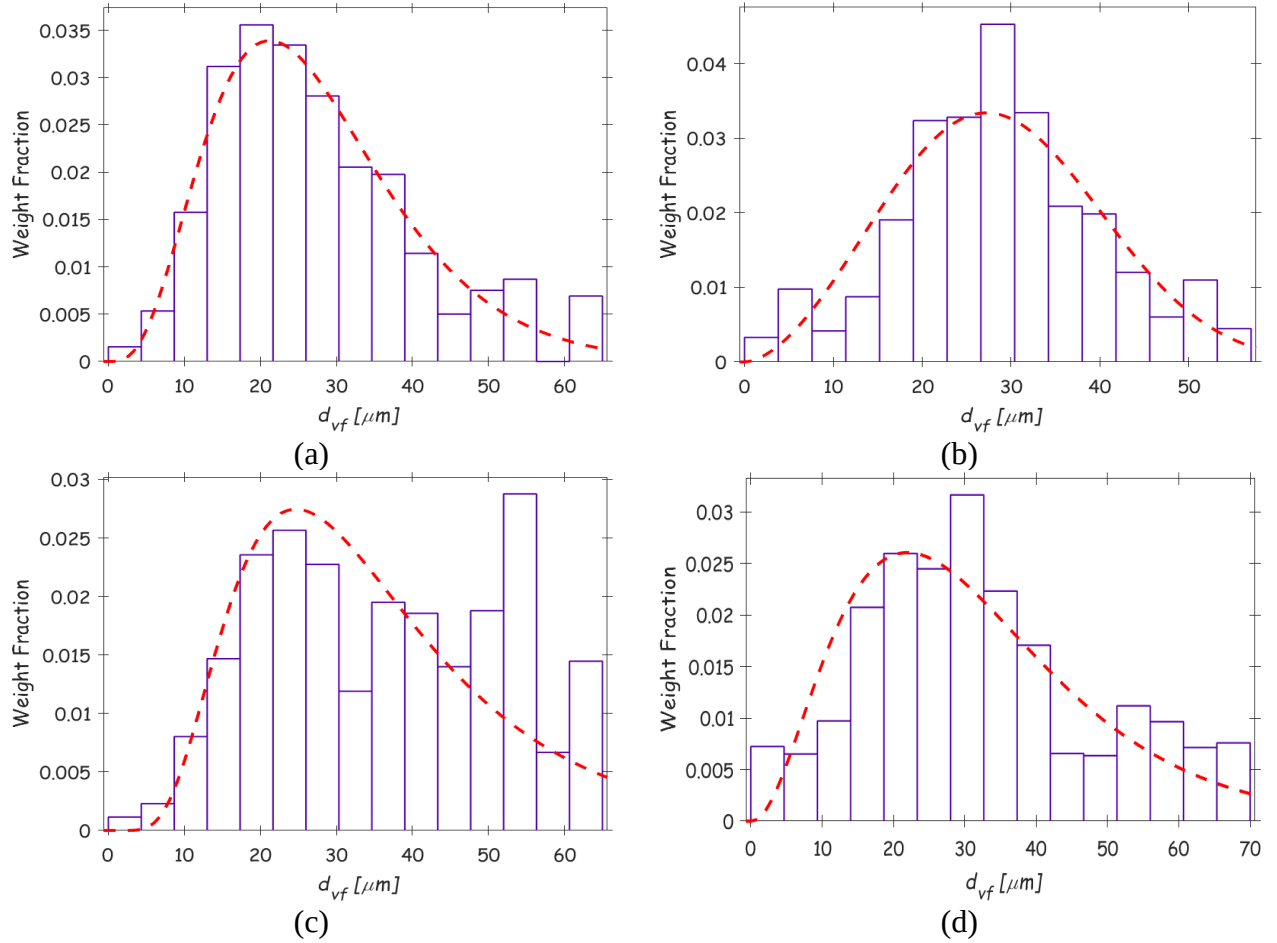


Figure 3.14: Distribution of the average void diameter across all regions of interest (ROI) of the 13% CF/ABS EDAM printed bead. (a) ROI-I (b) ROI-II (c) ROI-III (d) ROI-IV.

The isolated micro-voids have a higher overall mean sphericity value $\Phi_{vm} = 0.735$ with a standard deviation less than 0.01 compared to the overall mean sphericity value for the micro-voids in contact with fibers, $\Phi_{vf} = 0.6$ with a standard deviation less than 0.02. The distribution of the sphericity for micro-voids in contact with fibers Φ_{vf} can be represented well with a Weibull

probability distribution function in all four ROIs of the bead where all peaks are near the mean value as shown in Figure 3.15a-d. The parameters of the Weibull pdfs for the various ROIs appear in Table 3.4. Overall, the pdfs indicate that the bulk of the sphericity for micro-voids in contact with fibers are centered between about 0.62-0.67 across all ROIs.

Table 3.4: Average sphericity of the microstructural voids features with and without fiber interaction across all ROIs.

Symbol	Definition	ROI-I	ROI-II	ROI-III	ROI-IV
Φ_{vm}	Average sphericity of voids isolated in matrix	0.73	0.73	0.74	0.74
Φ_{vf}	Average sphericity of voids touching fiber (s)	0.58	0.60	0.59	0.62
α	Weibull pdf Scale Parameter	0.62	0.65	0.64	0.67
β	Weibull pdf Shape Parameter	5.51	6.18	5.27	6.30

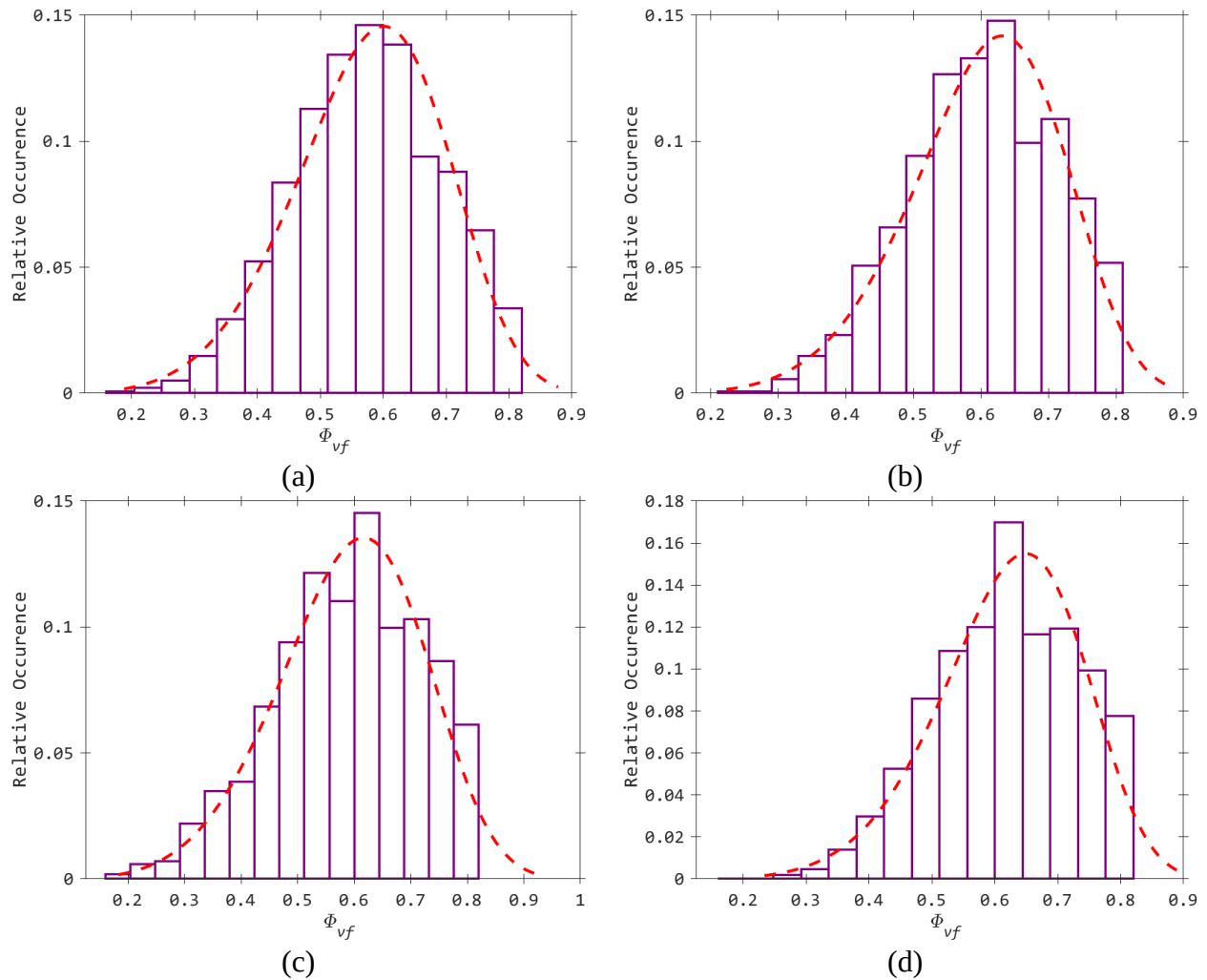


Figure 3.15: Distribution of the average void sphericity across all ROIs of the 13% CF/ABS EDAM printed bead. (a) ROI-I (b) ROI-II (c) ROI-III (d) ROI-IV.

We see from the results of the ensemble average values of the fiber orientation principal components presented in Table 3.5 that there is higher degree of fiber alignment in the z direction close to the bead edges (ROI-III) while the fibers near the bead's center (ROI-II) are more randomly oriented with a significant value of a_{xx} which is in agreement with published data [31], [52]. Figure 3.16a-d shows the post-processed result of segmented fiber microstructural features for the various ROI volumes which provides visualization of the fiber orientation distribution within each ROI volume.

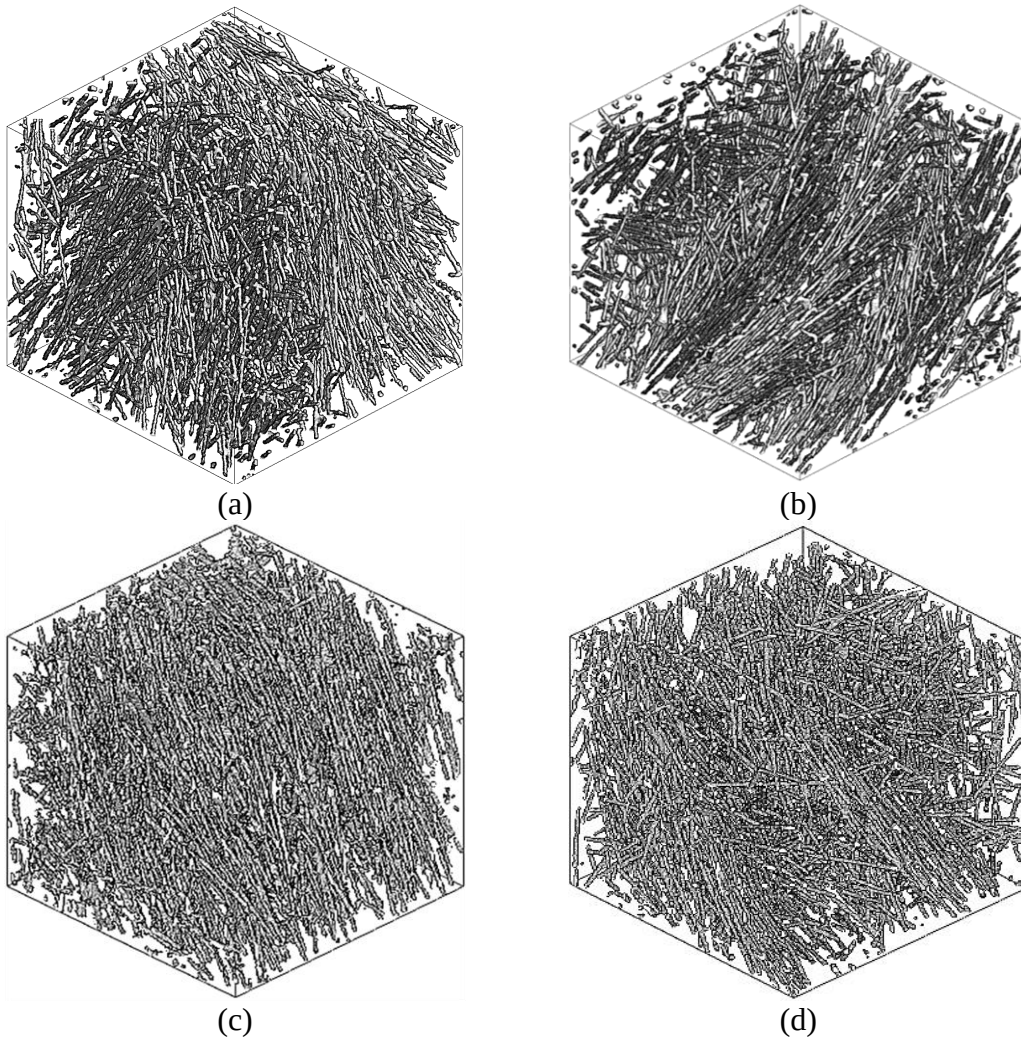


Figure 3.16: Fiber features within the microstructure of the different regions of interest of the CF/ABS bead (a) ROI-I (b) ROI-II (c) ROI-III (d) ROI-IV. (Cubic Envelope Size:0.35mm x 0.35mm x 0.35mm).

Table 3.5: Average values of the fiber orientation principal components in the various ROIs.

	ROI-I	ROI-II	ROI-III	ROI-IV
a_{xx}	0.41	0.32	0.09	0.21
a_{yy}	0.04	0.19	0.10	0.16
a_{zz}	0.55	0.49	0.81	0.63

Computed values of the a_{zz} fiber orientation tensor component for all four (4) ROIs shows that ROI-I near the print bed (cf. Figure 3.17a) shows a more random orientation as evidenced by the a_{zz} components as compared to other ROIs (cf. Figure 3.17c-d). This more random fiber arrangement can be seen in the magnified 3D sub-volume of the central microstructure of ROI-I (cf. Figure 3.18a) which also reveals a relatively low fiber volume fraction. In ROI-II close to the bead's center (cf. Figure 3.17b) and ROI-IV near the beads surface (cf. Figure 3.17d), the fibers are mostly either planarly or randomly oriented, although there is higher fiber alignment in the print direction for ROI-IV as compared to ROI-II. However, in ROI-III (cf. Figure 3.17b), the histogram of a_{zz} is skewed to the right ($a_{zz} \rightarrow 1$) that indicates most of the fibers are highly aligned with the print direction. This high degree of fiber alignment is evident from the magnified 3D central sub-volume of the ROI-IV microstructure where the fibers are seen to be mostly oriented in the z-direction with a considerably high fiber volume fraction as compared to other ROIs (cf. Figure 3.18c).

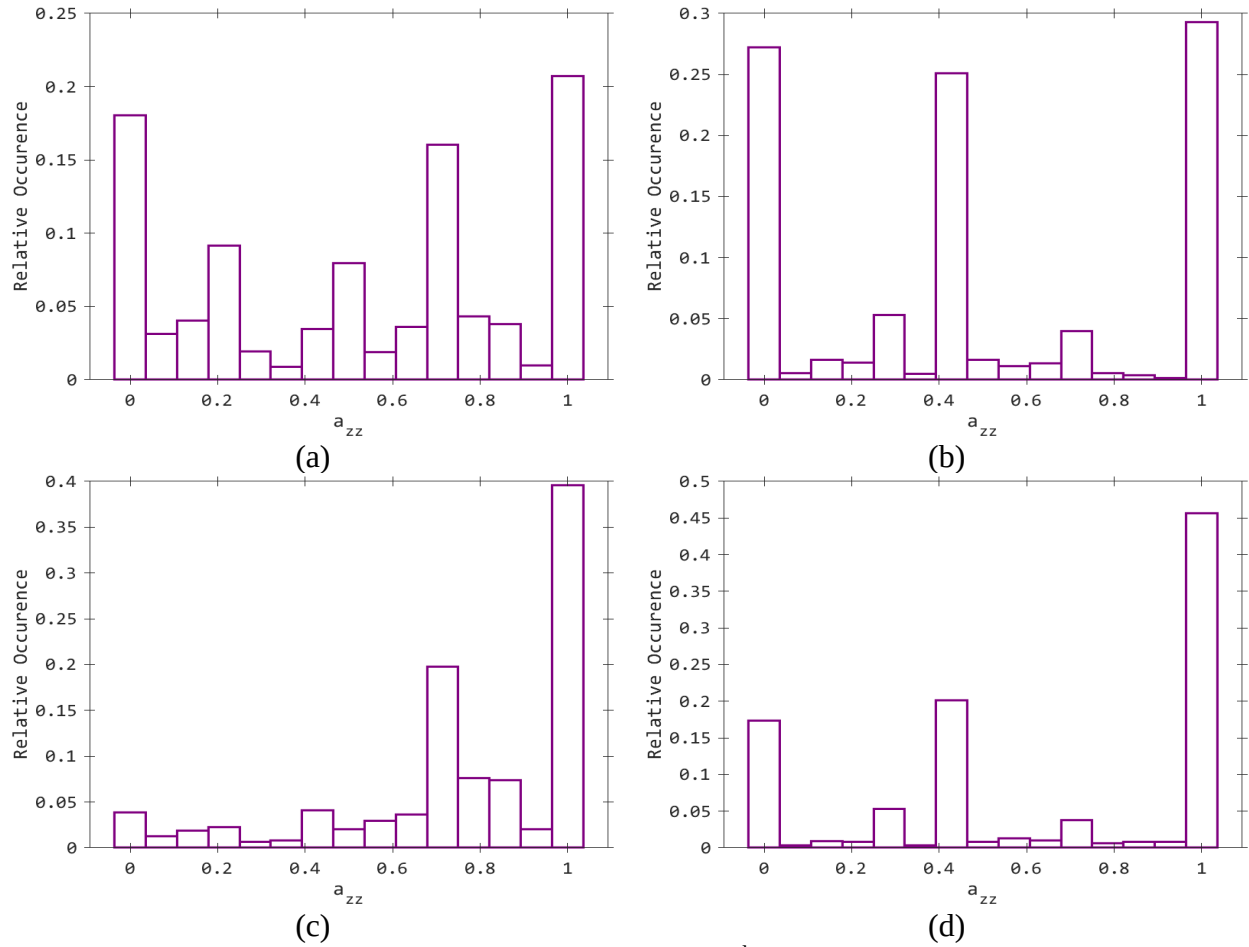


Figure 3.17: Distribution of the A_{zz} component of the 2nd order fiber orientation tensor across all ROIs of the 13% CF/ABS EDAM printed bead. (a) ROI-I (b) ROI-II (c) ROI-III (d) ROI-IV.

Consequently, the associated micro-void content with fiber tip contact is seen to be higher in ROI-III and IV which also has a relatively high degree of fiber alignment in the flow direction (z-direction); as compared to the same in ROI-I and II which both have a higher degree of fiber alignment normal to the flow direction. These results suggest that a high degree of fiber alignment allows for a more compact arrangement of the fibers. Further, higher alignment appears to reduce the propensity for larger micro-void formation between fibers and hence the fiber tips provide more favorable sites for void formation [3]. We observe that fewer isolated voids (red) form between fibers in ROI-III (cf. Figure 3.18c) due to the highly compact fiber arrangement as

compared to that in ROI-I (cf. Figure 3.18a) which has more isolated voids between fibers due to lower packing.

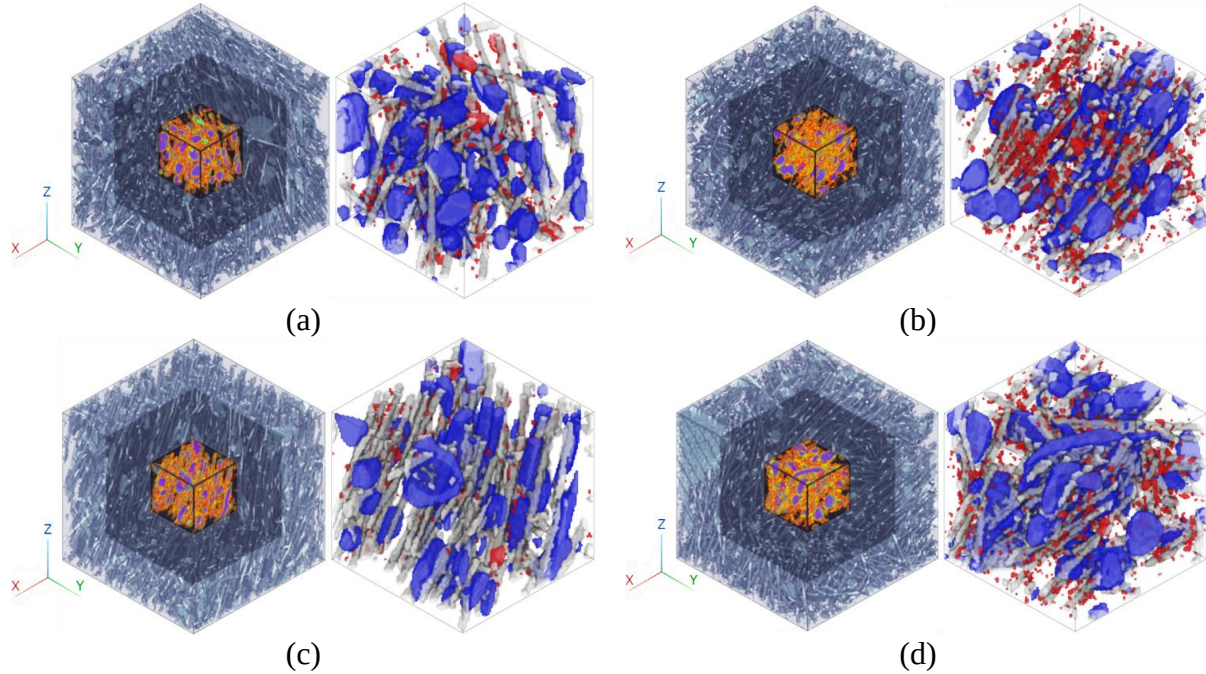


Figure 3.18: Volumetric plot showing extracted sub-volume view of the relevant microstructural features across all regions of interest (ROI) of the CF/ABS bead specimen. (a) ROI-I (b) ROI-II (c) ROI-III (d) ROI-IV.

3.3 Conclusion

In conclusion, microstructural features including micro-void content, micro-void sphericity, and fiber orientation within a 13% CF/ABS bead specimen produced with EDAM has been evaluated using μ -CT scanning and image processing techniques. The results reveal an extremely high percentage of the micro-void contents forms at the ends of the fibers, identified here as tip-voids. On average, the voids that nucleate at the fiber/matrix interface are relatively large compared to those that are isolated within the ABS matrix and are also less spherical in shape. Moreover, regions with a higher degree of fiber alignment with the flow direction have lower interstitial small-sized isolated voids possibly due to increased compactness. However, these regions have a higher micro-void content at the fiber tips due to the increased number of fiber

terminations which was also observation by Telkinalp et al. [3]. As we would see in **Chapter four**, the inherent microstructural characteristics of the bead specimen affect the resulting thermo-mechanical properties and ultimately the part performance. Computational simulation studies that reveal mechanisms potentially responsible for the experimentally observed high volume content of micro-voids and the various factors that may influence their formation are presented in later chapter of this dissertation.

4 CHAPTER FOUR

Numerical Evaluation of the Effective Thermo-Mechanical Properties of Large Scale Additively Manufactured Short-Fiber Reinforced Polymer Composite

In this chapter, we evaluate the effective thermo-mechanical properties of 13% carbon fiber filled ABS (13% CF-ABS) SFRP composite manufactured from typical LAAM technology using the same regions of interest (ROI) presented in the previous chapter (Chapter 3) and assess the impact of micro-structural voids on the resulting effective properties. We employ a finite element based numerical homogenization approach in our study using realistic RVEs developed from actual reconstructed 3D X-Ray μ -CT voxelated grayscale images of a 13% CF-ABS print bead specimen. Microstructural characterization of the printed bead specimen based on binary segmentation of the 3D grayscale voxelated data is performed to identify unique phases and microstructural features within the sample. Finite element analysis is carried out based on the derived realistic RVEs to compute the effective thermo-mechanical properties for different bead's region. To ensure domain continuity across the RVE boundaries, periodic constraints are imposed on opposing boundary entities which ensures effective transfer of load across boundary surfaces. The effective elastic stiffness is derived from the resulting homogenized macro-stresses and macro-strains and for different load cases through a least-square linear regression fitting algorithm and the average thermal expansion coefficient is computed using a procedure similar to Wang [296]. The ETC is computed from homogenized heat flux and temperature gradient obtained from steady-state heat transfer FE analysis based on the Fourier's law. Results of the effective properties obtained from solutions to the numerical based homogenization scheme are compared to results derived from analytical mean-field homogenization approach based on the Mori-Tanaka-Benveniste's formulation. The effects of the porosity on the effective properties are also quantified in the current assessment. By utilizing a discrete minimization approach to obtain characteristic

RVE instance from a given ROI volume with matching microstructural characteristics, the effective thermo-mechanical properties across different regions of the LAAM printed bead specimen is also computed and compared.

4.1 Methodology

In the current study, the effective properties for four (4) ROI across the 13% CF/ABS bead specimen shown in Figure 3.8 are evaluated which includes: (a) ROI-I close to the bed, (b) ROI-II close to the bead's center, (c) ROI-III close to the edge of the bead and (d) ROI-IV close to the top surface of the bead. The dimensions of each ROI's were each $0.35mm \times 0.35mm \times 0.35mm$. The CF/ABS bead was printed using Baylor University Strangpresse Model 19 single-screw extruder LAAM system (Strangpresse, Youngstown, OH, USA). More details on the LAAM printing parameters and operating conditions can be found in [297].

The average isotropic properties of the microconstituents of the 13% CF-ABS SFRP composite used in the homogenization analysis are presented in Table 4.1 [139]. We assume for the microstructural properties of Tourayaca® T300 (Touray Industries, Tokyo, Japan) PAN based carbon fiber, and for the matrix material, we assume properties of Lustran ® 433 ABS (INEOS Olefins & Polymers, London, UK).

Table 4.1: Average isotropic properties of the microconstituents of the 13% CF-ABS SFRP material

	E [GPa]	ν	$\alpha \times 10^{-6}$ [m/m – K]	κ [W/m – K]	ρ [g/cc]	ζ_s [J/kg · K]
Fiber	230.0	0.20	-0.61	3.060	1.76	777.
Matrix	2.55	0.35	90.1	0.175	1.05	1865

In Table 4.1 above, E is the elastic modulus, ν is the Poisson ratio, κ is the thermal conductivity, ρ is the density and ζ_s is the specific heat capacity.

4.1.1 Numerical FEA Homogenization Method

FEA model development of different RVE's were generated from reconstructed 3D X-ray μ -CT voxel-based radiographs of the different ROIs. Binary segmentation of the ROI volume into the three (3) microstructural constituents (matrix, fiber and voids) is achieved via grayscale data thresholding with detailed procedures provided in [297]. Sufficient image resolution that accurately captures the inherent microstructural features is achieved by selecting a voxel cube with side length of $1.4\mu m$ which yields a total of 250 voxels per length or 15,625,000 voxels per ROI. More details on the X-ray μ -CT image acquisition and analysis technique can be found in [297]. The FEA models were generated directly from the scripting interface of Abaqus/Standard (Abaqus 2023, Simulia, Dassault Systemes, Waltham, Massachusetts) using the voxel-data of the RVE which are directly imported as 3D solid 8-node fully integrated iso-parametric continuum brick elements (C3D8), with separate element sets created for the different segmented microstructural constituents. For the heat transfer analysis, diffusive-C3D8 elements (i.e. DC3D8) are used. Relevant material property definitions for the individual microstructural phases are also created and assigned to their respective material sections through the section assignment input syntax. Figure 4.1a&b shows a sample RVE block extracted from the ROI closer to the bead center (i.e. ROI-II) and highlighting the different microstructural phases including fibers (gray), micro-voids (red), and the ABS matrix (transparent volume).

Figure 4.1c shows the resulting FEA model created from directly importing the segmented voxelated data of the RVE into the Abaqus GUI and highlighting the different sections. Figure 4.1d - f shows the individual element sets of the different microstructural phases including the ABS matrix (cf. Figure 4.1d), the inherent micro-voids (cf. Figure 4.1e) and the fiber reinforcements (Figure 4.1f). The principal investigation compares results for three (3) different

RVE sizes using ROI-II as a case study. The smallest sized RVE (RVE-I) has a cube side length of $70\mu\text{m}$ with 125,000 elements and 125 RVE realizations (cf. Figure 4.2a), while the mid-sized RVE (RVE-II) has a side-length of $116.2\mu\text{m}$ with 571,787 elements and 27 RVE realizations (cf. Figure 4.2b). The largest RVE partitioning (RVE-III) has side-length of $175\mu\text{m}$, a total of 1,953,125 elements and 8 RVE realizations (cf. Figure 4.2c). Complete adhesion between the filler and matrix constituent is assumed. In all cases, the element side length equals the length of the voxel cube of $1.4\mu\text{m}$ which is one-fifth ($1/5$) the average fiber diameter of $7.0\mu\text{m}$.

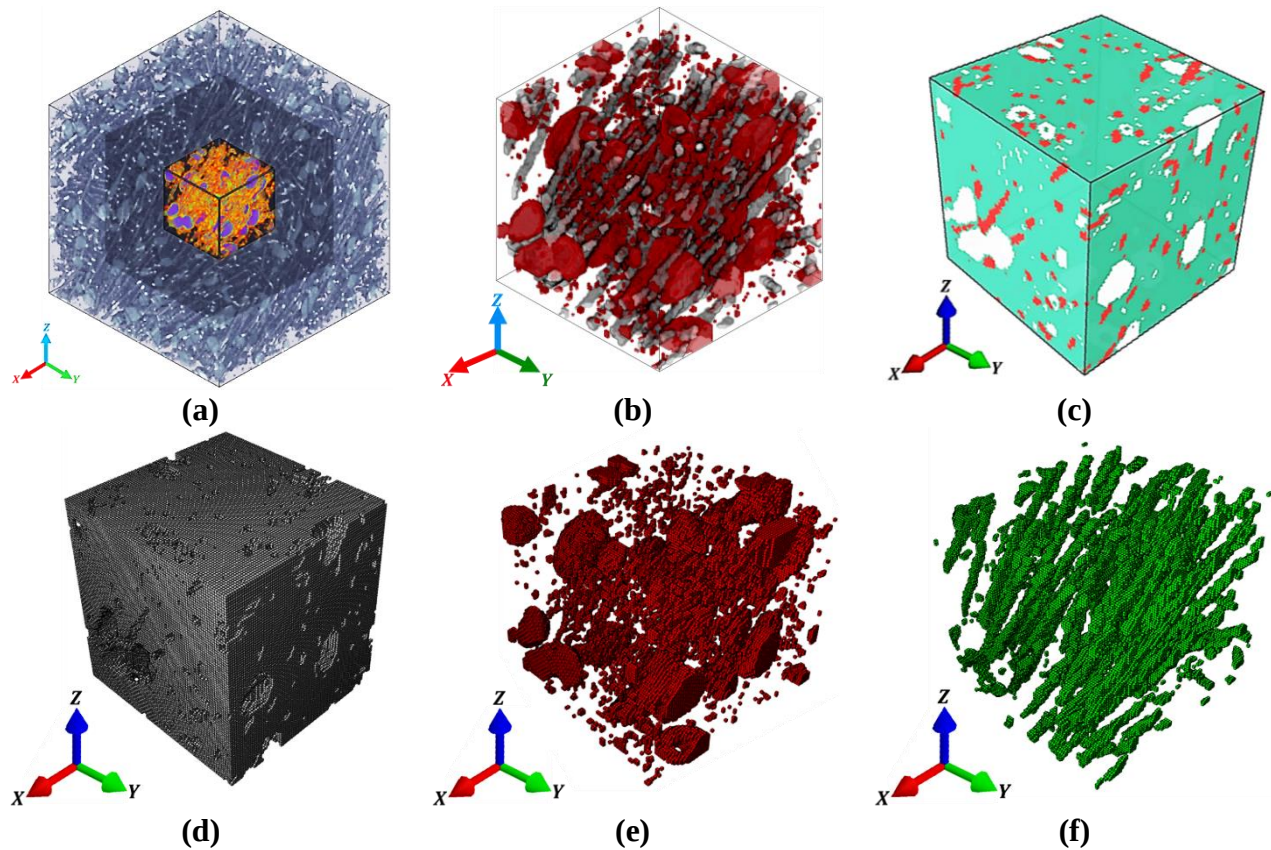


Figure 4.1: (a) Reconstructed μCT 3D-scans of ROI-II highlighting mid-block RVE volume (b) magnified view of the mid-block RVE showing segmented micro-constituents, (c) FEA model imported from μCT voxel data of the RVE, (d) ABS matrix FEA elements (e) micro-voids FEA elements and (f) fiber reinforcement FEA elements.

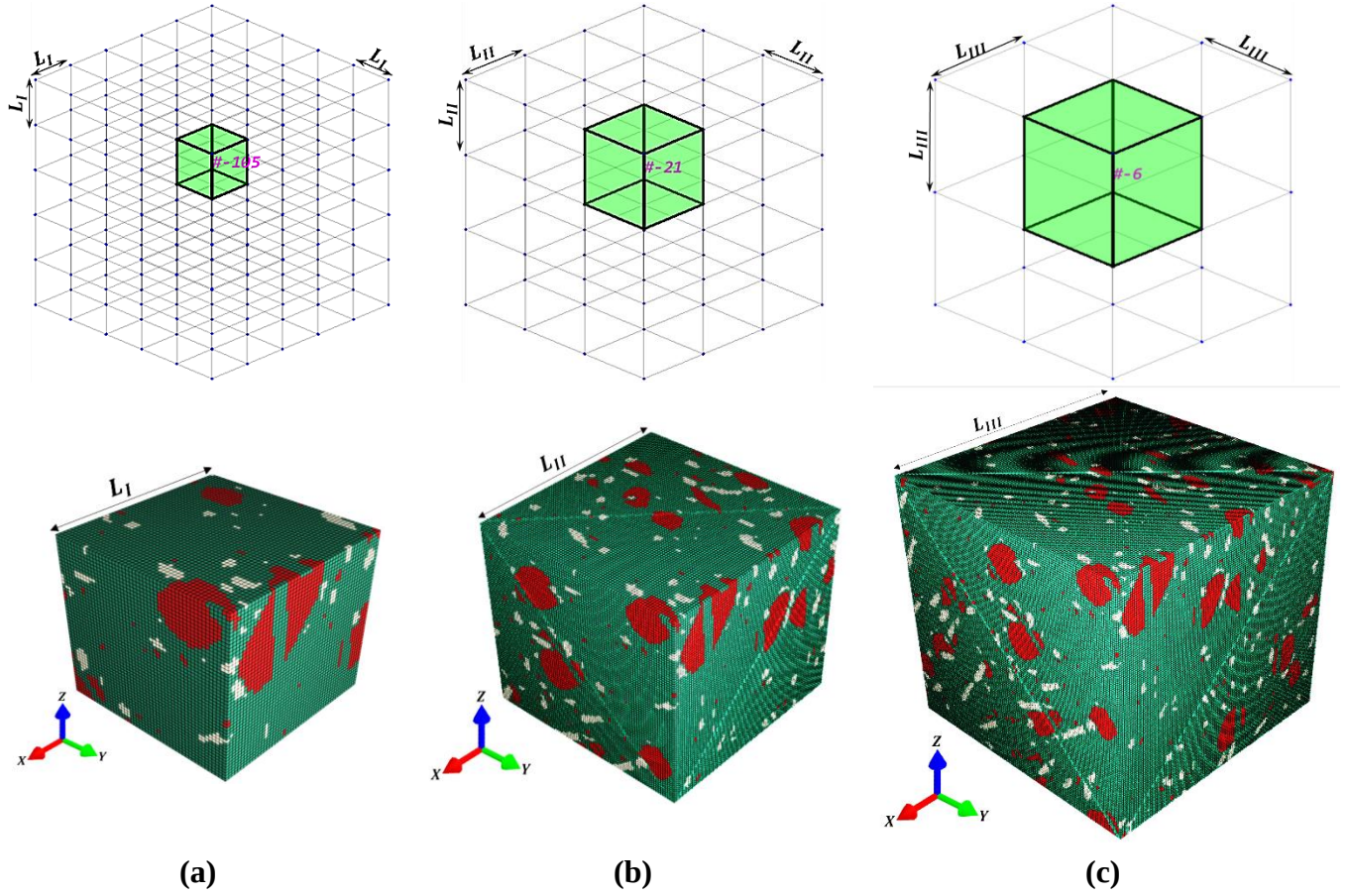


Figure 4.2: ROI-II partitioning into (a) RVE - I: 125 realizations with 125,000 elements per cube and side-length $L_I = 70 \mu m$ (b) RVE - II: 27 realizations with 571,787 elements per cube and side-length $L_{II} = 116.2 \mu m$ (c) RVE - III: 8 realizations with 1,953,125 elements per cube and side-length $L_{III} = 175 \mu m$.

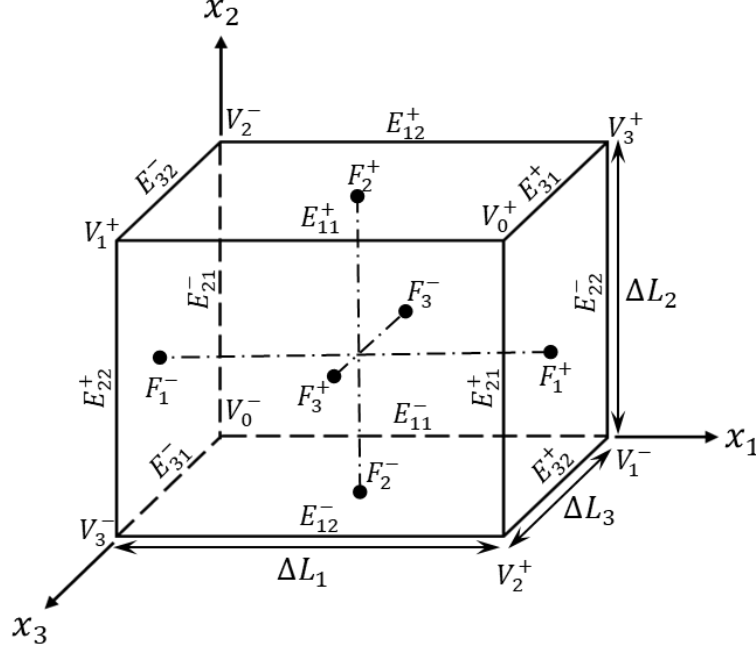


Figure 4.3: Single periodic RVE structure showing definitions of entities and coordinate directions used in the PBC formulations (cf. eqns. (4.1)-(4.3))

The set of equations defining the periodic boundary conditions (PBC) on the parallelepiped RVE boundary entities of the periodic microstructure according to formulations found in literature [298], [299], [300] are summarized in eqns. (4.1)-(4.3) as follows:

$$u_i^{F_k^+} - u_i^{F_k^-} = c_F \hat{\varepsilon}_{ij} \Delta x_j^k \quad \mathcal{T}^{F_k^+} - \mathcal{T}^{F_k^-} = c_F \hat{\nabla}_j \mathcal{T} \Delta x_j^k \quad c_F = \delta_{jk} \quad (4.1)$$

$$u_i^{\Sigma_{kn}^+} - u_i^{\Sigma_{kn}^-} = c_\Sigma \hat{\varepsilon}_{ij} \Delta x_j^k \quad \mathcal{T}^{\Sigma_{kn}^+} - \mathcal{T}^{\Sigma_{kn}^-} = c_\Sigma \hat{\nabla}_j \mathcal{T} \Delta x_j^k \quad c_\Sigma = (-e_{jkm})^n \quad (4.2)$$

$$u_i^{V_k^+} - u_i^{V_k^-} = c_V \hat{\varepsilon}_{ij} \Delta x_j^k \quad \mathcal{T}^{V_k^+} - \mathcal{T}^{V_k^-} = c_V \hat{\nabla}_j \mathcal{T} \Delta x_j^k \quad c_V = (-1)^{\delta_{jk}} \quad (4.3)$$

where $u_i^{N_k}$ is the i^{th} displacement degree of freedom component and $\mathcal{T}^{N_k}$ the temperature degree of freedom component, both in the k^{th} direction on opposing entities N of the periodic structure including faces ($N = F^\pm$), n^{th} edges ($N = \Sigma_n^\pm$) and vertices ($N = V^\pm$). δ_{ij} is the Kronecker delta and e_{ijk} is the Levi-Civita permutation tensor. The quantity $\hat{\varepsilon}_{ij}$ is the average macro-strain tensor of the periodic RVE microstructure. Δx_j^k is the projection of the j^{th} dimension of the RVE along the k^{th} direction and for our orthogonal shaped RVE, $\Delta x_j^k = \Delta L_j$. Indices $i, j, k \in \{1, 2, 3\}$ represent the cartesian degrees of freedoms and $m = 6 - j - k$ in eqn. (4.2) above. Except stated otherwise,

summation is implied by repeated subscript indices from this point onward. The PBC multi-point constraints (MPC) on opposing entities are defined in the model via the equation input syntax. To avoid redundancy, edges and vertices are excluded from the facet node sets definition, and vertices are excluded from the edge node sets definition. The prescribed macro-strains $\hat{\epsilon}_{ij}$ or temperature gradient $\hat{\nabla}_j \mathcal{T}$ are imposed through an extra set of dummy nodes that are coupled to degrees of freedom MPC nodes on opposing PBC entities ($N^\pm$) with a displacement or temperature magnitude equal to the RHS value of the constraint equation.

4.1.1.1 Evaluating the Effective Engineering Constants

For the stress analysis, six (6) deformation load cases with permutation indices kl (11, 22, 33, 23, 13, 12) applied through the displacement PBC constraints are considered. For load case kl , the applied strain $\hat{\epsilon}_{ij}^{kl}$ is given as:

$$\hat{\epsilon}_{ij}^{kl} = \frac{\epsilon}{2} (1 + \delta_{ij}) \delta_{ik} \delta_{lj} \quad (4.4)$$

In eqn. (4.4) above, ϵ is the magnitude of the imposed strain, and repeated indices do not imply summation. We assume a for the imposed strain $\epsilon = 0.25$. The homogenized equivalent macro-stresses $\hat{\sigma}_{ij}$ and macro-strains $\hat{\epsilon}_{ij}$ of the heterogenous RVE volume (Ω) is obtained by volume averaging and is based on satisfaction of the Hill-Mandel condition of equivalent strain energy [301], [302] between the idealized homogenized and heterogenous compounds given as

$$\frac{1}{2} \int_{\Omega} \sigma_{ij}(x) \epsilon_{ij}(x) d\Omega = \frac{1}{2} \hat{\sigma}_{ij} \hat{\epsilon}_{ij} \Omega \quad (4.5)$$

The equivalent macro-stresses $\hat{\sigma}_{ij}$ and macro-strains $\hat{\epsilon}_{ij}$ are thus derived from eqn. (4.5) and given as

$$\hat{\sigma}_{ij} = \frac{1}{\Omega} \int_{\Omega} \sigma_{ij}(x) d\Omega, \quad \hat{\varepsilon}_{ij} = \frac{1}{\Omega} \int_{\Omega} \varepsilon_{ij}(x) d\Omega, \quad i, j = 1, 2, 3; \quad x \in \Omega \quad (4.6)$$

σ_{ij} and ε_{ij} are components of the local stress and strain tensor at material point x of the RVE. The effective elastic constant $\hat{C}_{ijkl}$ is obtained from the homogenized quantities according to the constitutive relation given as

$$\hat{\sigma}_{ij} = \hat{C}_{ijkl} \hat{\varepsilon}_{kl}, \quad i, j, k, l = 1, 2, 3 \quad (4.7)$$

In contracted notation, the above can be rewritten as

$$\hat{\sigma}_m = \hat{C}_{mn} \hat{\varepsilon}_n, \quad m = f_1(i, j), \quad n = f_1(k, l) \quad (4.8)$$

The material constant $\hat{C}_{mn}$ is computed from a least-square linear regression fitting algorithm that minimizes the relative error in the components of the stress tensor of eqn. (4.8) above. Let the i^{th} component of the stress and strain tensors for j^{th} load case be denoted as $\hat{\Psi}_{ij} = \hat{\sigma}_i^j$ and $\hat{\Xi}_{ij} = \hat{\varepsilon}_i^j$ respectively, we define $\hat{D}_{ij}$ such that $\hat{D}_{ij} = \hat{\Xi}_{ik} \hat{\Xi}_{jk}$ and the block diagonal matrices $\hat{P}_{ijkl}$ and $\hat{Q}_{ijkl}$ such that

$$\hat{P}_{ijkl} = \hat{D}_{ij} \delta_{kl}, \quad \text{and}, \quad \hat{Q}_{ijkl} = \hat{\Xi}_{ij} \delta_{kl} \quad (4.9)$$

The material constant $\hat{C}_{mn}$ is thus computed from the linear algebraic expression given as

$$\hat{P}_{ijkl} \hat{C}_{kl} = \hat{b}_{ij}, \quad \hat{b}_{ij} = \hat{Q}_{iljk} \hat{\Psi}_{kl} \quad (4.10)$$

For simplicity the expression of eqn. (4.10) can be represented in the reduced order form given as:

$$\hat{P}_{mn} \hat{C}_n = \hat{b}_m, \quad m = f_2(i, j), \quad n = f_2(k, l) \quad (4.11)$$

The reduced order tensor $\hat{P}_{mn}$ is a 36 x 36 matrix and the tensors $\hat{C}_n$ and $\hat{b}_m$ are 36 x 1 vectors. Depending on the requirements of material property behavior desired from the least square fitting of the elastic constant $\hat{C}_n$, such as matrix and material symmetry, orthogonality, isotropy etc., the imposition of constraints is defined through a constraint matrix X_{vn} that satisfies the equation given as

$$X_{vn}\hat{C}_n = 0, \quad n = 1 \dots 36 \quad (4.12)$$

In the current study, only two material requirements are needed for complete definition of the constraint matrix X_{vn} which include the condition of material symmetry and material orthogonality defined through sets of linear equation constraint submatrices X'_{rn} and X''_{sn} respectively such that

$$X_{vn} = \begin{bmatrix} X'_{rn} \\ X''_{sn} \end{bmatrix} \quad (4.13)$$

The necessary condition of material symmetry $\hat{C}_{ij} = \hat{C}_{ji}$ in anisotropic materials requires the definition of 15 essential constraints, and thus X'_{rn} is a 15 x 36 sized submatrix such that:

$$X'_{rn} = \delta_{np} - \delta_{nq} \quad (4.14)$$

$$r = f_3(i, j), \quad p = f_2(i, j), \quad q = f_2(j, i), \quad i = 1 \dots 5, \quad j = i + 1 \dots 6$$

Further conditions for material orthotropy necessitating three (3) orthogonal planes of symmetry requires the definition of 12 additional constraints that sets the shear-extension (9) and biplanar shear-shear (3) coupling terms of the elastic constants to zero. i.e.

$$\hat{C}_{ij} = 0 \quad (4.15)$$

$$i = 1 \dots 5, \quad j = \begin{cases} 4 \dots 6 & i \leq 3 \\ i + 1 \dots 6 & i > 3 \end{cases}$$

The constraint submatrix X''_{sn} is thus a 12 x 36 tensor given as

$$X''_{sn} = \delta_{np}, \quad s = f_4(i, j), \quad p = f_2(i, j) \quad (4.16)$$

The linear indices transformation functions $f_1(i, j)$, $f_2(i, j)$, $f_3(i, j)$ and $f_4(i, j)$ that appear in eqn. (4.8) - (4.16) are given as

$$\begin{aligned} f_1(i, j) &= r\delta_{ij} + (1 - \delta_{ij})(9 - i - j), & f_2(i, j) &= i + 6(j - 1) \\ f_3(i, j) &= j - .5(i^2 - 11i + 12), & f_4(i, j) &= \begin{cases} 3i + j - 6 & i \leq 3 \\ i + j + 1 & i > 3 \end{cases} \end{aligned} \quad (4.17)$$

A Lagrange multiplier method is used to combine constitutive relation of eqn. (4.11) and the constraint definition of eqn. (4.12) to obtain a final linear algebraic system of equation given as

[296]

$$\begin{bmatrix} \hat{P}_{mn} & X_{nv} \\ X_{vn} & 0_{vv} \end{bmatrix} \begin{bmatrix} \hat{C}_n \\ \lambda_v \end{bmatrix} = \begin{bmatrix} \hat{b}_m \\ 0_v \end{bmatrix} \quad (4.18)$$

λ_v is a vector of the Lagrange multipliers for each imposed constraint definition. The accuracy of the regression fit is assessed by the coefficient of determination for each j^{th} load case R_j^2 given as

$$R_j^2 = 1 - St^j / Sr^j \quad (4.19)$$

where,

$$St^j = \sum_{vi} [\hat{\sigma}_i^j - \hat{C}_{ik} \hat{\epsilon}_k^j]^2, \quad Sr^j = \sum_{vi} [\hat{\sigma}_i^j - \langle \sigma \rangle^j]^2, \quad \langle \sigma \rangle^j = \frac{1}{n} \sum_{vi} \hat{\sigma}_i^j \quad (4.20)$$

The computed solution of $\hat{C}_n$ transformed to the 6 x 6 matrix form $\hat{C}_{mn}$ and inverted yields the effective compliance matrix $\hat{S}_{mn}$ from which we can derive the 9 independent engineering constants according to eqn. (4.21) below.

$$\hat{S}_{mn} = [\hat{C}^{-1}]_{mn} = \begin{bmatrix} \frac{1}{E_{11}} & -\frac{\nu_{21}}{E_{22}} & -\frac{\nu_{31}}{E_{33}} & & & \\ -\frac{\nu_{12}}{E_{11}} & \frac{1}{E_{22}} & -\frac{\nu_{32}}{E_{33}} & & & \\ -\frac{\nu_{13}}{E_{11}} & -\frac{\nu_{23}}{E_{22}} & \frac{1}{E_{33}} & & & \\ & & & \frac{1}{G_{23}} & & \\ & & & & \frac{1}{G_{13}} & \\ & & & & & \frac{1}{G_{12}} \end{bmatrix} \quad (4.21)$$

4.1.1.2 Evaluating the Effective Coefficient of Thermal Expansion

Estimation of the ECTE is based on the Duhamel-Neumann law [303], [304]. Fundamental constitutive expression that relates the mechanical stress $\hat{\sigma}_{ij}$ to the strains in a thermally loaded material is given as

$$\hat{\sigma}_{ij} = \hat{C}_{klij}[\hat{\varepsilon}_{kl} - \hat{\alpha}_{kl}\Delta\mathcal{T}] \quad (4.22)$$

where $\hat{C}_{klij}$ is the effective elastic stiffness tensor of the homogenized material, $\hat{\varepsilon}_{kl}$ is the average total strain tensor, $\hat{\alpha}_{kl}$ is the ECTE tensor and $\Delta\mathcal{T}$ is the applied steady state temperature difference.

In contracted notation, this is given as

$$\hat{\sigma}_m = \hat{C}_{nm}[\hat{\varepsilon}_n - \hat{\alpha}_n\Delta\mathcal{T}] \quad (4.23)$$

In our analysis, we assume $\Delta\mathcal{T} = 65^\circ C$. In evaluating the ECTE, the total strain ε_{ij}^{kl} in the sets of defined PBC constraints in eqn. (4.1) - (4.3) above is set to zero, thus the resulting equivalent macro-strain tensor $\hat{\varepsilon}_{kl} = 0$. eqn. (4.23) above reduces to

$$\hat{\sigma}_m = -\hat{C}_{nm}\hat{\alpha}_n\Delta\mathcal{T} \quad (4.24)$$

From the results of the homogenized equivalent macro-stresses $\hat{\sigma}_m$ derived from the thermal expansion analysis based on an applied temperature difference $\Delta\mathcal{T}$ to the entire RVE volume, the ECTE can be computed by rearranging eqn. (4.24) above to give

$$\hat{\alpha}_n = -\hat{S}_{nm}\hat{\sigma}_m/\Delta\mathcal{T} \quad (4.25)$$

4.1.1.3 Evaluating the Effective Thermal Conductivity

For the heat transfer analysis, three (3) thermal load cases with permutation indices kk (11, 22, 33) applied through the temperature PBC constraints are considered which are basically the orthogonal temperature gradients such that for case k

$$\hat{V}_j^k \mathcal{T} = \delta_{jk} \tau \quad (4.26)$$

where τ is the magnitude of the imposed temperature. In our analysis, we assume $\tau = 100^\circ C$. The general heat conduction energy conservation equation at an arbitrary material point within the RVE volume is given as

$$\nabla_i \kappa_{ij} \nabla_j \mathcal{T} + \dot{q}_v = \rho c_p \dot{\mathcal{T}} \quad (4.27)$$

where ∇_i is the gradient operator vector, κ_{ij} is the thermal conductivity of the material, $\mathcal{T}$ is the temperature, $\dot{q}_v$ is the rate of internal heat generation within the material, ρ & c_p are the density and specific heat capacity respectively, all quantities evaluated at specified material point within the RVE volume. In the FEA analysis for evaluating of the ETC tensor $\hat{\kappa}_{ij}$, we assumed steady state ($\dot{\mathcal{T}} = 0$) and there is no internal heat generation ($\dot{q}_v = 0$). The resulting temperature field distribution is the solution to the equation given as

$$\nabla_i \kappa_{ij} \nabla_j \mathcal{T} = 0 \quad (4.28)$$

From Fourier's law of steady state heat conduction, the heat flux q_i at any material point within the conducting medium is given as:

$$q_i = -\kappa_{ij} \nabla_j \mathcal{T} \quad (4.29)$$

By integrating eqn. (4.29) above and applying the Gauss divergence theorem making appropriate substitution for the heat flux q_i yields

$$\int_{\Omega} \nabla_i \kappa_{ij} \nabla_j \mathcal{T} \, d\Omega = \int_{\Lambda} \kappa_{ij} \nabla_j \mathcal{T} \, \vec{n}_i \, d\Lambda = - \int_{\Lambda} q_i \, \vec{n}_i \, d\Lambda = 0 \quad (4.30)$$

The Hill-Mandel condition of equivalent thermal dissipation between the homogenous and heterogenous compounds is given as [113]

$$-\hat{q}_i \hat{\nabla}_i \mathcal{T} = -\widehat{\{q_i \nabla_i\}} \mathcal{T} = -\frac{1}{\Omega} \int_{\Omega} q_i(x) \nabla_i \mathcal{T}(x) d\Omega \quad (4.31)$$

Applying the same macrohomogeneity principle as was done with the stress analysis previously described, we obtain spatial averages of the local heat flux $\hat{q}_i$ and temperature gradient $\hat{\nabla}_i \mathcal{T}$ given as [109], [126], [305]

$$\hat{q}_i = \frac{1}{\Omega} \int_{\Omega} q_i d\Omega, \quad \hat{\nabla}_i \mathcal{T} = \frac{1}{\Omega} \int_{\Omega} \nabla_i \mathcal{T} d\Omega \quad (4.32)$$

The ETC tensor of the RVE volume can thus be obtained from the Fourier's law of steady heat conduction given as:

$$\hat{q}_i^k = -\hat{\kappa}_{ij} \widehat{\nabla}_j^k \mathcal{T} \quad (4.33)$$

For simplicity, let the i^{th} component of the equivalent homogenized heat flux $\hat{q}_i^k$ and temperature gradient quantity $\widehat{\nabla}_j^k \mathcal{T}$ for the k^{th} load case be denoted as $\hat{Q}_{ik} = \hat{q}_i^k$ and $\widehat{\nabla T}_{jk} = \widehat{\nabla}_j^k \mathcal{T}$ respectively.

Then eqn. (4.33) above can be rewritten in tensorial notation as

$$\hat{Q}_{ik} = -\hat{\kappa}_{ij} \widehat{\nabla T}_{jk} \quad (4.34)$$

Rearranging eqn. (4.34) above, we can compute the ETC tensor $\hat{\kappa}_{ij}$ given as

$$\hat{\kappa}_{ij} = -\hat{Q}_{ik} [\widehat{\nabla T}^{-1}]_{jk} \quad (4.35)$$

4.1.2 Analytical Mean-Field Homogenization Method

Traditionally, the analytical approach to determine the properties of randomly distributed misaligned discontinuous fiber reinforced composite first developed by Advani and Tucker [17], [57] involves a two (2) step micromechanics homogenization approach, a first step that estimates average properties of decomposed pseudo-grains of unidirectionally aligned, uniform length fiber reinforced composite microscale RVE using any of the available mean-field theories [59] or numerical FEA analysis and a second step that involves orientation and length averaging of the aggregates using either the Voight's or Reuss' assumption, to account for the randomly dispersed spatially varying fiber orientation and length distribution in the heterogenous macro-scale volume of injection molded or extrusion-deposited polymer composites. In subsequent section, we present the Mori-Tanaka-Benveniste's analytical mean field homogenization approach for estimating the effective quantities, i.e. the 4th order elastic stiffness tensor $\hat{C}_{ijkl}$, the 2nd order coefficient of thermal expansion (ECTE), $\hat{\alpha}_{ij}$ and the 2nd order thermal conductivity tensor $\hat{\kappa}_{ij}$.

4.1.2.1 Estimating the Effective Engineering Constants

Various empirical micromechanics models have been developed to predict the elastic properties of unidirectionally aligned discontinuous short fiber reinforced polymer composite such as those briefly discussed in the introduction section of the current article and summarized in [59]. One such model which has been used for our validation is the Eshelby based – Mori-Tanaka formulations for calculating the effective composite stiffness. The general equation for the mean-field homogenized composite stiffness C_{ijkl} is given as:

$$C_{ijkl} = \left[\vartheta_m C_{ijkl}^m + \sum_p \vartheta_p C_{ijrs}^p B_{rsuv}^p \right] [\tilde{B}^{-1}]_{uvkl}, \quad \tilde{B}_{ijkl} = \left[\vartheta_m \delta_{ijkl} + \sum_p \vartheta_p B_{ijkl}^p \right] \quad (4.36)$$

where C_{ijkl}^m and C_{ijkl}^p are the isotropic matrix and particulates (fiber & voids) stiffness tensors. The particle's strain concentration tensor B_{ij}^p according to the Mori-Tanaka model corresponds to the Hashin-Shtrikman-Willis lower bounds solution for the stiffness tensor and is computed from eqn. (4.37) given as

$$[B^{-1}]_{ijkl}^p = \delta_{ijkl} + \Pi_{ijrs}^m ([C^{-1}]_{rsuv}^m C_{uvkl}^p - \delta_{rskl}) \quad (4.37)$$

where Π_{ijkl} is the Eshelby's elasticity tensor given in APPENDIX A. We assume the micro-void inclusions are spherical shaped with unity aspect ratio. Length averaging of the stiffness tensor is first performed using the length distribution of the inclusions within the composite according to the eqn. (4.38) given as

$$C_{ijkl} = \sum_{\forall n} w^n C_{ijkl}^n(\hat{a}_r^n) \quad (4.38)$$

where w^n is the weight fraction of the n^{th} pseudo-grain of an inclusion phase with average aspect ratio $\hat{a}_r^n$. Subsequently, the orientation average of the fourth order transversely isotropic elastic

stiffness tensor $\hat{C}_{ijkl}$ is computed using the fourth-order fiber orientation averaging scheme [57] given as:

$$\begin{aligned}\hat{C}_{ijkl} = & \beta_1 a_{ijkl} + \beta_2 (a_{ij} \delta_{kl} + a_{kl} \delta_{ij}) + \beta_3 (a_{ik} \delta_{jl} + a_{il} \delta_{jk} + a_{jk} \delta_{il} + a_{jl} \delta_{ik}) \\ & + \beta_4 (\delta_{ij} \delta_{kl}) + \beta_5 (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})\end{aligned}\quad (4.39)$$

where a_{ijkl} is the 4th order orientation tensor of an inclusion phase computed using any of the suitable closure approximations detailed in [17], [57], [251] and β_i vector are invariants of the unidirectional transversely isotropic elasticity tensor C_{mn} in contracted notation given as

$$\begin{aligned}\beta_1 &= C_{11} + C_{22} - 2C_{12} - 4C_{66}, & \beta_2 &= C_{12} - C_{23} \\ \beta_3 &= C_{66} + \frac{1}{2}(C_{23} - C_{22}), & \beta_4 &= C_{23}, & \beta_5 &= \frac{1}{2}(C_{22} - C_{23})\end{aligned}\quad (4.40)$$

The engineering constants is computed from the orthotropic stiffness matrix based on eqn. (4.21).

4.1.2.2 Estimating the Effective Coefficient of Thermal Expansion

Like the method adopted in the elastic stiffness prediction, a two-step homogenization approach is employed to estimate ECTE tensor for the discontinuous fiber reinforced polymer composite. The orientation averaged ECTE tensor $\hat{\alpha}_{ij}$ for the misaligned discontinuous fiber reinforced composite is computed from the expression given as [57], [139], [296]

$$\hat{C}_{ijkl} \hat{\alpha}_{kl} = [\widehat{C:\alpha}]_{ij}, \quad [C:\alpha]_{ij} = C_{ijkl} \alpha_{kl} \quad (4.41)$$

The orientation average for tensor product $[\widehat{C:\alpha}]_{ij}$ is calculated using the second-order orientation averaging scheme by Advani & Tucker [17], [57] given as

$$[\widehat{C:\alpha}]_{ij} = \gamma_1 a_{ij} + \gamma_2 \delta_{ij} \quad (4.42)$$

where γ_1 and γ_2 are the invariants of the tensor product $[C:\alpha]_{ij}$ obtained from the double contraction of the transversely isotropic elasticity tensor C_{ijkl} and CTE tensor α_{kl} for aligned discontinuous fiber composite respectively given as

$$\gamma_1 = [C:\alpha]_{11} - [C:\alpha]_{22}, \quad \gamma_2 = [C:\alpha]_{22} \quad (4.43)$$

For consistency, we utilize the Mori-Tanaka-Benveniste's equation for estimating the tensor product $C_{ijkl}\alpha_{kl}$ of unidirectional aligned short fiber reinforced polymer composite with isotropic constituents given as [92], [93], [94], [95]

$$[C:\alpha]_{ij} = C_{ijkl}\alpha_{kl} = \left[\vartheta_m C_{uvkl}^m \alpha_{kl} + \sum_p \vartheta_p C_{uvrs}^p B_{rskl}^p \alpha_{kl}^p \right] [\tilde{B}^{-1}]_{uvij} \quad (4.44)$$

Length averaging of the tensor product $[C:\alpha]_{ij}$ is carried out prior to orientation averaging according to eqn. (4.45) below.

$$[C:\alpha]_{ij} = \sum_{\forall n} w^n [C:\alpha]_{ij}^n(\hat{a}_r^n) \quad (4.45)$$

4.1.2.3 Estimating the Effective Thermal Conductivity

The Mori-Tanaka's model presented earlier for predicting the homogenized 4th order elasticity tensor for unidirectional particulate composite has been extended by several authors [97], [108], [109] to estimate other 2nd order tensor properties of the composite material including the thermal conductivity tensor. The reformulated expression for the composite's thermal conductivity tensor κ_{ij} is given as

$$\kappa_{ij} = \left[\vartheta_m \kappa_{is}^m + \sum_{\forall p} \vartheta_p \kappa_{ir}^p A_{rs}^p \right] [\tilde{A}^{-1}]_{sj}, \quad \tilde{A}_{ij} = \vartheta_m \delta_{ij} + \sum_{\forall p} \vartheta_p A_{ij}^p \quad (4.46)$$

where κ_{ij}^m and κ_{ij}^p are the isotropic matrix and particulates (fiber & void) thermal conductivity tensors. The intensity-concentration tensor A_{ij}^p so-called that couples the mean temperature gradients between the particulate inclusions and the matrix corresponds to the lower bound

solution based on Hashin-Shtrikman-Willis single variational principle and is computed from eqn. (4.47) given as

$$[A^{-1}]_{ij}^p = \delta_{ij} + \mathcal{K}_{ir} \{ [\kappa^{-1}]_{rs}^m \kappa_{sj}^p - \delta_{rj} \} \quad (4.47)$$

In eqn. (4.46) - (4.47) above, $\mathcal{K}_{ij}$ is the Eshelby's thermal conductance tensor having only non-zero diagonal components and for spheroidal inclusions, this is given as

$$\mathcal{K}_{22} = \mathcal{K}_{33} = 0.5a_r\chi_r^3\{a_r\chi_r^{-1} - \ln[a_r + \chi_r^{-1}]\}, \quad \mathcal{K}_{11} = 1 - 2\mathcal{K}_{22} \quad (4.48)$$

In our calculations, we assume the void particles are spherical shaped for simplicity, in this case the Eshelby's tensor is simply one-third the identity tensor, i.e. $\mathcal{K}_{ij} = 1/3 \delta_{ij}$. Similar to the procedure adopted in elasticity stiffness tensor homogenization, length averaging of the computed transversely isotropic thermal conductivity tensor using the length distribution of the fiber inclusion phase within the SFRP composite is carried out on a weight-based averaging scheme according to eqn. (4.49)

$$\kappa_{ij} = \sum_{\forall n} w^n \kappa_{ij}^n(\hat{a}_r^n) \quad (4.49)$$

The invariants of κ_{ij} i.e. ψ_i are computed in the usual manner given as

$$\psi_1 = \kappa_{11} - \kappa_{22}, \quad \psi_2 = \kappa_{22} \quad (4.50)$$

Subsequently, the second order orientation averaged thermal conductivity tensor for the misaligned fiber reinforced composite is calculated using eqn. (4.51) given as [57], [108]

$$\hat{\kappa}_{ij} = \psi_1 a_{ij} + \psi_2 \delta_{ij} \quad (4.51)$$

4.1.2.4 Density and Specific Heat Estimation

To determine the average density $\hat{\rho}$ and specific heat capacity $\hat{\zeta}_s$ of the composite material, the basic rule of mixture equation would suffice in estimating the scalar quantities since the average quantities are only dependent on the phase fractions and independent of the spatial

variations and characteristics of the RVE microstructural constituents. The average density $\hat{\rho}$ is given as

$$\hat{\rho} = \vartheta_m \rho_m + \sum_{\forall p} \vartheta_p \rho_p \quad (4.52)$$

where ρ_m & ρ_p are the matrix and particulates (fiber and void) isotropic density. Likewise, the average specific heat capacity $\hat{\zeta}_s$ is given as

$$\hat{\zeta}_s = \vartheta_m \zeta_s^m + \sum_{\forall p} \vartheta_p \zeta_s^p \quad (4.53)$$

where ζ_s^m & ζ_s^p are the matrix and particulates (fiber and void) isotropic specific heat capacity values.

4.1.3 Evaluating the Magnitude of the Effective Quantities

The effective elastic modulus magnitude E^{eff} and effective Poisson ratio ν^{eff} are given as [306]:

$$\frac{1}{E^{eff}} = \frac{1}{3G^{eff}} + \frac{1}{9K^{eff}}, \quad \nu^{eff} = \frac{3K^{eff} - 2G^{eff}}{6K^{eff} + 2G^{eff}} \quad (4.54)$$

where K^{eff} is the apparent effective bulk modulus defined as the average between the Voigt upper K_V and Reuss lower K_R first order bounds on the bulk modulus and is given as

$$K^{eff} = \frac{1}{2}[K_V + K_R], \quad \frac{1}{K_R} = \hat{S}_{iijj}, \quad K_V = \frac{1}{9}\hat{C}_{iijj} \quad (4.55)$$

The effective shear modulus G^{eff} is obtained from the average of the Voigt upper G_V and Reuss lower G_R first order bounds on the shear modulus and is given as

$$G^{eff} = \frac{1}{2}[G_V + G_R], \quad \frac{1}{G_R} = \frac{2}{5}\left[\hat{S}_{ijij} - \frac{1}{3}\hat{S}_{iijj}\right], \quad G_V = \frac{1}{10}\left[\hat{C}_{ijij} - \frac{1}{3}\hat{C}_{iijj}\right] \quad (4.56)$$

Likewise, the apparent ECTE magnitude α^{eff} is computed from the Hill's average of the Voight lower bound α_V and Reuss upper bound α_R values of the CTE tensor [307]. i.e.

$$\alpha^{eff} = \frac{1}{2}[\alpha_V + \alpha_R], \quad \alpha_V = \frac{1}{3}\hat{\alpha}_{ii}, \quad \alpha_V = \frac{1}{9K_V}\hat{C}_{iikl}\hat{\alpha}_{kl} \quad (4.57)$$

The apparent effective thermal conductivity magnitude is given as [113], [116]:

$$\kappa^{eff} = \frac{1}{3}\hat{\kappa}_{ii} \quad (4.58)$$

4.2 Results & Discussion

The results of the thermo-mechanical properties including the engineering constants, ECTE and ETC obtained from both FEA and numerical homogenization approaches are presented in the sections following. The impact of the inherent microstructural porosity on the resulting thermomechanical properties are also evaluated and quantitative assessment of the macroscale property anisotropy due to spatial variation of the microstructural configurations across various ROIs are presented. The measure of dispersion in the computed effective properties for each ROI is quantified using the coefficient of variation statistical parameter $\xi = \sigma/\mu$. ξ quantifies the suitability of the selected RVE size in representing the ROI volume. It is worth noting however that suitable RVE size selection is yet limited by the computational cost. In the current investigation, we choose a dispersion error tolerance of $\xi(Z) \leq 5\%$ as our acceptance criteria for selecting a suitable RVE size in accurately predicting a composite property Z . From this point onward, the effective composite properties are reported in their normalized form with respect to the equivalent properties of the isotropic matrix phase and are distinguished from actual non-dimensional quantities by an overbar accent. i.e. $\bar{Z} = Z/Z_m$.

4.2.1 Thermo-Mechanical Property Estimates for ROI-II

The first set of results presented here are solutions obtained from all RVE cases for ROI-II based on numerical FEA homogenization method and for the three (3) RVE sizes considered. Numerically computed quantities are compared to results obtained from corresponding analytical estimates based on the Mori-Tanaka's model. The effect of the inherent micro-voids on the resulting effective thermo-mechanical properties is also quantified. Using the same methodology as [297], the average microstructural properties of ROI-II used for the first set of analytical and numerical homogenization analysis are estimated. The computed values include the fiber volume fraction $\vartheta_f^{\text{II}} = 6.65\%$, the void volume fraction $\vartheta_v^{\text{II}} = 12.27\%$, and the ensemble average fiber orientation tensor $\hat{\mathbf{a}}_{ij}^{\text{II}}$. The region average 2nd order fiber orientation tensor for ROI-II is given as [297]

$$\hat{\mathbf{a}}_{ij}^{\text{II}} = \langle p_i p_j \rangle = \begin{bmatrix} 0.32 & 0.04 & 0.12 \\ 0.04 & 0.17 & -0.02 \\ 0.12 & -0.02 & 0.51 \end{bmatrix}$$

From the region averaged orientation, we observe higher degree of fiber alignment with the print direction (z-direction) followed by the x-direction parallel to the direction of substrates translation. The distribution of the fiber aspect ratio for this region (ROI-II) is presented in Figure 4.4 below. The distribution of the aspect ratio within the region can be fitted to a Weibull function with a shape parameter $\phi_1 = 22.72$ and a scale parameter $\phi_2 = 1.65$. The weighted average aspect ratio for this region is $\hat{a}_r^{\text{II}} = 20.31$. The average fibers aspect ratio is limited by the ROI envelope which may under-represent the specimens true mean value. Partitioning of the ROI into several RVE realizations further limits the average fiber aspect ratio within the region. The mean fiber aspect ratio ($\hat{a}_r$) and coefficient of variation (ξ_r) from the complete sets of realizations of each RVE sizes (RVE- I, II, & III) of ROI-II are presented in Table 4.2.

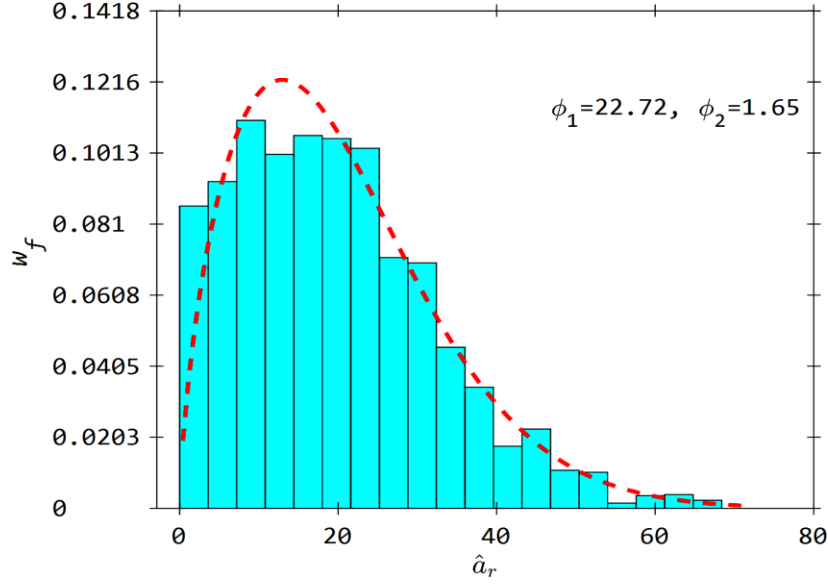


Figure 4.4: Fiber Aspect Ratio Distribution across extracted for ROI-II

Table 4.2: showing the mean fiber aspect ratio ($\hat{a}_r$) and coefficient of variation (ξ_r) from the complete sets of realizations of the different RVE cases from ROI-II region.

	RVE-I	RVE-II	RVE-III
$\hat{a}_r$	8.58	12.38	15.69
ξ_r [%]	10.50	7.19	7.01

The volume information of the microstructural characteristics for the center ROI (ROI-II) is used to estimate the thermo-mechanical properties of the region by averaging the results of all realizations of a select RVE size within the ROI.

4.2.1.1 Effective Stiffness & Engineering Constants

Typical displacement contours of a single RVE cube (RVE-II, #14) extracted from partitioning of ROI-II under tensile and shear loading are shown in Figure 4.5a-c below. As previously stated, the homogenized stresses and strains from the six (6) different load cases are used to compute the effective stiffness and compliance of the RVE volume.

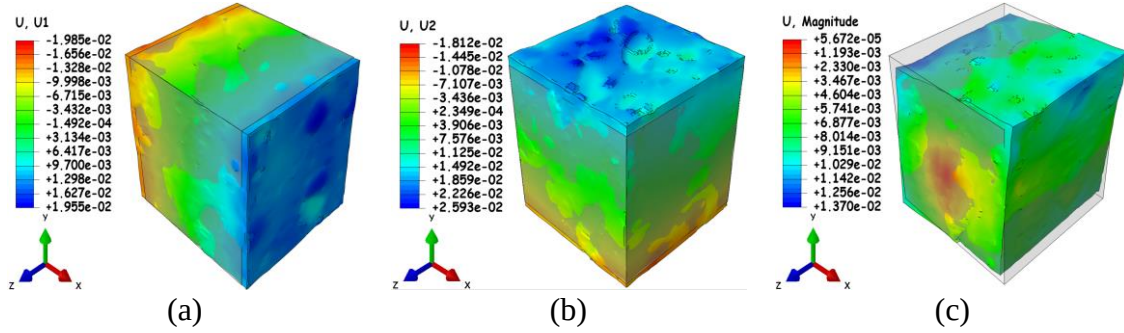


Figure 4.5: Isometric view of the deformation contours of RVE-II, #14 from ROI-II overlaid over the undeformed volume under different loading (a) tensile response in x-direction (b) tensile response in y-direction (c) shear response in xy-plane.

From Figure 4.6a-c we see mirrored contortion of opposing facets of the RVE in the direction of the applied load for x and y tensile deformations and the x-y shear deformation load cases which verifies correct implementation of the periodic constraints on the boundaries of the RVE. The periodic constraints enforce domain continuity without overlaps or separation among neighboring RVE boundaries that ensures effective transfer of loads between adjacent RVE boundaries.

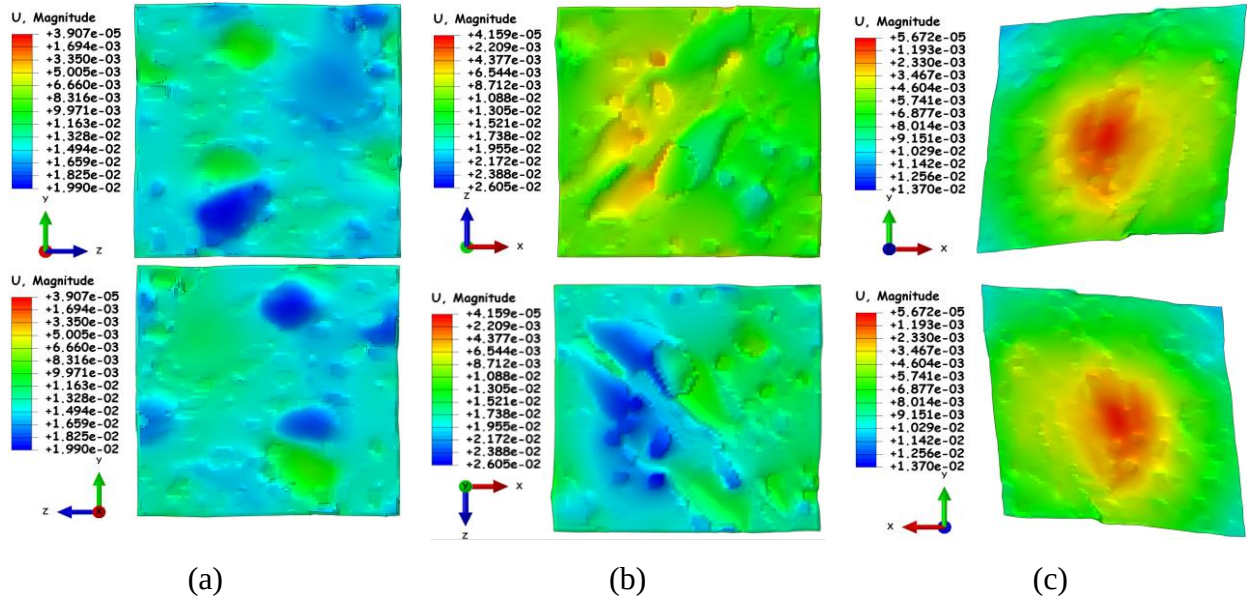


Figure 4.6: Deformation contours of RVE-II, #14 from ROI-II showing topography of opposing facet pairs to validate the implementation of the PBC for loading in (a) x-direction (b) y-direction (c) xy-plane.

Table 4.3 below shows the results of the mean elastic stiffness $\bar{C}_{mn}$ derived from the orthotropic regression fitting procedure of the homogenization macro-stresses and macro-strains obtained from the FEA simulations for the complete sets of RVE-II realizations of ROI-II compared to the Mori-Tanaka's mean-field estimate using the length distribution in Figure 4.4 and the region averaged orientation of ROI-II. Results show a close alignment between both homogenization methods. The results of the stiffness tensor in Table 4.3 and elastic moduli in Table 4.4 reveals that the 13% CF/ABS bead specimen exhibits a somewhat transversely isotropic material macro-behavior along the z-plane. It can also be observed that the largest component of the stiffness tensor $\bar{C}_{33}$ (Table 4.3) or the largest elastic moduli $\bar{E}_{33}$ (Table 4.4) coincides with the largest average fiber orientation component $\hat{a}_{33}$ which suggest that material stiffness increases with increasing degree of fiber alignment. It is evident from the results of Table 4.3 and Table 4.4 what we expect otherwise from literature [97], [110], [128] that there is a clear reduction in the magnitude of the predicted elastic stiffness components with the consideration of micro-void inclusions. We observe moderately high accuracy of the regression fitting procedure based on the average values of the computed coefficient of determination ($\widehat{R}^2 > 0.80$) from the set of realizations of the various RVE sizes reported in Table 4.5.

Table 4.3: Results of the average elastic stiffness $\bar{C}_{mn}$ for ROI-II region obtained from numerical (FE) homogenization approach based on RVE-II and the orientation averaged Mori-Tanaka (MT) method for 13% CF/ABS SFRP composite considering (a) non-porous microstructure (b) porous microstructure.

(a)				(b)			
FE	1.34	1.20	1.33	0.98	0.75	0.89	
	1.20	1.28	1.21	0.75	0.89	0.77	
	1.33	1.21	1.51	0.89	0.77	1.15	
			1.51				1.21
			1.79				1.48
			1.49				1.18

MT	1.36	1.18	1.32				0.99	0.74	0.83			
	1.18	1.21	1.21				0.74	0.87	0.75			
	1.32	1.21	1.61				0.83	0.75	1.21			
				1.44						1.20		
					1.71						1.45	
						1.37						1.13

Table 4.4: Mean values of the engineering constants for ROI-II computed from the numerical FEA homogenization schemes for all RVE cases (RVE- I, II & III) and considering (a) non-porous microstructure (b) porous microstructure.

Cases	$\bar{E}_{11}$	$\bar{E}_{22}$	$\bar{E}_{33}$	$\bar{G}_{23}$	$\bar{G}_{13}$	$\bar{G}_{12}$	$\bar{\nu}_{23}$	$\bar{\nu}_{13}$	$\bar{\nu}_{12}$
(a) RVE-I	1.39	1.38	1.58	1.45	1.67	1.42	0.82	0.93	0.97
(a) RVE-II	1.42	1.41	1.65	1.51	1.79	1.49	0.78	0.93	0.98
(a) RVE-III	1.45	1.43	1.70	1.55	1.87	1.52	0.76	0.94	0.97
(b) RVE-I	1.08	1.04	1.27	1.15	1.34	1.11	0.72	0.86	0.90
(b) RVE-II	1.12	1.07	1.35	1.21	1.48	1.18	0.78	0.93	0.98
(b) RVE-III	1.14	1.09	1.40	1.26	1.56	1.21	0.76	0.94	0.97

Table 4.5: Mean values and standard deviation of the coefficient of determination R^2 for ROI-II computed from the least square regression fitting procedure for all RVE cases (RVE- I, II & III) and considering (a) non-porous microstructure (b) porous microstructure.

Cases	(a)			(b)		
	RVE-I	RVE-II	RVE-III	RVE-I	RVE-II	RVE-III
$\widehat{R^2}$	0.92	0.90	0.88	0.87	0.84	0.81
σ_{std}	0.05	0.05	0.05	0.08	0.07	0.08

Figure 4.7a-c presents error-bar plots showing the mean value ('x' marker), the interquartile range (solid rectangle) consisting of the lower quartile, median line and upper quartile, the extremums of the data range (error-bars), and outliers (isolated dots) for the predicted engineering constants and from the complete set of realizations of the different RVE sizes considered. The results show a clear reduction in the dispersion of quantities with increasing RVE size. The mean values are seen to converge approximately for the mid-sized and largest RVE's, (i.e. RVE- II & III, cf. Figure 4.7b & c) for all engineering constants which validates the conclusions of Kanit et al. [116] that small but reasonable sized RVE with sufficient number of realizations can accurately predict effective properties as would larger sized RVE with smaller number of realizations. We observe quite a bit of deviation in the results of the engineering

constants for the smallest RVE (RVE- I). Although micro-voids are seen to reduce the elastic moduli for all RVE cases (up to 24% reduction observed), the mean Poisson ratios computed from RVE- II & III are seen to be unaffected by micro-void inclusions (cf. Figure 4.7b&c) contrary to what is observed from the results of RVE-I (cf. Figure 4.7a) which shows a remarkable impact of the voids on the Poisson's ratios as high as 11.6%. This suggests that RVE-I is insufficient in accurately predicting the elastic modulus of the CF/ABS composite.

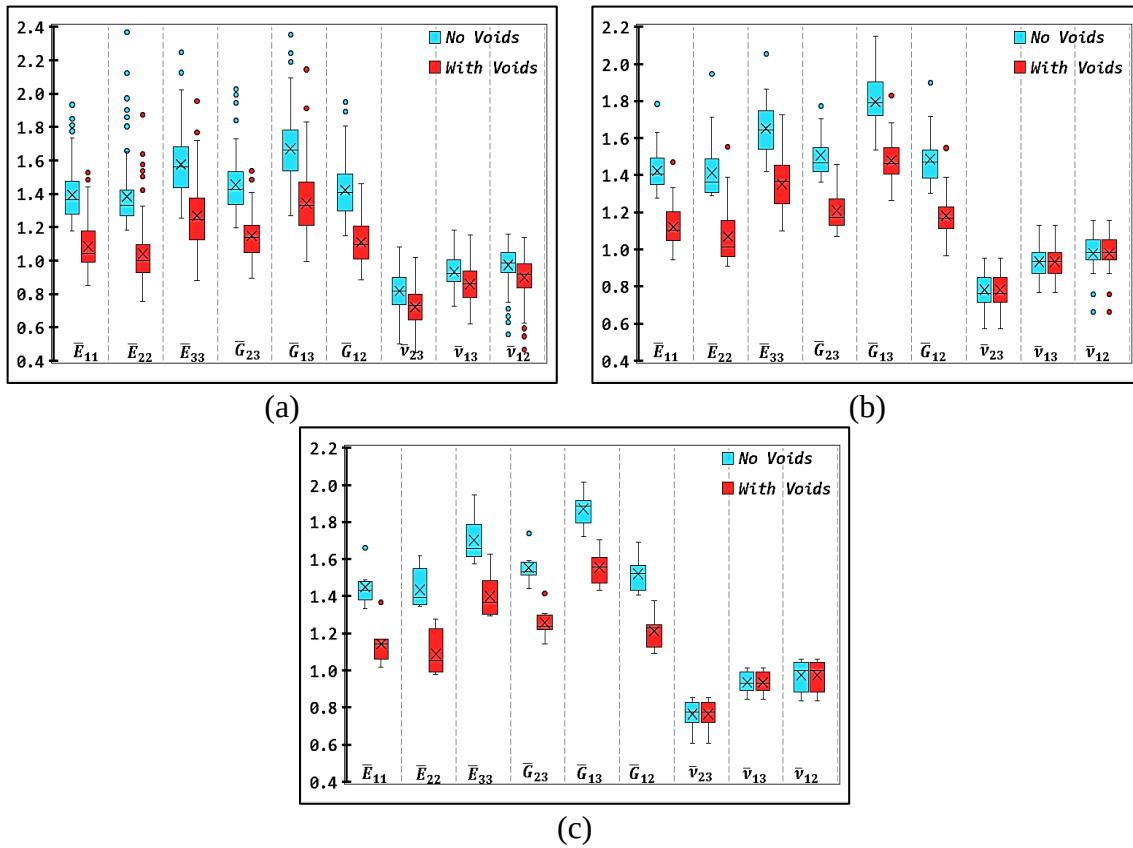


Figure 4.7: Error-bars showing the mean values, interquartile intervals, and outliers of the computed engineering constants from the RVE realization datasets for (a) RVE – I (b) RVE-II (c) RVE-III. Results are shown for analysis case with voids (red) and without voids (blue) present within the bead's microstructure.

Using volume information of ROI-II microstructural features including the distribution of the average fiber length, fiber orientation tensor and volume fractions of the fiber inclusions and micro-porosity within the complete set of realizations of each RVE cases (RVE- I, II & III), we

compute and compare the mean effective elastic modulus E^{eff} obtained from both Mori-Tanaka's mean-field homogenization approach and the numerical FEA approach using the method of Hills [306] according to eqn. (4.56). The relative error in the predicted values between both methods presented in Figure 4.6 below shows that the Mori-Tanaka estimates are comparable to the numerical predictions, mostly below 10% in the elastic moduli and the degree of accuracy is observed to improve somewhat with increasing RVE size. The original Mori-Tanaka-Benveniste model was formulated for two-phase composites with ellipsoidal inclusion and Norris A. N [112] has shown that the model's extension to multiphase inclusions may perform poorly and may violate the Hashin - Shtrikman stiffness bounds. Moreover, the Mori-Tanaka's predictions have been reported by Mortazavi et al. [56] and Breuer et al. [114] to deviate significantly from numerical estimates with increasing aspect ratio and volume fraction of the fiber inclusions.

Table 4.6: Relative error $\bar{\epsilon}$ [%] in the predicted effective elastic modulus magnitude, $\bar{E}^{eff}$ of ROI-II between Mori-Tanaka's analytical model and numerical FEA homogenization schemes for all RVE cases (RVE- I, II &III) and considering (a) non-porous microstructure (b) porous microstructure.

$\bar{E}^{eff}$	(a)			(b)		
	RVE-I	RVE-II	RVE-III	RVE-I	RVE-II	RVE-III
FE	1.46	1.52	1.56	1.13	1.20	1.23
MT	1.31	1.41	1.47	1.08	1.16	1.22
$\bar{\epsilon}$ [%]	10.21	8.20	5.86	4.49	3.38	1.49

The coefficient of variation ξ from the different RVE computations of ROI-II presented in Table 4.7 below shows that RVE – II is a sufficient for the purpose of predicting the elastic modulus of the CF/ABS composite based on the stipulated acceptance criteria ($\xi \leq 5\%$). Although the largest RVE, i.e. RVE-III yields slightly more accurate results, the computational requirements are excessive, and the gains of higher accuracy do not match the computational cost.

Table 4.7: Coefficient of Variation ξ [%] in the effective elastic modulus magnitude, $\bar{E}^{eff}$ for all RVE cases (I, II & III) of ROI-II and for both Mori-Tanaka's analytical model and numerical FEA homogenization schemes considering (a) non-porous microstructure (b) porous microstructure.

ξ	(a)			(b)		
	I	II	III	I	II	III
FE	7.13	3.91	2.37	7.78	4.23	2.47
MT	3.54	2.18	2.12	4.17	2.26	2.39

Partitioning of the ROI into smaller RVE volumes results in increased variability in average microstructural characteristics across the RVE realizations which potentially leads to increased dispersion in the predicted effective properties of the ROI volume as observed from the error-bar plots of Figure 4.7a-c.

4.2.1.2 Effective Coefficient of Thermal Expansion (ECTE)

In this section, we present the result of the effective coefficient of thermal expansion (ECTE) evaluated based on the numerical FE homogenization scheme. Representative displacement contour plot of a sample RVE instance (RVE – II, #14) from ROI-II region superposed over the undeformed structure subjected to a thermal load of $\Delta\theta = 65^\circ\text{C}$ appears in Figure 4.8 below. The mean values of the diagonal components of the ECTE tensor for the different RVE sizes, considering the non-porous and porous microstructure cases of the CF/ABS SFRP bead are presented in Table 4.8. The maximum observed discrepancy in the ECTE component values between consecutive RVE sizes is seen to drop from 6.3% between RVE-I & II to 3.3% between RVE-II & III.

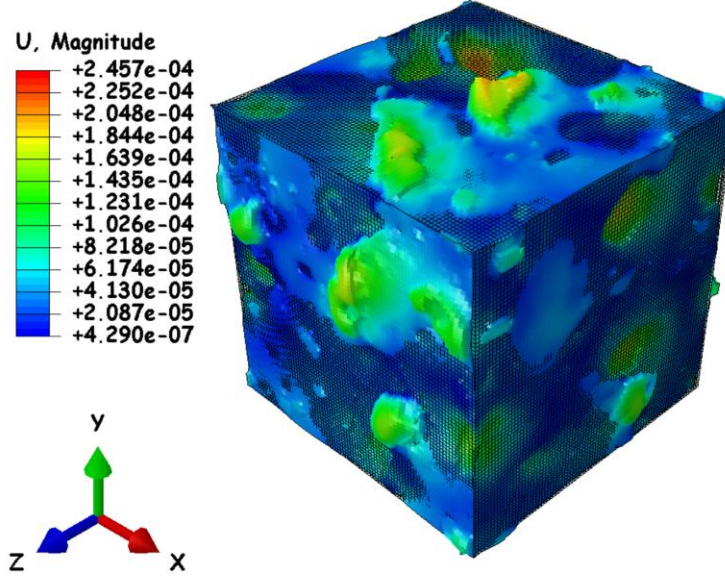


Figure 4.8: Deformation contour of RVE-II, #14 from ROI-II region overlayed on the undeformed mesh geometry and showing the bulk response of the volume under thermal load of $\Delta\theta = 65^\circ\text{C}$.

Table 4.8: Mean values of the diagonal components of the ECTE tensor for all RVE cases (I, II, & III) of ROI-II computed from the numerical FEA homogenization schemes and considering (a) non-porous microstructure (b) porous microstructure.

	(a)			(b)		
	$\bar{\alpha}_{11}$	$\bar{\alpha}_{22}$	$\bar{\alpha}_{33}$	$\bar{\alpha}_{11}$	$\bar{\alpha}_{22}$	$\bar{\alpha}_{33}$
RVE-I	0.83	0.91	0.70	0.81	0.89	0.67
RVE-II	0.82	0.90	0.66	0.79	0.88	0.63
RVE-III	0.81	0.89	0.64	0.79	0.87	0.61

From the error-bar plots in Figure 4.9a-c it is evident that the degree of dispersion in the predicted ECTE quantities reduces with increasing RVE size. We record higher variances in the predicted quantities with the smallest RVE (RVE-I) having more outliers outside the interquartile range (cf. Figure 4.9a) compared to largest RVE (RVE-III) with shorter error-bars and minimal dispersion in predicted ECTE quantities. From the results, the presence of porosity within the bead microstructure only slightly reduces the predicted ECTE values in all RVE cases (less than 5%). The estimated effects of the porosity on the volumetric ECTE values α_V are much lower (less than 3.25%). These conclusions are consistent with the conclusions of various literature [95], [128], [308].

The Mori-Tanaka's mean-field estimates of the apparent ECTE magnitude, $\bar{\alpha}^{eff}$ are also within range of the numerical FE predictions with a maximum observed discrepancy of about 5.5% for the non-porous composite and 7.0% for the porous composite (cf. Table 4.9).

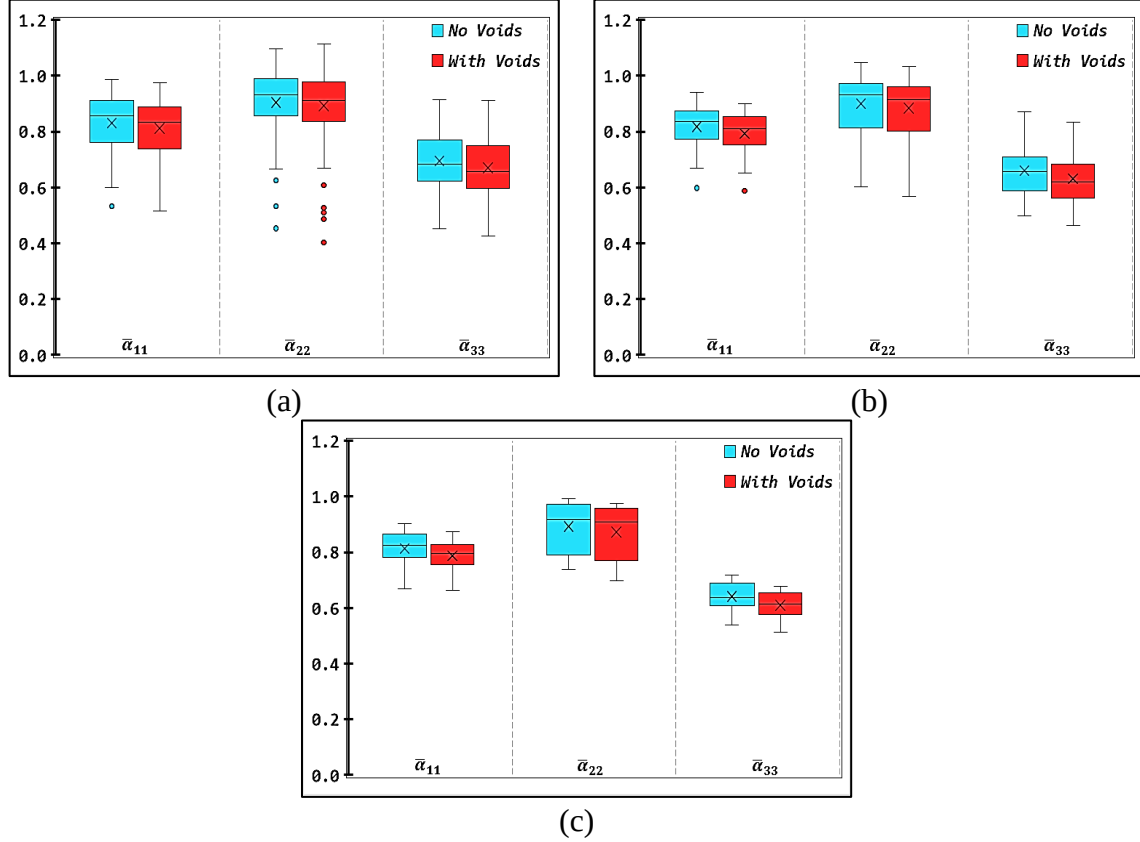


Figure 4.9: Error-bars showing the mean values, interquartile intervals, and outliers of the computed components of the ECTE tensor from the RVE realization datasets for (a) RVE – I (b) RVE-II (c) RVE-III. Results are shown for analysis case with voids (red) and without voids (blue) present within the bead's microstructure.

The accuracy of the analytical estimates is dependent on the accuracy of the calculated stiffness tensor and are observed to improve with increasing RVE size dropping to about 2.0% for the non-porous composite and 4.6% for the porous composite. Like the numerical FE results, we observe the same effect of the porosity on the analytical MT estimates of the apparent ECTE which leads to a reduction in estimated quantities generally below 1%.

Table 4.9: Relative error $\bar{\epsilon}$ [%] in the predicted normalized apparent ECTE magnitude ($\bar{\alpha}^{eff}$) of ROI-II between the numerical (FE) homogenization scheme and Mori-Tanaka's (MT) analytical estimate for all RVE sizes (RVE - I, II, & III) and considering (a) non-porous microstructure (b) porous microstructure.

$\bar{\alpha}^{eff}$	(a)			(b)		
	RVE-I	RVE-II	RVE-III	RVE-I	RVE-II	RVE-III
FE	0.81	0.79	0.78	0.79	0.76	0.75
MT	0.85	0.82	0.79	0.84	0.81	0.79
$\bar{\epsilon}$ [%]	5.45	3.85	2.00	7.02	6.07	4.59

Based on the results of the calculated coefficient of variation ξ for the apparent ECTE magnitude, $\bar{\alpha}^{eff}$ computed from the different realizations of the various RVEs of ROI-II presented in Table 4.10, we see that RVE – II is suitable for predicting the coefficient of thermal expansion based on the chosen error tolerance ($\xi \leq 5\%$).

Table 4.10: Coefficient of variation ξ [%] in the normalized apparent ECTE magnitude ($\bar{\alpha}^{eff}$) for all RVE cases (RVE- I, II &III) of ROI-II and for Mori-Tanaka's analytical model and numerical FEA homogenization schemes considering (a) non-porous microstructure (b) porous microstructure.

ξ	(a)			(b)		
	RVE-I	RVE-II	RVE-III	RVE-I	RVE-II	RVE-III
FE	4.00	2.28	1.12	5.36	3.16	1.61
MT	2.89	1.86	1.91	3.23	2.03	1.99

The impact assessment of voids on the ECTE although leads to a minimal reduction of the computed quantities, lower ECTE magnitudes may be desirable due to improved part dimensional stability. The impact studies may thus be more relevant to the effective composite stiffness and thermal conductivity.

4.2.1.3 Effective Thermal Conductivity (ETC)

The results of the ETC of the 13% CF/ABS composite based on the numerical evaluation procedures described in methodology sections are reported here. Typical contour plots of the temperature distribution for a sample non-porous RVE instance (RVE -II, #14) from ROI-II region

subjected to thermal gradient along the three principal references axes are shown in Figure 4.10a-c below. The plots reveal a non-uniform distribution of the temperature gradients along the principal coordinate axes due to the inherent microstructural heterogeneity across the composite coupled with the relatively high contrast in the isotropic thermal conductivity between both fiber and matrix phases ($\bar{\kappa}^f \sim 17.5$).

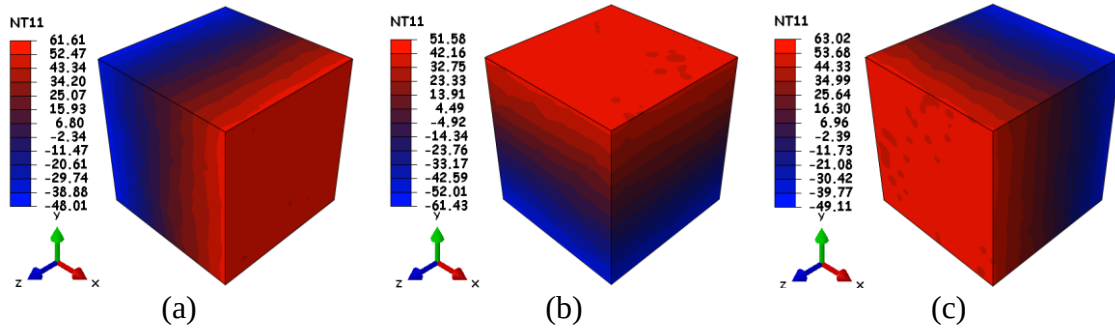


Figure 4.10: Temperature contours of RVE-II, #14 from ROI-II for different thermal loading (a) thermal gradient in x-direction (b) thermal gradient in y-direction (c) thermal gradient in z-direction.

Table 4.11 shows the principal components of the predicted ETC tensor for all three (3) RVE sizes, for both non-porous and porous composite microstructure. The effective mean values of all RVE considerations are seen to be within close range of each other. The inherent microvoids are seen to reduce the component values of the ETC by about 10% - 12%. Determination of sufficient RVE size is known to be dependent on the property being evaluated [116]. The maximum component of the conductivity tensor (i.e. $\bar{\kappa}_{33}$) is observed to coincide with the component of maximum average fiber orientation (i.e. $\hat{a}_{33}$).

Table 4.11: Mean values of the diagonal components of the ETC tensor for all RVE cases (RVE-I, II, & III) of ROI-II computed from the numerical FEA homogenization schemes and considering (a) non-porous microstructure (b) porous microstructure.

	(a)			(b)		
	$\bar{\kappa}_{11}$	$\bar{\kappa}_{22}$	$\bar{\kappa}_{33}$	$\bar{\kappa}_{11}$	$\bar{\kappa}_{22}$	$\bar{\kappa}_{33}$
RVE-I	1.39	1.32	1.47	1.22	1.15	1.31
RVE-II	1.40	1.33	1.49	1.24	1.16	1.33
RVE-III	1.41	1.33	1.50	1.24	1.16	1.35

From the error-bar plot of Figure 4.11a-c we see a clear reduction in the dispersion of quantities as the RVE size increases from RVE-I (cf. Figure 4.11a) to RVE-III (cf. Figure 4.11c). The error-bar shrinks considerably for largest RVE case, i.e. RVE-III, although the mean values for all three RVE cases are within close range to each other with maximum observed discrepancy in all quantities between consecutive RVE sizes for both porous and non-porous microstructural considerations dropping from about 1.8% between RVE-I & II to a value of only 0.8% between RVE II & III.

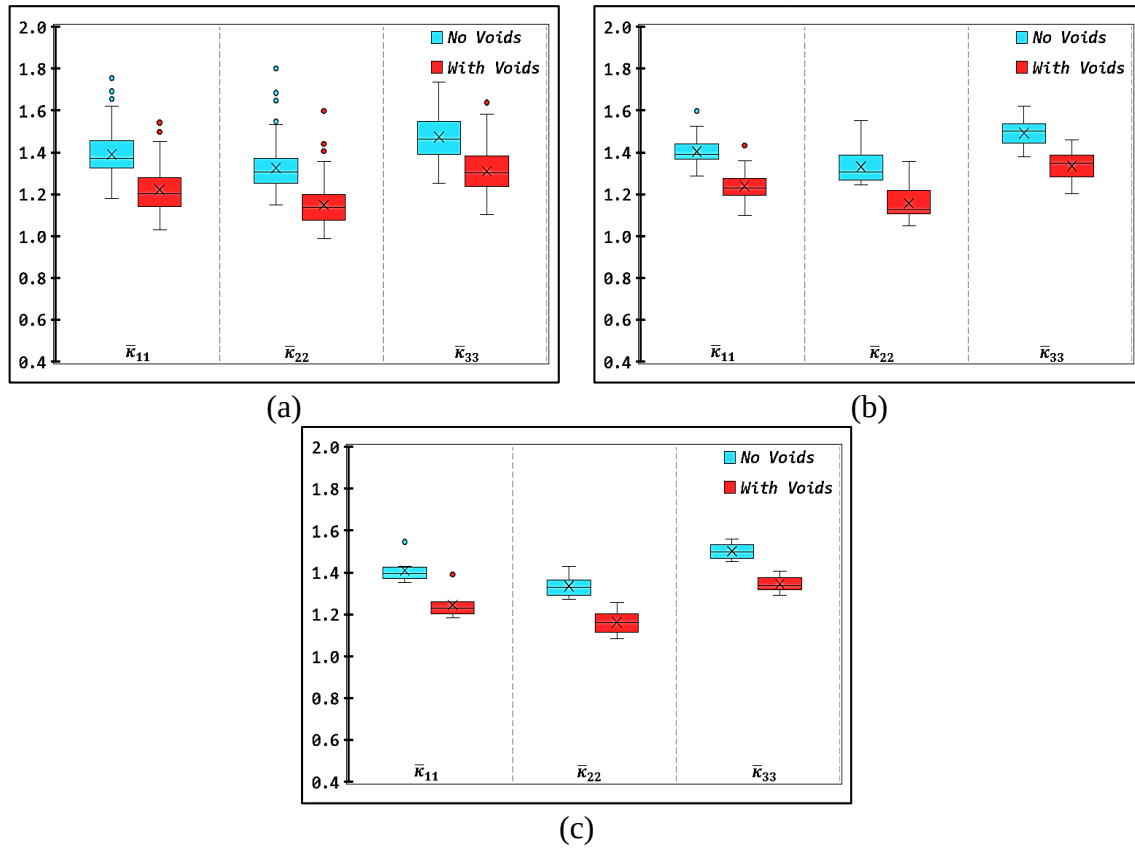


Figure 4.11: Error-bars showing the mean values, interquartile intervals, and outliers of the diagonal components of the ETC tensor from the RVE realization datasets for (a) RVE-I (b) RVE-II (c) RVE-III. Results are shown for analysis case with voids (red) and without voids (blue) present within the bead's microstructure.

The results of the apparent ETC magnitude κ^{eff} presented in Table 4.12 below shows very good agreement between the values obtained from both numerical (FE) homogenization and the

Mori-Tanaka (MT) analytical methods with a maximum discrepancy of only 3.5% recorded for the smallest sized RVE (RVE-I) which reduces with increasing RVE size to about 1% for RVE-III.

Table 4.12: Relative error $\bar{\epsilon}$ [%] in the predicted ETC magnitude ($\bar{\kappa}^{eff}$) of ROI-II between the numerical FEA homogenization scheme and Mori-Tanaka's (MT) analytical estimate for all RVE sizes (RVE - I, II, & III) and considering (a) non-porous microstructure (b) porous microstructure.

$\bar{\kappa}^{eff}$	(a)			(b)		
	RVE-I	RVE-II	RVE-III	RVE-I	RVE-II	RVE-III
FE	1.40	1.41	1.42	1.23	1.24	1.25
MT	1.35	1.38	1.40	1.18	1.22	1.24
$\bar{\epsilon}$ [%]	3.48	1.79	0.98	3.46	1.95	1.16

Although there is minimal disparity in the results of the mean values of the computed ETC tensor components among the different RVE sizes in Table 4.11, however results of the dispersion in the measured ETC quantities from Table 4.13 shows that RVE-I is insufficient in predicting the ETC of the ROI-II based on the given dispersion tolerance criteria ($\xi \leq 5\%$). Thus RVE-II is the minimum sufficient size for predicting the ETC quantity ($\xi < 3\%$) for both porous and non-porous microstructure consideration. Although we observe less dispersion in the computed ETC quantities for RVE-III ($\bar{\xi} < 2\%$), the gain in accuracy does not measure up to the added cost of computation due to the increased RVE size and associated mesh points.

Table 4.13: Coefficient of variation ξ [%] in the predicted normalized apparent ETC ($\bar{\kappa}^{eff}$) of ROI-II for both Mori-Tanaka's analytical model and numerical FEA homogenization schemes and for all RVE cases (RVE- I, II & III) and considering (a) non-porous microstructure (b) porous microstructure.

ξ	(a)			(b)		
	RVE-I	RVE-II	RVE-III	RVE-I	RVE-II	RVE-III
FE	5.63	2.92	1.78	5.49	2.80	1.83
MT	4.85	2.59	1.83	4.86	2.47	1.93

4.2.2 Thermo-Mechanical Property Estimates for Different Bead Regions

To better understand the impact of the spatial variation in the bead microstructure on the effective thermo-mechanical properties across the bead specimen, including the variations in the concentrations and characteristics of the micro-constituent phases, analysis is performed by selecting a single characteristic RVE instance of type II (i.e. RVE-II) from all four (4) ROIs [297] with matching microstructural characteristics in order to evaluate their effective properties that reflects the overall properties of their respective ROI volume. We have previously established that RVE-II is sufficient in predicting the effective properties of the 13% CF/ABS SFRP composite based on our acceptance criteria. The characteristic RVE instance is selected such that its microstructural characteristics only minimally deviate from that of the corresponding ROI volume. We argue that if the average value of a mathematical descriptor that defines the microstructural characteristics of a RVE instance chosen from a particular ROI volume deviate minimally from the corresponding value of the overall ROI volume, then the average effective properties of the characteristic RVE instance should also deviate minimally from the overall ROI volume. We define the measure of deviation in the effective property Z of the j^{th} ROI-RVE instance (i.e. δZ_j) as

$$\delta Z_j = \frac{Z_j^{RVE} - Z^{ROI}}{Z^{ROI}}, \quad Z = \vartheta_f, \quad \vartheta_v, \quad \bar{E}^{eff}, \quad \bar{\alpha}^{eff}, \quad \bar{\kappa}^{eff} \quad (4.59)$$

For the average fiber orientation $\underline{\hat{p}}$, the measure of the deviation in the average fiber orientation of the j^{th} ROI-RVE instance, δp_j is defined as

$$\delta p_j = 1 - \cos \delta \varphi_j = 1 - \frac{\underline{\hat{p}}_j^{RVE} \cdot \underline{\hat{p}}^{ROI}}{|\underline{\hat{p}}_j^{RVE}| |\underline{\hat{p}}^{ROI}|} \quad (4.60)$$

The 3D regression plots of Figure 4.12a-c shows the relationship between the deviation in the evaluated effective properties with the deviation in the volume fractions and average fiber

orientation for the various non-porous RVE-II instances of ROI-II volume. In Figure 4.12a, the deviation in the effective modulus of the non-porous RVE-II instances from the ROI-II mean modulus drops to a instance minimum value of about $\delta \bar{E}_{24}^{eff} = 0.03\%$ for RVE-II, #24 instance in ROI-II region where the maximum instance deviation in $\bar{E}^{eff}$, (i.e. $\delta \bar{E}_{max}^{eff}$) reaches 8.12%. The RVE-II instance with the minimum $\delta \bar{E}^{eff}$ corresponds to the RVE-II instance with the minimum deviation in the fiber volume fraction $\delta \vartheta_{f_{24}} = 0.13\%$ (i.e. RVE-II, #24) in ROI-II, region where $\delta \vartheta_f$ reaches a maximum value of 16.68%. The deviation in the average fiber orientation vector of the corresponding RVE-II, #24 instance at the discrete minimum location is seen to be about $\delta p_{24} = 0.004$ (or $\delta \varphi_{24} = 5.33^\circ$) at the minimum point although $\delta \varphi_{24}$ is not the minimum value of the complete set of RVE-II instances in ROI-II region. $\delta \varphi$ reaches a maximum instance value of $\delta p_{max} = 0.04$ (or $\delta \varphi_{max} = 16.20^\circ$) within the ROI-II region. We assume all effective properties ($\bar{E}^{eff}$, $\bar{\alpha}^{eff}$, $\bar{\kappa}^{eff}$) are weighted equally in terms of their importance in determining a suitable RVE instance. As such we define an objective function $\delta Y^{eff} = [\delta \bar{E}^{eff} + \delta \bar{\alpha}^{eff} + \delta \bar{\kappa}^{eff}]/3$ to minimize. Based on the given objective function, δY^{eff} , we yet arrive at the same RVE instance that yields the minimum value of the objective function (i.e. ROI-II, RVE-II, #24) where the fiber volume fraction $\delta \vartheta_f$ is minimum. The associated values of the deviation in the effective properties (cf. Figure 4.12 a-c) are well below 1% ($\delta \bar{\alpha}_{24}^{eff} = 0.33\%$, $\delta \bar{\kappa}_{24}^{eff} = 0.11\%$). At the minimum point of the regression lines for the deviation in all effective properties where $\delta \bar{E}^{eff} = \delta \bar{\alpha}^{eff} = \delta \bar{\kappa}^{eff} = 0$ (cf. Figure 4.12a-c), the deviation in the fiber volume fraction $\delta \vartheta_f$ approaches zero ($\delta \vartheta_f = 0.18\%$) however the deviation in the average fiber orientation is small but not zero ($\delta p = 0.007$ or $\delta \varphi = 6.98^\circ$). RVE-II, #24 instance has the

closest matching characteristics to the minimum point of the regression line amongst other instances in ROI-II volume.

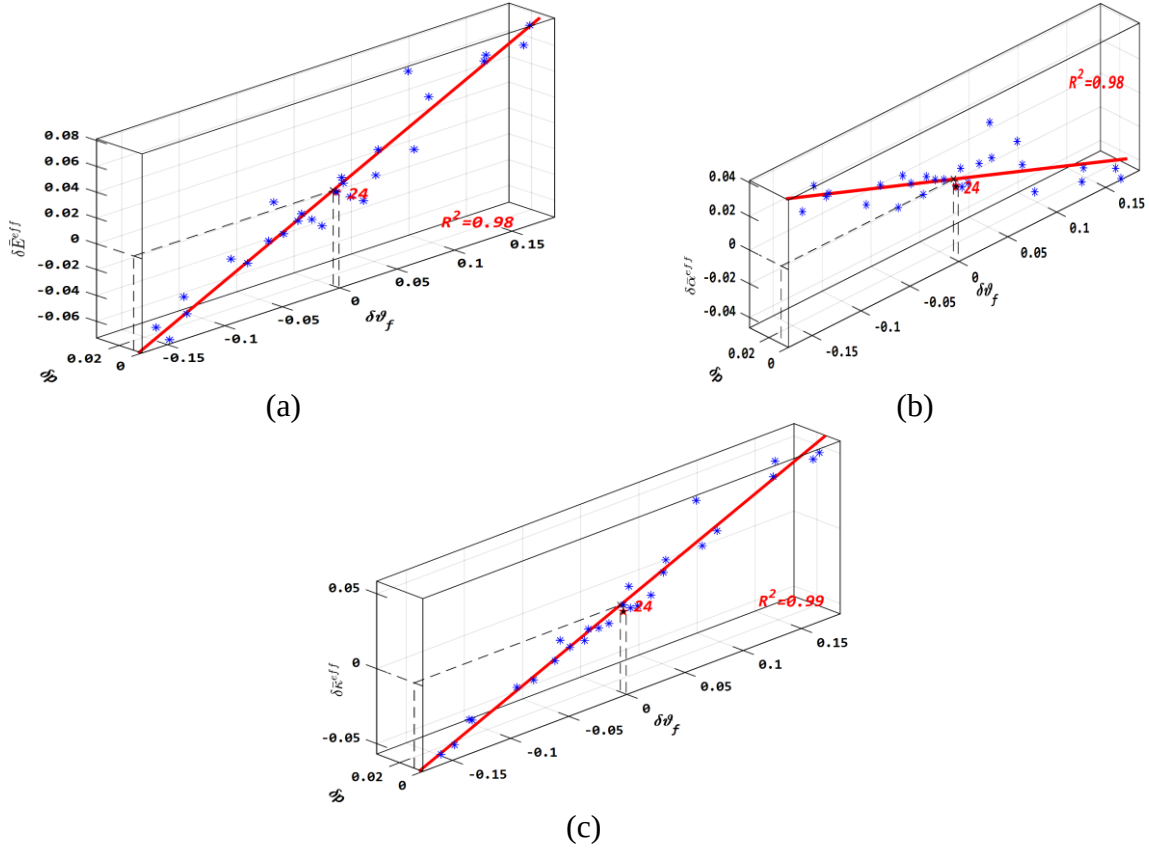


Figure 4.12: 3D correlation plot between the distribution of the deviation in the fiber fraction $\delta\vartheta_f$ and orientation vector $\delta\rho$ versus the deviation in the (a) apparent effective modulus, $\delta\bar{E}^{eff}$ (b) apparent ECTE, $\delta\bar{\alpha}^{eff}$ and (c) apparent ETC, $\delta\bar{\kappa}^{eff}$, for the various RVE-II instances of ROI-II.

Minimization of $\delta\vartheta_f$ takes precedence to minimizing δp (or $\delta\varphi$) in selecting an appropriate RVE instance from a given ROI based on the foregoing arguments. On the contrary, minimizing δp prior to minimizing $\delta\vartheta_f$, gives a non-optimal RVE instance (RVE-II, #7) that yields unacceptable objective function values, i.e. at $\delta p_{min} = 0.0003$, or $\delta\varphi_{min} = 1.46^\circ$, $\delta\vartheta_f = 15.95\%$, $\delta\bar{E}_7^{eff} = 5.69\%$, $\delta\bar{\alpha}_7^{eff} = 3.01\%$, and $\delta\bar{\kappa}_7^{eff} = 5.03\%$ which shows significant deviations in the microstructural characteristics between RVE-II, #7 and ROI-II region. Moreover, the effective properties are known from literature to depend strongly on the fiber volume fraction and very weakly on the average fiber orientation [114], [125].

Accordingly, we select characteristic RVE-II instances from the various ROI volumes following the minimal parameter deviation approach using volume information of the deviation in the inherent microstructure of the various RVE instances from their respective ROI volumes as presented in scatter plots of Figure 4.13a-d below. The selected characteristic RVE-II instances for the different ROI volumes include (a) ROI-I, RVE-II, #14 (b) ROI-II, RVE-II, #24 (c) ROI-III, RVE-II, #23 (d) ROI-IV, RVE-II, #3.

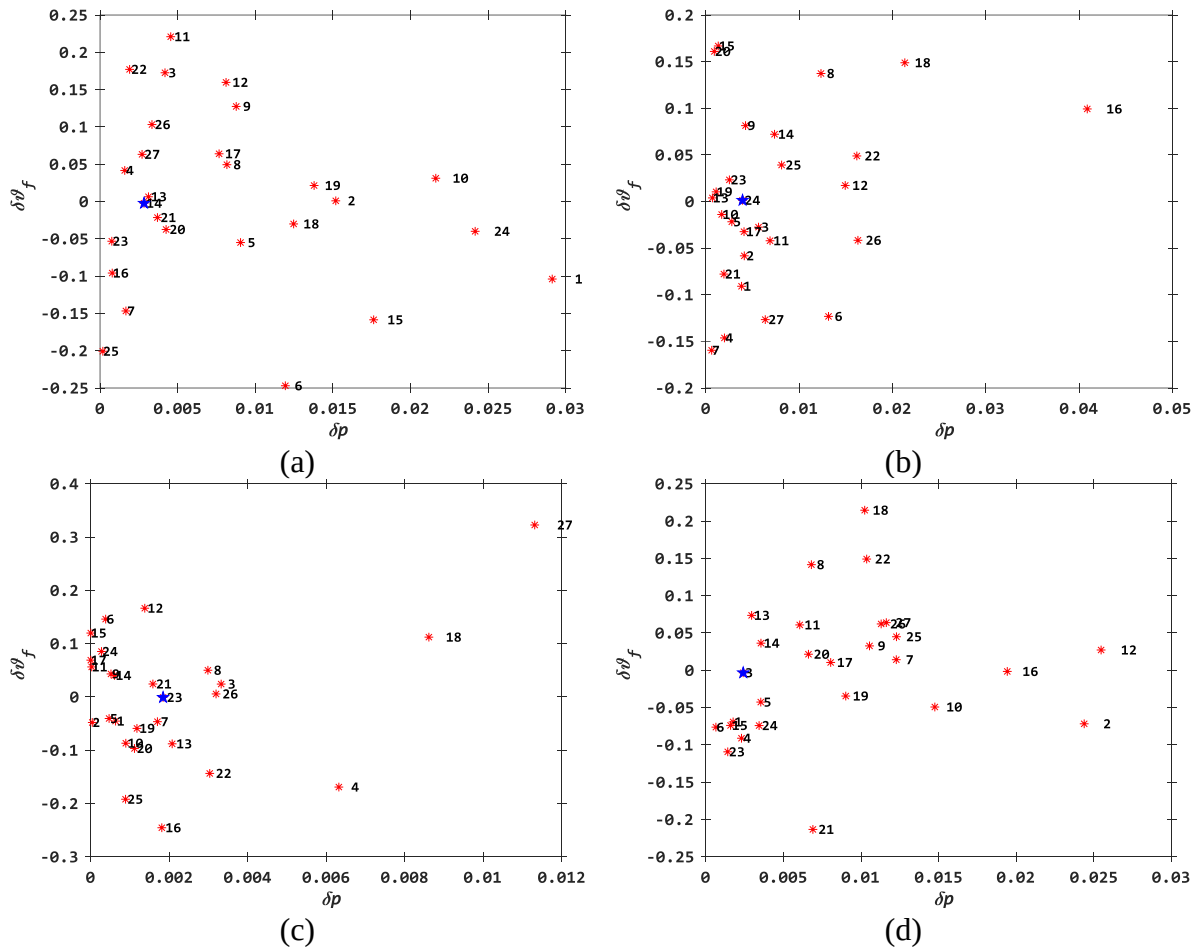


Figure 4.13: Scatter showing the deviations in the fiber volume fraction and average fiber orientation for the different RVE-II realizations of the various ROI volumes (a) ROI-I, (b) ROI-II, (c) ROI-III, and (d) ROI-IV.

The resulting deviations in the important microstructural parameters for the selected characteristic RVE instances from the respective non-porous ROI volumes are presented in Table

4.14 For all selected cases, the minimum deviation in the fiber volume fraction $\delta\vartheta_f$ is seen to be below 0.35% and the minimum deviation in the average fiber orientation vector $\delta\varphi$ is seen to be below 5.5° .

Table 4.14: Deviation in the relevant microstructural parameters for the non-porous microstructure between the characteristic RVE instance of the different ROI volumes (a) ROI-I, RVE-II, #14 (b) ROI- II, RVE-II, #24, (c) ROI- III, RVE-II, #23, and (d) ROI- IV, RVE-II, #3

	(a)	(b)	(c)	(d)
$\delta\vartheta_f$ [%]	-0.23	0.13	-0.10	-0.33
$\delta\vartheta_v$ [%]	3.73	-10.26	18.92	-9.31
$\delta\varphi$ [deg]	4.45	5.33	3.86	3.56

Figure 4.14a-d shows the microstructure of the selected characteristic RVE-II instances from the four (4) ROI volumes obtained from 3D X-ray μ CT imaging. The figures show that the characteristic RVE instances are representative of the various ROI regions. The estimated average fiber volume fractions ϑ_f and the diagonal components of the second order fiber orientation tensor for the various ROI - RVE's are presented in Table 4.15 below. By mere visual inspection of Figure 4.14a-d, the reported values in Table 4.15 can be corroborated. ROI-III, RVE-II, #23 instance and ROI- IV, RVE-II, #3 are seen to be more densely packed than the other RVE volumes hence their high fiber volume fractions. Likewise, ROI- III, RVE-II, #23 can be seen to have the highest alignment with the z-direction appearing almost vertical, compared to other region instances.

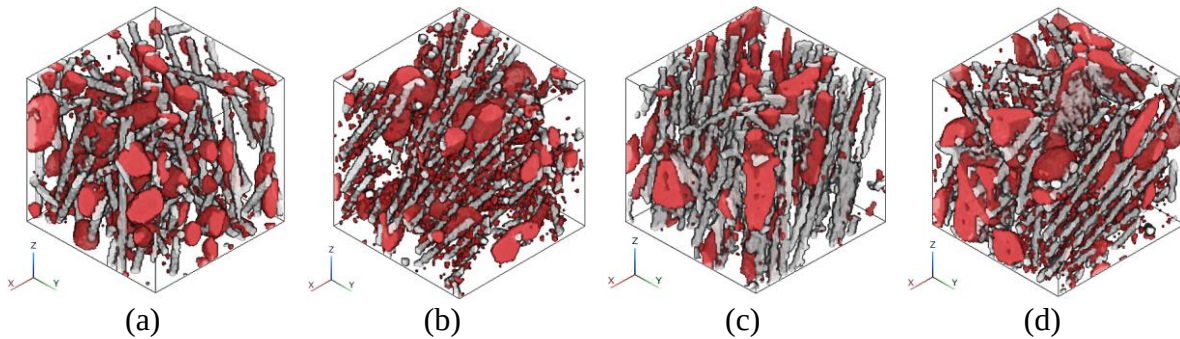


Figure 4.14: 3D-μCT volume view showing internal microstructure (fiber -gray, voids – red) of the characteristic RVE-II instances of the various ROI volumes (a) ROI-I, RVE-II, #14 (b) ROI-II, RVE-II, #24, (c) ROI-III, RVE-II, #23, and (d) ROI-IV, RVE-II, #3.

Table 4.15: Average values of the fiber volume fraction and diagonal orientation components for the characteristic RVE instances (a) ROI-I, RVE-II, #14 (b) ROI-II, RVE-II, #24, (c) ROI-III, RVE-II, #23, and (d) ROI-IV, RVE-II, #3

	(a)	(b)	(c)	(d)
ϑ_f [%]	6.95	6.66	7.24	7.51
ϑ_v [%]	11.10	11.01	11.96	10.13
$\langle p_x p_x \rangle$	0.34	0.25	0.08	0.22
$\langle p_y p_y \rangle$	0.04	0.23	0.08	0.20
$\langle p_z p_z \rangle$	0.62	0.52	0.84	0.58

Using the same numerical FE homogenization procedure detailed in the methodology section, we evaluate the effective properties of the characteristic RVE instances from each ROI shown in Figure 4.14a-d for the non-porous microstructure which are presented in Table 4.16 below. The results of the predicted quantities computed based on the single RVE instances chosen from each ROI volume are expected to be within range of the actual mean values computed from the complete set of RVE realizations of each ROI since the calculated RVE/ROI deviations in the relevant microstructural properties (cf. Table 4.14) are within the acceptable tolerance (i.e. $|\delta\vartheta_f| \leq 0.33\%$, $\delta\varphi \leq 5.33^\circ$). The reported effective quantities for the non-porous microstructure of the various single characteristics RVE-II instances from their respective ROIs in Table 4.16 were found to be comparable with average values of the effective quantities obtained from the overall set of realizations of RVE-II for each ROI with a maximum observed discrepancy of only 1.7 %. As expected, the results show that the effective modulus $\bar{E}^{eff}$ and thermal conductivity $\bar{\kappa}^{eff}$ increase with increasing fiber volume fraction and increasing degree of fiber alignment with the print direction (z-axis) across the different bead regions. Conversely, the ECTE $\bar{\alpha}^{eff}$ is seen to decrease with increasing fiber volume fraction and increasing fiber alignment with the print direction.

Table 4.16: (a) Estimated values of effective thermo-mechanical properties for the various non-porous microstructure of the selected RVE-II instances of the ROI volumes (a) ROI-I, RVE-II, #14 (b) ROI-II, RVE-II, #24, (c) ROI-III, RVE-II, #23, and (d) ROI-IV, RVE-II, #3.

	(a)	(b)	(c)	(d)
$\bar{E}^{eff}$	1.54	1.52	1.55	1.60
$\bar{\alpha}^{eff}$	0.76	0.79	0.77	0.74
$\bar{\kappa}^{eff}$	1.43	1.41	1.45	1.47

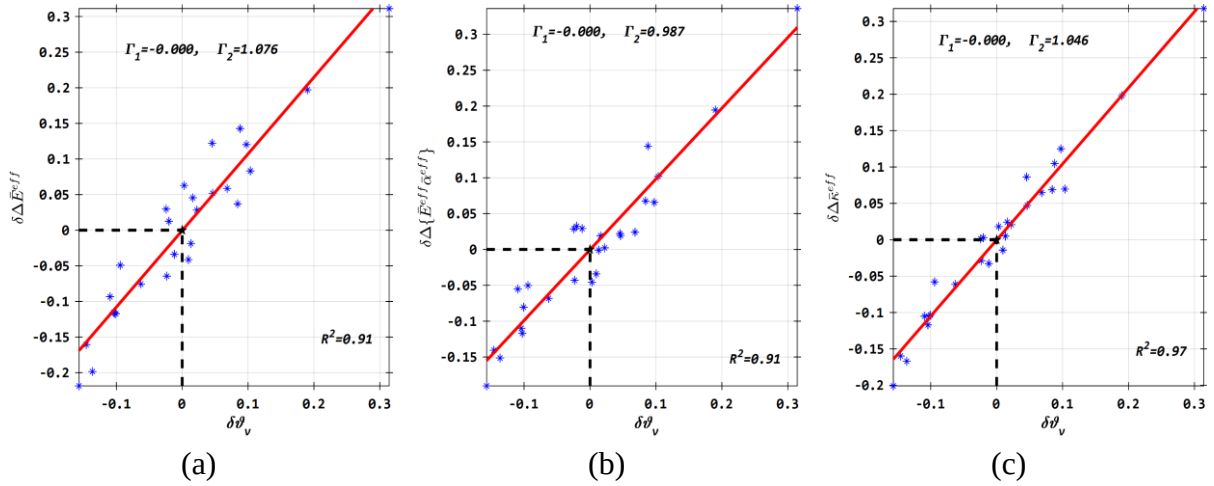


Figure 4.15: Correlation plot between the distribution of the deviation in the (a) net effective modulus (b) net product of the effective modulus and thermal expansion coefficient (c) net thermal conductivity; versus the deviation in the void fraction for the various RVE-II instances of ROI-II volume.

Once the effective properties of the different non-porous ROIs have been computed using relevant RVE instances, the effect of the inherent porosity on the properties can be approximated using established linear relationships that correlate the deviation in the effective properties difference, $\delta\Delta Z$ due to the inherent porosity to the deviation in volume fraction of the porosity, $\delta\vartheta_v$ between the selected RVE instances and their respective ROI volumes. (i.e. ΔZ is the difference in the magnitude of effective property Z between the porous and non-porous microstructure). The inherent porosity is assumed to impact primarily the integrity of the polymer matrix and the contribution of its properties to the overall behavior of the composite material. Accordingly, developed linear relationship, $\delta\Delta Z = \Gamma_1 + \Gamma_2\delta\vartheta_v$ in Figure 4.15a-c for ROI-II would apply to other ROI regions across the entire bead section. The ECTE values are weighted with

their corresponding elastic modulus values when developing its linear relationship. Given the difference between the porous and non-porous effective properties for the characteristic RVE-II instances of the various ROI volumes, ΔZ^{RVE} (cf. Table 4.17), we can backtrack the associated difference between the porous and non-porous effective properties of the various ROI volumes, ΔZ^{ROI} from eqn. (4.59) using the linear relationships presented in Figure 4.15a-c. Consequently, given ΔZ^{ROI} and the effective properties of the non-porous microstructure of the various ROI volumes Z^{ROI} (cf. Table 4.16), we compute the approximate effective properties of the porous microstructure of the various ROI volumes (cf. Table 4.18). Again, the evaluated effective quantities for the porous microstructure presented in Table 4.18 based on volume information of the various single characteristics RVE-II instances from their respective ROI were found to match closely with the mean values of the effective quantities obtained from the complete set of RVE-II realizations for each ROI with a maximum observed discrepancy of 2.5%. As expected, the porosity is observed to reduce the effective properties in the different ROI regions of the bead.

Table 4.17: (a) Estimated values of difference in the effective thermo-mechanical properties (ΔZ^{RVE}) between the porous and non-porous microstructure for the selected RVE-II instances of the various ROI volumes (a) ROI-I, RVE-II, #14 (b) ROI-II, RVE-II, #24, (c) ROI- III, RVE-II, #23, and (d) ROI- IV, RVE-II, #3.

	(a)	(b)	(c)	(d)
$\Delta \bar{E}^{eff}$	0.31	0.29	0.40	0.29
$\Delta \bar{\alpha}^{eff}$	0.04	0.02	0.02	0.05
$\Delta \bar{\kappa}^{eff}$	0.15	0.15	0.17	0.14

Table 4.18: (a) Approximate values of effective thermo-mechanical properties for the porous microstructure of the various ROI volumes (a) ROI-I, RVE-II, #14 (b) ROI- II, RVE-II, #24, (c) ROI- III, RVE-II, #23, and (d) ROI- IV, RVE-II, #3.

	(a)	(b)	(c)	(d)
$\bar{E}^{eff}$	1.24	1.20	1.23	1.27
$\bar{\alpha}^{eff}$	0.72	0.76	0.76	0.68
$\bar{\kappa}^{eff}$	1.28	1.24	1.31	1.31

4.2.3 Effective Property Correlation Studies

To better understand the variation of the effective quantities with variation in microstructural features across the 13% CF/ABS bead we evaluate the effective quantities for the complete set of RVE-II realizations for the various ROI volumes shown in Figure 4.16, and correlate the computed quantities with the relevant microstructural information of the various instances across each ROI volumes.

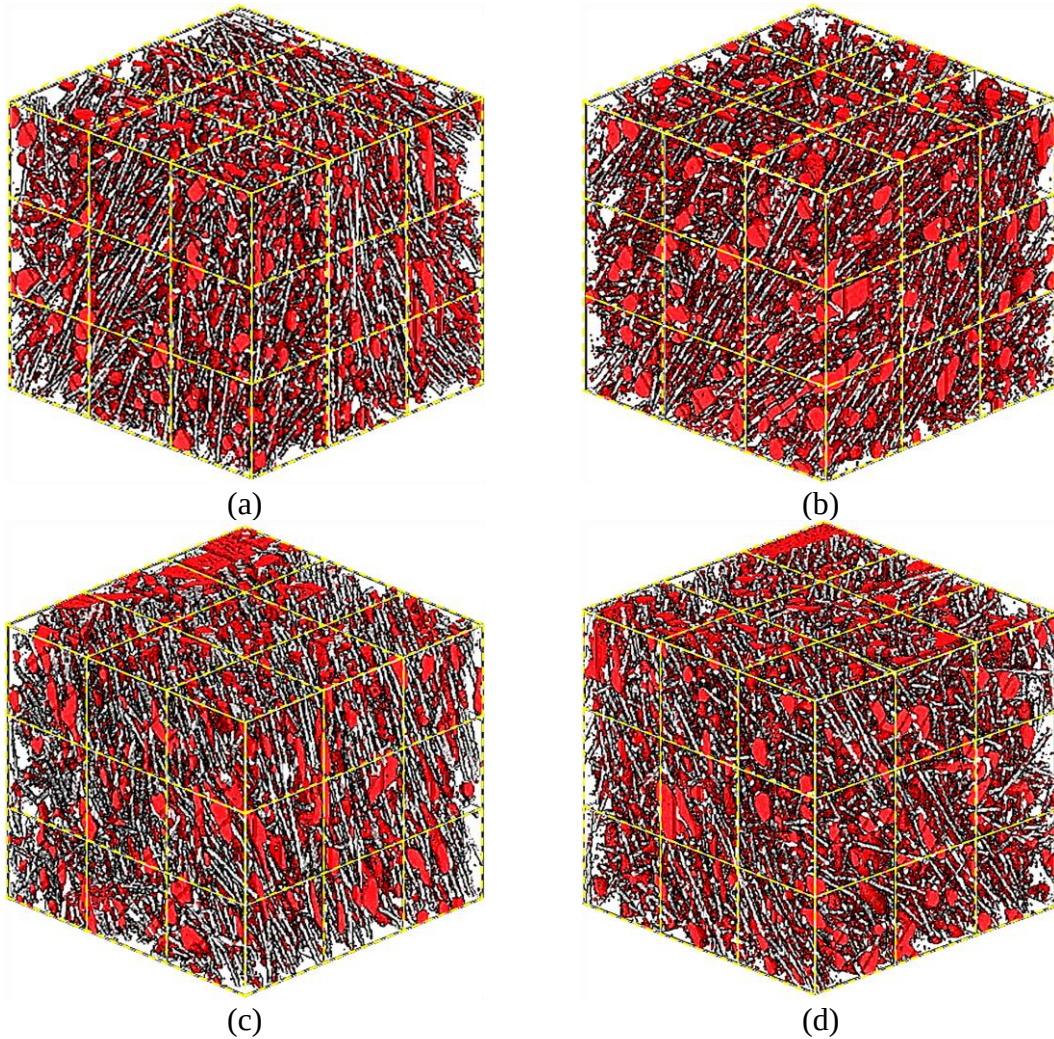


Figure 4.16: 3D- μ CT volume view showing internal microstructure (fiber -gray, voids – red) of the various ROI volumes and their partitioning into the various RVE-II instances for (a) ROI-I, (b) ROI- II, (c) ROI- III (d) ROI- IV.

Figure 4.17a presents linear correlation fits of the computed values of the effective elastic modulus $\bar{E}^{eff}$ with the average fiber volume fraction ϑ_f and Figure 4.17b shows the correlation between the $\bar{E}_{33}$ elastic modulus component and $\hat{a}_{33}$ average fiber orientation tensor component obtained from realization datasets for the non-porous RVE-II instances of the various ROIs. From the results, we observe good correlation between the fiber volume fraction and effective elastic modulus with a correlation coefficient $R^2 = 0.94$ which implies that the fiber volume fraction is a salient microstructural parameter for predicting the modulus of SFRP composites. We likewise observe reasonable correlation between the $\bar{E}_{33}$ elastic modulus component and $\hat{a}_{33}$ average fiber orientation tensor component with a correlation coefficient $R^2 = 0.75$, which suggests that the degree of fiber alignment with the print direction directly relates to the resulting effective elastic properties of the SFRP composite.

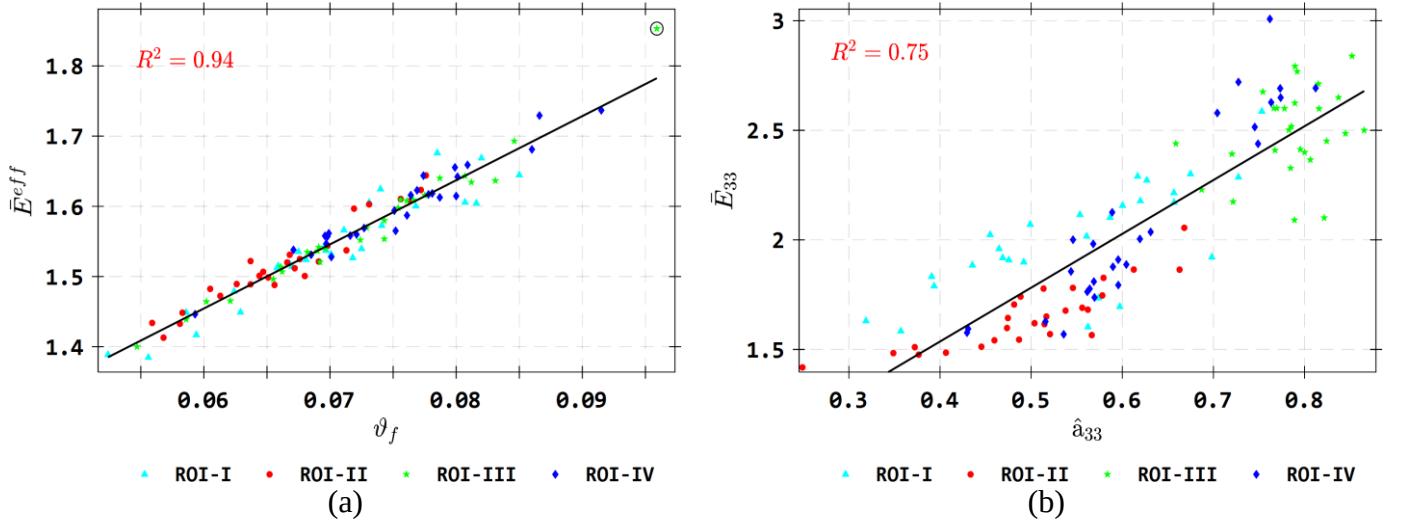


Figure 4.17: Linear correlation plots between the (a) effective elastic modulus $\bar{E}^{eff}$ and the fiber volume fraction ϑ_f (b) elastic modulus component $\bar{E}_{33}$ and the average fiber orientation tensor component $\hat{a}_{33}$; for non-porous RVE-II instances of the various ROIs.

Linear correlations fit of the apparent ECTE magnitude $\bar{\alpha}^{eff}$ with the average fiber volume fraction ϑ_f for the non-porous RVE-II realizations of the various ROIs shown in Figure 4.18a reveals an inverse relation between both quantities. With higher concentration of the reinforcing

particles, the ECTE decreases in magnitude. The relatively lower ECTE values of the fiber inclusions effectively reduces the average homogenized ECTE of the composite material in line with the conclusions of various literature [95], [128], [308]. The result shows good correlation between $\bar{\alpha}^{eff}$ and ϑ_f with correlation coefficient $R^2 = 0.81$. We likewise observe good correlation between the average fiber orientation component in the print direction $\hat{a}_{33}$ and the $\bar{\alpha}_{33}$ component of ECTE tensor ($R^2 = 0.85$) in Figure 4.18b. The degree of fiber alignment in the print direction $\hat{a}_{33}$ is observed to vary inversely with the $\bar{\alpha}_{33}$ component of the ECTE tensor along the same direction. Higher levels of fiber alignment results in high packing density which invariably results in higher fractions of fiber contained within the volume.

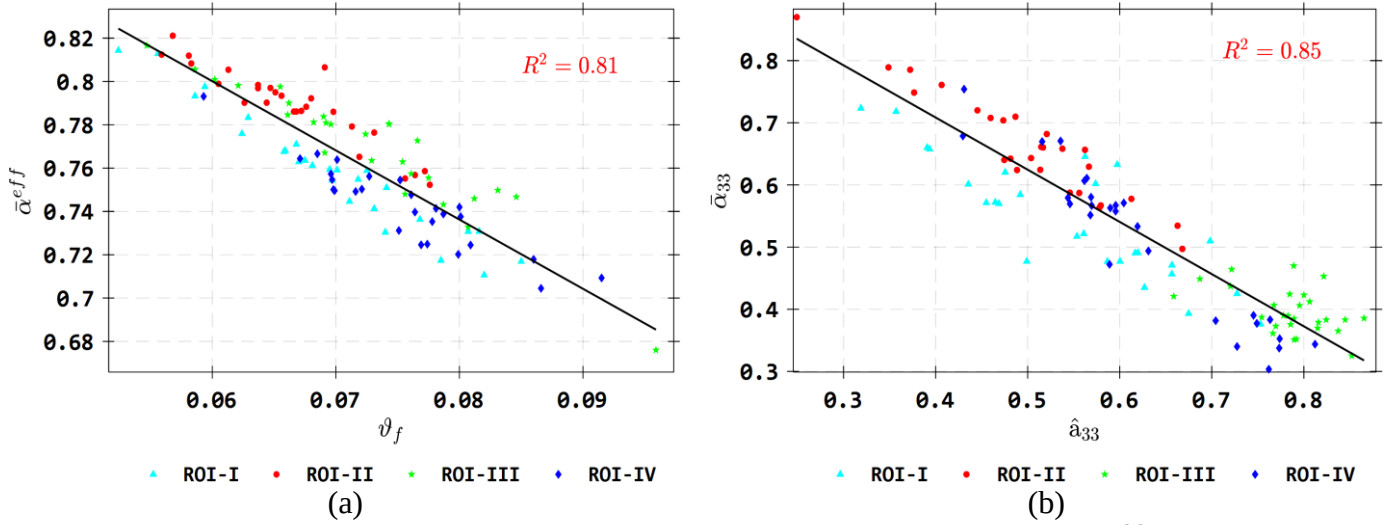


Figure 4.18: Linear correlation plots between (a) the apparent ECTE magnitude $\bar{\alpha}^{eff}$ with the fiber volume fraction ϑ_f (b) the ECTE tensor component $\bar{\alpha}_{33}$ and the average fiber orientation tensor component $\hat{a}_{33}$; for non-porous RVE-II instances of the various ROIs.

Additionally, we observe there is a very high correlation ($R^2 = 1.0$) between the fiber volume fraction ϑ_f and apparent ETC magnitude $\bar{\kappa}^{eff}$ for the non-porous RVE-II instances of various ROIs (cf. Figure 4.19a), however a very weak correlation ($R^2 = 0.66$) between the average fiber orientation component in the print direction $\hat{a}_{33}$ and the $\bar{\kappa}_{33}$ component of the ETC tensor (cf. Figure 4.19b). This implies that for the two-phase SFRP composite, the apparent ETC

$\bar{\kappa}^{eff}$ has a strong linear dependence on the volume fraction of the fiber reinforcement. This conclusion is consistent with the findings of Tian et. al [305].

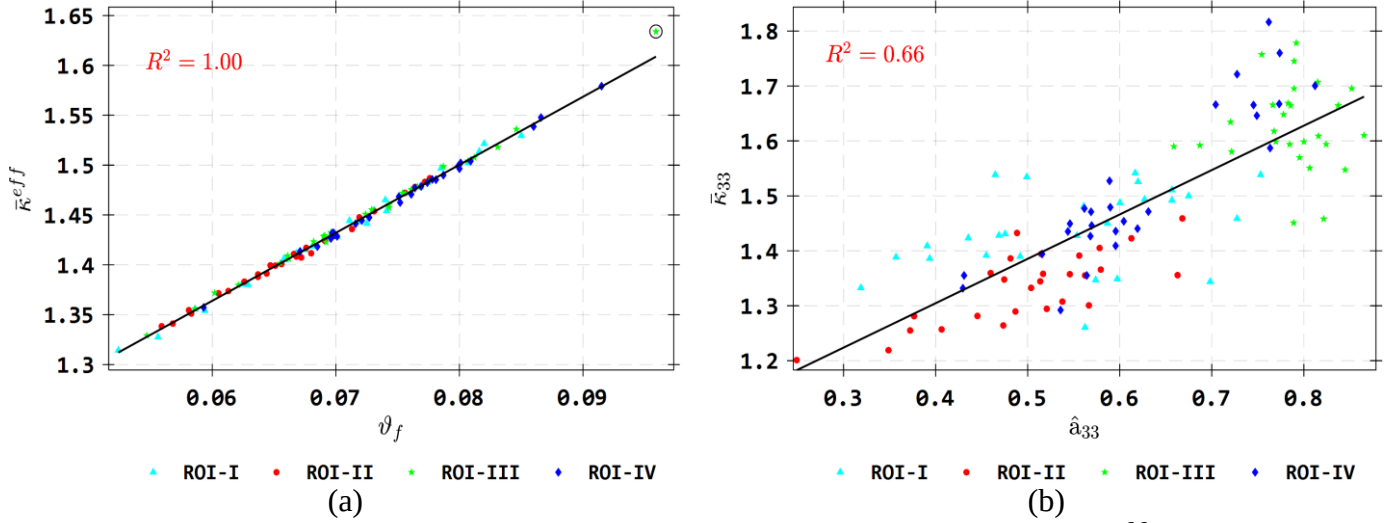


Figure 4.19: Linear correlation plots between (a) the apparent ETC magnitude $\bar{\kappa}^{eff}$ and the fiber volume fraction ϑ_f (b) the ETC tensor component $\bar{\kappa}_{33}$ and the average fiber orientation tensor component $\hat{a}_{33}$; for non-porous RVE-II instances of the various ROIs.

4.3 Conclusion

Numerical FEA homogenization method was utilized in the current investigation to evaluate effective thermo-mechanical properties of a 13% CF-ABS SFRP composite using X-ray μ -CT microstructural characterization technique to generate 3D voxel based realistic, periodic RVEs. Although, the stepped-like surface of micro-features within RVE generated with voxel data are likely to induce stress concentrations, Guven et al. [115] has shown that the impact on evaluated effective properties for small displacement analysis are only minimal. Sensitivity analysis was carried out to determine a suitable computationally efficient RVE from three (3) different RVE sizes and realizations sets that yield effective properties within acceptable dispersion tolerance limit. Predicted effective properties obtained from numerical FEA approach were comparable to estimated properties based on Mori-Tanaka mean-field homogenizations technique. Impact

assessment of the micro-porosities on the material behavior of the SFRP composite revealed an overall reduction in the equivalent properties of the composite however the measured effects on the ECTE were minimal. Parameter dependent studies carried out revealed that the evaluated effective properties had a linear correlation with the fiber volume fraction and the average fiber orientation within the bead specimen consistent with literature [114], [124], [125]. The effective modulus and thermal conductivity were observed to vary proportionally with the fiber volume fraction and degree of fiber alignment with the print direction. Conversely the effective thermal expansion coefficient was observed to vary inversely with fiber volume fraction and degree of fiber alignment with the print direction. For increased simplicity and computational efficiency, a microstructure-based minimization approach that involves selection of a single characteristic RVE instance from a given realization set with matching microstructural properties as the overall parent ROI region was used to obtain quick estimate of the effective thermo-mechanical properties across regions of the non-porous printed bead strand and with very small prediction error tolerance. Overall, the effective modulus and thermal conductivity were predicted to be higher at the edges and top surface of the print bead where the volume fraction and degree of fiber alignment with the print direction are seen to be highest and the properties were lower closer to the bead center with less densely packed and more randomly oriented fibrous microstructure. The opposite behavior was observed for the thermal expansion coefficient across the bead sections.

5 CHAPTER FIVE

Simulating Particle Motion in Viscous Homogenous Suspension Flow

Sections of this chapter published as: Awenlimobor, A. and Smith, D.E., 2024. Effect of shear-thinning rheology on the dynamics and pressure distribution of a single rigid ellipsoidal particle in viscous fluid flow. *Physics of Fluids*, 36(12).

From the numerical evaluation carried out in the preceding chapter and in line with numerous literature [110], [113], [128], the inherent bead microstructure including the fibrous and porous structure significantly affect the resulting effective material properties and print quality. As such, understanding the mechanisms that are responsible for the development of the bead's microstructure, especially the micro-void formation mechanisms, is crucial. Presently, there is limited understanding on the known cause of micro-voids and the factors responsible for their formation in SFRP composites. Not until recently has there been increasing research interest in understanding mechanisms responsible for process-induced micro-void formation using computational-based simulation approach [52], [230], [297], [309]. Simulating the EDAM polymer composite melt flow-field process can provide valuable insight into potential mechanisms responsible for the microstructural development within the print beads, especially the micro-voids formation. Since intra-bead void nucleation is a localized (microscale) transport phenomena (occurring on the order of the fiber dimension) and is known to be heterogenous in nature forming at the particle-fluid interface, coupled multiscale simulation is required to accurately study this phenomenon. Previous studies [5], [9], [10], [11], [12], [13], [14], [15] have revealed that the local surrounding fluid pressure is a relevant process variable that significantly influences the nucleation of micro-voids in polymeric materials which itself depends on the particle dynamics. While previous numerical studies on fiber suspension flows have mainly focused on the particle dynamics that are mostly based on linear shear flow, the local flow-field surrounding the particle, including the velocity and pressure distribution has received very little attention which are particularly

relevant in understanding complex flow processes involved in physical rheological systems. Moreover, existing studies that also investigated development of the pressure field surrounding a particle are based on flow analysis around fixed particle in space [180], [216], [310] that do not consider the influence of the particle's dynamics on the pressure distribution.

As in many previous works on short fiber composites, it is helpful to consider Jeffery [19] when studying the dynamics of short fiber suspensions. Jeffery's equation has been widely used to evaluate particle dynamics in viscous, low Reynolds number, Newtonian fluid flow. Most physical processes involving the flow of particle suspension like EDAM polymer composites melt flow process possess non-linear suspension rheology and contains arbitrary shaped deformable particles with complex hydrodynamic interactions which are unaccounted for in Jeffery's model. Recently, Jeffery's equation has been extended to capture various effects neglected by the assumptions made in his initial work such as the influence of fiber's shape and symmetry [244], [311], the effect of a fiber's flexibility/deformability [196], [197], [212], [293], the influence of neighboring particles in a concentrated suspension [17], [256], the effect of a non-Newtonian and visco-elastic fluid rheology [185], [189], [312], etc. In these prior studies, model advancement and application of Jeffery's equations has been primarily focused on particle dynamics, and has yet to be employed to better understand micro-void formation within print beads. Moreover, the various extensions to Jeffery's equation have not specifically addressed the flow-field velocity and pressure surrounding the fiber surface during its motion.

The current chapter presents the 3D FEM model development which is used to investigate the effect of non-standard Jeffery's condition including the effect of generalized Newtonian fluid (GNF) rheology on the dynamics and surface pressure distribution of a single particle suspended in viscous homogenous flows. Firstly, we explore the effect of various factors such as the fibers

geometric aspect ratio and initial fiber angle on the single particle motion and surface pressure distribution for a single particle suspended in Newtonian homogenous flow-field using Jeffery's equation. Typical size of particles encountered during Extrusion Deposition Additive Manufacturing (EDAM) polymer composite processing are on average hundreds of microns in magnitude depending on the particles concentration and system's scale, usually around 50 – 100 μm for small scale EDAM systems and $\sim 300\mu m$ for large scale EDAM systems [313]. The rotary Peclet number that characterizes these polymeric melt flow through an EDAM nozzle are orders of magnitude high (i.e. $Pe_r \gg 1$). Brownian effects arising from particle interaction with the surrounding fluid molecules are thus insignificant and have been ignored in the current investigation since the hydrodynamic forces are expected to dominate the particle's motion. Jeffery's equations are a good starting point for studying particles behavior in these Newtonian flows. More rigorous stochastic statistical analysis accounting for Brownian disturbance such as that conducted by Leal et al. [314] and Zhang et al. [229] is a relevant study for future consideration. The generalized Newtonian FEA single fiber motion model development is a non-linear extension to the Newtonian formulations of Zhang et al. [225], [229], [291] and Awenlimobor et al. [52], [230] assuming a power-law non-Newtonian fluid behavior for fiber suspension rheology. A two (2) stage Newton Raphson numerical algorithm is used in our simulation, firstly to solve for the steady-state flow-field velocities and pressure distribution within the flow domain and secondly to compute the resulting translational and rotational velocities of the rigid spheroidal particle during its motion in various homogenous flow fields by equilibrating the net force and couple acting on the particles surface and the fiber's instantaneous positions and orientations are updated using a numerical ordinary differential equation (ODE) solution technique. FEA model validation is achieved by comparing steady state responses at a single time

step of the quasi-transient analysis of a single particle motion along Jeffery's orbit obtained from a custom-built FEA simulation with results obtained Jeffery's Equations. Likewise, the behavior of the particle (kinematics and surface pressure response) in various Newtonian homogenous flow fields are benchmarked for both Jeffery's Model and FEA simulation.

Finally, we investigate the resulting effect of particle shape and the shear-thinning fluid rheology on the particle's dynamics and evolution of the pressure distribution response on the fibers' surface in the various homogenous flow fields using our validated FEA model. These findings are particularly useful in controlling process parameters to optimize the microstructure of particulate polymer composites to improve print properties.

5.1 Methodology

This section provides in detail the methods used for predicting the behavior of a single three-dimensional (3D) rigid ellipsoidal particle suspended in Newtonian and non-Newtonian viscous homogenous shear-extension flows. The first section presents Jeffery's formulation for the flow-field development around an ellipsoid and explicit derivations for the particle motion (angular velocities and orientation angles) in a special class of linear homogenous flow with combined extension and shear rate velocity gradient components that idealizes typical flow conditions found in various sections of an EDAM extruder-nozzle. The second section details the FEA model development for obtaining particle angular velocities, orientation angles and field velocities and pressure distribution surrounding a particle suspended in non-linear creeping shear flow with a power-law fluid definition. Subsequent section presents results of the model validation by comparing the evolution of the particle's angular velocities and surface pressure distribution obtained from both Jeffery's analytical equations and FEA numerical model for different Newtonian flow cases and particle aspect ratio. Except stated otherwise, we consider a geometric

aspect ratio of $r_e = 6$ for the prolate spheroid, a consistency index of $m = 1 \text{ Pa} \cdot \text{s}^n$ for the power-law fluid or a viscosity of $\mu_1 = 1 \text{ Pa} \cdot \text{s}$ for Newtonian fluid, and shear rate of $\dot{\gamma} = 1 \text{ s}^{-1}$ for the various flow cases.

5.1.1 Standard Jeffery Analytical Model

Jeffery [19] derived analytical equations for the motion of a single 3D ellipsoidal particle suspended in a Newtonian homogenous viscous creeping flow by linearization of the Navier Stokes equations assuming a zero Reynolds number. The following includes a summary of Jeffery's particle-fluid interaction dynamics model where he obtained expressions for the velocity and pressure field within the fluid surrounding the particle. The equations for the pressure and velocity within a Newtonian fluid having viscosity μ_1 are respectively given as

$$p = p_0 + 2\mu_1 \Lambda_{ij}^{III} \nabla_{X_i} \nabla_{X_j} \Omega \quad (5.1)$$

and

$$\dot{X}_i = \dot{X}_i^\infty + \nabla_{X_i} \Lambda_j^I \chi_j + \epsilon_{ijk} \nabla_{X_j} \Lambda_{km}^{II} X_m + \Lambda_{jk}^{III} X_k \nabla_{X_i} \nabla_{X_j} \Omega - \Lambda_{ij}^{III} \nabla_{X_j} \Omega \quad (5.2)$$

where the position vector $\underline{X}$, gradient operator $\underline{\nabla}$ and integral function $\underline{\chi}$ are given respectively as

$$\underline{X} = [X_1 \quad X_2 \quad X_3]^T, \quad \underline{\nabla}_X = [\partial/\partial X_1 \quad \partial/\partial X_2 \quad \partial/\partial X_3]^T, \quad \underline{\chi} = [\chi_1 \quad \chi_2 \quad \chi_3]^T \quad (5.3)$$

In the above, the Laplace function Ω is defined in terms of the independent position vector variables $\underline{X}$ and λ as

$$\Omega = \Omega(\underline{X}, \lambda) = \int_{\lambda}^{\infty} \frac{1}{\Delta} \left\{ \sum_{j=1}^3 \frac{X_j^2}{\mathbb{H}_j^2 + \lambda} - 1 \right\} d\lambda, \quad \Delta^2 = \prod_{j=1}^3 (\mathbb{H}_j^2 + \lambda) \quad (5.4)$$

where λ is an arbitrary offset distance from the particle's surface obtained from the positive real roots of

$$\sum_{j=1}^3 \frac{X_j^2}{W_j^2 + \lambda} = 1, \quad \lambda \geq 0 \quad (5.5)$$

The undisturbed fluid velocity $\dot{X}_i^\infty$ in eqn. (5.2) above is given as

$$\dot{X}_i^\infty = L_{ij} X_j \quad (5.6)$$

where L_{ij} is the velocity gradient tensor. The constant-coefficient tensors $\Lambda_i^I, \Lambda_{ij}^{II}$ & Λ_{ij}^{III} that appear in eqns. (5.1) - (5.2) above are given as

$$\underline{\Lambda}^I = \begin{bmatrix} R \\ S \\ T \end{bmatrix}, \quad \underline{\Lambda}^{II} = \begin{bmatrix} U & & \\ & V & \\ & & W \end{bmatrix}, \quad \underline{\Lambda}^{III} = \begin{bmatrix} A & H & G' \\ H' & B & F \\ G & F' & C \end{bmatrix} \quad (5.7)$$

where expressions for the components shown here are given in APPENDIX B **(B.1)**. The terms in Λ_{ij}^{III} are simply the stresslet and torque acting on the rigid ellipsoid suspended in linear ambient flow-field [315]. The tensors $\Lambda_i^I, \Lambda_{ij}^{II}$ & Λ_{ij}^{III} are functions of the symmetric rate of deformation tensor Γ_{ij} and the antisymmetric vorticity tensor $\Xi_{ij} = \epsilon_{imn} \Xi_m \delta_{nj}$ obtained by decomposing the velocity gradient tensor L_{ij} according to

$$L_{ij} = \nabla_{X_j} \dot{X}_i = \Gamma_{ij} + \Xi_{ij}, \quad \Gamma_{ij} = \frac{1}{2} [L_{ij} + L_{ji}], \quad \Xi_{ij} = \frac{1}{2} [L_{ij} - L_{ji}] \quad (5.8)$$

The velocity gradient L_{ij} is given with respect to the particle's local coordinate axis and is thus a function of the independent particle orientation angle vector $\underline{\Theta} = [\phi \quad \theta \quad \psi]^T$ obtained by a transformation operation according to

$$L_{ij} = Z_{X_{mi}} \underline{L}_{mn} Z_{X_{nj}} \quad (5.9)$$

where $\underline{L}_{mn}$ is the velocity gradient in the global reference frame axis. The transformation tensor $Z_{\theta_{ij}}$ is given in terms of the Euler angles as:

$$Z_{X_{ij}} = \Pi_{mi}^{(1)} \Pi_{nm}^{(2)} \Pi_{jn}^{(3)} \quad (5.10)$$

where,

$$\Pi_{ij}^{(k)} = \delta_{in}\delta_{jn} + (1 - \delta_{in})(1 - \delta_{jn})[\delta_{ij} \cos \Theta_k + (j - i) \sin \Theta_k], \quad n = 2 + -1^k \quad (5.11)$$

At the particle's surface, the field velocity is given by

$$\dot{X}_i^p = \dot{X}_i \big|_{\lambda=0} = \epsilon_{ijk} \dot{\Psi}_j X_k \quad (5.12)$$

The particle's angular velocity $\dot{\Psi}_i$ in the local reference frame is given by the expression.

$$\dot{\Psi}_i = \Xi_i + M_{ij} D_j \quad (5.13)$$

where Ξ_i is the vorticity vector, D_i contains non-diagonal terms of the symmetric rate of deformation tensor Γ_{ij} , i.e.

$$D_k = \Gamma_{ij}, \quad k = 6 - i - j \mid i \neq j$$

and the constant coefficient matrix M_{ij} is defined as

$$M_{ij} = M_i \delta_{ij}, \quad M_1 = \frac{W_2^2 - W_3^2}{W_2^2 + W_3^2}, \quad M_2 = \frac{W_3^2 - W_1^2}{W_3^2 + W_1^2}, \quad M_3 = \frac{W_1^2 - W_2^2}{W_1^2 + W_2^2} \quad (5.14)$$

The angular velocities in the global reference coordinate axis $\underline{\dot{\Theta}}$ based on Euler's definition are obtained by the transformation operation

$$Z_{\Theta ij} \dot{\Theta}_j = \dot{\Psi}_i \quad (5.15)$$

where the transformation operator $\underline{\underline{Z}}_{\Theta}$ is given as (cf. Figure 5.1a) for the Euler definition of orientation angles

$$\underline{\underline{Z}}_{\Theta} = \begin{bmatrix} \cos \theta & 0 & 1 \\ -\sin \theta \cos \psi & \sin \psi & 0 \\ \sin \theta \sin \psi & \cos \psi & 0 \end{bmatrix} \quad (5.16)$$

Figure 5.1a illustrates the ellipsoidal particle of interest suspended in simple shear flow as shown.

The normal and shear stress components at any point in the flow field may be evaluated for incompressible fluid as

$$\sigma_{ij} = -p \delta_{ij} + \mu_1 \left[\nabla_{X_i} \dot{X}_j + \nabla_{X_j} \dot{X}_i \right] \quad (5.17)$$

On the particle's surface, the stress reduces to $\sigma_{ij} = -p\delta_{ij}$ implying that the only active stresses on the particle's surface are the hydrostatic pressure acting normal to the surface.

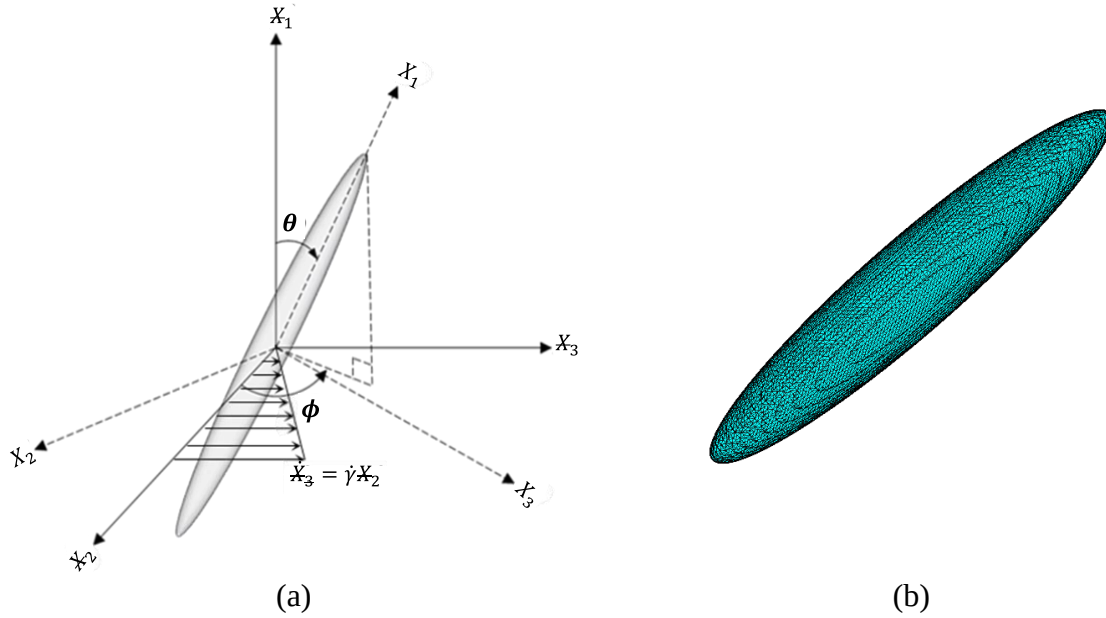


Figure 5.1: (a) Fiber orientation angles definition (b) Mesh refinement on the fiber surface.

The two-dimensional (2D) contraction of Jeffery's expression for the field velocities and pressure surrounding a rigid particle of elliptical shape in planar homogenous flow field can be found in APPENDIX B **(B.2)**. Our main interest here is to evaluate the motion, and surface pressure and velocity of the ellipsoidal inclusion using Jeffery's equations given above. To compute surface pressure and velocity distribution on the particle surface, the ellipsoidal surface is discretized using MATLAB's inbuilt PDE modeller (MathWorks, Natick, MA, USA) where vertices were imposed at ends of the ellipsoid to enable the calculation of particle tip pressure (cf. Figure 5.1b). At the mesh points, the flow-field pressure and velocities are evaluated using eqns. (5.1)-(5.2) respectively. The degree of mesh refinement is critical to obtaining accurate pressure extremities and locations on the particle surface. A 4th order explicit Runge-Kutta ordinary differential equation (ODE) technique is used to numerically integrate the particle's angular velocities (cf. eqn. (5.13)) with time to obtain solutions of the particle orientation angles, and the

associated field state (pressure and velocities on each node of the particle surface) based on Jeffery's model equations.

5.1.2 Optimization of Jeffery's Pressure

The current objective is to minimize the pressure p on the surface of the fiber and about all possible orientation configuration defined by the surface of the unit sphere S such that:

$$\oint d\rho = \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \sin \theta d\theta d\phi \quad (5.18)$$

The minimization objective function is thus the pressure p given in eqn. (5.1) above which we can rewrite as a function of the dependent variables $\underline{\Theta}$, and $\underline{X}$, such that:

$$p(\underline{X}, \underline{\Theta}) = p_0 + \varpi_j \mathcal{A}_j \quad (5.19)$$

and the constraints are thus the surface of the ellipsoidal fiber, and the surface of the unit sphere S that defines all possible fiber orientation, i.e.,

$$\sum_{j=1}^3 \frac{X_j^2}{N_j^2} - 1 = 0 \quad \text{on the fiber's surface} \quad (5.20)$$

$$0 \leq \phi \leq 2\pi, \quad 0 \leq \theta \leq \pi \quad \text{on the surface of the unit sphere } S \quad (5.21)$$

The constant coefficient vector $\underline{\varpi}$ is given as:

$$\underline{\varpi} = [A \quad B \quad C \quad F + F' \quad G + G' \quad H + H'] \quad (5.22)$$

Definition of the constants A, B, C, F, G, H have been provided in APPENDIX B **(B.1)** and are functions of the components of the deformation tensor Γ_{ij} , and the vorticity tensor Ξ_{ij} given in eqn. (5.8) above. The vector $\mathcal{A}_n$ in form contracted notation contains components of the hessian of the Laplace function Ω that appear in the pressure equation (cf. eqn. (5.19)) given as

$$\mathcal{A}_n = \nabla_{X_i} \nabla_{X_j} \Omega, \quad n = f(i, j) \quad (5.23)$$

where the function $f(i, j)$ is given as

$$f(i, j) = i\delta_{ij} + (1 - \delta_{ij})(9 - i - j) \quad (5.24)$$

Moreover, the Laplace equation must be satisfied. i.e.,

$$\nabla^2 \Omega = \nabla_{X_k} \nabla_{X_k} \Omega = 0 \quad (5.25)$$

Derivation of the exact gradient and hessian of the pressure used in optimization process are provided in detail in the succeeding section.

5.1.2.1 Obtaining Exact Derivatives of Jeffery's Pressure

The optimization operation requires the gradient and hessian of the pressure which we obtain explicitly by taking first and second derivatives of the pressure equation with respect to the independent variable vectors $\underline{X}$ and $\underline{\Theta}$. i.e., the gradient of the pressure is given as

$$\underline{\nabla} p = \begin{bmatrix} \underline{\nabla}_X \\ \underline{\nabla}_\Theta \end{bmatrix} p \quad (5.26)$$

where

$$\nabla_{X_i} p = \varpi_j \nabla_{X_i} \Delta_j, \quad \nabla_{\Theta_i} p = \Delta_j \nabla_{\Theta_i} \varpi_j, \quad i = 1 - 3, \quad j = 1 - 6 \quad (5.27)$$

and the hessian is given as

$$\underline{\underline{\nabla^2}} p = \begin{bmatrix} \underline{\nabla}_X \underline{\nabla}_X^T & \underline{\nabla}_X \underline{\nabla}_\Theta^T \\ \underline{\nabla}_\Theta \underline{\nabla}_X^T & \underline{\nabla}_\Theta \underline{\nabla}_\Theta^T \end{bmatrix} p \quad (5.28)$$

Since the hessian is symmetric, the relevant components of $\underline{\underline{\nabla^2}} p$ are given as

$$\begin{aligned} \nabla_{X_i} \nabla_{X_j} p &= \varpi_k \left[\nabla_{X_i} \nabla_{X_j} \Delta_k \right], & \nabla_{X_i} \nabla_{\Theta_j} p &= \left[\nabla_{X_i} \Delta_k \right] \left[\nabla_{\Theta_j} \varpi_k \right], \\ \nabla_{\Theta_i} \nabla_{\Theta_j} p &= \Delta_k \left[\nabla_{\Theta_i} \nabla_{\Theta_j} \varpi_k \right], & i, j &= 1 - 3, \quad k = 1 - 6 \end{aligned} \quad (5.29)$$

The derivative operators are distributive over the differentiable elements and sub-elements of the constant coefficient vector $\underline{\varpi}$ and can be assembled from the derivatives of its individual

components. Typical first and second order derivatives of the constants in $\underline{\omega}$ are presented in eqn.

(5.30) below from which the others can be surmised.

$$\begin{aligned}\nabla_{\theta_i} \omega_1 &= \nabla_{\theta_i} A = \frac{1}{6} \left\{ \frac{2\mathcal{V}_{1_0}'' \nabla_{\theta_i} \Gamma_{11} - \mathcal{V}_{2_0}'' \nabla_{\theta_i} \Gamma_{22} - \mathcal{V}_{3_0}'' \nabla_{\theta_i} \Gamma_{33}}{\mathcal{V}_{2_0}'' \mathcal{V}_{3_0}'' + \mathcal{V}_{3_0}'' \mathcal{V}_{1_0}'' + \mathcal{V}_{1_0}'' \mathcal{V}_{2_0}''} \right\} \\ \nabla_{\theta_i} \nabla_{\theta_j} \omega_1 &= \nabla_{\theta_i} \nabla_{\theta_j} A = \frac{1}{6} \left\{ \frac{2\mathcal{V}_{1_0}'' \nabla_{\theta_i} \nabla_{\theta_j} \Gamma_{11} - \mathcal{V}_{2_0}'' \nabla_{\theta_i} \nabla_{\theta_j} \Gamma_{22} - \mathcal{V}_{3_0}'' \nabla_{\theta_i} \nabla_{\theta_j} \Gamma_{33}}{\mathcal{V}_{2_0}'' \mathcal{V}_{3_0}'' + \mathcal{V}_{3_0}'' \mathcal{V}_{1_0}'' + \mathcal{V}_{1_0}'' \mathcal{V}_{2_0}''} \right\}\end{aligned}\quad (5.30)$$

The components of $\underline{\omega}$ are functions of the components of the deformation rate tensor Γ_{ij} and the vorticity tensor $\mathcal{E}_{ij}$ which are obtained from the decomposition of the velocity gradient L_{ij} in the local fiber reference frame according to the transformation operation of eqn. (5.8) and is thus function of the fiber orientation angles $\underline{\Theta}$. i.e.

$$\langle \nabla_{\theta_k} \Gamma_{ij}, \nabla_{\theta_k} \mathcal{E}_{ij} \rangle = \frac{1}{2} [\nabla_{\theta_k} L_{ij} \pm \nabla_{\theta_k} L_{ji}] \quad (5.31)$$

likewise, the second derivatives can be written as

$$\langle \nabla_{\theta_l} \nabla_{\theta_k} \Gamma_{ij}, \nabla_{\theta_l} \nabla_{\theta_k} \mathcal{E}_{ij} \rangle = \frac{1}{2} [\nabla_{\theta_l} \nabla_{\theta_k} L_{ij} \pm \nabla_{\theta_l} \nabla_{\theta_k} L_{ji}] \quad (5.32)$$

where the operator $\nabla_{\theta_k} = \partial / \partial \theta_k$. The first derivative of the velocity gradient tensor with respect to k^{th} component of $\underline{\Theta}$ in the fibers local coordinate axis is obtained by the product rule and expressed in indicial notation as

$$\nabla_{\theta_k} L_{ij} = \nabla_{\theta_k} Z_{X_{mi}} \mathcal{L}_{mn} Z_{X_{nj}} + Z_{X_{mi}} \mathcal{L}_{mn} \nabla_{\theta_k} Z_{X_{nj}} \quad (5.33)$$

The derivative of the transformation tensor $Z_{X_{ij}}$ with respect to $\underline{\Theta}$, i.e. $\nabla_{\theta_k} Z_{X_{ij}}$ is a third order tensor given as

$$\nabla_{\theta_k} Z_{X_{ij}} = \delta_{k1} \nabla \Pi_{mi}^{(1)} \Pi_{nm}^{(2)} \Pi_{jn}^{(3)} + \delta_{k2} \Pi_{mi}^{(1)} \nabla \Pi_{nm}^{(2)} \Pi_{jn}^{(3)} + \delta_{k3} \Pi_{mi}^{(1)} \Pi_{nm}^{(2)} \nabla \Pi_{jn}^{(3)} \quad (5.34)$$

The derivative $\partial Z_{X_{ij}} / \partial \theta_k$ is trivial. Since $\Pi_{ij}^{(k)}$ is conveniently represented in indicial notation as given in eqn. (5.11), it is easy to differentiate $\Pi_{ij}^{(k)}$ with respect to θ_k , i.e.

$$\nabla \Pi_{ij}^{(k)} = (1 - \delta_{in})(1 - \delta_{jn})[-\delta_{ij} \sin \Theta_k + (j - i) \cos \Theta_k] \quad (5.35)$$

Following from eqn. (5.33) above, the second derivative of the velocity gradient L_{ij} with respect to $\underline{\Theta}$ in the fibers local coordinate axis via product rule is given as

$$\begin{aligned} \nabla_{\Theta_l} \nabla_{\Theta_k} L_{ij} = & \nabla_{\Theta_l} \nabla_{\Theta_k} Z_{X_{mi}} \underline{L}_{mn} Z_{X_{nj}} + \nabla_{\Theta_k} Z_{X_{mi}} \underline{L}_{mn} \nabla_{\Theta_l} Z_{X_{nj}} + \nabla_{\Theta_l} Z_{X_{mi}} \underline{L}_{mn} \nabla_{\Theta_k} Z_{X_{nj}} \\ & + Z_{X_{mi}} \underline{L}_{mn} \nabla_{\Theta_l} \nabla_{\Theta_k} Z_{X_{nj}} \end{aligned} \quad (5.36)$$

The second derivative of the transformation tensor $Z_{X_{ij}}$ with respect to $\underline{\Theta}$, i.e. $\nabla_{\Theta_l} \nabla_{\Theta_k} Z_{X_{ij}}$ that appear in eqn. (5.36) above is a fourth order tensor given as

$$\begin{aligned} \nabla_{\Theta_l} \nabla_{\Theta_k} Z_{X_{ij}} = & \delta_{l1} \delta_{k1} \nabla^2 \Pi_{mi}^{(1)} \Pi_{nm}^{(2)} \Pi_{jn}^{(3)} + \delta_{l2} \delta_{k1} \nabla \Pi_{mi}^{(1)} \nabla \Pi_{nm}^{(2)} \Pi_{jn}^{(3)} \\ & + \delta_{l3} \delta_{k1} \nabla \Pi_{mi}^{(1)} \Pi_{nm}^{(2)} \nabla \Pi_{jn}^{(3)} + \delta_{l1} \delta_{k2} \nabla \Pi_{mi}^{(1)} \nabla \Pi_{nm}^{(2)} \Pi_{jn}^{(3)} \\ & + \delta_{l2} \delta_{k2} \Pi_{mi}^{(1)} \nabla^2 \Pi_{nm}^{(2)} \Pi_{jn}^{(3)} + \delta_{l3} \delta_{k2} \Pi_{mi}^{(1)} \nabla \Pi_{nm}^{(2)} \nabla \Pi_{jn}^{(3)} \\ & + \dots \delta_{l1} \delta_{k3} \nabla \Pi_{mi}^{(1)} \Pi_{nm}^{(2)} \nabla \Pi_{jn}^{(3)} + \delta_{l2} \delta_{k3} \Pi_{mi}^{(1)} \nabla \Pi_{nm}^{(2)} \nabla \Pi_{jn}^{(3)} \\ & + \delta_{l3} \delta_{k3} \Pi_{mi}^{(1)} \Pi_{nm}^{(2)} \nabla^2 \Pi_{jn}^{(3)} \end{aligned} \quad (5.37)$$

From eqn. (5.35) above, we can conveniently obtain second derivatives of $\Pi_{ij}^{(k)}$ with respect to $\underline{\Theta}$.

i.e.

$$\nabla^2 \Pi_{ij}^{(k)} = -(1 - \delta_{in})(1 - \delta_{jn})[\delta_{ij} \cos \Theta_k + (j - i) \sin \Theta_k] \quad (5.38)$$

The $\underline{\Theta}$ – derivatives of the fiber angular velocities with respect to its local coordinate axis are linear superposition of the derivatives of the individual terms in eqn. (5.13) and given as

$$\nabla_{\Theta_i} \dot{\Psi}_j = \nabla_{\Theta_i} \mathcal{E}_j + M_{jk} \nabla_{\Theta_i} D_k \quad \left| \quad \nabla_{\Theta_m} \nabla_{\Theta_n} \dot{\Psi}_j = \nabla_{\Theta_m} \nabla_{\Theta_n} \mathcal{E}_j + M_{jk} \nabla_{\Theta_m} \nabla_{\Theta_n} D_k \quad (5.39)$$

where the operator $\underline{\nabla}_{\underline{\Theta}}^{(n)}$ is distributive over the components of $\mathcal{E}_j$ and D_j as in the usual manner.

For instance,

$$\begin{aligned} \nabla_{\Theta_i} \mathcal{E}_1 &= \nabla_{\Theta_i} \xi, & \nabla_{\Theta_i} D_1 &= \nabla_{\Theta_i} \Gamma_{23} \\ \nabla_{\Theta_m} \nabla_{\Theta_n} \mathcal{E}_1 &= \nabla_{\Theta_m} \nabla_{\Theta_n} \xi, & \nabla_{\Theta_m} \nabla_{\Theta_n} D_1 &= \nabla_{\Theta_m} \nabla_{\Theta_n} \Gamma_{23} \end{aligned} \quad (5.40)$$

The first derivatives of the coefficient vector $\underline{D}$ containing terms of the derivatives Laplace function $\nabla_{\underline{X}}^2 \Omega$ with respect to $\underline{X}$ is given as

$$\nabla_{X_m} D_n = \nabla_{X_m} \nabla_{X_j} \nabla_{X_k} \Omega, \quad n = f(j, k) \quad (5.41)$$

Similarly, the second derivative of the coefficient vector $\underline{D}$ with respect to $\underline{X}$ is a third order tensor $\nabla_{X_r} \nabla_{X_s} D_m$ given as

$$\nabla_{X_r} \nabla_{X_s} D_n = \nabla_{X_r} \nabla_{X_s} \nabla_{X_j} \nabla_{X_k} \Omega, \quad n = f(j, k) \quad (5.42)$$

The terms of the higher order derivatives of the Laplace function Ω found in expressions for $\nabla_{X_m} D_n$ and $\nabla_{X_r} \nabla_{X_s} D_m$ in eqns. (5.41) - (5.42) above are given in the next section below.

5.1.2.2 Obtaining higher-order derivatives of the Laplace Function Ω

The Laplace function Ω found in Jeffery's equations for the field velocities and pressure distribution is an integral function in terms of position descriptor variable λ and is given as

$$\Omega = \int_{\lambda}^{\infty} f(\underline{X}, \lambda) d\lambda, \quad f(\underline{X}, \lambda) = \frac{1}{\Delta} \left[\sum_{j=1}^3 \frac{X_j^2}{\mathbb{H}_j^2 + \lambda} - 1 \right] \quad (5.43)$$

To obtain derivatives of Ω , the well-known Leibnitz integral theorem finds particular use in differentiating definite integral functions with limits that are function of the differentiable variable. For instance, the first-order partial derivative of the Laplace function with respect to X_j using the Leibnitz theorem can be evaluated from the expression in eqn. (5.44) below:

$$\frac{d\Omega}{dX_j} = \int_{\lambda}^{\infty} \frac{\partial}{\partial X_j} \{f(\underline{X}, \lambda)\} d\lambda + f(\underline{X}, \infty) \frac{d\infty}{dX_j} - f(\underline{X}, \lambda) \frac{d\lambda}{dX_j} \quad (5.44)$$

By definition, $f(\underline{X}, \lambda) = 0$ since

$$\left[\sum_{j=1}^3 \frac{X_j^2}{H_j^2 + \lambda} - 1 \right] = 0, \quad \text{and}, \quad \frac{d\infty}{dX_j} = 0 \quad (5.45)$$

Therefore

$$\frac{d\Omega}{dX_j} = \int_{\lambda}^{\infty} \frac{\partial}{\partial X_j} \{f(\underline{X}, \lambda)\} d\lambda = 2X_j \int_{\lambda}^{\infty} \frac{1}{\Delta} \left[\frac{d\lambda}{H_j^2 + \lambda} \right] = 2\mathfrak{U}_j X_j \quad (5.46)$$

In eqn. (5.46) above, repeated indices do not imply summation. For subsequent higher order derivatives of Ω , it is important to make some necessary definitions. Firstly, we define $\mathcal{P}^{(n)}$ such that:

$$\frac{1}{\mathcal{P}^{(n)}} = \sum_{j=1}^3 \frac{X_j^2}{(H_j^2 + \lambda)^n}, \quad \mathcal{P}^{(2)} = \frac{1}{2} X_j \nabla_{X_j} \lambda \quad (5.47)$$

Also, we define $\mathfrak{Y}^{(n)}$ such that:

$$\frac{1}{\mathfrak{Y}^{(n)}} = \sum_{j=1}^3 \frac{1}{(H_j^2 + \lambda)^n}, \quad \mathfrak{Y} = \mathfrak{Y}^{(1)} \quad (5.48)$$

Additionally, the first and second derivatives of λ with respect to components of $\underline{X}$ and its permutations are important in concisely obtaining higher order derivatives of Ω . By differentiating $\Delta f(\underline{X}, \lambda)$, and making necessary substitutions, we obtain for the first derivatives of λ thus:

$$\nabla_{X_j} \lambda = \frac{2X_j}{H_j^2 + \lambda} \mathcal{P}^{(2)} \quad (5.49)$$

Similarly, the second derivatives of λ with respect to X_j are given as

$$\begin{aligned} \nabla_{X_j}^2 \lambda &= \frac{1}{X_j} \nabla_{X_j} \lambda - \left[2 \frac{1}{H_j^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \nabla_{X_j} \lambda^2 \\ \nabla_{X_i} \nabla_{X_j} \lambda &= - \left[\frac{1}{H_i^2 + \lambda} + \frac{1}{H_j^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \nabla_{X_i} \lambda \nabla_{X_j} \lambda \end{aligned} \quad (5.50)$$

With the above definitions we can concisely present expressions for typical forms of the second-order partial derivatives of Ω with respect to permutations of X_j vector using Leibnitz integral theorem thus from which derivatives with respect to other permutations of the differentiable variables are implicit.

$$\nabla_{X_j}^2 \Omega = 2\mathcal{H}_j - \frac{1}{\mathcal{P}^{(2)}\Delta} [\nabla_{X_j} \lambda]^2, \quad \nabla_{X_i} \nabla_{X_j} \Omega = -\frac{1}{\mathcal{P}^{(2)}\Delta} \nabla_{X_i} \lambda \nabla_{X_j} \lambda \quad (5.51)$$

Likewise, third-order partial derivatives of Ω with respect to permutations of components of X_j are conveniently presented in eqns. (5.52) - (5.54) from which components of $\nabla_{X_m} \mathcal{A}_n$ can be deduced.

$$\nabla_{X_j}^3 \Omega = -3 \frac{1}{X_j} \frac{1}{\mathcal{P}^{(2)}\Delta} [\nabla_{X_j} \lambda]^2 + \frac{1}{\mathcal{P}^{(2)}\Delta} \left[\frac{1}{2\mathbb{Y}} + 3 \frac{1}{\mathbb{H}_j^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] [\nabla_{X_j} \lambda]^3 \quad (5.52)$$

$$\nabla_{X_i}^2 \nabla_{X_j} \Omega = -\frac{1}{X_i} \frac{1}{\mathcal{P}^{(2)}\Delta} \nabla_{X_i} \lambda \nabla_{X_j} \lambda + \frac{1}{\mathcal{P}^{(2)}\Delta} \left[\frac{1}{2\mathbb{Y}} + 2 \frac{1}{\mathbb{H}_i^2 + \lambda} + \frac{1}{\mathbb{H}_j^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] [\nabla_{X_i} \lambda]^2 \nabla_{X_j} \lambda \quad (5.53)$$

$$\nabla_{X_i} \nabla_{X_j} \nabla_{X_k} \Omega = + \frac{1}{\mathcal{P}^{(2)}\Delta} \left[\frac{1}{2\mathbb{Y}} + \frac{1}{\mathbb{H}_i^2 + \lambda} + \frac{1}{\mathbb{H}_j^2 + \lambda} + \frac{1}{\mathbb{H}_k^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \nabla_{X_i} \lambda \nabla_{X_j} \lambda \nabla_{X_k} \lambda \quad (5.54)$$

Additionally, fourth – order partial derivatives of Ω with respect to permutations of components of X_j are given in eqns. (5.55) - (5.58) from which components of $\nabla_{X_r} \nabla_{X_s} \mathcal{A}_n$, can be deduced.

$$\begin{aligned} \nabla_{X_j}^4 \Omega = \frac{1}{\mathcal{P}^{(2)}\Delta} \left\{ 15 \frac{1}{X_j^2} [\nabla_{X_j} \lambda]^2 - 36 \left[\frac{1}{\mathbb{H}_j^2 + \lambda} - \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \frac{1}{X_j} [\nabla_{X_j} \lambda]^3 \right. \\ \left. + \left[-\frac{1}{4\mathbb{Y}^2} - \frac{1}{2\mathbb{Y}^{(2)}} - 3 \frac{1}{\mathbb{Y}} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} + 2 \frac{1}{\mathbb{Y}} \frac{1}{\mathbb{H}_j^2 + \lambda} + 16 \frac{1}{(\mathbb{H}_j^2 + \lambda)^2} - 36 \frac{1}{\mathbb{H}_j^2 + \lambda} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right. \right. \\ \left. \left. + 12 \left[\frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right]^2 + 6 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(4)}} \right] [\nabla_{X_j} \lambda]^4 \right\} \quad (5.55) \end{aligned}$$

$$\begin{aligned} \nabla_{X_i}^3 \nabla_{X_j} \Omega = \frac{1}{\mathcal{P}^{(2)}\Delta} \left\{ 3 \left[\frac{1}{2\mathbb{Y}} + 2 \frac{1}{\mathbb{H}_i^2 + \lambda} + \frac{1}{\mathbb{H}_j^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \frac{1}{X_i} [\nabla_{X_i} \lambda]^2 \nabla_{X_j} \lambda \right. \\ \left. + \left[-\frac{1}{4\mathbb{Y}^2} - \frac{1}{2\mathbb{Y}^{(2)}} - 3 \frac{1}{\mathbb{Y}} \frac{1}{\mathbb{H}_i^2 + \lambda} - \frac{1}{\mathbb{Y}} \frac{1}{\mathbb{H}_j^2 + \lambda} + 3 \frac{1}{\mathbb{Y}} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} - 12 \frac{1}{(\mathbb{H}_i^2 + \lambda)^2} \right. \right. \\ \left. \left. - 2 \frac{1}{(\mathbb{H}_j^2 + \lambda)^2} - 6 \frac{1}{\mathbb{H}_i^2 + \lambda} \frac{1}{\mathbb{H}_j^2 + \lambda} + 18 \frac{1}{\mathbb{H}_i^2 + \lambda} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} + 6 \frac{1}{\mathbb{H}_j^2 + \lambda} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right. \right. \\ \left. \left. - 12 \left[\frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right]^2 + 6 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(4)}} \right] [\nabla_{X_i} \lambda]^3 \nabla_{X_j} \lambda \right\} \quad (5.56) \end{aligned}$$

$$\begin{aligned}
\nabla_{X_i}^2 \nabla_{X_j}^2 \Omega = \frac{1}{\mathcal{P}^{(2)} \Delta} & \left\{ -\frac{1}{X_i} \frac{1}{X_j} \nabla_{X_i} \lambda \nabla_{X_j} \lambda + \left[\frac{1}{2\mathfrak{V}} + \frac{1}{\mathfrak{H}_i^2 + \lambda} + 2 \frac{1}{\mathfrak{H}_j^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \frac{1}{X_i} \nabla_{X_i} \lambda \left[\nabla_{X_j} \lambda \right]^2 \right. \\
& + \left[\frac{1}{2\mathfrak{V}} + 2 \frac{1}{\mathfrak{H}_i^2 + \lambda} + \frac{1}{\mathfrak{H}_j^2 + \lambda} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \frac{1}{X_j} \left[\nabla_{X_i} \lambda \right]^2 \nabla_{X_j} \lambda \\
& + \left[-\frac{1}{4\mathfrak{V}^2} - \frac{1}{2\mathfrak{V}^{(2)}} - 2 \frac{1}{\mathfrak{V}} \frac{1}{\mathfrak{H}_i^2 + \lambda} - 2 \frac{1}{\mathfrak{V}} \frac{1}{\mathfrak{H}_j^2 + \lambda} - 6 \frac{1}{(\mathfrak{H}_i^2 + \lambda)^2} - 6 \frac{1}{(\mathfrak{H}_j^2 + \lambda)^2} \right. \\
& - 8 \frac{1}{\mathfrak{H}_i^2 + \lambda} \frac{1}{\mathfrak{H}_j^2 + \lambda} + 3 \frac{1}{\mathfrak{V}} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} + 12 \frac{1}{\mathfrak{H}_i^2 + \lambda} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} + 12 \frac{1}{\mathfrak{H}_j^2 + \lambda} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} - 12 \left[\frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right]^2 \\
& \left. \left. + 6 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(4)}} \right] \left[\nabla_{X_i} \lambda \right]^2 \left[\nabla_{X_j} \lambda \right]^2 \right\} \quad (5.57)
\end{aligned}$$

$$\begin{aligned}
\nabla_{X_i}^2 \nabla_{X_j} \nabla_{X_k} \Omega = \frac{1}{\mathcal{P}^{(2)} \Delta} & \left\{ \left[\frac{3}{2} \frac{1}{\mathfrak{V}} - 2 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right] \frac{1}{X_i} \nabla_{X_i} \lambda \nabla_{X_j} \lambda \nabla_{X_k} \lambda \right. \\
& + \left[-\frac{9}{4} \frac{1}{\mathfrak{V}^2} - \frac{3}{2} \frac{1}{\mathfrak{V}^{(2)}} - 3 \frac{1}{\mathfrak{V}} \frac{1}{\mathfrak{H}_i^2 + \lambda} - 2 \frac{1}{(\mathfrak{H}_i^2 + \lambda)^2} + 6 \frac{1}{\mathfrak{H}_i^2 + \lambda} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} + 9 \frac{1}{\mathfrak{V}} \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right. \\
& \left. \left. - 12 \left[\frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(3)}} \right]^2 + 6 \frac{\mathcal{P}^{(2)}}{\mathcal{P}^{(4)}} \right] \left[\nabla_{X_i} \lambda \right]^2 \nabla_{X_j} \lambda \nabla_{X_k} \lambda \right\} \quad (5.58)
\end{aligned}$$

Lastly, the derivatives of the variables χ_i with respect to components of the position vector χ_j that appear in Jeffery's expressions for the field velocities are given in eqns. (5.59)-(5.60) below, where repeated indices do not imply summation.

$$\nabla_{X_j} \chi_j = -2 \frac{\mathcal{P}^{(2)}}{\Delta^3} \prod_{j=1}^3 \mathfrak{H}_j = \mathfrak{K} \quad (5.59)$$

$$\nabla_{X_j} \chi_i = \mathfrak{V}_i' X_k + \mathfrak{K} \frac{(\mathfrak{H}_i^2 + \lambda) X_j}{(\mathfrak{H}_j^2 + \lambda) X_i}, \quad k = 6 - i - j \mid i \neq j \quad (5.60)$$

5.1.2.3 Finite Difference Validation of the Gradient and Hessian of Jeffery's Pressure

The derived gradients and hessian of the pressure obtained in the preceding sections are validated by simple finite difference approximations for an arbitrary design variable vector $\underline{\mathbf{y}} = [\underline{X} \quad \underline{\Theta}]^T$. Given a small perturbation $\mathfrak{g}$ the central finite difference (FD) gradient of the objective function $p(\mathbf{y}_j)$ can be approximated as

$$\nabla_k p = \frac{p(y_j + g\delta_{jk}y_j) - p(y_j - g\delta_{jk}y_j)}{2gy_k} + O(g^2) \quad (5.61)$$

Likewise, the hessian approximation of $p(y_j)$ is obtained via the same central finite difference method. i.e.

$$\nabla_{jk}^2 p = \frac{\nabla_j p(y_i + g\delta_{ik}y_i) - \nabla_j p(y_i - g\delta_{ik}y_i)}{2gy_k} + O(g^2) \quad (5.62)$$

In the eqns. (5.61) - (5.62) above, there is no summation over repeated indices. The metric adopted to assess the accuracy of the derived gradient and hessian tensors is the Frobenius norm of the difference between the exact values and finite difference approximations. i.e., the error of the gradient, $\varsigma^{(1)}$ and the error of the hessian, $\varsigma^{(2)}$ are estimated according to the respective the expressions in eqn. (5.63) below:

$$\varsigma^{(1)} = \|\underline{\nabla} p^{exact} - \underline{\nabla} p^{FD}\|_2, \quad \varsigma^{(2)} = \|\underline{\nabla^2} p^{exact} - \underline{\nabla^2} p^{FD}\|_2 \quad (5.63)$$

Given a random fiber orientation state $\underline{\Theta}^i$ and any arbitrary spatial position $\underline{X}^i$ at an instant t_i within the flow domain such that $\lambda \geq 0$, say,

$$\underline{\Theta}^i = [\pi/4 \quad -\pi/3 \quad 2\pi/5]^T, \quad \underline{X}^i = [5.45 \quad 0.85 \quad 0.25]^T \quad (5.64)$$

and considering a flow field with a fluid viscosity $\mu = 1 \text{ Pa.s}$, and random velocity gradient $\underline{L}$ say

$$\underline{L} = \begin{bmatrix} 0.8143 & 0.3500 & 0.6160 \\ 0.2435 & 0.1966 & 0.4733 \\ 0.9293 & 0.2511 & 0.3517 \end{bmatrix}$$

Based on expression of eqn. (5.63) above, assuming $g = 10^{-4}$, we obtain for the error estimates of the gradient and hessian i.e., $\varsigma^{(1)}$ & $\varsigma^{(2)}$ the following respective values

$$\varsigma^{(1)} = 9.0047 \times 10^{-8}, \quad \varsigma^{(2)} = 7.7596 \times 10^{-7}$$

5.1.3 Homogenous Flow Considerations

Various homogenous flows similar to those used in short fiber composite fiber orientation simulations [316] are considered here which serve as input for our particle motion studies. These homogenous flows also serve as a basis for understanding the flow fields development in common extrusion-deposition additive manufacturing (EDAM) polymer composite processing that involves a combination of shearing and extensional components within the flow (cf. APPENDIX B, B.3). The following flows are considered in this study:

- (i) Simple Shear flow (SS), i.e., $L_{23} = \dot{\gamma}$
- (ii) Two Stretching/Shearing flows (SUA), including simple shear in X_2X_3 plane superimposed with uniaxial elongation in the X_3 -direction, i.e., $L_{11} = L_{22} = -\dot{\epsilon}$, $L_{33} = 2\dot{\epsilon}$, $L_{23} = \dot{\gamma}$. Two cases are considered, balanced shear/stretch, $\dot{\gamma}/\dot{\epsilon} = 10$, and dominant stretch, $\dot{\gamma}/\dot{\epsilon} = 1$
- (iii) Uniaxial Elongation flow (UA) in the X_3 direction, i.e., $L_{11} = L_{22} = -\dot{\epsilon}$, $L_{33} = 2\dot{\epsilon}$
- (iv) Biaxial Elongation (BA) flow in the $X_2 - X_3$ plane, i.e., $L_{11} = -2\dot{\epsilon}$, $L_{22} = L_{33} = \dot{\epsilon}$
- (v) Two shear/planar-elongation flows (PST), including simple shear in $X_2 - X_3$ plane superimposed on planar elongation in $X_1 - X_3$ plane, i.e., $L_{11} = -\dot{\epsilon}$, $L_{33} = \dot{\epsilon}$, $L_{23} = \dot{\gamma}$. Two cases are considered including balanced shear-planar elongation with $\dot{\gamma}/\dot{\epsilon} = 10$, and dominant planar elongation with $\dot{\gamma}/\dot{\epsilon} = 1$.
- (vi) Balanced shear/bi-axial elongation flow (SBA), simple shear in the $X_2 - X_3$ plane superimposed on biaxial elongation, i.e., $L_{33} = \dot{\epsilon}$, $L_{22} = \dot{\epsilon}$, $L_{23} = \dot{\gamma}$, $L_{11} = -2\dot{\epsilon}$. Two cases are considered which include $\dot{\gamma}/\dot{\epsilon} = 1$ and $\dot{\gamma}/\dot{\epsilon} = 10$
- (vii) Triaxial Elongation flow (TA), i.e., $L_{11} = L_{22} = L_{33} = \dot{\epsilon}$

- (viii) Balanced shear/tri-axial elongation flow (STA), including simple shear in the $\mathcal{X}_2 - \mathcal{X}_3$ plane superimposed on biaxial elongation, i.e., $\mathcal{L}_{11} = \mathcal{L}_{22} = \mathcal{L}_{33} = \dot{\epsilon}$, $\mathcal{L}_{23} = \dot{\gamma}$, Two cases are considered i.e. $\dot{\gamma}/\dot{\epsilon} = 1$, and $\dot{\gamma}/\dot{\epsilon} = 10$

Classification of the various combined homogenous flow regimes based on the flow parameter $\bar{\nu}$ (cf. APPENDIX B, B.3) is given in Table 5.1 below

Table 5.1: Flow parameter values $\bar{\nu}$ for the combined homogenous flow types

$\dot{\gamma}/\dot{\epsilon}$	SUA	PST	SBA	STA
1	0.5657	0.3820	0.5657	0.4514
10	0.0283	0.0098	0.0283	0.0146

For visualization purposes and to better interpret the results that follows in later section, typical flow streamlines around a particle suspended in the mixed mode flow conditions are presented in Figure 5.2. In all flow types, simple shear is applied in the $\mathcal{X}_2 - \mathcal{X}_3$ plane and the particle is initially oriented in the $\mathcal{X}_2$ direction. The SUA flow (cf. Figure 5.2a) tends to orient the particle such that its major axis aligns with the $\mathcal{X}_3$ direction of stretching, thus mitigating the tumbling motion in the $\mathcal{X}_2 - \mathcal{X}_3$ shear plane that occurs under simple shear flow alone. The inward flow in the y-direction initially accelerates the particle, aiding the tumbling motion into the direction of applied extension. High shear to extension rate dominance is thus required to prevent the particle from stalling in the $\mathcal{X}_3$ direction. In the PST flow type shown in Figure 5.2b, the $\mathcal{X}_1$ direction inward flow tends to constrain particle tumbling motion in the $\mathcal{X}_2 - \mathcal{X}_3$ shear plane and promotes preferential alignment of the particle in the z-direction and there is no flow in the y-direction that influence the particles initial motion. Unlike the SUA flow condition, in the SBA flow regime (cf. Figure 5.2c), the $\mathcal{X}_1$ direction inward flow limits particle tumbling motion in the $\mathcal{X}_2 - \mathcal{X}_3$ shear plane without promoting directional preference for the particle alignment in the shear plane. Hence there is no tendency for particle stall to occur irrespective of the shear-extension rate dominance. Since the STA flow type has equal applied extension in all principal directions,

the deviator of the velocity gradient has no principal component, and the particle's behavior under this flow type is similar to that under simple shear flow.

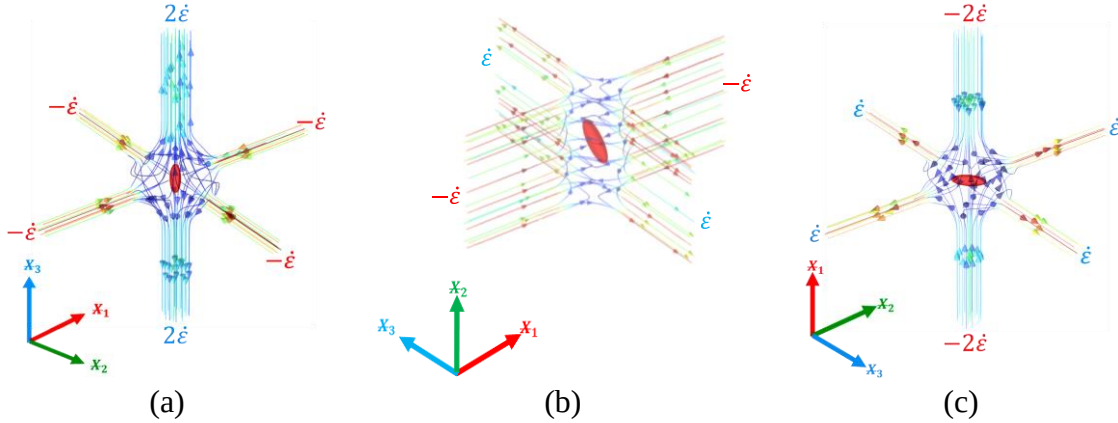


Figure 5.2: Visualization of the suspended particle in the combined shearing in $X_2 - X_3$ plane and (a) uniaxial elongation (SUA), (b) planar stretching (PST), and (c) biaxial elongation (SBA) flow conditions.

For the case of an axisymmetric ellipsoidal particle suspended in unconfined simple shear flow (see type (i) flow above) with velocity gradient $\mathbf{L}_{23} = \dot{\gamma}$, Jeffery [19] derived analytical expressions for the particle's angular velocities given as

$$\begin{aligned}\dot{\phi}(t) &= \frac{\dot{\gamma}}{2} [\kappa \cos 2\phi + 1], & \dot{\theta}(t) &= \frac{\dot{\gamma}}{2} \frac{(\kappa \sin 2\phi) \sqrt{(\kappa \cos 2\phi + 1) \zeta^2 (1 + \kappa)}}{[(\kappa \cos 2\phi + 1) + \zeta^2 (1 + \kappa)]}, \\ \dot{\psi}(t) &= -\frac{\dot{\gamma}}{2} (\kappa \cos 2\phi) \cos \theta\end{aligned}\quad (5.65)$$

where the precession $\dot{\phi}$ is observed to be independent of θ and ζ is the orbit constant. By integrating the angular velocities, Jeffery further obtained expressions for the corresponding particle orientation angles which may be written as

$$\begin{aligned}\phi(t) &= \tan^{-1} \left\{ \sqrt{\frac{1+\kappa}{1-\kappa}} \tan \left[\frac{\dot{\gamma}}{2} \sqrt{1-\kappa^2} t \right] \right\}, & \theta(t) &= \tan^{-1} \left\{ \zeta \frac{\sqrt{1+\kappa}}{\sqrt{\kappa \cos 2\phi + 1}} \right\}, \\ \psi(t) &= \int_0^t \left(\frac{\dot{\gamma}}{2} - \dot{\phi} \right) \cos \theta dt\end{aligned}\quad (5.66)$$

where $\dot{\gamma}$ is the shear-rate, κ is a shape factor given as $\kappa = (r_e^2 - 1)/(r_e^2 + 1)$. The orbit constant of integration ζ can be shown to become $\zeta = \tan \theta_0$ when $\phi_0 = 0$ and $\theta_0 \leq \theta \leq \tan^{-1}\{r_e \zeta\}$ [19]. For in-plane particle rotation, $\zeta = +\infty$ such that $\theta = \pi/2$, $\psi = 0, \dot{\psi} = \dot{\theta} = 0$. Yamane et al. [200] provides a general equation for calculating the orbital constant ζ as a function of the orientation vector ρ_i given as

$$\zeta^2 = \frac{1}{r_e^2} \left(\frac{\rho_1}{\rho_3} \right)^2 + \left(\frac{\rho_2}{\rho_3} \right)^2 \quad (5.67)$$

The corresponding period for the in-plane particle tumbling motion in simple shear flow about the ellipsoid's polar axis is

$$\tau_1 = \frac{4\pi}{\dot{\gamma} \sqrt{1 - \kappa^2}} \quad (5.68)$$

As the ellipsoid rotates in the $X_2 - X_3$ plane of shear flow, $\dot{\phi}$ reaches a maximum value when the particle is oriented normal to the principal direction of the fluid motion, i.e., at $\phi = n\pi$, $|n| \geq 0$ (cf. Figure 5.1a), and attains a minimum value when it aligns in the flow direction i.e., at $\phi = n\pi/2$, $|n| \geq 1$ [256]. The limit of the precession is thus $0 \leq \dot{\phi} \leq \dot{\gamma}$ for ellipsoidal particles and $\dot{\phi} = \dot{\gamma}/2$ for spherical particles. The extremum of the nutation $\dot{\theta}$ occurs when $\phi = \text{Re}\{.5 \cos^{-1} q\}$, where q is the solution to the cubic equation defined as

$$\{q : \kappa^2 q^3 + 3\kappa(\beta + 1)q^2 + (\kappa^2 + 2\beta + 2)q + \kappa(1 - \beta) = 0\}, \quad \beta = \zeta^2(1 + \kappa) \quad (5.69)$$

The nutation ranges between $-\dot{\gamma}/4 \leq \dot{\theta} \leq \dot{\gamma}/4$ for spheroidal particles, and it is critical for rodlike particles when $\zeta = 1/\sqrt{2}$, and for disc-like particles when $\zeta = +\infty$. It attains a value of $\dot{\theta} = 0$ for spherical particles. Likewise, the particle spin rate, $\dot{\psi}$ reaches a minimum at $\phi = n\pi$, $n \geq 0$, and a maximum value at $\phi = .5 \cos^{-1} \left\{ \left[-(3\beta + 4) \pm \sqrt{B(9\beta + 8)} \right] / 4\kappa \right\}$. The spin-rate ranges between $-\dot{\gamma}/2 \leq \dot{\psi} \leq \dot{\gamma}/2$ and it is critical for rod-like particles when $\zeta = 0$ and for disc-

shaped particle when $\zeta = +\infty$. We now consider a more complicated flow condition and derive expressions for the case of an axisymmetric particle suspended in combined elongation and shear flow, i.e., flow types (ii, v, vi, & viii) given above following similar procedures adopted by Jeffery [19] for the case of simple shear flow. Consider a flow with velocity gradient of the form

$$\underline{\underline{L}} = \begin{bmatrix} \dot{\epsilon}_1 & 0 & 0 \\ 0 & \dot{\epsilon}_2 & 0 \\ 0 & \dot{\gamma} & \dot{\epsilon}_3 \end{bmatrix} \quad (5.70)$$

where the $trace(\underline{\underline{L}}) = 0$, i.e., $\dot{\epsilon}_1 + \dot{\epsilon}_2 + \dot{\epsilon}_3 = 0$. It can be shown that the angular velocities of a particle for this $\underline{\underline{L}}$ may be written as

$$\begin{bmatrix} \dot{\phi} \\ \dot{\theta} \\ \dot{\psi} \end{bmatrix} = \begin{bmatrix} \frac{\dot{\gamma}}{2} + \frac{\kappa}{2} \{ \dot{\gamma} \cos 2\phi - [\dot{\epsilon}_2 - \dot{\epsilon}_3] \sin 2\phi \} \\ \frac{\kappa}{4} \{ \dot{\gamma} \sin 2\phi + [\dot{\epsilon}_2 - \dot{\epsilon}_3] \cos 2\phi - [2\dot{\epsilon}_1 - \dot{\epsilon}_2 - \dot{\epsilon}_3] \} \sin 2\theta \\ -\frac{\kappa}{2} \{ \dot{\gamma} \cos 2\phi - [\dot{\epsilon}_2 - \dot{\epsilon}_3] \sin 2\phi \} \cos \theta \end{bmatrix} \quad (5.71)$$

where the in-plane angular velocity reduces to

$$\dot{\phi} = \frac{d\phi}{dt} = \frac{1}{2} \{ \dot{\gamma} (1 + \kappa \cos 2\phi) - [\dot{\epsilon}_2 - \dot{\epsilon}_3] \kappa \sin 2\phi \} \quad (5.72)$$

By integrating $\dot{\phi}$ in eqn. (5.72)(5.71), we obtain an expression for the in-plane orientation angle ϕ in these flow-types with characteristics velocity gradient $\underline{\underline{L}}$ given as

$$\tan \phi = \frac{k\kappa}{\kappa - 1} \tan \left[\tan^{-1} \frac{1}{k} \left[\frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}} + \frac{\kappa - 1}{\kappa} \tan \phi_0 \right] - \frac{1}{2} k \kappa \dot{\gamma} t \right] - \frac{\kappa}{\kappa - 1} \frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}} \quad (5.73)$$

where,

$$k = \sqrt{\frac{1}{\kappa^2} - \frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}}^2} - 1 \quad (5.74)$$

If the initial orientation $\phi_0 = 0$, then eqn. (5.73) reduces to

$$\tan \phi = -\frac{\kappa}{\kappa - 1} \left[\frac{k^2 + \left[\frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}} \right]^2}{k \cot[.5k\kappa\dot{\gamma}t] + \frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}}} \right] \quad (5.75)$$

By integrating $\dot{\theta}$ in eqn. (5.71), we can directly obtain an expression for θ as

$$\tan \theta = \left[\frac{\frac{1}{\kappa} + \cos 2\phi_0 - \frac{[\dot{\epsilon}_2 - \dot{\epsilon}_3]}{\dot{\gamma}} \sin 2\phi_0}{\frac{1}{\kappa} + \cos 2\phi - \frac{[\dot{\epsilon}_2 - \dot{\epsilon}_3]}{\dot{\gamma}} \sin 2\phi} \right]^{1/2} \tan \theta_0 e^{-\kappa/2[2\dot{\epsilon}_1 - \dot{\epsilon}_2 - \dot{\epsilon}_3]t} \quad (5.76)$$

It can be shown that for the special case of initial polar orientation angle $\phi_0 = 0$, then eqn. (5.76) reduces to

$$\tan \theta = \left[\frac{\frac{1}{\kappa} + 1}{\frac{1}{\kappa} + \cos 2\phi - \frac{[\dot{\epsilon}_2 - \dot{\epsilon}_3]}{\dot{\gamma}} \sin 2\phi} \right]^{1/2} \tan \theta_0 e^{-\kappa/2[2\dot{\epsilon}_1 - \dot{\epsilon}_2 - \dot{\epsilon}_3]t} \quad (5.77)$$

Further, the spin $\psi(t)$ for these flow conditions may be written in integral form as

$$\psi(t) = \int_0^t \left(\frac{\dot{\gamma}}{2} - \dot{\phi} \right) \cos \theta dt \quad (5.78)$$

The quarter-period of rotation may be derived from eqn. (5.75) by finding the pole of the above expression of $\tan \phi$ as

$$\tau_1^{0.25} = \frac{2}{k\kappa\dot{\gamma}} \left[\pi - \tan^{-1} \left[\frac{k\dot{\gamma}}{\dot{\epsilon}_2 - \dot{\epsilon}_3} \right] \right] \quad (5.79)$$

The period for a complete tumbling motion in this flow type is obtained by finding the zero of $\tan \phi$ in eqn. (5.75) above which is given as

$$\tau_1 = \frac{4\pi}{k\kappa\dot{\gamma}} \quad (5.80)$$

When $(\dot{\epsilon}_2 - \dot{\epsilon}_3)/\dot{\gamma} = 0$, the flow is essentially simple shear, and the period is as given in eqn. (5.68) above. The particle stalls when $k^2 \leq 0$, i.e., when

$$\frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}} \geq \frac{\sqrt{1 - \kappa^2}}{\kappa} \quad (5.81)$$

and the stall angle ϕ_s is derived by equating $\dot{\phi} = 0$ (cf. eqn. (5.73)) to obtain

$$\tan 2\phi_s = \left[\frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}} \pm i \frac{\kappa}{\kappa} \right] / \left[\frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}} - \frac{1}{\kappa^2} \right], \quad \phi_s = \begin{cases} \phi_s + \pi/2, & \phi_s < 0 \\ \phi_s, & \phi_s \geq 0 \end{cases} \quad (5.82)$$

Correspondingly, given a stall angle tolerance ϕ_{tol} , the time for particle stall is obtained by equating eqn. (5.75) and (5.82), i.e. $t_s : \phi(t_s) = \phi_s - \phi_{tol}$. When eqn. (5.82) is satisfied ($k = 0$), the stall angle may be shown to be

$$\phi_{onset} = \tan^{-1} r_e \quad (5.83)$$

The particle orientation at stall for the special class of homogenous flows (described as ii, v, vi, and viii above) can be obtained by using Newton-Raphson numerical iterative process to zero the angular velocities thus

$$\underline{\underline{\theta}}_s^{\rho+} = \underline{\underline{\theta}}_s^{\rho-} - \underline{\underline{J}}_{\theta_1}^{-1} \underline{\underline{\dot{\theta}}}^{\rho} \quad (5.84)$$

where $\underline{\underline{\theta}}_s^{\rho-} = [\phi_s \quad \theta_s]^T$, $\underline{\underline{\dot{\theta}}}^{\rho} = [\dot{\phi} \quad \dot{\theta}]^T$, and the Jacobian $\underline{\underline{J}}_{\theta}$ is given as

$$\underline{\underline{J}}_{\theta_1} = \begin{bmatrix} -4\dot{\theta} \operatorname{cosec} 2\theta - \kappa[2\dot{\epsilon}_1 - \dot{\epsilon}_2 - \dot{\epsilon}_3] & 0 \\ \left\{ \dot{\phi} - \frac{\dot{\gamma}}{2} \right\} \sin 2\theta & 2\dot{\theta} \cot 2\theta \end{bmatrix} \quad (5.85)$$

For particle motion in more general class of Newtonian homogenous flows with velocity gradient $\underline{\underline{L}}$ the stall angle can be obtained using the Newton-Raphson procedure in APPENDIX B (B.4).

Jeffery's model derivations are limited to the standard assumption of single rigid ellipsoidal shaped particle suspended in Newtonian viscous linear homogenous flows. Practically speaking, the pressure driven flow of polymer melt through EDAM nozzle contraction during material processing is more accurately characterized by a quadratic ambient flow-field such as given in Lubansky et al. [261]. As such, development of a more realistic solution would involve a velocity

gradient with higher order polynomial terms which is a relevant direction for future studies. For more general conditions, it is common to employ the Finite Element Analysis (FEA) which are not bound by the limitations of the Jeffery's model and can include inter and intra fibre forces, non-ellipsoidal fibre shape, non-Newtonian visco-elastic fluid rheology, confinement flows, and other deviations from standard conditions. Moving beyond Jeffery's model assumptions may result in a preferred particle configuration that is independent of its initial orientation and may cause the particle to align with the flow or vorticity direction [175], [176], [177]. In the sections following we describe an FEA modelling approach that may be used to investigate the effect of Generalized Newtonian Fluid (GNF) rheology on the particle dynamics and surface pressure response.

5.1.4 FEA Single Particle Model with GNF Rheology

In the FEA model analysis present here, we simulate the motion of a single rigid spheroidal particle suspended in homogenous viscous flow with GNF rheology. The flow domain ϑ for the single particle micromodel analysis is shown in Figure 5.3a. The model extends the Newtonian fluid single fiber model developed by Zhang et. al. [225], [229], [291] and implemented by Awenlimobor et al., [227], [228] to simulate GNF flow. In this approach, the governing equations are based on the Stokes assumption of creeping, incompressible, isothermal, steady state, low Reynolds number viscous flow where the mass and momentum conservation equations may be written as

$$\nabla_{x_i} \dot{X}_i = 0 \quad (5.86)$$

$$\nabla_{x_i} \sigma_{ij} + f_j = 0 \quad (5.87)$$

In the above, ∇_{x_i} is the gradient operator, $\dot{X}_i$ is the flow velocity vector, f_j is the body force vector, and σ_{ij} is the Cauchy stress tensor given as

$$\sigma_{ij} = \tau_{ij} - p\delta_{ij} \quad (5.88)$$

In eqn. (5.88), p is the hydrostatic fluid pressure, δ_{ij} is the kronecker delta, and τ_{ij} is the deviatoric stress tensor defined in terms of the strain rate tensor $\dot{\gamma}_{ij}$ by the constitutive relation

$$\tau_{ij} = 2\mu(\dot{\gamma})\dot{\gamma}_{ij} \quad (5.89)$$

where the viscosity μ is a function of the strain rate magnitude $\dot{\gamma} = \sqrt{2\dot{\gamma}_{ij}\dot{\gamma}_{ji}}$. The simulations presented below solve eqns. (5.86)-(5.89) for quasi-steady velocity and pressure within the fluid domain surrounding the ellipsoidal inclusion using our custom finite element analysis (FEA) program developed in MATLAB. We assume a non-porous particle surface with zero slip allowance and velocity boundary conditions are prescribed with respect to the particle's local coordinate reference axes.

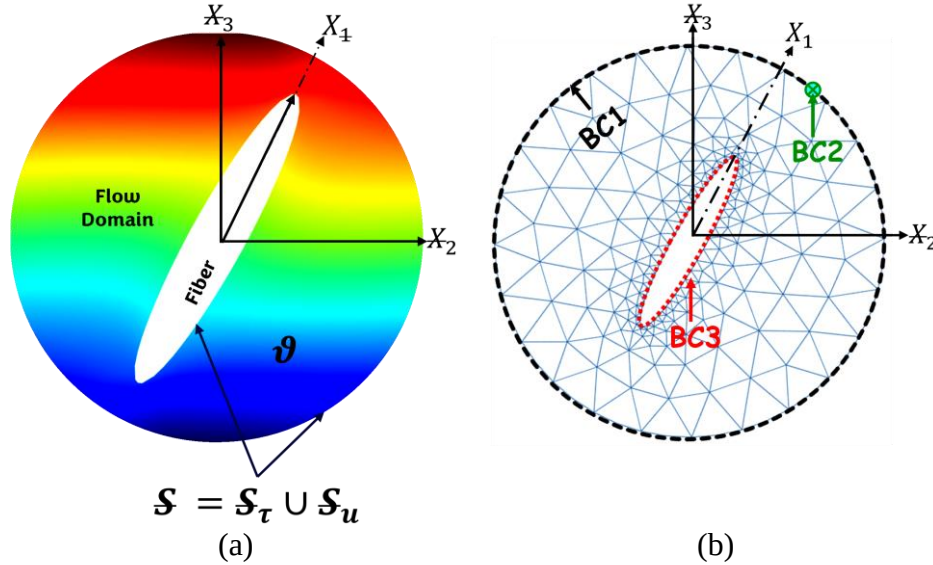


Figure 5.3: FEA model showing (a) flow domain (b) prescribed boundary conditions

Similar to previous single particle Newtonian fluid analyses [52], the velocities and velocity gradients of the prevailing flow are used to compute the far-field velocities on the fluid domain boundary $\dot{X}_i^{BC1}$ (cf. Figure 5.3b) of the micromodel as

$$\dot{X}_i^{BC1} = \dot{X}_i^\infty = Z_{X_{ji}}\dot{X}_j^\psi + Z_{X_{mi}}L_{mn}Z_{X_{nj}}\Delta X_j^{BC1} \quad (5.90)$$

where $Z_{X_{ij}}$ is the local to global transformation tensor, $\dot{\mathbf{X}}_j^\psi$ is the flow-field velocity vector, $\mathbf{L}_{mn}$ is the velocity gradient tensor in global reference frame and $\Delta\mathbf{X}_j$ is the position vector with respect to the particle's center. In 2D, $Z_{X_{ij}}$ is simply

$$Z_{X_{ij}} = \delta_{ij} \cos \phi - \mathbf{B}_{ij} \sin \phi \quad (5.91)$$

where $\mathbf{B}_{ij} = i - j$. Again, referring to Figure 5.3b, the velocity on the particle's surface $\dot{\mathbf{X}}_i^{BC3}$ is computed from the particle's center translational and rotational velocities assuming rigid body motion which is written with respect to the particle's local reference axis as

$$\dot{\mathbf{X}}_i^{BC3} = \dot{\mathbf{X}}_i^p = Z_{X_{ji}} \dot{\mathbf{X}}_j^c + \epsilon_{ijk} Z_{\Theta_{jn}} \dot{\Theta}_n \Delta\mathbf{X}_k^{BC3} \quad (5.92)$$

where $\dot{\mathbf{X}}_i^c$ is the particle's center translational velocity vector and $\dot{\Theta}_i$ is the particle's angular velocity vector. In 2D, eqn. (5.92) above can be simplified to

$$\dot{\mathbf{X}}_i^{BC3} = \dot{\mathbf{X}}_i^p = Z_{X_{ji}} \dot{\mathbf{X}}_j^c + \dot{\phi} \mathbf{B}_{ji} \Delta\mathbf{X}_j^{BC3}, \quad (5.93)$$

A pressure point constraint p_{BC2} is imposed at a node on the far-field fluid domain (see, e.g., BC2 in Figure 5.3b) with a magnitude equal to the prescribed static fluid pressure p_0 , i.e.

$$p_{BC2} = p_0 \quad (5.94)$$

We define a fluid domain size factor $\mathfrak{m} = d_f/2\mathcal{H}_3$ [52] (where d_f is the diameter of the flow domain and $\mathcal{H}_3$ is the major axis length of the particle). The flow domain size thus increases linearly with the size of the particle. In our analysis, we utilize a factor of $\mathfrak{m} = 10$ which is determined to be sufficiently large to yield accurate results. The fluid domain discretization for the base case having a particle geometric aspect ratio $r_e = 6$ appears in Figure 5.4a&b where an increasing mesh density is used near the particle and particles tip. All FEA simulations are performed with a 10-node quadratic, iso-parametric tetrahedral serendipity element as shown in Figure 5.4c.

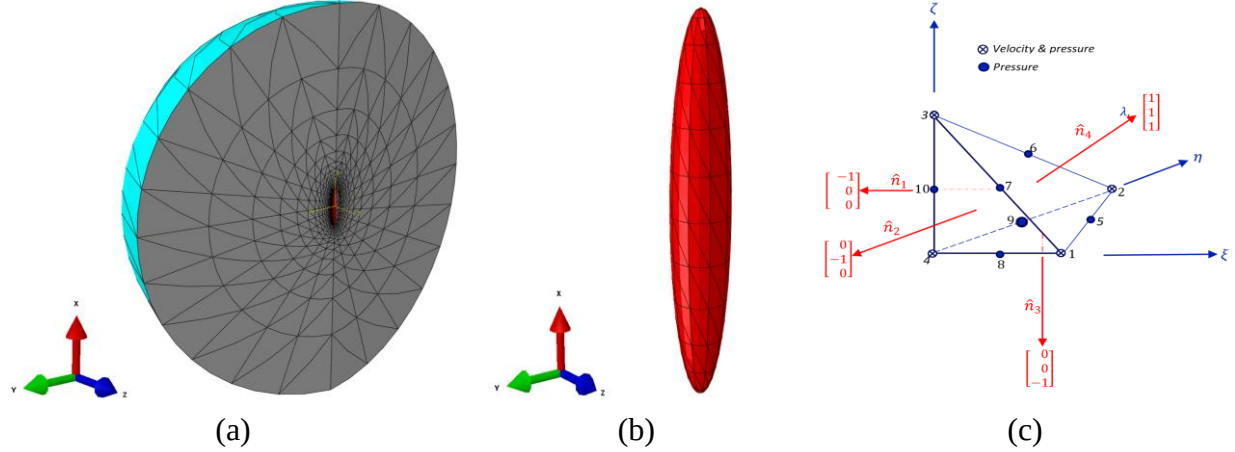


Figure 5.4: 3D Single suspended particle finite element model (a) Fluid domain discretization (b) magnified view of the domain mesh on the surface of the rigid particle (c) element selection with active degrees of freedom.

For the two-dimensional (2D) single fiber simulation, discretization of the micro-model fluid domain is achieved using a radial seed of 60-unit cells with a unidirectional geometric bias of 1.1 and circumferential seed of 60-unit cells resulting in a total of 1800 triangular elements as shown in Figure 5.5a. We employ a 6-node quadratic, iso-parametric triangle serendipity element (cf. Figure 5.5b) which has been found to give accurate results for low Reynolds number fluid flow problems [265].

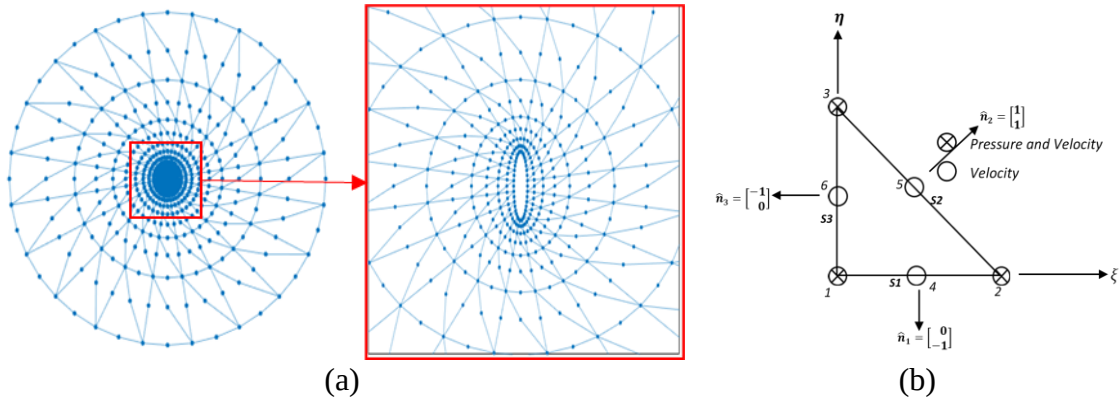


Figure 5.5: 2D Single suspended particle finite element model (a) Fluid domain discretization (b) element selection with active degrees of freedom.

For 2D sensitivity analysis involving very large fibers aspect ratio $r_e = 30$, the MATLAB inbuilt PDE modeler is used to discretize the fluid domain with increasing mesh density towards the fiber and fibers tip as shown in Figure 5.6a & b.

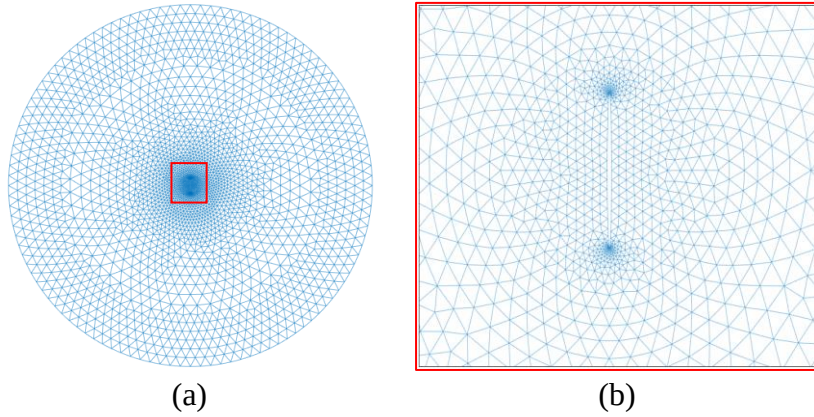


Figure 5.6: (a) Fluid domain discretization (b) magnified view of the domain mesh around the rigid fiber

The weak form of the nonlinear finite element equations may be transformed in the usual manner to a system of algebraic equations written in terms of the solution variable vector $\underline{u}$ and the global system residual vector $\underline{\Sigma}$ as

$$\underline{\Sigma} = \underline{K}(\underline{u})\underline{u} - \underline{f} \quad (5.95)$$

where $\underline{K}$ is the global system ‘stiffness’ matrix, $\underline{u} = [\underline{v} \quad \underline{p}]^T$ is the primary solution vector containing nodal velocities $\underline{v}$ and pressures $\underline{p}$ and $\underline{f}$ is the secondary variable vector containing the associated nodal reaction forces and flow rates. To simplify the solution procedure, the global system matrix is partitioned into essential ‘ e ’ (known) and free ‘ f ’ (unknown) degrees of freedom (dofs) as

$$\underline{\Sigma} = \begin{Bmatrix} \underline{\Sigma}_f \\ \underline{\Sigma}_e \end{Bmatrix} = \begin{Bmatrix} \underline{K}_{ff} & \underline{K}_{fe} \\ \underline{K}_{ef} & \underline{K}_{ee} \end{Bmatrix} \begin{Bmatrix} \underline{u}_f \\ \underline{u}_e \end{Bmatrix} - \begin{Bmatrix} \underline{f}_f \\ \underline{f}_e + \underline{g}_e \end{Bmatrix} \quad (5.96)$$

where $\underline{u}_f$ & $\underline{g}_e$ are the unknown quantities to be computed in the finite element analysis. The unknown free velocity and pressure dofs in $\underline{u}_f$ are computed via a Newton Raphson iterative

algorithm by zeroing the free residual vector $\underline{\Sigma}_f$. i.e $\underline{u}_f$ is iteratively updated until it approaches the actual solution according to

$$\underline{u}_f^+ = \underline{u}_f^- - \underline{J}_{ff}^{-1} \underline{\Sigma}_f \quad (5.97)$$

In the above, the Tangent Stiffness Matrix (TSM) or Jacobian $\underline{J}_{ff}$ is obtained by differentiating the free residual vector $\underline{\Sigma}_f$ defined in eqn. (5.96) with respect to the free degrees of freedom $\underline{u}_f$ to obtain

$$\underline{J}_{ff} = \frac{\partial \underline{\Sigma}_f}{\partial \underline{u}_f} = \frac{\partial \underline{K}_{ff}}{\partial \underline{u}_f} \underline{u}_f + \frac{\partial \underline{K}_{fe}}{\partial \underline{u}_f} \underline{u}_e + \underline{K}_{ff} - \frac{\partial \underline{f}_f}{\partial \underline{u}_f} \quad (5.98)$$

For the linear system, i.e. $\underline{K} \neq \underline{K}(\underline{u})$,

$$\underline{u}_f = \underline{K}_{ff}^{-1} (\underline{f}_f - \underline{K}_{fe} \underline{u}_e) \quad (5.99)$$

The unknown reactions forces and flow rates at the essential dofs in $\underline{g}_e$ are computed by setting the essential residual vector $\underline{\Sigma}_e = 0$ (cf. eqn. (5.96)) to obtain as

$$\underline{g}_e = \underline{K}_{ef} \underline{u}_f + \underline{K}_{ee} \underline{u}_e - \underline{f}_e \quad (5.100)$$

The global residual vector and Jacobian are assembled from individual element residual $\underline{\Sigma}^e$ and element tangent stiffness matrices $\underline{J}^e$ in the usual manner. The element residual vector $\underline{\Sigma}^e$ is written in terms of the FEA integral equations as

$$\underline{\Sigma}^e = \begin{Bmatrix} \underline{\Sigma}_1^e \\ \underline{\Sigma}_2^e \end{Bmatrix} = \begin{Bmatrix} \int_{\vartheta^e} \underline{\omega}_1 (\underline{\nabla} \cdot \underline{v}) d\vartheta \\ \int_{\vartheta^e} (\underline{v}_s \cdot \underline{\omega}_2)^T \mu(\dot{\gamma}) \underline{C}_o (\underline{v}_s \cdot \underline{v}) d\vartheta - \int_{\vartheta^e} \underline{p} (\underline{\nabla} \cdot \underline{\omega}_2) d\vartheta - \int_{\vartheta^e} \underline{p} \underline{\omega}_2^T \underline{f} d\vartheta - \int_{S_t^e} \underline{\omega}_2^T \underline{t} dS \end{Bmatrix} \quad (5.101)$$

where $\underline{\Sigma}_1^e$ & $\underline{\Sigma}_2^e$ are element residual vectors derived from mass and momentum conservation, respectively, $\underline{\omega}_1$ and $\underline{\omega}_2$ are the arbitrary FEA weighting functions on the continuity and

momentum equation, respectively, $\underline{\nabla}$ and $\underline{\nabla}_s$ are the gradient vector and symmetric gradient matrix operator, respectively, defined in [266], $\underline{p}$ and $\underline{v}$ are the pressure and velocity field variables, ρ is the fluid density, $\mu(\dot{\gamma})$ is the non-Newtonian fluid viscosity, $\underline{C}_o$ is a constant coefficient matrix, $\underline{\bar{t}}$ and $\underline{f}$ are the surface traction and the body force vectors, and S_τ^e and ϑ^e are the element surface and interior domains of integration, respectively. The element TSM $\underline{J}^e$ is obtained by differentiating the element residual vector $\underline{\Sigma}^e$ with respect to the element solution variables $\underline{u}^e$ which contains $\underline{p}^e$ and $\underline{v}^e$, i.e., $\underline{u}^e = [\underline{v}^e \quad \underline{p}^e]^T$ and

$$\underline{J}^e = \frac{\partial \underline{\Sigma}^e}{\partial \underline{u}^e} = \frac{\partial}{\partial \underline{u}^e} \left\{ \begin{matrix} \underline{\Sigma}_1^e \\ \underline{\Sigma}_2^e \end{matrix} \right\}, \quad \underline{J}^{eT} = \left[\frac{\partial}{\partial \underline{v}^e} \quad \frac{\partial}{\partial \underline{p}^e} \right]^T \left\{ \begin{matrix} \underline{\Sigma}_1^e \\ \underline{\Sigma}_2^e \end{matrix} \right\}^T \quad (5.102)$$

First order Façade derivatives are used to approximate the tangent stiffness matrix according to

$$\frac{\partial \underline{\Sigma}}{\partial \underline{u}} \Delta \underline{u} = \underline{\Sigma}(\underline{u} + \Delta \underline{u}) - \underline{\Sigma}(\underline{u}), \quad \underline{\Sigma} = \underline{\Sigma}(\underline{u}) \quad (5.103)$$

which we apply to the continuity residual term $\underline{\Sigma}_1^e$ to obtain derivatives with respect to the velocity and pressure as

$$\frac{d \underline{\Sigma}_1^e}{d \underline{v}} \Delta \underline{v} = \int_{\vartheta^e} \underline{\omega}_1 (\underline{\nabla} \cdot \Delta \underline{v}) d\vartheta, \quad \frac{d \underline{\Sigma}_1^e}{d \underline{p}} \Delta \underline{p} = 0 \quad (5.104)$$

Similarly, derivatives of the momentum conservation term with respect to the solution variables after algebraic manipulations are, respectively, given as

$$\frac{d \underline{\Sigma}_2^e}{d \underline{v}} \Delta \underline{v} = \int_{\vartheta^e} (\underline{\nabla}_s \underline{\omega}_2)^T \mu \underline{C}_o \underline{\nabla}_s \Delta \underline{v} d\vartheta + \int_{\vartheta^e} \frac{1}{\mu^2} \frac{1}{\dot{\gamma}} \frac{\partial \mu}{\partial \dot{\gamma}} \left[(\underline{\nabla}_s \underline{\omega}_2)^T \mu \underline{C}_o \underline{\nabla}_s \underline{v} \right] \left[(\underline{\nabla}_s \underline{v})^T \mu \underline{C}_o \underline{\nabla}_s \Delta \underline{v} \right] d\vartheta \quad (5.105)$$

$$\frac{d \underline{\Sigma}_2^e}{d \underline{p}} \Delta \underline{p} = - \int_{\vartheta^e} (\underline{\nabla} \cdot \underline{\omega}_2) \Delta \underline{p} d\vartheta \quad (5.106)$$

It follows that the Galerkin formulation written as the element residual vector $\underline{\Sigma}^e$ and tangent stiffness matrix $\underline{J}^e$ in tensorial representation are given respectively as

$$\underline{\underline{\Sigma}}^e = \begin{pmatrix} \int_{\vartheta^e} \underline{\underline{B}}_s^{eT} \mu(\dot{\gamma}) \underline{\underline{C}}_o \underline{\underline{B}}_s^e d\vartheta & - \int_{\vartheta^e} \underline{\underline{B}}^{eT} \underline{\underline{\Phi}}^e d\vartheta \\ - \int_{\vartheta^e} \underline{\underline{\Phi}}^{eT} \underline{\underline{B}}^e d\vartheta & \underline{\underline{0}} \end{pmatrix} \begin{pmatrix} \underline{\underline{v}}^e \\ \underline{\underline{p}}^e \end{pmatrix} - \begin{pmatrix} \int_{\vartheta^e} \underline{\underline{p}} \underline{\underline{N}}^{eT} \underline{\underline{f}} d\vartheta + \int_{S_r^e} \underline{\underline{N}}^{eT} \underline{\underline{t}} dS \\ \underline{\underline{0}} \end{pmatrix} \quad (5.107)$$

and

$$\underline{\underline{J}}^e = \frac{d\underline{\underline{\Sigma}}^e}{d\underline{\underline{u}}^e} = \begin{pmatrix} \int_{\vartheta^e} \underline{\underline{B}}_s^{eT} \mu \underline{\underline{C}}_o \underline{\underline{B}}_s^e d\vartheta + \int_{\vartheta^e} \frac{1}{\mu^2} \frac{1}{\dot{\gamma}} \frac{\partial \mu}{\partial \dot{\gamma}} \left(\underline{\underline{B}}_s^{eT} \mu \underline{\underline{C}}_o \underline{\underline{B}}_s^e \underline{\underline{v}}^e \right) \left(\underline{\underline{v}}^{eT} \underline{\underline{B}}_s^{eT} \mu(\dot{\gamma}) \underline{\underline{C}}_o^T \underline{\underline{B}}_s^e \right) d\vartheta & - \int_{\vartheta^e} \underline{\underline{B}}^{eT} \underline{\underline{\Phi}}^e d\vartheta \\ - \int_{\vartheta^e} \underline{\underline{\Phi}}^{eT} \underline{\underline{B}}^e d\vartheta & \underline{\underline{0}} \end{pmatrix} \quad (5.108)$$

where

$\underline{\underline{\Phi}}^e$ and $\underline{\underline{N}}^e$ are the pressure and velocity interpolation functions, respectively,
 $\underline{\underline{B}}^e$ and $\underline{\underline{B}}_s^e$ are ‘strain’ displacement matrices
 $\underline{\underline{v}}^e$ and $\underline{\underline{p}}^e$ are respectively the velocities and pressures degrees-of-freedom (dof) at the respective element nodes
 S^e and ϑ^e are the element boundary surfaces and domain of integration, respectively.

In eqn. (5.108), $\dot{\gamma}$ is the scalar magnitude of the strain rate tensor $\dot{\underline{\underline{\gamma}}}$ which may be written in terms

of FEA quantities as

$$\dot{\gamma} = \sqrt{\frac{1}{2} \dot{\underline{\underline{\gamma}}} : \dot{\underline{\underline{\gamma}}}} = \sqrt{\left(\underline{\underline{v}}_s \underline{\underline{v}} \right)^T \underline{\underline{C}}_o \left(\underline{\underline{v}}_s \underline{\underline{v}} \right)}, \quad \dot{\gamma} = \sqrt{\underline{\underline{v}}^{eT} \underline{\underline{B}}_s^{eT} \mu(\dot{\gamma}) \underline{\underline{C}}_o \underline{\underline{B}}_s^e \underline{\underline{v}}^e} \quad (5.109)$$

In this work, we consider the non-Newtonian viscosity $\mu(\dot{\gamma})$ as that of a power-law shear-thinning fluid given as

$$\mu = m \dot{\gamma}^{n-1} \quad (5.110)$$

where m is the flow consistency coefficient in $Pa \cdot s^n$ and n is the power-law index, and $\dot{\gamma}$ is the scalar magnitude of the deformation tensor $\dot{\gamma}_{ij}$. In the second integral of the momentum equation Jacobian in eqn. (5.108) above, it is convenient to introduce a variable $\alpha = 1/(\mu^2 \dot{\gamma}) (\partial \mu / \partial \dot{\gamma})$ to

simplify the expression and make it generally applicable to other GNF fluids. It follows that α can be written for the power-law fluid as

$$\alpha = \frac{1}{\mu^2} \frac{1}{\dot{\gamma}} \frac{\partial \mu}{\partial \dot{\gamma}} = \frac{1}{\mu \dot{\gamma}^2} (n - 1) \quad (5.111)$$

Alternatively, for a Carreau-Yasuda fluid, the expression for μ and α are, respectively,

$$\frac{\mu - \mu_\infty}{\mu_0 - \mu_\infty} = \{1 + (\lambda \dot{\gamma})^a\}^{(n-1)/a} \quad \text{and} \quad \alpha = \frac{1}{\mu^2} \frac{1}{\dot{\gamma}} \frac{\partial \mu}{\partial \dot{\gamma}} = \frac{1}{\dot{\gamma}^2} \frac{\mu - \mu_\infty}{\mu^2} \left\{ \frac{n-1}{1 + (\lambda \dot{\gamma})^{-a}} \right\} \quad (5.112)$$

where, μ_0 is the zero-shear viscosity, μ_∞ is an infinite-shear viscosity, λ is a time constant, and a is a fitting parameter.

5.1.5 Single Particle Motion with GNF Rheology

In our numerical approach, the particle's motion is computed based on an appropriate explicit numerical ordinary differential equation solution technique by calculating its linear and rotational velocities that results in a zero net hydrodynamic force and torque acting on the particle's surface. Again, we adopt the Newton-Raphson's iterative method to determine the nonlinear solution of particle's translational and rotational velocities as

$$\underline{\dot{Y}}^+ = \underline{\dot{Y}}^- - \underline{J}_H^{-1} \underline{\Sigma}_H \quad (5.113)$$

where $\underline{\dot{Y}}$ contains the particle's linear velocities $\underline{\dot{X}}^c$ and rotational velocity $\underline{\dot{\Psi}}$, i.e., $\underline{\dot{Y}} = [\underline{\dot{X}}^c \quad \underline{\dot{\Psi}}]^T$ and $\underline{\Sigma}_H$ is the particle hydrodynamic residual vector which is composed of the particle's hydrodynamic forces $\underline{F}_H$ and couple $\underline{Q}_H$, i.e., $\underline{\Sigma}_H = [\underline{F}_H \quad \underline{Q}_H]^T$ as a function of the particle's velocity, i.e., $\underline{\Sigma}_H = \underline{\Sigma}_H(\underline{\dot{Y}})$. Since calculations are performed with respect to the particle's local reference frame, the particle's velocity vector is transformed to global coordinate system according to the eqn. (5.114)

$$\underline{\dot{\underline{Y}}} = \underline{\underline{Z}}_{\underline{\dot{Y}}} \underline{\dot{Y}} \quad (5.114)$$

where variables on the global reference frame are accented by a strikethrough and the particle's velocity transformation tensor $\underline{\underline{Z}}_{\underline{\dot{Y}}}$ 3D and 2D are respectively given by

$$\underline{\underline{Z}}_{\underline{\dot{Y}}} = \begin{bmatrix} \underline{\underline{Z}}_X & \underline{\underline{0}} \\ \underline{\underline{0}}^T & \underline{\underline{Z}}_{\underline{\Theta}}^{-1} \end{bmatrix}, \quad \underline{\underline{Z}}_{\underline{\dot{Y}}} = \begin{bmatrix} \underline{\underline{Z}}_X & \underline{\underline{0}} \\ \underline{\underline{0}}^T & 1 \end{bmatrix} \quad (5.115)$$

We calculate the net hydrodynamic force vector $\underline{F}_H$ and couple $\underline{Q}_H$ on the particle by vector summation of the nodal reactions forces and torques on the particle surface as

$$\underline{F}_H = - \sum_k^{n_k} \underline{g}_e^{(k)}, \quad \underline{Q}_H = - \sum_k^{n_k} \underline{\Delta X}^{(k)} \times \underline{g}_e^{(k)} \quad (5.116)$$

where $\underline{\Delta X}^{(k)}$, and $\underline{g}_e^{(k)}$ are the position vector and the nodal reaction force vector at the k^{th} node on the particle surface ($BC3$), respectively, and n_k is the total number of nodes on $BC3$. The particle hydrodynamic Jacobian $\underline{J}_H$ in eqn. (5.113) above is obtained by differentiating the components of the particle hydrodynamic residual vector $\underline{\Sigma}_H$ with respect to components of the particle's velocity vector $\underline{\dot{Y}}$ as

$$\underline{J}_H = \frac{\partial \underline{\Sigma}_H}{\partial \underline{\dot{Y}}} = \frac{\partial}{\partial \underline{\dot{Y}}} [\underline{F}_H \quad \underline{Q}_H]^T = \left[- \sum_k^{n_k} \frac{\partial \underline{g}_e^{(k)}}{\partial \underline{\dot{Y}}} \quad - \sum_k^{n_k} \underline{\Delta X}^{(k)} \times \frac{\partial \underline{g}_e^{(k)}}{\partial \underline{\dot{Y}}} \right]^T \quad (5.117)$$

Differentiating the global system FEA residual vector $\underline{\Sigma}$ in eqn. (5.96) with respect to the particle velocity vector $\underline{\dot{Y}}$ we obtain the derivative of the nodal reaction force vector $d\underline{g}_e/d\underline{\dot{Y}}$ in eqn. (5.117) as

$$\frac{d\underline{g}_e}{d\underline{\dot{Y}}} = \left\{ \frac{\partial \underline{K}_{ef}}{\partial \underline{u}_e} \underline{u}_f + \frac{\partial \underline{K}_{ee}}{\partial \underline{u}_e} \underline{u}_e + 2\underline{K}_{ee} - \frac{d\underline{f}_e}{d\underline{u}_e} \right\} \frac{d\underline{u}_e}{d\underline{\dot{Y}}} + \left\{ \frac{\partial \underline{K}_{ef}}{\partial \underline{u}_f} \underline{u}_f + \frac{\partial \underline{K}_{ee}}{\partial \underline{u}_f} \underline{u}_e + 2\underline{K}_{ef} - \frac{d\underline{f}_e}{d\underline{u}_f} \right\} \frac{d\underline{u}_f}{d\underline{\dot{Y}}} \quad (5.118)$$

where the derivative $d\underline{u}_f/d\underline{\dot{Y}}$ is written in terms of the derivative $d\underline{u}_e/d\underline{\dot{Y}}$ as

$$\frac{d\underline{u}_f}{d\underline{\dot{Y}}} = - \left\{ \frac{\partial \underline{K}_{ff}}{\partial \underline{u}_f} \underline{u}_f + \frac{\partial \underline{K}_{fe}}{\partial \underline{u}_f} \underline{u}_e + 2\underline{K}_{ff} - \frac{d\underline{f}_f}{d\underline{\dot{Y}}} \right\}^{-1} \left\{ \frac{\partial \underline{K}_{ff}}{\partial \underline{u}_e} \underline{u}_f + \frac{\partial \underline{K}_{fe}}{\partial \underline{u}_e} \underline{u}_e + 2\underline{K}_{fe} - \frac{d\underline{f}_f}{d\underline{\dot{Y}}} \right\} \frac{d\underline{u}_e}{d\underline{\dot{Y}}} \quad (5.119)$$

To obtain the FEA model derivatives in the above, we differentiate the global FEA system residual $\underline{\Sigma}$ in eqn. (5.96) with respect to the solution variable $\underline{u}$ to obtain the global FEA system Jacobian $\underline{J}$ as

$$\begin{aligned} \underline{J} &= \frac{d\underline{\Sigma}}{d\underline{u}} = \begin{Bmatrix} \underline{J}_{ff} & \underline{J}_{fe} \\ \underline{J}_{ef} & \underline{J}_{ee} \end{Bmatrix} \\ &= \begin{Bmatrix} \left\{ \frac{\partial \underline{K}_{ff}}{\partial \underline{u}_f} \underline{u}_f + \frac{\partial \underline{K}_{fe}}{\partial \underline{u}_f} \underline{u}_e + \underline{K}_{ff} - \frac{\partial \underline{f}_f}{\partial \underline{u}_f} \right\} & \left\{ \frac{\partial \underline{K}_{ff}}{\partial \underline{u}_e} \underline{u}_f + \frac{\partial \underline{K}_{fe}}{\partial \underline{u}_e} \underline{u}_e + \underline{K}_{fe} - \frac{\partial \underline{f}_f}{\partial \underline{u}_e} \right\} \\ \left\{ \frac{\partial \underline{K}_{ef}}{\partial \underline{u}_f} \underline{u}_f + \frac{\partial \underline{K}_{ee}}{\partial \underline{u}_f} \underline{u}_e + \underline{K}_{ef} - \frac{\partial \underline{f}_e}{\partial \underline{u}_f} \right\} & \left\{ \frac{\partial \underline{K}_{ef}}{\partial \underline{u}_e} \underline{u}_f + \frac{\partial \underline{K}_{ee}}{\partial \underline{u}_e} \underline{u}_e + \underline{K}_{ee} - \frac{\partial \underline{f}_e}{\partial \underline{u}_e} \right\} \end{Bmatrix} \end{aligned} \quad (5.120)$$

where eqn. (5.120) has been expanded to include all free and essential degrees of freedom in $\underline{u} = \{\underline{u}_f \quad \underline{u}_e\}^T$. In addition, the nodal reaction force vector derivative $d\underline{g}_e/d\underline{\dot{Y}}$ in eqn. (5.118) is written in terms of the submatrices of the global FEA system Jacobian $\underline{J}$ as

$$\frac{d\underline{g}_e}{d\underline{\dot{Y}}} = \left\{ \underline{K}_{ee} + \underline{J}_{ee} \right\} \frac{d\underline{u}_e}{d\underline{\dot{Y}}} + \left\{ \underline{K}_{ef} + \underline{J}_{ef} \right\} \frac{d\underline{u}_f}{d\underline{\dot{Y}}} \quad (5.121)$$

Likewise, the derivative $d\underline{u}_f/d\underline{\dot{Y}}$ in eqn. (5.119) is also written in terms of the submatrices of the global system Jacobian $\underline{J}$ as

$$\frac{d\underline{u}_f}{d\underline{\dot{Y}}} = - \left\{ \underline{K}_{ff} + \underline{J}_{ff} \right\}^{-1} \left\{ \underline{K}_{fe} + \underline{J}_{fe} \right\} \frac{d\underline{u}_e}{d\underline{\dot{Y}}} \quad (5.122)$$

Again, for the linear consideration i.e. $\underline{K} \neq \underline{K}(\underline{u})$, $\underline{J} = 0$ the derivative of the nodal reaction force vector derivative $d\underline{g}_e/d\underline{\dot{Y}}$ in eqn. (5.118) reduces to

$$\frac{\partial \underline{g}_e}{\partial \underline{\dot{Y}}} = \left(\underline{K}_{ee} - \underline{K}_{ef} \underline{K}_{ff}^{-1} \underline{K}_{fe} \right) \frac{\partial \underline{u}_e}{\partial \underline{\dot{Y}}} \quad (5.123)$$

Given the initial condition of the particle, $\underline{Y}^{j-1}$ at any instant with an associated velocity $\dot{\underline{Y}}^{j-1}$ at each j^{th} time step, we update particle's position and orientation $\underline{Y}^j$ using on an explicit fourth order Runge-Kutta method. i.e.

$$\underline{Y}^j = \underline{Y}^{j-1} + \frac{\Delta t}{6} [\underline{\mathcal{K}}_1^{j-1} + 2\underline{\mathcal{K}}_2^{j-1} + 2\underline{\mathcal{K}}_3^{j-1} + \underline{\mathcal{K}}_4^{j-1}] \quad (5.124)$$

where

$$\begin{aligned} \underline{\mathcal{K}}_1^{j-1} &= f_Y(t^{j-1}, \underline{Y}^{j-1}) = \dot{\underline{Y}}^{j-1}, \quad \underline{\mathcal{K}}_2^{j-1} = f_Y(t^{j-1} + \Delta t/2, \underline{Y}^{j-1} + \Delta t/2 \underline{\mathcal{K}}_1^{j-1}) \\ \underline{\mathcal{K}}_3^{j-1} &= f_Y(t^{j-1} + \Delta t/2, \underline{Y}^{j-1} + \Delta t/2 \underline{\mathcal{K}}_2^{j-1}), \quad \underline{\mathcal{K}}_4^{j-1} = f_Y(t^{j-1} + \Delta t, \underline{Y}^{j-1} + \Delta t \underline{\mathcal{K}}_3^{j-1}) \end{aligned} \quad (5.125)$$

and the function f_Y is used to evaluate the particles velocities $\dot{\underline{Y}}$ at time t and position $\underline{Y}$

5.1.6 Cylindrical Particle Geometry

In reality, the geometry of pristine particle consolidations present within a typical polymer composite bead are not ellipsoidal in shape with smooth edges but are better represented by cylindrical particles. Moreover, the chopped ends of the particles reinforcement do not possess a clearly defined tip as the ellipsoid but are rather characterized by sharp geometrical transitions at the particle terminations that likely results in pressure singularities. Unfortunately, besides other drawbacks, Jeffery's model equations are only applicable to ellipsoidal shaped particles and cannot model arbitrary shaped particles, however, our FEA simulation has the advantage of modelling complex particle shapes. To investigate the existence of exacerbated pressure extremes at the particle ends, we consider a cylindrical shaped particle in our FEA simulation choosing a cylindrical aspect ratio that yields the equivalent hydrodynamic ellipsoidal aspect ratio for the base case (i.e. $r_e = 6$). We develop a fluid domain mesh using ABAQUS Std. (Simulia ABAQUS, Dassault Systemes SE, Velizy-Villacoublay, France) for the single cylinder suspension using similar fluid domain size ratio, 10 times the cylinder length as shown in Figure 5.7a below. Mesh refinement zone close the cylinder surface is defined to accurately capture the field response on

the particles surface (cf. Figure 5.7b). As would become evident in subsequent chapters, the pressure at the particles tip is dependent on the tip curvature and aspect ratio. With ellipsoidal shaped particles, both geometric attributes are interdependent and cannot be decoupled which limits our understanding of the individual contribution of both attributes to the surface pressure at the particles tip. With cylindrical shaped particles however, we can independently study the individual contribution of both geometric attributes to the tip pressure response. In our analysis, we consider different end conditions (i.e. edge curvature radii - r_k) as shown in Figure 5.7c ranging from small fillet radius to perfectly hemispherical (i.e. $0.05 \leq \bar{r}_k \leq 0.5$) where $\bar{r}_k = r_k/U_1$ is the normalized curvature radius and U_1 is the cylinder diameter. By adjusting the cylindrical height while maintaining a constant diameter, U_1 we determine their respective cylinders geometric aspect ratio that are hydrodynamically equivalent to the ellipsoidal aspect ratio of $r_e = 6$ by matching output of the dynamic response. After careful determination of cylinder appropriate heights, the results of the evolution of the angular velocity along Jeffery's orbit for the various cylindrical particles with different end conditions are benchmarked with angular velocity of the ellipsoidal particle with aspect ratio of $r_e = 6$ using the same fluid viscosity and shear rate. To study the independent effect of the cylinder aspect ratio on the pressure response while maintaining a constant particle end curvature, the cylinder with the hemispherical end was chosen. For objectivity, the aspect ratio was varied by adjusting the length of the straight section of the cylinder while retaining a constant mesh for the hemispherical curved surface. Moreover, since only the cylinder with the hemispherical end has a clearly defined unique tips where the surface pressure extremes are expected to occur, it provides a biased means for studying the decoupled effect of aspect ratio on the tip pressure response compared to the ellipsoid. In our investigation, we

consider five (5) aspect ratios for the cylinder with the hemispherical end ranging from $7.0 \leq r_c \leq 7.4$ in steps of 0.1.

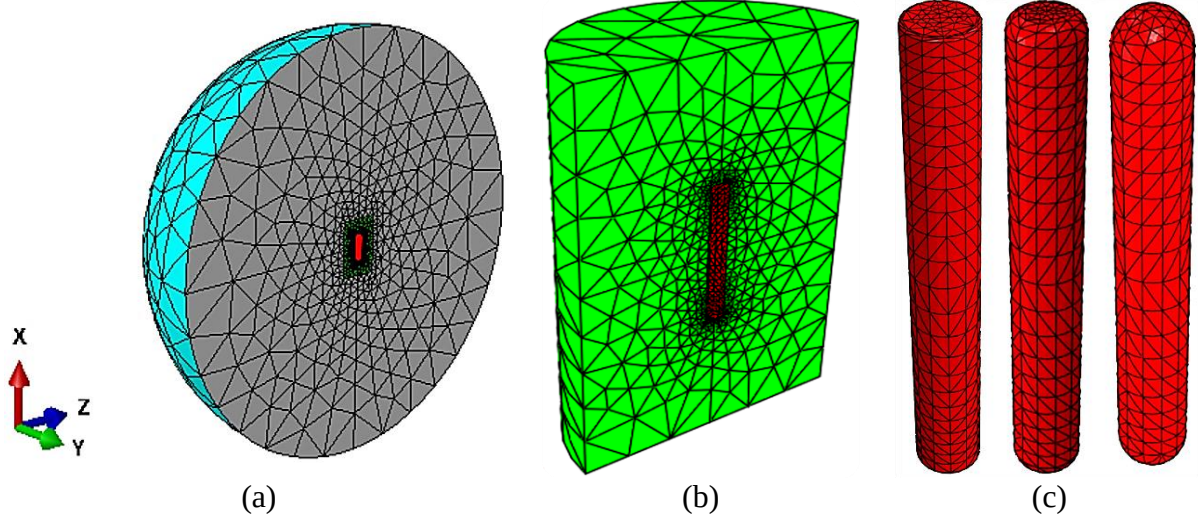


Figure 5.7: (a) Fluid domain discretization around the cylindrical particle (b) mesh refinement around the cylindrical particle surface (c) cylindrical particle with different end conditions (edge curvature radius).

5.1.7 Validation of FEA Model Development

To validate our FEA model-based particle motion simulations to calculations performed with Jeffery's equations, we first define the particle surface pressure $\bar{p}$ in dimensionless form as

$$\bar{p} = \frac{p - p_0}{\mu_1 \dot{\gamma}_c} \quad (5.126)$$

where $\dot{\gamma}_c$ is a characteristic strain rate of the flow-field. For a given μ_1 and $\dot{\gamma}_c$, $\bar{p}$ is evaluated from eqn. (5.126) where p is computed from Jeffery's model (cf. eqn. (5.1)) and similarly from the nodal pressure solution of the FEA model described above. Likewise, the flow-field velocity magnitude is normalized with respect to the tangential velocity at the particle's tip is given as

$$\bar{v} = |\dot{\underline{x}}|/|\dot{\underline{x}}^t|, \quad \dot{\underline{x}}^t = \underline{\Theta} \times \underline{x}^t \quad (5.127)$$

where $\underline{x}^t$ is the position vector at particle's tip defined by the major axis length. To ensure consistency between the Jeffery's model equations and Finite Element Analysis (FEA) simulation results, we consider the particle's motion and surface pressure distribution for the case of a single

rigid ellipsoidal particle suspended in viscous homogenous Newtonian (i.e., power-law index $n = 1$) flow. The FEA model is shown to exactly match Jeffery's results for a range of particle aspect ratios including $r_e = 1, 2, 3, 6$, and 10 (cf. Figure 5.8a for $\dot{\phi}$ and Figure 5.8b for $\bar{P}$). $\bar{P}$ is the dimensionless pressure at the particle's tip. Additionally, Jeffery's orbit exactly matches our FEA results for the various flow conditions described above as shown in Figure 5.8c&d which show components of the particle unit vector ρ_i , and maximum and minimum normalized surface pressure $\tilde{p}$. Results in Figure 5.8a & b are for one period of Jeffery's orbit, however, given that values at the end point exactly match within 0.25%, we expect the accuracy of our numerical approach to remain as particle rotations continue.

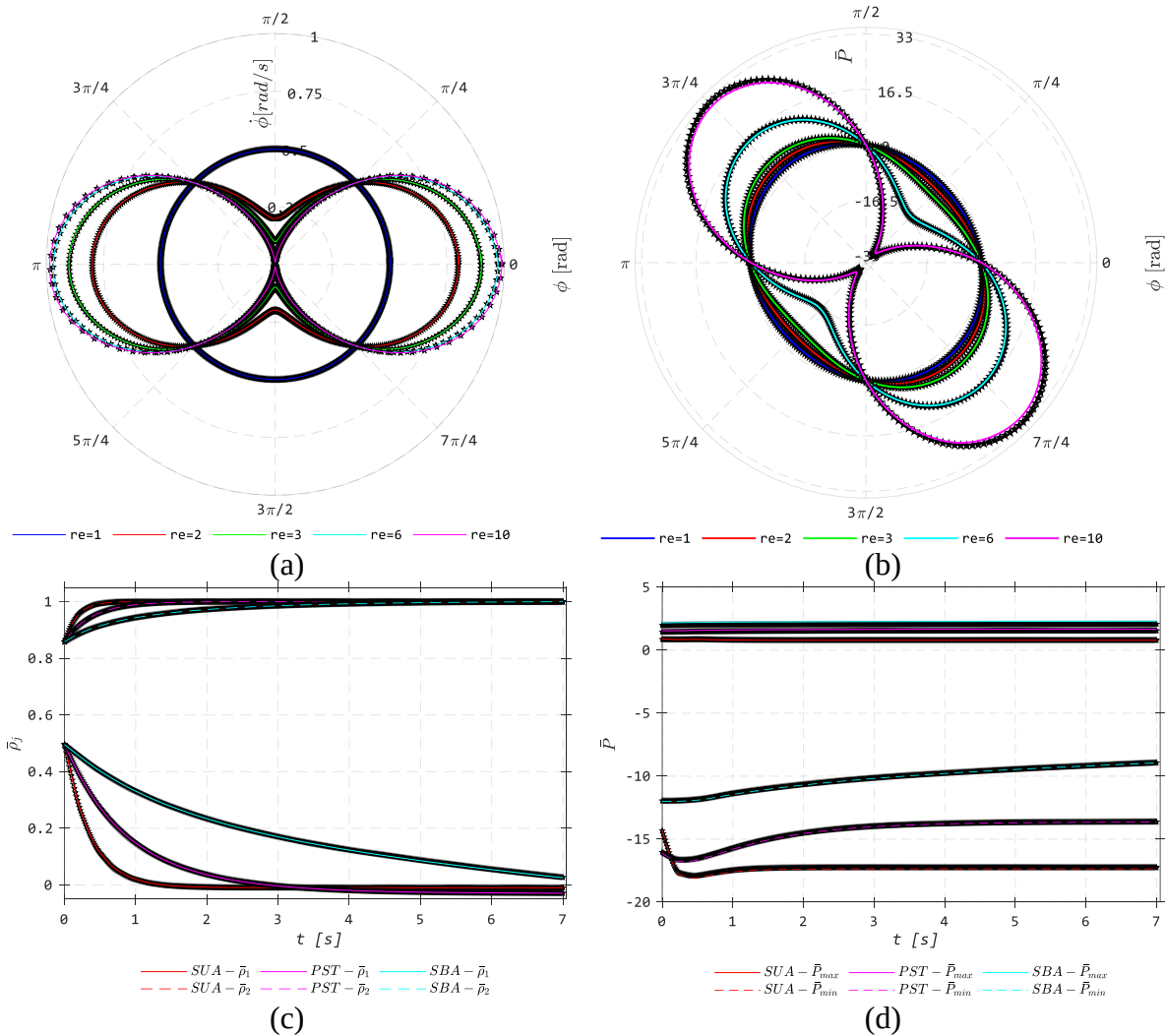
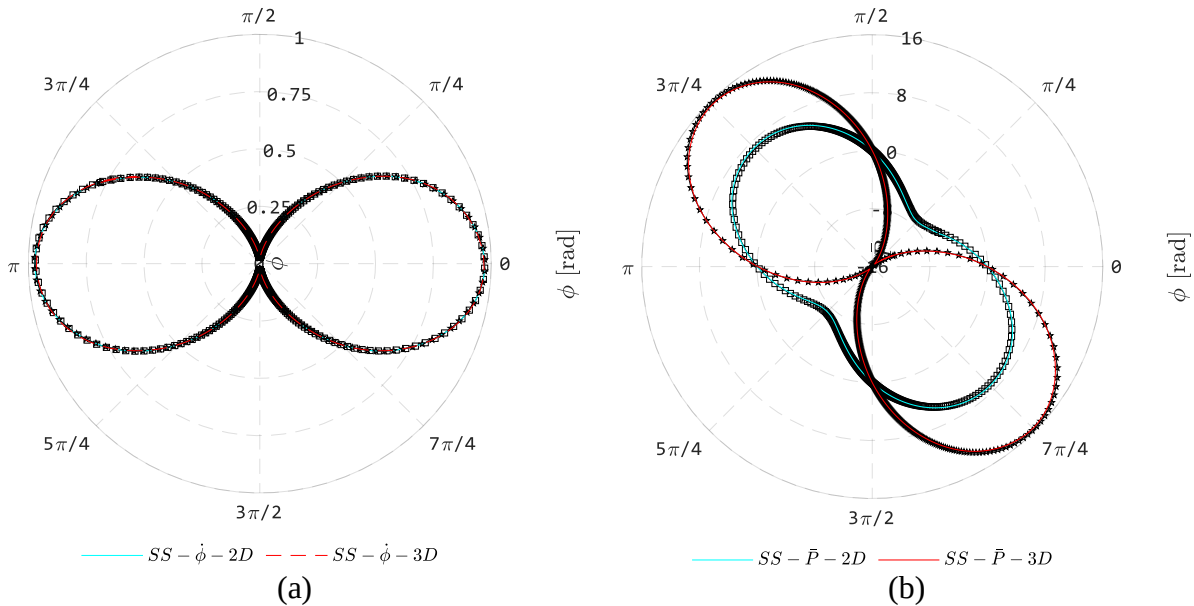


Figure 5.8¹: FEA (colored lines) and Jeffery's results (black markers) of the evolution of the particle's (a) angular velocity, & (b) tip pressure, in simple shear flow for particle tumbling in the shear plane with different aspect ratios $1 \leq r_e \leq 10$; (c) orientation components, and (d) minimum (dashed) and maximum (continuous) surface pressure for particle with initial orientation, $\phi^0 = \pi/3, \theta^0 = 11\pi/24, \psi^0 = 0$ suspended in different combined flow types - SUA (red), PST (pink) and SBA (cyan) with $\dot{\gamma}/\dot{\epsilon} = 1$.

The FEA results of the particle's angular velocity and tip surface pressure for the suspended rigid particle motion in simple shear flow in both 2D and 3D space are compared and validated against their corresponding reference counterparts computed from Jeffery's analytic equations as shown in Figure 5.9. We likewise observe very good agreement in the responses obtained from both FEA and Jeffery's solution irrespective of the dimensional space. While the particle's in-plane angular velocity is unaffected by the dimensional space evident from the overlapping curves in Figure 5.9a, the same is not the case for the particle's surface pressure as the pressure response magnitude is observed to reduce significantly with reduction in dimensionality of the computational space as can be observed from Figure 5.9b.



¹ Results of the 3rd component of the particle's orientation vector (i.e. $\bar{\rho}_3$) is implicit given the normalization condition $\bar{\rho}_i \bar{\rho}_i = 1$.

Figure 5.9: FEA (colored lines) and Jeffery's results (black markers) of the evolution of the particle's (a) in-plane angular velocity (b) tip pressure, in simple shear flow and for in-plane tumbling of the particle in both 2D space (cyan curve) and 3D space (red curve) with $r_e = 6$.

Figure 5.10 shows the evolution of the maximum (red trend) and minimum (cyan trend) pressure on the fiber surface over the tumbling period from Jeffery's (black markers) and FEA simulation (colored trends) in both 2D (dashed trend) and 3D (continuous trend) space. The location of the pressure extremes varies from point to point on the fiber's surface during its motion along Jeffery's orbit. As a result, the pressure extreme depends on the mesh refinement on the fiber surface which results in minor discrepancies observed between the extreme pressure profiles obtained from Jeffery's exact solution and FEA simulation in Figure 5.10.

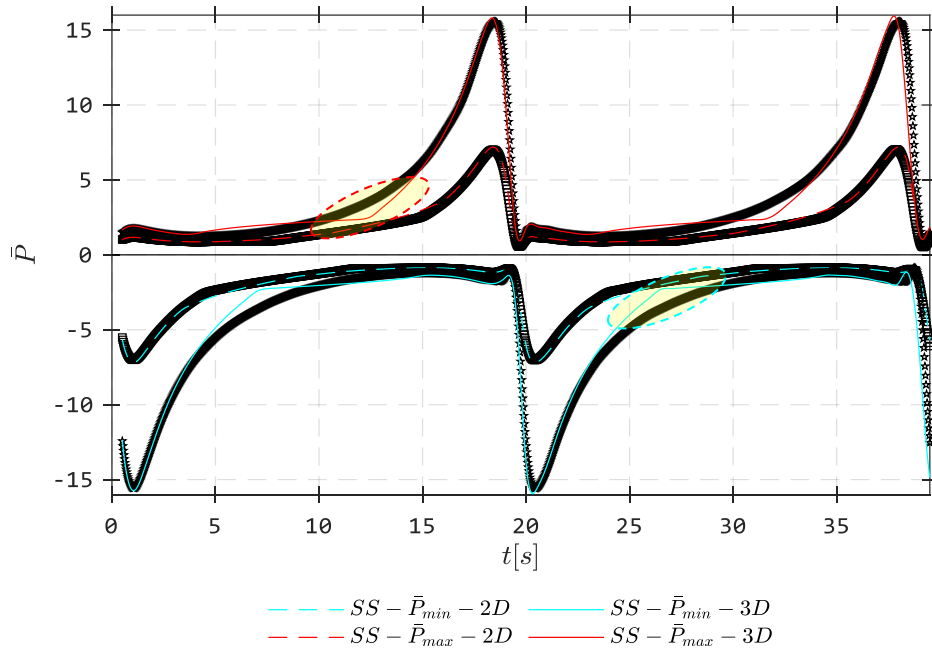


Figure 5.10: FEA (colored lines) and Jeffery's results (black markers) of the evolution of the maximum (red lines) and minimum (cyan lines) surface pressure for particle tumbling in in 2D (dashed line) and 3D (continuous line) space.

The GNF power law FEA model development is validated by benchmarking pressure response obtained from the custom-built MATLAB FEA simulation for a single steady state condition and fiber configuration with outputs obtained from a similar simulation developed with

COMSOL Multiphysics software (COMSOL, Inc., Burlington, MA, USA) using same model input. A fibers geometric aspect ratio $r_e = 6$ is used for the validation exercise, and a simple shear flow field with a shear rate of $\dot{\gamma} = 1 \text{ s}^{-1}$ is imposed. We consider two different power law fluid definition with different flow behavior index n , for the first case (a) $n = 0.2$, and for the second case (b) $n = 1.$, both cases having a consistency coefficient $m = 1 \text{ Pa} \cdot \text{s}^n$. An initial fiber configuration corresponding to an orientation $\phi_0 = -0.7762 \text{ rad}$ and angular velocity of $\dot{\phi}_0 = -0.5087 \text{ rads}^{-1}$ has been used for the steady state analysis which is where the first minimum pressure peak occurs on the fibers surface during its evolution along Jeffery's orbit. The result of the pressure distribution for both cases presented in Figure 5.11 and the pressure extremes on the fiber's surface in Table 5.2 shows there is good agreement between COMSOL simulation and inbuilt MATLAB FEA simulations. We observe a maximum discrepancy in pressure limits of about 6%.

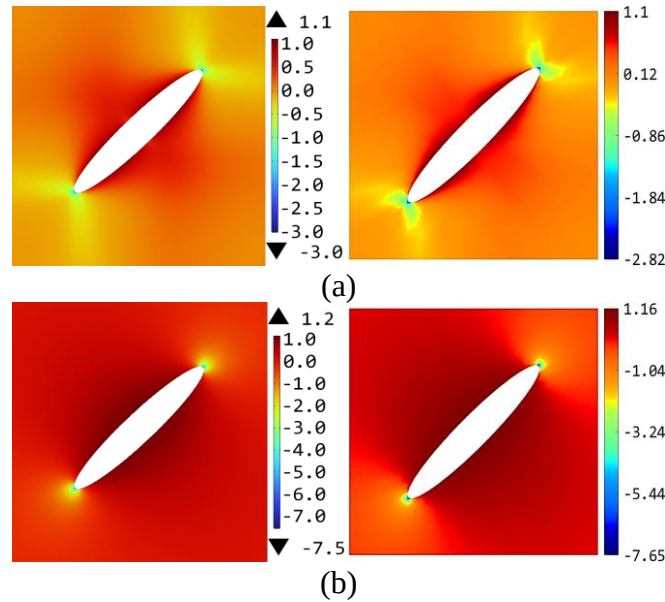


Figure 5.11: Figure showing pressure distribution around the fiber for power law index corresponding to (a) $n = 0.2$, (b) $n = 1.0$, for COMSOL (left of each case) and MATLAB (right of each case) simulations.

Table 5.2: Table comparing results of minimum and maximum fiber surface pressure obtained from both COMSOL and inbuilt MATLAB FEA simulations for both cases of power law indices (i.e., $n = 0.2$, & $n = 1.0$)

	$n = 0.2$		$n = 1.0$	
	P_{min}	P_{max}	P_{min}	P_{max}
MATLAB	-2.83	1.11	-7.65	1.17
COMSOL	-3.01	1.10	-7.55	1.17

The 3D FEA formulations are direct extensions in dimensionality to the 2D FEA model and the results of the computed responses are likewise expected to agree with results obtained from COMSOL Multiphysics.

5.1.8 Validation of Jeffery's Pressure Optimization Scheme

The implementation of the optimization scheme to obtain the minimum surface pressure on the particles surface using exact derivatives of the Jeffery's pressure equation is validated by comparing outputs of optimum spatial location X_j^{opt} and fiber orientation, ϕ_{opt} where the minimum particle surface pressure, $\bar{P}_{min}$ occurs with outputs obtained from the IVP-RK4 method, ϕ_{RK4} for the different homogenous flow cases and for $\dot{\gamma}/\dot{\epsilon} = 1$ as shown in Table 5.3 below. The peak pressure magnitude on the particle's surface occurs when the particle tumbles in the shear plane. The results of the in-plane particle orientation angle at minimum pressure is seen to match closely from both numerical methods and this fiber orientation at the optimum location is seen to corresponds to the principal flow direction, ϕ_{prin} (cf. Table 5.3). Moreover, the optimum location on the particle's surface, X_j^{opt} shows that the minimum pressure occurs at the particle's tip.

Table 5.3: Results of the in-plane particle orientation angle, the location of peak minimum pressure occurrence on the particles surface and the associated minimum pressure for the different homogenous flow conditions ($\dot{\gamma}/\dot{\epsilon} = 1$).

	ϕ_{RK4}	ϕ_{opt}	ϕ_{prin}	$X_1^{opt} [mm]$	$X_2^{opt} [mm]$	$X_3^{opt} [mm]$	$\bar{P}_{min}$
SS	0.7863	0.7853	0.7854	0.0600	0.0000	0.0000	-15.7059
SUA	1.3842	1.4110	1.4099	0.0600	0.0000	0.0000	-18.1227
UA			***No minimum found				9.0701
BA			***No minimum found				-9.0701

PST	1.1776	1.1781	1.1781	0.0600	0.0000	0.0000	-16.9603
SBA	0.7977	0.7854	0.7854	0.0600	0.0000	0.0000	-13.0741
TA			***No minimum found				0.0000
STA	0.7864	0.7853	0.7854	0.0600	0.0000	0.0000	-5.9363

5.2 Results and Discussion

The Results and Discussion section is divided into two sub-sections. The first sub-section presents particles behavior (orientation dynamics and surface pressure distribution) in a Newtonian fluid, considering the various homogenous flows described above and the effect of geometric aspect ratio and particles initial orientation on the particles motion and evolution of the surface pressure. The subsequent sub-section presents in detail the effect of shear-thinning power-law fluid rheology on the particles behavior in the various combined homogenous flows and for different shear-to-extension rate ratio ($\dot{\gamma}/\dot{\epsilon} = 1$ and 10). The section also presents the results of sensitivity studies on the influence of the ellipsoidal aspect ratio and initial particle orientation on the particles behavior in non-Newtonian simple shear flow.

5.2.1 Particle Motion in Newtonian Homogenous Flows

5.2.1.1 Effect of Particle Aspect Ratio and Flow Conditions

The 2D FEA sensitivity analysis on the particle's geometric aspect ratio r_e carried out showed that the magnitude of r_e varies directly with the max and min pressures on the particle's surface as it rotates through Jeffery's orbit in simple shear flow. Figure 5.12 illustrates that the minimum pressure on the particle surface drops as the shape of the ellipsoid oblates from a prolate spheroid to a perfect sphere at which point there are no noticeable pressure peaks on the particle surface during its evolution, as expected. A closer inspection of the pressure contour plots

appearing in Figure 5.13 shows the location of minimum pressure on the particle surface and that these low-pressure sites occur at the particle tip.

The shear rate magnitude and Newtonian viscosity is observed to influence computed pressure response as that for particle aspect ratio, i.e., higher shear rate and viscosity result in a higher peak pressure at sites where they occur on the particle surface as shown in Figure 5.14 and Figure 5.15. These factors (particle aspect ratio, viscosity, and flow shear rate), however, affect Jeffery's period differently. By mere inspection of the definition of Jeffery's tumbling period (cf. eqn. (5.68)), the period is observed to vary directly with aspect ratio (i.e., implying faster tumbling for shorter particles) the reverse is the case with the shear rate magnitude which varies inversely with the period as higher shear rate results in higher particle angular velocities, as predicted by Jeffery. However, Jeffery's period is unaffected by the viscosity magnitude. In summary, higher geometric aspect ratios, shear rate magnitude and viscosity result in lower particle surface pressure drop for suspended particle in simple shear flow.

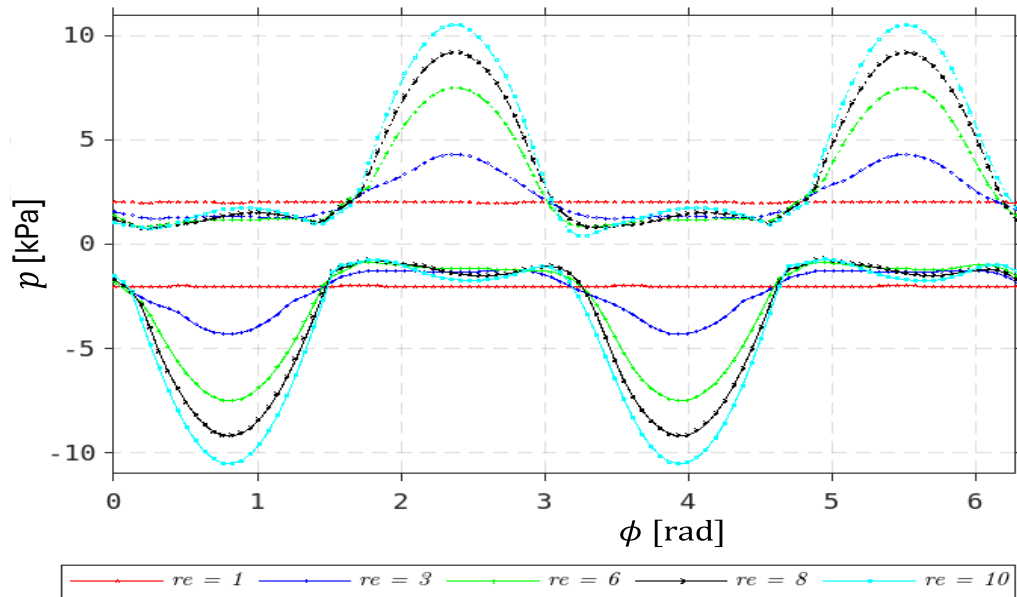


Figure 5.12: Maximum (upper curves $p > 0$) and minimum (lower curves $p < 0$) particle surface pressures for various aspect ratio in simple shear flow ($\dot{\gamma} = 1\text{s}^{-1}$).

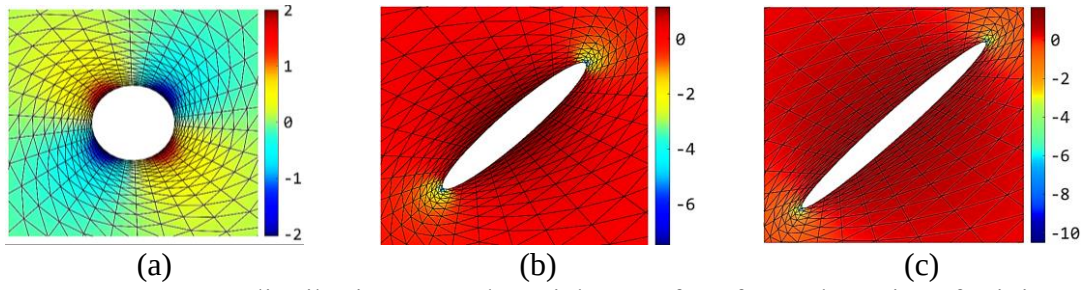


Figure 5.13: Pressure distribution around particle's surface for at the point of minimum pressure drop for different particle's aspect ratio (a) $r_e = 1$ (b) $r_e = 6$ (c) $r_e = 10$.

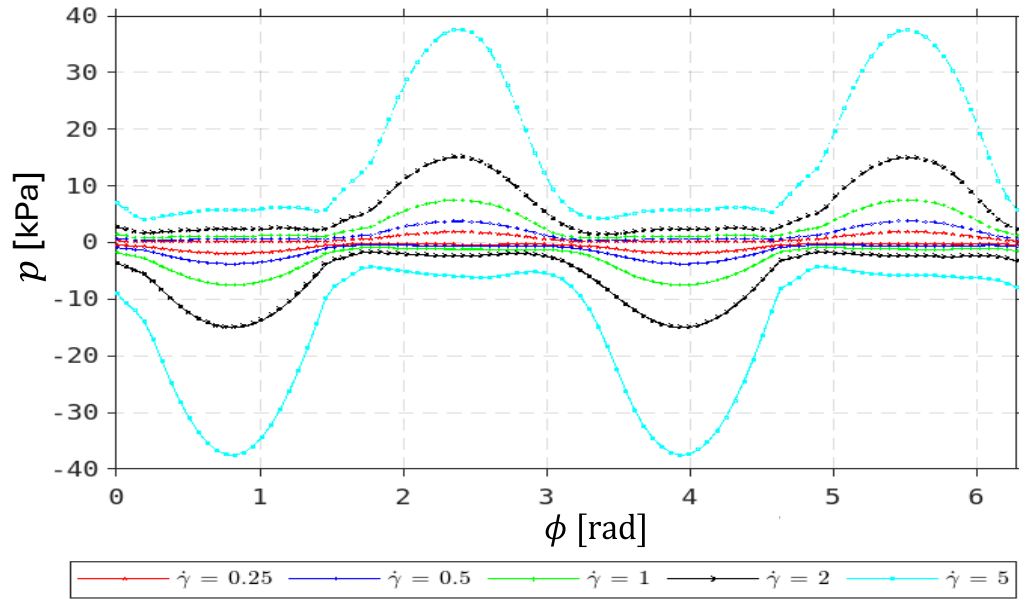


Figure 5.14: Maximum (upper curves $p > 0$) and minimum (lower curves $p < 0$) particle surface pressures for various shear rate values in simple shear flow ($r_e = 6$). The units for $\dot{\gamma}$ are s^{-1} .

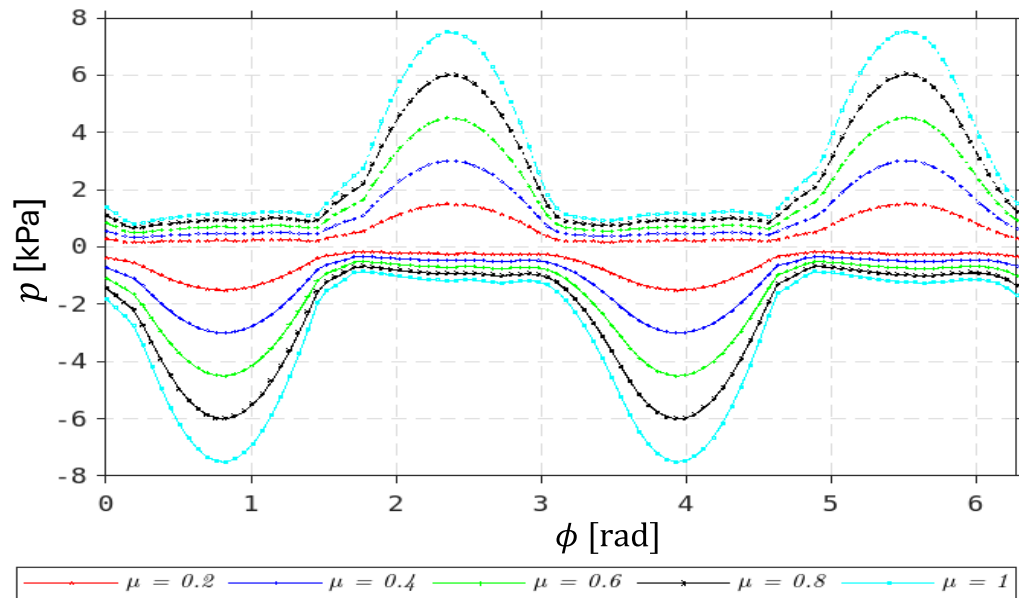


Figure 5.15: Maximum (upper curves $p > 0$) and minimum (lower curves $p < 0$) particle surface pressure limits for various Newtonian viscosities in simple shear flow ($r_e = 6$). The units for μ are $\text{Pa} \cdot \text{s}$.

From the foregoing preliminary pressure sensitivity 2D Newtonian studies, we see that the peak pressure extreme on the surface of a particle suspended in Newtonian purely viscous simple shear flow is influenced by the fluid viscosity μ_1 , the magnitude of the shear rate $\dot{\gamma}$, and the particle aspect ratio r_e . For completeness, we further explore 3D particle behavior in Newtonian purely viscous flow using Jeffery's equations. For a given aspect ratio, the net pressure $p - p_0$, computed from eqn. (5.1) is seen to have a linear dependence on the Newtonian viscosity μ_1 and shear rate $\dot{\gamma}$, i.e. $(p - p_0)/\mu_1\dot{\gamma}$ is constant. However, as r_e increases, so does the extreme tip pressure. Figure 5.8b shows that the particle's tip pressure magnitude is proportional to the r_e of the ellipsoidal particle, which is likely due to the increased particle length, the reduced particle tip curvature which occurs as r_e is increased, or both. From eqns. (5.71) & (5.72), it can be shown that the particle's tip pressure extremes occur at an orientation angle of $\phi = \pm \pi/4$ when the angular velocity $\dot{\phi} = \dot{\gamma}/2$ which also corresponds to the principal flow directions for simple shear flow. Further, at the position where the particle's precession approaches extremum at $\phi = n\pi/2, |n| \geq 0$, the particles tip pressure goes to zero irrespective of the geometric aspect ratio. Figure 5.16 shows the pressure distribution on the surface of rigid spheroidal particles at the location of orbital minimum surface pressure extreme for different aspect ratios and for particle motion in the plane of shear flow. It is evident that the minimum pressure on the particles surface occurs at the particle tips and the pressure peak magnitudes increases with the geometric aspect ratio.

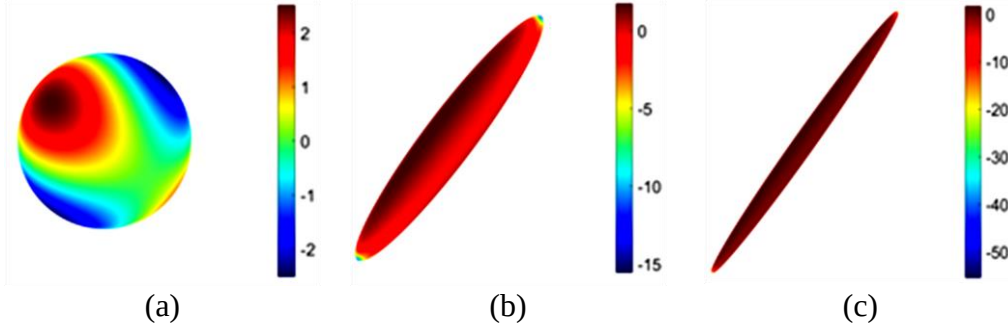


Figure 5.16: Pressure Distribution around the particle surface at the point of minimum peak pressure occurrence ($\phi = \pi/4$) for different aspect ratio (a) $r_e = 1$ (b) $r_e = 6$ (c) $r_e = 15$.

With increased ellipsoidal aspect ratio, the curvature radius at the particle's tip reduces. It is important to understand the relation of the tip pressure magnitude with the tip geometry (i.e. the curvature radius, $r_\kappa = 1/r_e$) and with the relative positioning of the tip in the constant velocity gradient flow-field (defined by the particles geometric parameter, κ). Figure 5.17a shows the relationship between the spheroidal orbital minimum tip pressure, $\bar{P}_{min,\kappa}$ normalized with respect to the spherical reference values, $\bar{P}_{min,0}$, (i.e. $\kappa = 0$) and the curvature radius for a prolate spheroid with unity minor axis length. This relationship obtained through a typical curve fitting procedure can be represented by eqn. (5.128). The Newtonian orbital minimum tip pressure ratio is seen to decrease exponentially with increasing tip curvature radius as

$$\bar{P}_{min,\kappa}/\bar{P}_{min,0} = 0.63 + 0.39r_\kappa^{-1.53} - 4.81 \exp(14.47r_\kappa) \quad (5.128)$$

Alternatively, the Newtonian orbital minimum tip pressure ratio can be represented in terms of the geometric parameter κ as shown in Figure 5.17b and can be written as

$$\bar{P}_{min,\kappa}/\bar{P}_{min,0} = 1.87\kappa + 10.74\kappa^{19.56} + 0.82 \exp(4.54\kappa^{56.62}) \quad (5.129)$$

Figure 5.17b shows that as κ tends to unity approaching a slender rod, the particle tip orbital minimum pressure goes to infinity. Note that the mean aspect ratio of short fiber fillers experimentally measured in 13% CF/ABS large scale EDAM printed bead were found to be about $r_e = 45$, $\kappa = 0.999$ [317], [318], that would theoretically yield high pressure spikes at the particle

tips in the polymer suspension during polymer composite processing based on Jeffery's model assumption, which have been suggested by Awenlimobor et al. [52] to be potentially responsible for micro-void nucleation at the fiber tips.

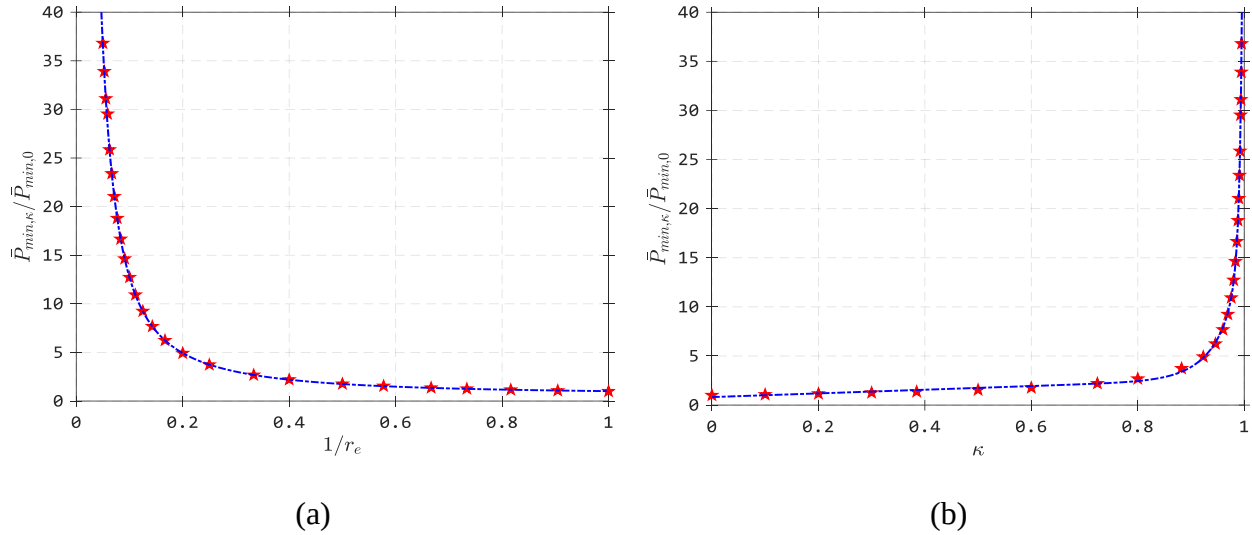
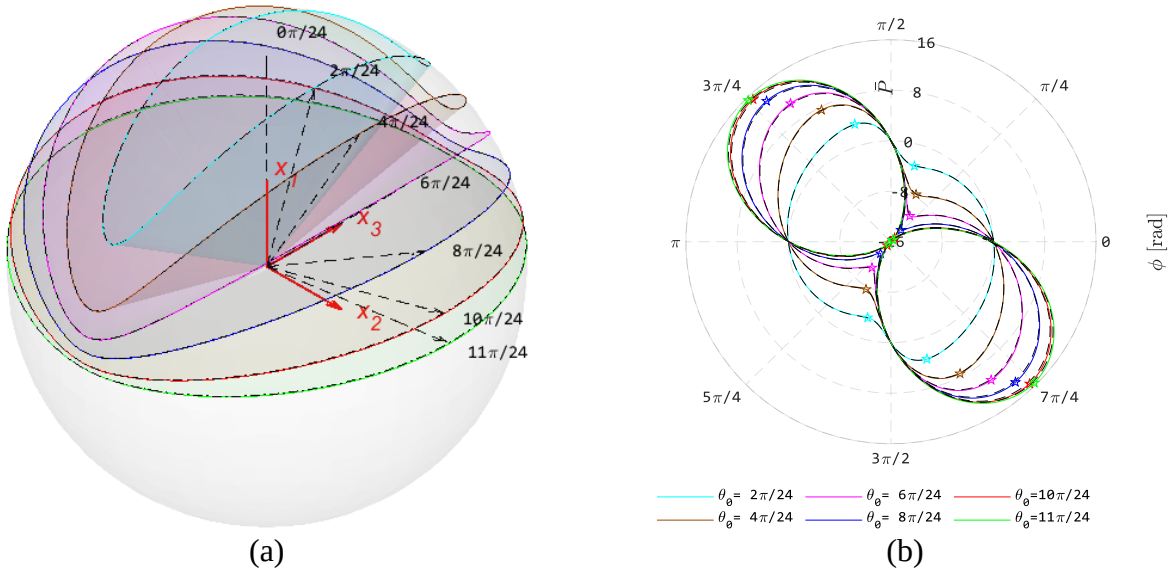


Figure 5.17: Relationship between the particle's orbital minimum pressure normalized with respect to the minimum surface pressure on a sphere in Newtonian fluid flow as a function of (a) radius of curvature (r_c), and (b) geometric parameter κ . Results are shown for particle tumbling in simple shear flow with $\mu_1 = 1 \text{ Pa} \cdot \text{s}$ and $\dot{\gamma} = 1 \text{ s}^{-1}$.

5.2.1.2 Effect of Initial Particle Orientation

In Figure 5.18a, we present the particle's motion in simple shear flow for various initial particle azimuth angle $\theta_0 = 2\pi/24 \leq \theta \leq 11\pi/24$ ($\phi_0 = 0$) based on Jeffery's solution given above. As expected, the particle's motion is periodic, and the period is same for all orbits. The orbit becomes narrower as we increase the initial out-of-plane orientation angle which reduces the effective aspect ratio (seen as that projected to the shear plane), resulting in lower peak pressure extremes. Figure 5.18b shows that the angle at which the particle pressure extreme occurs shifts as the particle is oriented further out of the shear plane. Eventually, setting the initial out of plane orientation to zero would lead to the particle spinning about its axis in a log-rolling position with near-zero surface pressure due to negligible disturbance velocity. The phase diagrams (cf. Figure 5.18c&d) reveals a symmetric behavior in particle dynamics. As the particle moves further out of

plane (i.e. $\zeta \rightarrow 0$), the location of the tip pressure extremes converges towards the location of minimum precession at $\phi = \pm \pi/2$, but as the particle moves towards the shear-plane, the pressure extreme locations coincide with the direction of the principal axis of the flow ($\phi = \pm \pi/4$). Figure 5.19 shows the particle's configuration at the location of minimum particle tip pressure along select Jeffery's orbits with various initial azimuth angle θ_0 . For the particle tumbling in the shear plane of the flow ($\theta_0 = -\pi/2$) we see that the particle's orientation coincides with the principal direction of the flow ($\phi = \pi/4$) but as it moves further out of plane, the peak pressure location moves closer towards the upper limit of azimuthal inclination for each orbit (i.e. $\phi \rightarrow \pi/2$).



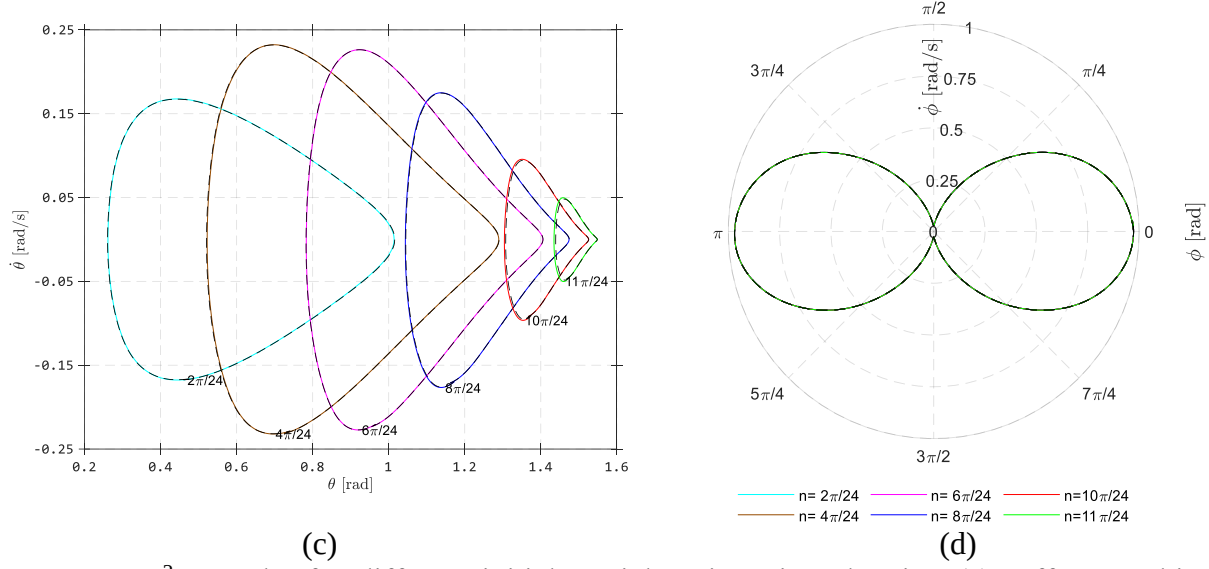


Figure 5.18²: Results for different initial particle orientation showing (a) Jeffery's orbits (b) particle tip pressure evolution where the asterisk (*) indicates location of the tip pressure extreme (c) phase diagram of azimuth angle θ vs nutation $\dot{\theta}$ (d) polar plot of the precession $\dot{\phi}$ vs polar angle ϕ . Results are shown for $-2\pi/24 \leq \theta_0 \leq -12\pi/24$ and for simple shear flow with $\mu_1 = 1 \text{ Pa} \cdot \text{s}$ and $\dot{\gamma} = 1 \text{ s}^{-1}$, $r_e = 6$.

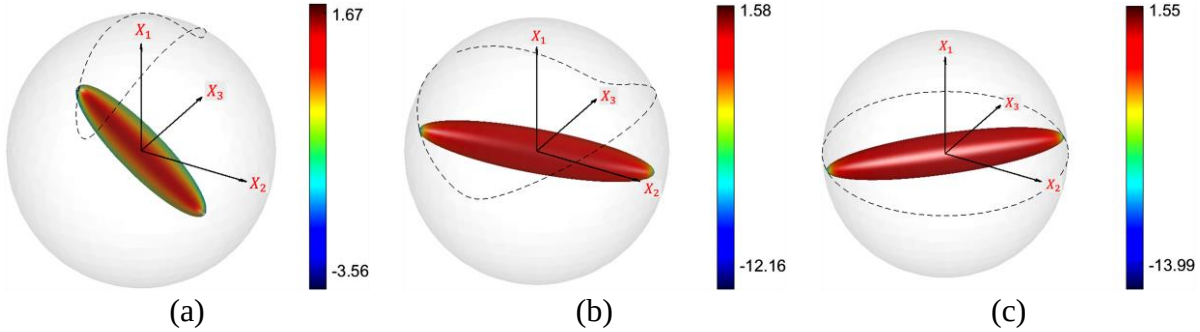


Figure 5.19: Spatial configuration of the particle at the point of minimum pressure occurrence and for various initial azimuthal angle θ_0 of (a) $\theta_0 = 2\pi/24$, (a) $\theta_0 = 8\pi/24$, (a) $\theta_0 = 12\pi/24$. Results are shown for $\mu_1 = 1 \text{ Pa} \cdot \text{s}$ and $\dot{\gamma} = 1 \text{ s}^{-1}$, $r_e = 6$.

Figure 5.20a shows a nearly linear relationship between the particle's orbital minimum tip pressure and the polar angle location along the corresponding Jeffery's orbits. As noted above, when the particle is tumbling in shear plane (i.e., $\zeta = +\infty$), the location of the particle's surface extreme pressure coincides with the ellipsoidal tip location. However, as the particle becomes

² The results presented in Figure 5.18a-d are also validated with both FEA and Jeffery's analytical calculation. The black dashed lines are results obtained from Jeffery's equation and the continuous colored lines are results from FEA computations.

oriented more out-of-plane (i.e. $\zeta \rightarrow 0$), the location of minimum pressure on the particle surface at the orientation of peak pressure occurrence is slightly shifted away from the tip down the leeward side trailing the flow. Figure 5.20b shows the difference between the minimum pressure on the fibers surface and tip pressure ($\delta\bar{P}$) at the instant when the peak occurs along Jeffery's orbit. The result shows that a higher initial out of plane orientation leads to greater deviation of the fiber tip pressure from its surface pressure extreme magnitude.

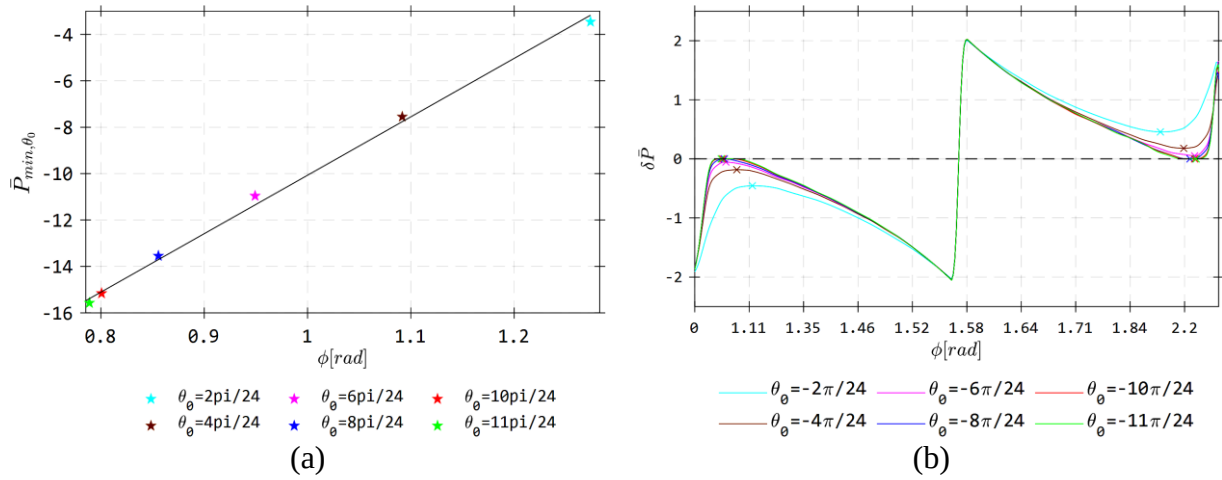


Figure 5.20: Tip pressure results (a) Orbital minimum particle tip pressure versus polar angle, and (b) difference in the instantaneous particle tip pressure and actual surface pressure extremum, for different Jeffery's orbit and for $\mu_1 = 1 \text{ Pa} \cdot \text{s}$ and $\dot{\gamma} = 1 \text{ s}^{-1}$, $r_e = 6$.

The particle orbital maximum nutation $\dot{\theta}$ itself peaks at a Jeffery's orbit that passes through $(\phi, \theta = \pm \pi/4)$ irrespective of the aspect ratio. In Figure 5.21a, the continuous lines trace the paths of orbital maximum nutation across the degenerate spectrum of Jeffery's orbit for different aspect ratios, and the dashed lines are the Jeffery's orbit that cuts across the location of peak nutation for different ellipsoidal aspect ratios. From Figure 5.21b, the peak nutation across the spectrum of Jeffery's orbit is observed to increase with the aspect ratio and approaches the critical value at $\dot{\theta} = \dot{\gamma}/4$.

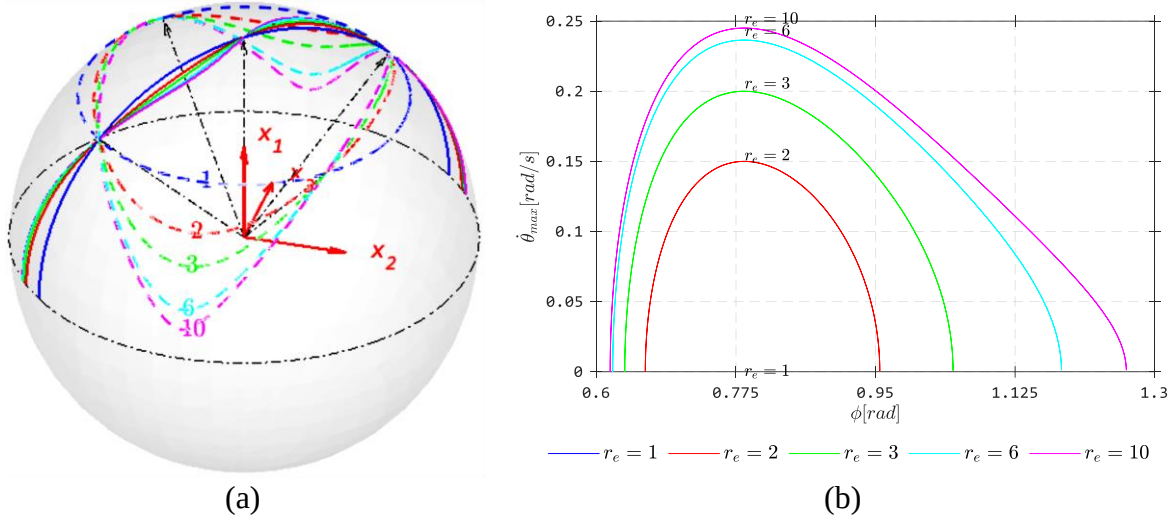


Figure 5.21: Out-of-plane Jeffery orbits (a) the path of orbital maximum nutation across degenerate spectrum of Jeffery's orbit for aspect ratios of 1, 2, 3, 6, and 10 (continuous lines) and critical Jeffery's orbit at which the orbital maximum nutation attains peak magnitude for the same aspect ratios (dashed lines). (b) phase plot of the orbital maximum nutation across degenerate spectrum of Jeffery's orbit for different aspect ratios.

5.2.1.3 Effect of Particle Shape

As we earlier stated, a drawback of Jeffery's equation is its inability to model arbitrary shaped particles with different end conditions which can be accounted for in our FEA simulation. Most chopped fibers used to reinforce polymer composites are cylindrical shaped with different end conditions. Cylindrical shaped particle allows us to study the decoupled effect of edge curvature radius and aspect ratio on the particle surface pressure response. We present results of the cylindrical particle's responses (cf. Figure 5.22) for different end curvature radius ranging from $0.05 \leq \bar{r}_\kappa \leq 0.5$ for a cylinder of diameter $\mathcal{H}_1 = 1.0$, individually calibrated to be hydrodynamically equivalent to an ellipsoid with an aspect ratio $r_e = 6$. The fibers orientation angle, rotational velocity, and surface pressure were computed using flow parameters of $\mu_1 = 1 \text{ Pa.s}$, $\dot{\gamma} = 1 \text{ s}^{-1}$. Figure 5.22a shows that the evolution of the cylinders' angular velocity for the different edge curvature cases with different geometric aspect ratios (colored lines) are perfectly superposed on the angular velocity profile of the ellipsoid with $r_e = 6$ (black dotted line).

The corresponding cylindrical geometric aspect ratio, r_c for the different end cases is seen to vary inversely with the edge curvature radius, $\bar{r}_\kappa$ (cf. Figure 5.22c). For an equivalent ellipsoidal aspect ratio of $r_e = 6$, the cylindrical geometric aspect ratio r_c was approximated as a cubic function of the tip curvature $\bar{r}_\kappa$ according to

$$r_c = 7.806 - 1.282 \bar{r}_\kappa - 1.463 \bar{r}_\kappa^2 + 3.859 \bar{r}_\kappa^3 \quad (5.130)$$

Expectedly, the pressure extremes on the particle's surface are observed to increase with decreasing edge curvature radius (cf. Figure 5.22b). The minimum surface pressure is observed to drop in magnitude from a value of $\bar{P}_{min} = -19.41$ when $\bar{r}_\kappa = 0.05$ to about $\bar{P}_{min} = -8.68$ when $\bar{r}_\kappa = 0.5$. Recall that for the ellipsoidal particle of dynamically equivalent aspect ratio, the orbital minimum pressure occurs at a value of $\bar{P}_{min} = -15.71$. It follows that unlike the ellipsoidal particle, the cylindrical hydrodynamic equivalent could have higher or lower pressure magnitudes at the ends depending on the edge curvature radius. As the particle tumbles in and out of alignment with the principal direction of the flow, the minimum and maximum pressure cycles through mesh-points along the surface of the particle in the plane of the shear flow. Figure 5.22d shows the pressure distribution on the cylinder particle surface at the instant of orbital minimum surface pressure for different edge curvatures. The instantaneous pressure extremes occur at the terminations of the curved section at the cylinder's end in comparison to the ellipsoid where the instantaneous pressure extremes occur at its vertices.

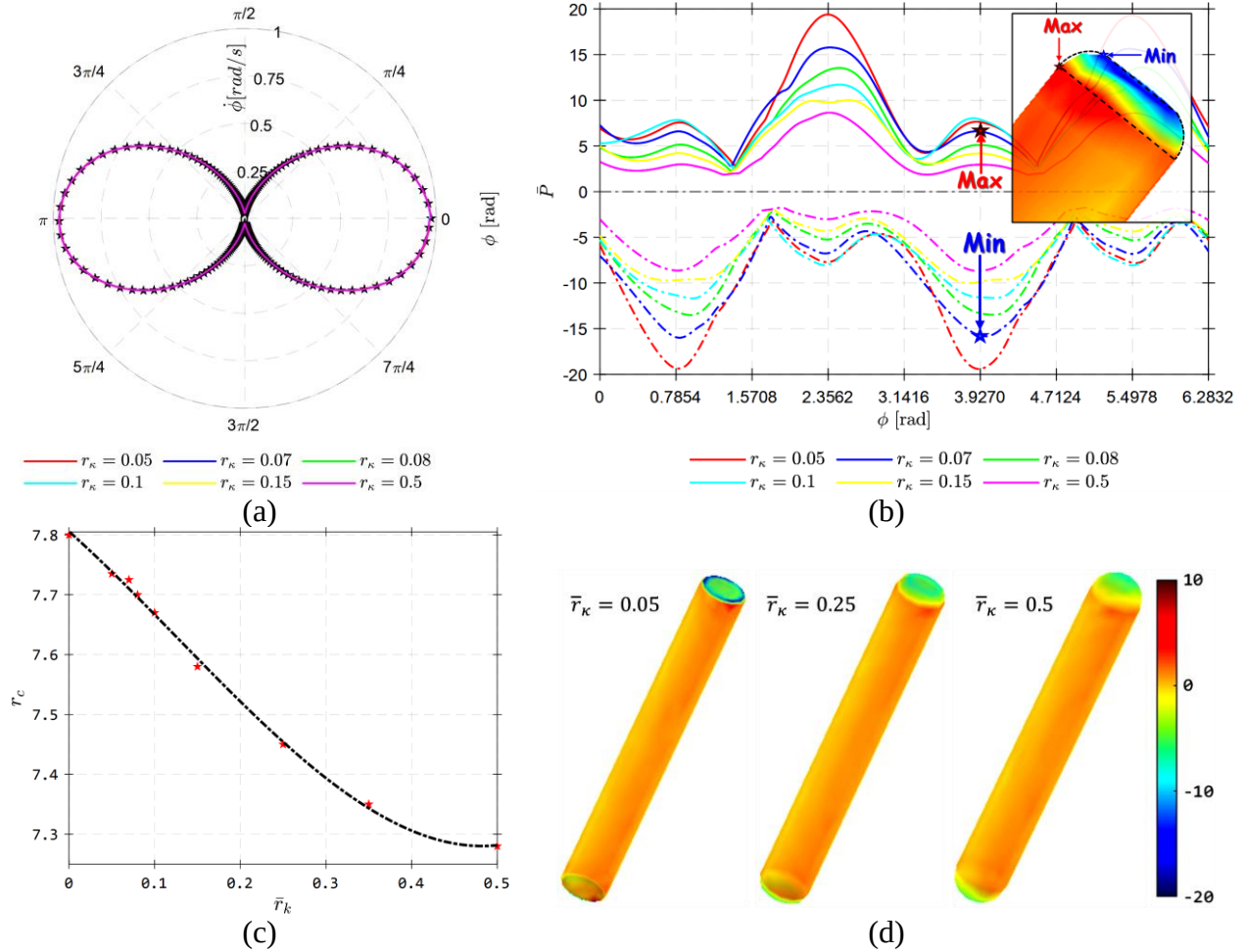


Figure 5.22: Evolution of (a) the angular velocity (b) pressure extremes; on the surface of the cylindrical particle for different curvature radius $0.05 \leq \bar{r}_\kappa \leq 0.5$. (c) Relation between the cylindrical geometric aspect ratio and edge curvature radius, and (d) Pressure distribution on the surface of the particle at the instant of orbital minimum surface pressure occurrence for different cylinder edge curvatures. Results are shown for cylinders with hydrodynamic equivalent ellipsoidal aspect ratio of $r_e = 6$ tumbling in simple shear flow ($\mu_1 = 1 \text{ Pa.s}$, $\dot{\gamma} = 1 \text{ s}^{-1}$).

Although the aspect ratio of the cylinder is slightly adjusted for each end curvature cases to hydrodynamically match the aspect ratio of the reference ellipsoid aspect ratio, we argue that the observed change in the pressure extreme magnitude is mainly a result of the change in the edge curvature radius rather than the aspect ratio. To validate this, we perturb the geometric aspect ratio of the cylinder by about 6 % (i.e. $7.0 \leq r_c \leq 7.4$) for a constant edge curvature, $\bar{r}_\kappa \leq 0.5$ (hemispherical end case) similar to the range of adjustment in aspect ratio obtained for different curvature cases in Figure 5.22b. For objectivity, we adjust only the length of the straight section

of the cylinder to ensure the curvature and mesh integrity of the curved section is unaffected which is where we expect pressure extreme would occur. Figure 5.23a&b shows that the angular velocity and pressure extremes on the cylinder surface are not significantly affected by the perturbation in the aspect ratio. This is because the flow-field is symmetric and of a constant velocity gradient, and the perturbation in cylinder aspect ratio only slightly and linearly perturbs the disturbance velocity $\delta\dot{X}_i^d$ on the surface of the particle and the corresponding pressure field such that $\delta\dot{X}_i^d = \mathbb{E}_{ik}\delta X_k$ where $\mathbb{E}_{ik} = \epsilon_{ijk}\dot{\Psi}_j - L_{ik}$ is constant.

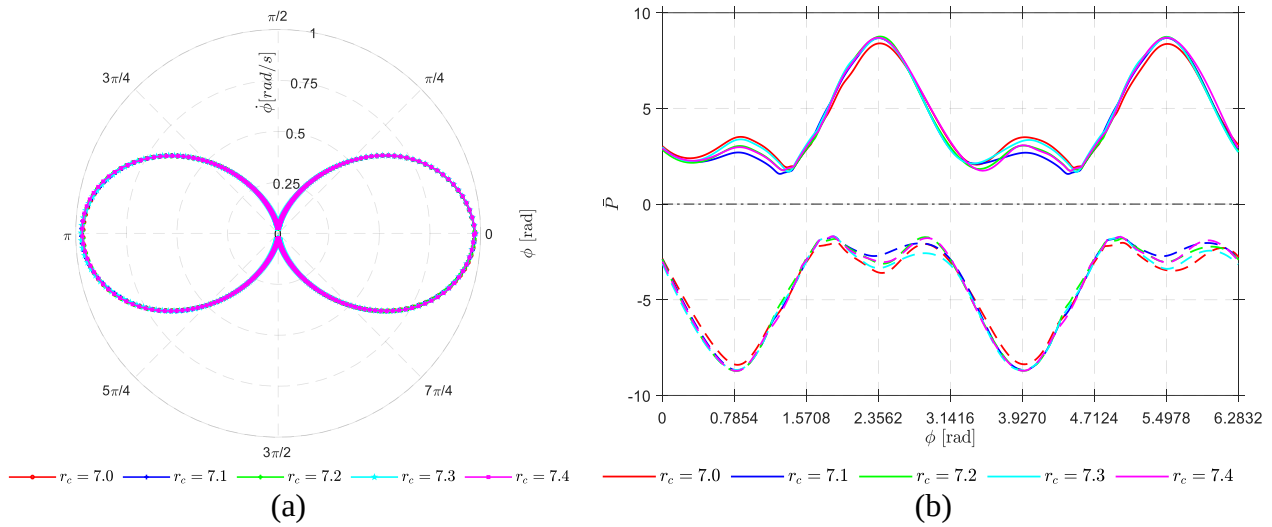


Figure 5.23: Evolution of (a) the angular velocity (b) pressure extremes; on the surface of the cylindrical particle for cylinder with different geometric aspect ratio $7.0 \leq r_c \leq 7.4$ and with constant end curvature radius, $\bar{r}_\kappa \leq 0.5$. Results are shown for cylindrical particles tumbling in simple shear flow ($\mu_1 = 1 \text{ Pa.s}$, $\dot{\gamma} = 1 \text{ s}^{-1}$).

5.2.1.4 Effect of Flow-type & Elongational ratio

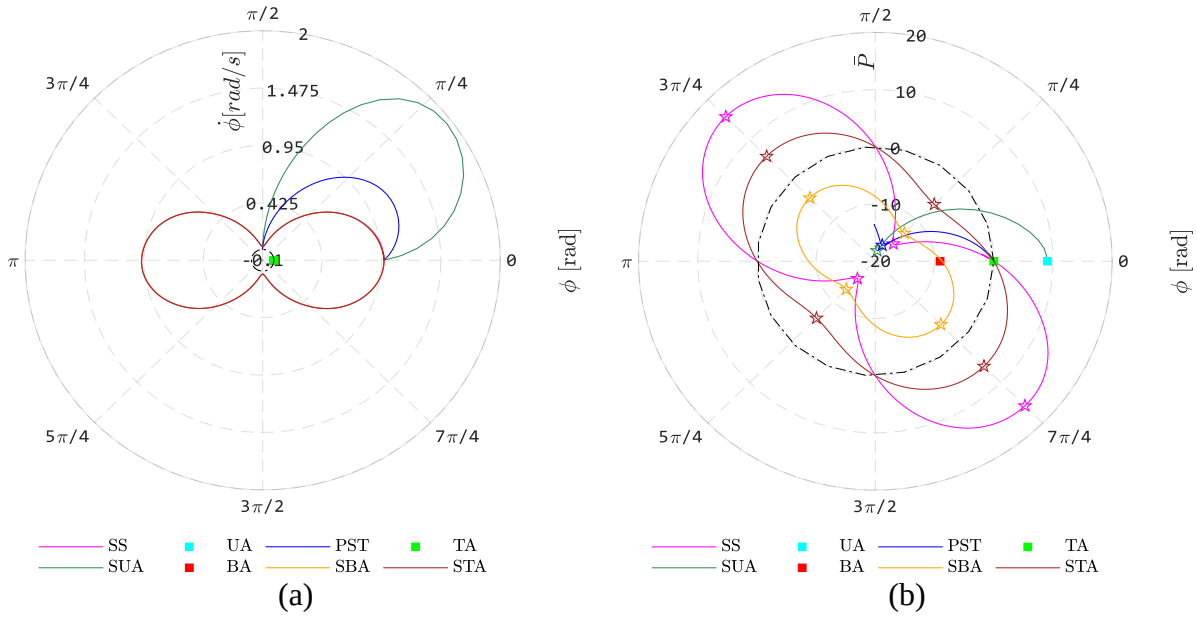
5.2.1.4.1 Steady Homogenous Flows

For the investigation of the behaviour of single rigid spheroidal particle suspended in Newtonian homogenous flows, Jeffery's equations are sufficient and computationally more efficient than our numerical solutions. The basic homogenous flows discussed in the methodology

section above that consider various combinations of stretching and shearing rate are expected in polymer composite melt flow applications such as material extrusion/deposition additive manufacturing (see e.g., Awenlimobor et al. [52]). In all Newtonian flow analyses considered here, we employ an aspect ratio of $r_e = 6$, a viscosity of $\mu_1 = 1 \text{ Pa} \cdot \text{s}$ and a shear rate of $\dot{\gamma} = 1 \text{ s}^{-1}$ where applicable. The particle is initially oriented in the $\underline{X}_2$ -direction (i.e. $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$) and rotates in the $\underline{X}_2 - \underline{X}_3$ shear plane.

Figure 5.24 shows the calculated particle in-plane angular velocity (ϕ) and particle tip pressure ($\bar{P}$) in the various homogenous flows for two cases of shear-to-extension rate ratio ($\dot{\gamma}/\dot{\epsilon}$) where applicable. Here we use the overbar to indicate a dimensionless pressure as in eqns. 75 and 76. In the planar extensional flows (i.e. UA, BA, & TA flows), we observe an absence of particle motion, however, the particle begins to rotate with the introduction of a non-zero shear velocity gradient component (cf. Figure 5.24a). In the extension-shear SUA flow (i.e., $\dot{\gamma}/\dot{\epsilon} = 1$), the particle is initially accelerated by the combined action of the inward flow in the $\underline{X}_2$ -direction and the shear flow in the $\underline{X}_2 - \underline{X}_3$ plane. The particle eventually stalls at $\phi_s = 1.58 \text{ rad}$ as it aligns with the $\underline{X}_3$ -direction due to the applied stretching and relatively low shear rate. In the PST flow case, there is no flow in the $\underline{X}_2$ -direction that influences the initial particle motion, however the inflow in the $\underline{X}_1$ -direction keeps the particle motion in the $\underline{X}_2 - \underline{X}_3$ shear plane. Like the SUA flow case, the applied stretching and relatively high extensional dominance causes the particle to stall at $\phi_s = 1.60 \text{ rad}$ as it turns to align in the $\underline{X}_3$ -direction. The SUA and PST mixed mode flow types are asymmetric in the $\underline{X}_2 - \underline{X}_3$ plane. In the SBA flow regime, the inward flow in the $\underline{X}_1$ -direction prevents out-of-plane motion of the particle, and there is no provision for preferential orientation in the $\underline{X}_2 - \underline{X}_3$ plane due to uniform stretching in the $\underline{X}_2 - \underline{X}_3$ -shear plane. As a result, the particle tumbles continuously. The STA and SS flow types are essentially similar in terms of their influence

on the particle's behavior. The only difference observed between these flow types is in the calculated particle tip pressure. At the onset of particle motion at $\phi^0 = 0$ the net pressure at the particle tip is zero ($\bar{P} = 0$) for cases with no net flow in the $\underline{X}_2$ -direction. However, the particle tip has a net positive pressure ($\bar{P} = +31.4$) for the UA/SUA flows due to the inflow in the $\underline{X}_2$ -direction, and the outflow in the $\underline{X}_2$ -direction creates a net negative pressure on the particles tip ($\bar{P} = -31.4$) for the BA/SBA cases. As the shear flow induces particle rotation, the tip pressure drops gradually until it reaches a minimum, at which point the particles orientation coincides with a principal flow direction (cf. Figure 5.24b).



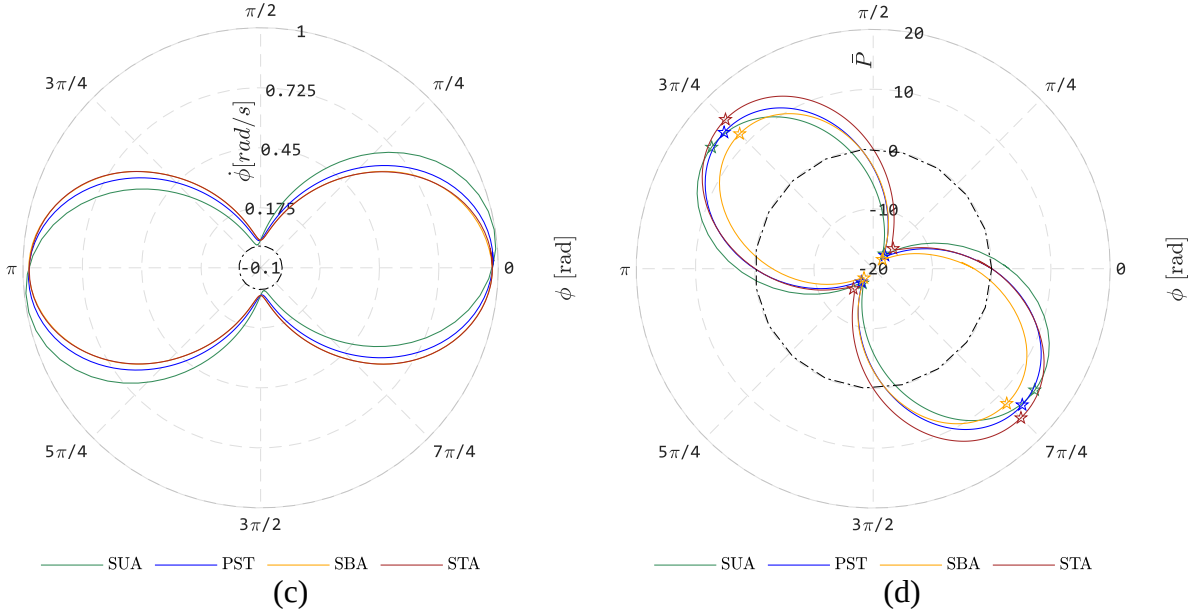


Figure 5.24: Polar plot of the evolution of the particle's (a) precession $\dot{\gamma}/\dot{\epsilon} = 1$ (where applicable) (b) tip pressure $\dot{\gamma}/\dot{\epsilon} = 1$ (c) precession $\dot{\gamma}/\dot{\epsilon} = 10$ (d) tip pressure $\dot{\gamma}/\dot{\epsilon} = 10$ for particle in the various homogenous flow types. In all cases, $\dot{\gamma} = 1 \text{ s}^{-1}$, $\mu_1 = 1 \text{ Pa} \cdot \text{s}$.

In the event where the particle does not stall, the pressure on the particle tip fluctuates between its minimum and maximum limits at locations where its orientations coincide with the principal flow directions. For the axisymmetric flows, the particle tip pressure extremes occur at $\phi = \pm \pi/4$, while for the SUA asymmetric flow (i.e., $\dot{\gamma}/\dot{\epsilon} = 1$), this occurs at $\phi = +1.41 \text{ rad}$. Alternatively, for the PST asymmetric flow, the pressure extreme occurs at $\phi = +1.18 \text{ rad}$. Cessation of the particles motion under the combined SUA and PST flow conditions is lifted once the conditions of eqn. (5.81) are violated, i.e. when $\dot{\gamma}/\dot{\epsilon} \geq 3\kappa/\sqrt{1-\kappa^2}$ for the SUA flow condition and $\dot{\gamma}/\dot{\epsilon} \geq \kappa/\sqrt{1-\kappa^2}$ for the PST flow conditions. In the current study where we assumed $\kappa = .9459$, the particle does not stall when $\dot{\gamma}/\dot{\epsilon} \geq 8.75$ for SUA flow condition and when $\dot{\gamma}/\dot{\epsilon} \geq 2.92$ for the PST flow condition. With increased shear strain rate (i.e., for $\dot{\gamma}/\dot{\epsilon} = 10$), the particle rotates periodically for all combined flow conditions (cf. Figure 5.24c). Since $\dot{\epsilon}_2 = \dot{\epsilon}_3 = \dot{\epsilon}$, for the axisymmetric combined flow cases, the particle does not stall regardless of the magnitude of $\dot{\gamma}/\dot{\epsilon}$. One exception is seen for ellipsoidal particles with small but finite thickness such as in the

case of a thin rod when $\kappa \rightarrow 1$ or in the case of a circular disc when $\kappa \rightarrow 0$, both of which are degenerate cases as described by Jeffery [19]. As the shear rate increases, the asymmetric flows become more symmetrical and the particle's surface pressure magnitudes are increased (cf. Figure 5.24d). Additionally, increased shear rate also moves the orientation where tip pressure extremes occur (i.e. at the point where it coincides with the principal flow directions). For example, in the SUA flow case, the orientation where pressure extremes occurs are at $\phi = -0.640, +0.931$ rad while the same occurs at $\phi = -0.736, +0.835$ rad for the PST flow case. Figure 5.25a&b shows that the particles orbital minimum surface pressure and corresponding orientation approaches a stable equilibrium value with increasing shear rates.

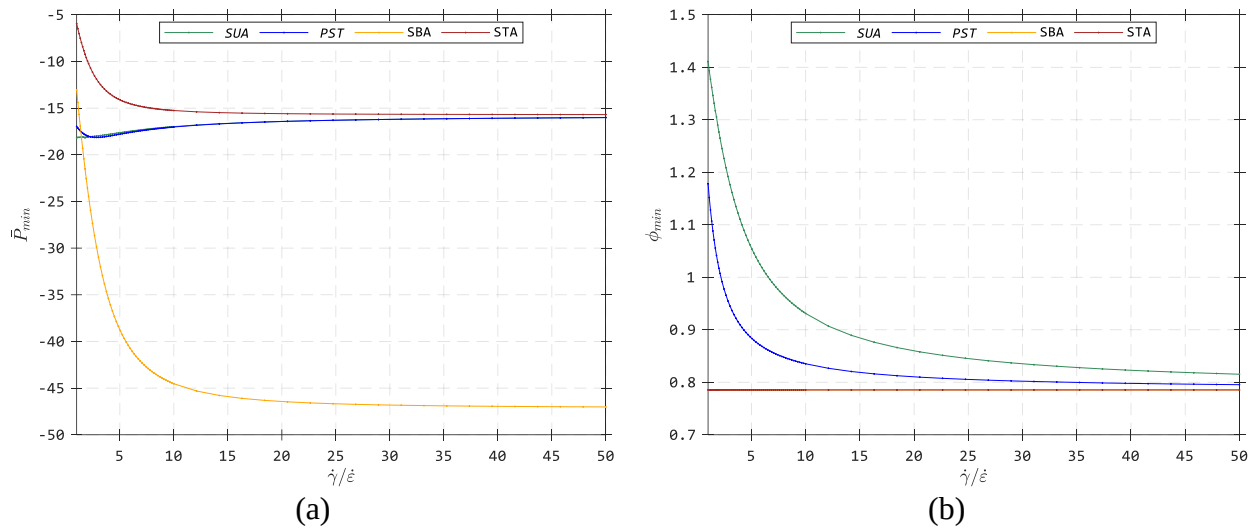


Figure 5.25: (a) In-plane particle orientation angle at the instant of peak minimum pressure occurrence on the particles surface and (b) corresponding minimum pressure, for different shear dominance factor $\dot{\gamma}/\dot{\epsilon}$ and for the combined homogenous flow conditions.

Particle motion analyses show that cessation of the rotation depends on the value of $\dot{\gamma}/\dot{\epsilon}$, i.e. for the SUA and PST flows as shown in Figure 5.26. The tumbling period is seen to asymptote from either direction to the orientation where conditions for the onset of particle stall is satisfied which is seen to occur at a limit stall angle of approximately $\phi_p = 1.72$ rad. To the left of the red-dashed vertical limit lines in Figure 5.26a, or beneath the red-dashed horizontal line in Figure

5.26b, defining the asymptote events, the particle would stall, however the reverse situation is expected beyond these limits.

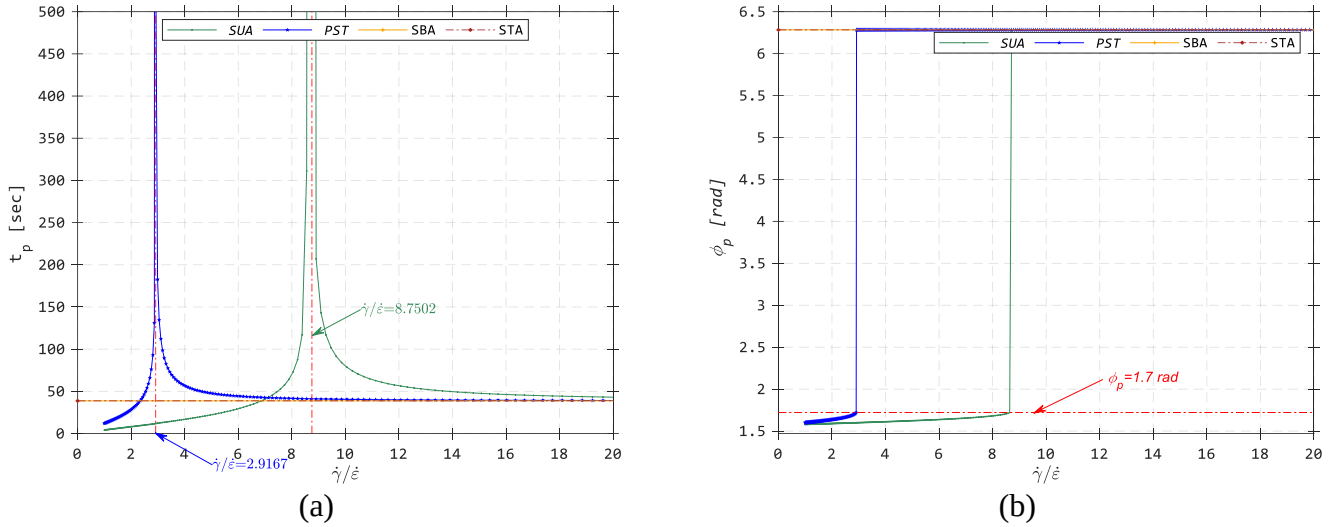


Figure 5.26: Particle motion analysis (a) particle tumbling period (or stall time where applicable) and (b) corresponding particle rotation angle, for different shear dominance factor $\dot{\gamma}/\dot{\epsilon}$ and for the combined homogenous flow conditions.

5.2.1.4.2 Unsteady Center Gated Disk Flow

The results of the pressure field around the fiber surface for this flow type are applicable to understanding the micro-void formations in injection molding of polymer composite materials. The flow conditions necessitate a negative pressure gradient along the radial flow direction. The flow characteristics involve a combination of spatially varying shear and planar elongation deformation rates. Closer to the plates, the flow is dominated by shear while regions closer to the centerline of the axisymmetric flow, are dominated by extensional flow conditions and the transition zones involves a combination of both shear and extensional velocity gradient driven flow [142]. The shear dominance increases with layer height. Numerical and experimental studies have shown that within a thin inner layer lining the wall, fibers are randomly oriented in the flow plane, and within thicker outer shells but close to the wall, the fibers are mostly aligned with the flow direction in the shear plane. In the core regions near the centerline where extensional flow is

dominant, the fibers are mostly aligned in the direction perpendicular to the shear plane and the fiber orientation distribution in the transition regions are indeterminate [142], [319]. The calculations here are performed based on a half gap height (half disk spacing) h of 1.5mm and fiber dimensions of $60\mu\text{m}$ major axis length and $10\mu\text{m}$ minor axis radius for the ellipsoid. An inlet radius of $r_1 = h$ and outlet radius of $r_2 = 30h$ is assumed.

The results of the minimum pressure on the fibers surface over the possible range of fiber orientation configuration described by the surface of the unit sphere at the flow outlet and at layer heights $X_{0_3} = 0., .5, 1$ and the associated fiber orientation where the peak occurs which has been obtained from the optimization analysis are compared to results of the minimum pressure obtained by multidimensional grid analysis based on a discretization of 100 elements in the radial axis X_{0_r} , 96 elements along the longitudinal axis ϕ and 48 elements along the latitudinal axis θ and computed at the fibers tip where it has been pre-determined to occur from the optimization analysis at the different layer heights. Figure 5.27 also shows that the direction at which the fiber orientation at peak pressure magnitude aligns with one of the principal directions of the flow at the associated spatial position. The pressure magnitudes are highest at the wall lining and the centerline of the flow (about -10.36), and lower at intermediate regions (about -9.63 at $X_{0_3} = .25$). Figure 5.28a-c shows the evolution of the fiber orientation for a fiber with initial random orientation state at different layer heights including the flow centerline $X_{0_3} = 0.$, the inner wall lining of the disk $X_{0_3} = 1.$, and at an intermediate region $X_{0_3} = .5$. The results show that at the centerline characterized by stretch dominant flow (cf. Figure 5.28a) the fiber abruptly reorients almost parallel to the transverse flow direction and stalls while at intermediate region with mixed stretching and shearing flow, the fibers gradually re-orient in the flow plane more favorably to the flow direction. At the walls, where shear is dominant, the fiber gradually but continuously

tends to align with the flow direction (cf. Figure 5.28c) in line with the conclusion of Ferec et al. [1]. From the evolution of the fibers tip pressure magnitude in Figure 5.28d, at distance away from the flow inlet, the pressure magnitude increases from the walls to the centerline of the flow. The fibers trajectories are relatively longer at higher vertical distance from the centerline.

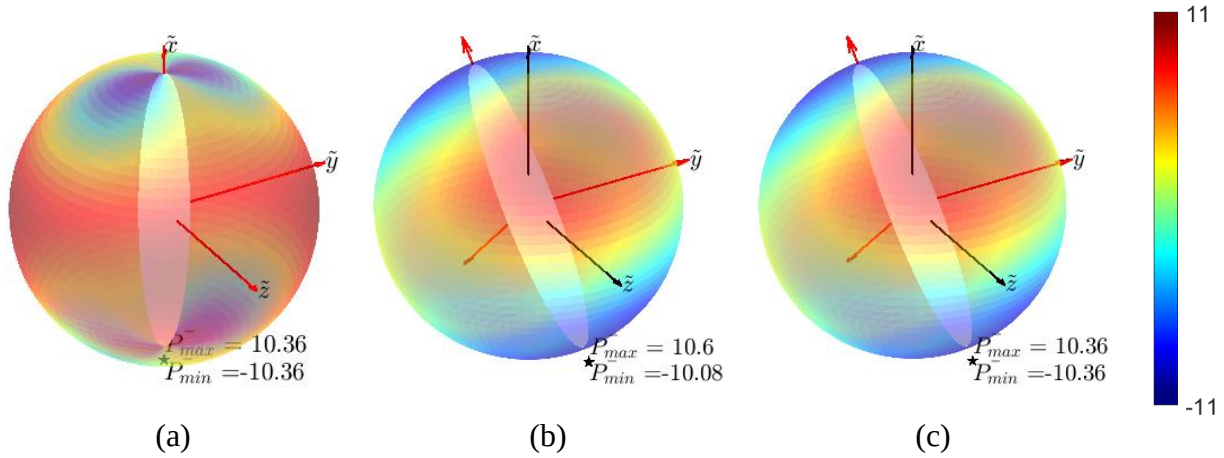
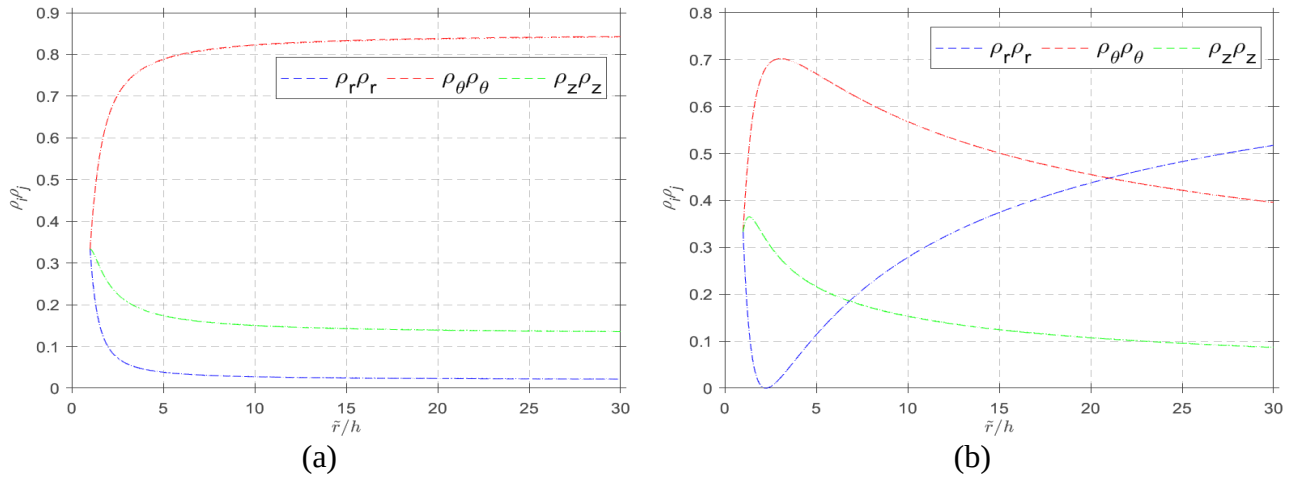


Figure 5.27: Distribution of the fiber surface “dimensionless” pressure over all possible orientation at the instant of peak minimum pressure occurrence on the fiber surface at the flow outlet at $X_{0_r} = .25$ and for different layer heights (a) $X_{0_3} = 0$. (b) $X_{0_3} = .5$ and (c) $X_{0_3} = 1$.



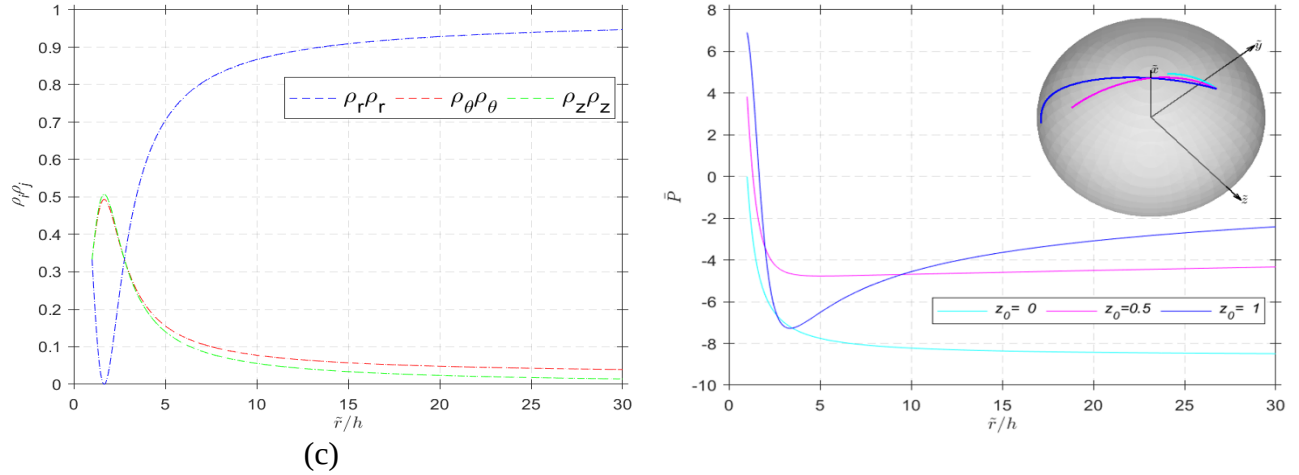


Figure 5.28: Evolution of the fiber orientation component at different layer heights (a) $X_{0_3} = 0$. (b) $X_{0_3} = .5$ and (c) $X_{0_3} = 1$ for a fiber with initial random orientation at $r_0 = h$ (d) Evolution of the fibers tip pressure at the different layer heights (i.e. $X_{0_3} = 0, .5, 1$.)

Figure 5.29a shows the distribution of the minimum fiber tip pressure over all possible direction at various layer heights. The values are seen to be relatively less severe than those observed in the shear dominant homogenous flow discussed in earlier sections. The corresponding instantaneous directions of peak minimum fiber tip pressure for the various layer heights occurs in the shear plane and lies between the flow axis direction and an azimuthal inclination of $\theta_0 = -\pi/4$ which are the in-plane principal directions for pure elongation and pure shear respectively. The deviation of the instantaneous fiber orientation vector from the corresponding instantaneous direction of minimum fiber tip pressure would be the magnitude of the extreme pressure at the fiber tips. For the case of a fiber initially orientated randomly at the flow inlet, Figure 5.29b shows the cosine of angle between the instantaneous fiber orientation and the corresponding unit direction of instantaneous minimum fiber tip pressure for varying layer heights. The result shows minimal deviation angle at the flow centerline and gradually increasing deviation angle with increasing layer height and radial distances.

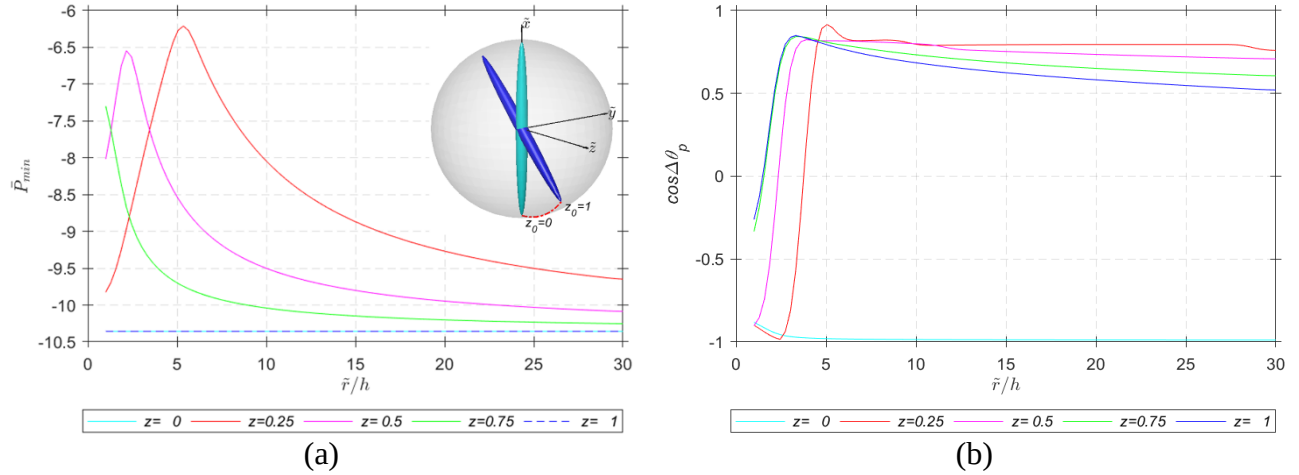


Figure 5.29: (a) Distribution of the instantaneous minimum fiber tip pressure over all possible fiber configuration (b) Evolution of the cosine of angle between the fiber orientation vector and direction of peak minimum pressure for a fiber with initial random orientation.

5.2.2 Particle Motion in Non-Newtonian Homogenous Flows

The results presented above focused on a single rigid ellipsoidal particle in various combined extensional and shear Newtonian homogenous flows that are considered typical of those in an EDAM nozzle during polymer composite processing. It is well understood, however, that thermoplastic polymer materials are inherently non-Newtonian. Moreover, the addition of filler reinforcements to polymers are known to increase the melt viscosity and the shear-thinning fluid behavior in the nozzle. Additionally, high shear regions of complex flows such as the lubrication zone near the screw edge or regions of flow acceleration near the nozzle are known to result in flow segregation of highly shear-thinning polymer melt suspension into resin lean highly viscous domains and resin rich low-viscosity domains. As such understanding the particle behavior in shear-thinning fluid within various flow regimes is important in understanding microstructural development within polymer composite beads. The sections to follow present results obtained with the nonlinear FEA modeling approach presented above which considers a non-Newtonian shear-thinning power-law fluid rheology.

5.2.2.1 Effect of Flow-type & Elongational ratio

In this section, we consider the response of a single 3D ellipsoidal particle in simple homogeneous power-law fluid flows computed using the FEA method described above. The results presented in Figure 15 are for an ellipsoid with geometric ratio $r_e = 6$ rotating in a power-law fluid with a flow shear rate of $\dot{\gamma} = 1 \text{ s}^{-1}$ and power-law indices ranging from 0.2 to 1.0.

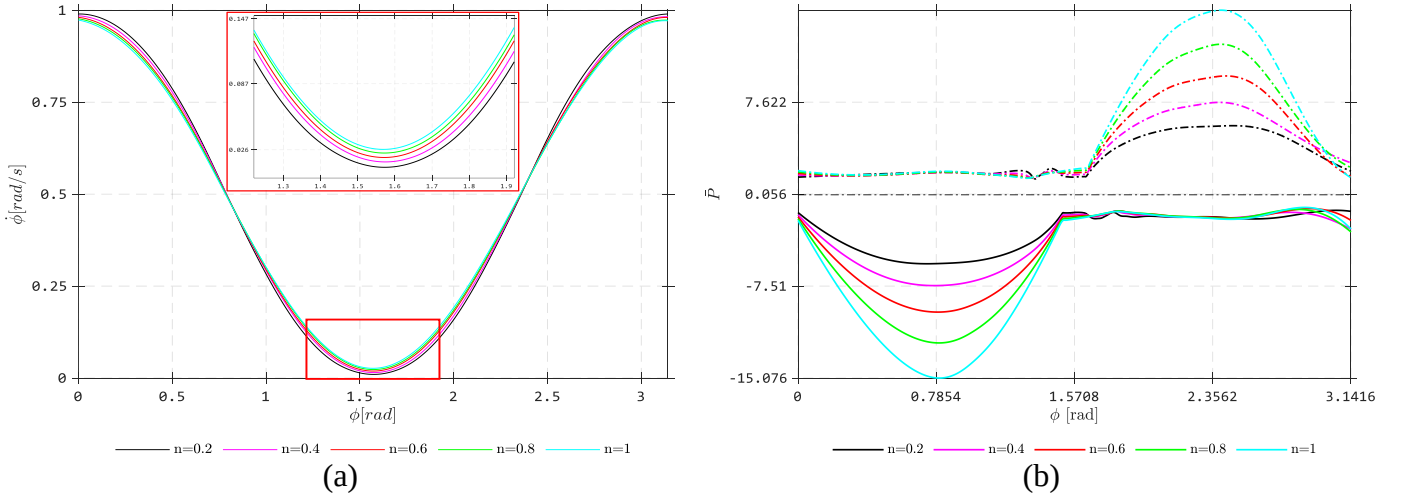


Figure 5.30: FEA computed shear-thinning response of (a) particle polar angle ϕ vs precession $\dot{\phi}$ and (b) surface pressure extremes for particle motion in simple shear flow. Results are shown, for $r_e = 6$, $0.2 \leq n \leq 0.8$, $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$.

Figure 5.30a shows that the shear-thinning behavior has a slight influence on the particle's dynamic motion as reduction in the power-law index slows down the particle. The limits of the particle's in-plane angular velocity are observed to increase with increasing power-law index. Further, Figure 5.30b shows that the particle surface pressure extremes increase with decreased shear-thinning. Additionally, it is interesting to note that even though the orbit formed from particle tumbling in the shear-plane appears to exhibit little noticeable difference due to shear-thinning, Figure 5.31a shows that the tumbling period significantly increases with increasing shear-thinning.

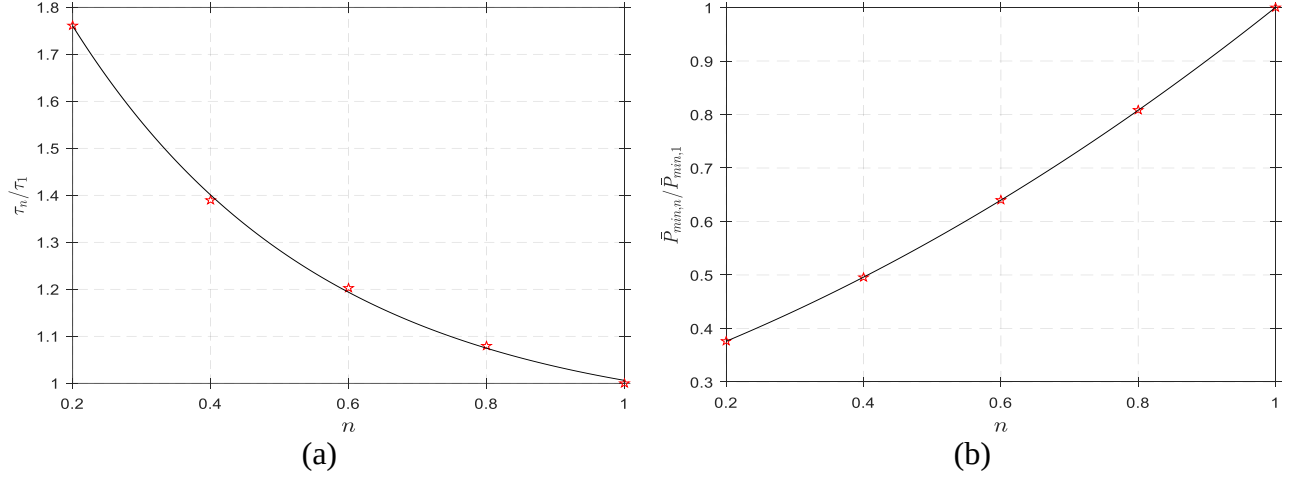


Figure 5.31: Non-Newtonian to Newtonian ratio of the (a) particle's in-plane tumbling period (b) Orbital minimum particle tip pressure. Results are shown for $r_e = 6$, $0.2 \leq n \leq 0.8$, $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$.

The relationship between the particle tumbling period τ_n and the power-law index n under simple shear flow conditions was determined through a typical curve fitting procedure to follow

$$\tau_n = \tau_1(0.9135 + 1.4724e^{-2.7645n}) \quad (5.131)$$

where τ_n is the tumbling period in a shear-thinning fluid with power-law index n and τ_1 is the particle tumbling period for the Newtonian case, i.e. when $n = 1$. Figure 5.31b shows that the orbital minimum particle tip pressure has a quadratic variation with the flow behavior index as described as

$$\bar{P}_{min,n} = \bar{P}_{min,1}(0.28 + 0.42n + 0.30n^2) \quad (5.132)$$

which implies that the shear-thinning effect on particle pressure distribution can be interpreted as having the same effect as would a modification of the Newtonian viscosity, agreeing with the findings of Ji et al.[216] and Awenlimobor et al. [227].

Figure 5.32 shows the pressure field around the ellipsoidal particle at various instants during the particle tumbling motion in the plane of the shear flow. The contours show an intensification of the pressure on the particle surface as the power-law index increases from $n = 0.2$ to $n = 1.0$. The pressure intensification is observed to be higher at orientations of peak orbital

pressure extreme magnitudes (i.e. at $\phi = \pm \pi/4$). These observations can be explained from the plot of the disturbance in the velocity $\dot{X}_i^d$ [189] around the surface of the particle due to the particles motion defined as the difference between the flow-field velocity and free stream velocity, i.e. $\dot{X}_i^d = \dot{X}_i - \dot{X}_i^\infty$ (cf. Figure 5.33). We observe a higher magnitude of the velocity disturbance around same location on the particles surface where pressure extremes are observed to occur (i.e. at the particle tips). Likewise, the intensity of the disturbance is seen to increase with increasing power-law index and the magnification is higher at critical orientation angles where the orbital peak pressure extremes occur during alignment with the principal flow directions (i.e. at $\phi = \pm \pi/4$). The lower pressure intensities are thus a result of lower disturbance in the velocity field around the particle caused by the deceleration of the particles motion in the shear-thinning fluid.

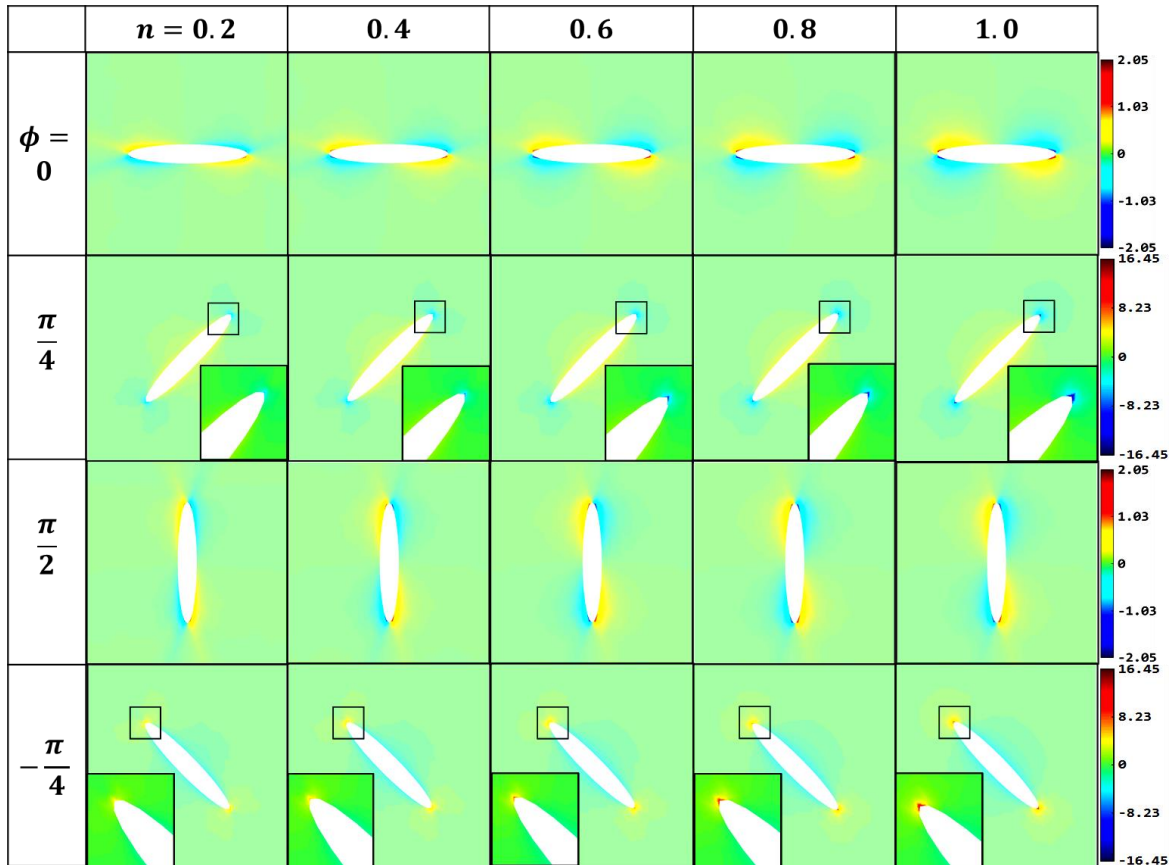


Figure 5.32: Mid - sectional plot of the pressure distribution around the ellipsoidal particle for at different instants during the particle's in-plane tumbling motion ($\phi = 0, \pi/4, \pi/4, \pi/4$) and for different power-law indices ($0.2 \leq n \leq 0.8$). Results are shown for $r_e = 6$, $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$.

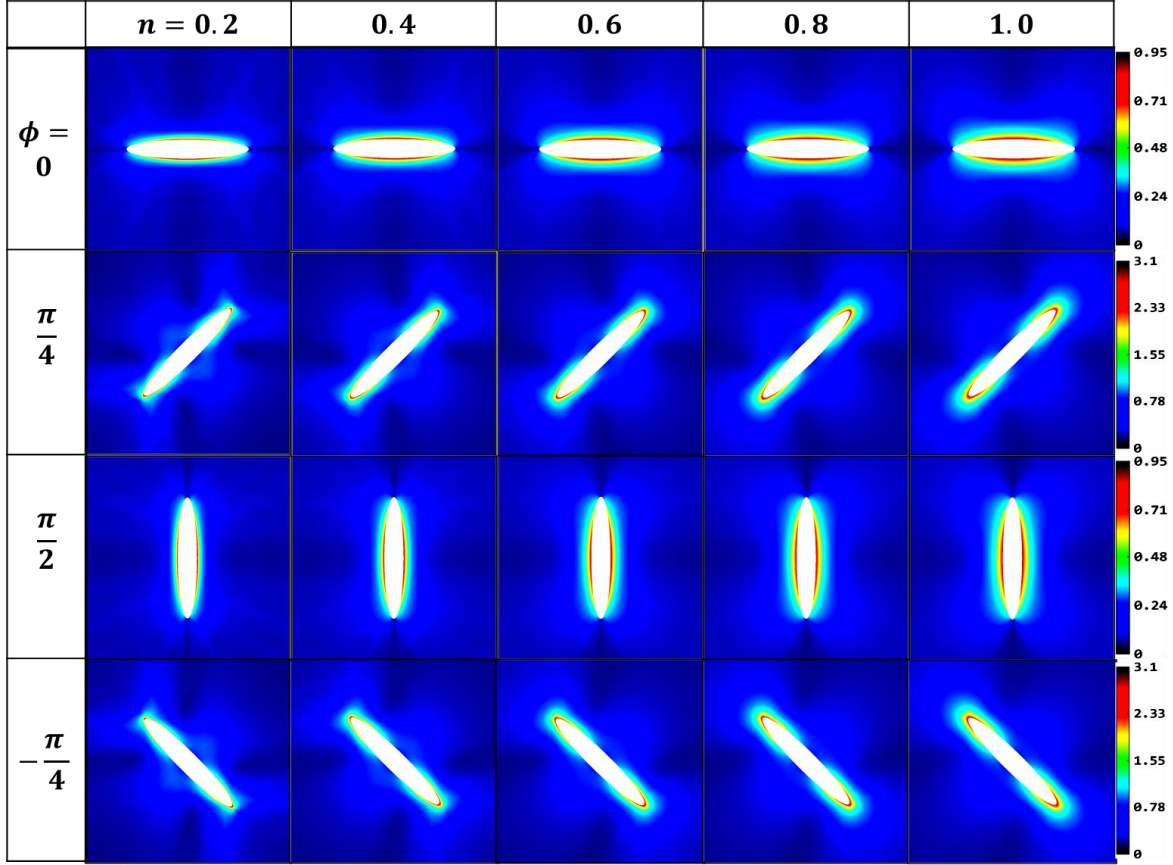


Figure 5.33: Mid - sectional plot of the disturbance velocity around the ellipsoidal particle for at different instants during the particle's in-plane tumbling motion ($\phi = 0, \pi/4, \pi/4, \pi/4$) and for different power-law indices ($0.2 \leq n \leq 0.8$). Results are shown for $r_e = 6$, $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$.

Figure 5.34a-d shows the computed results of the single rigid ellipsoidal particle in combined shear and uniaxial extension (SUA) flow type with a power-law index n ranging from 0.2 to 1 while considering two shear-extension rate ratios (i.e., $\dot{\gamma}/\dot{\epsilon} = 1$ and 10). Figure 5.34a and Table 5.4 shows that the particle stalls in the SUA flow with $\dot{\gamma}/\dot{\epsilon} = 1$) and the shear-thinning fluid behavior slightly increases particle rotation speed and shortens the trajectory which is evident from the slight reduction in the time to particle stall and the stall angle with decreasing power-law index. Figure 5.34b shows that the shear-thinning fluid reduces the magnitude of the particle surface

pressure extremes in the SUA flow, however, the shear-thinning rheology does not affect the orbital angle location where the minimum peak magnitude pressure occurs (i.e. at $\phi = +1.41$ rad).

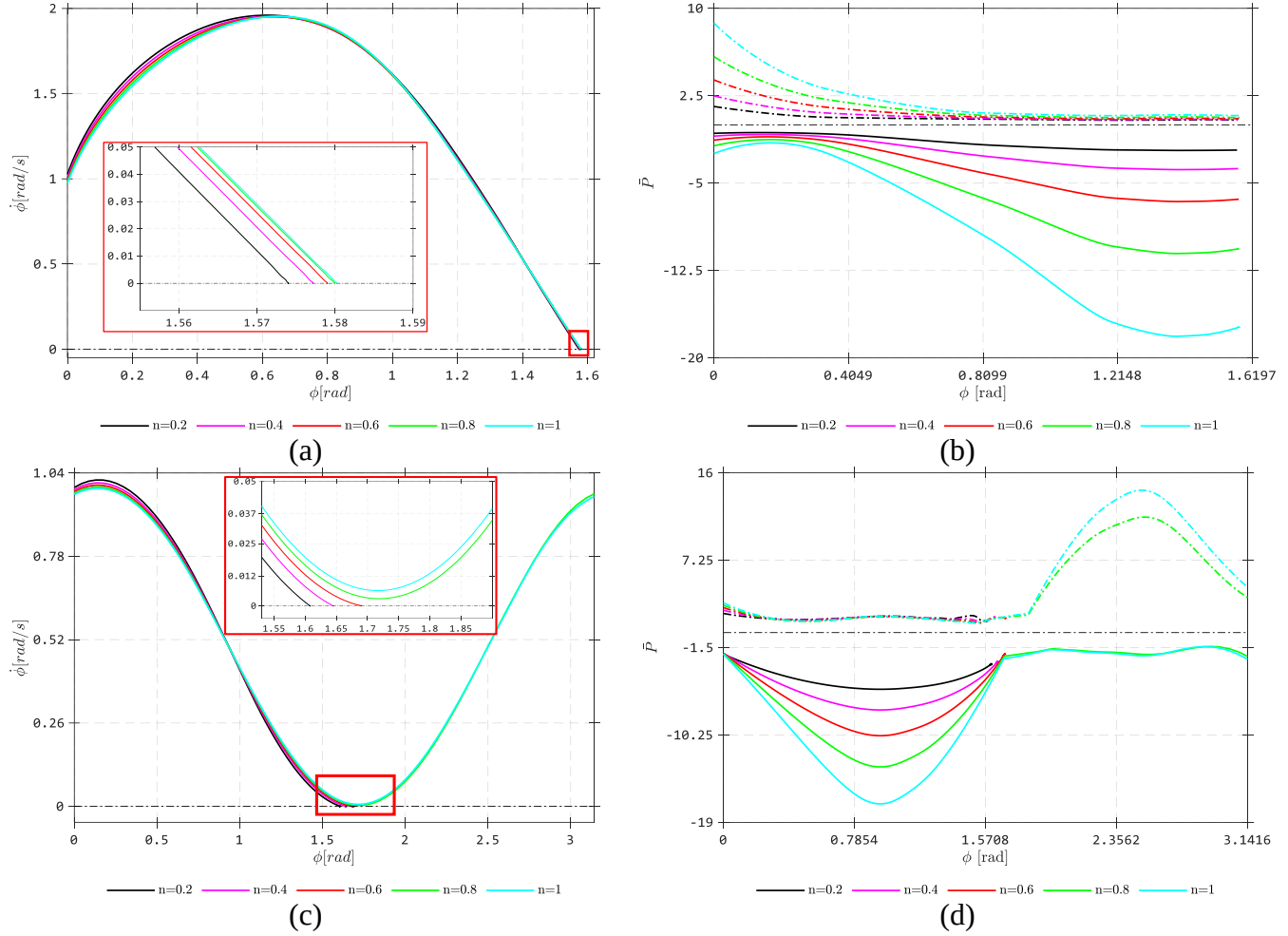


Figure 5.34: Phase diagram of the particles polar angle ϕ vs (a) precession $\dot{\phi} - \dot{\gamma}/\dot{\epsilon} = 1$ (b) surface pressure maximum (dashed) and minimum (continuous) - $\dot{\gamma}/\dot{\epsilon} = 1$ and (c) precession $\dot{\phi} - \dot{\gamma}/\dot{\epsilon} = 10$, (d) surface pressure maximum (dashed) and minimum (continuous) - $\dot{\gamma}/\dot{\epsilon} = 10$, for particle motion in combined shear and uniaxial extension (SUA) flow. Results are shown, for $0.2 \leq n \leq 0.8$, $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$.

Table 5.4: Particle stall time τ_s and particle stall angle ϕ_s for single ellipsoidal particle motion in SUA shear-thinning flow for different flow behavior index $0.2 \leq n \leq 1.0$ with $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\dot{\gamma}/\dot{\epsilon} = 1$.

n	0.2	0.4	0.6	0.8	1.0
τ_s	3.922	3.982	4.012	4.032	4.032
ϕ_s	1.574	1.577	1.579	1.580	1.580

In the shear dominant flow condition when $\dot{\gamma}/\dot{\epsilon} = 10$, the particle tumbles periodically under slightly non-Newtonian rheological fluid behavior ($n \geq 0.8$), however further reduction in the power-law index ($n < 0.8$) causes the particle to eventually stall in a preferred orientation along the direction of stretching (cf. Figure 5.34c). This implies that the conditions for particle stall in a shear-thinning fluid is dependent on the competing influence of the shear-extensional rate factor and the intensity of the shear-thinning fluid behavior. Table 5.5 shows that the particle stall time (τ_s) and stall angle (ϕ_s) when $n < 0.8$, and half period ($\tau_n^{0.5}$) for the cases where the particle tumbles periodically (i.e. when $n \geq 0.8$). As expected, at the location of the orbital extreme pressure magnitude where the particle orientation coincides with the principal flow direction (at $\phi = +0.931, +2.502$ rad), the surface extreme pressure magnitudes are observed to decrease with the intensity of the shear-thinning fluid rheology. The pressure fluctuations on the particle's tip as it tumbles continuously in the shear dominant flow or the local pressure that subsist at particle's tip as it stalls in the extension dominant flow condition are important in understanding the final microstructural formations within printed polymer composite beads [52].

Table 5.5: Half-period/stall time (where applicable) τ_s and stall angle ϕ_s (where applicable) for single ellipsoidal particle motion in SUA shear-thinning flow for different flow behavior index $0.2 \leq n \leq 1.0$ with $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\dot{\gamma}/\dot{\epsilon} = 10$.

n	0.2	0.4	0.6	0.8	1.0
$\tau_n^{0.5}$ or τ_s	31.930	39.777	65.146	59.070	40.156
ϕ_s	1.607	1.644	1.689	-	-

In the combined shearing/planar stretching (PST) flow, the shear-thinning fluid rheology does not deter the particle's acquiescence into preferred orientation state under the extension-rate dominant flow condition (i.e. $\dot{\gamma}/\dot{\epsilon} = 1$). However, the shear-thinning is observed to decelerate the particles motion, prolong the stall event and extend the particles trajectory to stall contrary to what was observed in the SUA flow. Figure 5.35a reveals a slight reduction in the peak in-plane angular velocity with decreasing power-law index and Table 5.6 shows that the stall time and stall angle

both of which increase with increased shear-thinning. The particle tip pressure magnitudes are nonetheless observed to decrease with increased shear-thinning as expected (cf. Figure 5.35b). The particle in-plane orientation at the location of orbital minimum surface pressure (i.e. at $\phi = +1.18$) is unaltered by the shear-thinning effect.

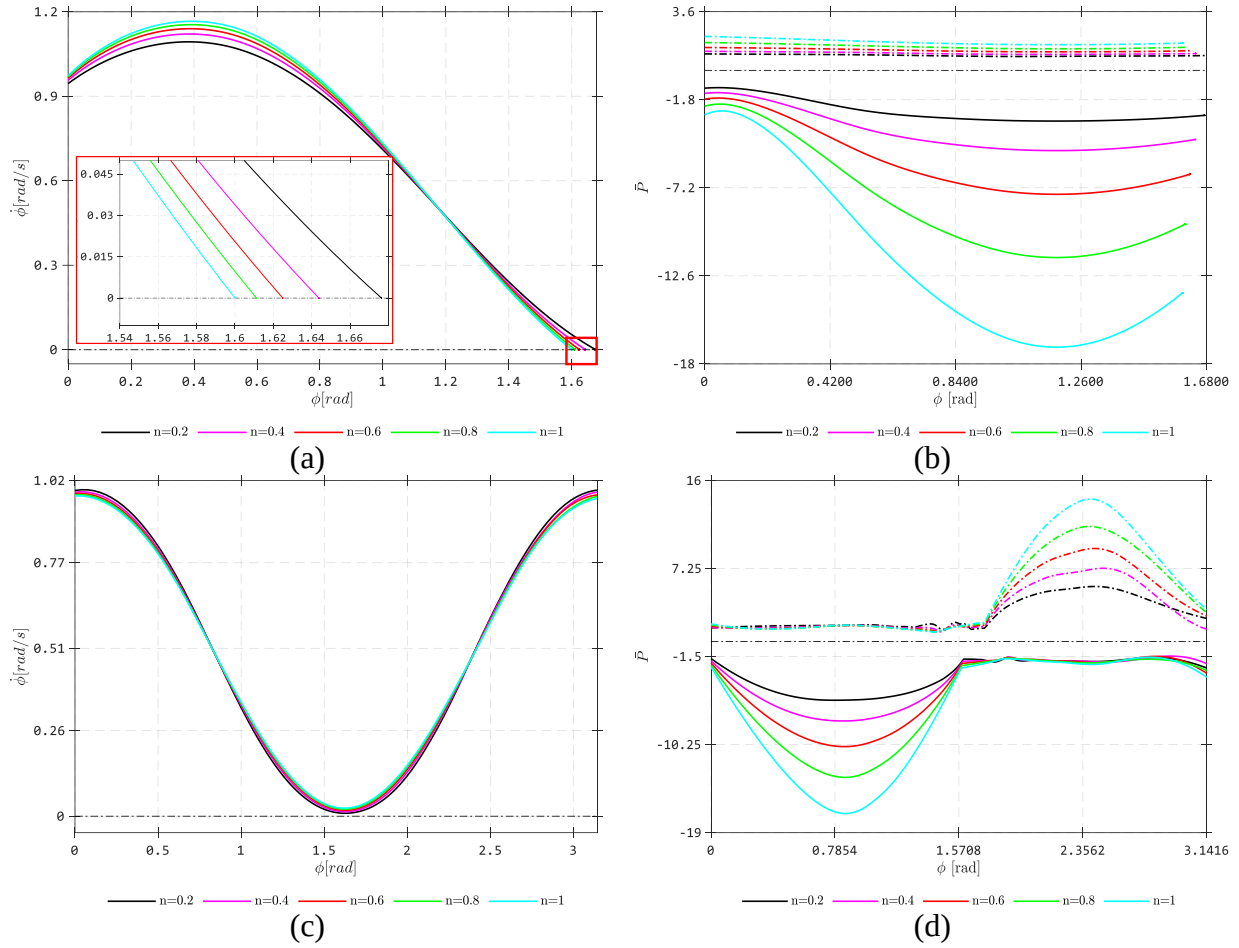


Figure 5.35: Phase diagram of the particles polar angle ϕ vs (a) precession $\dot{\phi}$ - $\dot{\gamma}/\dot{\epsilon} = 1$ (b) surface pressure maximum (dashed) and minimum (continuous) - $\dot{\gamma}/\dot{\epsilon} = 1$ and (c) precession $\dot{\phi}$ - $\dot{\gamma}/\dot{\epsilon} = 10$, (d) surface pressure maximum (dashed) and minimum (continuous) - $\dot{\gamma}/\dot{\epsilon} = 10$, for particle motion in combined shear and planar stretching (PST) flow. Results are shown, for $0.2 \leq n \leq 0.8$, $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$.

Table 5.6: Particle stall time τ_s and particle stall angle ϕ_s for single ellipsoidal particle motion in PST shear-thinning flow for different flow behavior index $0.2 \leq n \leq 1.0$ with $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\dot{\gamma}/\dot{\epsilon} = 1$.

n	0.2	0.4	0.6	0.8	1.0
τ_s	13.337	11.796	11.026	10.515	10.135

ϕ_s	1.676	1.644	1.625	1.611	1.600
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The shear-thinning effect does not stall the particle under the shear-rate dominant condition (i.e. when $\dot{\gamma}/\dot{\epsilon} = 10$) in the PST flow contrary to what was observed in the SUA flow. However, at the local minima of the particle's angular velocity evolution curve when its deceleration approaches zero (cf. Figure 5.35c), the increased shear-thinning effect is observed to further decelerate particle motion and bring it closer to stall condition. Table 5.7 shows that the particles tumbling period increases with decreasing power-law index indicating the deceleration of the particle rotation with increased shear-thinning. The sustained particle motion allows for continuous fluctuations between particle surface pressure extremes at the particle tip. As would be expected, the pressure magnitudes are observed to decrease with increased shear-thinning (cf. Figure 5.35d). Further, the in-plane orientation at the orbital location of particle surface tip pressure extremum (i.e. at $\phi = +0.835, +2.406\text{rad}$) is unaltered by the shear-thinning effect.

Table 5.7: Half-period for single ellipsoidal particle motion in PST shear-thinning flow for different flow behavior index $0.2 \leq n \leq 1.0$ with $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\dot{\gamma}/\dot{\epsilon} = 10$.

n	0.2	0.4	0.6	0.8	1.0
$\tau_n^{0.5}$	36.181	28.045	24.169	21.839	20.280

Under the balanced shear and bi-axial elongation (SBA) flow condition, inward flow normal to the shear plane coupled with uniform stretching along the shear plane promotes particle in-plane tumbling motion. Under this flow condition, the particle does not stall irrespective of the magnitude of the extension rate. However, while the increased shear-thinning is observed to accelerate the particles motion when $\dot{\gamma}/\dot{\epsilon} = 1$, it is shown to slightly decelerate the particles motion under a higher shear rate i.e. $\dot{\gamma}/\dot{\epsilon} = 10$ (cf. Figure 5.36a & c). When $\dot{\gamma}/\dot{\epsilon} = 1$ the limits of particle in-plane angular velocity are observed to decrease with increased shear-thinning and vice versa when $\dot{\gamma}/\dot{\epsilon} = 10$. The shear-thinning effect decreases the particle tumbling period when

$\dot{\gamma}/\dot{\epsilon} = 1$ and increases the period when $\dot{\gamma}/\dot{\epsilon} = 10$ (cf. Table 5.8). Under a lower shear rate ($\dot{\gamma}/\dot{\epsilon} = 1$), there are no noticeable peaks in the evolution of the particle maximum surface pressure, contrary to what is observed when $\dot{\gamma}/\dot{\epsilon} = 10$. As would be expected, the particle surface pressure extremes are observed to decrease with increased shear-thinning and the location of orbital minimum surface pressure at $\phi = \pm \pi/4$ is unaffected by the shear-thinning rheology (cf. Figure 5.36b & d).

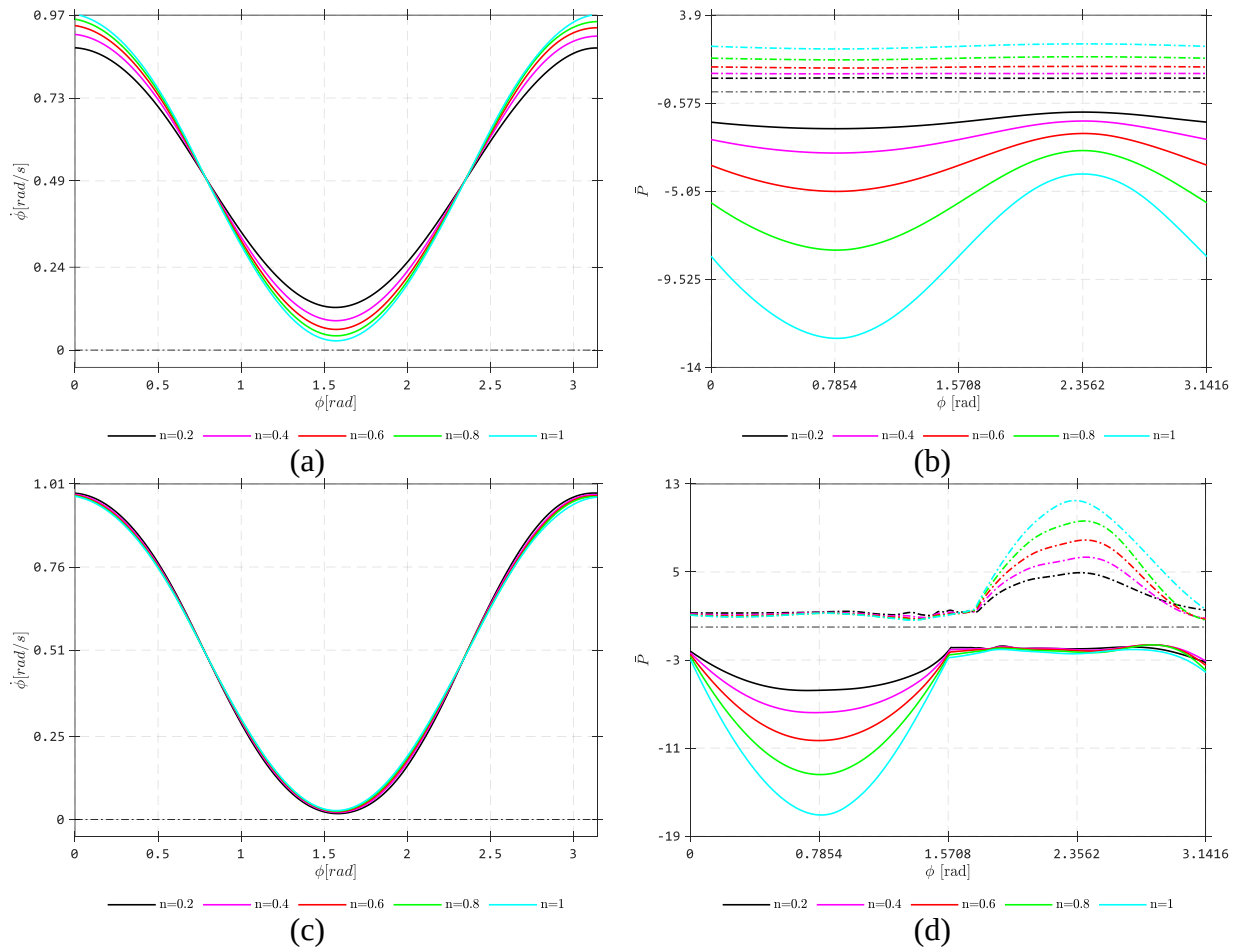


Figure 5.36: Phase plots of the particles polar angle ϕ vs (a) precession $\dot{\phi}$ - $\dot{\gamma}/\dot{\epsilon} = 1$ (b) surface pressure maximum (dashed) and minimum (continuous) - $\dot{\gamma}/\dot{\epsilon} = 1$ and (c) precession $\dot{\phi}$ - $\dot{\gamma}/\dot{\epsilon} = 10$, (d) surface pressure maximum (dashed) and minimum (continuous) - $\dot{\gamma}/\dot{\epsilon} = 10$, for particle motion in combined shear and biaxial extension (SBA) flow. Results are shown, for $0.2 \leq n \leq 0.8$, $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$ and $\phi^0 = 0, \theta^0 = -\pi/2, \psi^0 = 0$.

Table 5.8: Half tumbling period $\tau_n^{0.5}$ for single ellipsoidal particle motion in SBA shear-thinning flow for different flow behavior index $0.2 \leq n \leq 1.0$ and different shear to extension rate ratio ($\dot{\gamma}/\dot{\epsilon}$) with $m = 1 \text{ Pa} \cdot \text{s}^n$, $\dot{\gamma} = 1 \text{ s}^{-1}$.

$\tau_n^{0.5}$		n				
		0.2	0.4	0.6	0.8	1.0
$\dot{\gamma}/\dot{\epsilon}$	1	9.558	11.273	13.266	15.799	19.453
	10	25.265	22.650	21.155	20.138	19.423

Observation of particle behavior in the flow types considered here as applied to polymer melt flow conditions during EDAM processing suggests that the shear-thinning effect increases the particle stall tendency closer to the EDAM nozzle center where a higher extension rate dominance is seen. Shear-thinning is seen here to have a similar effect as decreasing the shear-to-extension rate ($\dot{\gamma}/\dot{\epsilon}$), thus shifting the boundaries of the extension dominant region outward (cf. APPENDIX B, B.3). Irrespective of the flow regime, the shear-thinning rheology reduces the pressure magnitude which has a similar effect to reducing the viscosity magnitude in a Newtonian fluid. Additionally, in high shear dominant flow regions of the EDAM nozzle, the shear-thinning effect is generally expected to slow down the particles motion, while close to the nozzle center, dominated by high extension-rate, the particle's stall event is expected to be promoted by shear-thinning effects.

5.2.2.2 *Effect of Initial Particle Orientation*

In earlier sections we showed that the pressure magnitudes on the surface of a particle suspended in a Newtonian simple shear flow reduces as the orbit constant ζ (cf. eqn. (5.65) & (5.66)) goes from $\zeta = +\infty$ where the particle is tumbling in the shear plane to $\zeta = 0$ where the particle is spinning about its axis perpendicular to the shear plane. It was also shown that the tumbling period was unaffected by Jeffery's orbit. The effect of shear-thinning rheology on the particle motion for various Jeffery orbits are presented in this section. We consider particle motion in simple shear flow with shear rate of $\dot{\gamma} = 1 \text{ s}^{-1}$ and for a GNF power-law fluid rheology with a

power-law index of $n = 0.5$ and a consistency index of $m = 1 \text{ Pa} \cdot \text{s}^n$. The same geometric aspect ratio of $r_e = 6$ as was previously used is considered here.

The 2D sensitivity analysis on the fibers' initial condition showed that the angular velocity of the fiber is unaltered by the initial condition (cf. Figure 5.37a), nor is its limit pressure peaks on the fiber's surface affected (cf. Figure 5.37b) in a shear-thinning fluid with strong non-Newtonian characteristics (flow behavior index $n = 0.2$).

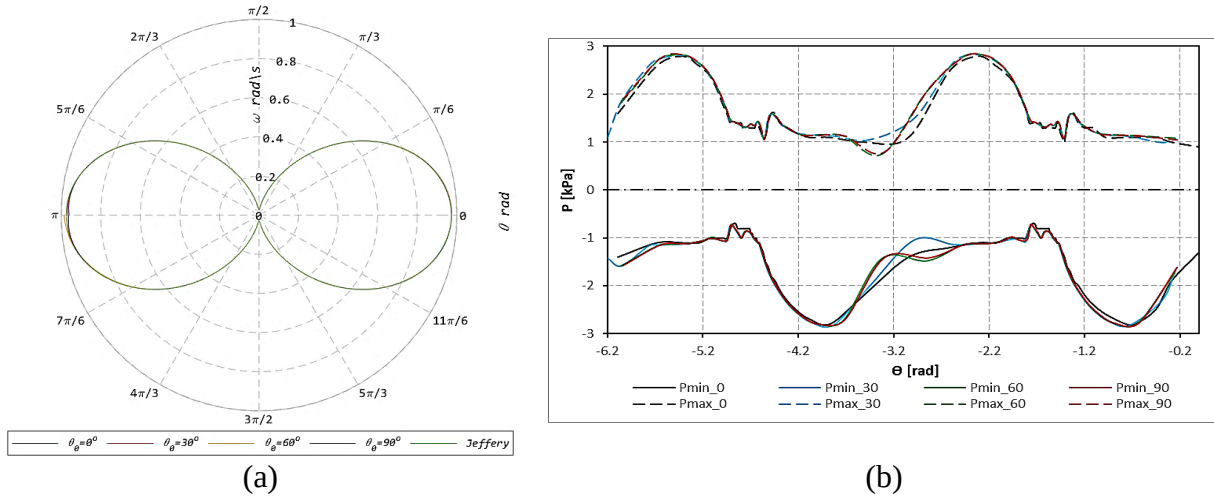


Figure 5.37: Figure showing results of (a) evolution of the fibers angular velocity in space along Jeffery's orbit (b) evolution of fibers limit surface pressure in space along Jeffery's orbit. Results are presented for different fiber initial orientation ($\theta_0 = 0^\circ, 30^\circ, 60^\circ, 90^\circ$) and for flow shear rate $\dot{\gamma} = 1 \text{ s}^{-1}$, fiber aspect ratio $r_e = 6$, and flow behavior index $n = 0.2$.

In the 3D analysis, the Jeffery's orbits are altered slightly by the shear-thinning fluid which occurs to a greater extent as the fiber is oriented further out of the shear plane (i.e., as $\zeta = +\infty$ is moved to $\zeta = 0$) as shown in Figure 5.38a. The initial particle polar angle on a particular Newtonian Jeffery's orbit is observed to also modify the particle trajectory. Figure 5.38a and b also show that trajectory of the particle motion in an orbit with initial azimuth angle of $\theta^0 = 2\pi/24$ with two initial starting positions at the vertices of the Newtonian conical orbit. With an initial starting position at the vertex of the directrix of the Newtonian conical orbit on the major axis (at $\phi^0 = \pi/2$), the particle path is seen to dilate outwardly defined by the outer curve (dashed

cyan line) from the Newtonian orbit (continuous black line). However, starting the particle from the co-vertex of the directrix of the Newtonian orbit on the minor axis (i.e. $\phi^0 = 0$), the orbit constricts inwardly defined by the inner curve (continuous cyan line). Both curves clearly illustrate the extent of deviation in the particle path from the Newtonian orbit and that for a given power-law index and set of flow parameters. The fluid shear-thinning is seen to influence the particles motion similar to elongating or shortening the particle, depending on the initial position on the orbit. This observed behavior is consistent with conclusions by Abtahi et al.[189].

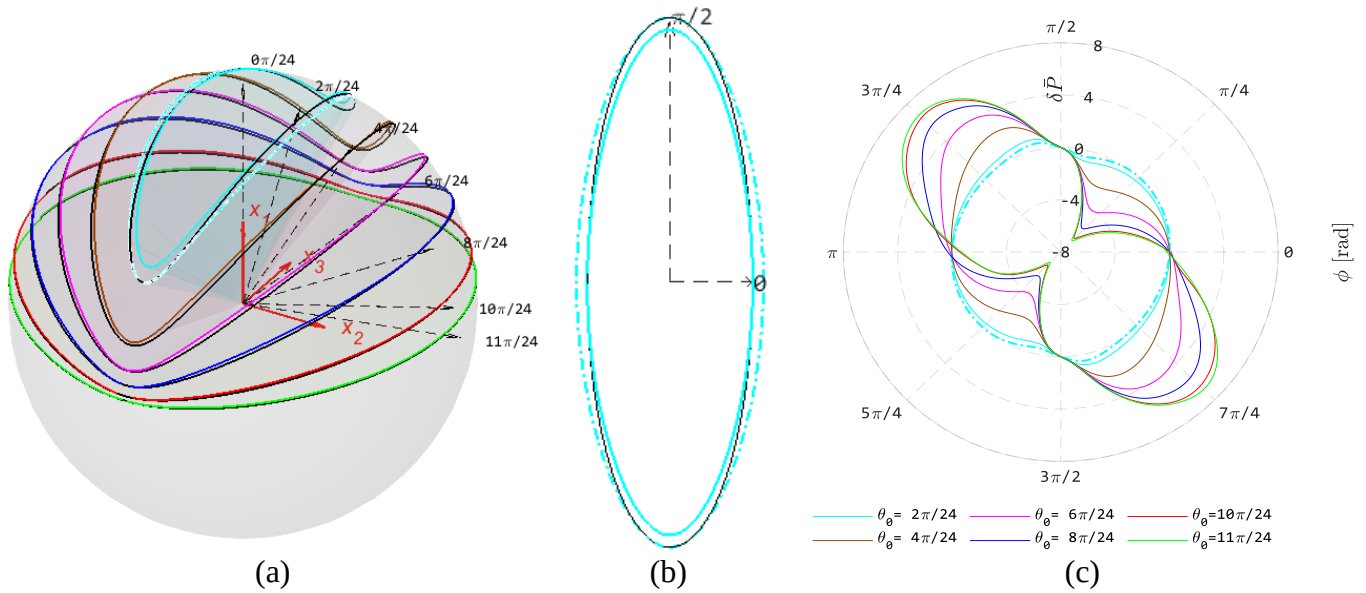
The fluid shear-thinning is seen to have a more profound effect on the surface pressure of particles on Jeffery orbit closer to the shear plane ($\zeta \rightarrow +\infty$) as compared to orbits farther out of plane (i.e. close to $\zeta \rightarrow 0$). The net pressure drop ($\delta\bar{P}$) due to the shear-thinning effect is seen to be proportional to the magnitude of the particle surface pressure as shown in Figure 5.38c. Likewise, the net pressure drop of particle tip pressure is seen to depend on its initial starting position as is evident from the net pressure curves shown for each initial polar angle on the orbit farthest from the shear plane ($\theta^0 = 2\pi/24$), i.e. dashed cyan line for $\phi^0 = 0$ and continuous cyan line for $\phi^0 = \pi/2$.

As expected, the particle dynamics are also affected by the shear-thinning rheology. The envelope of the phase diagram of the particle's nutation (cf. Figure 5.38d) contract inwardly from the Newtonian envelope due to the shear-thinning effect irrespective of the initial position on the orbit. The shear-thinning rheology appears to have less effect on the particle's precession as the Jeffery's orbit is oriented further out of plane, i.e. when $\zeta \rightarrow 0$ (cf. Figure 5.38e), however, this effect on the particle's nutation is more profound as $\zeta \rightarrow 0$. Although, the particle's period of tumbling is independent on the Jeffery's orbit in Newtonian flow, the tumbling period is observed to be influenced by the Jeffery's orbit under shear-thinning flow conditions. Figure 5.39a shows

the relationship between the tumbling period $\tau_{0.5}$ and the initial azimuth angle, θ_0 for the particle motion in non-Newtonian power-law fluid, with flow behaviour index of $n = 0.5$. The relationship in Figure 5.39a can be described as

$$\tau_{0.5} = \tau_1(1.2976 - 0.7358e^{-3.8495\theta_0}) \quad (5.133)$$

which has been obtained using a typical curve fitting procedure. Overall, the shear-thinning fluid rheology slows down a particle's motion which occurs to a greater degree as the tumbling orbit approaches the shear plane (i.e. $\zeta \rightarrow +\infty$). Additionally, the reduction in the minimum surface pressure magnitudes due to shear-thinning becomes more significant as $\zeta \rightarrow +\infty$ and vice-versa. The relationship between the particles orbital minimum tip pressure $\bar{P}_{min}$ and the initial particles out-of-plane orientation θ^0 appearing in Figure 5.39b clearly shows a gradual widening of the gap between the Newtonian and non-Newtonian pressure profiles.



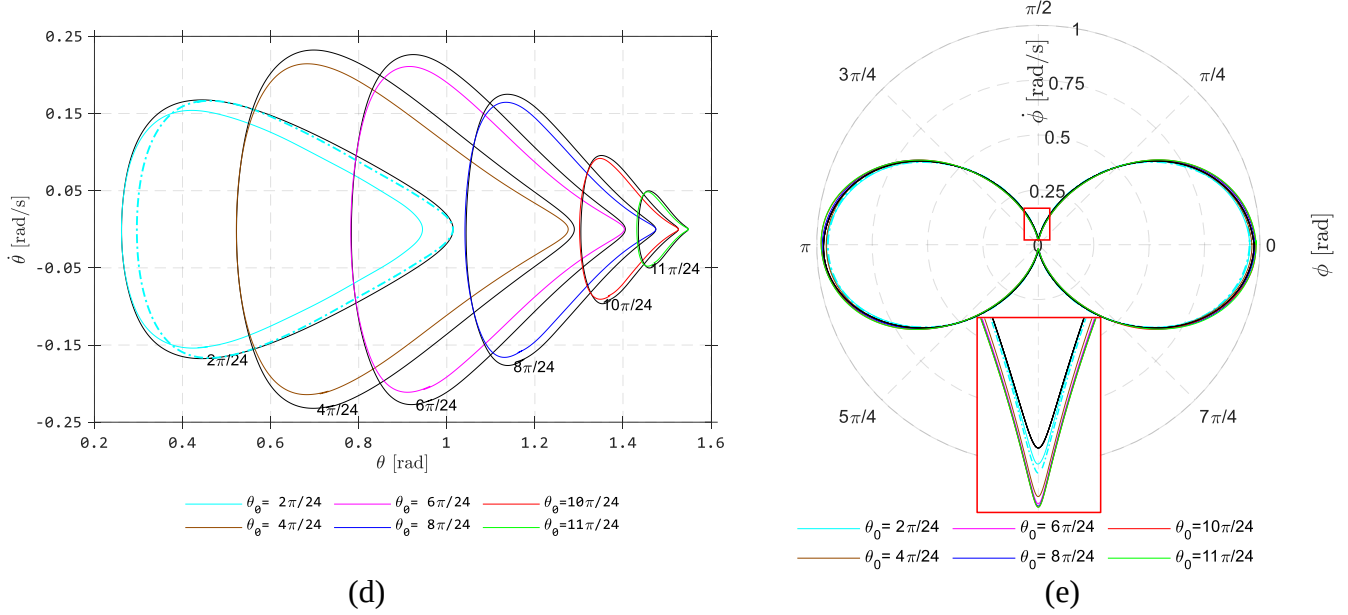


Figure 5.38: Effect of fluid shear-thinning on Jeffery's orbit: (a) particle's orbits in simple shear flow (b) dilated orbit (dashed cyan line $\phi^0 = \pi/2$), constricted orbit (continuous cyan line, $\phi^0 = 0$) and Newtonian orbit (black line) for $\theta^0 = 2\pi/24$ (c) difference in particle tip pressure between NT and GNF fluid (d) phase diagram of azimuth angle θ vs nutation $\dot{\theta}$ (e) polar plot of precession $\dot{\phi}$ vs polar angle ϕ , for different initial particle orientation between $-2\pi/24 \leq \theta^0 \leq -12\pi/24$, $\phi^0 = 0$, $\psi^0 = 0$ and for NT fluid (dashed) and GNF power-law fluids (continuous) with $m = 1 \text{ Pa} \cdot \text{s}^n$, $n = 0.5$.

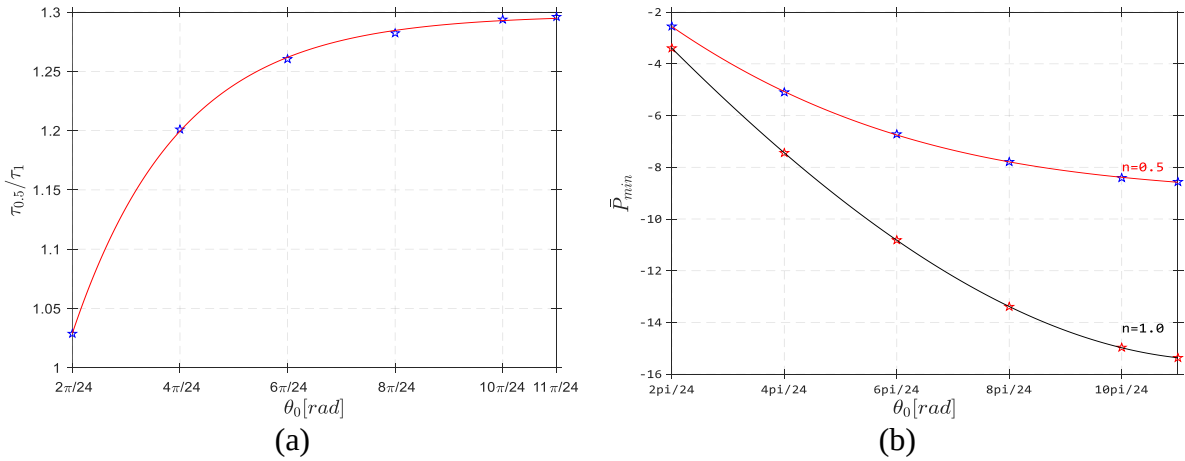
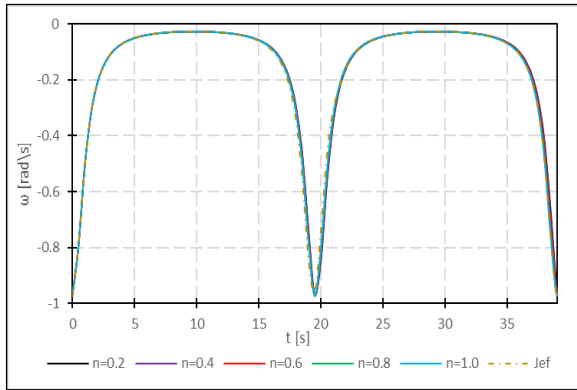


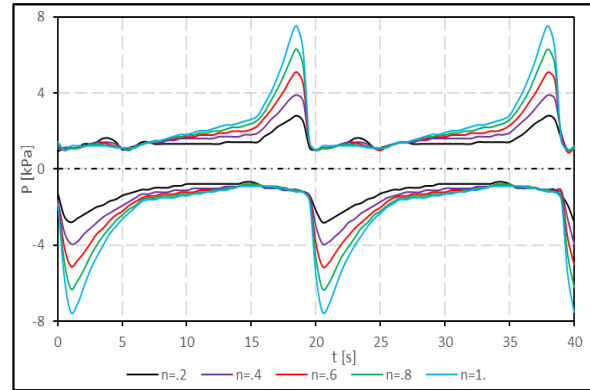
Figure 5.39: Effect of shear-thinning comparing (a) the non-Newtonian to Newtonian tumbling period $\tau_{0.5}/\tau_1$, and (b) the non-Newtonian (red line) and Newtonian (black line) particles orbital minimum tip pressure $\bar{P}_{min}$, versus the initial azimuth angle θ_0 , considering GNF power-law fluid, with $m = 1 \text{ Pa} \cdot \text{s}^n$, $n = 0.5$ and initial orbit position $\phi^0 = 0$.

5.2.2.3 Effect of Geometric Aspect Ratio

The result for the evolution of the 2D rigid ellipsoidal fiber along Jeffery's orbit in viscous fiber suspension simple shear flow with shear thinning fluid rheology having flow behavior index ranging from 0.2 to 1.0 are presented in Figure 5.40 below for two (2) cases of fibers geometric aspect ratio, i.e., a prolate spheroid with geometric ratio $r_e = 6$ and a slender fiber with geometric ratio $r_e = 30$. For the first case, a shear rate of $\dot{\gamma} = 1 \text{ s}^{-1}$ is used however to reduce the orbit period for the case with high aspect, a shear rate of $\dot{\gamma} = 3 \text{ s}^{-1}$ was used given the definition of the Jeffery's orbit period (cf. eqn. (5.68)). The result in Figure 5.40a & b shows that irrespective of the fibers aspect ratio, the fiber's motion is unaffected by the shear-thinning fluid rheology in agreement with the findings of numerous literatures [226]. Although only a single periodic motion was computed, there was no significant effect of the shear thinning fluid rheology observed on the fiber's motion that would result in a departure from Jeffery's orbit within a finite timescale. The minimum and maximum pressure peaks on the fiber's surface were however observed to increase proportionally with the flow behavior index for both fiber aspect ratio cases (cf. Figure 5.40c&d).



(a)



(c)

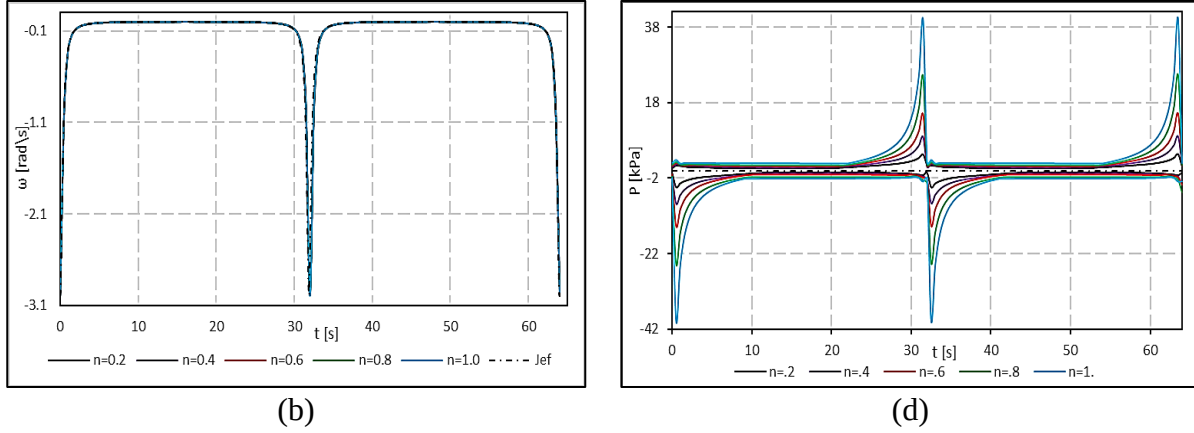


Figure 5.40: Figure showing results of evolution of fibers angular velocity in time along Jeffery's orbit for (a) fiber with aspect ratio $r_e = 6$, and flow with shear rate $\dot{\gamma} = 1 \text{ s}^{-1}$ (b) fiber with aspect ratio $r_e = 30$, and flow with shear rate $\dot{\gamma} = 3 \text{ s}^{-1}$. Also shown are results of evolution of fibers limit surface pressure in time along Jeffery's orbit for (c) fiber with aspect ratio $r_e = 6$, and flow with shear rate $\dot{\gamma} = 1 \text{ s}^{-1}$ (d) fiber with aspect ratio $r_e = 30$, and flow with shear rate $\dot{\gamma} = 3 \text{ s}^{-1}$. (Results are presented for different shear-thinning fluid with flow behavior index ranging from $n = 0.2 - 1.0$).

Unlike the 2D results, the 3D sensitivity study on the influence of the particle geometric aspect ratio on its field state showed that the aspect ratio significantly influences the observed particle kinematic behaviour and the surface pressure distribution in Newtonian shear flow. For completeness, we now consider the effect of the geometric aspect ratio on particle behaviour in shear-thinning simple shear flow making comparisons to the behaviour in a Newtonian fluid. Previous studies showed that the shear-thinning effect on the particle's orbit are magnified with increasing initial out of plane orientation θ^0 [189]. As such we consider Jeffery's orbit with initial particle orientation of $\phi^0 = 0, \theta^0 = 2\pi/24$, and $\psi^0 = 0$. Figure 5.41 shows the deviation in particle trajectories, pressure and dynamic responses between the shear-thinning and Newtonian fluid for various particle aspect ratios. For spherical shaped particles, shear-thinning has no significant effect on the particles orbit, or the evolution of the particle's surface pressure and dynamic responses. However, as the particle aspect ratio increases up to $r_e = 6$, we observe considerable deviation in the particle trajectory (cf. Figure 5.41a) consistent with the findings of Abtahi et al. [189]. Similar to results that appear above, the particle trajectory is elongated or

constricted depending on the initial starting position on a particular Newtonian Jeffery's orbit. With a further increase in the particle's slenderness, i.e. as $\kappa \rightarrow 1$, modification of the particle's trajectory due to shear-thinning becomes negligible as was also observed by Ferec et al. [226].

The shear-thinning effect on the pressure response however continues to increase with the particle length (cf. Figure 5.41b) which can be attributed to the hydrostatic stress intensification at the particle's tip arising from the increased particle length and/or the related decrease in the tip curvature. Likewise, the impact of shear-thinning on particle angular velocities is initially observed to increase with increasing aspect ratio. The non-linear effects, however, gradually declines with further increase in ellipsoid's slenderness. The shear-thinning behaviour is observed to slightly decrease the particles orbit period with slight increase in the aspect ratio. Further increases in the particle's slenderness, however, results in the shear-thinning behaviour prolonging the tumbling period. At lower aspect ratios, the pressure drag which does not depend on the local viscosity dominates the hydrodynamic resistance, however, with longer particles, the skin friction drag becomes significant due to the increased surface area and change in apparent viscosity [180]. Since a decrease in the apparent viscosity is known to slow down particle motion, we experience longer tumbling periods with considerable increase in the particle aspect ratio (cf. Table 5.9).

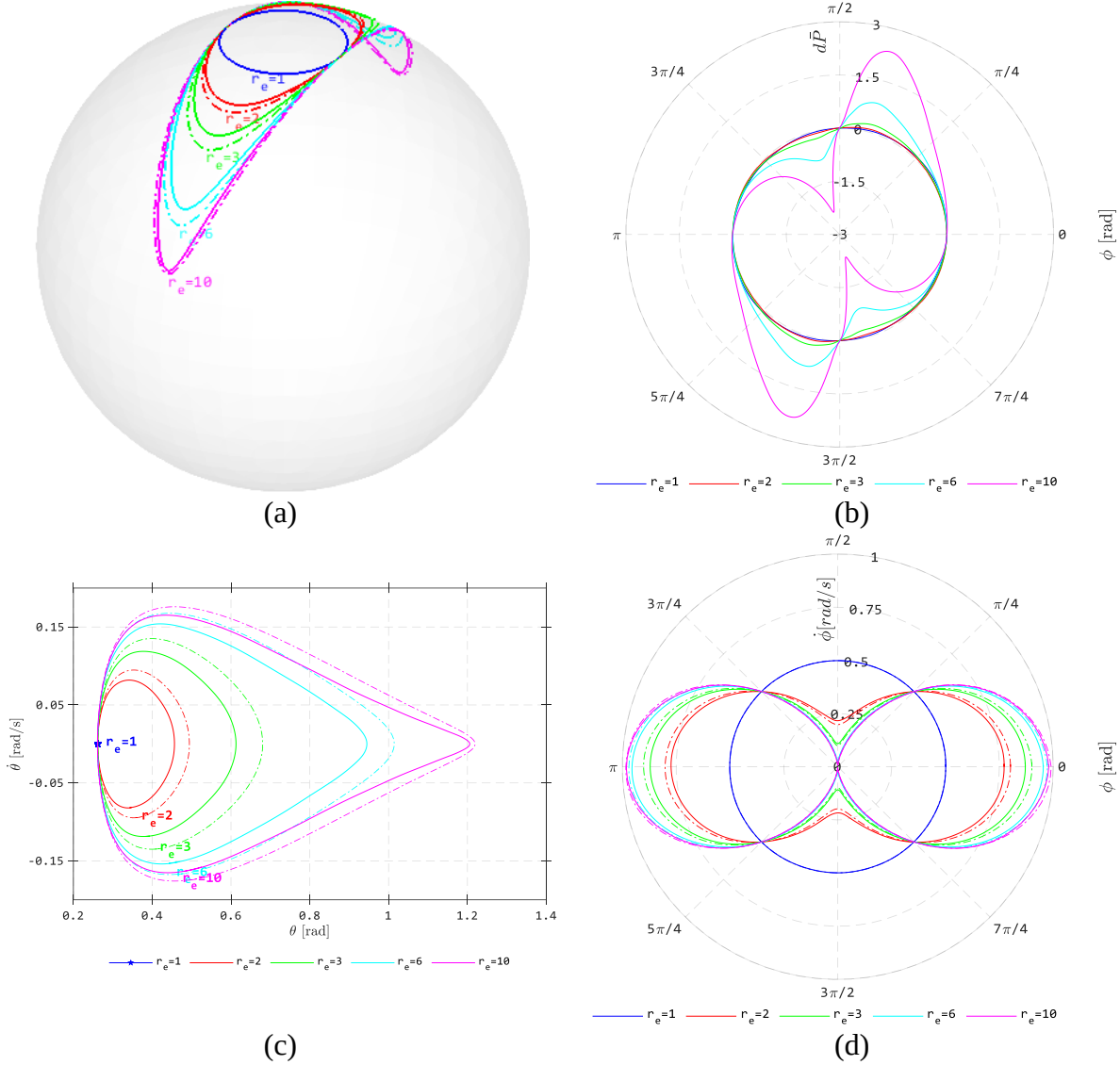


Figure 5.41: Showing (a) particle's orbits in simple shear flow (b) difference in particle tip pressure evolution between NT and GNF fluid (c) phase diagram of azimuth angle θ vs nutation $\dot{\theta}$ (d) polar plot of the polar angle ϕ vs precession $\dot{\phi}$, for different particle aspects r_e and for NT fluid (dashed) and GNF power-law fluids (continuous) with $m = 1 \text{ Pa} \cdot \text{s}^n, n = 0.5$. Initial particle orientation is $\phi^0 = 0, \theta^0 = 2\pi/24, \psi^0 = 0$.

Table 5.9: Ratio of the particle's tumbling period in shear-thinning simple shear flow with $n = 0.5$ and $m = 1 \text{ Pa} \cdot \text{s}^n$, and $\dot{\gamma} = 1 \text{ s}^{-1}$ and with to the reference Newtonian quantity, τ_1 . Initial particle orientation is $\phi^0 = 0, \theta^0 = 2\pi/24, \psi^0 = 0$.

r_e	1	2	3	6	10
$\tau_{0.5}/\tau_1$	1.000	0.966	0.951	1.027	1.250

Since typical EDAM printed fiber-filled polymer composites are known to have very high aspect ratios $r_e > 45$ [317], [318], the shear-thinning rheology is expected to have negligible

effects on particle angular velocity and trajectory. However, we expect the non-Newtonian fluid slows down the particles kinematics and reduces the surface pressure distribution.

5.2.2.4 *Effective viscosity of shear-thinning suspension*

The flow behavior index of the shear-thinning fluid has an effect analogous to the influence of a Newtonian fluid viscosity on the pressure response on the fiber surface. Figure 5.15 shows the variation of the 2D fiber surface limit pressure response with the Newtonian viscosity μ (or consistency coefficient m for power law fluid with behavior index of $n = 1$). We earlier observed that the pressure magnitude on the fibers surface increases with increasing Newtonian viscosity like the influence of the flow behavior index on the pressure response (Figure 5.40b). This suggests the occurrence of regions of low and high viscosities extremes on the fibers surface during the fibers tumbling motion within the non-Newtonian fluid. Figure 5.42 shows extracted data points (blue dots) of the instantaneous shear viscosity and shear rate scalar magnitude on the fibers surface over the complete period of fiber tumbling motion and for a power law index $n = 0.2$. The average viscosity η_1 and the viscosity corresponding to the average shear rate magnitude η_2 on the fibers surface over the entire period are also shown. From both values, η_2 is observed to be a better representation (definition) of the ‘effective’ mean viscosity on the fibers surface with an order of magnitude like the flow viscosity due to the imposed shear rate on the flow-field.

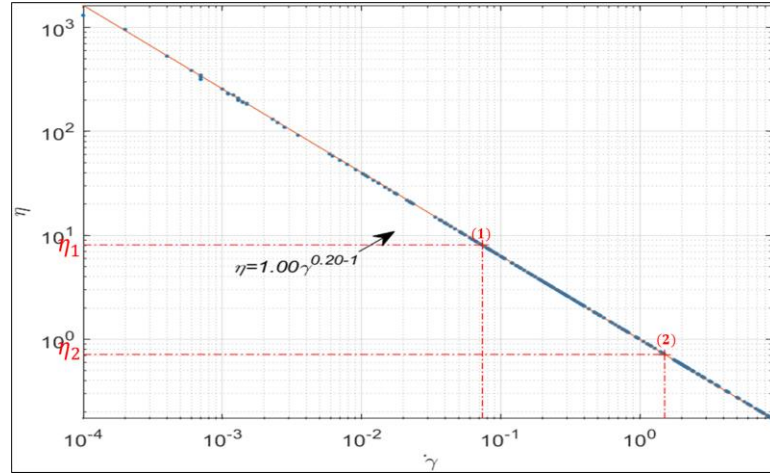
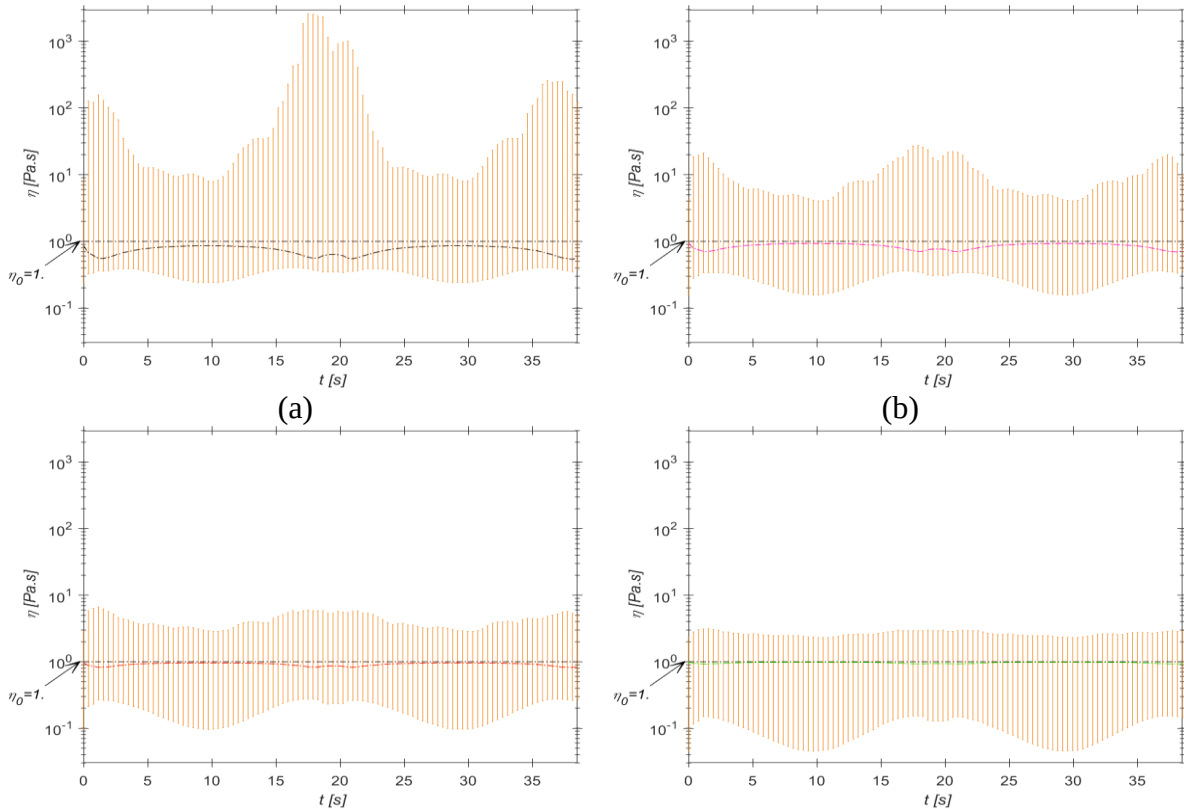


Figure 5.42: Scatter plot of the shear rates and resulting shear viscosities on the 2D fibers surface over the complete period of fiber tumbling motion. Indicated on the plot are the mean value points (1 & 2) of the shear rate and viscosities.

To gain a better understanding on the dynamics of the shear viscosities on the fiber surface during its tumbling motion and its influence on the fibers surface limit pressures, we present transient profiles of the evolution of the effective mean shear viscosity η_2 and the corresponding viscosity limits at each time interval for different flow behavior index (cf. Figure 5.43).



(c) (d)

Figure 5.43³: Figure showing time evolution of the average and limits of the non-Newtonian viscosity on the fiber's surface for different flow behavior index (a) $n = 0.2$, (b) $n = 0.4$, (c) $n = 0.6$, (d) $n = 0.8$. For the Newtonian case, all upper & lower limits of the viscosities and effective mean viscosity transient profile all coincide with the far-field viscosity at $\eta_0=1$.

From the profiles, we see that although the limits of shear rates magnitudes and resulting viscosities increases with decreasing low behavior index, the effective mean viscosity η_2 on the fibers surface only slightly shifts below the actual farfield viscosity η_0 . Following the effect of the Newtonian viscosity observed on the pressure limits on the fiber surface (cf. Figure 5.42a), we can infer in a qualitative sense that the decreasing trend in the effective mean viscosities with decreasing flow behavior index observed over the tumbling period in Figure 5.43a-d above are responsible for the low pressure limit magnitudes on the fiber surface.

5.3 Conclusion

In conclusion, a non-linear FEM numerical approach has been implemented to investigate the effects of shear-thinning fluid rheology in combination with other factors including the particles aspect ratio and initial particle orientation on the dynamics and surface pressure distribution on a particle suspended in viscous homogenous flow. The particles behavior in a special class of homogenous flows that typifies conditions found in melt flow regions of the of an extrusion nozzle during polymer composite additive manufacturing processing is also studied.

In the Newtonian flow, the ellipsoidal particle stalls in extension dominant asymmetric flow regimes but tumbles periodically in axisymmetric flows irrespective of the magnitude of the extension rate. The stall event in asymmetric flows is dictated by the shear-to-extension rate ratio.

³ The plots indicate viscosity limits on the fiber surface are exacerbated as the flow behavior index decreases due to the power law relationship. i.e., $\lim_{\substack{n \rightarrow 0 \\ \gamma \rightarrow \infty}} \eta \rightarrow 0$ and $\lim_{\substack{n \rightarrow 0 \\ \gamma \rightarrow 0}} \eta \rightarrow \infty$.

Increased shear dominance increases flow symmetry and tendency for continuous and periodic particle tumbling. The tumbling period in the asymmetric flows is expectedly dependent on the shear-to-extension rate ratio. The tumbling period increases asymptotically with increasing extension dominance until the conditions for stall based on Jeffery's equation are satisfied. On the other hand, the evolution time to particle stall is shown to increase asymptotically with increased shear dominance until the conditions for stall are violated. With sustained particle motion, the particle tip pressure fluctuates between extremums at the instants where its orientation aligns with the principal flow directions. An increase in the ellipsoidal particle aspect ratio was shown to affect the particles dynamics and increase the tumbling period. It also was shown to exacerbate the pressure extremes at the particle tip which could be caused by the increased aspect ratio alone, or the related reduction in tip curvature, or both. With a narrowing of Jeffery's orbit as the particle tumbles further out of plane, the particle surface pressure extremes are observed to decrease and the surface location of the pressure extreme further deviates from the particle's tip location. The orbital peak particle tip pressure magnitude follows a somewhat linear relationship with the polar location on the orbit across spectrum of degenerate Jeffery's orbit.

Although in the 2D study, the particle's dynamics is unaffected by the shear-thinning fluid, the behavior of the suspended particle in the 3D study is shown to be affected by the shear-thinning fluid rheology. In the axisymmetric flows where the particle motion ensues periodically, the shear-thinning fluid rheology slows down the particles motion and increases the tumbling period. Cessation of particle motion (i.e., a stall condition) in the asymmetric homogenous flows is shown to be dictated by a competing influence of the shear-thinning intensity and shear to extension rate dominance. The shear-thinning was found to have similar effect has decreasing the shear-rate dominance of the prevailing flow on the particles motion. Irrespective of the homogenous flow

type, the magnitude of the particle surface pressure distribution was observed to significantly decrease with increased shear-thinning intensity due to an accompanying decrease in the effective viscosity of the fluid around the particle surface. The orbital location at which the pressure magnitude extremes on the particles surface are, however, unaffected by the shear-thinning rheology. On the shear-plane, shear-thinning rheology has no noticeable effect on the particles orbit, however, with a narrowing of the Jeffery orbit as we move further out of plane, the particle's trajectory deviates further from the Newtonian reference path. The shear-thinning rheology may either constrict or dilate the Newtonian orbit depending on the initial starting location of the particle on the orbit. The elongation of the particle's motion and the lowering of the pressure on the surface of the particle by the shear-thinning effect is augmented with widening of Jeffery's orbit as the particle tumbles closer to the shear plane. For spherical particles, the shear-thinning fluid has no significant effect on the dynamics or surface pressure distribution, but with increased aspect ratio, modification of the particle's trajectory and dynamics due to the non-linear effects becomes significant until a critical point, where the non-linear effects are reversed. With excessive particle slenderness, the impact of the shear-thinning fluid on the particle's trajectory and dynamics diminishes. On the contrary, the effects of the shear-thinning on lowering of the particle's surface pressure magnitude is proportionally elevated with increasing aspect ratio.

6 CHAPTER SIX

Determination of Steady State Fiber Orientation State based on an Exact Jacobian Newton – Raphson Numerical Scheme

Mechanical performance of printed parts depends on the inherent process-induced microstructural properties. Of the different bead microstructural descriptors for short fiber reinforced composite polymer, the fiber orientation is a key parameter that is useful for accurately predicting the material behavior. As such, appropriate macroscopic modelling of the average fiber orientation distribution is crucial in evaluating these properties. Various analytical models have been developed over time for estimating the average flow induced fiber orientation during polymer composite processing. Traditionally, Jeffery's equation [19] has been used to simulate fiber orientation evolution in dilute fiber suspension. However, Jeffery's model fails when considering semi-dilute or concentrated fiber suspension or confinement lubrication flows involving long and short-range hydrodynamic fiber interaction forces [177]. This has led to the development of more advanced advection-diffusion macroscopic fiber orientation evolution models that account for the neglected effects of momentum diffusion due to inter fiber interactions in concentrated suspensions such as the Folgar-Tucker model [256], [257] or the various variants of the Advani-Tuckers even-order moment tensor model [17], [20]. The preceding chapter (Chapter 5) presented extensive study on the behavior of single particle in dilute viscous homogenous suspension flow with GNF fluid rheology without considering the effects of rotary diffusion due to hydrodynamic fiber interaction forces.

As would appear in subsequent chapter, a novel numerical approach that relates the momentum diffusion phenomenological interaction coefficient and the effective fluid domain radius of influence utilized in the single fiber model is developed to capture the effect of fiber-fiber interaction. The first step involves relating the interaction coefficient with the steady state fiber

orientation tensor using one of the available advection-diffusion fiber orientation tensor evolution models. In a separate study involving a series of single fiber motion FEA simulations, a relation between the fiber stall orientation angles and the effective fluid domain size that results in cessation of the fibers rotary motion due flow disturbance introduced by the hydrodynamic interaction between the fiber wall and the adjacent far-field flow is established. Subsequently, the relationship between the steady state fiber orientation and the fiber interaction coefficient is determined from the established correlations. It is thus apparent that a numerical method for determining the steady state fiber orientation for a range of diffusion interaction coefficients using any of the available fiber orientation evolution models which vary in degree of prediction accuracy is pertinent to the obtaining the relevant relationship which is the focus of the current chapter. The steady state fiber orientation tensor values have traditionally been computed with time evolution numerical IVP-ODE techniques like the famous 4th order Runge-Kutta method or predictor-corrector methods. Here we present a computationally efficient and faster method based on Newton Raphson algorithm for determining the steady state or preferred orientation using explicit derivatives of the 2nd order tensor equation of change with respect to its orientation tensor components for different macroscopic fiber orientation models considering various closure approximations and their performance in complex flow fields. We benchmark results of the explicit derivatives with those obtained using finite differences to ensure accuracy. The explicit derivatives are comparatively faster compared to the finite difference derivatives.

6.1 Determination of Preferred Orientation

The focus of this chapter is to develop a numerical based approach in determining the steady state orientation vector ρ_i or tensor a_{ij} that results in zero rate of change of the orientation state by

employing the iterative numerical Newton Raphson algorithm, setting the rate of change equation or residual to zero. i.e.,

$$\Sigma_i = \frac{D\rho_i}{Dt} = 0, \quad \Sigma_{ij} = \frac{Da_{ij}}{Dt} = 0 \quad (6.1)$$

based on Newton's algorithm, the successive iterative improvement to the approximation of a given root (in our case the orientation state) is given as [320]

$$\rho_i^+ = \rho_i^- - J_{ij}^{-1} \Sigma_j \quad (6.2)$$

and for the 2nd order tensor

$$a_{ij}^+ = a_{ij}^- - J_{mnij}^{-1} \Sigma_{mn}^- \quad (6.3)$$

The implication of this is the need to determine explicit derivatives for the time rate of change of the orientation tensor/vector with respect to its components to obtain the Jacobian. i.e.

$$J_{ij} = \frac{\partial \Sigma_i}{\partial \rho_j}, \quad J_{mnij} = \frac{\partial \Sigma_{mn}}{\partial a_{ij}} \quad (6.4)$$

In the succeeding sections, we present various existing models for rate of change equation of the orientation tensor based on a review by Kugler et al. [20] representing the residual and we derive the associated Jacobian for each model.

6.2 Macroscopic Fiber Orientation Modelling

Macroscopic fiber orientation modelling is usually required in polymers processing to predict the bulk response of chopped fibers in composites parts and ultimately determine part performance. The choice of macroscopic model depends on various processing parameters such as the concentration of fiber suspension, flow type and strength, fiber geometry and volume fraction, material rheology, etc. Our algorithm is based on fiber orientation modelling in the dilute and concentrated regime given the classification of fiber suspension concentration presented in detail in the literature review.

6.2.1 Macroscopic Fiber Orientation Model in Dilute Regime

Jeffery's hydrodynamic model for the motion of a single rigid ellipsoidal particle in incompressible, Newtonian viscous suspension forms the basis for fiber orientation determination in dilute suspension. Jeffery assumed that the particle is convected with bulk motion of the undisturbed surrounding fluid. Jeffery's model is valid for a particle whose linear dimensions multiplied by its velocity pales in comparison to the kinematic viscosity of the fluid. The equation describing Jeffery's motion is given by [19], [20], [254].

$$\dot{\rho}_i^{JF} = \Xi_{ij}\rho_j + \kappa(\Gamma_{ij}\rho_j - \Gamma_{kl}\rho_k\rho_l\rho_i) \quad (6.5)$$

where, Ξ_{ij} and Γ_{ij} are the anti-symmetric and symmetric decomposition of the deformation rate tensor $L_{ij} = \partial \dot{X}_i / \partial X_j$ and can be given respectively as

$$\Xi_{ij} = \frac{1}{2}(L_{ij} - L_{ji}), \quad \Gamma_{ij} = \frac{1}{2}(L_{ij} + L_{ji}) \quad (6.6)$$

Such that $L_{ij} = \Gamma_{ij} + \Xi_{ij}$, κ is a particle shape parameter given as $\kappa = (r_e^2 - 1)/(r_e^2 + 1)$, r_e is the geometric aspect ratio of the particle. The Newton Raphson residual Σ_i for Newtons model is thus:

$$\Sigma_i^{JF} = \dot{\rho}_i^{JF} \quad (6.7)$$

The Jacobian is obtained by taking derivatives with respect to the components of ρ_i , i.e.

$$J_{mn}^{JF} = \frac{\partial \dot{\rho}_m^{JF}}{\partial \rho_n} = \Xi_{mj}\delta_{jn} + \kappa[\Gamma_{mj}\delta_{jn} - \Gamma_{kl}(\delta_{kn}\rho_l\rho_m + \rho_k\delta_{ln}\rho_m + \rho_k\rho_l\delta_{mn})] \quad (6.8)$$

Noting that the derivative of the orientation vector with respect to its individual components is the identity matrix., i.e., $\partial \rho_i / \partial \rho_j = \delta_{ij}$. There are only 2 independent components of the orientation vector, i.e., Σ_i^{JF} is a 2×1 vector. Thus J_{mn}^{JF} is a 2×2 matrix. Jeffery's model has limited application because the polymer melt in the actual injection molding process is non-Newtonian

and the fiber's flexural and fracture properties are significant in contrast with Jefferies model assumption. Moreover, Jeffery's model ignores the effect due to fiber-fiber interaction, hence more elaborate models have been developed by researchers to capture these effects.

6.2.2 Macroscopic Fiber Orientation Model in Concentrated Regime

Various improvements to the Jeffery's single fiber model have been made to model the bulk behavior of fibers in semi-dilute and concentrated suspension due to fiber-fiber interaction. Although theoretically feasible, it is computationally expensive and impractical to simulate the behavior of every individual particle in the fiber suspension flow.

6.2.2.1 The Advani-Tucker's model

The effect of momentum diffusion due to short- and long-range fiber-fiber interaction are accounted for in suspension models in concentrated regime. The Advani and Tucker's moment-tensor equation for the evolution of the average fiber suspension orientation was an extension to the Folger-Tuckers PDF model and the equation of change for the 2nd order orientation tensors in terms of the 2nd and 4th order tensor is given as.

$$\frac{Da_{ij}}{Dt} = \dot{a}_{ij}^{FT} = \{\dot{a}_{ij}^{HD} + \dot{a}_{ij}^{IRD}\} \quad (6.9)$$

$\dot{a}_{ij}^{HD}$ is the hydrodynamic tensor component of the Folger-Tuckers that represents Jeffery's equation and given as

$$\dot{a}_{ij}^{HD} = -(\Xi_{ik}a_{kj} - a_{ik}\Xi_{kj}) + \kappa(\Gamma_{ik}a_{kj} + a_{ik}\Gamma_{kj} - 2\Gamma_{kl}a_{ijkl}) \quad (6.10)$$

and $\dot{a}_{mn}^{IRD}$ is the isotropic rotary diffusion term modelling fiber interaction and is given as

$$\dot{a}_{ij}^{IRD} = 2D_r(\delta_{ij} - \alpha a_{ij}) \quad (6.11)$$

α is a dimension factor, $\alpha = 3$ for 3D orientation and $\alpha = 2$ for 2D planar orientation. The residual for the Folger-Tuckers model is thus.

$$\Sigma_{mn}^{FT} = \dot{a}_{ij}^{FT} \quad (6.12)$$

The associated Jacobian J_{mni}^{FT} is obtained by differentiating the residual with-respect-to components of the 2nd order orientation tensor a_{ij} thus.

$$J_{mni}^{FT} = \frac{\partial \Sigma_{mn}^{FT}}{\partial a_{ij}} = \frac{\partial \dot{a}_{mn}^{HD}}{\partial a_{ij}} + \frac{\partial \dot{a}_{mn}^{IRD}}{\partial a_{ij}} \quad (6.13)$$

The derivative of the 2nd order orientation tensor with respect to its individual components is simply

$$\frac{\partial a_{mn}}{\partial a_{ij}} = \delta_{mi} \delta_{nj} \quad (6.14)$$

where,

$$\frac{\partial \dot{a}_{mn}^{HD}}{\partial a_{ij}} = (-\Xi_{mk} + \kappa \Gamma_{mk}) \delta_{ki} \delta_{nj} + (\Xi_{kn} + \kappa \Gamma_{kn}) \delta_{mi} \delta_{kj} - 2\kappa \Gamma_{kl} \frac{\partial a_{mnkl}}{\partial a_{ij}} \quad (6.15)$$

$$\frac{\partial \dot{a}_{mn}^{IRD}}{\partial a_{ij}} = -2D_r \alpha \delta_{mi} \delta_{nj} \quad (6.16)$$

Different closures approximation for the 4th order tensor and their derivatives have been investigated and discussed in later sections. Since there are only 5 independent components of the 2nd order tensor a_{ij} , in contracted notation, we can represent the residual Σ_{mn}^{FT} as a vector Σ_r^{FT} and the Jacobian J_{mni}^{FT} as a matrix J_{rs}^{FT} . Any reordering convention could be used. Here we employ.

$$r(m, n) = n - \frac{1}{2}(m-1)(m-6), \quad \text{for } n = m \dots 3, \quad \text{for } m = 1, 2 \quad (6.17)$$

The Advani-Tucker's nth order orientation evolution model is less accurate depending on the order of the tensor and thus requires a closure approximation. Due to experimentally observed disparity in the fiber orientation kinetics compared to those computed from traditional orientation models, different model corrections have been proposed to retard the evolution rate.

6.2.2.2 Strain Reduction Factor (SRF) Model

The SRF model was developed by Huynh [321] as an improvement to the FT model where he applied a strain reduction factor $1/\kappa$ directly to the FT model to slow down the orientation kinetics as observed experimentally. He based his premise on a reduced bulk strain of fiber clusters in a concentrated suspension flow. Although the predictions of the steady state orientation based on this model for simple shear flow with suitable determination of κ matched experimental results [322], however it gave an initial overshoot at small strain. The residual and Jacobian in this case is just a multiplication of κ with that previously obtained for the FT model.

$$\Sigma_{mn}^{SRF} = \kappa \Sigma_{mn}^{FT}, \quad J_{mni}^{FT} = \kappa J_{mni}^{FT}, \quad 0 < \kappa < 1 \quad (6.18)$$

The SRF model does not satisfy the rheological test of material non-objectivity and results are dependent on the coordinate system and cannot be applied to complex flows.

6.2.2.3 Reduced Strain Closure (RSC) Model

To address the material non-objectivity drawback of the SRF model, Wang et al. [322] developed a phenomenological reduced strain closure (RSC) model where he applied the reduction factor only to the evolution rate of the spectral decomposed principal directions of the orientation tensor $\underline{\dot{\lambda}}$, without modifying the rate of the rotation $\underline{\dot{\phi}}$ i.e.

$$\dot{\lambda}_i^{RSC} = \kappa \dot{\lambda}_i^{FT}, \quad \dot{\phi}_{ij}^{RSC} = \dot{\phi}_{ij}^{FT}, \quad a_{mn}|a_{mn} = \lambda_i \phi_{mi} \phi_{ni} \quad (6.19)$$

Based on this model, the modified material derivative is thus [322]

$$\dot{a}_{mn}^{RSC} = \dot{a}_{mn}^{FT} - (1 - \kappa) \dot{a}_{mn}^{\Delta FT} \quad (6.20)$$

$$\dot{a}_{mn}^{\Delta FT} = 2\kappa \Gamma_{kl} (\mathbb{L}_{mnkl} - \mathbb{M}_{mnrs} a_{rskl}) + \dot{a}_{mn}^{IRD}$$

where,

$$\mathbb{L}_{mnkl} = \dot{\lambda}_i \phi_{mi} \phi_{ni} \phi_{ki} \phi_{li}, \quad \mathbb{M}_{mnkl} = \phi_{mi} \phi_{ni} \phi_{ki} \phi_{li} \quad (6.21)$$

The Newton Raphson residual is thus $\Sigma_{mn}^{RSC} = \dot{a}_{mn}^{RSC}$ and the Jacobian is obtained by taking partial derivatives thus

$$J_{mni j}^{RSC} = J_{mni j}^{FT} - (1 - k) \frac{\partial \dot{a}_{mn}^{\Delta FT}}{\partial a_{ij}} \quad (6.22)$$

where,

$$\frac{\partial \dot{a}_{mn}^{\Delta FT}}{\partial a_{ij}} = 2\kappa \Gamma_{kl} \frac{\partial}{\partial a_{ij}} \{ \mathbb{L}_{mnkl} - \mathbb{M}_{mnrs} a_{rskl} \} + \frac{\partial \dot{a}_{mn}^{IRD}}{\partial a_{ij}} \quad (6.23)$$

expanding eqn. (6.23) above based on the distributive properties of differentiation we obtain

$$\frac{\partial \dot{a}_{mn}^{\Delta FT}}{\partial a_{ij}} = \kappa \Gamma_{kl} \left[\frac{\partial \mathbb{L}_{mnkl}}{\partial a_{ij}} - a_{rskl} \frac{\partial \mathbb{M}_{mnrs}}{\partial a_{ij}} - \mathbb{M}_{mnrs} \frac{\partial a_{rskl}}{\partial a_{ij}} \right] + \frac{\partial \dot{a}_{mn}^{IRD}}{\partial a_{ij}} \quad (6.24)$$

Applying the product rule of differentiation, we obtain the derivatives of 4th order tensors $\mathbb{M}_{mnkl}$ and $\mathbb{L}_{mnkl}$ respectively.

$$\begin{aligned} \frac{\partial \mathbb{M}_{mnkl}}{\partial a_{rs}} &= \frac{\partial}{\partial a_{rs}} \{ \Phi_{mi} \Phi_{ni} \Phi_{ki} \Phi_{li} \} \\ &= \Phi_{ni} \Phi_{ki} \Phi_{li} \frac{\partial \Phi_{mi}}{\partial a_{rs}} + \Phi_{mi} \Phi_{ki} \Phi_{li} \frac{\partial \Phi_{ni}}{\partial a_{rs}} + \Phi_{mi} \Phi_{ni} \Phi_{li} \frac{\partial \Phi_{ki}}{\partial a_{rs}} \\ &\quad + \Phi_{mi} \Phi_{ni} \Phi_{ki} \frac{\partial \Phi_{li}}{\partial a_{rs}} \end{aligned} \quad (6.25)$$

and

$$\frac{\partial \mathbb{L}_{mnkl}}{\partial a_{rs}} = \Phi_{mi} \Phi_{ni} \Phi_{ki} \Phi_{li} \frac{\partial \dot{\lambda}_i}{\partial a_{rs}} + \dot{\lambda}_i \frac{\partial}{\partial a_{rs}} \{ \Phi_{mi} \Phi_{ni} \Phi_{ki} \Phi_{li} \} \quad (6.26)$$

where the procedure for obtaining the derivatives of the eigenvalues and eigenvectors kindly can be found in [323] APPENDIX C (C.1)

6.2.2.4 Retarding Principal Rate (RPR) Model

Tseng et al. [324], [325] likewise developed a retarding principal rate (RPR) model like the RSC model, to slow down the fiber orientation kinetics based on a coaxial modification to the principal directions of the orientation tensor evolution rate via a nonlinear correlation. The material derivative tensor of any standard model $\dot{a}_{mn}^X$ can be linearly superposed to its RPR correction to slow down the response rate. i.e.

$$\dot{a}_{mn}^{X-RPR} = \dot{a}_{mn}^X + \dot{a}_{mn}^{RPR} \quad (6.27)$$

where the RPR correction $\dot{a}_{mn}^{RPR}$ is given as

$$\dot{a}_{mn}^{RPR} = -\Phi_{mk} \mathbb{A}_{kl}^{IOK} \Phi_{nl}, \quad \mathbb{A}_{kl}^{IOK} = \mathbb{A}_{kl}^{IOK}(\dot{\underline{A}}^X) \quad (6.28)$$

Because the correction is coaxial, the rotation tensor growth rate is unaffected and is obtained from the spectral decomposition of $\dot{a}_{mn}^X$. i.e.,

$$\underline{\underline{\Phi}} \mid \mathbb{A}_{mn}^X = \Phi_{km} \dot{a}_{kl}^X \Phi_{ln} \quad (6.29)$$

The columns of the eigenmatrix obtained in this way are reordered in descending order with the magnitude of the eigenvalues i.e.,

$$\Phi_{ij} = \{ \Phi_{ij} \mid \Lambda_j^X = \mathbb{A}_{jj}^X, \quad \Lambda_1^X \geq \Lambda_2^X \geq \Lambda_3^X \} \quad (6.30)$$

The growth rate of the principal eigenvalues of the standard model $\mathbb{A}_{kl}^X$ is obtained from

$$\mathbb{A}_{kl}^X = \Phi_{km} \dot{a}_{kl}^X \Phi_{ln}, \quad \dot{\Lambda}_k^X = \mathbb{A}_{kk}^X \quad (6.31)$$

The correction to the principal values of the material derivative of the orientation tensor based on the IOK assumption $\mathbb{A}_{kk}^{IOK}$ is defined by a 2-parameter non-linear correlation to the principal values of the standard model $\mathbb{A}_{kl}^X$ such that.

$$\mathbb{A}_{kk}^{IOK} = \dot{\Lambda}_k^{IOK} = \alpha \left[\dot{\Lambda}_k^X - \beta \left(\{\dot{\Lambda}_k^X\}^2 + 2\dot{\Lambda}_l^X \dot{\Lambda}_m^X \right) \right], \quad \mathbb{A}_{kl}^{IOK} \big|_{k \neq l} = 0 \quad (6.32)$$

For an RPR corrected model, the NT residual Σ_{mn}^{X-RPR} is simply the material derivative, i.e.,

$$\Sigma_{mn}^{X-RPR} = \dot{a}_{mn}^{X-RPR} \quad (6.33)$$

and Jacobian J_{mni}^{X-RPR} is given as

$$J_{mni}^{X-RPR} = \frac{\partial \dot{a}_{mn}^X}{\partial a_{ij}} + \frac{\partial \dot{a}_{mn}^{RPR}}{\partial a_{ij}} \quad (6.34)$$

The partial derivative of the RPR correction term $\dot{a}_{mn}^{RPR}$ is given as

$$\frac{\partial \dot{a}_{mn}^{RPR}}{\partial a_{ij}} = - \left\{ \frac{\partial \Phi_{mk}}{\partial a_{ij}} \dot{\mathbb{A}}_{kl}^{IOK} \Phi_{nl} + \Phi_{mk} \frac{\partial \dot{\mathbb{A}}_{kl}^{IOK}}{\partial a_{ij}} \Phi_{nl} + \Phi_{mk} \dot{\mathbb{A}}_{kl}^{IOK} \frac{\partial \Phi_{nl}}{\partial a_{ij}} \right\} \quad (6.35)$$

and the partial derivative of the modified growth rate of eigenvalues tensor $\dot{\mathbb{A}}_{kl}^{IOK}$ is given as

$$\begin{aligned} \frac{\partial \dot{\mathbb{A}}_{kk}^{IOK}}{\partial a_{ij}} &= \frac{\partial \dot{\mathbb{A}}_k^{IOK}}{\partial a_{ij}} = \alpha \left[\frac{\partial \dot{\mathbb{A}}_k^X}{\partial a_{ij}} - 2\beta \left(\dot{\mathbb{A}}_k^X \frac{\partial \dot{\mathbb{A}}_k^X}{\partial a_{ij}} + \frac{\partial \dot{\mathbb{A}}_l^X}{\partial a_{ij}} \dot{\mathbb{A}}_m^X + \dot{\mathbb{A}}_l^X \frac{\partial \dot{\mathbb{A}}_m^X}{\partial a_{ij}} \right) \right], \\ \left. \frac{\partial \dot{\mathbb{A}}_{kl}^{IOK}}{\partial a_{ij}} \right|_{k \neq l} &= 0 \end{aligned} \quad (6.36)$$

where

$$\frac{\partial \dot{\mathbb{A}}_{kl}^X}{\partial a_{ij}} = \left\{ \frac{\partial \Phi_{km}}{\partial a_{ij}} \dot{\mathbb{A}}_{kl}^X \Phi_{ln} + \Phi_{km} \frac{\partial \dot{\mathbb{A}}_{kl}^X}{\partial a_{ij}} \Phi_{ln} + \Phi_{km} \dot{\mathbb{A}}_{kl}^X \frac{\partial \Phi_{ln}}{\partial a_{ij}} \right\}, \quad \frac{\partial \dot{\mathbb{A}}_k^X}{\partial a_{ij}} = \frac{\partial \dot{\mathbb{A}}_{kk}^X}{\partial a_{ij}} \quad (6.37)$$

$\partial \dot{a}_{mn}^X / \partial a_{ij}$ is the partial derivative material derivative of the 2nd order orientation tensor with respect to its components obtained a priori and the partial derivatives of the eigenmatrix with respect to the same (*i.e.*, $\partial \Phi_{mn} / \partial a_{ij}$) can be obtained through any method in [323] (cf. APPENDIX C, C.1).

While the IRD models were experimental observed to be accurate in predicting the orientation state of short-fiber/thermoplastic composites (SFT) with fiber length typically in the range of 0.2mm to 0.4mm [326], they were ineffective in accurately predicting the complete set of orientation tensor components for the long-fiber/thermoplastic composites (LFT) with typical size between 10mm to 13mm which was the motivation for ARD model development. For long-

fiber/thermoplastic composites (LFT), the IRD models possess unidirectional prediction effectiveness. Different researchers have proposed models that involve modifying the rotary diffusion term for an all-round competency in accurately predicting the components of the orientation tensor. Ranganathan et al. [327] assumed an isotropic rotary diffusivity that inversely varies with the degree of alignment of the orientation tensor with a phenomenological interaction parameter that depends on the reciprocal of the inter fiber spacing. Their model application was limited to the transient orientation state and suited for long range fiber-fiber interaction. Their model was however unsuitable for LFTs steady state orientation prediction as with other IRD models since its diffusivity was isotropic.

6.2.2.5 Anisotropic Rotary Diffusion (ARD) Models

Various ARD models with different modifications have been developed based on the definition of the spatial diffusion tensor. Fan et al. [312] and Phan-Thien [260], were the first to propose an anisotropic rotary diffusion (ARD) moment-tensor model by substituting the scalar phenomenological interaction parameter with a second order anisotropic rotary diffusion tensor. Their model was however exclusive and restricted in application. At the same time, Koch [328] developed an ARD model suited for semi-dilute suspension with an anisotropic spatial diffusion tensor that depends on the orientation state and the rate of deformation tensor. However, their model was based on the more complicated PDF form for the orientation tensor representation rather than the moment-tensor form and proved ineffective in LFT modelling. Phelps et. al [326] built on the work of Fan [312] and Phan-Thien et al. [260] and developed a more general moment-tensor anisotropic diffusion model that depends on the spatial diffusion tensor and orientation tensor state. The derivation of the spatial diffusion tensor was based on similar representation by Hand [329] as a function of the orientation state and rate of deformation tensor. Phelps's model

had remarkable improvements in predicting orientation state of LFTs. Most recent models utilize the moment-tensor form for the ARD representation developed by Phelps and Tucker [326]. The general expression for the 2nd order orientation tensor evolution rate is a linear combination of the Jeffery's model and the rotary diffusion term given as

$$\dot{\mathbf{a}}_{mn}^{PT} = \dot{\mathbf{a}}_{mn}^{HD} + \dot{\mathbf{a}}_{mn}^{ARD} \quad (6.38)$$

where the rotary diffusion term $\dot{\mathbf{a}}_{mn}^{ARD}$ is defined in terms of the spatial diffusion coefficient and the orientation state and is given as

$$\dot{\mathbf{a}}_{mn}^{ARD} = \dot{\gamma} [2\mathbb{C}_{mn} - 2\mathbb{C}_{rs}\delta_{rs}\mathbf{a}_{mn} - 5(\mathbb{C}_{mk}\mathbf{a}_{kn} + \mathbf{a}_{mk}\mathbb{C}_{kn}) + 10\mathbf{a}_{mnkl}\mathbb{C}_{kl}] \quad (6.39)$$

and $\mathbb{C}$ is the spatial diffusion tensor. Based on this model, the NT residual and Jacobian are respectively given as

$$\Sigma_{mn}^{PT} = \dot{\mathbf{a}}_{mn}^{PT} \quad (6.40)$$

$$J_{mnij}^{PT} = \frac{\partial \dot{\mathbf{a}}_{mn}^{PT}}{\partial \mathbf{a}_{ij}} = \frac{\partial \dot{\mathbf{a}}_{mn}^{HD}}{\partial \mathbf{a}_{ij}} + \frac{\partial \dot{\mathbf{a}}_{mn}^{ARD}}{\partial \mathbf{a}_{ij}} \quad (6.41)$$

Where the derivative of the rotary diffusion (ARD) term is obtained using product rule as

$$\begin{aligned} \frac{\partial \dot{\mathbf{a}}_{mn}^{ARD}}{\partial \mathbf{a}_{ij}} = \dot{\gamma} & \left[2 \frac{\partial \mathbb{C}_{mn}}{\partial \mathbf{a}_{ij}} - 2 \left(\frac{\partial \mathbb{C}_{rs}}{\partial \mathbf{a}_{ij}} \delta_{rs} \mathbf{a}_{mn} + \mathbb{C}_{rs} \delta_{rs} \delta_{mi} \delta_{nj} \right) + \dots \right. \\ & - 5 \left(\frac{\partial \mathbb{C}_{mk}}{\partial \mathbf{a}_{ij}} \mathbf{a}_{kn} + \mathbb{C}_{mk} \delta_{ki} \delta_{nj} + \delta_{mi} \delta_{kj} \mathbb{C}_{kn} + \mathbf{a}_{mk} \frac{\partial \mathbb{C}_{kn}}{\partial \mathbf{a}_{ij}} \right) + \dots \\ & \left. + 10 \left(\frac{\partial \mathbf{a}_{mnkl}}{\partial \mathbf{a}_{ij}} \mathbb{C}_{kl} + \mathbf{a}_{mnkl} \frac{\partial \mathbb{C}_{kl}}{\partial \mathbf{a}_{ij}} \right) \right] \quad (6.42) \end{aligned}$$

Bakharev [330] proposed a moldflow rotational diffusion (MRD) model based on reduction of the terms of the generic ARD model by Phelps to just linear terms with a spatial diffusion tensor like

Tseng's model. In the mold-flow ARD (*mARD*) model developed by Bakharev [330], the Phelps & Tucker's rotary diffusion (*ARD*) expression is truncated to include just the linear terms. i.e.

$$\dot{a}_{mn}^{mARD} = \dot{\gamma}[2\mathbb{C}_{mn} - 2\mathbb{C}_{kl}\delta_{kl}a_{mn}] \quad (6.43)$$

$$\frac{\partial \dot{a}_{mn}^{mARD}}{\partial a_{ij}} = \dot{\gamma} \left[2 \frac{\partial \mathbb{C}_{mn}}{\partial a_{ij}} - 2 \left(\frac{\partial \mathbb{C}_{kl}}{\partial a_{ij}} \delta_{kl} a_{mn} + \mathbb{C}_{kl} \delta_{kl} \delta_{mi} \delta_{nj} \right) \right] \quad (6.44)$$

The corresponding evolution rate equation for the 2nd order orientation tensor based on *mARD* model is given as

$$\dot{a}_{mn}^{mPT} = \dot{a}_{mn}^{HD} + \dot{a}_{mn}^{mARD} \quad (6.45)$$

Various models for the spatial diffusion coefficient $\mathbb{C}_{mn}$ used in the ARD model have been developed by various researchers. The basic representation of $\mathbb{C}_{mn}$ by Phelps and Tucker [326] based on a modification of Hand's anisotropic tensor [329] is given as a function of the rate of deformation tensor and orientation state as

$$\mathbb{C}_{mn}^{PT} = b_1 \delta_{mn} + b_2 a_{mn} + b_3 a_{mk} a_{nk} + \frac{b_4}{\dot{\gamma}} \Gamma_{mn} + \frac{b_5}{\dot{\gamma}^2} \Gamma_{mk} \Gamma_{nk} \quad (6.46)$$

where b_i are dimensionless constants obtained from regression analysis of experimental data. For this model, the derivative of the $\mathbb{C}_{mn}^{PT}$ with respect to a_{ij} is given as

$$\frac{\partial \mathbb{C}_{mn}^{PT}}{\partial a_{ij}} = b_2 \delta_{mi} \delta_{nj} + b_3 (\delta_{mi} \delta_{kj} a_{nk} + a_{mk} \delta_{ni} \delta_{kj}) \quad (6.47)$$

The sensitivity of the PT model parameters b_i to ensure numerical stability of the model response coupled with the complicated process involved in their determination were the major limitations to this model application. Tseng et al. [331] developed an improved anisotropic rotary diffusion model (*iARD*) based on a definition of a two-parameter spatial diffusion tensor model in terms of the rate of deformation tensor that couples the effect of fiber-matrix interaction and fiber-fiber interaction given as

$$\mathbb{C}_{mn}^{iARD} = C_I \left(\delta_{mn} - 4C_M \frac{\Gamma_{mk}\Gamma_{nk}}{\dot{\gamma}^2} \right) \quad (6.48)$$

where C_I & C_M are the fiber-fiber and fiber-matrix interaction parameters respectively. An alternate definition is given as

$$\mathbb{C}_{mn}^{iARD} = C_I (\delta_{mn} - C_M \tilde{L}_{mn}), \quad \tilde{L}_{mn} = (L_{mk}L_{nk})/(L_{rs}L_{rs}) \quad (6.49)$$

The derivative of the spatial diffusion tensor with respect to the 2nd order orientation tensor is simply zero. i.e.,

$$\frac{\partial \mathbb{C}_{mn}^{iARD}}{\partial a_{ij}} = 0 \quad (6.50)$$

Because of the material non-objectivity of the rate of deformation tensor L_{mn} used in the definition of the spatial diffusion tensor $\mathbb{C}_{mn}$ in the *iARD* model, Tseng et. al [332] developed an improved objective principal spatial tensor ARD model (*pARD*) that corotates with the orientation tensor given as

$$\mathbb{C}_{mn}^{pARD} = \left\{ C_I \Phi_{mk} \mathbb{D}_{kl} \Phi_{nl}, \quad \underline{\underline{\Phi}} \mid a_{mn} = \Phi_{mk} \mathbb{A}_{kl} \Phi_{nl} \right\} \quad (6.51)$$

Where the tensor $\mathbb{D}_{kl}$ contains only diagonal terms and its trace is unity. i.e.

$$\mathbb{D}_{kl} \delta_{kl} = \mathbb{D}_{kk} = 1, \quad D_{kl} \mid_{k \neq l} = 0 \quad (6.52)$$

The derivative of $\mathbb{C}_{mn}^{pARD}$ with respect to the 2nd order orientation tensor is given as

$$\frac{\partial \mathbb{C}_{mn}^{pARD}}{\partial a_{ij}} = C_I \left\{ \frac{\partial \Phi_{mk}}{\partial a_{ij}} \mathbb{D}_{kl} \Phi_{nl} + \Phi_{mk} \mathbb{D}_{kl} \frac{\partial \Phi_{nl}}{\partial a_{ij}} \right\} \quad (6.53)$$

Another ARD model reduction suggested by Wang [333] called the WPT model involved truncating the terms of the PT model to just the first and third term such that,

$$\mathbb{C}_{mn}^{WPT} = b_1 \delta_{mn} + b_3 a_{mk} a_{nk} \quad (6.54)$$

Falvoro et al. [254] provided an alternative form of the spatial diffusion tensor where he replaced the coefficients with a weighted superposition of the interaction coefficient, i.e.

$$\mathbb{C}_{mn}^{WPT} = C_I((1 - w)\delta_{mn} + wa_{mk}a_{nk}) \quad (6.55)$$

where w is the weighting factor. The derivative of $\mathbb{C}_{mn}^{pARD}$ with respect to the 2nd order orientation tensor is given as

$$\frac{\partial \mathbb{C}_{mn}^{WPT}}{\partial a_{ij}} = wC_I(\delta_{mk}a_{nk} + a_{mk}\delta_{nk}) \quad (6.56)$$

Lastly, we consider the D_z ARD model development (cf. Falvoro et al. [254]) by Moldflow for simulating 2.5D flow processes. Their model is defined in terms of the interaction coefficient, C_I , a moment of interaction thickness parameter D_z , and the unit normal to the mold surface $\hat{n}$. The expression for $\mathbb{C}_{mn}$ here is given as

$$\mathbb{C}_{mn}^{Dz} = C_I^{Dz}(\delta_{mn} - (1 - D_z)\hat{n}_m\hat{n}_n) \quad (6.57)$$

and

$$\frac{\partial \mathbb{C}_{mn}^{Dz}}{\partial a_{ij}} = 0 \quad (6.58)$$

6.2.2.6 Nematic Potential (NEM) Model

Latz et al. [334] developed a fully coupled flow-orientation tensor model for concentrated suspensions utilizing a two-parameter nematic potential (*NEM*) effective collision ARD tensor for the diffusion term that couples the phenomenological effect of the momentum diffusion due to fiber-fiber interaction and a topological interaction effect of diffusion due to an exclusion volume mechanism. i.e.

$$\dot{\mathbf{a}}_{mn}^{IRD-MS} = \dot{\gamma}[C_I(\delta_{mn} - \alpha a_{mn}) + U_0(a_{mk}a_{kn} - a_{kl}a_{mnkl})] \quad (6.59)$$

where U_0 is the ‘Onsager’ nematic topological interaction coefficient of the Maier-Saupe potential. Typically, for stability, $U_0 \leq 4C_I$ for 2D analysis and $U_0 > 8C_I$ for 3D analysis. The material derivative of the 2nd order orientation tensor based on the nematic diffusion model is thus given as

$$\dot{a}_{mn}^{nem} = \dot{a}_{mn}^{HD} + \dot{a}_{mn}^{IRD-MS} \quad (6.60)$$

The NT residual and Jacobian are respectively given as

$$\Sigma_{mn}^{nem} = \dot{a}_{mn}^{nem} \quad (6.61)$$

$$J_{mni}^{nem} = \frac{\partial \dot{a}_{mn}^{HD}}{\partial a_{ij}} + \frac{\partial \dot{a}_{mn}^{IRD-MS}}{\partial a_{ij}} \quad (6.62)$$

where the derivative of the nematic diffusion term is given as

$$\begin{aligned} \frac{\partial \dot{a}_{mn}^{IRD-MS}}{\partial a_{ij}} = \dot{\gamma} & \left[-C_I \alpha \delta_{mi} \delta_{nj} \right. \\ & \left. + U_0 \left(\delta_{mi} \delta_{kj} a_{kn} + a_{mk} \delta_{ki} \delta_{nj} - \delta_{ki} \delta_{lj} a_{mnkl} - a_{kl} \frac{\partial a_{mnkl}}{\partial a_{ij}} \right) \right] \end{aligned} \quad (6.63)$$

Latz et al. [334] found the influence of the topological interaction on the fiber orientation state to be flow dependent having significant effect on channel and contraction flow with relatively lesser influence on flow around cylinder. Kugler et al. [20], Favaloro et al. [254], Agboola et al. [335] and Park et al. [336] presents detailed review and comparison of existing fiber orientation models. The foregoing ARD models find useful application in polymer composite industry and have been incorporated in mold-filling flow computations in injection molding process simulations [218], [219], [337], [338], [339], [340], [341]. Most commercial software used in simulation of the injection molding process such as Autodesk Moldflow, Moldex3D usually combines multiple models in predicting the orientation state for improved accuracy. One such combination is the ARD-RSC models whose material derivative is expressed as

$$\dot{a}_{mn}^{pARD-RSC} = \dot{a}_{mn}^{RSC} - k \dot{a}_{mn}^{IRD} + \dot{a}_{mn}^{ARD} + \dot{a}_{mn}^{\Delta RSC} \quad (6.64)$$

where,

$$\dot{a}_{mn}^{\Delta RSC} = -2\dot{\gamma}(1 - k)[M_{mnkl} - \delta_{kl}a_{mn} - 5(L_{mnkl} - M_{mnrs}a_{rskl})]C_{kl} \quad (6.65)$$

and $\dot{a}_{mn}^{RSC}$, $\dot{a}_{mn}^{IRD}$, $\dot{a}_{mn}^{ARD}$ have been defined in preceding sections. In this case the NT residual $\Sigma_{mn}^{pARD-RSC}$ is the material derivative $\dot{a}_{mn}^{pARD-RSC}$, i.e.,

$$\Sigma_{mn}^{pARD-RSC} = \dot{a}_{mn}^{pARD-RSC} \quad (6.66)$$

And the Jacobian is obtained by taking partial derivatives with respect to the 2nd order tensor as usual and can be expressed as

$$J_{mnij}^{pARD-RSC} = \frac{\partial \dot{a}_{mn}^{RSC}}{\partial a_{ij}} - k \frac{\partial \dot{a}_{mn}^{IRD}}{\partial a_{ij}} + \frac{\partial \dot{a}_{mn}^{ARD}}{\partial a_{ij}} + \frac{\partial \dot{a}_{mn}^{\Delta RSC}}{\partial a_{ij}} \quad (6.67)$$

where

$$\frac{\partial \dot{a}_{mn}^{\Delta RSC}}{\partial a_{ij}} = -2\dot{\gamma}(1 - k) \left\{ \left[\frac{\partial \mathbb{M}_{mnkl}}{\partial a_{ij}} - \delta_{kl}\delta_{mi}\delta_{nj} - 5 \frac{\partial}{\partial a_{ij}} \{ \mathbb{L}_{mnkl} - \mathbb{M}_{mnrs}a_{rskl} \} \right] \mathbb{C}_{kl} + \dots \right\} \quad (6.68)$$

$$\left[\mathbb{M}_{mnkl} - \delta_{kl}a_{mn} - 5(\mathbb{L}_{mnkl} - \mathbb{M}_{mnrs}a_{rskl}) \right] \frac{\partial \mathbb{C}_{kl}}{\partial a_{ij}} \right\}$$

All terms of the partial derivatives have been previously derived in preceding section.

6.2.3 Closure Approximations and Their Explicit Derivatives

Due to the absence of exact solutions for orientation state for inhomogeneous flows involving momentum diffusion, various closure approximations with different degree of accuracy have been developed for higher orders of the moment-tensor fiber orientation equation. Derivatives of the orientation tensor closure approximation are used in the Newton-Raphson iteration method to compute the steady-state fiber orientation tensor state. These derivatives for the various closure approximation used in this study appear below.

6.2.3.1 Quadratic Closure Approximation

The quadratic closure, $\tilde{a}_4$ was introduced by Doi [342] and Lipscomb [242] and defined as dyadic product of the 2nd order orientation tensor. We denote the quadratic closure approximate $\tilde{a}_4$ and is mathematically given as

$$\tilde{a}_4 = \tilde{a}_{ijkl} = a_2 a_2 = a_{ij} a_{kl} \quad (6.69)$$

The derivative of $\tilde{a}_4$ above with respect to the 2nd order tensor a_{mn} is simply.

$$\frac{\partial \tilde{a}_{ijkl}}{\partial a_{mn}} = \frac{\partial a_{ij}}{\partial a_{mn}} a_{kl} + a_{ij} \frac{\partial a_{kl}}{\partial a_{mn}} = \delta_{im} \delta_{jn} a_{kl} + a_{ij} \delta_{km} \delta_{ln} \quad (6.70)$$

6.2.3.2 Linear Closure Approximation

The linear 4th order orientation tensor closure approximation, $\hat{a}_4$ was first proposed by Hand [329] using all the products of a_2 and δ is given as

$$\begin{aligned} \hat{a}_4 = \hat{a}_{ijkl} = & -h_1 (\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \\ & + h_2 (a_{ij} \delta_{kl} + a_{ik} \delta_{jl} + a_{il} \delta_{jk} + \delta_{ij} a_{kl} + \delta_{ik} a_{jl} + \delta_{il} a_{jk}) \end{aligned} \quad (6.71)$$

The derivative of $\hat{a}_4$ above with respect to components of the 2nd order tensor a_{mn} is given as

$$\begin{aligned} \frac{\partial \hat{a}_{ijkl}}{\partial a_{mn}} = & h_2 (\delta_{im} \delta_{jn} \delta_{kl} + \delta_{im} \delta_{kn} \delta_{jl} + \delta_{im} \delta_{ln} \delta_{jk} + \delta_{im} \delta_{jn} \delta_{kl} + \delta_{im} \delta_{kn} \delta_{jl} \\ & + \delta_{im} \delta_{ln} \delta_{jk}) \end{aligned} \quad (6.72)$$

where h_1 and h_2 are numerical factors which varies based on spatial dimensionality and given in

Table 6.1 below

Table 6.1: Numerical factors of the linear closure		
	<i>Solid (3D)</i>	<i>Planar (2D)</i>
h_1	1/35	1/24
h_2	1/7	1/6

The QDR closure inherently lacks symmetry property requirement but preserves the symmetry of the computed lower order tensor. The LIN closures are exact for random orientation distribution while the QDR closures are exact for uniaxially aligned fiber orientation.

6.2.3.3 Hybrid Closure Approximation

The hybrid closure approximation, a_4 is simply a weighted combination of both linear $\hat{a}_{ijkl}$ and quadratic $\tilde{a}_{ijkl}$ closure approximation above by some scalar measure of orientation f given as [17]

$$a_4 = a_{ijkl} = f\tilde{a}_{ijkl} + (1 - f)\hat{a}_{ijkl} \quad (6.73)$$

where f is a generalization of Herman's Orientation factor. Advani & Tucker [17] proposed an appropriate approximation of the weighting factor as an invariant of the orientation state given as $f = a_f a_{ij} a_{ji} - b_f$, where a_f and b_f are constants that depends on the spatial dimension given in Table 6.2 below

	Table 6.2: Constants of the hybrid closure	
	<i>Solid (3D)</i>	<i>Planar (2D)</i>
a_f	3/2	2
b_f	1/2	1

the derivative of the hybrid closure approximation a_4 with respect to components of the 2nd order tensor a_{mn} is given as

$$\frac{\partial a_{ijkl}}{\partial a_{mn}} = f \frac{\partial \tilde{a}_{ijkl}}{\partial a_{mn}} + (1 - f) \frac{\partial \hat{a}_{ijkl}}{\partial a_{mn}} + \frac{\partial f}{\partial a_{mn}} (\tilde{a}_{ijkl} - \hat{a}_{ijkl}) \quad (6.74)$$

where,

$$\frac{\partial f}{\partial a_{mn}} = a_f (\delta_{im} \delta_{jn} a_{ji} + a_{ij} \delta_{jm} \delta_{in}) \quad (6.75)$$

An alternative estimation of the factor f by Advani & Tucker [17] is given as

$$f = 1 - \alpha^\alpha e_{ijk} a_{i1} a_{j2} a_{k3} \quad (6.76)$$

$$\frac{\partial f}{\partial a_{mn}} = -\alpha^\alpha e_{ijk} \{ \delta_{im} \delta_{1n} a_{j2} a_{k3} + a_{i1} \delta_{jm} \delta_{2n} a_{k3} + a_{i1} a_{j2} \delta_{km} \delta_{3n} \} \quad (6.77)$$

The hybrid model is observed to perform better for transient state orientation prediction; however, the hybrid closure tends to over-predict the orientation tensor compared with the more accurate

distribution function closure (DFC). DFC are however computationally involved since they require finite difference grid in space and time.

6.2.3.4 Hinch and Leal Closure Approximation

Hinch and Leal [241] developed numerous composite closure approximations for the 4th order tensor in precontracted forms with the deformation rate tensor and the accuracy of their predictions were dependent on flow type and strength. The Hinch and Leal closure approximations were not explicit expressions of the 4th order orientation tensor a_{ijkl} but were in contracted form with the deformation rate tensor i.e., $\gamma_{kl}a_{ijkl}$. Advani and Tucker developed a general explicit expression of a_{ijkl} (eqn. (6.78)) summarizing all the Hinch and Leal closures forms given as

$$a_{ijkl} = \beta_1(\delta_{ij}\delta_{kl}) + \beta_2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \beta_3(\delta_{ij}a_{kl} + a_{ij}\delta_{kl}) + \beta_4(a_{ik}\delta_{jl} + \dots + a_{jl}\delta_{ik} + a_{il}\delta_{jk} + a_{jk}\delta_{il}) + \beta_5(a_{ij}a_{kl}) + \beta_6(a_{ik}a_{jl} + a_{il}a_{jk}) + \dots + \beta_7(\delta_{ij}a_{km}a_{ml} + a_{im}a_{mj}\delta_{kl}) + \beta_8(a_{im}a_{mj}a_{kn}a_{nl}) \quad (6.78)$$

and the partial derivative of the above expression with respect to components of the 2nd order orientation tensor a_{rs} based on product rule is given as

$$\begin{aligned} \frac{\partial a_{ijkl}}{\partial a_{rs}} = & \left[\frac{\partial \beta_1}{\partial a_{rs}}(\delta_{ij}\delta_{kl}) + \frac{\partial \beta_2}{\partial a_{rs}}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{\partial \beta_3}{\partial a_{rs}}(\delta_{ij}a_{kl} + a_{ij}\delta_{kl}) \right. \\ & + \frac{\partial \beta_4}{\partial a_{rs}}(a_{ik}\delta_{jl} + a_{jl}\delta_{ik} + a_{il}\delta_{jk} + a_{jk}\delta_{il}) + \frac{\partial \beta_5}{\partial a_{rs}}(a_{ij}a_{kl}) \\ & + \frac{\partial \beta_6}{\partial a_{rs}}(a_{ik}a_{jl} + a_{il}a_{jk}) + \frac{\partial \beta_7}{\partial a_{rs}}(\delta_{ij}a_{km}a_{ml} + a_{im}a_{mj}\delta_{kl}) \\ & \left. + \frac{\partial \beta_8}{\partial a_{rs}}(a_{im}a_{mj}a_{kn}a_{nl}) \right] \\ & + \left[\beta_3(\delta_{ij}\delta_{kr}\delta_{ls} + \delta_{ir}\delta_{js}\delta_{kl}) \right. \\ & + \beta_4(\delta_{ir}\delta_{ks}\delta_{jl} + \delta_{jr}\delta_{ls}\delta_{ik} + \delta_{ir}\delta_{ls}\delta_{jk} + \delta_{jr}\delta_{ks}\delta_{il}) \\ & + \beta_5(\delta_{ir}\delta_{js}a_{kl} + a_{ij}\delta_{kr}\delta_{ls}) \\ & + \beta_6(\delta_{ir}\delta_{ks}a_{jl} + a_{ik}\delta_{jr}\delta_{ls} + \delta_{ir}\delta_{ls}a_{jk} + a_{il}\delta_{jr}\delta_{ks}) \\ & + \beta_7(\delta_{ij}\delta_{kr}\delta_{ms}a_{ml} + \delta_{ij}a_{km}\delta_{mr}\delta_{ls} + \delta_{ir}\delta_{ms}a_{mj}\delta_{kl} + a_{im}\delta_{mr}\delta_{js}\delta_{kl}) \\ & + \beta_8(\delta_{ir}\delta_{ms}a_{mj}a_{kn}a_{nl} + a_{im}\delta_{mr}\delta_{js}a_{kn}a_{nl} + a_{im}a_{mj}\delta_{kr}\delta_{ns}a_{nl} \\ & \left. + a_{im}a_{mj}a_{kn}\delta_{nr}\delta_{ls}) \right] \end{aligned} \quad (6.79)$$

Mullens [343] provided a summary Table (cf. Table 6.3) for the β_i factors of the Hinch and Leal closures subdivided into weak flow (WF - Isotropic, Linear and Quadratic), strong flow (SF), and Hinch and Leal composite flows (HL – HL1&HL2) closure forms.

Table 6.3: Summary of the Hinch and Leal closure β_i factors for the different flow classifications

		β_1	β_2	β_3	β_4	β_5	β_6	β_7	β_8
	ISO	$\frac{1}{15}$	$\frac{1}{15}$	...	...	...	...	...	...
WF	LIN	$-\frac{1}{35}$	$-\frac{1}{35}$	$\frac{1}{7}$	$\frac{1}{7}$	...	...	...	...
	QDR	...	...	...	...	1	...	...	...
SF	SF2	...	...	...	...	1	1	...	$-\frac{2}{\langle a^2 \rangle}$
	HL1	...	...	$\frac{2}{5}$	...	$-\frac{1}{5}$	$\frac{3}{5}$	$-\frac{2}{5}$	...
HL	HL2	$\frac{26}{315}\alpha$	$\frac{26}{315}\alpha$	$\frac{16}{63}\alpha$	$-\frac{4}{21}\alpha$	1	1	...	$-\frac{2}{\langle a^2 \rangle}$

where the parameters $\langle a^2 \rangle$ and α are respectively

$$\langle a^2 \rangle = a_{ij}a_{ji}, \quad \alpha = \exp \left[2 \frac{1 - 3\langle a^2 \rangle}{1 - \langle a^2 \rangle} \right] \quad (6.80)$$

and the partial derivatives are respectively given as

$$\begin{aligned} \frac{\partial \langle a^2 \rangle}{\partial a_{rs}} &= \delta_{ir}\delta_{js}a_{ji} + a_{ij}\delta_{jr}\delta_{is}, \quad \frac{\partial \alpha}{\partial a_{rs}} = -\frac{4\alpha}{(1 - \langle a^2 \rangle)^2} \frac{\partial \langle a^2 \rangle}{\partial a_{rs}}, \quad \frac{\partial}{\partial a_{rs}} \left\{ \frac{k}{\langle a^2 \rangle} \right\} \\ &= -\frac{k}{\langle a^2 \rangle^2} \frac{\partial \langle a^2 \rangle}{\partial a_{rs}} \end{aligned} \quad (6.81)$$

Recently, more accurate higher order polynomial closure approximations have been developed including the eigenvalue-based fitting (EBF) that involves principal axis transformation and the Invariant-based fitting (IBF).

6.2.3.5 Eigenvalue based Orthotropic Fitted (EBF) Closure Approximations

The idea of orthotropic closure approximations for the 4th order tensor was to impose objectivity such that the approximation is independent of the coordinate frame selection. In

essence, the principal axes of the closure approximation and second order tensor must coincide. The orthotropic smooth (ORS), orthotropic fitted (ORF) closures and ORF closures for low fiber-fiber interaction coefficient (ORL) fall under the class of EBF closures and were developed by Cintra and Tucker [316]. The (9x9) term 4th order tensor can be represented in (6 x 6) contracted notation like in structural analysis of composite material based on symmetry property. i.e.

$$A_{rs} = a_{ijkl} \quad (6.82)$$

where, the index of the contracted notation is related to the index notation according to

$$r = \begin{cases} i = j & \delta_{ij} = 1 \\ (9 - i - j) & \delta_{ij} = 0 \end{cases} \quad \& \quad s = \begin{cases} k = l & \delta_{kl} = 1 \\ (9 - k - l) & \delta_{kl} = 0 \end{cases} \quad (6.83)$$

The derivative of the 4th order tensor with respect to the 2nd order tensor is such that

$$\frac{\partial A_{rs}}{\partial a_{mn}} = \frac{\partial a_{ijkl}}{\partial a_{mn}} \quad (6.84)$$

Symmetry property of the 4th order tensor requires $a_{ijkl} = a_{klij}$ which implies that $A_{rs} = A_{sr}$.

The contracted tensor A_{rs} transformed to the principal axes has the orthotropic form $\bar{A}_{rs}$ given thus.

$$\underline{\bar{A}} = \begin{bmatrix} \bar{A}_{11} & \bar{A}_{12} & \bar{A}_{13} & & & \\ \bar{A}_{21} & \bar{A}_{22} & \bar{A}_{23} & & & \\ \bar{A}_{31} & \bar{A}_{32} & \bar{A}_{33} & & & \\ & & & \bar{A}_{44} & & \\ & & & & \bar{A}_{55} & \\ & & & & & \bar{A}_{66} \end{bmatrix} \quad (6.85)$$

The contracted tensor transforms from its principal reference frame to the original coordinate axes according to

$$A_{rs} = \mathcal{V}_{ri} \mathcal{V}_{sj} \bar{A}_{ij} \quad (6.86)$$

The 6x6 transformation matrix $\mathcal{V}_{ij}$ is given as $\mathcal{V}_{ij} = \mathbb{U}_{im} \mathbb{U}_{mn} \mathbb{U}_{nj}^{-1}$, where $\mathbb{U}_{ij} = k\delta_{ij}$, $k = \begin{cases} 1 & i \leq 3 \\ 2 & i > 3 \end{cases}$ and $\mathbb{U}_{rs} = \Phi_{ik} \Phi_{jl} + (1 - \delta_{kl}) \Phi_{jk} \Phi_{il}$. The modal matrix $\underline{\underline{\Phi}}$ whose k^{th} column are the corresponding eigenvectors $\underline{x}^k$ of eigenvalues $\lambda_k = \mathbb{A}_{kk}$ is obtained from the spectral decomposition of $\mathbb{a}_{ij}$ is such that:

$$\underline{\underline{\Phi}} | \mathbb{a}_{mn} = \Phi_{mk} \mathbb{A}_{kl} \Phi_{nl} \quad (6.87)$$

The indices of the contracted 4th order modal tensor $\mathbb{U}_{rs}$ relates to the those of the 2nd order modal matrix $\underline{\underline{\Phi}}$ according to above equation. A more direct way is to reconstruct the 4th order orientation tensor $\bar{\mathbb{a}}_{mnpq}$ from the contracted form A_{rs} and transform from the principal reference frame to the original axes according to eqn. (6.88) below.

$$\mathbb{a}_{ijkl} = \Phi_{im} \Phi_{jn} \Phi_{kp} \Phi_{lq} \bar{\mathbb{a}}_{mnpq} \quad (6.88)$$

and using the product rule

$$\begin{aligned} \frac{\partial \mathbb{a}_{ijkl}}{\partial \mathbb{a}_{rs}} &= \Phi_{im} \Phi_{jn} \Phi_{kp} \Phi_{lq} \frac{\partial \bar{\mathbb{a}}_{mnpq}}{\partial \mathbb{a}_{rs}} \\ &+ \left(\frac{\partial \Phi_{im}}{\partial \mathbb{a}_{rs}} \Phi_{jn} \Phi_{kp} \Phi_{lq} + \Phi_{im} \frac{\partial \Phi_{jn}}{\partial \mathbb{a}_{rs}} \Phi_{kp} \Phi_{lq} + \Phi_{im} \Phi_{jn} \frac{\partial \Phi_{kp}}{\partial \mathbb{a}_{rs}} \Phi_{lq} \right. \\ &\left. + \Phi_{im} \Phi_{jn} \Phi_{kp} \frac{\partial \Phi_{lq}}{\partial \mathbb{a}_{rs}} \right) \bar{\mathbb{a}}_{mnpq} \end{aligned} \quad (6.89)$$

Derivative of the eigentensor $\underline{\underline{\Phi}}$ can be found in [323] (cf. APPENDIX C, C.1). Symmetry requirements of the transformed orthotropic tensor reduces the total number of independent non-zero components to 9, and additional special symmetry properties of the exact 4th order tensor requires that $\bar{\mathbb{a}}_{ijkl} = \bar{\mathbb{a}}_{kjil} = \bar{\mathbb{a}}_{ljki} = \bar{\mathbb{a}}_{ikjl} = \bar{\mathbb{a}}_{iljk}$ reduces the non-zero independent components to the 6 diagonal terms. i.e.

$$\bar{A}_{ij} = \bar{A}_{kk} \quad \{k : k = 9 - i - j, \quad i \neq j\} \quad (6.90)$$

The normalization property $\mathbb{a}_{ijkk} = \mathbb{a}_{ij}$ of the exact 4th order tensor further requires that:

$$\begin{bmatrix} \bar{A}_{44} \\ \bar{A}_{55} \\ \bar{A}_{66} \end{bmatrix} = \underline{\underline{\beta}}^{-1} \left\{ \begin{bmatrix} \lambda_1 \\ \lambda_2 \\ \lambda_3 \end{bmatrix} - \begin{bmatrix} \bar{A}_{11} \\ \bar{A}_{22} \\ \bar{A}_{33} \end{bmatrix} \right\} \quad (6.91)$$

where, λ_i are the eigenvalues of the 2nd order orientation tensor a_{ij} , $\sum_i \lambda_i = 1$ and $\beta_{ij} = 1 - \delta_{ij}$. Based on the foregoing conditions, the only three surviving non-zero independent terms are $\bar{A}_{11}$, $\bar{A}_{22}$ & $\bar{A}_{33}$. The general form for orthotropic closure is to express the three surviving non-zero independent components ($\bar{A}_{11}$, $\bar{A}_{22}$, $\bar{A}_{33}$) of the contracted 4th order tensor in the principal reference frame after imposing all symmetric and normalization conditions of the exact 4th order tensor, as a scalar function $F_k(\lambda_1, \lambda_2)$ of the two largest eigenvalues (λ_1, λ_2) of the 2nd order tensor. Most fitted closures take the form of an n^{th} -order binomial function in λ_1 & λ_2 to represents the scalar function i.e.,

$$\bar{A}_{kk} = F_k(\lambda_1, \lambda_2) = f_k^{(n)}(\lambda_1, \lambda_2), \quad \lambda_1 \geq \lambda_2 \geq \lambda_3, \quad k = 1, 2, 3 \quad (6.92)$$

Polynomial order exceeding $n \geq 4$ fall under the class of eigenvalue based optimal fitting (EBOF) closures. Generally, we can represent the function $f_k^{(n)}$ as a tensor product of a constant coefficient matrix $\underline{\underline{\mathfrak{C}}}^{(n)}$ and a n^{th} order permuted bivariate polynomial vector $\underline{\underline{\mathbb{A}}}^{(n)} = \underline{\underline{\mathbb{A}}}^{(n)}(\lambda_1, \lambda_2)$, i.e.

$$f^{(n)}(\lambda_1, \lambda_2) = \underline{\underline{\mathfrak{C}}}^{(n)} \underline{\underline{\mathbb{A}}}^{(n)}(\lambda_1, \lambda_2) = \mathfrak{C}_{kj}^{(n)} \mathbb{A}_j^{(n)}(\lambda_1, \lambda_2) \quad (6.93)$$

Different representation of $\underline{\underline{\mathfrak{C}}}^{(n)}$ and $\underline{\underline{\mathbb{A}}}^{(n)}$ depending on the polynomial order fit (n) can be found in APPENDIX C, C.2 . The derivative of the components of the orthotropic closure with respect to the 2nd order tensor are thus:

$$\frac{\partial \bar{A}_{kk}}{\partial a_{rs}} = \frac{\partial \bar{A}_k}{\partial a_{rs}} = \mathfrak{C}_{kj}^{(n)} \frac{\partial \mathbb{A}_j^{(n)}}{\partial a_{rs}} = \mathfrak{C}_{kj}^{(n)} \mathbb{A}'_{jrs} = \mathfrak{C}_{kj}^{(n)} \tilde{\mathbb{A}}_{jl}^{(n)} \lambda'_{lrs}, \quad k = 1, 2, 3, \quad l = 1, 2 \quad (6.94)$$

The n th order binomial permutation vector $\underline{\mathbb{A}}^{(n)}$ and its derivative coefficient matrix $\underline{\widetilde{\mathbb{A}}}^{(n)}$ for the quadratic closure are given from terms of binomial expansion respectively as

$$\left\{ \begin{array}{l} \mathbb{A}_k^{(n)}(\lambda_1, \lambda_2) = \lambda_1^{i-j} \lambda_2^j \\ \widetilde{\mathbb{A}}_{kl}^{(n)} = \frac{\partial \mathbb{A}_k^{(n)}}{\partial \lambda_l} = \begin{cases} (i-j) \cdot \lambda_1^{i-j-1} \lambda_2^j & l = 1, k|k = j + \frac{1}{2}i(i+1), j = 0 \cdots i, i = 0 \cdots n \\ j \cdot \lambda_1^{i-j} \lambda_2^{j-1} & l = 2 \end{cases} \end{array} \right. \quad (6.95)$$

For a special case of orthotropic fitted closure called rational ellipsoid closure (REC) by Wetzel and Tucker [344], the scalar function for the 3 independent tensor component is given as

$$F(\lambda_1, \lambda_2) = \frac{f^{(n)}(\lambda_1, \lambda_2)}{f^{(m)}(\lambda_1, \lambda_2)} \quad (6.96)$$

The derivative of the components of the above with respect to the 2nd order tensor based on the quotient rule is thus:

$$\frac{\partial \bar{A}_{kk}}{\partial a_{rs}} = \frac{1}{[f^{(m)}]^2} \left[f^{(m)} \frac{\partial f^{(n)}}{\partial a_{rs}} - f^{(n)} \frac{\partial f^{(m)}}{\partial a_{rs}} \right] \quad (6.97)$$

From normalization condition of the 4th order tensor, we obtain for the derivative of $\bar{A}_{kk}$, ($k = 4, 5, 6$)

$$\frac{\partial \bar{A}_{kk}}{\partial a_{rs}} = \beta_{ki}^{-1} \left\{ \frac{\partial \lambda_i}{\partial a_{rs}} - \frac{\partial \bar{A}_{ii}}{\partial a_{rs}} \right\}, \quad \frac{\partial \lambda_i}{\partial a_{rs}} = \varepsilon_{il} \lambda'_{lrs}, \quad \underline{\underline{\varepsilon}} = \begin{bmatrix} 1 & 0 & -1 \\ 0 & 1 & -1 \end{bmatrix}^T \quad (6.98)$$

$$i = 1, 2, 3, \quad l = 1, 2$$

For the partial derivative of the eigenvalues with respect to the components of the 2nd order orientation tensor, kindly refer to APPENDIX C, C.1.

6.2.3.6 Invariant Based Optimal Fitting Closure (IBOF) Approximations

EBF closures are computationally more involved in numerical calculations of actual flows because of the principal axis transformation. Of the class of IBF closures, the natural (NAT) closure approximation of Verleye and Dupret [345] was built on the work of Lipscomb et al. [242]

and formed the basis of other IBF developments. They developed a general expression for the full symmetric 4th order tensor in terms of the 2nd order tensor, the identity matrix and fitted coefficients as functions of the tensor invariant which were derived from analytical calculations based on a least square fitting process. The NAT closure assumed the absence of fiber-fiber interaction and infinitely long fiber geometry. The closure is exact based on the foregoing assumptions however it has been reported to possess singularities for axisymmetric orientation states. The IBOF closure approximation was developed by Chung et al. [346] and combined the qualities of the natural closure representation of the 4th order closure approximation by Verleye & Dupret [345] and optimal fitting of invariants of the 4th order tensor based on actual flow data obtained from distribution function (DFC) to obtain unknown coefficients similar to the orthotropic fitted closures by Cintra and Tucker [316]. In contracted form the 4th order tensor based on symmetry properties is given as

$$\underline{\underline{A}} = \begin{bmatrix} A_{11} & A_{12} & A_{13} & A_{14} & A_{15} & A_{16} \\ & A_{22} & A_{23} & A_{24} & A_{25} & A_{26} \\ & & A_{33} & A_{34} & A_{35} & A_{36} \\ & & & A_{44} & A_{45} & A_{46} \\ & & & & A_{55} & A_{56} \\ \dots Sym & & & & & A_{66} \end{bmatrix} \quad (6.99)$$

Based on special symmetry requirement.

$$\begin{aligned} A_{44} &= A_{23}, & A_{45} &= A_{36}, & A_{46} &= A_{25}, & A_{55} &= A_{13}, & A_{56} &= A_{14}, \\ A_{66} &= A_{12} \end{aligned} \quad (6.100)$$

and from the normalization condition

$$\sum_{n=1}^3 A_{nm} = a_m, \quad a_m = a_{ij}, \quad m = \begin{cases} i=j & i=j \\ 9-i-j & i \neq j \end{cases} \quad (6.101)$$

Or more explicitly we derive the sets of equations in eqn. (6.102) below.

$$A_{11} + A_{12} + A_{13} = a_{11} \quad A_{12} + A_{22} + A_{23} = a_{22} \quad A_{13} + A_{23} + A_{33} = a_{33}$$

$$A_{14} + A_{24} + A_{34} = a_{23} \quad A_{15} + A_{25} + A_{35} = a_{13} \quad A_{16} + A_{26} + A_{36} = a_{12} \quad (6.102)$$

Taking partial derivatives of eqns. (6.100) & (6.101) we obtain in indicial representation.

$$\frac{\partial A_{mn}}{\partial a_{rs}} = \frac{\partial A_{ij}}{\partial a_{rs}}, \quad \&, \quad \sum_{n=1}^3 \frac{\partial A_{nm}}{\partial a_{rs}} = \frac{\partial a_m}{\partial a_{rs}} \quad (6.103)$$

There are thus only 9 independent components for the 4th order tensor. The IBOF is developed in terms of the full symmetric 4th order expansion of a_{ijkl} as a combination of the 2nd order tensor a_{ij} and identity matrix δ_{kl} based on Cayley-Hamilton theory is given as

$$a_{ijkl} = \beta_1 \mathbb{S}(\delta_{ij} \delta_{kl}) + \beta_2 \mathbb{S}(\delta_{ij} a_{kl}) + \beta_3 \mathbb{S}(a_{ij} a_{kl}) + \beta_4 \mathbb{S}(\delta_{ij} a_{km} a_{ml}) + \beta_5 \mathbb{S}(a_{ij} a_{km} a_{ml}) + \beta_6 \mathbb{S}(a_{im} a_{mj} a_{kn} a_{nl}) \quad (6.104)$$

where the S operator represents the symmetric permutation expansion of its argument, for example,

$$\begin{aligned} \mathbb{S}(\mathbb{T}_{ijkl}) = \frac{1}{24} [& \mathbb{T}_{ijkl} + \mathbb{T}_{ijlk} + \mathbb{T}_{ikjl} + \mathbb{T}_{iklj} + \mathbb{T}_{iljk} + \mathbb{T}_{ilkj} + \mathbb{T}_{jilk} + \mathbb{T}_{jilk} \\ & + \mathbb{T}_{jkil} + \mathbb{T}_{jkli} + \mathbb{T}_{jlki} + \mathbb{T}_{jlki} + \mathbb{T}_{kijl} + \mathbb{T}_{kilj} + \mathbb{T}_{kjil} + \mathbb{T}_{kjli} \\ & + \mathbb{T}_{klji} + \mathbb{T}_{klji} + \mathbb{T}_{lijk} + \mathbb{T}_{likj} + \mathbb{T}_{ljik} + \mathbb{T}_{ljki} + \mathbb{T}_{lki} \\ & + \mathbb{T}_{lkji}] \end{aligned} \quad (6.105)$$

We obtain the derivative of the 4th order tensor with respect to components of 2nd order tensor by product rule thus.

$$\begin{aligned} \frac{\partial}{\partial a_{rs}} \{a_{ijkl}\} = & \left[\frac{\partial \beta_1}{\partial a_{rs}} \mathbb{S}(\delta_{ij} \delta_{kl}) + \frac{\partial \beta_2}{\partial a_{rs}} \mathbb{S}(\delta_{ij} a_{kl}) + \frac{\partial \beta_3}{\partial a_{rs}} \mathbb{S}(a_{ij} a_{kl}) \right. \\ & + \frac{\partial \beta_4}{\partial a_{rs}} \mathbb{S}(\delta_{ij} a_{km} a_{ml}) + \frac{\partial \beta_5}{\partial a_{rs}} \mathbb{S}(a_{ij} a_{km} a_{ml}) \\ & \left. + \frac{\partial \beta_6}{\partial a_{rs}} \mathbb{S}(a_{im} a_{mj} a_{kn} a_{nl}) \right] + \dots \\ & + \left[\beta_2 \mathbb{S}(\delta_{ij} \delta_{kr} \delta_{ls}) + \beta_3 \{ \mathbb{S}(\delta_{ir} \delta_{js} a_{kl}) + \mathbb{S}(a_{ij} \delta_{kr} \delta_{ls}) \} \right. \\ & + \beta_4 \{ \mathbb{S}(\delta_{ij} \delta_{kr} \delta_{ms} a_{ml}) + \mathbb{S}(\delta_{ij} a_{km} \delta_{mr} \delta_{ls}) \} \\ & + \beta_5 \{ \mathbb{S}(\delta_{ir} \delta_{js} a_{km} a_{ml}) + \mathbb{S}(a_{ij} \delta_{kr} \delta_{ms} a_{ml}) + \mathbb{S}(a_{ij} a_{km} \delta_{mr} \delta_{ls}) \} \\ & + \beta_6 \{ \mathbb{S}(\delta_{ir} \delta_{ms} a_{mj} a_{kn} a_{nl}) + \mathbb{S}(a_{im} \delta_{mr} \delta_{js} a_{kn} a_{nl}) \\ & \left. + \mathbb{S}(a_{im} a_{mj} \delta_{kr} \delta_{ns} a_{nl}) + \mathbb{S}(a_{im} a_{mj} a_{kn} \delta_{nr} \delta_{ls}) \} \right] \end{aligned} \quad (6.106)$$

The β_i coefficients are expressed as functions of the second and third invariants (II & III) of the 2nd order tensor a_{ij} . Based on normalization condition and full symmetry requirement coupled with the Cayley-Hamilton theorem, there remains only 3 independent coefficients to determine.

The expressions for the IBOF dependent coefficients ($\beta_1, \beta_2, \beta_5$) are given as

$$\begin{aligned}\beta_1 &= \frac{3}{5} \left[-\frac{1}{7} + \frac{1}{5} \beta_3 \left(\frac{1}{7} + \frac{4}{7} \text{II} + \frac{8}{3} \text{III} \right) - \beta_4 \left(\frac{1}{5} - \frac{8}{15} \text{II} - \frac{14}{15} \text{III} \right) + \dots \right. \\ &\quad \left. - \beta_6 \left(\frac{1}{35} - \frac{4}{35} \text{II} - \frac{24}{105} \text{III} + \frac{16}{15} \text{II III} + \frac{8}{35} \text{II}^2 \right) \right] \\ \beta_2 &= \frac{6}{7} \left[1 - \frac{1}{5} \beta_3 (1 + 4\text{II}) + \frac{7}{5} \beta_4 \left(\frac{1}{6} - \text{II} \right) - \beta_6 \left(-\frac{1}{5} + \frac{4}{5} \text{II} + \frac{2}{3} \text{III} - \frac{8}{5} \text{II}^2 \right) \right] \\ \beta_5 &= -\frac{4}{5} \beta_3 - \frac{7}{5} \beta_4 - \frac{6}{5} \beta_6 \left(1 - \frac{4}{3} \text{II} \right)\end{aligned}\tag{6.107}$$

We obtain the explicit derivatives of the dependent coefficients via the product rule thus

$$\begin{aligned}\frac{\partial \beta_1}{\partial a_{rs}} &= \frac{3}{5} \left[\frac{1}{5} \frac{\partial \beta_3}{\partial a_{rs}} \left(\frac{1}{7} + \frac{4}{7} \text{II} + \frac{8}{3} \text{III} \right) - \frac{\partial \beta_4}{\partial a_{rs}} \left(\frac{1}{5} - \frac{8}{15} \text{II} - \frac{14}{15} \text{III} \right) + \dots \right. \\ &\quad \left. - \frac{\partial \beta_6}{\partial a_{rs}} \left(\frac{1}{35} - \frac{4}{35} \text{II} - \frac{24}{105} \text{III} + \frac{16}{15} \text{II III} + \frac{8}{35} \text{II}^2 \right) \right] + \dots + \frac{3}{5} \left[\frac{4}{35} \beta_3 - \beta_4 \right. \\ &\quad \left. + \beta_6 \left(\frac{4}{35} - \frac{16}{35} \text{II} - \frac{16}{15} \text{III} \right) \right] \frac{\partial \text{II}}{\partial a_{rs}} \\ &\quad \left. + \left[\frac{1}{5} \frac{8}{3} \beta_3 + \frac{14}{15} \beta_4 + \beta_6 \left(\frac{24}{105} - \frac{16}{15} \text{II} \right) \right] \frac{\partial \text{III}}{\partial a_{rs}} \right] \\ \frac{\partial \beta_2}{\partial a_{rs}} &= \frac{6}{7} \left[-\frac{1}{5} \frac{\partial \beta_3}{\partial a_{rs}} (1 + 4\text{II}) + \frac{7}{5} \frac{\partial \beta_4}{\partial a_{rs}} \left(\frac{1}{6} - \text{II} \right) - \frac{\partial \beta_6}{\partial a_{rs}} \left(-\frac{1}{5} + \frac{4}{5} \text{II} + \frac{2}{3} \text{III} - \frac{8}{5} \text{II}^2 \right) \right] \\ &\quad + \frac{6}{7} \left[-\frac{1}{5} [4\beta_3 + 7\beta_4 + \beta_6(4 - 16\text{II})] \frac{\partial \text{II}}{\partial a_{rs}} - \frac{2}{3} \beta_6 \frac{\partial \text{III}}{\partial a_{rs}} \right] \\ \beta_5 &= -\frac{4}{5} \frac{\partial \beta_3}{\partial a_{rs}} - \frac{7}{5} \frac{\partial \beta_4}{\partial a_{rs}} - \frac{6}{5} \frac{\partial \beta_6}{\partial a_{rs}} \left(1 - \frac{4}{3} \text{II} \right) + \frac{8}{5} \beta_6 \frac{\partial \text{II}}{\partial a_{rs}}\end{aligned}\tag{6.108}$$

The independent coefficients ($\beta_3, \beta_4, \beta_6$) by Chung et al. [346] were obtained from a 5th order binomial fitted function in terms of II & III thus:

$$\beta_m = \sum_{i=0}^5 \sum_{j=0}^i a_k^m \cdot \text{II}^{i-j} \text{III}^j, \quad k = j + \frac{1}{2} i(i+1)\tag{6.109}$$

Where the coefficients of the binomial terms can be found in Table C. 1 (APPENDIX C). The non-unity invariants of a_2 are respectively given as

$$\text{II} = \lambda_1 \lambda_2 + \lambda_2 \lambda_3 + \lambda_3 \lambda_1, \quad \text{III} = \lambda_1 \lambda_2 \lambda_3 \quad (6.110)$$

The derivative of the independent coefficient with respect to the components of the 2nd order tensor is

$$\frac{\partial \beta_m}{\partial a_{rs}} = \sum_{i=0}^5 \sum_{j=0}^i a_k^m \left\{ (i-j) \cdot \text{II}^{i-j-1} \text{III}^j \frac{\partial \text{II}}{\partial a_{rs}} + j \cdot \text{II}^{i-j-1} \text{III}^{j-1} \frac{\partial \text{III}}{\partial a_{rs}} \right\} \quad (6.111)$$

where,

$$\begin{aligned} \frac{\partial \text{II}}{\partial a_{rs}} &= (\lambda_2 + \lambda_3) \frac{\partial \lambda_1}{\partial a_{rs}} + (\lambda_1 + \lambda_3) \frac{\partial \lambda_2}{\partial a_{rs}} + (\lambda_1 + \lambda_2) \frac{\partial \lambda_3}{\partial a_{rs}} \\ \frac{\partial \text{III}}{\partial a_{rs}} &= (\lambda_2 \lambda_3) \frac{\partial \lambda_1}{\partial a_{rs}} + (\lambda_1 \lambda_3) \frac{\partial \lambda_2}{\partial a_{rs}} + (\lambda_1 \lambda_2) \frac{\partial \lambda_3}{\partial a_{rs}} \end{aligned} \quad (6.112)$$

Other highly accurate closure approximations include the neural network based fitted closures by Jack et al. [347] and the 6th order Invariant based orthotropic fitted closure by Jack [18], [348].

6.3 Error Estimate

The performance of the Newton-Raphson (NR) method in accurately predicting the steady-state values of the 2nd order orientation tensor component, is accessed based on the relative absolute error between results of the focus NR method and a reference method, in this case the explicit 4th order Runge-Kutta (RK4) numerical method. We define the error percent as

$$err = \frac{a_{mn}^{NR} - a_{mn}^{ref}}{a_{mn}^{ref}} \times 100\% \quad (6.113)$$

6.4 Results and Discussion

We present results of validation carried out for the derived partial derivatives of material derivative for the 2nd order tensor with respect to its components for each model and closure approximations discussed in preceding sections using finite differences. We also present result of the validation for the steady state orientation obtained using the Newton Raphson method by

comparing with those obtained using the explicit 4th order Runge-Kutta ODE method. Validation exercise is carried out for different flow conditions.

6.4.1 Validation of Derivatives based on Finite Difference Approximation

The results of the validation based on comparison of the Jacobian obtained with the exact derivative to the finite difference approximation is presented below. We present the error defined as the Euclidean norm of the difference between the results obtained from both methods. i.e.

$$err = \left\| \underline{J}^{exact} - \underline{J}^{FD} \right\|_2 \quad (6.114)$$

The central difference finite difference approximation is used according to

$$J_{mni}^{FD} = \frac{\Sigma_{mn}(a_{ij} + \delta a_{ij}) - \Sigma_{mn}(a_{ij} - \delta a_{ij})}{2\delta a_{ij}} + O(\delta^2) \quad (6.115)$$

The model parameters used here can be found in Table 6.7. The results of the error are shown for different models and closure approximations below. We assume for this validation exercise a ‘randomly’ generated orientation state $\underline{a}^0$ given below:

$$\underline{a}^0 = \begin{bmatrix} 0.0622 & 0.0765 & 0.0398 \\ 0.0765 & 0.5521 & 0.0186 \\ 0.0398 & 0.0186 & 0.3857 \end{bmatrix}$$

Table 6.4: Result of error obtained for different evolution models and different permutation closure approximations.

	HYB_1	HYB_2	ISO	LIN	QDR	SF2	HL1	HL2
FT	6.4360e-09	9.3849e-09	2.2195e-09	4.1879e-09	2.6914e-09	2.0949e-08	9.6178e-09	4.3940e-08
PT	8.0880e-09	7.5487e-09	5.8367e-09	5.0032e-09	4.2438e-09	1.6776e-08	8.2414e-09	3.4809e-08
iARD	5.7369e-09	1.2712e-08	3.4444e-09	6.3363e-09	5.7281e-09	5.1004e-09	8.7737e-09	1.6148e-08
pARD	7.1691e-09	5.4745e-09	2.7221e-09	2.4378e-09	4.1547e-09	1.4805e-08	9.8182e-09	3.6185e-08
WPT	8.5633e-09	1.0386e-08	3.7731e-09	2.5253e-09	2.9256e-09	1.4543e-08	9.6324e-09	3.5284e-08
Dz	5.8987e-09	8.2475e-09	2.3728e-09	5.4843e-09	3.2334e-09	7.1374e-09	1.0594e-08	2.7732e-08
NEM	6.4903e-09	9.3064e-09	4.0116e-09	4.3136e-09	1.6118e-09	2.1012e-08	9.8464e-09	4.4062e-08
pARD-RSC	1.0030e-08	1.3343e-08	1.3062e-08	1.0506e-08	1.3699e-08	5.3778e-09	1.3441e-08	1.6482e-08
iARD-RPR	5.6451e-09	6.8998e-09	3.4777e-09	3.6868e-09	5.2224e-09	1.0731e-08	1.0324e-08	2.0512e-08

Table 6.5: Result of error obtained for different evolution models and different orthotropic fitted and IBOF closure approximations.

	IBOF	ORS	ORT	NAT_1	ORW	NAT_2
FT	6.1748e-07	4.5725e-08	3.3508e-08	3.5570e-08	5.9942e-08	5.8124e-08
PT	5.6437e-07	3.3269e-08	2.5174e-08	3.0756e-08	5.6934e-08	3.9459e-08
iARD	4.4942e-07	2.1292e-08	1.9339e-08	2.5119e-08	4.7618e-08	2.6629e-08
pARD	5.5458e-07	3.2158e-08	2.4788e-08	2.9749e-08	5.3542e-08	3.9564e-08
WPT	5.6412e-07	3.4300e-08	2.6217e-08	3.0395e-08	5.7198e-08	4.0679e-08
Dz	6.4226e-07	2.8050e-08	2.9196e-08	3.5323e-08	6.7044e-08	3.2757e-08
NEM	6.1978e-07	4.6150e-08	3.3879e-08	3.6486e-08	6.0619e-08	5.8685e-08
pARD-RSC	4.1821e-07	2.0742e-08	1.8016e-08	2.5292e-08	4.7718e-08	2.7998e-08
iARD-RPR	3.4882e-07	1.6010e-08	1.4040e-08	1.9636e-08	3.6873e-08	1.7086e-08

Table 6.6: Result of error obtained for different evolution models and different EBOF closure approximations.

	WTZ	LAR32	ORW3	VST	FFLAR4	LAR4
FT	4.3147e-07	5.0800e-07	6.4960e-08	3.1567e-07	4.3188e-07	4.3101e-07
PT	4.0286e-07	4.7162e-07	5.2842e-08	2.9443e-07	4.0435e-07	3.9967e-07
iARD	3.0776e-07	3.6782e-07	4.2132e-08	2.2662e-07	3.0115e-07	3.0665e-07
pARD	3.8741e-07	4.5415e-07	5.2287e-08	2.8548e-07	3.8500e-07	3.8818e-07
WPT	4.0391e-07	4.7234e-07	5.6117e-08	2.9350e-07	4.0486e-07	3.9851e-07
Dz	4.8010e-07	5.5454e-07	6.0103e-08	3.3924e-07	4.8016e-07	4.6691e-07
NEM	4.3254e-07	5.0936e-07	6.5200e-08	3.1653e-07	4.3271e-07	4.3182e-07
pARD-RSC	2.9401e-07	3.4878e-07	4.1548e-08	2.1358e-07	2.9236e-07	2.9070e-07
iARD-RPR	2.4509e-07	2.9082e-07	2.8202e-08	1.7359e-07	2.3590e-07	2.4110e-07

6.4.2 Validation using explicit 4th-order Runge-Kutta (RK4) Method

In this section, results for the steady state values of the preferred orientation states obtained for various cases using the Newton Raphson algorithm are compared to those obtained based on the 4th order explicit Runge-Kutta method. Three (3) sample cases were studied here, the first set of models are based on study by Falvoro et al. [254] and the two (2) other model set were based on study by Tseng et al. [324]. The EBOF closure approximation of Verweyst [341] has been utilized for all analysis. The following data have been used for the different models considered in the first case study [254].

Table 6.7: Case Study 1 parameters for the *FT*, *Dz*, *iARD*, *pARD*, *WPT*, *MRD* and *PT* models [254]

	C_I	ARD Parameters
<i>FT</i>	0.0311	-
<i>Dz</i>	0.0258	$D_z = 0.051, \hat{n} = [0 \ 0 \ 1]$
<i>iARD</i>	0.0562	$C_M = 0.9977$
<i>pARD</i>	0.0169	$\Omega = 0.9868$
<i>WPT</i>	0.0504	$w = 0.9950$
<i>MRD</i>	0.0198	$\begin{bmatrix} D_1 & D_2 & D_3 \\ 1.000 & 0.7946 & 0.0120 \end{bmatrix}$
-		$\begin{bmatrix} b_1 & b_2 & b_3 & b_4 & b_5 \\ 1.924 & 58.39 & 400 & 0.1168 & 0 \end{bmatrix}$
<i>PT</i>		$\times 10^{-4}$

A random orientation state was considered for the initial tensor in the RK4 while for the NR method we consider an initial guess value $\underline{\underline{a}}^0$ for the 2nd order orientation tensor below.

$$\underline{\underline{a}}^0 = \begin{bmatrix} 0.30 & 0.00 & 0.00 \\ 0.00 & 0.60 & 0.10 \\ 0.00 & 0.10 & 0.10 \end{bmatrix}$$

The transient profiles for the component of the 2nd order orientation tensor based on RK4 method for the models presented in Table 6.7 are shown in Figure 6.1 below.

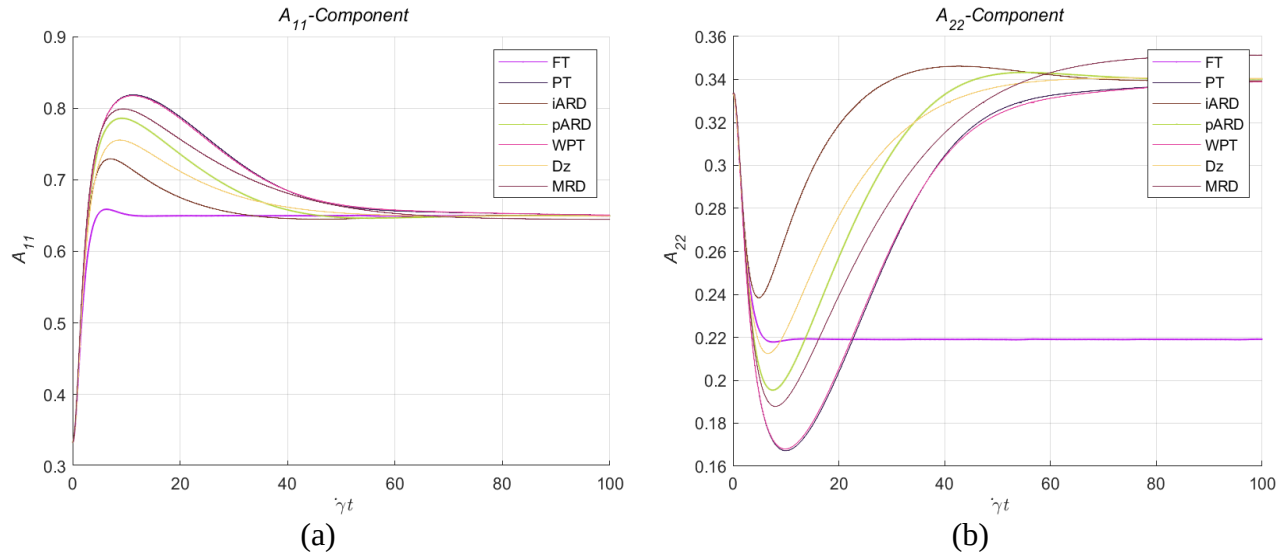


Figure 6.1: Time evolution of the 2nd order orientation tensor for calibrated FT, PT, iARD, pARD, WPT, *Dz* and MRD models for (a) a_{11} component (b) a_{22} component.

Table 6.8 below shows result of the error estimate of the steady state values of the orientation tensor components obtained by the NR method for the various models considered using the RK4 values as reference. From the result we see the NR predictions possess good accuracy.

Table 6.8: Error estimates of the a_{11} , a_{22} & a_{13} steady-state orientation tensor component values for FT, Dz, iARD, pARD, WPT, MRD and PT models

	<u>A_11</u>	<u>A_22</u>	<u>A_13</u>
FT	0.0014	0.0009	0.0053
PT	0.0038	0.0022	0.0067
iARD	0.0032	0.0015	0.0033
pARD	0.0073	0.0035	0.0534
WPT	0.0026	0.0015	0.0099
Dz	0.0297	0.0155	0.0086
MRD	0.0017	0.0009	0.0000

The second case study is based on work by Tseng et al. [324], the calibrated data based on the different model improvements for slow orientation kinetics which they utilized are presented in Table 6.9 below.

Table 6.9: Case Study 2 parameters for the *FT*, *SRF*, *RSC* and *RPR* models [324]

	<i>FT</i>	<i>SRF</i>	<i>RSC</i>	<i>RPR</i>
C_I	0.01	0.01	0.01	0.01
k_ξ	—	0.1	0.1	—
α	—	—	—	0.9
β	—	—	—	0

A random orientation state was used as the starting orientation for the RK4 analysis while the initial guess $\underline{\underline{a}}^0$ given below was used for the Newton Raphson method.

$$\underline{\underline{a}}^0 = \begin{bmatrix} 0.35 & 0.00 & 0.00 \\ 0.00 & 0.55 & 0.10 \\ 0.00 & 0.10 & 0.10 \end{bmatrix}$$

Two flow cases were considered:

1. Simple shear flow in the 1-2 plane, $L_{12} = \dot{\gamma}$ (L1).

- Balanced shear/planar-elongation flow, simple shear in 1-2 plane superimposed on planar elongation in 1-2 plane. $L_{11} = -\dot{\epsilon}, L_{22} = \dot{\epsilon}, L_{12} = \dot{\gamma}$ given $\dot{\gamma}/\dot{\epsilon} = 10$ (L2).

The time evolution of the components of the 2nd order orientation tensor based on the RK4 method are shown in Figure 6.2 below.

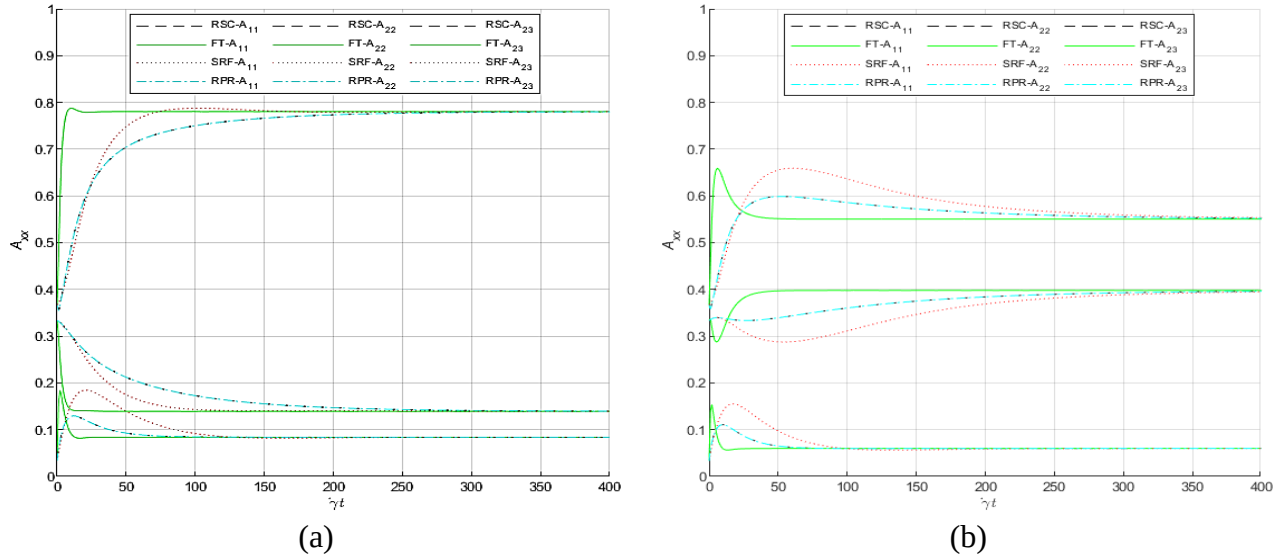


Figure 6.2: Time evolution of the a_{11} , a_{22} & a_{13} components of the 2nd order orientation tensor for calibrated FT, SRF, RSC and FT-RPR models for (a) simple shear flow and (b) shearing/stretching combination flow.

The percentage error estimate between the NR steady state values and the reference RK4 values are presented in Table 6.10 below. Results show a high level in accuracy in prediction based on the NR method.

Table 6.10: Error estimates of the a_{11} , a_{22} & a_{13} steady-state orientation tensor component values for RSC, FT, SRF, and RPR models and for the 2 different flow fields (L1 & L2)

	<u>A_11_L1</u>	<u>A_22_L1</u>	<u>A_13_L1</u>	<u>A_11_L2</u>	<u>A_22_L2</u>	<u>A_13_L2</u>
RSC	0.0000	0.0000	0.0000	0.0003	0.0007	0.0167
FT	0.0022	0.0021	0.0263	0.0005	0.0009	0.0017
SRF	0.0022	0.0013	0.0048	0.0075	0.0085	0.0167
RPR	0.0000	0.0000	0.0000	0.0000	0.0002	0.0033

In the third case, we consider more complex model development usually involving the combination of two models typically found in injection molding simulation packages such as

Moldex3D. The different cases are based on [324] and the model parameter used for the analysis are given in Table 6.11, & Table 6.12 below. We assume a random initial orientation state for the reference RK4 method and the same initial guess as with case study 2 for the NR method. The result of the steady state values based on the RK4 method for the different methods are shown in Figure 6.3. The percentage error estimate of the NR steady state values with respect to the RK4 reference values are given in Table 6.13 and the results show negligible discrepancy in values obtained.

The results shown in Table 6.13 reveals good performance in terms of accuracy for the NR method based on the calculated error estimates of the steady state orientation values for the 3-tensor components and for the various models.

Table 6.11: ARD-RSC Parameters [324]

	40 wt. % glass-fiber/PP	31 wt. % carbon-fiber/PP	40 wt. % glass-fiber/nylon
k	1/30	1/30	1/20
b_1	3.842×10^{-4}	3.728×10^{-3}	4.643×10^{-4}
b_2	-1.786×10^{-3}	-1.695×10^{-2}	-6.169×10^{-4}
b_3	5.250×10^{-2}	1.750×10^{-1}	1.900×10^{-2}
b_4	1.168×10^{-5}	-3.367×10^{-3}	9.650×10^{-4}
b_5	-5.000×10^{-4}	-1.000×10^{-2}	7.000×10^{-4}

Table 6.12: *iARD-RPR* & *pARD-RPR* Parameters [324]

	40 wt. % glass-fiber/PP	31 wt. % carbon-fiber/PP	40 wt. % glass-fiber/nylon
C_I	0.0165	0.0630	0.0060
C_M	0.9990	1.0100	0.9000
Ω	0.9880	0.9650	0.9000
α	0.9650	0.9650	0.9500
β	0.0000	0.0000	0.0000

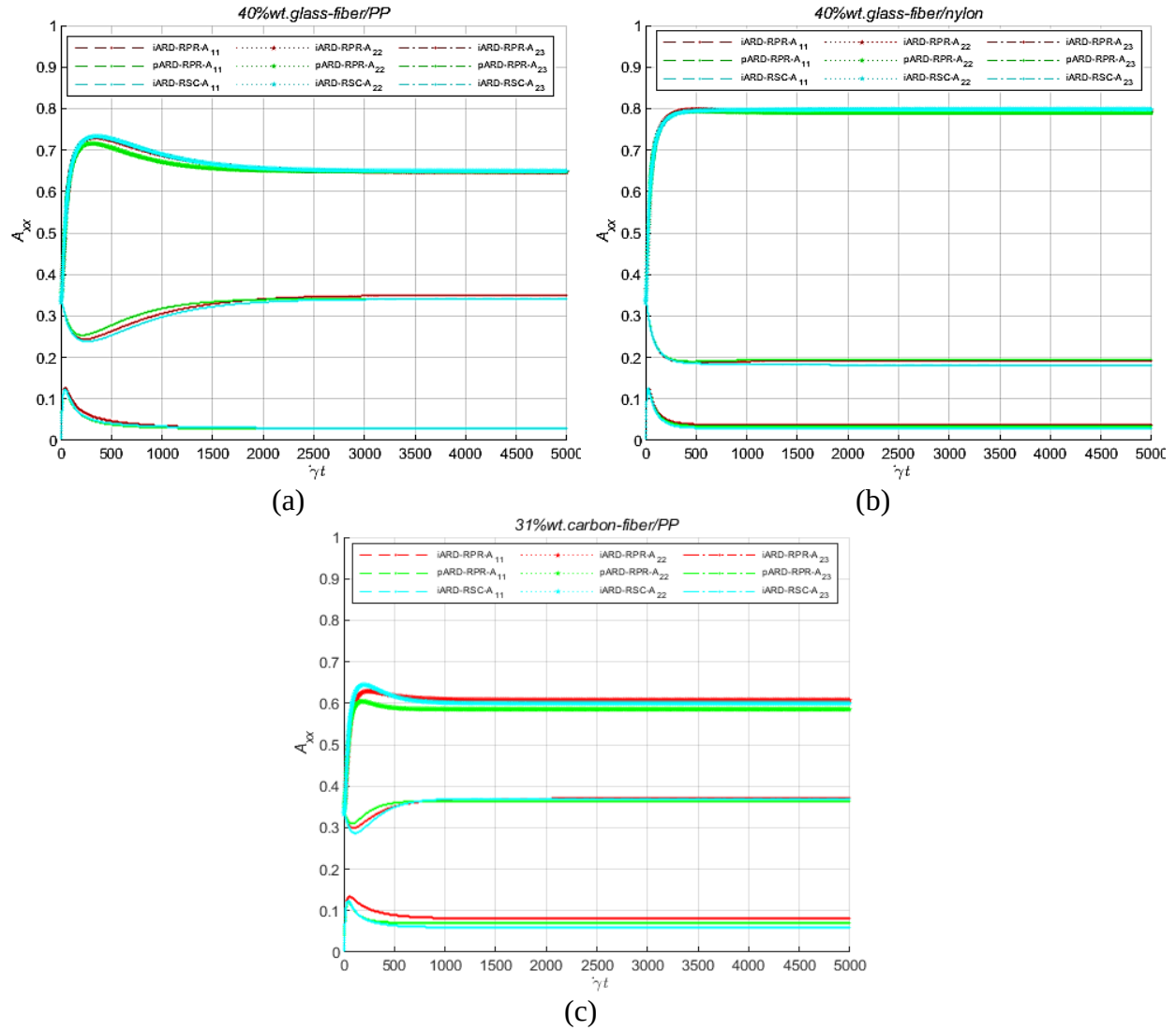


Figure 6.3: Time evolution of the a_{11} , a_{22} & a_{13} components of the 2nd order orientation tensor for calibrated iARD-RPR, iARD-RSC, pARD-RPR models for (a) 40% wt. glass-fiber/PP (b) 40% wt. glass-fiber/nylon (c) 31% wt. carbon-fiber/PP.

Table 6.13: Error estimates of the a_{11} , a_{22} & a_{13} steady-state orientation tensor component values for iARD-RPR, iARD-RSC, pARD-RPR models for (1) 40% wt. glass-fiber/PP (2) 40% wt. glass-fiber/nylon (3) 31% wt. carbon-fiber/PP

	<u>A_11</u>	<u>A_22</u>	<u>A_13</u>
iARD-RPR1	0.0060	0.0032	0.0036
pARD-RPR1	0.0009	0.0006	0.0311
iARD-RSC1	0.0070	0.0037	0.0134
iARD-RPR2	0.0003	0.0002	0.0012
pARD-RPR2	0.0003	0.0007	0.0156
iARD-RSC2	0.0008	0.0017	0.0551
iARD-RPR3	0.0000	0.0001	0.0053
pARD-RPR3	0.0000	0.0001	0.0059
iARD-RSC3	0.0066	0.0014	0.0067

6.4.3 Performance of the different Closure Approximations

The performance of the NR method in obtaining the steady state values for different closure approximations of the 4th order orientation tensor in terms of accuracy and stability has also been assessed. We consider for this assessment the FT model with a $C_I = 0.01$. The initial orientation state for the RK4 reference method is assumed to be random and we assume the same initial guess for the NR method as that of the preceding section. From Table 6.14, except for the HL2 closure approximation all other Hinch and Leal closures behaved well. By reason of the inherent nature of the transient behavior of the orientation tensor based on the HL2 closure approximation which shows a sudden transition in steady state values at a time fraction of about 100 (cf. Figure 6.4), we observe a discrepancy in the result for this closure since the NR method has no memory of the history of the orientation state and the accuracy of its prediction is based on the initial guess. The NR method predicts the initial steady state values of $a_{11} = 0.6103$, $a_{12} = 0.0206$ while the RK4 method transitions to a final steady state orientation of $a_{11} = 0.5759$, $a_{12} = 0.0467$. The higher order fitted closure approximations behave well in the NR methods and show good accuracy in predictions (cf. Table 6.15).

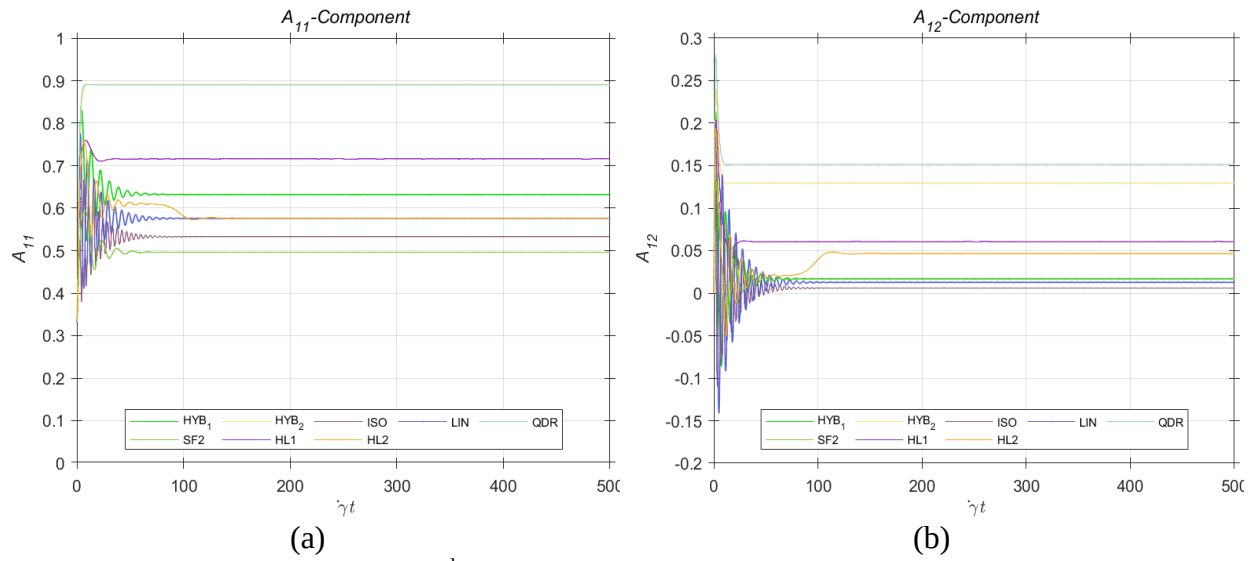


Figure 6.4: Transient profiles of 2nd order orientation tensor evolution for (a) component a_{11} and (b) component a_{12} for the various Hinch and Leal closure approximations of the 4th order orientation tensor.

Table 6.14: Error estimates of the a_{11} , a_{22} & a_{12} steady-state orientation tensor component values based on the various Hinch and Leal closure approximations of the 4th order orientation tensor.

	A_11	A_22	A_12
HYB_1	0.0000	0.0005	0.2306
HYB_2	0.0151	0.0017	0.0223
ISO	0.0000	0.0049	0.6305
LIN	0.0004	0.0003	0.4655
QDR	0.0036	0.0006	0.0053
SF2	0.0027	0.0101	0.0419
HL1	0.0042	0.0059	0.0313
HL2	25.9705	5.9662	55.8952

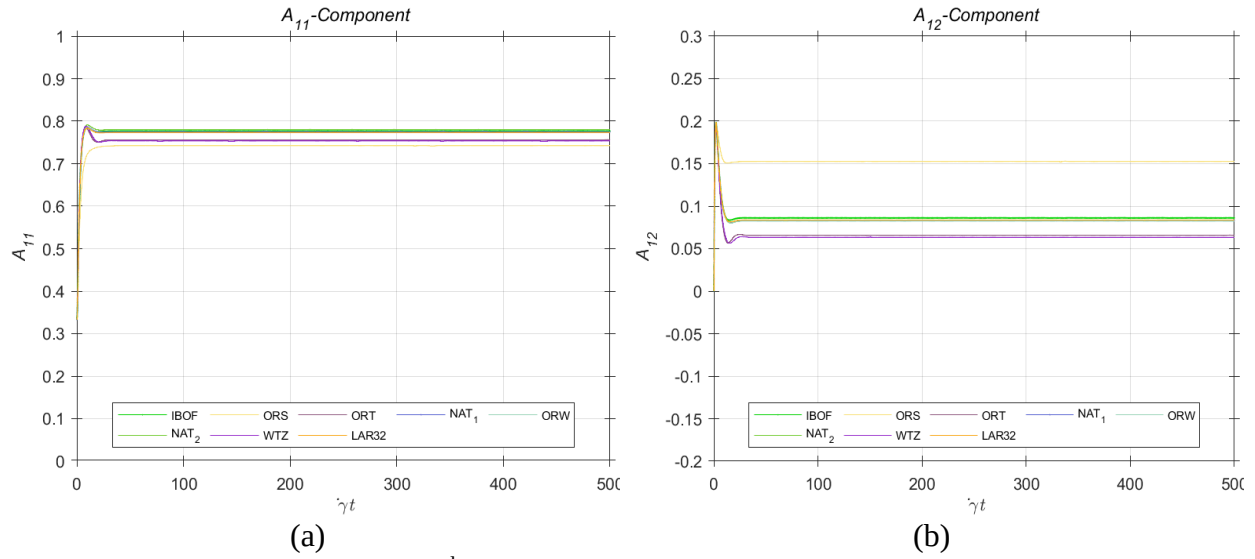


Figure 6.5: Transient profiles of 2nd order orientation tensor evolution for (a) component a_{11} and (b) component a_{12} for the higher order orthotropic fitted, IBOF and EBOF closure approximations of the 4th order orientation tensor.

Table 6.15: Error estimates of the a_{11} , a_{22} & a_{12} steady-state orientation tensor component values based on the higher order fitted closure approximations of the 4th order orientation tensor.

	A_11	A_22	A_12
IBOF	0.0007	0.0015	0.0232
ORS	0.0050	0.0055	0.0151
ORT	0.0781	0.0415	0.1600
NAT_1	0.0000	0.0004	0.0096
ORW	0.0007	0.0001	0.0061
NAT_2	0.0022	0.0017	0.0131
WTZ	0.0000	0.0003	0.0079
LAR32	0.0046	0.0013	0.0203
ORW3	0.0013	0.0003	0.0036
VST	0.0257	0.0123	0.0240
FFLAR4	0.0068	0.0029	0.0327
LAR4	0.0020	0.0008	0.0036

6.4.4 Homogenous Flow Considerations

We consider different homogenous flows to ensure the stability of the Newtons method in finding stable roots. The following flows were considered:

- (i) Simple Shear, $L_{12} = \dot{\gamma}$

- (ii) Two Stretching/Shearing flow, simple shear in 1-2 plane superimposed with uniaxial elongation in 3-direction. $L_{11} = -\dot{\epsilon}$, $L_{22} = \dot{\epsilon}$, $L_{33} = 2\dot{\epsilon}$, $L_{12} = \dot{\gamma}$. Two cases consider, balanced shear/stretch, $\dot{\gamma}/\dot{\epsilon} = 10$, dominant stretch, $\dot{\gamma}/\dot{\epsilon} = 1$
- (iii) Uniaxial Elongation, $L_{11} = 2\dot{\epsilon}$, $L_{22} = L_{33} = -\dot{\epsilon}$
- (iv) Biaxial Elongation, $L_{11} = L_{22} = \dot{\epsilon}$, $L_{33} = -2\dot{\epsilon}$
- (v) Two shear/planar-elongation flow, simple shear in 1-3 plane superimposed on planar elongation in 1-2 plane. $L_{11} = -\dot{\epsilon}$, $L_{22} = \dot{\epsilon}$, $L_{12} = \dot{\gamma}$. Two cases consider, balanced shear-planar elongation, $\dot{\gamma}/\dot{\epsilon} = 10$, & dominant planar elongation, $\dot{\gamma}/\dot{\epsilon} = 1$
- (vi) Balanced shear/bi-axial elongation flow, simple shear in 1-3 plane superimposed on biaxial elongation. $L_{11} = \dot{\epsilon}$, $L_{22} = \dot{\epsilon}$, $L_{12} = \dot{\gamma}$, $L_{33} = -2\dot{\epsilon}$, A range for $\dot{\gamma}$ is used such that, $2 \leq \dot{\gamma}/\dot{\epsilon} \leq 5$
- (vii) Triaxial Elongation, $L_{11} = L_{22} = L_{33} = \dot{\epsilon}$
- (viii) Balanced shear/tri-axial elongation flow, simple shear in 1-3 plane superimposed on biaxial elongation. $L_{11} = \dot{\epsilon}$, $L_{22} = L_{33} = \dot{\epsilon}$, $L_{12} = \dot{\gamma}$, A range for $\dot{\gamma}$ is used such that, $2 \leq \dot{\gamma}/\dot{\epsilon} \leq 5$

The initial orientation state for the RK4 reference method is assumed to be random and the initial guess $\underline{\underline{a}}^0$ assumed for each flow consideration is presented in Table 6.16 below.

Table 6.16: NR initial guess values for different flow conditions

(i)			(ii) & (v)			(iii)			(iv, vii & viii)			(vi)		
0.35	0.00	0.0	0.70	0.00	0.0	0.10	0.00	0.0	0.40	0.00	0.0	0.20	0.00	0.0
0.00	0.55	0.0	0.00	0.20	0.0	0.00	0.10	0.0	0.00	0.40	0.0	0.00	0.70	0.0
0.00	0.00	0.1	0.00	0.00	0.1	0.00	0.00	0.8	0.00	0.00	0.2	0.00	0.00	0.1

Among all closure approximations, the natural closure approximations (exact midpoint fit and extended quadratic fit (cf. Kuzmin [1]), and the Wetzel rational ellipsoid closures behaved well in all flows while the other orthotropic closures had stability issues for one or more of the complex

flows and gave non-physical roots. The ability of the NR method to predict accurate results depends on a reasonable initial guess based on the flow type and a suitable closure approximation.

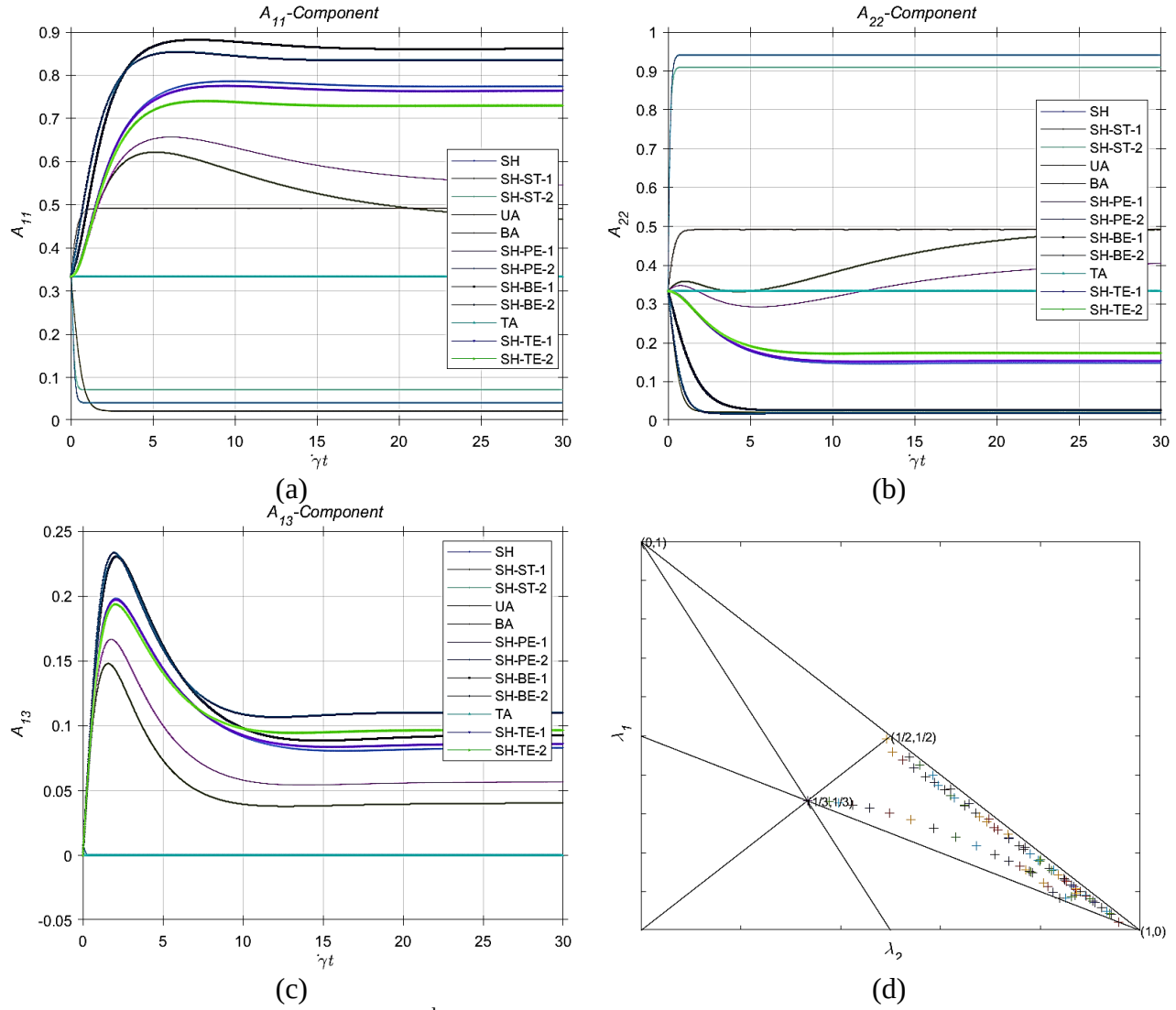


Figure 6.6: Transient profiles of 2nd order orientation tensor evolution for (a) component a_{11} and (b) component a_{22} (c) component a_{12} for the various flow considerations. (d) shows the eigenspace for the steady state values obtained for the different flow conditions based on NR method.

Table 6.17: Error estimates of the a_{11} , a_{22} & a_{12} steady-state orientation tensor component values for the various flow considerations.

	A_11	A_22	A_13
SH	0.0027	0.0014	0.0012
SH-ST-1	0.0004	0.0004	0.0073
SH-ST-2	0.0001	0.0000	0.0000
UA	0.0046	0.0046	0.0000
BA	0.0175	0.0175	0.0000
SH-PE-1	0.0019	0.0013	0.0053
SH-PE-2	0.0004	0.0024	0.1631
SH-BE-1	0.0148	0.0003	0.0000
SH-BE-2	0.0053	0.0000	0.0000

6.5 Conclusion

In conclusion, a Newton-Raphson method has been successfully implemented in determining the steady state 2nd order orientation tensor component values by determining exact partial derivatives of the material derivative of the orientation tensor with respect to its individual components. Different macroscopic fiber orientation moment-tensor models and closure approximations of the 4th order orientation tensor has also considered and the performance of the NR method in different flows have been studied. Like with any typical application of the NR root finding method, a good initial guess of the steady state orientation is required to yield non-physical values. The numerical stability of the NR method depends on complexity of the flow and the closure approximations. The Natural orthotropic and the IBOF closure approximations performed best for very complex flows. The NR method is comparatively faster compared to the RK4 method. Although obtaining exact derivatives of the 2nd order moment-tensor equation of change can be very cumbersome, once they are modelled, they are computational more efficient since they require less function evaluations compared to a higher order finite difference method of matching accuracy. Moreover, round off error and truncation error may become significant when dealing with relatively small quantities that may lead to instability of the numerical scheme.

7 CHAPTER SEVEN

2D Multi-Scale Extrusion-Deposition Polymer Composite Melt Flow Process Simulation

Sections of this chapter published as: Awenlimobor, A., Smith, D.E. and Wang, Z., 2024. Simulation of fiber-induced melt pressure fluctuations within large scale polymer composite deposition beads. *Additive Manufacturing*, 80, p.103980.

The phenomenon of heterogenous micro-void segregation at the interface between fiber and matrix during polymer melt flow processing has been shown to be significantly influenced by the local surrounding fluid pressure [9], [10], [11], [12], [13], [14], [15]. Note that in the moisture induced void nucleation mechanism [9], [10], [11], [12], [13] and the restrained volumetric shrinkage mechanism [14], [15], the onset of void nucleation occurs once the local fluid pressure drops below a critical value. Once micro-voids nucleate, their growth is driven primarily by the pressure difference between the micro-void internal pressure and external pressure in the surrounding fluid [5], [8]. Simulating the local pressure distribution around the fiber's surface during polymer processing can provide useful insight into the underlying mechanisms responsible for void formation especially at the tips of fiber where they are observed to mostly segregate. [52], [230].

Fiber suspension simulation, particularly those performed for polymer composite melt extrusion-deposition processes, have almost exclusively focused on fiber orientation and spatial distribution within the microstructure. However, little attention has been given to micro-void formation and evolution during extrusion-deposition or to understand how the suspended fibers influence micro-void development. The flow of polymer-melt through the nozzle during typical EDAM processing is characterized by a complex combination of shear and extensional flows that are dependent on temperature, the viscoelastic polymer melts rheology and the geometry of the extruder nozzle. High shear rates tend to occur on the nozzle walls and the flow is more uniaxial elongation at the nozzle centreline [136], [349]. The shear dominant flow condition has been

shown [52], [230] to be responsible for creating pressure extremes at the fiber surface where heterogenous void nucleation likely occurs. The main objective of this chapter is to present a computational approach aimed at understanding mechanisms that may promote moisture/volatile induced micro-void nucleation on or near suspended fibers within the bead microstructure produced by polymer extrusion-deposition process using a multiscale modelling methodology. While our approach would be applicable to both filaments based FFF and LAAM systems and other extrusion-based processes, our focus here is on the large-scale polymer composite deposition. In the macroscale model, we develop a two-dimensional (2D) planar flow model for predicting melt flow velocity and fiber orientation within the polymer melt during the extrusion-deposition process in a LAAM extruder nozzle. Then a micro-scale model is developed following the approach in Zhang et al. [225], [229], [291] which is based on Jeffery's model assumptions for suspended particles [19]. We simulate the evolution of a single ellipsoidal fiber along streamlines of the polymer melt flow through the nozzle and onto the print platform utilizing the field responses (velocity, velocity gradients and pressure) obtained from the macroscale model which define boundary conditions in the micro-model. Then, a single fiber's translational and rotational velocities are computed by zeroing the net hydrodynamic forces and torques on the fiber's surface where its orientation and evolution along the flow path are updated based on an explicit iterative numerical algorithm which incorporates velocities and pressures from the macro-model. The micro-model is validated by comparing results of fiber motion and pressure distribution on the fiber surface with Jeffery's analytical model equations [19] for the motion of a single particle suspended in purely viscous shear flow. We account for rotary diffusivity due to short-range fiber-fiber interaction in the micro-model FEA simulation by determining an effective fluid domain size that mitigates Jeffery's rotation to match that predicted by the Advani-Tucker fiber orientation

evolution equation. We also consider the fiber's evolution along various flow paths based on a given set of random initial fiber conditions to determine pressure bounds on the fiber surface across the melt flow. The pressure distribution on the fiber's surface as it travels along streamlines through the LAAM nozzle and onto the print bed, particularly within the regions of die swell at the nozzle exit, provides insight into a potential mechanism that could promote micro-void formation within printed beads.

Knowledge of the relationship between process operating parameters and void formation and evolution can be used to control the quality of printed parts [5], [31].

7.1 Methodology

A multiscale modelling approach is developed in this work to better understand micro-void initiation within the beads printed with the LAAM extrusion-deposition process. The computational method here includes a macro-scale model which is used to calculate velocities and pressure along streamlines from the polymer melt flow solution in the extrusion-deposition process, and a micro-scale model which simulates the motion of a single rigid ellipsoidal particle based on the fluid flow solution along the macro-model streamlines. Our approach is a one-way coupling where computed velocities and pressures calculated along macro-model streamlines serve as inputs to define boundary conditions in the micro-model. A Newtonian fluid is assumed in both models. The material properties of the polymer melt employed in this study are taken from Heller et al. [21] and Wang et al. [22] which include a density of 1154 kgm^{-3} and kinematic viscosity of $817 \text{ Pa} \cdot \text{s}$ (i.e., 13% by weight carbon fiber filled ABS at 230°C with a shear rate of 100 s^{-1}). In all of the discussion to follow, a 'fiber' is a rigid two-dimensional ellipsoidal solid having an aspect ratio of $r_e = U_1/U_2$ where U_1 and U_2 are the lengths of the major and minor ellipsoidal axes.

7.1.1 Macroscale Model - Planar deposition flow simulation

A typical extrusion-deposition process of fiber filled polymer through a LAAM extrusion nozzle and the subsequent single bead deposition on a translating substrate is shown in Figure 3.1. The internal nozzle geometry used in this study is based on the Strangpresse (Strangpresse, LLC, Youngstown, Ohio, USA) Model 19 LAAM single screw extruder nozzle where an annotated schematic representation of its internal nozzle geometry appears in Figure 3.1. The 2D planar flow domain consists of the internal nozzle geometry region and a single bead layer deposited on the substrate that translates laterally with respect to the nozzle. (cf. Figure 3.1a). The FEM formulation is briefly described here where additional modelling details of planar deposition flow can be found in Zhang, et al. [22].

The governing equations of mass and momentum conservation for polymer melt flow within the nozzle and the printed bead are defined by Stokes's equation (eqns. (5.86)-(5.89)) based on the assumptions of no inertia in the fluid, the polymer melt is a creeping flow with a low Reynolds number (i.e., $Re \ll 1$), and the polymer melt is an isothermal, incompressible, Newtonian fluid [22]. Note that eqn. (5.89) does not include the influence of fiber orientation on the deviatoric stress. The ANSYS Polyflow (Ansys, Canonsburg, PA, USA) commercial software is used for the macro-model polymer melt flow extrusion-deposition analysis. Figure 7.1a illustrates the quasi-steady fluid domain and boundary conditions for the 2D polymer melt flow model. Using data from Heller et al. [21] and Wang et al. [22], [263], the average normal velocity of 24mm/s is prescribed at the nozzle inlet Γ_1 , and the velocity of the moving substrate and deposited material is 101.6mm/s in the positive x-direction which is imposed on Γ_4 and Γ_5 . A no slip boundary condition is imposed on the nozzle inner wall Γ_2 and a free-surface boundary condition is

prescribed on the exposed surface Γ_3 of the deposited material. Figure 7.1b shows computed velocity streamlines that form between the nozzle inlet Γ_1 and the bead flow exit Γ_5 . Also shown in Figure 2b are feature streamlines 4, 10, and 18 in addition to zones of interest 1, 2, and 3 to be discussed below.

For the non-Newtonian simulation, a shear-thinning fluid with a power law index of $n \approx 0.45$ and a consistency coefficient of $m \approx 10^4 Pa \cdot s^n$ is used. As n approaches 1, the viscosity approaches the Newtonian value equal to the consistency coefficient corresponding to a shear-rate of unity. The computed streamlines and resulting velocity profile distribution across sections of the nozzle for the non-Newtonian studies in comparison to results of the Newtonian analysis [22], [263] are presented in Figure 7.1b. While the velocity profiles of the Newtonian analysis are parabolic in shape, the profiles of the non-Newtonian analysis are somewhat hyperbolic shaped with a velocity plateau towards the center tending towards a plug flow velocity distribution.

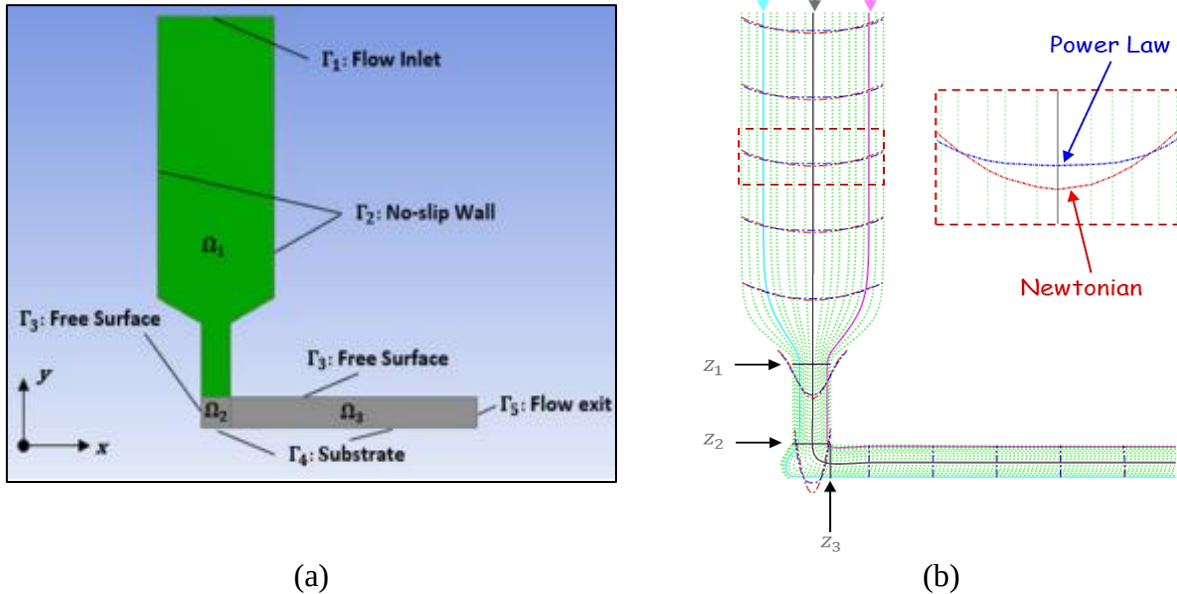


Figure 7.1: 2D Planar extrusion-deposition flow model a) fluid domain and boundary conditions, b) velocity streamlines of the polymer flow through the nozzle with feature streamlines highlighted.

7.1.2 Microscale Model - Single fiber motion simulation

Simulation of a rigid ellipsoidal fiber motion along streamlines of the polymeric melt flow is performed in this work using a custom FEM code developed in MATLAB (MathWorks, Natick, MA, USA). The single fiber micromodel is governed by Stokes's assumption of negligible inertia and negligible thermal effects and includes an isotropic homogenous Newtonian fluid that is the same as that used in the extrusion-deposition macro-model described above. Our algorithm for the micro-model simulation of a single 2D rigid ellipsoidal particle is derived from the work in Zhang et al. [225], [229], [291]. The flow domain for the 2D single fiber micro-model appears in axes (cf. Figure 5.3a) where we assume no slip occurs on the fiber surface and there is no flux across the fiber surface. Velocity, velocity gradient and pressure computed along streamlines of the extrusion-deposition macro-model described above are used to prescribe boundary conditions on the micro-model flow boundaries as a function of time. To impose these values in the micro-model, three essential boundary conditions are prescribed with respect to the fiber's local coordinate axes (cf. Figure 5.3b). FEM solutions are obtained by applying the essential boundary conditions to a fixed mesh which is rotated with the local fiber axes. Rotating the model in this manner significantly reduces computation time by maintaining a constant FEA system matrix, avoiding the need of remeshing the domain and/or recalculating the system matrix and its decomposed form at each iteration time step. The far-field velocities on the fluid domain boundary $\dot{X}_i^{BC1}$ (cf. Figure 5.3b) of the micro-model are defined from the streamline velocities $\dot{X}_i^\psi$ and velocity gradients L_{ij}^ψ based on eqn. (5.90) which are obtained from the macro-model velocity solution at each time t of the single fiber evolution solution. Likewise, the prescribed pressure p_{BC2} is defined

according to eqn. (5.94) on a far-field node BC2 located on the fluid domain surface where its value is computed from the macro-model streamline pressure p_ψ . The prescribed velocities $\dot{X}_i^{BC3}$ on the fiber's surface are obtained in the usual manner according to the equation of rigid body motion (cf. eqn. (5.93)).

The micro-model formulations are non-linear modifications to the model development by Zhang et. al. [225], [229], [291] and the governing equations are the same Stokes equations for mass and momentum conservation used in the macro-model given in eqns. (5.86)-(5.89), based on the same assumption of isothermal, incompressible, homogenous viscous flow with a non-Newtonian power-law fluid definition. The microscale model development for the single fiber motion along the streamlines of the GNF polymer melt flow has been provided in detail in CHAPTER FIVE of this dissertation. Similar to the Newtonian analysis [52], [230], the instantaneous velocities, velocity gradients and pressure of the streamline data obtained from velocity solutions of the GNF macro-model analysis are used to derive the far-field fluid domain boundary conditions.

7.1.3 Non-Dilute Fiber Suspension Motion

Jeffery's model assumes a Newtonian fluid and is valid for dilute suspension where fibers possess a relatively large radius of influence with neighboring fibers and contribute independently to the dissipation of energy in the form of a modified isotropic effective fluid viscosity μ^* for the suspension, such that $\mu^* = \mu (1 + \kappa \vartheta_f)$ [19], where κ is the modification factor dependent on the particles dimension which has been accounted for in our extrusion-deposition macro-model appearing above and ϑ_f is the volume fraction of the ellipsoidal fiber in the suspension. However, for semi-dilute and concentrated

suspensions, there exists some degree of stochasticity in an individual fiber's behavior due to momentum diffusion and fiber-fiber interactions as the distance between neighboring particles becomes small relative to its size. In this case, neighboring fibers would introduce some degree of disturbance in a particle's surrounding fluid. As a result, particle-particle interaction necessitates a coupling effect between fibers. In other words, interactions between fibers reduce the effective radii of influence between near neighbors, the proximity of which results in an increased energy dissipation within each fiber's sphere of influence [20], [291].

As the fiber volume fraction and/or aspect ratio increases, collision of particles creates momentum transfer between colliding particles. Kugler et. al [20] classified fiber-fiber interaction into long-range and short-range hydrodynamic interaction, the latter of which can be further sub-divided into short range lubrication regimes, direct mechanical contact and a transition regime. As a result of momentum diffusion, the fibers eventually assume a steady state orientation that depends on the initial condition in accordance with the indeterminacy described by Jeffery. Folgar and Tucker [256] extended Jeffery's analysis by accounting for a collection of interacting suspended particles by incorporating a rotary diffusion term D_r . The rotary diffusion term D_r is defined in terms of the scalar magnitude of deformation tensor $\dot{\gamma}$ according to $D_r = C_I \dot{\gamma}$, where C_I is the interaction coefficient which is an empirical constant. Kugler et. al [20] gives a review of existing orientation models that accounts inter-particle interaction such as nematic model, anisotropic and mold flow rotary diffusion model, retarding principal rate model, etc.

To capture fiber-fiber interactions in our single fiber model, we develop a relation between the Folgar-Tucker interaction coefficient C_I and the effective radius of influence in our micro-model. Firstly, we determine a relation between the stall angle of the fiber and the interaction coefficient C_I based on equation of change of the 2nd order orientation tensor by Advani and Tucker [17]. Here the stall angle is the fiber angle at which rotary motion ceases which has been found to be a function of the micro-model flow domain size (see e.g., Zhang et al. [291]). Subsequently we obtained a relation between the flow domain size and the fiber stall angle through a series of micro-model FEA simulations with fluid boundary domain BC1 of different sizes. As $\mathfrak{m}$ decreases, the ends of the fiber become nearer to the prescribed boundary BC1 such that the velocity field near the fiber tips hydrodynamically interacts with the flow adjacent to BC1. The prescribed boundary creates a flow disturbance as viewed from the fiber in a manner similar to that which would be expected by neighboring fibers in a semi-concentrated flow. We then determine the relationship between the steady-state orientation tensor and the interaction coefficient C_I for a given ellipsoidal aspect ratio. A relationship between C_I and the micro-model flow domain size is then established by equating fiber stall angle in the micro-model to the direction of the first eigenvector of the fiber orientation tensor at steady state. This approach provides a means to approximately account for the effect of fiber-fiber interaction in the FEA simulation of the single fiber evolution along streamlines for a given interaction coefficient.

Observations of the experimentally determined steady state orientation [176] show that the fibers tend to align with streamlines of the flow field irrespective of the initial conditions, contrary to Jeffery's idealization where suspended particles continue to rotate

in simple shear. Saffman [176] shows that non-Newtonian properties of the fluid, not considered by Jeffery, is responsible for a stall in the tumbling motion. Other factors not accounted for in Jeffery's model that adds to the indeterminacy of a particle's motion include the flexural tendency of the particle which would depend on its inherent elastic property, aspect ratio, fluid rheology of the medium and interacting flow field. Moreover, the fibers may eventually break when subject to severe interacting forces, however, fiber flexibility is beyond the scope of our work.

7.1.4 Determining Effective Fluid Domain Size

To quantify the effect of fiber-fiber interactions with our single fiber model, we first establish a relationship between a suspension's interaction coefficient C_I (cf. the Advani-Tucker model) and the stall angle in our single fiber FEA micro-model. The steady state orientation tensor values that correspond to a particular interaction coefficient can be determined from the Advani-Tucker 2nd order orientation tensor equation of change given as

$$\dot{a}_{ij} = \frac{1}{2}(\Xi_{im}a_{mj} - a_{im}\Xi_{mj}) + \frac{\kappa}{2}(\Gamma_{im}a_{mj} + a_{im}\Gamma_{mj} - 2a_{ijkl}\Gamma_{kl}) + 2D_r(\delta_{ij} - \alpha a_{ij}) \quad (7.1)$$

where, a_{ij} and a_{ijkl} are the 2nd and 4th order fiber's orientation tensors, respectively, κ is the shape parameter defined above, Γ_{ij} is the strain rate tensor given as $\Gamma_{ij} = [L_{ij} + L_{ji}]$, Ξ_{ij} is the vorticity tensor given as $\Xi_{ij} = [L_{ij} - L_{ji}]$ and α is a dimension factor (i.e., $\alpha = 3$ for 3D orientation and $\alpha = 2$ for 2D planar orientation). In the above, the fourth-order orientation tensor a_{ijkl} is computed from a_{ij} using a closure approximation as is common in polymer composite suspension simulations. We employ the orthotropic fitted closure of Verweyst et al. [350] in all the calculations to follow. The symmetry properties of the

orientation tensors require that $a_{ij} = a_{ji}$ and $a_{ijkl} = a_{jikl} = a_{kijl} = a_{lijk} = a_{klij}$. The normalization condition also requires that $a_{ii} = 1$ and $a_{ijkk} = a_{ij}$ where repeated indices imply summation in the usual manner here and in the following. We determine the steady state 2nd order orientation tensor that results in zero rate of change, i.e., $\dot{a}_{ij} = \mathbf{0}$ via a Newton Raphson iteration scheme given as

$$a_{ij}^+ = a_{ij}^- - J_{mni j}^- \backslash \Sigma_{mn}^- \quad (7.2)$$

where the residual $\Sigma_{mn} = \dot{a}_{mn}$ is

$$\begin{aligned} \Sigma_{mn} = & \frac{1}{2} (\Xi_{mk} a_{kn} - a_{mk} \Xi_{kn}) + \frac{\kappa}{2} (\Gamma_{mk} a_{kn} - a_{mk} \Gamma_{kn} - 2\Gamma_{kl} a_{mnkl}) \\ & + 2D_r (\delta_{mn} - \alpha a_{mn}) \end{aligned} \quad (7.3)$$

and the Jacobian $J_{mni j}$ is obtained by differentiating the residual with-respect-to components of the 2nd order orientation tensor a_{ij} as.

$$\begin{aligned} J_{mni j} = \frac{\partial \Sigma_{mn}}{\partial a_{ij}} = & \frac{1}{2} \left(\Xi_{mk} \frac{\partial a_{kn}}{\partial a_{ij}} - \frac{\partial a_{mk}}{\partial a_{ij}} \Xi_{kn} \right) \\ & + \frac{\kappa}{2} \left(\Gamma_{mk} \frac{\partial a_{kn}}{\partial a_{ij}} + \frac{\partial a_{mk}}{\partial a_{ij}} \Gamma_{kn} - 2\Gamma_{kl} \frac{\partial a_{mnkl}}{\partial a_{ij}} \right) - 2D_r \alpha \frac{\partial a_{mn}}{\partial a_{ij}} \end{aligned} \quad (7.4)$$

The derivative of the 2nd order orientation tensor with respect to its individual components is simply

$$\frac{\partial a_{rs}}{\partial a_{mn}} = \delta_{rm} \delta_{sn} \quad (7.5)$$

where δ_{ij} is the Kronecker delta. Derivatives of a_{ijkl} with respect to a_{ij} are provided elsewhere for various closures approximations that are commonly used with eqn. (7.4) (cf. Awenlimobor and Smith [351], to appear). We define a preferred direction of orientation as the principal direction of the steady state a_{ij} computed from the n^{th} eigenvector of $a_{ij}(\Phi_{mn})$ corresponding to the maximum eigenvalue λ_n which is obtained from

$$\underline{\underline{\Phi}} : \mathbb{A}_{ij} = \Phi_{ki} a_{kn} \Phi_{nj}, \quad \Lambda_k = \mathbb{A}_{kk}, \quad \underline{\underline{\Lambda}} : \epsilon_{ijk} [a_{ij} - \Lambda_n \delta_{ij}] = 0 \quad (7.6)$$

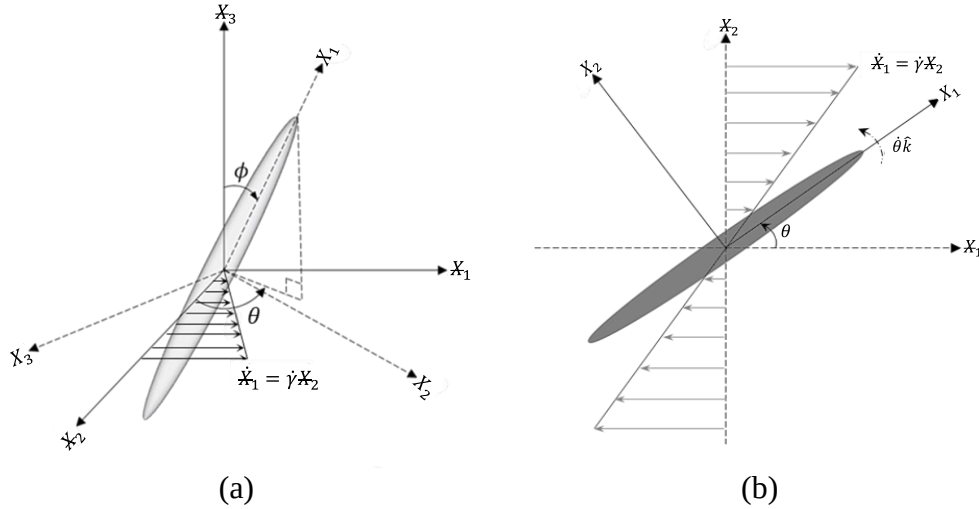


Figure 7.2: Fiber Orientation Angles: a) 3D coordinates used in fiber orientation tensor equations and b) 2D coordinates used in single fiber motion simulations.

Consider planar simple shear flow having $\dot{X}_1 = \dot{\gamma} X_2$ and $\dot{X}_2 = \dot{X}_3 = 0$ (cf. Figure 7.2b) with a fiber at $\phi = 90^\circ$ rotating in the $X_1 - X_2$ -plane. For this flow field, the in-plane steady state orientation angle θ was evaluated using eqn. (7.1) through (7.6) for various values of C_I and for different closure approximations as given in [351]. Alternatively, a series of FEA simulations were performed for an ellipsoidal fiber rotating through a modified Jeffery's orbit in simple shear for various fluid boundary domain sizes (cf. Figure 5.3). A corresponding pair of FEA simulation and orientation tensor evaluations were performed using the same fiber geometry and shear rate. Values of stall angle were then compared. Results of stall angle as a function of micro-model domain size factor $\mathfrak{m} = d_f/2H_1$ (where d_f is the diameter of the micromodel flow domain) and C_I appear in Figure 7.3.

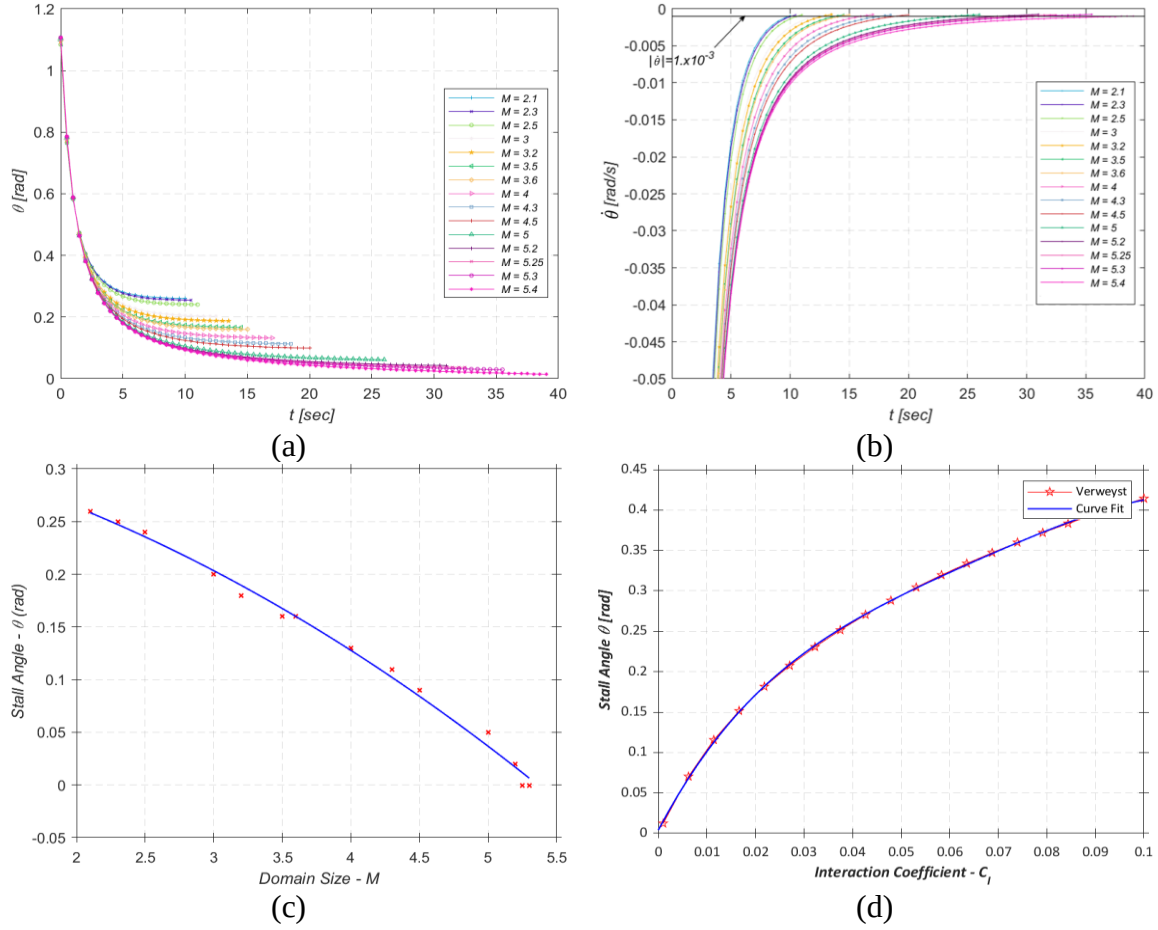


Figure 7.3: Single fiber angular motion and preferred orientation results for varying domain size $\mathbb{m}$ a) fiber orientation angle θ through its stall angle b) fiber angular velocity $\dot{\theta}$ simulated through a stall angular velocity tolerance of $|\dot{\theta}| = 1 \times 10^{-3} \text{ 1/s}$ c) relationship between fiber stall angle and domain size factor $\mathbb{m}$ from FEA analysis, d) relationship between fiber steady state angle θ and interaction coefficient C_I (Aspect ratio $r_e = 6$).

The influence of domain size appearing in Figure 7.3c shows a nearly linear relationship between the fiber stall angle and domain size from the micro-model simulations, given by eqn. (7.7) below.

$$\theta = 0.33839 - 0.022\mathbb{m} - 0.0077\mathbb{m}^2 \quad (7.7)$$

Additionally, results of the orientation angle computed from the eigenvectors of the steady state orientation tensor $\mathbf{a}_2$ show nonlinear relationship between stall angle and interaction coefficient (cf. Figure 7.3d) which can be represented as

$$\theta = \pi/2 - 1.57 + 11.4C_I - 183.5C_I^2 + 1773.4C_I^3 - 6680.1C_I^4 \quad (7.8)$$

Combining results from Figure 7.3c&d, we obtain a relationship between the fluid boundary domain size in our single fiber micro-model and C_I given as (cf. Figure 7.4)

$$\mathfrak{m} = -1.4285 + \sqrt{45.89 - 1.48 \times 10^3 C_I + 2.38 \times 10^4 C_I^2 - 2.30 \times 10^5 C_I^3 + 8.68 \times 10^5 C_I^4} \quad (7.9)$$

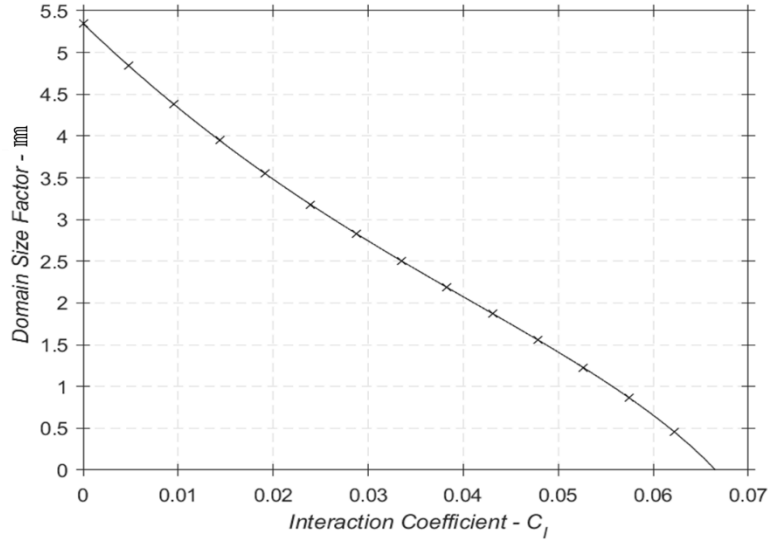


Figure 7.4: Fitted relationship between domain size factor $\mathfrak{m}$ vs interaction coefficient C_I .

Assuming an ellipsoidal fiber aspect ratio $r_e = 6$ which corresponds to a shape parameter $\kappa = 0.9459$ and given a volume fraction $\vartheta_f = 8.4\%$ by volume (13% by weight) CF/ABS polymer composite, we obtain an interaction coefficient of $C_I = 0.0128$ using Bay's correlation that relates C_I to ϑ_f and r_e [258]. It follows from Equation (7.9), that the effective domain size based on our C_I is $\mathfrak{m} = 4.08$ (~ 4.0) which we have used in the Newtonian simulations. Given that fiber suspensions are classified into 3 concentration regimes based on v_f and r_e as [57], [240] our simulations are within or nearly within the concentrated regime for the suspension where $C_I = .0128$ and $\mathfrak{m} = 4$ are used in the results section below.

7.2 Results and Discussion

7.2.1 Multi-scale Newtonian melt flow simulation

The result of the velocity magnitude $|\underline{\dot{X}}|$ and scalar magnitude of deformation tensor $\dot{\gamma}$ from the macroscale Newtonian analysis appear in Figure 7.5a & b, respectively. Computed velocities in Figure 3a show an increase in velocity magnitude from the edge of the nozzle to its center as expected. It follows that material along streamlines near the edges of the nozzle have a higher extrusion-deposition time compared to those closer to the center. The velocity contours (see for example, Figure 5 and 6 in Ref. [22]) show a parabolic velocity distribution across transverse sections of extruder nozzle except near the entrance and exit of the straight capillary portion of the nozzle. Melt flow in these transition regions is characterized by sharp transitions of velocity and velocity gradients along the inside wall of the extrusion nozzle. Upon deposition onto the print bed, the melt flow attains a uniform velocity throughout the bead material where all stresses reduce to zero.

The plot of velocity gradient in Figure 7.6 shows unusually high values occurring at the sharp corners of the flow field due to singularities in the velocity solution where the polymer melt flow transitions from a no-slip to a free surface boundary condition, which we attribute a posteriori to be responsible for unexpected behavior of the fiber's motion along streamlines close to these locations. In this figure, as well as in all of the micro-model results, $\dot{X}_1$ and $\dot{X}_2$ are the components of the velocity vector $\underline{\dot{X}}$ in the X_1 - and X_2 -directions, respectively. We see from Figure 7.6 that the velocity gradient component - L_{12} dominates near the nozzle exit and is seen to increase in magnitude when moving outward from the center streamline towards those near the edge of the nozzle.

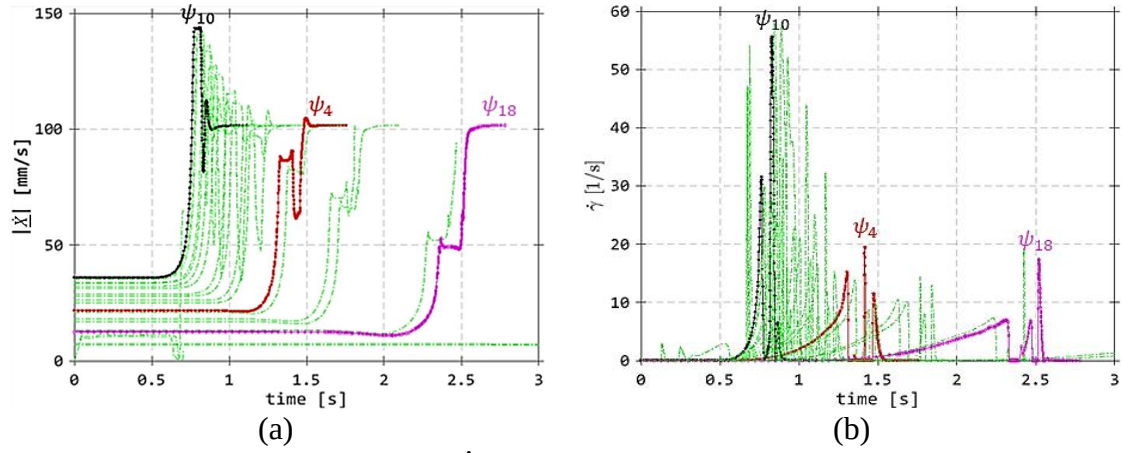


Figure 7.5: a) Velocity magnitude $|\dot{\mathbf{x}}|$ b) scalar magnitude of second order deformation tensor for various streamlines with feature streamlines highlighted⁴.

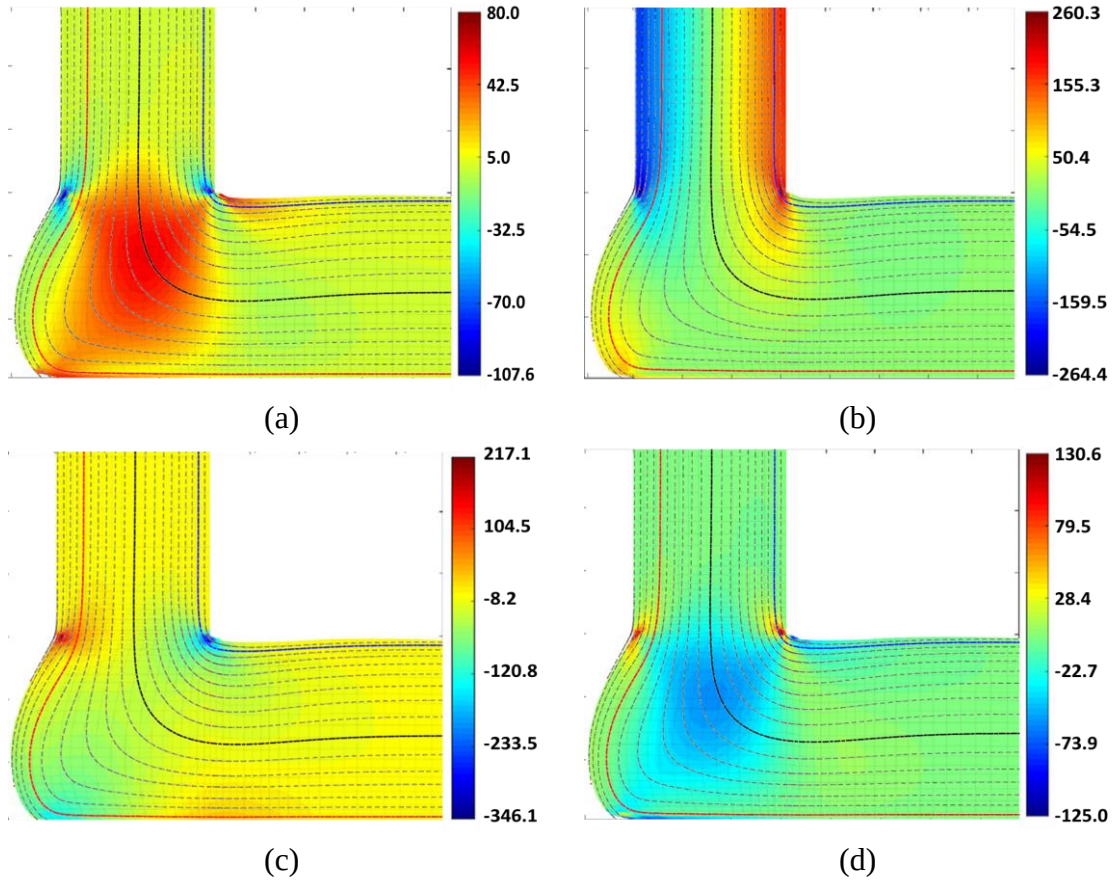


Figure 7.6: Velocity gradient contours near extrusion-deposition transition zones (a) L_{11} , (b) L_{12} , (c) L_{21} , (d) L_{22} . The units of the global velocity gradients are s^{-1}

⁴ ψ_n here refers to streamline identifier (n) and starts at 1 from the left edge of the nozzle increasing transversely to a maximum number of 22 at the right edge of the nozzle.

Chapter 5 describes the ability of our micro-model to reproduce Jeffery's result for single fiber motion, and the determination of an effective single ellipsoidal fluid domain size that approximates the effect of short-range fiber interaction in simple shear flow. All simulations included here use a fiber half-length of $a = 42\mu m$ and an ellipsoidal aspect ratio of $r_e = 6$ which corresponds to a cylindrical geometric aspect ratio of $r_c = 7.66$ using Equation (2.21) in Zhang [291]. Here we limit our discussion to results along streamlines ψ_4 , ψ_{10} , and ψ_{18} to capture effects along the lower, middle, and upper sections of the bead, respectively (cf. Figure 7.1b). The following simulations incorporate velocity, velocity gradients, and pressure computed in the 2D planar extrusion-deposition macro-model to define far field boundary conditions BC1 and BC2 in the single fiber micro-model. To assess the effect of initial conditions in the single fiber analysis, we run multiple simulations, each with its own initial fiber angle θ_0 over a range of $-\pi/2 \leq \theta_0 \leq \pi/2$ in increments of $\pi/12$. Simulating fiber motion over this range of initial angles and on various streamlines provides a comprehensive assessment of possible fiber responses and corresponding location where they occur across the extruder nozzle. To better display streamline results, subsequent figures presented in this section have been annotated to show three interest regions of the nozzle geometry appearing in Figure 7.1b which includes:

- (i) Zone 1: The entrance to the small capillary section of the nozzle at the point where the polymer- melt just exits the convergent zone.
- (ii) Zone 2: The exit from the nozzle where the polymer leaves the nozzle and enters the region of die swell, and the external pressure drops to atmospheric condition.

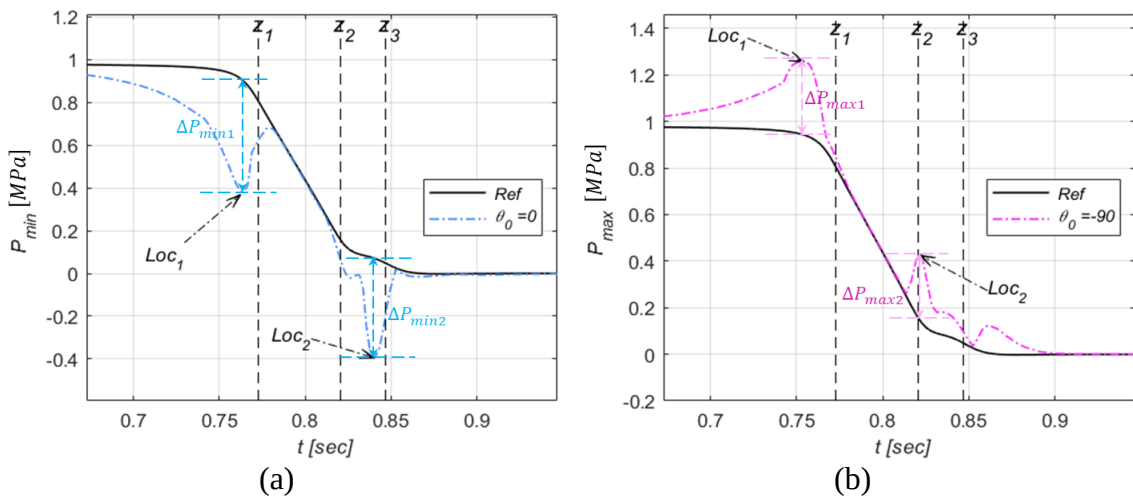
- (iii) Zone 3: The exit of die swell region below and to the side of the nozzle exit where the deposited material has made a complete 90° turn onto the translating bed below and attains a near uniform velocity equal to the print speed.

We consider the simulation of fibers in a concentrated suspension with $C_f = 0.0128$ using the reduced single fiber domain approach with $m = 4$ in the micro-model as described above. For each fiber motion simulation result (i.e., a fiber moving along a specific streamline with a designated initial angle), the overall minimum and maximum fiber surface pressure is calculated and the difference between the streamline pressure and overall minimum and maximum fiber surface pressures are noted. In addition, the corresponding coordinate locations where the minimum and maximum fiber surface pressures occur within extrusion-deposition flow are identified. Figure 7.7 shows a typical fiber surface pressure result along streamline ψ_{10} (starting at the centerline of the nozzle inlet) for a concentrated suspension where distinct extremes of minimum and maximum pressures identified as ΔP_{min} and ΔP_{max} , respectively, are plotted as a function time along with the streamline pressure from the macro-model. The first extreme pressure location, denoted here as Loc. 1, and the second extreme location, denoted as Loc. 2, appear in the pressure history for all streamlines and θ_0 with varying degrees of intensity and at slightly different locations as shown below. Note that the position along the streamline for Loc. 1 and Loc. 2 will occur at different locations depending on the streamline and initial fiber angle.

The initial extreme in minimum fiber surface pressure at Loc. 1 is observed to occur just prior to the entrance of the nozzle capillary section (i.e., zone 1) while the second pressure drop at Loc. 2 occurs within the die swell region between zones 2 & 3. Only at

the latter extreme fiber location does the absolute local minimum pressure on the fiber surface drop to a value that is below zero atmosphere (reaching -0.4MPa in the simulation appearing in Figure 7.7). This low pressure extreme is expected to provide a favorable condition for void nucleation to occur based on prior related research [9], [10], [11], [12], [13], [14], [15]. A closer inspection of the fiber's surface pressure distribution at this location shows that the peak sites occur at the fiber's tips (cf. Figure 7.7d) which is typical of all simulations presented in this work.

To gain a better understanding of the effect of streamline location on the fiber response during its motion through the extrusion-deposition flow in the concentrated regime, we present results of time-varying profiles for three select streamlines, one near the left edge - ψ_4 , the center streamline - ψ_{10} , and one at the far-right edge ψ_{18} (cf. Figure 7.1b), each with a range of initial fiber orientation as specified above. The computed results show that the fiber surface extreme pressures on the outer streamlines (ψ_4 and ψ_{18}) are less sensitive to initial fiber orientation over the entire deposition time as compared to the center streamline ψ_{10} where the initial fiber angle has much more pronounced effect on the characteristic pressure peak values.



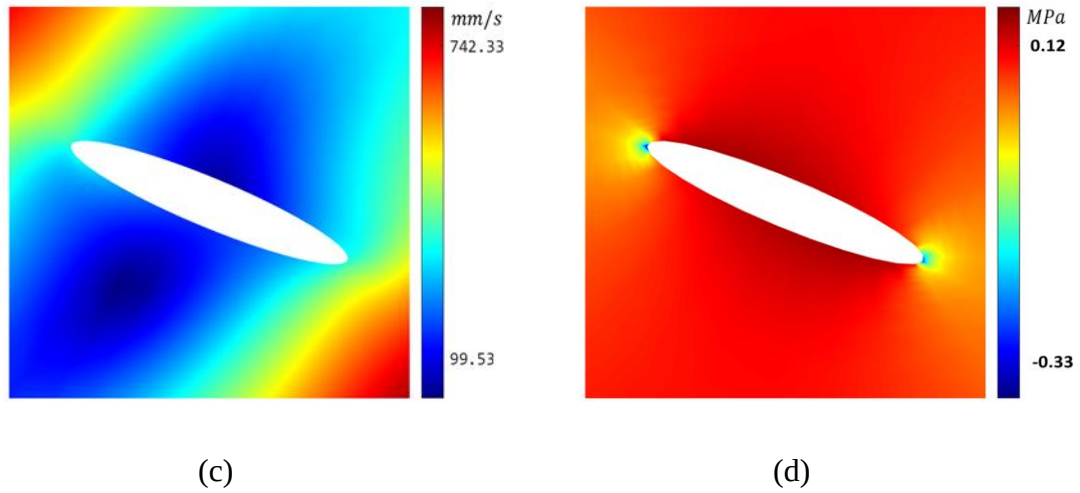
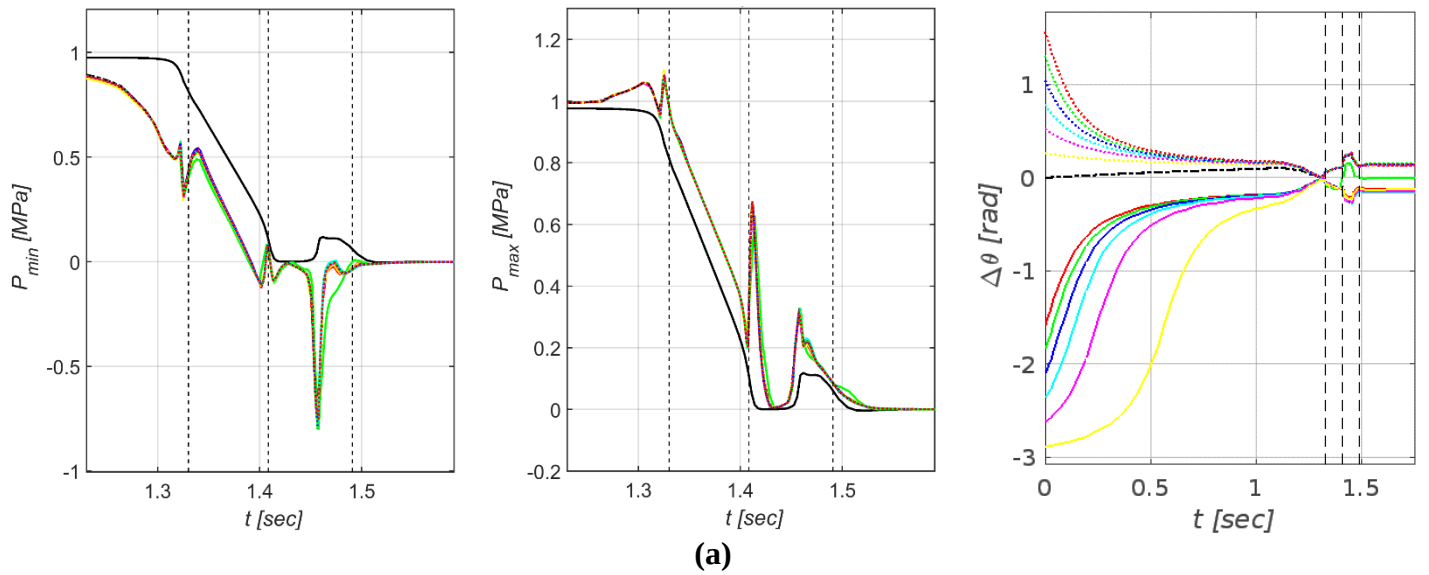


Figure 7.7: Selected computed results along center streamline ψ_{10} for the concentrated suspension ($C_I = 0.0128$ and $\text{mm} = 4$). Shown are the fiber's surface (a) minimum pressure ($\theta^0 = 0^\circ$) (b) maximum pressure ($\theta^0 = -90^\circ$) at peak locations (Loc. 1 & Loc. 2). Contour plot at the first location (Loc. 1) of minimum pressure drop showing (c) Velocity magnitude (d) Pressure near the fiber.

The results of the fiber orientation relative to the streamline direction presented in Figure 7.8 shows that the particle eventually tends to align with the streamlines of the flow irrespective of its initial starting angle and the degree of fiber alignment increases from the center streamline (ψ_{10}) to streamlines closer to edges of the nozzle (ψ_4 and ψ_{18}). The asymmetry in the results of the orientation for edge streamlines ψ_4 & ψ_{18} shown in Figure 7.8a & c, respectively, signifies that fibers on these streamlines undergoes uneven rotation prior to flow alignment depending on the degree and direction of initial misalignment relative to the prevailing vortex direction (ω) of the undisturbed flow which in turn depends on the relative positioning of the streamline with respect to the centerline.

To better depict the fiber rotation span for fibers initially inclined unfavorably with the flow, the orientation transient profiles have been vectorially added to π considering the fiber has no preferred ends (i.e., $\theta(t) = -\theta(t) - \pi$, $\theta_0 < 0$, $\omega > 0$ for streamline ψ_4 and $\theta(t) = -\theta(t) + \pi$, $\theta_0 > 0$, $\omega < 0$ for streamline ψ_{18}). Alternatively, the fiber

motion on the outer streamlines is more sensitive to the initial fiber orientation and possesses some degree of asymmetry with respect to the initial angle. This is due to the relatively high velocity gradients for streamlines closer to the nozzle edge as compared to the center streamline. Moreover, the transition time in the die swell region between zones 2 and 3 increases with streamline location from the right-hand edge to the left-hand edge due to correspondingly larger radius of curvature (cf. Figure 7.1b). Streamline 18 has a sharp 90° turn with negligible dwell time in the die swell region as zones 2 and 3 almost nearly overlaps unlike streamline 4 and 10 which experiences relatively higher dwell in the die swell region as the polymer melt gradually approaches the deposition plate surface.



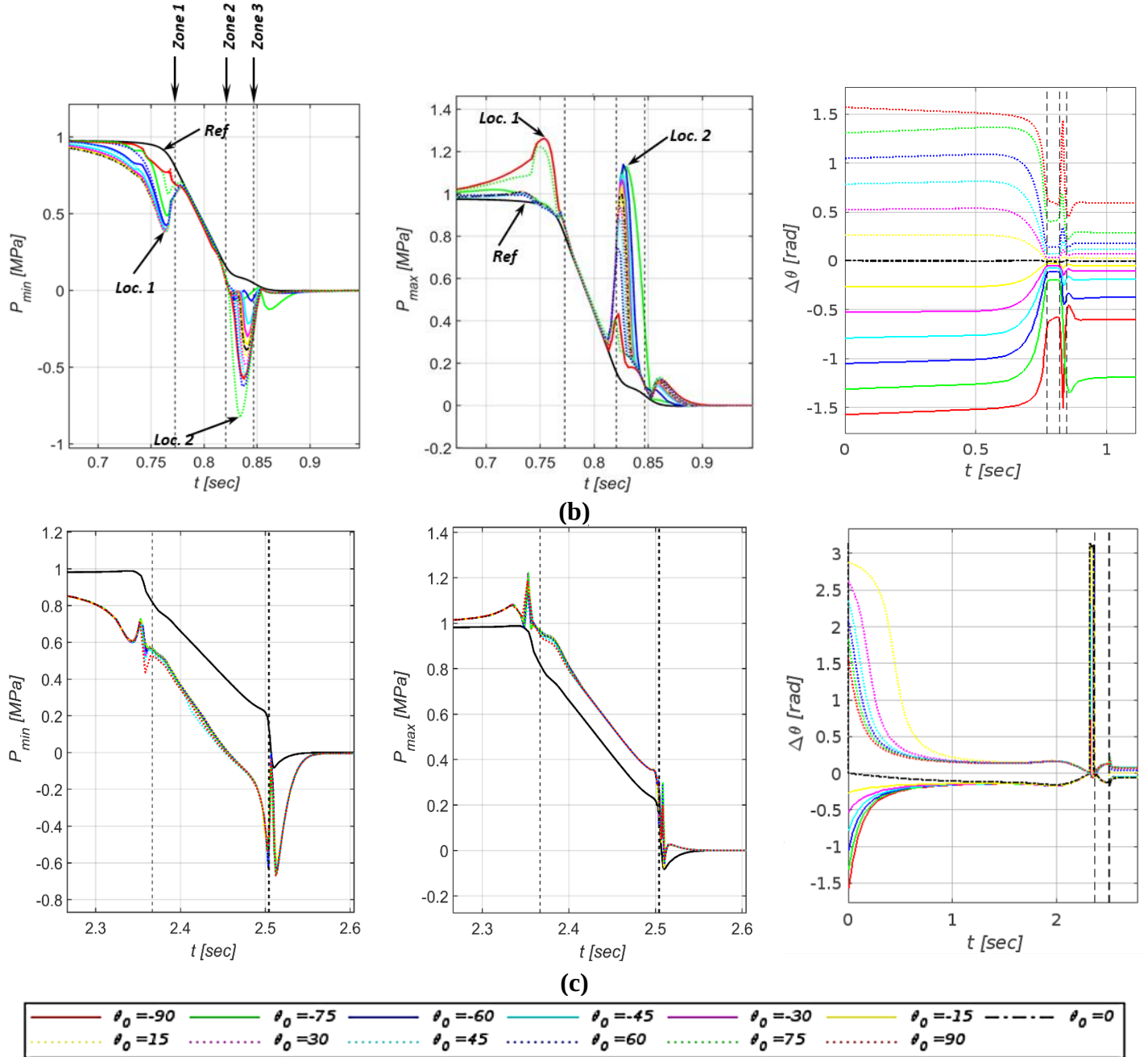


Figure 7.8: Quasi-transient profile plots of the fiber minimum pressure, maximum pressure and relative orientation angle, including various initial fiber angles for selected streamlines a) streamline-4 b) streamline-10 and c) streamline-18 ($C_I = 0.0128$ and $m = 4$).

For subsequent simulation results, we consider a range of initial fiber orientation and report the computed overall minimum and maximum pressure difference with respect to the streamline pressure across the nozzle at the important extreme pressure locations

(i.e., Loc. 1 and 2). In addition, we report the corresponding spatial positions where the minimum and maximum pressure extremes occur within extrusion-deposition flow for each of the various streamlines across the nozzle section. Lastly, we report the fiber's orientation relative to the streamline direction at three interest zones of the nozzle (zones 1-3).

Calculated results in Figure 7.9 show that the extreme pressures on center streamlines are more sensitive to initial fiber angle than that for the outer streamlines. We observe a drop in average minimum pressure of -0.5MPa at the first extreme occurrence (Loc. 1) which is almost uniform across all streamlines within the nozzle. Alternatively, the second average pressure extreme occurrence (Loc. 2) has a minimum streamline pressure of -0.8MPa at the left edge streamline and -0.1MPa at the right edge (cf. Figure 7.9b). The spatial position where the first extreme in the minimum pressure drop occurs across the nozzle is seen to be well-above the entrance to the straight nozzle capillary (zone 1) but at the second pressure extreme location, the mean minimum extreme pressure occurs across the die swell region of the flow as shown in Figure 7.10b. This would indicate that the likelihood of void nucleation decreases from the bottom to the upper free surface of the bead. The average extreme maximum pressure at the first peak location (Loc. 1) across streamlines of the nozzle just before zone 1 is seen to be generally less severe than pressure values at the second peak location (Loc. 2), and the mean extreme pressure magnitudes decline asymmetrically with a trough-like appearance from streamlines closer to the edges towards the centerline (cf. Figure 7.9c). The opposite behavior is observed at the second extreme site (Loc. 2) where there is an unsymmetrical rise in the mean extreme pressure magnitude from the edges to the centerline in a crest-like manner (cf. Figure 7.9d), and the

spatial position where this occurs is seen just after the nozzle exit, about .5mm beneath zone 2 almost nearly evenly across the flow (cf. Figure 7.10d). This behavior may be attributed to the relatively high shear rates at the wall just before exiting the nozzle compared to the center streamline which transitions abruptly at the edges.

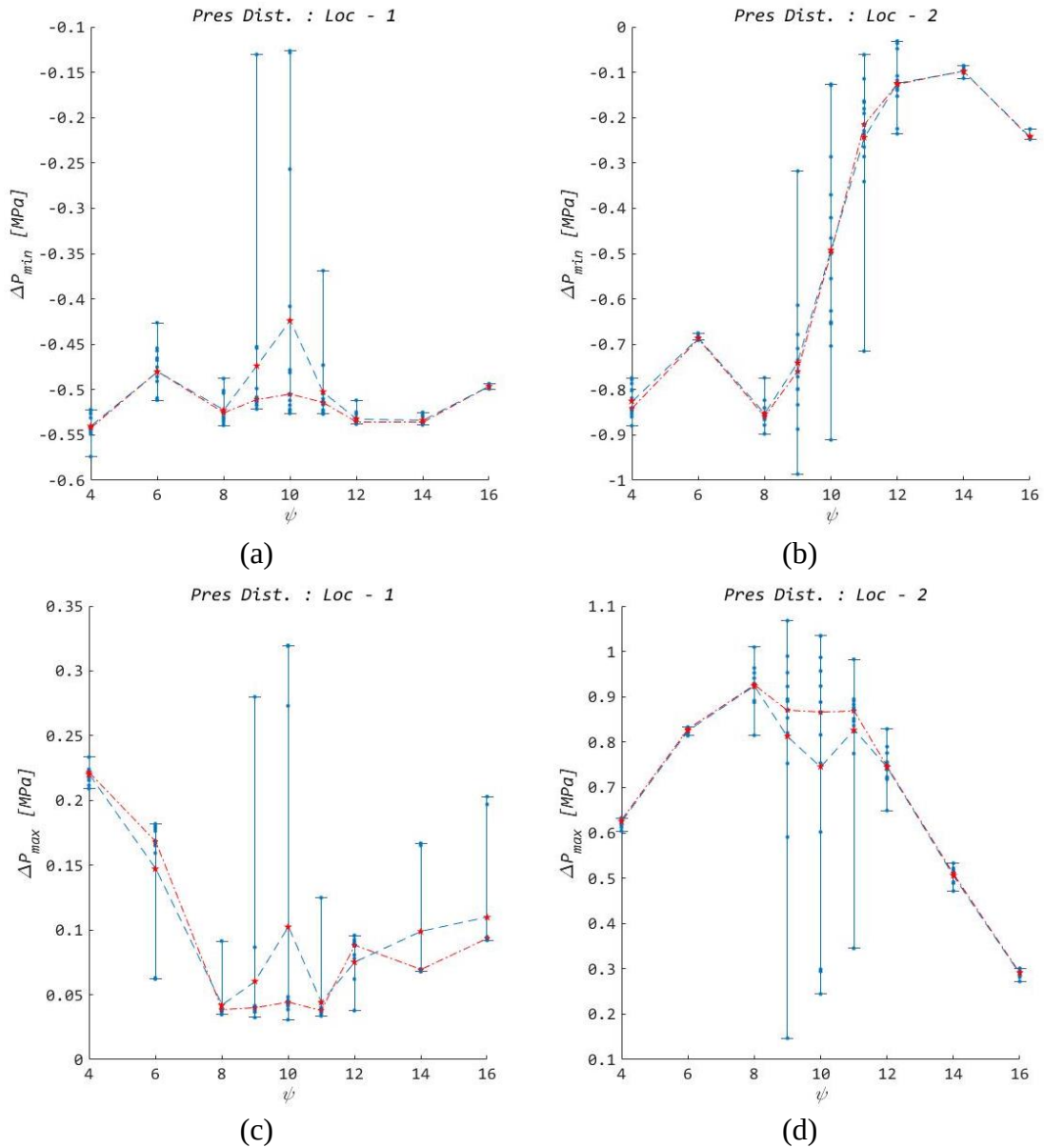


Figure 7.9: Overall pressure extremes on the fiber surface over the complete period of deposition (the blue trendline represents the mean and the red trendline is the median): (a) overall minimum at Loc. 1 (b) overall minimum at the Loc. 2 (c) overall maximum at Loc. 1 (d) overall maximum at Loc. 2.

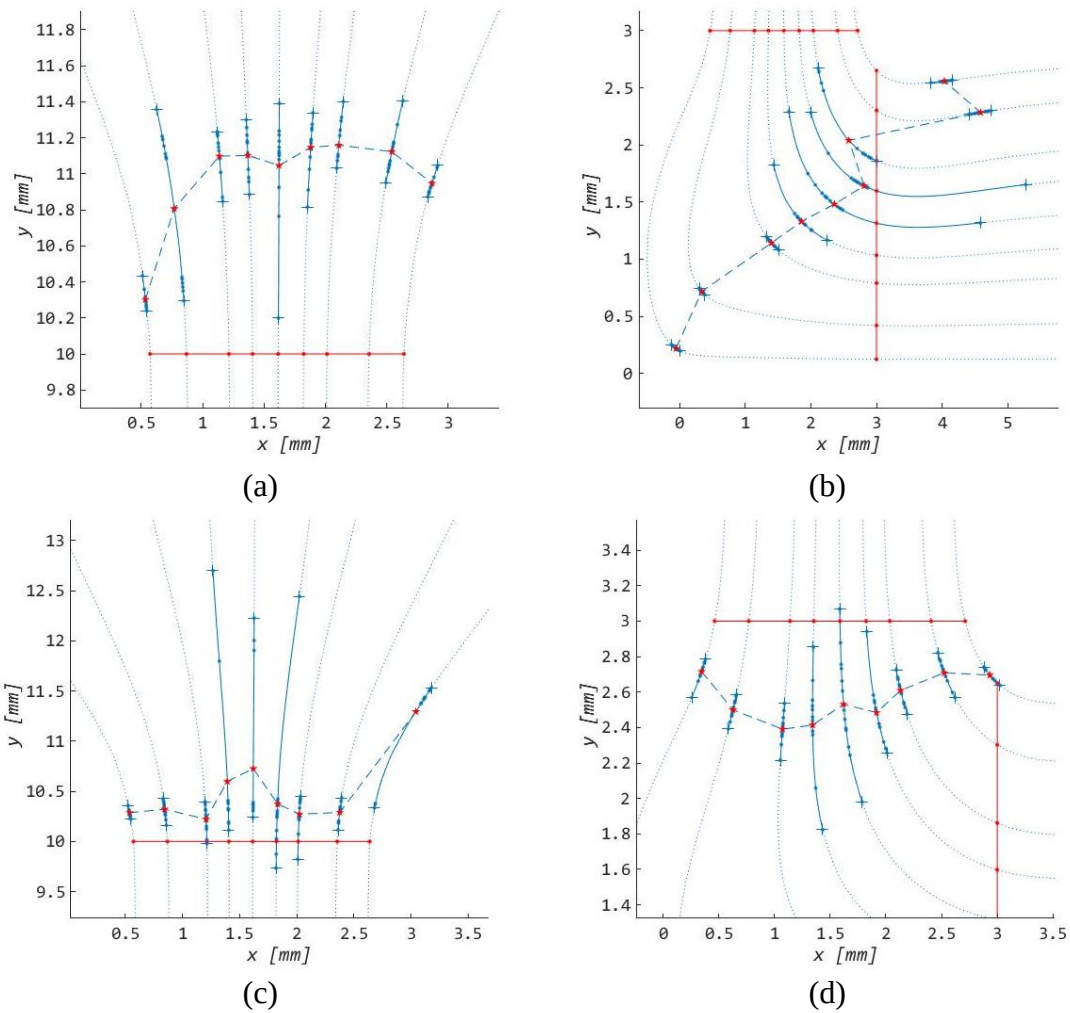


Figure 7.10: Distribution of location within the nozzle where the pressure extremes on the fiber surface occurs over the complete period of deposition and for all computed streamlines: (a) overall minimum at Loc. 1 (b) overall minimum at the Loc. 2 (c) overall maximum at Loc. 1 (d) overall maximum at Loc. 2.

The result of the fiber's orientation distribution relative to the streamline direction at the 3 regions of interest shows that the fiber is almost nearly aligned with the streamlines of the flow across the nozzle section and the degree of alignment increases towards the edge of the nozzle as we observe from Figure 7.11a-c. This is consistent with the conclusion of Saffman [176] who observed that the fibers tend to align with the flow. The

error bounds of the fiber's orientation across the nozzle due to the variation of initial fiber angle in all three locations are also similar.

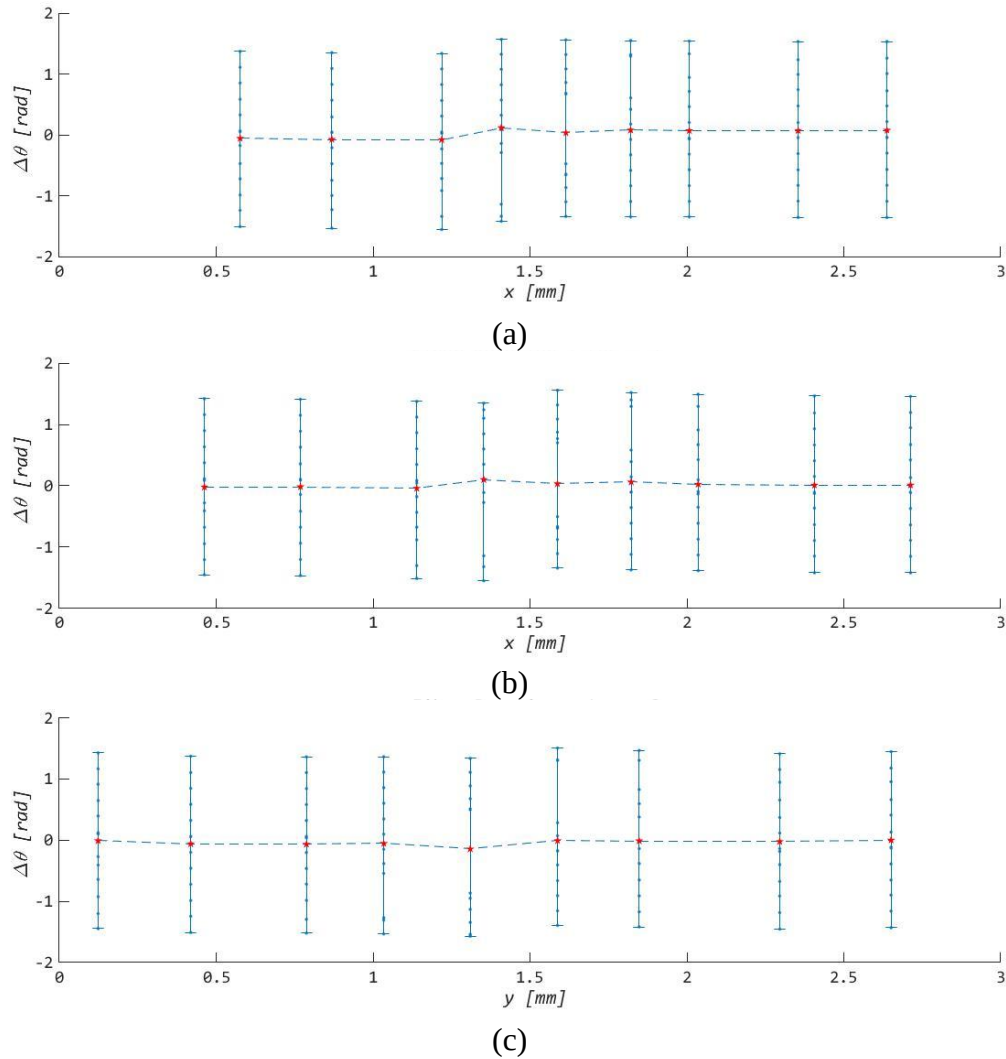


Figure 7.11: Distribution of fiber orientation angle at the region of interest within the extruder nozzle: (a) Zone 1 (b) Zone 2 (c) Zone 3.

7.2.2 Multi-scale non-Newtonian melt flow simulation

Computed results of the velocity magnitudes and shear-rates from the non-Newtonian macroscale analysis are shown in Figure 7.12a & b together with the results from the Newtonian simulation for comparison. The results show relatively higher velocity and shear rate magnitudes for streamlines at the nozzle edges (ψ_4 & ψ_{18}) and lower values towards the centerline (ψ_{10}) for the shear-thinning fluid compared to the Newtonian fluid.

Correspondingly, the deposition times are relatively shorter for streamlines closer to the nozzle edge and relatively longer for streamlines closer to the centerline for the shear-thinning fluid compared to the Newtonian fluid. Likewise, the pressure-drop and pressure gradients across the nozzle are less severe for the shear-thinning fluid compared to the Newtonian fluid [227].

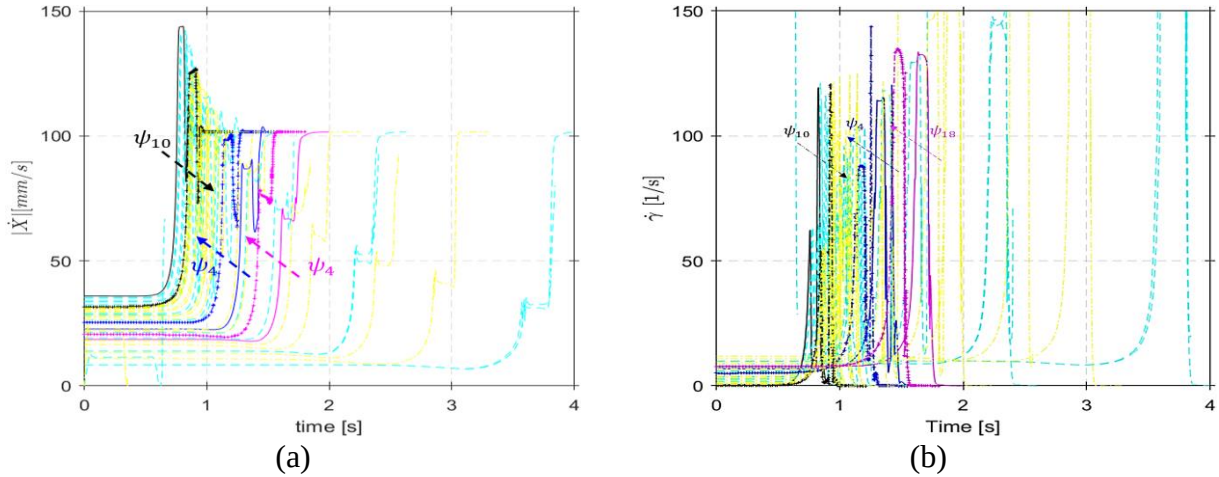


Figure 7.12: showing a) relationship between shear-thinning fluid viscosity and flow shear-rate. Also shown are time-varying profiles along streamline ψ_4 (blue), ψ_{10} (black) and ψ_{18} (pink) for the both Newtonian (continuous line) and non-Newtonian (dotted line) analysis results for b) velocity magnitude c) shear-rate scalar magnitude, and d) pressure distribution.

We present results of the rigid ellipsoidal fiber's motion and surface limit pressure evolution along streamlines of the extrusion-deposition flow for dilute fiber suspension with shear-thinning fluid rheology based on the micro-model non-Newtonian analysis. The results are presented for three (3) feature streamlines i.e. streamline ψ_4 closer the left edge of the nozzle, streamline ψ_{10} at the nozzle center and streamline ψ_{18} at the right edge of the nozzle (cf. Figure 7.1b).

Like the 2D Jeffery studies, we see from Figure 7.13a-c that the fibers angular velocities are unaffected by the shear-thinning fluid rheology irrespective of the non-uniform velocity gradients that characterizes the extrusion-deposition flow-field especially

at the nozzle edges and a Newtonian analysis is sufficient to predict fiber's motion. This is evident from the overlap of the angular velocity profiles for all flow-behavior indices considered. On streamline ψ_4 , the fiber experiences a spin reversal upon exiting the nozzle within the region of die swell due to counter-rotation in the 90° bend that opposes the local shear-vorticity direction at the left inner wall of the straight capillary before returning to steady state during bed deposition. On streamline ψ_{10} , the fiber's motion is steady within the straight capillary due to the uniform flow-field at the center streamline however the angular velocity peaks within the die-swell region due to the change in flow direction. On streamline ψ_{18} the fiber experiences two (2) significant peaks in the angular velocity along the flowpath. The first peak occurs as a result of the severe velocity gradient at the right edge of the nozzle while the latter occurs due to abrupt change in flow direction at the sharp notch where the polymer exits the nozzle. Although, we expect the particle dynamics would be influenced by the shear-thinning fluid rheology in a 3D simulation based on our studies in Chapter 5, however our primary focus here is the particle's surface pressure distribution which our 2D GNF FEA model has been shown to be sufficient for understanding the shear-thinning effect on the pressure response in Chapter 5. Figure 7.13d-e shows that the shear-thinning fluid rheology reduces the magnitude of the fibers surface pressure peaks as the flow behavior index is reduced. The implication of this is that we expect lower probability of void nucleation with higher void formation times for fiber suspension with strong shear-thinning fluid characteristics than for weakly non-Newtonian fiber suspension. The magnitude of minimum pressure drops on the fiber surface are observed to be significantly higher on edge streamlines (ψ_4 , & ψ_{18}) compared to the center streamline (ψ_{10}). The net pressure extremes with respect to the instantaneous streamline

pressure are observed to be higher at the second peak location for streamlines (ψ_4 & ψ_{10}) except on streamline ψ_{18} where the net pressure magnitude is seen to be higher at the first peak location.

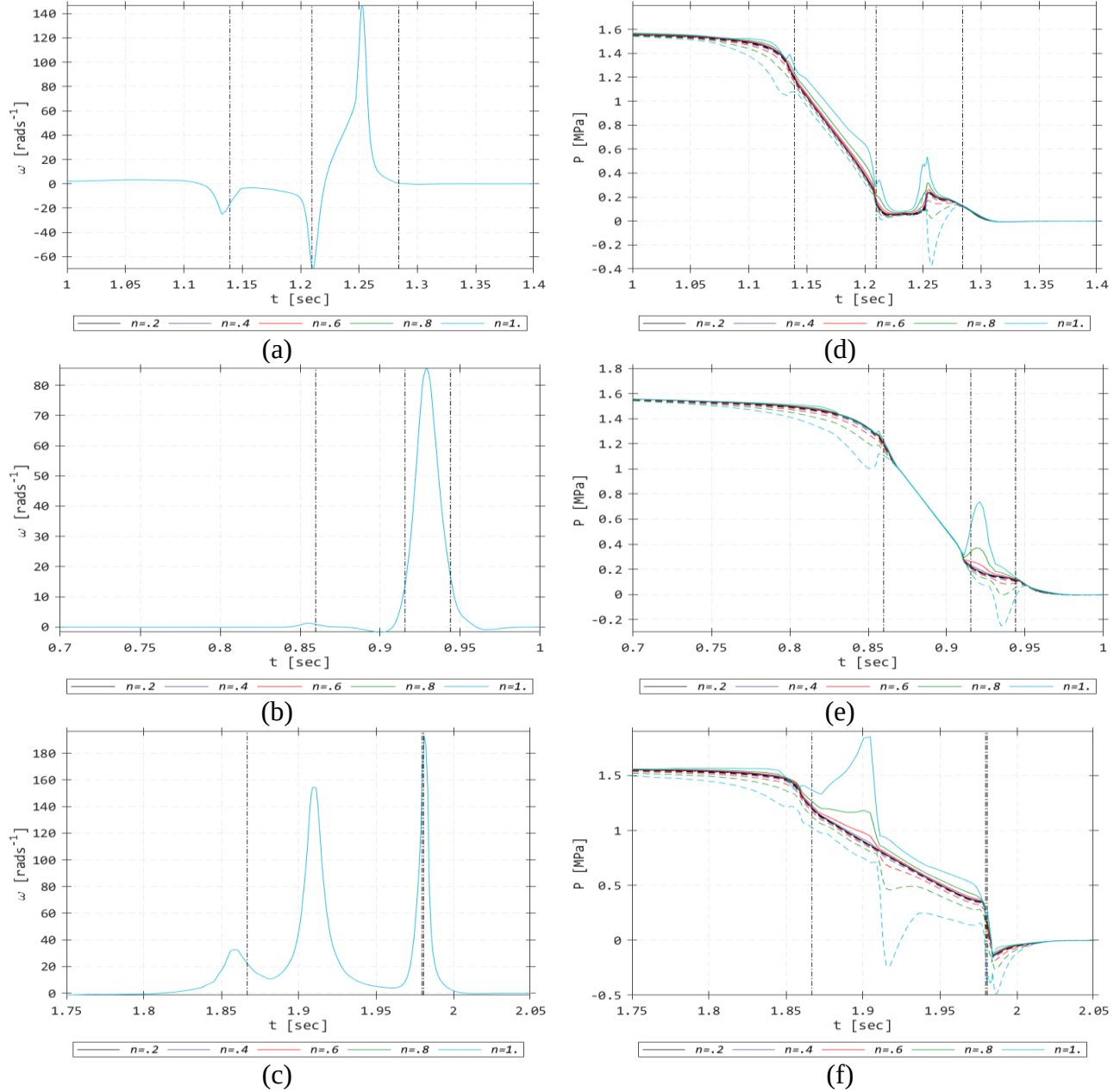


Figure 7.13: Figure showing the time evolution of the net fiber orientation relative to the streamline direction for (a) streamline ψ_4 , (b) streamline ψ_{10} and (c) streamline ψ_{18} . Also shown is the time evolution of the extreme pressure distribution on the fibers surface for (d) streamline ψ_4 , (e) streamline ψ_{10} and (f) streamline ψ_{18} . Results are presented for flow behavior index ranging from $n = 0.2 - 1.0$

Figure 7.14 shows that like the Newtonian case, the peak sites of minimum pressure drop are observed to occur at the fiber's tips when they do occur. Moreover, the deposition times at which the peak pressure magnitudes occur are only slightly modified by the shear-thinning fiber suspension. For the edge streamlines at the second peak location of minimum pressure drop, the time of occurrence are slightly shifted downstream the extrusion-deposition flow while for the center streamline, the time of occurrence is slightly shifted upstream the flow. As such the orientation angle at which the second peak minimum pressure drop on the fibers surface occurs is slightly modified.

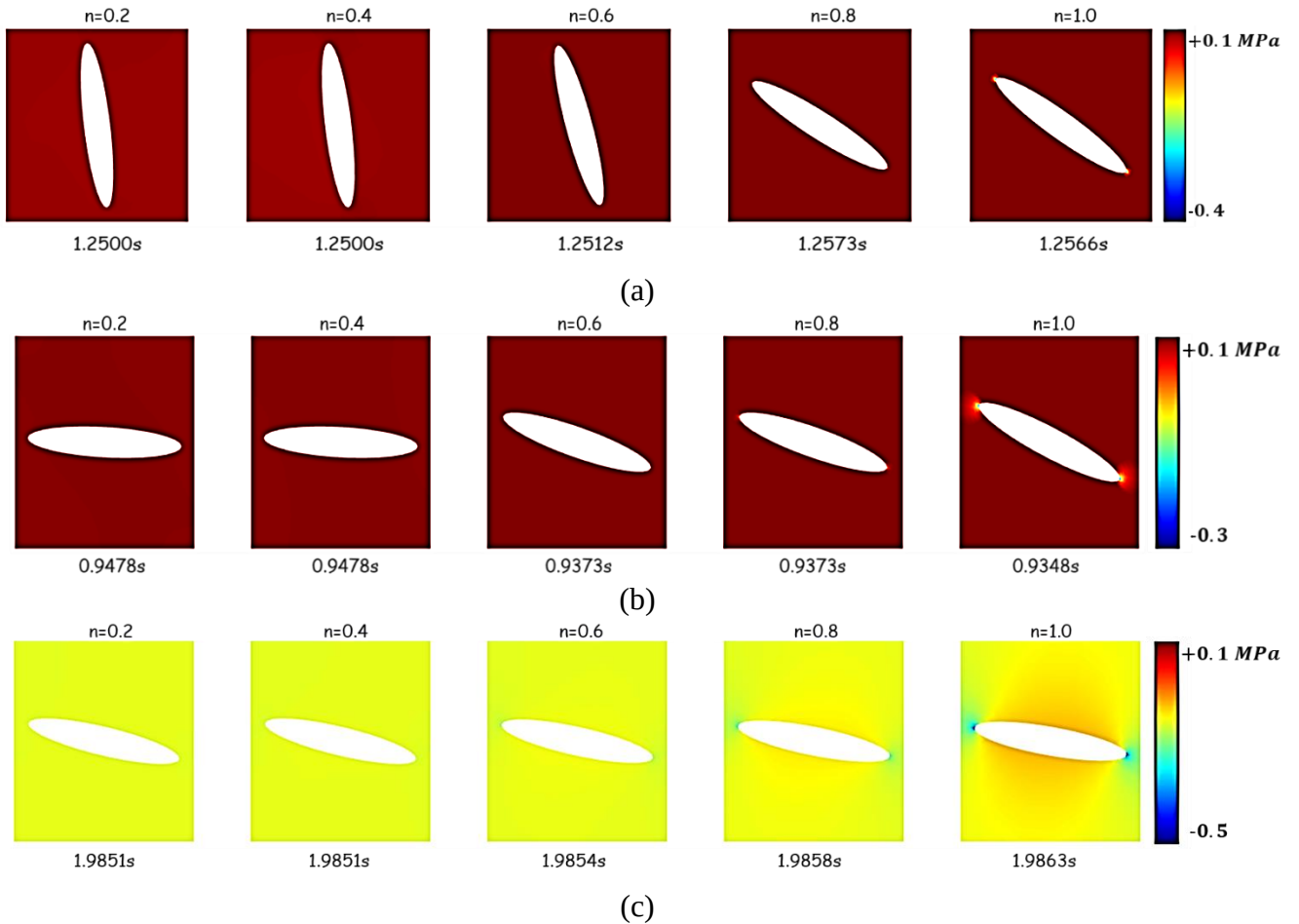


Figure 7.14: Figure showing the pressure distribution around the vicinity of the fiber at the second peak location of minimum pressure drop on the fibers surface for different flow behavior index ranging from $n = 0.2 - 1.0$ and (a) streamline ψ_4 , (b) streamline ψ_{10} and (c) streamline ψ_{18} .

The observed minimum tip surface pressure computed in the simulation above provides fundamental insight into the occurrence of significant tip-voids within CF/ABS EDAM polymer composites as presented in Table 3.2. The occurrence of extreme minimum surface pressure at the tips of a suspended rotating particle potentially explains the high-volume fraction of micro-voids that form at particle ends. Further, the negative fiber tip pressures in the shear dominated flow regions of the EDAM nozzle correspond directly with the observed larger micro-voids in regions of the printed bead specimen close to the bead edges (i.e. ROI-III), as compared to the regions closer to the bead center that encountered a high degree of stretching flow during processing (i.e. ROI-II), shown in Table 3.3 and Figure 3.13. Previous sensitivity studies presented in Chapter 5 revealed various factors that influences the pressure distribution on the fiber surface in CF/ABS EDAM which includes the fluid viscosity μ , shear rate $\dot{\gamma}$, and particle aspect ratio r_e . As we have shown in our non-Newtonian studies, the shear-thinning rheology has no impact on the fibers motion in the 2D simulations however for the 3D simulations (Chapter 5), the particle's motion is seen to be affected by the shear-thinning fluid rheology. In both cases, the shear-thinning fluid rheology influences the fiber surface pressure extremes which decreases as the power law index is reduced. Overall, our multiscale simulation that shows extreme low pressures at the tips of suspended particles as they travel down streamlines of the polymer melt flow and our μ -CT scan results (Chapter 3) indicating that a large majority of micro-voids occur at fiber tips in CF/ABS EDAM samples together provide unique insight into a potential mechanism for micro-void nucleation with short fiber polymer composites.

7.3 Conclusion

A computational multiscale FEA methodology has been developed to study the behavior of suspended rigid ellipsoidal fibers during polymer composite melt extrusion-deposition flow through a LAAM nozzle. Sensitivity analysis based on Jeffery's model assumption reveals a direct correlation between the extreme pressures on the fiber surface with its geometry aspect ratio and the rheological properties of the flow (shear rate and viscosity) and these pressure extremes are observed to occur at the fiber's tips. Further, extreme minimum pressures are shown to occur at the fiber tips as the fiber rotates into alignment with the principal direction of the flow. Results of the extrusion-deposition multi-scale analysis that considers the effect of rotary diffusion due to short-range fiber interaction reveals a dependence of the severity and sensitivity of the fibers extreme pressures to streamline location and the initial fiber orientation. In addition, the effect of increasing fiber concentration and aspect ratio increases the magnitude of the pressure extremes on the fiber surface. In the extrusion-deposition flow, a significant minimum pressure extreme occurs on the fiber surface at the entrance to the straight capillary section and across the die swell region immediately outside of the extruder nozzle which indicates an increased likelihood for micro-voids initiation at fiber ends in these regions. Results indicate that we would expect a higher probability of occurrence of micro-voids closer to the plate than the free surface. Results also confirm a high degree of fiber alignment in the extruded bead. The effect of shear-thinning is seen to decrease the fiber's surface pressure extremes with decreasing power-law index. Based on the classical nucleation theory, we expect lower probability of void nucleation and higher micro-void formation times for strongly non-Newtonian fiber suspension and vice versa. The non-uniform velocity

gradient that characterizes the LAAM nozzle extrusion-deposition flow does not influence the observed effect of the shear-thinning fluid rheology on the fiber dynamics or fiber responses and the peak sites of minimum pressure drop occurs at the fiber's tips as we observed in the Newtonian studies. However, the time interval and corresponding fiber orientation angle at which the peak pressure magnitudes occur are slightly modified in the shear-thinning simulations.

The foregoing discussion on the study of rigid spheroidal particle's motion along streamlines of EDAM polymer composite deposition flow provide insight into potential micro-void mitigation strategies that could exploit the fluid rheological behavior to improve component part quality. Based on the proposed micro-void nucleation mechanism, a potential way for mitigating their formation would leverage factors that reduce the pressure intensity at the fibers tip. One potential way of controlling these micro-voids formation would involve suitable rheological adjustment to reduce the local extreme pressure fluctuations on the fibers surface. On one hand, increasing the shear-thinning intensity may help control the void formations, however increased shear-thinning may increase the likelihood of multiphase flow segregation within nozzle and the create more anisotropy in the microstructure of the printed composite. Our findings from previous chapters also reveal that low curvature radius at the fiber ends is a more relevant parameter that results in exacerbated pressure extremes compared to the fiber length. Our simulation results to this regard agrees with our experimental observation of very high content of heterogenous micro-void nucleation at the tip of fibers compared to their formation elsewhere on the particles surface. Accordingly, proper fiber surface finishing that reduces abrupt changes in fiber geometry and the possible pressure singularities at these sharp

transitions is a possible way to mitigate the micro-void formation. Additionally, our simulation results show that the pressure extremes on the fiber surface across the nozzle are exacerbated at the die-swell region of the nozzle exit where the polymer melt flow makes a sharp 90° turn upon deposition unto the substrate. This agrees with the experimental observations of Yang et al. that found the micro-void content to be highest at the die-swell region compared to other polymer melt flow regions. As such, determining an optimal nozzle tilt angle design to control the flow angle at the nozzle exit may help reduce micro-void formation. A recent simulation-based investigation conducted by Guo et al. [309] showed that adjusting the nozzle tilt angle effectively modifies the flow-field pressure and velocity distribution in the die-swell region and the resulting shear rates and viscosity distribution within this region which could potential help control the micro-void formation within this region.

A future direction to our simulation effort would capture effects typically found in the actual structure of the fiber suspension such as the actual fiber geometry imported from the μ -CT data, the inter-particle and intra-particle hydrodynamic forces, and Brownian effects etc. which all potentially affect the resulting pressure distribution on the fibers surface. Although our simulation efforts have revealed process induced mechanisms that may cause micro-voids there is yet significant knowledge gap in predicting void formation, growth and characteristics via computational based approach which is potential future research opportunity.

APPENDIX

A APPENDIX A

A.1 The Eshelby's (Strain Concentration) Elasticity Tensor

The strain concentration tensor or Eshelby's tensor (Π_{ijkl}) that appears in the Mori Tanaka's model for predicting the homogenized elasticity tensor of the short fiber reinforced composite material is given in this section. Given

$$\eta_m = (1 - \nu_m)^{-1}, \quad \chi_r = (1 - a_r^2)^{-1}, \quad \omega_m = (1 + a_r) + a_r \begin{cases} (-\chi_r)^{1.5} \cos^{-1} a_r & a_r < 1 \\ -\chi_r^{1.5} \cosh^{-1} a_r & a_r > 1 \end{cases} \quad (\text{A. 1})$$

Then for spheroidal inclusions the non-zero components of Π_{ijkl} are given as:

$$\begin{aligned} \Pi_{1111} &= \Pi_{1111} = .50\eta_m[-\chi_r + (4 - 2\nu_m + 3\chi_r)(1 - \omega_m)] \\ \Pi_{1122} &= \Pi_{1133} = .25\eta_m[\chi_r - (2 - 4\nu_m + 3\chi_r)(1 - \omega_m)] \\ \Pi_{2222} &= \Pi_{3333} = .25\eta_m[1.5(1 + \chi_r) + (1 - 2\nu_m - 2.25\chi_r)\omega_m] \\ \Pi_{2233} &= \Pi_{3322} = .25\eta_m[0.5(1 + \chi_r) + (-1 + 2\nu_m - 0.75\chi_r)\omega_m] \\ \Pi_{2211} &= \Pi_{3311} = .25\eta_m[-2.0(1 + \chi_r) + (2 + 2\nu_m + 3.00\chi_r)\omega_m] \\ \Pi_{2323} &= \Pi_{3232} = .25\eta_m[0.5(1 + \chi_r) + (1 - 2\nu_m - 0.75\chi_r)\omega_m] \\ \Pi_{1212} &= \Pi_{1313} = .25\eta_m[-2.(\nu_m + \chi_r) + (1 + \nu_m + 3.00\chi_r)\omega_m] \end{aligned} \quad (\text{A. 2})$$

For spherical shaped inclusions, the non-zero components of the Eshelby tensor are given as

$$\begin{aligned} \Pi_{1111} &= \Pi_{2222} = \Pi_{3333} = \eta_m[7 - 5\nu_m]/15 \\ \Pi_{1122} &= \Pi_{1133} = \Pi_{2211} = \Pi_{3311} = \Pi_{2233} = \Pi_{3322} = \eta_m[-1 + 5\nu_m]/15 \\ \Pi_{1212} &= \Pi_{1313} = \Pi_{2323} = \eta_m[4 - 5\nu_m]/15 \end{aligned} \quad (\text{A. 3})$$

B APPENDIX B

B.1 Definition of Constants in Jeffery's Equation

The expressions of the components of the variable vector $\underline{\chi}$ and coefficient tensors $\underline{\underline{A}}^I$, $\underline{\underline{A}}^{II}$ & $\underline{\underline{A}}^{III}$ that appear in the definition of the Jeffery's velocity and pressure are defined in eqns. (B. 1) -(B. 5) below. For the variable vector $\underline{\chi}$ the components are given as

$$\chi_1 = \Psi'_1 X_2 X_3, \quad \chi_2 = \Psi'_2 X_3 X_1, \quad \chi_3 = \Psi'_3 X_1 X_2 \quad (\text{B. 1})$$

The components in $\underline{\underline{A}}^I$ vector is likewise given as

$$R = -\frac{\Gamma_{23}}{\Psi''_{1_0}}, \quad S = -\frac{\Gamma_{13}}{\Psi''_{2_0}}, \quad T = -\frac{\Gamma_{12}}{\Psi''_{3_0}} \quad (\text{B. 2})$$

The components in $\underline{\underline{A}}^{II}$ tensor is given as

$$U = 2[\mathcal{H}_2^2 B - \mathcal{H}_3^2 C], \quad V = 2[\mathcal{H}_3^2 C - \mathcal{H}_1^2 A], \quad W = 2[\mathcal{H}_1^2 A - \mathcal{H}_2^2 B] \quad (\text{B. 3})$$

where the coefficients A, B, C in eqn. (B. 3) above are also components of tensor $\underline{\underline{A}}^{III}$ containing the stresslets and torque acting on the rigid ellipsoidal particle suspended in linear ambient flow-field [315]. given in eqn. (B. 4) below

$$\begin{aligned} A &= \frac{1}{6} \left\{ \frac{2\Psi''_{1_0}\Gamma_{11} - \Psi''_{2_0}\Gamma_{22} - \Psi''_{3_0}\Gamma_{33}}{\Psi''_{2_0}\Psi''_{3_0} + \Psi''_{3_0}\Psi''_{1_0} + \Psi''_{1_0}\Psi''_{2_0}} \right\}, & F &= \frac{\Psi_{2_0}\Gamma_{23} - \mathcal{H}_3^2\Psi'_{1_0}(\mathcal{E}_1 - \Psi_1)}{2\Psi'_{1_0}(\mathcal{H}_2^2\Psi_{2_0} + \mathcal{H}_3^2\Psi_{3_0})}, \\ F' &= \frac{\Psi_{3_0}\Gamma_{23} + \mathcal{H}_2^2\Psi'_{1_0}(\mathcal{E}_1 - \Psi_1)}{2\Psi'_{1_0}(\mathcal{H}_2^2\Psi_{2_0} + \mathcal{H}_3^2\Psi_{3_0})} \\ B &= \frac{1}{6} \left\{ \frac{2\Psi''_{2_0}\Gamma_{22} - \Psi''_{3_0}\Gamma_{33} - \Psi''_{1_0}\Gamma_{11}}{\Psi''_{2_0}\Psi''_{3_0} + \Psi''_{3_0}\Psi''_{1_0} + \Psi''_{1_0}\Psi''_{2_0}} \right\}, & G &= \frac{\Psi_{3_0}\Gamma_{13} - \mathcal{H}_1^2\Psi'_{2_0}(\mathcal{E}_2 - \Psi_2)}{2\Psi'_{2_0}(\mathcal{H}_3^2\Psi_{3_0} + \mathcal{H}_1^2\Psi_{1_0})}, \\ G' &= \frac{\Psi_{1_0}\Gamma_{13} + \mathcal{H}_3^2\Psi'_{2_0}(\mathcal{E}_2 - \Psi_2)}{2\Psi'_{2_0}(\mathcal{H}_3^2\Psi_{3_0} + \mathcal{H}_1^2\Psi_{1_0})} \\ C &= \frac{1}{6} \left\{ \frac{2\Psi''_{3_0}\Gamma_{33} - \Psi''_{1_0}\Gamma_{11} - \Psi''_{2_0}\Gamma_{22}}{\Psi''_{2_0}\Psi''_{3_0} + \Psi''_{3_0}\Psi''_{1_0} + \Psi''_{1_0}\Psi''_{2_0}} \right\}, & H &= \frac{\Psi_{1_0}\Gamma_{12} - \mathcal{H}_2^2\Psi'_{3_0}(\mathcal{E}_3 - \Psi_3)}{2\Psi'_{3_0}(\mathcal{H}_1^2\Psi_{1_0} + \mathcal{H}_2^2\Psi_{2_0})}, \\ H' &= \frac{\Psi_{2_0}\Gamma_{12} + \mathcal{H}_1^2\Psi'_{3_0}(\mathcal{E}_3 - \Psi_3)}{2\Psi'_{3_0}(\mathcal{H}_1^2\Psi_{1_0} + \mathcal{H}_2^2\Psi_{2_0})} \end{aligned} \quad (\text{B. 4})$$

The integral constants $\mathcal{V}_j$ and their symmetric forms are defined as [19]

$$\mathcal{V}_j = \int_{\lambda}^{\infty} \frac{1}{\Delta} \frac{d\lambda}{(\mathcal{H}_j^2 + \lambda)}, \quad \mathcal{V}'_j = \int_{\lambda}^{\infty} \frac{1}{\Delta^3} (\mathcal{H}_j^2 + \lambda) d\lambda, \quad \mathcal{V}''_j = \int_{\lambda}^{\infty} \frac{1}{\Delta^3} (\mathcal{H}_j^2 + \lambda) \lambda d\lambda \quad (\text{B. 5})$$

A constant $\mathcal{V}_j$ subscripted with 0, i.e. $\mathcal{V}_{j_0}$ implies that the lower limit of integration $\lambda = 0$.

B.2 Two-Dimensional (2D) Reduced Form of Jeffery's Equation

The 2D contracted form of the Jeffery's pressure and velocity [19] can be expressed by eqns. (B. 6) & (B. 7) respectively given as

$$p = p_0 + 2\mu_1 \underline{\underline{A}}_{ij}^V \nabla_{X_i} \nabla_{X_j} \Omega \quad (\text{B. 6})$$

$$\dot{X}_i = \dot{X}_i^{\infty} + \underline{\underline{A}}_{ij}^{IV} \nabla_{X_j} \chi_3 + \underline{\underline{A}}_{jk}^V X_k \nabla_{X_i} \nabla_{X_j} \Omega - \underline{\underline{A}}_{ij}^V \nabla_{X_j} \Omega \quad (\text{B. 7})$$

where p_0 is the constant mean pressure at a distance from the ellipsoid, $\dot{X}_i$ are the velocity components at arbitrary position $\underline{X} = [X_1 \ X_2]^T$ and $\dot{X}_i^{\infty}$ is the velocity of the undisturbed fluid at $\underline{X}$ given as

$$\dot{X}_i^{\infty} = L_{ij} X_j \quad (\text{B. 8})$$

coefficient matrices $\underline{\underline{A}}^{IV}$ and $\underline{\underline{A}}^V$ and the gradient operators $\underline{\nabla}_X$ are respectively given as

$$\underline{\underline{A}}^{IV} = \begin{bmatrix} Y & W \\ -W & Y \end{bmatrix}, \quad \underline{\underline{A}}^V = \begin{bmatrix} A & H \\ H' & B \end{bmatrix}, \quad \underline{\nabla}_X = [\partial/\partial X_1 \ \partial/\partial X_2]^T$$

The 2D strain deformation tensor in the local fiber reference frame L_{ij} is decomposed in the usual way to obtain the 2D symmetric component Γ_{ij} and anti-symmetric components Ξ_{ij} according to

$$L_{ij} = \Gamma_{ij} + \Xi_{ij} \quad (\text{B. 9})$$

$$\Gamma_{ij} = L_{ij} + L_{ji}, \quad \Xi_{ij} = [L_{ij} + L_{ji}] = \Xi_3^{-1} \mathcal{B}_{ij}, \quad \mathcal{B}_{ij} = i - j \quad (\text{B. 10})$$

The 2D Laplace Function Ω that appears in eqns. (B. 6) & (B. 7) is defined as

$$\Omega = \int_{\lambda}^{\infty} \frac{1}{\Delta} \left\{ \sum_{j=1}^2 \frac{X_j^2}{\mathcal{H}_j^2 + \lambda} - 1 \right\} d\lambda \quad (\text{B. 11})$$

where

$$\Delta^2 = \prod_{j=1}^2 (\mathbb{H}_j^2 + \lambda), \quad \text{and}, \quad \lambda : \sum_{j=1}^2 \frac{X_j^2}{\mathbb{H}_j^2 + \lambda} = 1 \quad (\text{B. 12})$$

At the fiber's surface where $\lambda = 0$, the field velocity must equal the fiber's surface velocity assuming no slip at the fiber's surface, i.e.

$$\dot{X}_i^p = \dot{X}_i \big|_{\lambda=0} = \Psi_3 \mathbb{E}_{ij} X_j \quad (\text{B. 13})$$

The constants that appear in $\underline{\underline{A}}^{IV}$, $\underline{\underline{A}}^V$ above are thus obtained as

$$A = -B = \frac{\Gamma_{11}}{4\mathbb{Q}_{3_0}''}, \quad H = H' = \frac{1}{2} \left[\frac{\Xi_3 - \Psi_3}{\mathbb{Q}_{1_0} - \mathbb{Q}_{2_0}} \right], \quad Y = -\frac{\Gamma_{12}}{\mathbb{Q}_{3_0}'}, \quad W = 2(\mathbb{H}_1^2 + \mathbb{H}_2^2)A \quad (\text{B. 14})$$

where $\mathbb{Q}_j$, $\mathbb{Q}_j'$, $\mathbb{Q}_j''$ & χ_3 retain their usual definition given in eqn. (B. 1) & (B. 5). above. The fibers angular velocity is derived as

$$\Psi_3 = \Xi_3 + M_{33}D_3, \quad D_3 = \Gamma_{12} \quad (\text{B. 15})$$

B.3 Flow-Regimes in Typical EDAM Nozzle

Polymer composite melt flow through the nozzle in typical EDAM polymer composite processing is characterized by complex combination of shear and extensional deformation rate components that are dependent on the viscoelastic polymer melt rheology and the geometry of the extrusion nozzle. The flow condition at the nozzle wall is pure shear and at the nozzle centreline is pure uniaxial elongation (cf. Figure B. 1) [136], [349]. Away from the convergent zone in the lubrication zone defined by the clearance between the screw edge and the nozzle walls, the flow is predominantly shear dominant while close to the centreline and near the entrance of the nozzle where the flow undergoes acceleration due to geometric constriction, the flow is dominated by extensional rate, and at the vortices created near the notch edges with sharp transitions due to elastic instabilities, the flow is mainly rotational [136]. The flow contraction region consists of a complex combination of the various flow categories with varying dominance.

A simple metric used to classify the flow regimes is based on a flow parameter $\bar{v}$ given by [136].

$$\bar{v} = \frac{\dot{\gamma}_c + j\omega_c}{\dot{\gamma}_c - j\omega_c} \quad (\text{B. 16})$$

where $\dot{\gamma}_c$ is the magnitude of deformation rate tensors defined as $\dot{\gamma}_c = \sqrt{2\Gamma_{ij}\Gamma_{ji}}$ and ω_c is the magnitude of the vorticity tensor given as $\omega_c = \sqrt{2\Xi_{ij}\Xi_{ji}}$. The flow is pure shear when $\bar{v} = 0$, pure elongational when $\bar{v} = 1$, and purely rotational when $\bar{v} = -1$. Typical flow patterns within the convergent zone results in $\bar{v}$ lying between $-1 \leq \bar{v} \leq 1$.

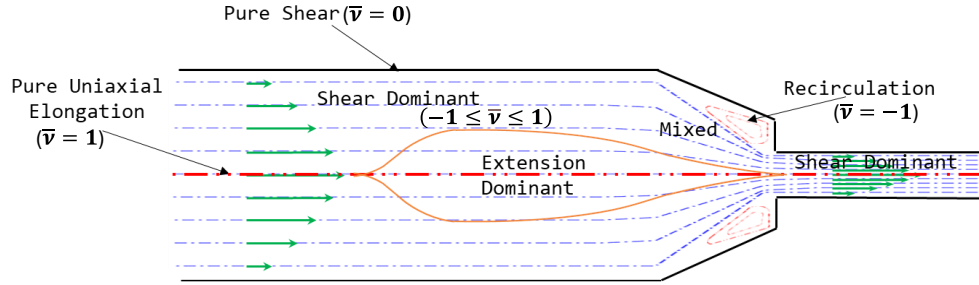


Figure B. 1 Schematic showing flow regimes within a typical EDAM nozzle during polymer processing.

B.4 Obtaining Particle Stall Orientation Angles in Newtonian Homogenous Flows

The particle stall angles under favorable conditions in general class of homogenous flows can be obtained using the tensorial representation for the particle orientation of an axisymmetric ellipsoidal particle in viscous suspension with velocity gradient $\underline{\underline{L}}$ developed by Dinh et al. [244] based on Jeffery's model assumptions and is given as

$$\dot{\rho}_i = -\Xi_{ij}\rho_j + \kappa(\Gamma_{ij}\rho_j - \Gamma_{kl}\rho_k\rho_l\rho_i) \quad (\text{B. 17})$$

where $\underline{\rho}$ is the particle orientation defined by the vector:

$$\underline{\rho} = [\cos \theta \quad \sin \theta \sin \phi \quad \sin \theta \cos \phi]^T \quad (\text{B. 18})$$

The Euler angles and angular velocities can be backtracked from the rate of the orientation vectors $\underline{\dot{\rho}}$ thus:

$$\phi = \tan^{-1} \frac{\rho_2}{\rho_3}, \quad \theta = \cos^{-1} \rho_1, \quad \dot{\phi} = \frac{\dot{\rho}_3}{\rho_3} \left[\frac{\dot{\rho}_2}{\dot{\rho}_3} - \frac{\rho_2}{\rho_3} \right] \left[1 + \frac{\rho_2^2}{\rho_3^2} \right]^{-1}, \quad \dot{\theta} = -\dot{\rho}_1 (1 - \rho_1^2)^{-1/2} \quad (\text{B. 19})$$

Considering the normalization condition, the independent components of the particle orientation at stall can likewise be obtained via the Newton-Raphson numerical iterative process according to eqn. (B. 20) below

$$\underline{\rho}_s^+ = \underline{\rho}_s^- - \underline{J}_{\theta_2}^{-1} \underline{\dot{\theta}}^\rho \quad (\text{B. 20})$$

where $\underline{\rho}_s = [\rho_2^s \ \rho_3^s]^T$, $\sum_j \rho_j = 1$, $\underline{\dot{\theta}}^\rho = [\dot{\phi} \ \dot{\theta}]^T$, and the components of the Jacobian $\underline{J}_{\theta_2}$

are explicitly defined in eqns. (B. 21) - (B. 24) below

$$\underline{J}_{\theta_2,11} = \frac{1}{\rho_3} \left\{ J_{\rho,21} - J_{\rho,31} \frac{\rho_2}{\rho_3} - \frac{\dot{\rho}_3}{\rho_3} \left[1 + 2 \left(\frac{\dot{\rho}_2}{\dot{\rho}_3} - \frac{\rho_2}{\rho_3} \right) \left(\frac{\rho_2}{\rho_3} + \frac{\rho_3}{\rho_2} \right)^{-1} \right] \right\} \left[1 + \frac{\rho_2^2}{\rho_3} \right]^{-1} \quad (\text{B. 21})$$

$$\underline{J}_{\theta_2,12} = \frac{1}{\rho_3} \left\{ J_{\rho,22} - J_{\rho,32} \frac{\rho_2}{\rho_3} - \frac{\dot{\rho}_3}{\rho_3} \frac{\dot{\rho}_2}{\rho_3} \right. \quad (\text{B. 22})$$

$$\left. + 2 \frac{\dot{\rho}_3}{\rho_3} \frac{\rho_2}{\rho_3} \left[1 + \left(\frac{\dot{\rho}_2}{\dot{\rho}_3} - \frac{\rho_2}{\rho_3} \right) \left(\frac{\rho_2}{\rho_3} + \frac{\rho_3}{\rho_2} \right)^{-1} \right] \right\} \left[1 + \frac{\rho_2^2}{\rho_3} \right]^{-1}$$

$$\underline{J}_{\theta_2,21} = [-J_{\rho,11}(1 - \rho_1^2) + \dot{\rho}_1](1 - \rho_1^2)^{-3/2} \quad (\text{B. 23})$$

$$\underline{J}_{\theta_2,22} = [-J_{\rho,12}(1 - \rho_1^2) + \dot{\rho}_1](1 - \rho_1^2)^{-3/2} \quad (\text{B. 24})$$

and the tensor $\underline{J}_\rho$ is computed from eqn. (B. 25) below

$$\underline{J}_\rho = -\underline{\underline{\varepsilon}} \underline{\underline{\Omega}} + \kappa \left[\underline{\underline{\Gamma}} - \underline{\rho} \underline{\rho}^T \left(\underline{\underline{\Gamma}} + \underline{\underline{\Gamma}}^T \right) - \left(\underline{\rho}^T \underline{\underline{\Gamma}} \underline{\rho} \right) \underline{\underline{I}} \right] \underline{\underline{\Omega}}, \quad \underline{\underline{\Omega}}^T = \begin{bmatrix} -1 & 1 & 0 \\ -1 & 0 & 1 \end{bmatrix} \quad (\text{B. 25})$$

B.5 Principal Flow Directions

The principal flow directions can be obtained by spectral decomposition of the symmetric part of the velocity gradient tensor $\underline{\underline{\Gamma}}$. The respective eigenvectors $\underline{\Phi}^k$ are the principal flow directions. i.e.

$$\underline{\underline{\Phi}} \mid \Gamma_{mn} = \Phi_{mk} \Lambda_{kl} \Phi_{nl}, \quad \Lambda_{kl} = \begin{cases} \lambda_k & k = l \\ 0 & k \neq l \end{cases}, \quad \underline{\underline{\Phi}} \mid \underline{\underline{\Gamma}} = \underline{\underline{\Phi}} \underline{\underline{\Lambda}} \underline{\underline{\Phi}}^T \quad (\text{B. 26})$$

Considering the in-plane homogenous flow velocity gradient of eqn. (5.70), the principal flow directions in the shear plane irrespective of coordinate reference frame are obtained as

$$\tan \phi_p = \frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}} \pm \sqrt{\frac{\dot{\epsilon}_2 - \dot{\epsilon}_3}{\dot{\gamma}}^2 + 1} \quad (\text{B. 27})$$

As would be seen from the simulation results, for a particle tumbling in the flow shear-plane, the particle orientation at the location of minimum pressure extreme on particle's surface corresponds to position of particle alignment with one of the principal flow directions in the flow shear plane. i.e.

$$\underline{\rho} \Big|_{p=p_{max}} = \underline{\Phi}^k \quad \theta = 0 \quad (\text{B. 28})$$

Hence the peak pressure occurs at an instant t_p such that $\phi(t_p) = \phi_p$.

B.6 Optimization of Jeffery's equation for Center Gated Disk Flow

The center gated disk axisymmetric flow finds application in fiber orientation modelling in injection molding system and involves a pressure-gradient that drives the flow of fluid between two parallel plates such that the flow diverges radially outwardly from the inlet gate [352]. The velocity solutions are developed from the lubrication approximation based on the assumption of Newtonian fluid property and constant flow rate with no temperature gradient and the solutions are valid at radial distance X_{0r} much greater than the gap thickness $\mathbb{h}$ (i.e., $X_{0r}/\mathbb{h} \gg 1$) [291]. The velocity profiles are fully developed at the disk inlet with reduced curvature at greater radial distances. The velocity gradient varies with time and is characterized by combined spatially dependent shearing and planar elongation components. Analytical and numerical solutions for accurately predicting the fiber orientation currently exists and have been well developed by various researchers [291], [332], [352].

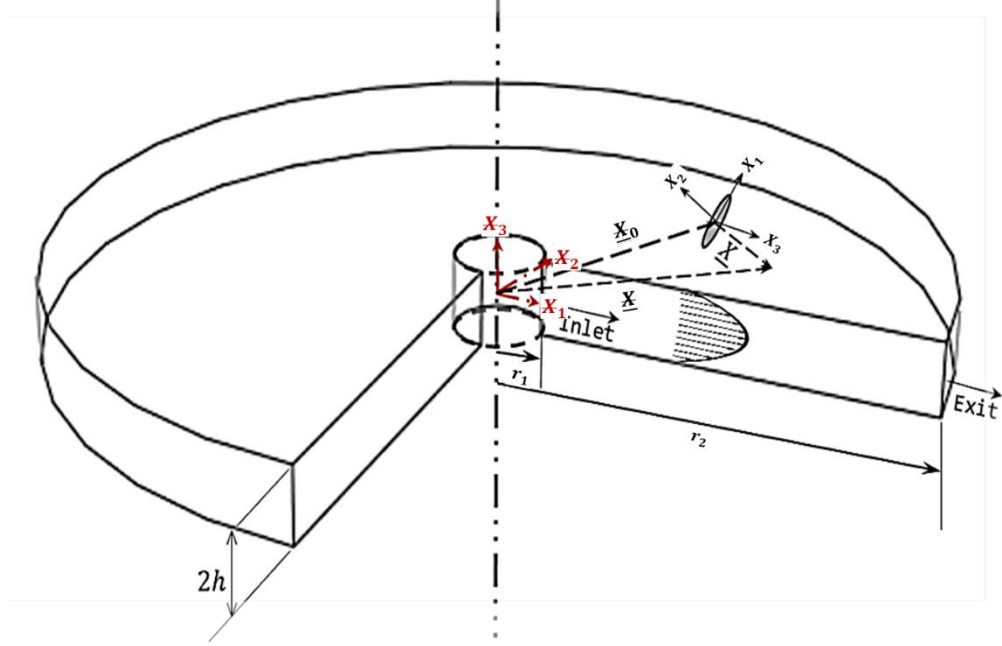


Figure B. 2 Schematic representation of a center gated disk with relevant annotations

Here we utilize Jeffery's model development to study the pressure distribution at the fiber's interface based on this flow condition that may provide insight into micro-void formation injection molding processing of polymer composites. For this flow type, the Jeffery' pressure are not only dependent on the spatial position vector $\underline{X}$ in the local fiber reference frame relative to its center and the fiber orientation angles $\underline{\Theta}$, but also the position vector of the fiber centroid $\underline{X}_0$ from the global origin at the gate entry is an additional unknown. We thus need to find the partial derivative of the pressure p with respect to $\underline{X}_0$ in addition to $\underline{X}$ and $\underline{\Theta}$. In the global reference frame, the absolute position vector of an arbitrary point away from the fiber surface is given as

$$\underline{X}_j = \underline{X}_{0j} + Z_{X_{jk}} X_k \quad (\text{B. 29})$$

where the spatial quantities have been normalized with the gap height $\mathbb{h}$. The normalized velocity components at $\underline{X}_0$ for this flow type is given in cylindrical coordinates as

$$\dot{X}_{0r} = \frac{1}{X_{0r}} [1 - X_{0z}^2], \quad \dot{X}_{0\theta} = \dot{X}_{03} = 0 \quad (\text{B. 30})$$

where X_{0_r} and X_{0_3} are the normalized radial and vertical distance and $\mathbb{X}_{0_r} = \sqrt{\mathbb{X}_{0_1}^2 + \mathbb{X}_{0_2}^2}$. The actual non dimensionless velocity components are $\dot{\mathbb{X}}'_{0_r} = \overline{\mathbb{U}} \dot{\mathbb{X}}_{0_r}$, $\dot{\mathbb{X}}'_{0_\theta} = \dot{\mathbb{X}}'_{0_3} = 0$, where $\overline{\mathbb{U}} = 3\mathbb{Q}/8\pi\mathbb{h}^2$ is the average velocity. The non-zero components of the velocity gradient and its partial derivative with respect to the radial distance in terms of the normalized variables is thus given as [352].

$$L_{11} = -L_{22} = \frac{\partial \dot{\mathbb{X}}_{0_r}}{\partial \mathbb{X}_{0_r}} = -\frac{\dot{\mathbb{X}}_{0_r}}{\mathbb{X}_{0_r}}, \quad L_{13} = \frac{\partial \dot{\mathbb{X}}_{0_r}}{\partial \mathbb{X}_{0_3}} = -2 \frac{\mathbb{X}_{0_3}}{\mathbb{X}_{0_r}} \quad (\text{B. 31})$$

The non-dimensionless equivalent of velocity gradient L_{ij} in eqn. (B. 31) above can be obtained from $L'_{ij} = \overline{\mathbb{U}}/\mathbb{h} L_{ij}$. Additional optimization constraints to those given in eqns. (5.20) - (5.21) for this flow type would include.

$$r_1 \leq \mathbb{X}_{0_r} \leq r_2, \quad 0 \leq \mathbb{X}_{0_3} \leq 1 \quad (\text{B. 32})$$

The velocity gradient L for this flow type has been provided in eqn. (B. 31). The first derivative of the non-zero component of the of the velocity gradient L with respect to the global position vector $\underline{X}_0$ is given as

$$\underline{\nabla}_{\underline{X}_0} L_{11} = -\underline{\nabla}_{\underline{X}_0} L_{22} = 2 \frac{\dot{\mathbb{X}}_{0_r}}{\mathbb{X}_{0_r}^3} \underline{X}_0, \quad \underline{\nabla}_{\underline{X}_0} L_{13} = 2 \frac{\mathbb{X}_{0_3}}{\mathbb{X}_{0_r}^3} \begin{bmatrix} \mathbb{X}_{0_1} \\ \mathbb{X}_{0_2} \\ -\mathbb{X}_{0_r}^2 \mathbb{X}_{0_3}^{-1} \end{bmatrix} \quad (\text{B. 33})$$

Likewise, the second derivatives of the non-zero component of the of the velocity gradient L with respect to the global position vector $\underline{X}_0$ is given as

$$\begin{aligned} \underline{\nabla}_{\underline{X}_0}^2 L_{11} = -\underline{\nabla}_{\underline{X}_0}^2 L_{22} &= -2 \frac{1}{\mathbb{X}_{0_r}^5} \begin{bmatrix} \dot{\mathbb{X}}_{0_r} (3\mathbb{X}_{0_1}^2 - \mathbb{X}_{0_2}^2) & 4\dot{\mathbb{X}}_{0_r} \mathbb{X}_{0_1} \mathbb{X}_{0_2} & 2\dot{\mathbb{X}}_{0_r} \mathbb{X}_{0_1} \mathbb{X}_{0_3} \\ & \dot{\mathbb{X}}_{0_r} (3\mathbb{X}_{0_2}^2 - \mathbb{X}_{0_1}^2) & 2\dot{\mathbb{X}}_{0_r} \mathbb{X}_{0_2} \mathbb{X}_{0_3} \\ & & -\mathbb{X}_{0_r}^3 \end{bmatrix} \\ \underline{\nabla}_{\underline{X}_0}^2 L_{13} &= -2 \frac{1}{\mathbb{X}_{0_r}^5} \begin{bmatrix} (2\mathbb{X}_{0_1}^2 - \mathbb{X}_{0_2}^2) \mathbb{X}_{0_3} & 3\mathbb{X}_{0_1} \mathbb{X}_{0_2} \mathbb{X}_{0_3} & -\mathbb{X}_{0_r}^2 \mathbb{X}_{0_1} \\ & (2\mathbb{X}_{0_2}^2 - \mathbb{X}_{0_1}^2) \mathbb{X}_{0_3} & -\mathbb{X}_{0_r}^2 \mathbb{X}_{0_2} \\ & & 0 \end{bmatrix} \end{aligned} \quad (\text{B. 34})$$

The non-dimensionless equivalent of the above derivatives in eqn. (B. 33) & (B. 34) above are given as

$$\underline{\nabla}_{\underline{X}_0} \underline{\mathcal{L}}'_{ij} = \frac{\overline{\mathbb{U}}}{\mathbb{h}^2} \underline{\nabla}_{\underline{X}_0} \underline{\mathcal{L}}_{ij}, \quad \underline{\nabla}_{\underline{X}_0}^2 \underline{\mathcal{L}}'_{ij} = \frac{\overline{\mathbb{U}}}{\mathbb{h}^3} \underline{\nabla}_{\underline{X}_0}^2 \underline{\mathcal{L}}_{ij} \quad (\text{B. 35})$$

The Jeffery's pressure equation in terms of the independent variable vectors of differentiation $\underline{X}_0$, $\underline{X}$ and $\underline{\Theta}$ for this flow type as

$$p(\underline{X}_0, \underline{X}, \underline{\Theta}) = p_0 + \mathbb{h}^{-1} \varpi_j(\underline{X}_0, \underline{\Theta}) \Delta_j(\underline{X}) \quad (\text{B. 36})$$

The gradient vector of the Jeffery's pressure with respect to $\underline{X}_0$, $\underline{X}$ and $\underline{\Theta}$ is thus given as

$$\underline{\nabla} p = \mathbb{h}^{-1} \begin{bmatrix} \underline{\nabla}_{\underline{X}_0} \\ \underline{\nabla}_{\underline{X}} \\ \underline{\nabla}_{\underline{\Theta}} \end{bmatrix} p \quad (\text{B. 37})$$

where

$$\nabla_{X_{0i}} p = \Delta_k \nabla_{X_{0i}} \varpi_k, \quad \nabla_{X_i} p = \varpi_k \nabla_{X_i} \Delta_k, \quad \nabla_{\Theta_i} p = \Delta_k \nabla_{\Theta_i} \varpi_k \quad (\text{B. 38})$$

and the corresponding hessian is given as

$$\underline{\underline{\nabla}}^2 p = \mathbb{h}^{-1} \begin{bmatrix} \underline{\nabla}_{\underline{X}_0} \underline{\nabla}_{\underline{X}_0}^T & \underline{\nabla}_{\underline{X}_0} \underline{\nabla}_{\underline{X}}^T & \underline{\nabla}_{\underline{X}_0} \underline{\nabla}_{\underline{\Theta}}^T \\ \underline{\nabla}_{\underline{X}} \underline{\nabla}_{\underline{X}_0}^T & \underline{\nabla}_{\underline{X}} \underline{\nabla}_{\underline{X}}^T & \underline{\nabla}_{\underline{X}} \underline{\nabla}_{\underline{\Theta}}^T \\ \underline{\nabla}_{\underline{\Theta}} \underline{\nabla}_{\underline{X}_0}^T & \underline{\nabla}_{\underline{\Theta}} \underline{\nabla}_{\underline{X}}^T & \underline{\nabla}_{\underline{\Theta}} \underline{\nabla}_{\underline{\Theta}}^T \end{bmatrix} p \quad (\text{B. 39})$$

By exploiting the symmetric nature of the hessian, the only six (6) relevant components of $\underline{\underline{\nabla}}^2 p$

are given as

$$\begin{aligned} \nabla_{X_{0i}} \nabla_{X_{0j}} p &= \Delta_k \nabla_{X_{0i}} \nabla_{X_{0j}} \varpi_k, \quad \nabla_{X_i} \nabla_{X_j} p = \varpi_k \nabla_{X_i} \nabla_{X_j} \Delta_k, \quad \nabla_{\Theta_i} \nabla_{\Theta_j} p = \Delta_k \nabla_{\Theta_i} \nabla_{\Theta_j} \varpi_k \\ \nabla_{X_{0i}} \nabla_{X_j} p &= [\nabla_{X_{0i}} \varpi_k] [\nabla_{X_j} \Delta_k], \quad \nabla_{X_{0i}} \nabla_{\Theta_j} p = \Delta_k \nabla_{X_{0i}} \nabla_{\Theta_j} \varpi_k, \quad \nabla_{X_i} \nabla_{\Theta_j} p = [\nabla_{X_i} \Delta_k] [\nabla_{\Theta_j} \varpi_k] \end{aligned} \quad (\text{B. 40})$$

In the usual manner, the $\underline{\Theta}$ & $\underline{X}_0$ derivative operators are distributive over the non-constant elements and sub-elements of $\underline{\varpi}$. For conciseness, we can write a general expression for permutations of m^{th} order $\underline{\Theta}$ – derivative and n^{th} order $\underline{X}_0$ – derivative of $\underline{\varpi}$ as

$$\underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \underline{\varpi} = \left[\underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n A \quad \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n B \quad \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n C \quad \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n (F + F') \quad \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n (G + G') \quad \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n (H + H') \right] \quad (\text{B. 41})$$

where

$$\underline{\nabla}_{\underline{\Theta}}^m = \nabla_{\Theta_i}^{(1)} \nabla_{\Theta_j}^{(2)} \nabla_{\Theta_k}^{(3)} \dots \nabla_{\Theta_r}^{(p)} \dots \nabla_{\Theta_s}^{(m)}, \quad i, j, k, r, s = 1, 2, 3 \quad (\text{B. 42})$$

$\underline{\nabla}_{\underline{\Theta}}^{(p)}$ is p^{th} instance gradient operation for the m^{th} order gradient operator $\underline{\nabla}_{\underline{\Theta}}^m$ for instance,

$$\underline{\nabla}_{\underline{\Theta}}^2 = \nabla_{\Theta_j} \nabla_{\Theta_k}, \quad \underline{\nabla}_{\underline{X}_0}^2 = \nabla_{X_{0j}} \nabla_{X_{0k}}, \quad \underline{\nabla}_{\underline{\Theta}} \underline{\nabla}_{\underline{X}_0} = \nabla_{\Theta_j} \nabla_{X_{0k}}, \quad j, k = 1, 2, 3 \quad (\text{B. 43})$$

Typical partial derivative for the components of $\underline{Q}$ are given in eqn. (B. 44) below, and implicit in their expressions are the definition of other component derivatives.

$$\begin{aligned} \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n A &= \frac{1}{6} \left\{ \frac{2\Upsilon''_{1_0} \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Gamma_{11} - \Upsilon''_{2_0} \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Gamma_{22} - \Upsilon''_{3_0} \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Gamma_{33}}{\Upsilon''_{2_0} \Upsilon'_{3_0} + \Upsilon'_{3_0} \Upsilon''_{1_0} + \Upsilon'_{1_0} \Upsilon''_{2_0}} \right\} \\ \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n F &= \frac{\Upsilon_{2_0} \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Gamma_{23} - \mathcal{H}_3^2 \Upsilon'_{1_0} (\underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Xi_1 - \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Psi_1)}{2\Upsilon'_{1_0} (\mathcal{H}_2^2 \Upsilon_{2_0} + \mathcal{H}_3^2 \Upsilon_{3_0})} \end{aligned} \quad (\text{B. 44})$$

The derivative of the tensors Γ_{ij} and Ξ_{ij} can be derived from the derivative of the symmetric and antisymmetric decomposition of L_{ij} in the usual manner according to.

$$\langle \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Gamma_{ij}, \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Xi_{ij} \rangle = \frac{1}{2} \left[\underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n L_{ij} \pm \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n L_{ji} \right] \quad (\text{B. 45})$$

where the velocity gradient in the local fibers reference frame L_{ij} is obtained in the usual manner from the global definition $\mathcal{L}_{ij}$ through the transformation operation according to eqn. (5.9) given as $L_{ij} = Z_{X_{mi}} \mathcal{L}_{mn} Z_{X_{nj}}$. We require the first and second partial derivatives of L_{ij} with respect to the components of $\underline{X}_0$ and $\underline{\Theta}$. The first derivatives in indicial notation are given as

$$\nabla_{X_{0k}} L_{ij} = Z_{X_{mi}} \nabla_{X_{0k}} \mathcal{L}_{mn} Z_{X_{nj}} \quad (\text{B. 46})$$

$$\nabla_{\Theta_k} L_{ij} = \nabla_{\Theta_k} Z_{X_{mi}} \mathcal{L}_{mn} Z_{X_{nj}} + Z_{X_{mi}} \mathcal{L}_{mn} \nabla_{\Theta_k} Z_{X_{nj}}$$

Likewise, the second derivative of L_{ij} with respect to the components of $\underline{X}_0$ and $\underline{\Theta}$ in like manner are given as

$$\begin{aligned} \nabla_{X_{0r}} \nabla_{X_{0s}} L_{ij} &= Z_{X_{mi}} \nabla_{X_{0r}} \nabla_{X_{0s}} \mathcal{L}_{mn} Z_{X_{nj}} \\ \nabla_{\Theta_r} \nabla_{\Theta_s} L_{ij} &= \nabla_{\Theta_r} \nabla_{\Theta_s} Z_{X_{mi}} \mathcal{L}_{mn} Z_{X_{nj}} + Z_{X_{mi}} \mathcal{L}_{mn} \nabla_{\Theta_r} \nabla_{\Theta_s} Z_{X_{nj}} + \nabla_{\Theta_r} Z_{X_{mi}} \mathcal{L}_{mn} \nabla_{\Theta_s} Z_{X_{nj}} \\ &\quad + \nabla_{\Theta_s} Z_{X_{mi}} \mathcal{L}_{mn} \nabla_{\Theta_r} Z_{X_{nj}} \end{aligned} \quad (\text{B. 47})$$

$$\nabla_{\Theta_r} \nabla_{X_{0s}} L_{ij} = Z_{X_{mi}} \nabla_{\Theta_r} \nabla_{X_{0s}} \underline{L}_{mn} Z_{X_{nj}} + \nabla_{\Theta_r} Z_{X_{mi}} \nabla_{X_{0s}} \underline{L}_{mn} Z_{X_{nj}} + Z_{X_{mi}} \nabla_{X_{0s}} \underline{L}_{mn} \nabla_{\Theta_r} Z_{X_{nj}}$$

The higher order $\underline{\Theta}$ & $\underline{X}_0$ derivatives of the angular velocity $\underline{\Psi}$ found in eqn. (B. 44) can be

derived in similar fashion to eqn. (B. 42) above given as

$$\underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \underline{\Psi}_j = \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \underline{\Xi}_j + M_{jk} \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n D_k \quad (\text{B. 48})$$

In the usual manner, the $\underline{\Theta}$ & $\underline{X}_0$ derivatives are distributive over the individual components of the tensors $\underline{\Xi}_j$, and D_j , for instance

$$\underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \underline{\Xi}_1 = \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \xi, \quad \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n D_1 = \underline{\nabla}_{\underline{\Theta}}^m \underline{\nabla}_{\underline{X}_0}^n \Gamma_{23} \quad (\text{B. 49})$$

The derivatives of the Laplace function Ω used to obtain the gradient and hessian of $\underline{\Delta}$ where previously derived in terms of the actual quantities. Recall the dimensionless form for the Jeffery's pressure $\bar{p}$ given in eqn. (5.126). For this flow, $\dot{\gamma}_c$ is not constant but depends on the independent differentiable variables. As such it must be considered when obtaining derivatives of the dimensionless pressure. The first and second derivatives of $\bar{p}$ is given in indicial notation as

$$\nabla_k \bar{p} = \nabla_k (\dot{\gamma}_c^{-1}) \frac{p - p_0}{\mu} + \dot{\gamma}_c^{-1} \frac{\nabla_k p}{\mu}, \quad \dot{\gamma}_c = \sqrt{2\Gamma_{ij}\Gamma_{ji}} \quad (\text{B. 50})$$

$$\nabla_m \nabla_n \bar{p} = \nabla_m \nabla_n (\dot{\gamma}_c^{-1}) \frac{p - p_0}{\mu} + \nabla_m (\dot{\gamma}_c^{-1}) \frac{\nabla_n p}{\mu} + \nabla_n (\dot{\gamma}_c^{-1}) \frac{\nabla_m p}{\mu} + \dot{\gamma}_c^{-1} \frac{\nabla_m \nabla_n p}{\mu} \quad (\text{B. 51})$$

where

$$\nabla_k (\dot{\gamma}_c^{-1}) = -2\dot{\gamma}_c^{-3} \nabla_k \Gamma_{ij} \Gamma_{ji}, \quad \Gamma_{ij} = \Gamma_{ji} \quad (\text{B. 52})$$

$$\begin{aligned} \nabla_m \nabla_n (\dot{\gamma}_c^{-1}) = & 12\dot{\gamma}_c^{-5} [\Gamma_{ij} \nabla_m \Gamma_{ji}] [\Gamma_{pq} \nabla_n \Gamma_{qp}] \cdots \\ & - 2\dot{\gamma}_c^{-3} [\Gamma_{ij} \nabla_m \nabla_n \Gamma_{ji} + \nabla_m \Gamma_{ij} \nabla_n \Gamma_{ji} + \nabla_n \Gamma_{ij} \nabla_m \Gamma_{ji} + \Gamma_{ij} \nabla_m \nabla_n \Gamma_{ji}] \end{aligned} \quad (\text{B. 53})$$

The same validation exercise of the gradient and hessian for this flow type using finite difference such as that described in Appendix IV is carried out. Assuming the same orientation state $\underline{\Theta}^i$ and relative spatial position $\mathbb{h}^{-1} \underline{X}^i$ as provided in (5.64) for a unity $\mathbb{h}$ value and given an arbitrary fiber centroidal position $\underline{\mathbf{X}}_0^i = [12 \quad 9 \quad 0.75]^T$, We obtain the following non-zero component of the velocity gradient as $\underline{L}_{11} = -\underline{L}_{22} = -0.011575$, $\underline{L}_{13} = 0.25053$. Subsequently,

the pressure and the gradient and hessian error estimates based on definition of eqn. (5.63) are obtained as $\zeta^{(1)} = 1.0763 \times 10^{-15}$, $\zeta^{(2)} = 3.9542 \times 10^{-13}$.

C APPENDIX C

C.1 Eigenvalues and Eigenvector Derivatives

The eigenvalue definition for any system is typically given in terms of the eigen-values λ^k and corresponding eigen-vectors $\underline{\Phi}^k$ as [323]

$$\left[\underline{K} - \lambda_k \underline{M} \right] \underline{\Phi}^k = \underline{F}^k \quad (\text{C. 1})$$

For most undamped systems, $\underline{F}^k = \mathbf{0}$, and since $\underline{\Phi}^k \neq 0$ to yield non-trivial solutions, then by setting $\left| \underline{K} - \lambda_k \underline{M} \right| = 0$, we can obtain solutions for λ^k . By reason of the nature of the system matrix $[K_{mn} - \lambda^k M_{mn}]$ being rank deficient with one order less than the matrix size one may adopt a scaling algorithm to obtain the corresponding eigen-vectors $\underline{\Phi}^k$ usually by defining a Mode I normalization technique for scaling the eigen-vectors $\underline{\Phi}^k$ via a scalar functions $G^k(\underline{\Phi}^k)$ such that $G^k = 0$, which may be non-linear in nature. The eigenvectors are thus obtained by replacing the n^{th} row of the residual column vector $\Sigma_i^k = (K_{ij} - \lambda^k M_{ij})\Phi_j^k - F_i^k$ with $G^k(\underline{\Phi}^k)$ and solving for $\underline{\Phi}^k$ from the equation $\underline{\Sigma}^k(\underline{\Phi}^k) = 0$ through any iterative algorithm or explicit solvers. Here we employ the Newton-Raphson's method to obtain $\underline{\Phi}^k$ such that:

$$\underline{\Phi}^{k+} = \underline{\Phi}^{k-} - J^{k-1} \underline{\Sigma}^k \quad (\text{C. 2})$$

$$\Sigma_i^k = (1 - \delta_{in})[S_{ij}^k \Phi_j^k - F_i^k] + \delta_{in} G^k(\underline{\Phi}^k) \quad (\text{C. 3})$$

$$J_{ij}^k = \frac{\partial \Sigma_i^k}{\partial \Phi_j^k} = (1 - \delta_{in})S_{ij}^k + \delta_{in} \frac{\partial G^k}{\partial \Phi_j^k} \quad (\text{C. 4})$$

where

$$S_{ij}^k = K_{ij} - \lambda^k M_{ij} \quad (\text{C. 5})$$

The derivative of the eigen values with respect to components $\underline{a}$ can thus be obtained by

differentiating eqn. (C. 3) assuming symmetry of system matrix, i.e., $S_{ij}^k = S_{ji}^k$ such that:

$$\Phi_i^k \frac{\partial S_{ij}^k}{\partial a_{rs}} \Phi_j^k + F_i^k \frac{\partial \Phi_i^k}{\partial a_{rs}} - \Phi_i^k \frac{\partial F_i^k}{\partial a_{rs}} = 0 \quad (\text{C. 6})$$

$$\Phi_i^k S_{ij}^k = S_{ji}^k \Phi_j^k = S_{ij}^k \Phi_j^k = \underline{F}_i^k \quad (\text{C. 7})$$

where:

$$\frac{\partial S_{ij}^k}{\partial a_{rs}} = \frac{\partial K_{ij}}{\partial a_{rs}} - \frac{\partial \lambda^k}{\partial a_{rs}} M_{ij} - \lambda^k \frac{\partial M_{ij}}{\partial a_{rs}} \quad (\text{C. 8})$$

Since $\underline{F}_i^k = 0$ for undamped systems

$$\frac{\partial \lambda^k}{\partial a_{rs}} = \frac{1}{(\Phi_i^k M_{ij} \Phi_j^k)} \left\{ \Phi_i^k \left[\frac{\partial K_{ij}}{\partial a_{rs}} - \lambda^k \frac{\partial M_{ij}}{\partial a_{rs}} \right] \Phi_j^k \right\} \quad (\text{C. 9})$$

Consequently, we can obtain the corresponding derivatives $\partial \Phi_i^k / \partial a_{rs}$ given $\partial \lambda^k / \partial a_{rs}$ from

$$S_{ij}^k \frac{\partial \Phi_i^k}{\partial a_{rs}} = - \frac{\partial S_{ij}^k}{\partial a_{rs}} \Phi_j^k \quad (\text{C. 10})$$

Recalling the system matrix $[\underline{K} - \lambda^k \underline{M}]$ is inherently singular, we can substitute an n^{th} row of the above equation adopting one the normalization techniques with the equation [323].

$$\frac{\partial G^k}{\partial \Phi_j^k} \frac{\partial \Phi_j^k}{\partial a_{rs}} = \frac{\partial G^k}{\partial a_{rs}} \quad (\text{C. 11})$$

The modified differential equation can be recast as given in equation xx below allowing the inversion of the modified system matrix.

$$J_{ij}^k \frac{\partial \Phi_i^k}{\partial a_{rs}} = \frac{\partial Q_i^k}{\partial a_{rs}} \quad (\text{C. 12})$$

where:

$$\frac{\partial Q_i^k}{\partial a_{rs}} = -(1 - \delta_{in}) \frac{\partial S_{ij}^k}{\partial a_{rs}} \Phi_j^k - \delta_{in} \frac{\partial G^k}{\partial a_{rs}} \quad (\text{C. 13})$$

Smith et. al [323] presents 3 common Mode *I* normalization techniques thus:

1. Mass Normalization

$$G^k(\underline{\Phi}^k) = \Phi_i^k M_{ij} \Phi_j^k - 1, \quad \frac{\partial G^k}{\partial \Phi_j^k} = 2\Phi_i^k M_{ij}, \quad \frac{\partial G^k}{\partial a_{rs}} = \Phi_i^k \frac{\partial M_{ij}}{\partial a_{rs}} \Phi_j^k \quad (\text{C. 14})$$

2. Preassigning an m^{th} Component of $\underline{\Phi}^k$

$$G^k(\underline{\Phi}^k) = \delta_{mj}\Phi_j^k - \alpha, \quad \frac{\partial G^k}{\partial \Phi_j^k} = \delta_{mj}, \quad \frac{\partial G^k}{\partial a_{rs}} = 0 \quad (\text{C. 15})$$

3. Predefining the Euclidean norm of $\underline{\Phi}^k$

$$G^k(\underline{\Phi}^k) = \sqrt{\Phi_j^k \Phi_j^k} - \beta, \quad \frac{\partial G^k}{\partial \Phi_j^k} = \Phi_j^k, \quad \frac{\partial G^k}{\partial a_{rs}} = 0 \quad (\text{C. 16})$$

A more direct and efficient approach by Nelson [353] which utilizes mass normalization technique is given below:

$$\frac{\partial \Phi_i^k}{\partial a_{rs}} = V_i^k + c^k \Phi_i^k, \quad c^k = -\Phi_i^k M_{ij} V_j^k - \frac{1}{2} \Phi_i^k \frac{\partial M_{ij}}{\partial a_{rs}} \Phi_j^k, \quad (\text{C. 17})$$

$$V_i^k = \left\{ \underline{\underline{SP}}^{k-1} \right\}_{ij} \frac{\partial QP_j^k}{\partial a_{rs}}$$

Given a pre-selected fixed index – m and noting in eqns. (C. 18) - (C. 19) below that repeated indices do not imply a summation,

$$SP_{ij}^k = (K_{ij} - \lambda^k M_{ij})(1 - \delta_{mi})(1 - \delta_{mj}) + \delta_{mi}\delta_{mj} \quad (\text{C. 18})$$

$$\frac{\partial QP_i^k}{\partial a_{rs}} = -\frac{\partial S_{ij}^k}{\partial a_{rs}} \Phi_j^k (1 - \delta_{mi}) \quad (\text{C. 19})$$

C.2 Optimal Fitted Closure Approximation Constants/Coefficients

C.2.1 Eigenvalue Based Optimal Fitting Closure (EBOF) Approximation

We consider 4 fitted closures approximations of this form. Linear, quadratic, cubic and quartic binomial fitted closures with $(n+1)(n+2)/2$ number of parameters and their constants below.

C.2.1.1 Linear Orthotropic Fitted Closure ($n = 1$)

For the general linear orthotropic closure, the constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ is given as

$$\underline{\underline{\mathfrak{C}}}^{(1)} = \frac{1}{7} \begin{bmatrix} -3/5 & 6 & 0 \\ -3/5 & 0 & 6 \\ 27/5 & -6 & -6 \end{bmatrix}$$

and for the smooth orthotropic closure by Cintra and Tucker (cf. [316]), the constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ is given as

$$\underline{\underline{\mathfrak{C}}}^{(1)} = \begin{bmatrix} -0.15 & 1.15 & -0.10 \\ -0.15 & 0.15 & 0.90 \\ 0.60 & -0.60 & -0.60 \end{bmatrix}$$

C.2.1.2 Quadratic Orthotropic Fitted Closures ($n = 2$)

The simple general orthotropic quadratic closure has constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ given as

$$\underline{\underline{\mathfrak{C}}}^{(2)} = \begin{bmatrix} 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \\ 1 & -2 & -2 & 1 & 2 & 1 \end{bmatrix}$$

The orthotropic natural closure - exact midpoint fit [251] has constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ given as

$$\underline{\underline{\mathfrak{C}}}^{(2)} = \begin{bmatrix} 0.0708 & 0.3236 & -0.3776 & 0.6056 & 0.4124 & 0.3068 \\ 0.0708 & -0.2792 & 0.2252 & 0.2084 & 0.4124 & 0.7040 \\ 1.1880 & -2.0136 & -2.1264 & 0.8256 & 1.7640 & 0.9384 \end{bmatrix}$$

The ORF independent coefficients are derived from a 2nd order polynomial fit of the principal axis data obtained from DFC via a minimization technique. For the orthotropic fitted closure by Cintra and Tucker (ORF) (cf. [316]), the constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ is given as

$$\underline{\underline{\mathfrak{C}}}^{(2)} = \begin{bmatrix} 0.060964 & 0.371243 & -0.369160 & 0.555301 & 0.371218 & 0.318266 \\ 0.124711 & -0.389402 & 0.086169 & 0.258844 & 0.544992 & 0.796080 \\ 1.228982 & -2.054116 & -2.260574 & 0.821548 & 1.819756 & 1.053907 \end{bmatrix}$$

The ORF had better performance compared to non-fitted closure approximations however the ORF behaved poorly for flows with very low interaction coefficients and sometimes gave non-physical oscillations like the behavior of the Hinch and Leal closure (HL2) in same condition. Though the ORL behaves better for low interaction coefficient in simple shear flow yet overpredicts the flow direction orientation tensor and is unstable for radial diverging flows. Chung and Kwon [354],

improved the ORF and developed the 2nd order ORW closure for wide interaction coefficients that is stable for all flow conditions using new flow data from distribution function calculation (DFC). The improved orthotropic fitted closure (ORW2) by Chung and Kwon (cf. [354]), has constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ given as

$$\underline{\underline{\mathfrak{C}}}^{(2)} = \begin{bmatrix} 0.070055 & 0.339376 & -0.396796 & 0.590331 & 0.411944 & 0.333693 \\ 0.115177 & -0.368267 & 0.094820 & 0.252880 & 0.535224 & 0.800181 \\ 1.249811 & -2.148297 & -2.290157 & 0.898521 & 1.934914 & 1.044147 \end{bmatrix}$$

Kuzmin [251] presents details on derivations of some orthotropic fitted closures via a numerical bottom top approach.

C.2.1.3 Cubic Orthotropic Fitted Closures ($n = 3$)

Recently higher order polynomial fitted closures were developed for improved accuracy. The orthotropic natural closure - extended quadratic fit though of cubic order is essentially quadratic.

1. Non-rational Fitted Closure

The constant coefficient matrix $\underline{\underline{\mathfrak{C}}}^{(3)}$ for this closure approximation is given as

$$\underline{\underline{\mathfrak{C}}}^{(3)} = \begin{bmatrix} 0 & 0.5 & 0 & 0.5 & -0.6 & 0 & 0.6 & 0.6 & 0 \\ 0 & 0 & 0.5 & 0 & -0.6 & 0.5 & 0 & 0.6 & 0.6 \\ 1 & -1.5 & -1.5 & 0.5 & 0.4 & 0.5 & 0 & 0.6 & 0.6 \end{bmatrix}$$

Chung and Kwon [354], also extended the 2nd order ORW to develop 3rd order polynomial ORW3 closure using new flow data from distribution function calculation (DFC). For the improved 3rd order orthotropic fitted closure ORW3 by Chung and Kwon [354], the constant coefficient matrix is given as

$$\underline{\underline{\mathfrak{C}}}^{(3)} = \begin{bmatrix} -0.1480648093 & -0.2106349673 & 0.4868019601 \\ 0.8084618453 & 0.9092350296 & 0.5776328438 \\ 0.7765597096 & 1.1104441966 & 0.4605743789 \\ 0.3722003446 & -1.2840654776 & -2.2462007509 \\ -1.7366749542 & -2.5375632310 & -4.8900459209 \\ -1.3431772379 & 0.1260059291 & -1.9088154281 \\ -0.0324756095 & 0.5856304774 & 1.1817992322 \\ 0.8895946393 & 1.9988098293 & 4.0544348937 \\ 1.7367571741 & 1.4863151577 & 3.8542602127 \end{bmatrix}^T$$

2. Rational Fitted Closure 0.6631716575 -0.0756740034 0.9512305286

The rational elliptical (RE) closure developed by Wetzel [41] is a higher order extension to the ORF using Carlson elliptical integrals. The rational ellipsoid fitted closure has two permutation vectors, the denominator being one order less than the numerator, i.e., $m = n - 1$ which is of cubic order, $n = 3$. The corresponding constant coefficient matrix for the Wetzel numerator ($n = 3$) is [344]

$$\begin{aligned} \left[\underline{\underline{\mathfrak{C}}}^{(3)} \right]^T \\ = \end{aligned} \begin{vmatrix} 0.1433751825 & 0.1433751825 & 0.9685744898 \\ -0.6566650339 & -0.5209453949 & -2.5526857671 \\ -0.5106016916 & -0.6463213306 & -2.5756669706 \\ 3.5295952199 & 0.6031924921 & 2.2044050704 \\ 4.4349137241 & 2.3303190917 & 4.4520903005 \\ 0.1229618909 & 5.1539592511 & 2.2485545147 \\ -2.9144388828 & -0.2256222796 & -0.6202937932 \\ -5.5556896198 & -1.6481269200 & -1.8811803355 \\ -2.8284365891 & -5.4494528976 & -1.9023485762 \\ 0.2292109036 & -3.7461520908 & -0.6414620339 \end{vmatrix}$$

And for the denominator ($m = n - 1 = 2$)

$$\left[\underline{\underline{\mathfrak{C}}}^{(2)} \right]^T = \begin{vmatrix} 1.0000000000 & 1.0000000000 & 1.0000000000 \\ 0.7257989503 & 0.6916858207 & -1.2134964928 \\ 3.0941511876 & 3.1282643172 & -1.2128608265 \\ -1.6239324646 & -1.5898193351 & 0.2393747647 \\ -4.7303686308 & -4.7303686308 & 0.6004510415 \\ -3.1742364608 & -3.2083495904 & 0.2162486576 \end{vmatrix}$$

Mullens et al. [42] developed several high order polynomials fitted closures for short fiber composites and introduced the time derivative fitted closures. For the LAR32 closure by Mullens [343] the corresponding constant coefficient matrix for the numerator ($n = 3$) is

$$\begin{aligned} \left[\underline{\underline{\mathfrak{C}}}^{(3)} \right]^T \\ = \end{aligned} \begin{vmatrix} 0.087602233 & 0.156805152 & 1.072423739 \\ 0.028205550 & -0.577818864 & -2.803554028 \\ -0.426784335 & -0.514280920 & -2.661576129 \\ 1.274677110 & 0.684250887 & 2.389379765 \\ 0.876469059 & 2.132305029 & 4.566728489 \\ 0.602031647 & 3.454835266 & 2.097523143 \\ -1.066583115 & -0.263237143 & -0.658248930 \\ -1.918931146 & -1.614122610 & -1.904704744 \\ -0.934291306 & -4.005261132 & -1.754978355 \\ -0.262854903 & -2.228133231 & -0.508282668 \end{vmatrix}$$

And for the denominator ($m = n - 1 = 2$)

$$\left[\underline{\underline{\mathfrak{C}}}^{(2)} \right]^T = \begin{vmatrix} 1.000000000 & 1.000000000 & 1.000000000 \\ -0.244001948 & 0.365652907 & -1.068512526 \\ -0.574150861 & 1.385725477 & -0.771356469 \\ -0.432097367 & -1.359687152 & 0.067386858 \\ -0.895226091 & -2.866357848 & 0.206908269 \\ -0.462709527 & -1.518996192 & -0.248999874 \end{vmatrix}$$

3. Quartic Orthotropic Fitted Closures ($n = 4$)

The constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ based on regression analysis by Verweyst [350] developed from Carlson elliptic integrals.

$$\left[\underline{\underline{\mathfrak{C}}}^{(4)} \right]^T = \begin{vmatrix} 0.6363 & 0.6363 & 2.7405 \\ -1.8727 & -3.3153 & -9.1220 \\ -4.4797 & -3.0371 & -12.2571 \\ 11.9590 & 11.8273 & 34.3199 \\ 3.8446 & 6.8815 & 13.8295 \\ 11.3421 & 8.4368 & 25.8685 \\ -10.9583 & -15.9121 & -37.7029 \\ -20.7278 & -15.1516 & -50.2756 \\ -2.1162 & -6.4873 & -10.8802 \\ -12.3876 & -8.6389 & -26.9637 \\ 9.8160 & 9.3252 & 27.3347 \\ 3.4790 & 7.7468 & 15.2651 \\ 11.7493 & 7.4815 & 26.1135 \\ 0.5080 & 2.2848 & 3.4321 \\ 4.8837 & 3.5977 & 10.6117 \end{vmatrix}$$

The constant coefficient matrix $\underline{\underline{\mathfrak{C}}}'$ based on regression analysis for the FFLAR4 closure by Mullens [343]

$$\begin{aligned} \left[\underline{\underline{\mathfrak{C}}}^{(4)} \right]^T &= \begin{vmatrix} 0.678225884 & 0.748226727 & 3.167356369 \\ -3.834359034 & -4.249612053 & -13.288266400 \\ -2.664862865 & -2.987266447 & -11.680179330 \\ 9.746185193 & 8.641488072 & 23.788431340 \\ 14.209962670 & 14.938209410 & 43.700607680 \\ 2.700369681 & 5.974489008 & 17.383121430 \\ -8.013024236 & -7.521216405 & -19.959054610 \\ -22.447252700 & -21.757217160 & -58.354308000 \end{vmatrix} \\ \left[\underline{\underline{\mathfrak{C}}}^{(4)} \right]^T &= \begin{vmatrix} -13.078649640 & -15.798676320 & -49.513705640 \\ -0.125467689 & -3.616551654 & -11.755525930 \\ 2.417857515 & 2.376441613 & 6.291273472 \\ 10.563248410 & 10.222185780 & 25.844317920 \\ 12.689484570 & 12.640352670 & 35.425354130 \\ 2.487386515 & 4.788201652 & 18.226443930 \\ -0.328195677 & 1.056519961 & 2.925785795 \end{vmatrix} \end{aligned}$$

The constant coefficient matrix $\underline{\underline{\mathfrak{C}'}}$ based on regression analysis for the LAR4 closure by Mullens

[343] is

$$\left[\underline{\underline{\mathfrak{C}^{(4)}}}\right]^T = \begin{bmatrix} 0.813175172 & 1.768619587 & 4.525066937 \\ -3.065410883 & -9.826017151 & -19.259137620 \\ -4.659333003 & -6.484058476 & -17.650178090 \\ 6.329870878 & 19.986994700 & 33.901239610 \\ 14.747639770 & 28.905936750 & 61.543979540 \\ 9.739797775 & 10.759963010 & 28.467355970 \\ -4.216519964 & -17.715409270 & -27.768082700 \\ -15.922240910 & -40.492387100 & -76.738638810 \\ -20.818571900 & -27.442217500 & -68.977583290 \\ -8.993993112 & -7.230748101 & -22.399036130 \\ 1.138888034 & 5.785725498 & 8.600822308 \\ 5.834142985 & 18.709047480 & 32.480679940 \\ 11.470974520 & 19.729631240 & 43.875135630 \\ 9.874209286 & 8.882877701 & 26.928320210 \\ 3.100457733 & 2.224834058 & 7.101978254 \end{bmatrix}$$

C.2.2 Invariant Based Optimal Fitting Closure (IBOF) Approximation

The unknown independent coefficients of the binomial expansion for the six parameters in the IBOF closure representation based on regression fitting by Chung et al. [346] of actual flow data obtained from the distribution function closure considering different flow types like EBF closures are given in Table C. 1 below.

Table C. 1: 5th order binomial fitting coefficients for the IBOF closure approximation

$k \backslash m$	1	2	3
0	2.49409081657860E+01	-4.97217790110754E-01	2.34146291570999E+01
1	-4.35101153160329E+02	2.34980797511405E+01	-4.12048043372534E+02
2	7.03443657916476E+03	1.53965820593506E+02	5.73259594331015E+03
3	3.72389335663877E+03	-3.91044251397838E+02	3.19553200392089E+03
4	-1.33931929894245E+05	-2.13755248785646E+03	-6.05006113515592E+04
5	8.23995187366106E+05	1.52772950743819E+05	-4.85212803064813E+04
6	-1.59392396237307E+04	2.96004865275814E+03	-1.10656935176569E+04
7	8.80683515327916E+05	-4.00138947092812E+03	-4.77173740017567E+04
8	-9.91630690741981E+06	-1.85949305922308E+06	5.99066486689836E+06
9	8.00970026849796E+06	2.47717810054366E+06	-4.60543580680696E+07
10	3.22219416256417E+04	-1.04092072189767E+04	1.28967058686204E+04
11	-2.37010458689252E+06	1.01013983339062E+05	2.03042960322874E+06
12	3.79010599355267E+07	7.32341494213578E+06	-5.56606156734835E+07
13	-3.37010820273821E+07	-1.47919027644202E+07	5.67424911007837E+08
14	-2.57258805870567E+08	-6.35149929624336E+07	-1.52752854956514E+09

15	-2.32153488525298E+04	1.38088690964946E+04	4.66767581292985E+03
16	2.14419090344474E+06	-2.47435106210237E+05	-4.99321746092534E+06
17	-4.49275591851490E+07	-9.02980378929272E+06	1.32124828143333E+08
18	-2.13133920223355E+07	7.24969796807399E+06	-1.62359994620983E+09
19	1.57076702372204E+09	4.87093452892595E+08	7.92526849882218E+09
20	-3.95769398304473E+09	-1.60162178614234E+09	-1.28050778279459E+10

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