Polyelectrolytes and Water-Soluble Polymers: AI/ML-Driven Materials and Applications

Rigoberto C. Advincula*[1,2] and Jihua Chen[1]

[1]Center for Nanophase Materials Sciences, Oak Ridge National Laboratory (ORNL)
1 Bethel Valley Road, Oak Ridge, TN 37830

[2]Department of Chemical and Biomolecular Engineering,
University of Tennessee at Knoxville
1512 Middle Dr, Knoxville, TN 37996

* To whom correspondence should be addressed: radvincu@utk.edu

Abstract

Water-soluble polymers are a class of macromolecules that includes natural, synthetic, and polyelectrolyte polymers. Their study is fascinating, and increasingly, new applications are being discovered from membranes to drug delivery. How do we apply artificial intelligence and machine learning (AI/ML) to this class of polymers? An AI/ML-polymer redesign and the application of large language models (LLMs) for agentic AI tasks can transform these macromolecules into libraries and workflows that can answer some of the most interesting questions in microstructure, phase stability, ion transport, and self-assembly, furthering discovery science and new technologies. This article examines the background of natural polymers, water-soluble polymers, polyelectrolytes, polyelectrolyte complexes (PEC), and polyelectrolyte multilayers (PEM). Active ML and creative materials design by generative AI will create opportunities for new structure-composition-processing-property (SCPP) correlations. Methods for scaling synthesis using continuous-flow chemistry (CFC) in autonomous, self-driving labs (SDL) will be examined for parametrization and optimization in reaction engineering. Finally, 3D printing, an advanced manufacturing method, will be reviewed, with a focus on these materials. Perspectives on the future impact of this approach will generate high interest in accelerating research and scientific discovery.

1. Introduction

Polymers are high molecular-weight (MW) molecules, or macromolecules, where structure-composition-properties (SCP) are predicted by specific principles governing their solution properties, crystallinity, morphology, and thermomechanical properties.[1] The linear polymer chain follows the degree of polymerization (DPn) or molecular-weight-based scalar properties.[2] Various microstructures, such as grafts, hyperbranching, dendrimers, and macrocycles, extend scalar properties to topological parameters. Overall, we understand polymers as a combination of chemistry, physics, and engineering disciplines—a true interdisciplinary field. As the largest class of materials used in modern industry, it includes natural (renewable) and synthetic options.

Natural polymers come from earth-abundant plant, animal, marine, and bacterial sources.[3,4] Feedstocks require mostly harvesting, extraction, purification, and some chemical modification.[3] While some are used directly in industrially scaled applications, such as paper, fibers, and ropes, many derivatives or intermediates are used as additives for paint, cement, food, and drilling fluids. Another category of polymers is synthetic water-soluble polymers. These are polymers whose main utility lies in their aqueous-soluble properties and hydrogel or hygroscopic properties. Their sources are both biomass and fossil-fuel-based. A key category of well-studied charged polymers is the polyelectrolytes. It carries electrostatic charges that can be cationic, anionic, zwitterionic, ampholytic, and amphoteric. Their complexation and coacervation properties are now attracting significant attention for potential applications in membranes, batteries, packaging, and related areas.

It is important to apply artificial intelligence and machine learning (AI/ML) to research and development on these classes of natural, water-soluble, and polyelectrolyte polymers.[5] Simulation and theory have predicted unique properties of polyelectrolytes, including coarse-grain molecular dynamics (MD) simulations and self-consistent field theories (SCFT). Natural polymers and various microstructures are abundant in nature, and generative AI can predict some of their unique supramolecular structures and properties. Water-soluble polymers are now widely used in formulation and packaging. ML-driven characterization and the use of autonomous, self-driving laboratories (SDLs) can be key to greater industrial adoption and environmental friendliness. The ability to 3D print polyelectrolyte complexes while optimizing their viscoelastic behavior with ML-driven feedback loop can be key to their utility in many applications. These are some of the reasons why it is important to bring AI/ML workflows into their study.

2.Classification of Polymers

2.1 Natural Polymers

Natural polymers are the most abundant polymers on Earth.[4] (Figure 1) Cellulose and polysaccharides are considered renewable and sustainable.[6] Lignocellulose is the basic compound composition that makes up plant sources, pulp, and eventually paper or fibers. But classically, the major classes based on monomers, sequence, and legacy names are: polynucleotides (DNA, RNA), polypeptides or proteins (gelatin, gluten, silk, wool, etc.), polysaccharides and carbohydrates (starch, cellulose, chitosan, etc.), polyoxoesters - poly(hydroxyalkanoic acids), rosin (pine), polyisoprenoides (natural rubber, inorganic polymers - inorganic polyesters polyphosphate, polythioesters, polyphenols (such as lignin or humic acids), poly(malic acid), poly(3-mercaptopropionate). Polysulfides or sulfur polymers are abundantly derived from extracted sub-surface heavy oil, unlike the others, which are abundant on Earth's surface.[4]

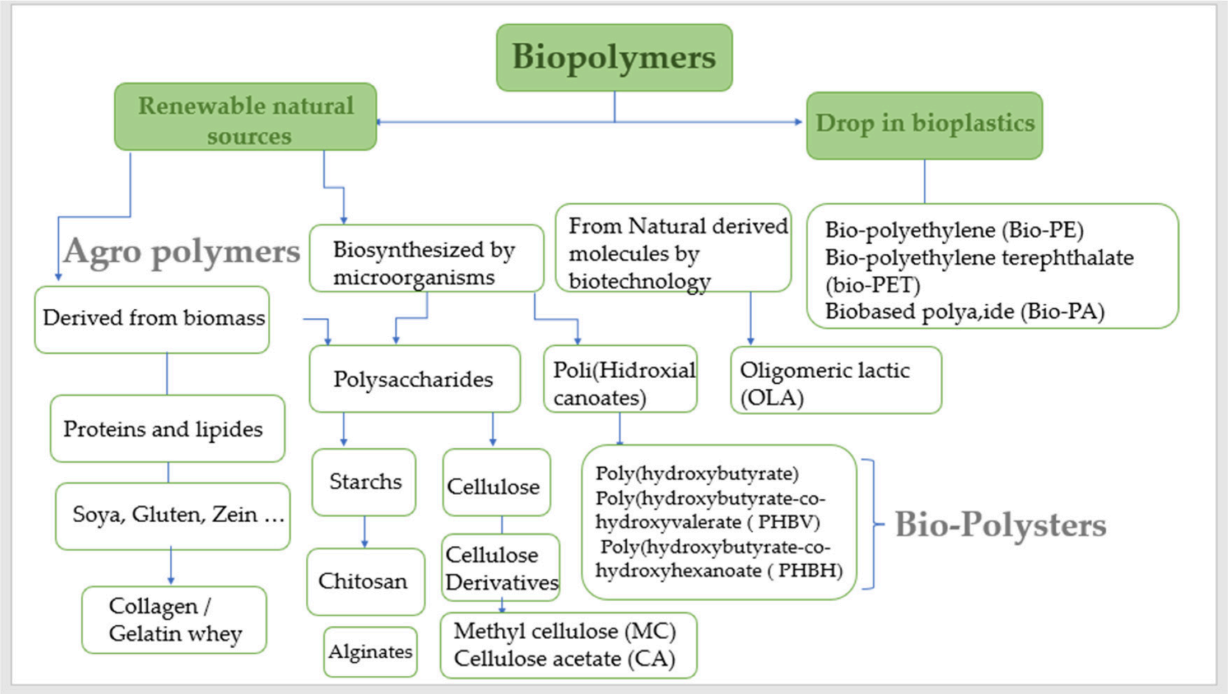

Figure 1. Classification of natural polymers. (Figure and caption from reference[7] with no change under CC-BY 4.0 license[8])

The distinct properties of natural or biopolymers include non-toxicity, biodegradability, biocompatibility, and water solubility or dispersibility.[9] This makes natural polymers useful for many applications that are considered environmentally friendly. Their strong thermo-mechanical properties can be based on strong chain alignment and crystallinity, as in fibers. Their degradation or biodegradation is also a distinct property. Their classification, properties, synthesis methods, low cost, and diverse sources still make them the polymer of choice for many applications, e.g., the food industry, cosmetics, packaging, adhesives, coatings, etc. Their renewable and sustainable feed stocks make them an integral part of the agriculture, biomass, and marine industries. Many natural polymers can also be classified as synthetic polymers because they are modified with synthetic functional groups via polymer-analogous reactions. We have recently written a review on advanced manufacturing with bio-derived (renewable) polymers and outlined the importance of AI/ML workflows and process optimization.[10] Some of their interesting properties, challenges, and opportunities are outlined as follows.

In many formulation industries, they are used as texture modifiers, thickeners, emulsifiers, gelling agents, and polymer excipients, i.e., in the food, coatings, and medicine industries.[11] Specific examples are polysaccharides (starch, cellulose, chitosan, carrageenan, pullulan, hyaluronic acid, fucoidan, alginate, guar gum, and agar) and proteins (lysozyme, albumin). They can be developed in final form factors as processed products for composites, coatings, adhesives, membranes, and polymer patches, but are derived from bio-based or renewable sources.[3]

Many polymers are derived from biobased monomers, such as polyesters and polyurethanes.[10,12–14] Polylactic acid (PLA) polymers are made from bio-based monomers. They can be used as multifunctional, biodegradable nanocomposites with many applications in biomedicine and as thermoplastics for packaging, molded objects, and 3D printing.[15] Interesting synthesis routes and copolymerization through step-growth and ring-opening reactions give them versatile compositions. For example, we reported an interesting supramolecular polymer based on catenated poly(ε-caprolactone) and copolymers with poly(l-lactide) via ring-opening polymerization and ring-expansion strategy.[16]

A specific class of marine-sourced polymers is chitosan, derived from deacetylated chitin (from crab or shrimp shells).[17,18] They can be made into films, fibers, and nanofibers that are biocompatible and exhibit antibacterial activity.[17] They can be derivatized to have both antioxidant and chelating properties.[18] The synthesis of crosslinked graphene oxide-chitosan (CS-GO) films as nanocomposite has been reported, and their potential for food processing and packaging applications has been described.[19] We recently reported a review on chitosan-based scaffolds and their biomedical applications for the treatment of myocardial infarction.[20]

Lignin, an important class of bio-derived polymers, is separated during pulp production and is the subject of valorization research, as it has long been considered a by-product.[6] We have converted it into sustainable shape-memory elastomers by blending, which results in materials with decreased melt viscosity and improved thermo-mechanical properties.[21] Dynamic mechanical properties were observed when interlocking Kraft lignin together with blends of ultrahigh molecular weight poly(ethylene oxide) or UHMWPE. It exhibited shape memory and self-healing properties.[22]

Fibers or natural fibers, isolated directly from wood pulp or agro-based biomass, are more complex.[23–26] But they can be used for many purposes as additives, rheological modifiers, binders, etc. We reported using *Miscanthus x. Giganteus* agro-waste particles as a filtrate reducer in water-based drilling fluids.[27]

Hydrogels can absorb large volumes of water, and can be derived from physical or chemical crosslinking.[28–30] Polysaccharides and proteins contain numerous electrostatic charges, reactive groups, and hydrogen-bonding interactions, making them ideal for membrane or controlled-release applications, including water treatment.[28] We reported a mussel-inspired, triple-responsive (pH, temperature, and NIR light) adhesive hydrogel, which combines catechol-functionalized chitosan with catechol-terminated poly(N-isopropylacrylamide) and polypyrrole nanoparticles to achieve dual heat- and photo-responsivity.[30]

Natural polymers, together with their synthetic equivalents, are also increasingly used for sustainable construction and geo-materials.[31] Stimuli-responsive materials can be designed to associate or dissociate as cross-linked structures in response to external or environmental conditions, enabling tunable disintegration and dissolution rates.[12] The use of cellulose-ether derivatives and welan gum additives for 3D-printed cement and cementitious materials, together with synthetic polymers such as polyvinyl alcohol (PVA), has been reported.[32,33]

2.2 Water-soluble Polymers

Water-soluble polymers include both natural and synthetic polymers. As a class, they are considered to have tunability in solubility with the aqueous solvent. Polymer hydration significantly contributes to the free energy of solvation of polymers and is important for understanding the thermodynamics of solubility, self-assembly, aggregation behavior, and phase separation properties.[34]

From Newtonian solutions to viscoelastic hydrogels, their solubility properties are essential for applications in personal care formulations, food, medicine, drilling fluids, and paints. etc.[18,34–36] An abundant water-soluble natural polymer is starch, while the most produced synthetic water-soluble polymers include polyvinyl alcohol (PVA) and variants of polyacrylic acid and polyacrylamide.[36–39] The most well-known biomedical-grade water-soluble polymers are the polyethylene glycols, or polyethylene oxide (PEO).[40] They are used as polymer excipients for pharmaceutical formulations. A large number of synthetic-natural polymers are the polycarboxylate or hydroxy cellulose derivatives.[3,4,6] Many of their properties, synthesis methods, cost, and predictable chemical production make them the polymer of choice for numerous applications, e.g., cosmetics, packaging, adhesives, paints, coatings, etc. However, their environmental impact and biodegradability are a concern.[35] It is important to use standardized tests to assess water-soluble polymer biodegradation and environmental impact to avoid incorrect assumptions and false conclusions.[39,41]

Some recent studies, reviews and applications are as follows:

Polyvinyl alcohol (PVA), produced by hydrolyzing vinyl esters like poly(vinyl acetate), is a versatile polymer used as a thickener, stabilizer, and hydrogel inducer; its applications span packaging, textiles, and adhesives, where it enhances the processability of gels and coatings as thermoplastics.[42] Blending with other components such as starch and cellulose also enhances degradability.[43] Tannic acid (TA), for example, a dynamic and adaptable catechol group modifier, when combined with PVA, enables multifunctional adhesives.[44] PVA can also be used as a cement additive for both construction and drilling.[45] Recently, the environmental fate, waste disposal and sewage, environmental persistence, and unintended biological consequences of many water-soluble polymers in commercial production have been a concern.[36,38] Enhancing the biodegradability, compatibility, water solubility, and thermal properties of PVA through blending with natural polymers can be key to many environmentally friendly applications.[46]

PVA has several applications in advanced and additive manufacturing or 3D printing.[47,48] In fused deposition modelling (FDM) 3D printing, PVA is often used as a water-soluble print support material, combined with other thermoplastics such as PLA, ABS, and PETG. For example, PVA dissolves after printing when used in a multimaterial print. Other sustainable materials include optimizing binder jetting of water-soluble, compostable, biodegradable, and recyclable polymers.[49]

Hydrogels are chemically or physically crosslinked, hydrophilic polymers that swell by absorbing significant amounts of water without dissolving.[29] Hydrocolloids can modify the viscosity and texture of fluids and formulations. They can be made from natural, semi-synthetic, and synthetic polymers. They can function as thickeners, gelling agents, binders, emulsifiers, and stabilizers, and have applications in packaging films and coating formulations. In the food industry, they are used for enhancing food systems with optimized rheology, compatibility, organoleptic properties, preservation, and nutritional value. Natural polymer-based hydrocolloids are highly preferred in the food and personal care industries due to their biodegradability, biocompatibility, and non-allergenic effects.[37] The integration of AI/ML methods for hydrogel design and synthesis can improve the precision of applications in biosensing, drug delivery, and tissue engineering.[50]

New macromolecular compositions and microstructures are key. A recently reported PISA (polymerization-induced self-assembly) method uses RAFT (reversible addition-fragmentation chain transfer) to produce high-molecular-weight, compatible, water-soluble polymers.[51]A low-viscosity route to high-MW water-soluble polymers was demonstrated with PISA and RAFT that exploits the sensitivity of poly(N-acryloylmorpholine) to various salt concentrations.[52] These methods, when combined with the production of block copolymers with other polymers such as polyphosphoesters, poly(amino acids), and modified polyesters, can also enhance hydrolytic and enzymatic degradation. The resulting functional nanostructures also have many applications in drug delivery and environmental remediation.[53] Hyperbranched or arborescent topologies enable high interfacial activity for multivalent, super-selective interactions at the end group. These properties open new applications for water-soluble polymers as interfacial materials, nanomaterials, and cooperative binders.[54]

Water-soluble polymers are used in the agro-industry for dispersing adjuvants in herbicides, pesticides, and fungicides to control disease and increase crop yield.[41,55] They are also used as seed coatings. They provide thickening, stabilizing, adhesive, and gel-forming properties in formulations and coatings. Synthetic polymers and natural polymers are used. For example, xanthan gum is considered an eco-friendly alternative to many synthetic products.[55]

Water-soluble polymers are also used in the oil and gas industry, both upstream and downstream.[53,56] These polymers are also used in polymer flooding, a widely used chemical enhanced oil recovery (CEOR) method in the oil and gas industry. It can increase oil sweep and displacement efficiencies in wells by controlling pressure and adsorption in porous media.[56] Reports on the application of water-soluble polymer inhibitors as corrosion inhibitors in metals and alloys are abundant.[57]

2.3 Polyelectrolyte Polymers

Polyelectrolytes have ionic or charged repeating units that dissociate in polar solvents like water (Figure 2). The charge is either part of the main chain (ionenes) or is part of a repeating unit side group. They can be classified by charge (anionic, cationic, amphoteric, or zwitterionic), strength (weak or strong), and origin (natural or synthetic). Most are also water-soluble. Table 1 provides a list of common polyelectrolytes. A more detailed classification is as follows:[58–63]

1) Charge type: anionic (polyanions) carry a negative charge, such as poly(acrylic acid) (PAA) and carboxyl methylcellulose. Cationic (polycations): carry a positive charge, such as polyethyleneimine (PEI) and chitosan. Amphoteric (polyampholytes): contain both positive and negative charges on the same chain or are zwitterionic. Mostly associated with polypeptides or synthetic polymers with acid-based conversion. Non-ionic: polymers that do not contain charges along their chain but are also charge-neutral, such as polyethylene glycol or polyethylene oxide (PEO) and polyoxazolines.

2) Dissociation strength: Strong polyelectrolytes dissociate completely in solution regardless of pH (e.g., polystyrene sulfonate). Weak polyelectrolytes have partial dissociation, with charge density dependent on solution pH or ionic strength (e.g., carboxylic acids).

3) Origin: Natural polyelectrolytes derived from biological sources, such as proteins, DNA, alginic acid, and chitosan. Synthetic polyelectrolytes involve polymers like polyacrylamide and poly(methacrylic acid). Chemically modified biopolymers are natural polymers with synthetically added functional groups (e.g., modified cellulose, xanthan gum).

4) Architecture and composition: Linear, branched, or cross-linked (e.g., hydrogels) and copolymers or grafts.

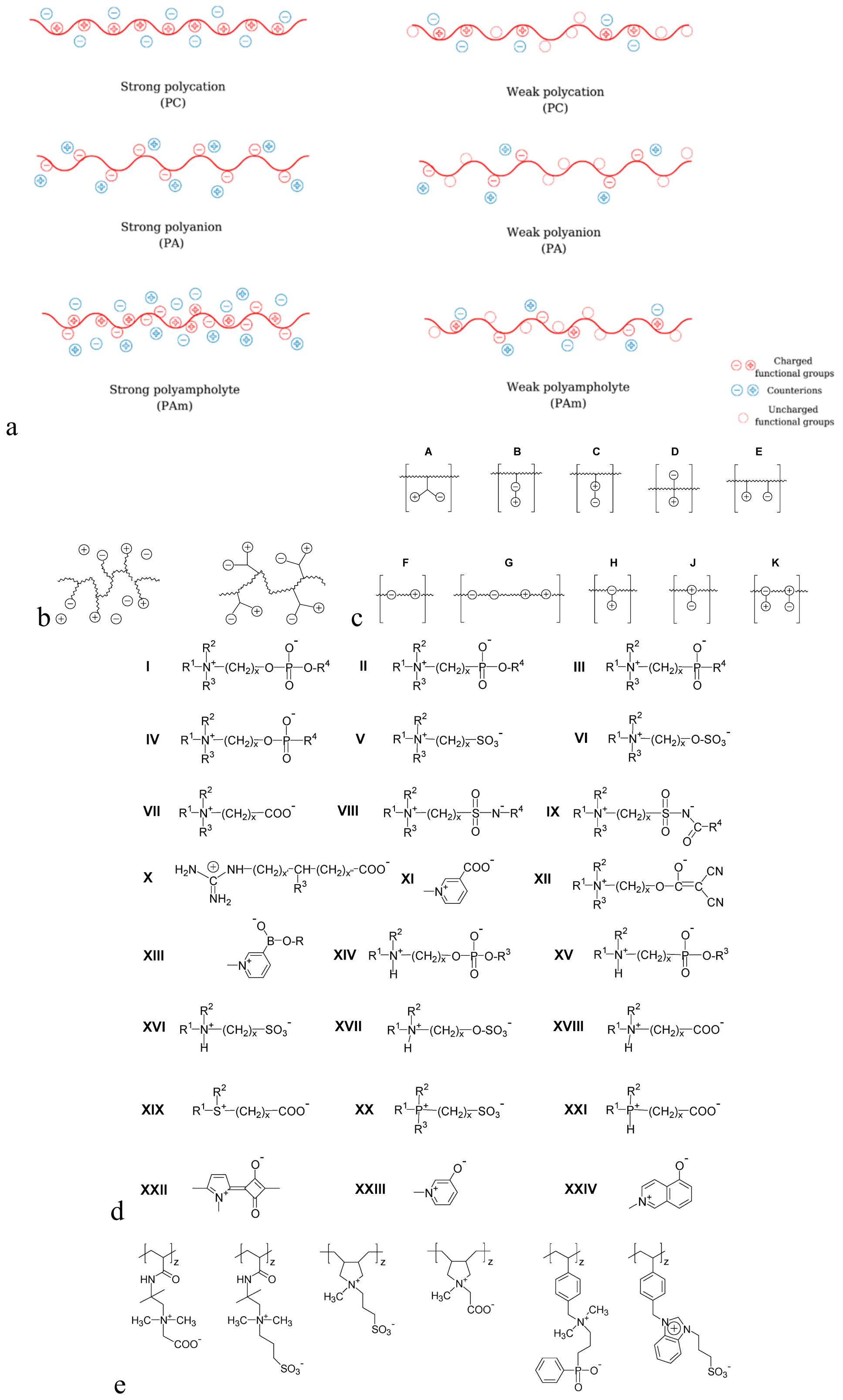

Figure 2. a. Classification of polyelectrolytes according to the nature of the functional groups and the ionic charge. (Figure and caption from reference[64] with no change under CC BY 4.0 license[8])

b. Simplistic model of polyampholytes (left) and polyzwitterions (right). c. Possible distribution of ionic groups within polyzwitterions. d. Zwitterionic groups, which have been incorporated in polymers: ammoniophosphates (phosphobetaines or lecithin analogues) I and XIV, ammoniophosphonates (phosphonobetaines) II, IV and XV, ammoniophosphinates (phosphinobetaines) III, ammoniosulfonates (sulfobetaines) V and XVI, ammoniosulfates VI and XVII, ammoniocarboxylates (carbo- or carboxybetaines) VII, X, XI, XVIII and XXI, ammoniosulfonamides VIII, ammoni-sulfon-imides IX, guanidiniocarboxylates (asparagine analogs) X, pyridiniocarboxylates XI, ammonio(alkoxy)dicyanoethenolates XII, ammonioboronates XIII, sulfoniocarboxylates XIX, phosphoniosulfonates XX, phosphoniocarboxylates XXI, squaraine dyes XXII, oxypyridine betaines XXIII and XXIV. e.Examples of permanent polyzwitterions with improved resistance to hydrolysis compared to (meth)acrylic polymers. (Figure and caption from reference[65] with no change under CC BY 3.0 license[66])

Table 1. Common Polyelectrolytes: Names and Abbreviations [59,60,64,67,68]

Category	**Full Chemical Name**	**Abbreviation**	**Charge**	**Strength**
Synthetic Anionic	Poly(sodium 4-styrenesulfonate)	PSS	Negative	Strong
	Poly(acrylic acid)	PAA	Negative	Weak
	Poly(methacrylic acid)	PMAA	Negative	Weak
	Poly(vinylsulfonic acid)	PVS	Negative	Strong
	Poly(2-acrylamido-2-methyl-1-propane sulfonic acid)	PAMPS	Negative	Strong
	Poly(vinylphosphonic acid)	PVPA	Negative	Weak
Synthetic Cationic	Poly(diallyldimethylammonium chloride)	PDDA / PDADMAC	Positive	Strong
	Poly(ethyleneimine)	PEI	Positive	Weak
	Poly(allylamine hydrochloride)	PAH	Positive	Weak
	Poly(4-vinylpyridine)	P4VP	Positive	Weak
	Poly(vinylbenzyltrimethylammonium chloride)	PVBTAC	Positive	Strong
	Poly(dimethylaminoethyl methacrylate)	PDMAEMA	Positive	Weak
Natural / Bio	Chitosan	CHI / CTS	Positive	Weak
	Sodium Alginate	ALG	Negative	Weak
	Hyaluronic acid	HA	Negative	Weak
	Deoxyribonucleic acid	DNA	Negative	Strong
	Heparin	HEP	Negative	Strong

	Chondroitin sulfate	CS / CSA	Negative	Strong
	Poly(L-lysine)	PLL	Positive	Weak
	Poly(L-glutamic acid)	PGA	Negative	Weak
Conjugated / Optical	Poly[9,9-bis(3'-tert-butylpropanoate)fluorene]	PFE	Anionic	Strong
	Poly[9,9-bis(3'-sulfonatopropyl)fluorene-alt-phenylene]	PFP	Anionic	Strong
	Poly[2,5-bis(3-sulfonatopropoxy)-1,4-phenylene]	PPP-SO3	Anionic	Strong
Specialized / Hybrid	Poly(amidoamine) Dendrimers	PAMAM	Amphoteric	pH-dependent
	Poly(sulfobetaine methacrylate)	PSBMA	Zwitterionic	Neutral
	Poly(carboxybetaine methacrylate)	PCBMA	Zwitterionic	Neutral
	PEDOT : Polystyrene sulfonate	PEDOT:PSS	Conductive	Complex
	Perfluorosulfonic acid (Nafion)	PFSA	Negative	Strong

They have many applications:[58–60,62,63] polyelectrolytes are widely used as flocculants and coagulants in water treatment to remove suspended solids and in drug delivery systems and functional coatings. Their ability to form complexes allows them to be classified as having properties suitable for membranes, battery applications, dielectric media, etc.

The following are some perspectives, reviews, and examples.

Polyelectrolytes, including their behavior in homogeneous solutions, thermodynamic instability, gelation, transitions (kinetic and multiphase), and biological implications, are outlined in a previous perspective article.[68] It is important to understand the roles of composition and monomer sequence in electrostatic interactions in synthetic polymers, as in biology.[69] The role of polyzwitterions is unique but it can be challenging to synthesize such materials.[65] Furthermore, diblock /multi-block polycations and polyanions have been studied to understand tunability, e.g., using styrene and PEO polymers.[62] Their surface adsorption properties are also of interest.[61] Our early studies included single-molecule fluorescence spectroscopy, which revealed heterogeneous transport mechanisms for molecular ions in a polyelectrolyte.[70] Single ion diffusive transport within polyelectrolytes such as a poly(styrene sulfonate) polymer brush matrix was also probed and modeled by fluorescence correlation spectroscopy.[71]

Amphiphilic and phase-segregated polymers are also significant. We reported that poly(vinylpyridine-N-oxide) block copolymers exhibit ion mobility with enhanced segregation strength, driven by a high Flory–Huggins interaction parameter and its impact on microphase-separated domains.[72] The investigation of [polystyrene-b-poly(2-vinylpyridine)]n star-block copolymers with poly(2-vinylpyridine) corona blocks has been reported as well.[73] Amphiphilic hydrophobically end-modified poly(2-isopropyl-2-oxazoline)s were studied at the air/water interface using the Langmuir method and on solid substrates via Langmuir-Blodgett (LB) film deposition.[74] Their temperature-dependent interfacial properties revealed specific transition states related to their morphology.

Topological control with branched polyelectrolytes, linear polyelectrolytes, and poly(ionic liquid)s yields unconventional assemblies and complex hierarchical structures.[60,62] They can also provide interesting viscoelastic characteristics and stimulus-responsive behavior. Electrostatic interaction with small molecules, linear polyelectrolytes, block copolymer polyelectrolytes, and branched polyelectrolytes can lead to new hybrid nanoparticle and multilayer ultrathin films. Possible applications encompass responsive microcapsules and ion-conductive membranes, as described previously.[60] We have reported on PEG-terminated crown-ether-based supramolecular adhesives that demonstrate ion interactions at ambient temperature, thus promoting the formation of high-performance materials.[75] In addition, polyelectrolyte Covalent Organic Frameworks (COFs) based on oligo(ethylene oxide) chains act as a Li-ion polyelectrolyte interface for investigating transport in nanochannels of solid-state ion conductors.[76] A high-entropy gel polymer electrolyte was demonstrated to function effectively at high temperatures for Li-metal battery applications.[77]

Understanding the role of charge-neutral polyzwitterionic polymers is important because of their interesting transport and complexation properties in solution and the solid state. We have conducted several studies at ORNL, including characterizing films of polyzwitterions in neutral and electric fields using Neutron and X-ray reflectometry. This was used to distinguish between electrostriction and mass loss mechanisms.[78] The examination of anti-polyelectrolyte and polyelectrolyte phenomena on polyzwitterionic polymer conformations was established, and the salt and concentration effects on dilute aqueous solutions were described.[79]

Gold nanoparticles with a terthiophene polyelectrolyte solution[80] and the quantitative adsorption process of multi-zwitterionic polymers on gold surfaces were reported.[81]

Cationic polyelectrolytes, with positively charged functional groups, are important for food solutions, water treatment, medical, cosmetic, textiles, etc., applications.[58,59,82] Quaternary ammonium salt polymers based on diallyl dimethyl ammonium chloride (DMDAAC) are commonly applied as biocides and coagulants for water treatment.[83] Polyelectrolytes are utilized in many water-treatment facilities as flocculants and coagulants to improve purification and water recycling, which involves common anionic and cationic polyacrylamide polymers.[84]

Various water-soluble and polyelectrolyte polymeric corrosion inhibitors used in the oil and gas industry were outlined previously, along with specific design principles and mechanisms of action.[85]

Finally, the differentiation between synthetic and bio-derived polymers is important.[82] For biological applications, it is important to recognize the role of polyelectrolytes in DNA or protein-based complexes with regard to structure, charge, and charge patterning.[86] Antifouling properties are important, particularly with the application of non-ionic water-soluble and polyzwitterionic polymers. We have examined the permeability of anti-fouling PEGylated substrates with surface-sensitive methods, including fluorescence correlation spectroscopy.[87]

2.4 Polyelectrolyte Complexes

Polyelectrolyte complex (PEC)-based materials are made from the complexation of oppositely charged polyelectrolytes (Figure 3).[88] Their coacervation and precipitation have become key to their possible applications in membrane or separation processes. The control of polyelectrolyte complexation and solubility is key to processing these materials as a class, or even as plastics. For example, the effect of solvent quality on the phase behavior of PECs is strongly influenced by salt concentration and non-coulombic interactions, as shown in phase diagrams.[89]
The mechanism of swelling-induced instability can also be attributed to counterions and their transport, with water acting as a plasticizer.[59] A practical way to view PECs is their processability as saloplastics.[90] Chemical specificity in polyzwitterion-polyelectrolyte coacervates was outlined previously, noting a distinct difference in behaviors between polycations and polyanions.[91]

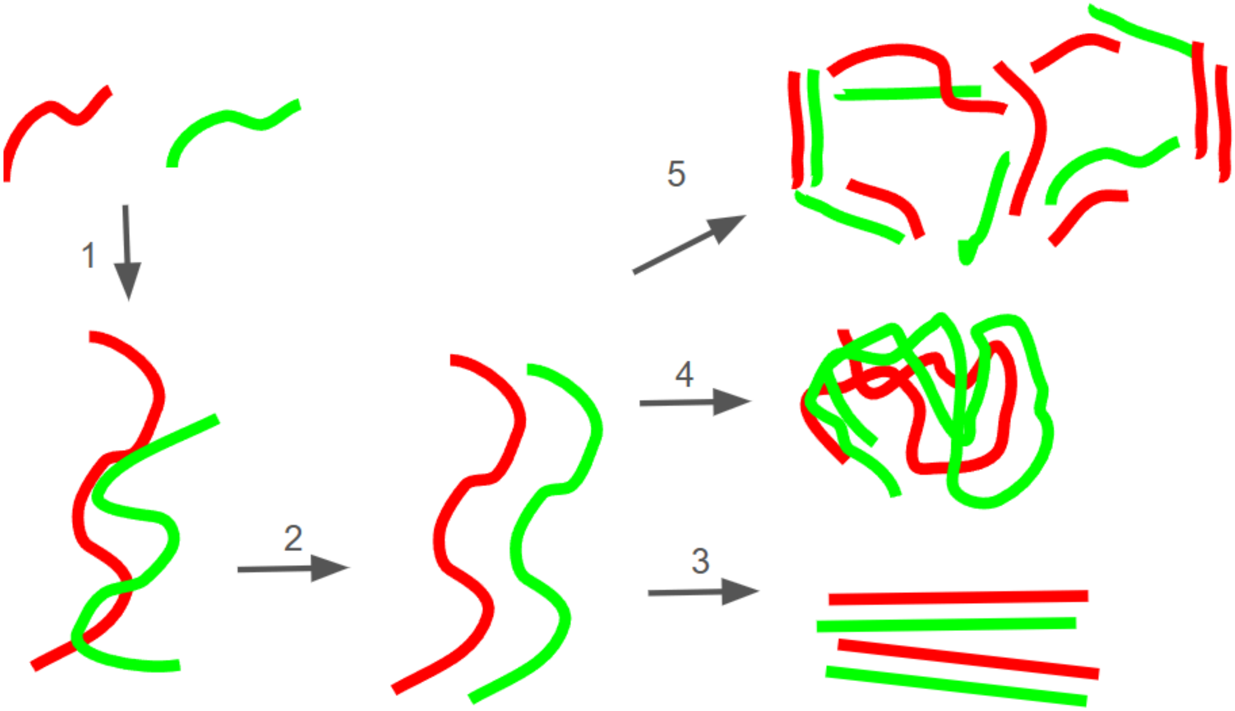

Figure 3. Formation of polyelectrolyte complex from cationic (red) and anionic polymer chains (green) via random complexation (1), rearrangement (2), and secondary complexations: fiber alignment (3), entanglement (4), or network formation (5). Inspired by reference. [92]

Control of the different parameters that affect polyelectrolyte complexation, including charged pairing, salt effect, concentration, and hydrogel formation, is essential for scalable synthesis as shown in Figure 3 and 4. A greater understanding can yield interesting PEC-based materials for applications in separation, using porous membranes, hydrogel and hydrocolloids, functional coatings, barrier materials, sealants, adhesives, saloplastics, and extraction media (as shown in Figure 5).[58,64,93] Accurate prediction of their complexation behavior employing multiscale modelling and AI/ML approaches can lead to future designs of high-performance PEC materials for novel, demanding applications.

A more significant transition in PECs is their application and convergence with bulk coacervate materials and ultrathin polyelectrolyte multilayers (PEMs).[94] Membrane applications are among the most widely anticipated uses of PECs and PEMS. Polyelectrolytes are interesting materials for next-generation membranes and separators.[59] PEM saloplastics have been reported as multiresponsive ion-exchange reservoirs and catalyst supports.[95] The roles of polycations and cross-linking in polyelectrolyte multilayer membranes have been investigated.[96]

Layer-by-layer (LBL)- fabricated polyelectrolyte multilayers (PEMs) have been explored by many groups (Figure 4).[58,63,97] Opposite-charge complexation, exchange, or ion displacement were driving forces for film formation. Additional non-covalent interactions, polarity, molecular weight, ion transport, charge density, and the glass transition (Tg) were found to be important parameters for nanostructuring layers and controlling thickness.[98–101]

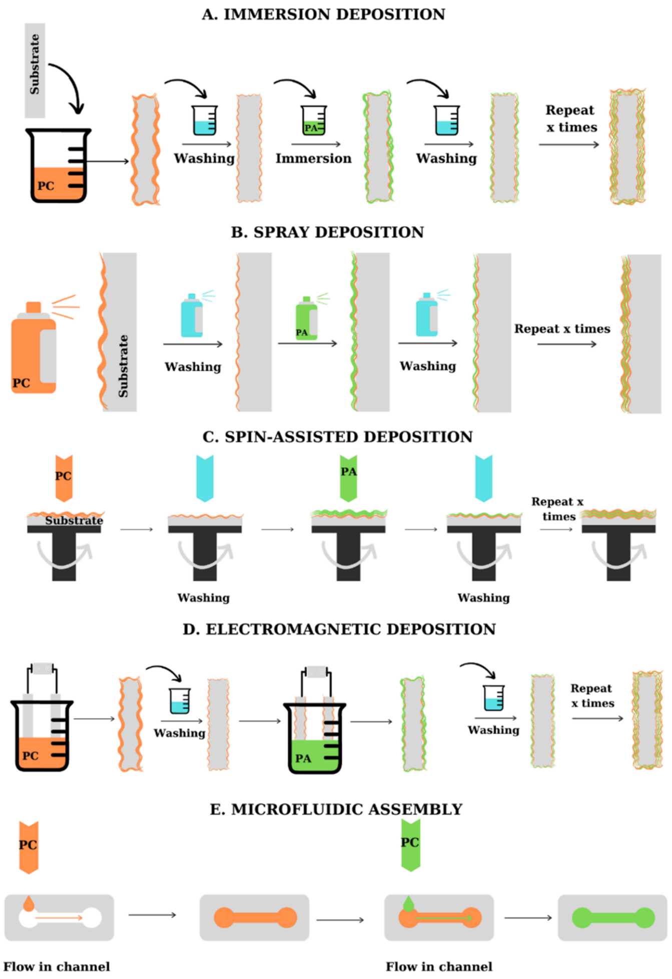

Figure 4. Schematic representation of the main LbL methods used for the fabrication of PEMs. (A) Conventional immersion deposition of PEs on flat substrates, (B) Spray deposition of PEs on flat substrates, (C) Spin-assisted deposition of PEs on flat substrates, (D) Electromagnetic deposition of PEs on flat substrates, (E) Microfluidic assembly of PEs on microfluidic materials. (Figure and caption from reference[64] with no change under a CC BY license 4.0[8])

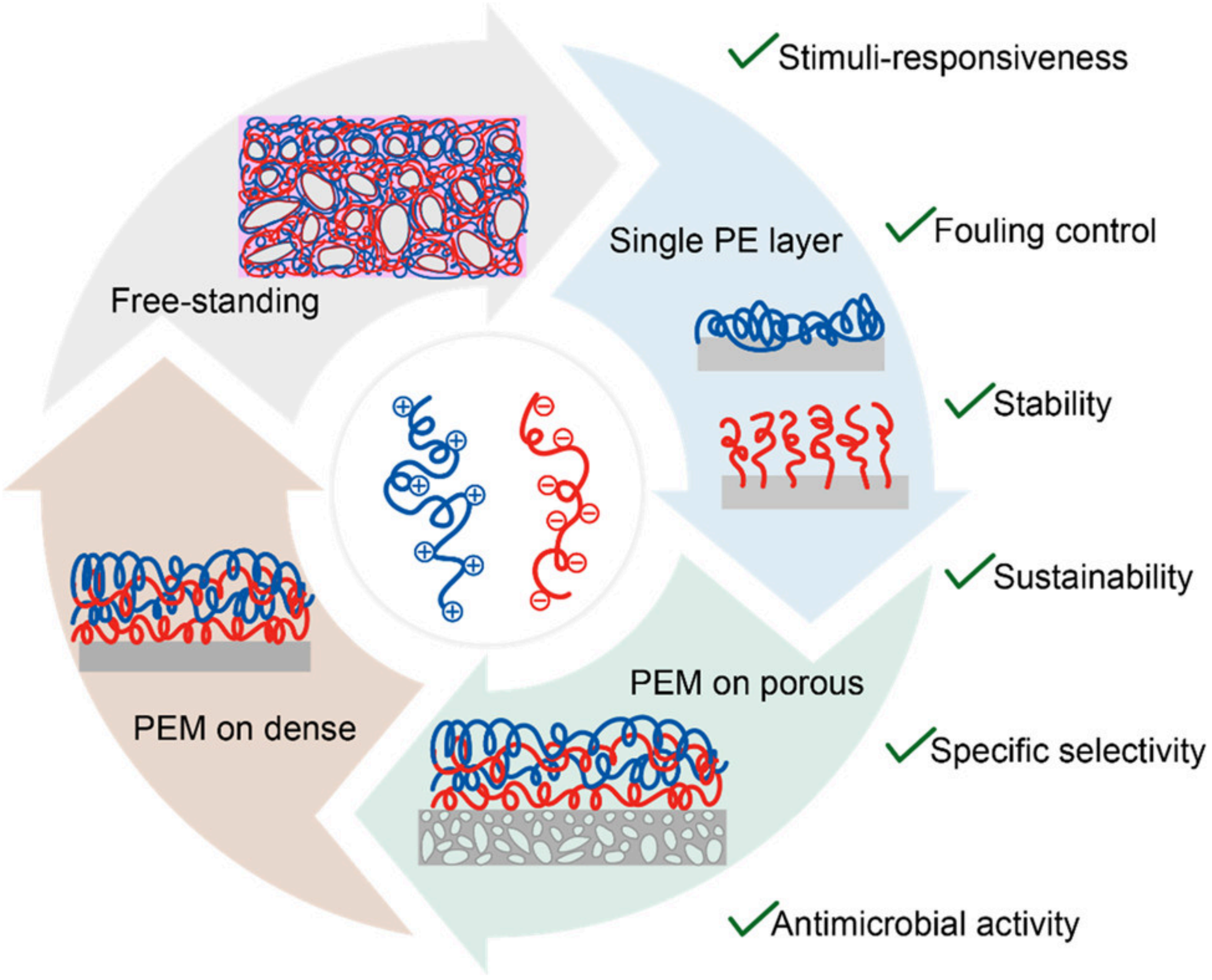

Figure 5. PE- and PEC-based membrane systems and their general functionalities. (Figure and caption from reference[59] with no change under a CC BY-NC-ND 4.0 license[102])

Most of the studies in our group were conducted early using PEM via alternating adsorption of oppositely charged polyelectrolytes. The ability to nanostructure and control the architecture and composition of PEMs via solution adsorption or wet-spinning is well documented. We have contributed much to this body of research:

Polyelectrolyte adsorption kinetics and alternate adsorption properties as ultrathin films can be monitored in situ and modeled using quartz crystal microbalance (QCM) techniques.[103] An earlier report using in-situ surface plasmon resonance (SPR) spectroscopy revealed surface assembly and kinetics.[104] We examined charge-dependent transport switching of single molecular ions across different diffusion media using a weak polyelectrolyte multilayer film.[105] The role of weak polyelectrolytes in pH-sensitive bipolar behavior and ion-permselective thin films was previously described.[106]

Initial work on using LBL approach to analyze optical properties in PEMs and dye assemblies was established by in-plane photoalignment of liquid crystals in films of azobenzene−polyelectrolyte fabricated by LBL techniques.[107] Investigations on molecularly ordered azobenzene dyes and polycation alternate multilayer films demonstrated aggregation states and layer order determined by polarized alignment.[108] Photoisomerization of the azobenzene dyes was the underlying mechanism responsible for this polarization behavior.[109] Attenuated total reflection (ATR) and SPR were used to study the dielectric, photoinduced alignment, and emission properties of LBL polyelectrolytes films with azo-dyes.[110,110–113] Other related studies included: a) polyelectrolyte thin films and complexes with viruses. This was based on the electrostatic LBL assembly of fibrous tobacco mosaic virus (TMV) biofilms.[114] b) a PEM method was used to fabricate the soft layers of a self-folding polymer–metal bilayer particle, which was then separated by transfer printing from an oxygen-reactive-ion-etched pattern.[115] and c) modulating electrochemical activities in polyaniline/titanium oxide hybrid combined using sol-gel processes for nanostructured LBL films.[116]

The ability to create nanostructure ultrathin films includes PEM and other self-assembly or polymerization processes; for example, surface-initiated polymerization (SIP) was combined with LBL and macroinitiators to fabricate grafted polymer brushes.[117] Using such methods, free-standing films were built with semi-fluorinated block copolymer brushes made using layer-by-layer polyelectrolyte macroinitiators.[118] A stimuli-responsive binary mixed polymer brush was grafted by employing the LBL-SIP method and fabricated similarly as free-standing films.[119] To vary the surface initiator density, a Langmuir−Schaefer (LS) macroinitiator was applied on a substrate by grafting a thermosensitive polymer brush via surface initiated-atom transfer radical polymerization (ATRP).[120] We demonstrated dual-stimuli-responsive polymer films using a binary architecture, combining an inner polyelectrolyte multilayer (PEM) with a surface-initiated polymerization (SIP) brush approach to achieve distinct pH- and temperature-responsive properties.[121]

Electropolymerization is a process where electrode surfaces act as substrates for fabricating PEM layers, utilizing potentiostatic or cyclic voltammetry (CV) methods to facilitate electrochemical reactions and cross-linking. We were the first to demonstrate the electropolymerization of LBL-PEM precursor polymer films.[122] Polyelectrolytes of opposite charges can be built up with the help of PEM layers and the electropolymerizable side group activated for cross-linking. The electrochemical cross-linking of polyelectrolyte−carbazole precursor films was additionally confirmed using conducting atomic force microscopy (C-AFM).[123] C-AFM also demonstrated the electro-nanopatterning of surface relief gratings on azobenzene LBL films.[124]

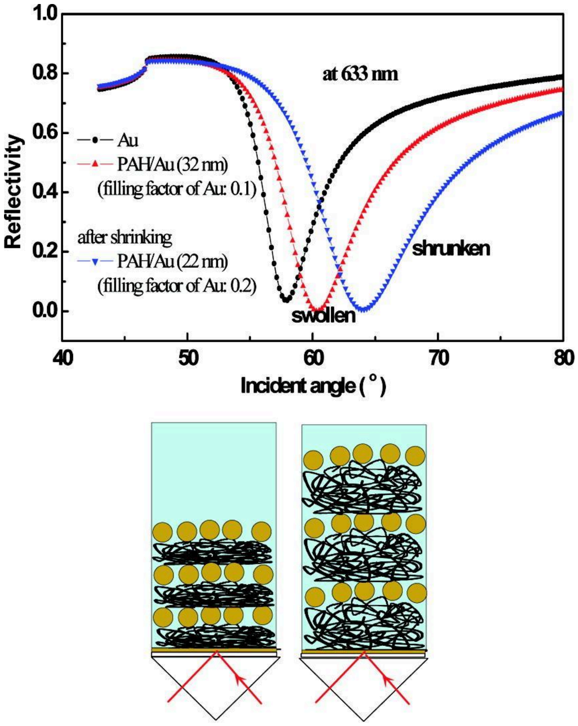

Figure 6. Calculated SPR curves of PAH/AuNPs before and after shrinking. (Figure and caption from reference[125] with no change under author reuse permission from ACS. Copyright © 2007 American Chemical Society)

PEM-type thin films, such as those with water-soluble and conducting polymers, can be combined with SPR, along with electrochemistry, for sensors and to investigate electron and ion transport behavior. One study demonstrated signal augmentation and SPR tuning in Au nanoparticle/polyelectrolyte films due to coupling with scattering and their localized SPR or LSPR behavior, as shown in the ATR setup of Figure 6.[125]

By using a gold substrate as an electrode, the *in situ* electropolymerization and doping/dedoping of polyaniline thin films were investigated, in which a combined electrochemical and surface plasmon resonance (EC-SPR) setup enables the study of *in situ* redox behavior and ion transport.[126] The simultaneous SPR optical and electrochemical examination of polyaniline and sulfonated polyaniline LBL conducting films is correlated to their ion transport and redox activity.[127] The same setup can be used to investigate the electroactivity of polyaniline multilayer films . This was carried out in neutral solution to exhibit electrocatalyzed oxidation of β-Nicotinamide Adenine Dinucleotide (NAD), and to demonstrate sensing.[128] PEDOT: PSS LBL-PEM films can be fabricated and nanopatterned as memory devices.[129] An innovative setup that enabled simultaneous in situ electrochemical, SPR, and AFM measurements has been described to explore the electropolymerization of conjugated polymers.[130]

The LBL-PEM assemblies of polyelectrolytes can be extended from nanostructured films to colloidal nanoparticle core-shell coating and hollow shells.[63] We reported on the LBL shell self-assembly of polyaniline (PANI) and sulfonated polystyrene (PSS) to generate multilayer-coated colloidal particles and hollow shells[131]. To further analyze a polyelectrolyte assembly, cross-linked, luminescent spherical colloidal particles and hollow-shell particles were built using polyfluorene-labeled polyelectrolytes.[132] A fuzzy ternary particle system was developed by combining SI-ATRP with an LBL-colloidal core−shell system and an outer macroinitiator shell.[133]

3.AI/ML Workflows in Polymers

Artificial Intelligence (AI) is a broad field dedicated to creating systems within biological intelligence or, in the future, replacing native intelligence in humans.[134–137] This includes reasoning, learning, problem-solving, and perception. At its core, AI seeks to simulate cognitive functions by using mathematical logic and computational power to process information. While "General AI" remains a theoretical goal of creating machines with human-like consciousness, today’s "Narrow AI" excels at specific functions, such as language translation or image recognition, by identifying patterns within vast datasets. Machine Learning (ML) is the primary subset of AI focused on building systems that improve their performance through experience rather than explicit programming. While data science provides the broader framework—incorporating statistics, data visualization, and domain expertise to extract insights—ML provides the specific algorithms that allow computers to "learn." These algorithms use historical data as input to make predictions or decisions, effectively turning raw information into actionable knowledge through iterative refinement.

Natural Language Processing (NLP) is a specialized category of AI that enables machines to understand, derivatize, and generate human language.[138,139] Modern NLP models, particularly Large Language Models (LLMs), utilize massive datasets to predict the next word in a sequence, allowing for fluid conversation and complex reasoning. Interestingly, these same principles are applied to chemistry through SMILES (Simplified Molecular Input Line Entry System). By treating chemical structures as strings of characters—much like words in a sentence—ML models can "read" and design new molecules, accelerating drug discovery and materials science.

Supervised learning is the most common ML classification, where models are trained on labeled datasets.[140,141] Key algorithms in this space include Decision Trees, which use a flowchart-like structure to reach conclusions, and Neural Networks, which mimic the interconnected structure of the human brain to process complex data. Support Vector Machines (SVMs) are also vital here; they work by finding the optimal "hyperplane" or boundary that best separates different classes of data in a high-dimensional space. Regression analysis is used when the goal is to predict a continuous numerical value rather than a category.[142–144] To handle complex, non-linear relationships, researchers often employ Kernel Functions, which map data into higher dimensions to make it easier to analyze. A sophisticated example of this is the Gaussian Process, a non-parametric Bayesian approach to regression.[145] Unlike standard models that give a single prediction, Gaussian processes provide a probability distribution, offering a measure of uncertainty for every forecast made. Neural networks form the backbone of modern AI, consisting of layers of "neurons" that weight and process information.[146–148] While Artificial Neural Networks (ANNs) are the standard foundation, specialized architectures like Graph Neural Networks (GNNs) represent data structured as graphs, such as social networks or molecular bonds. These systems excel at capturing the relationships between individual data points, allowing the model to understand the context and "topology" of the information it is processing.

Unsupervised learning enables the use of training models on data without pre-existing labels, tasking the AI with finding its own hidden structures.[5,140,141] The most prominent technique is Clustering, which is a method of algorithm groups based on similarities. By identifying natural partitions in data, businesses can segment customers, and scientists can categorize new species or genomic sequences without needing a human to tell the machine what to look for beforehand.

In many datasets, there are too many variables (or "dimensions") for a model to process efficiently. A solution is dimensionality reduction techniques like Principal Component Analysis (PCA) simplify this by condensing the data into its most important features while losing as little information as possible.[149] Other methods include Random Projection,[150] which is computationally faster for massive datasets, and Multi-Dimensional Scaling (MDS),[151] which focuses on preserving the relative distance between points to visualize high-dimensional data in 2D or 3D space.

The frontier of the field lies in Agentic AI, where models don't just predict text but take autonomous actions to achieve goals.[152–155] This is often powered by Reinforcement Learning (RL), where an agent learns through trial and error, receiving "rewards" for correct actions.[156–158] This is frequently paired with Active Learning,[145,159–161] a strategy where the model identifies which data it is most confused by and asks a human for specific labels. Together, these methods allow AI to move beyond static analysis and into active, goal-oriented problem-solving.

3.1 Polyelectrolytes and AI/ML Driven simulation and SDLs

3.2 AI and Machine Learning in Polymers

In the synthesis and optimization of water-soluble polymers, many laboratories still use a serial, multi-step process, which is closer to a trial-and-error approach. It is necessary to enable faster, higher-volume experiments to evaluate and rapidly optimize material designs through a hierarchy of simulations. Statistical methods such as linear regression, Bayesian optimization (BO), decision trees, and k-means clustering are useful for designing algorithms and experiments that yield well-defined structure-composition-property relationships. The objective is to develop more ML-driven models and training algorithms of data derived from LLMs that can lead to "water-soluble polymer informatics." This should encompass natural, synthetic, hydrogel, and polyelectrolyte polymers. By converting chemical structures into machine-readable formats, the chemistry domain will be more amplified with the ML domain in current structured datasets, e.g., PoLyInfo, PubChem, CAMPUS, PI1M, Khazana, and CROW.[141,162–168]

The development of these large polymer libraries will be invaluable for both academic research and industrial R&D, especially for more generative AI exploration. The use of chemical descriptors with artificial neural networks (ANNs) can enable the development of large-scale data sets for problem-solving. More complex or generative models can be employed, which include generative adversarial networks (GANs), graphical neural networks (GNNs), and Graph Convolutional Neural Networks (GCNNs).[137,169–172] Topological descriptors can be generated from SMILES,[173] where the repeat unit descriptor is primary. To represent 2D space and geometric descriptors in 3D, Dragon, Modred, and RDKit platforms can be used.[174–176] By developing a polymer natural language processing (NLP) framework, it becomes more adaptable to the nuances of macromolecular systems. For example, the polyBERT fingerprint has been developed and is derived from a deep learning (DL) model trained on polymers' SMILES (PSMILES) using a transformer-based language model.[177] Using more hierarchical descriptor models can accelerate the process. It can improve the prediction of polyelectrolyte SCP, based on constitutional repeat unit, connectivity, hierarchy, and topological features that align with the PoLyInfo database, for example.[164]

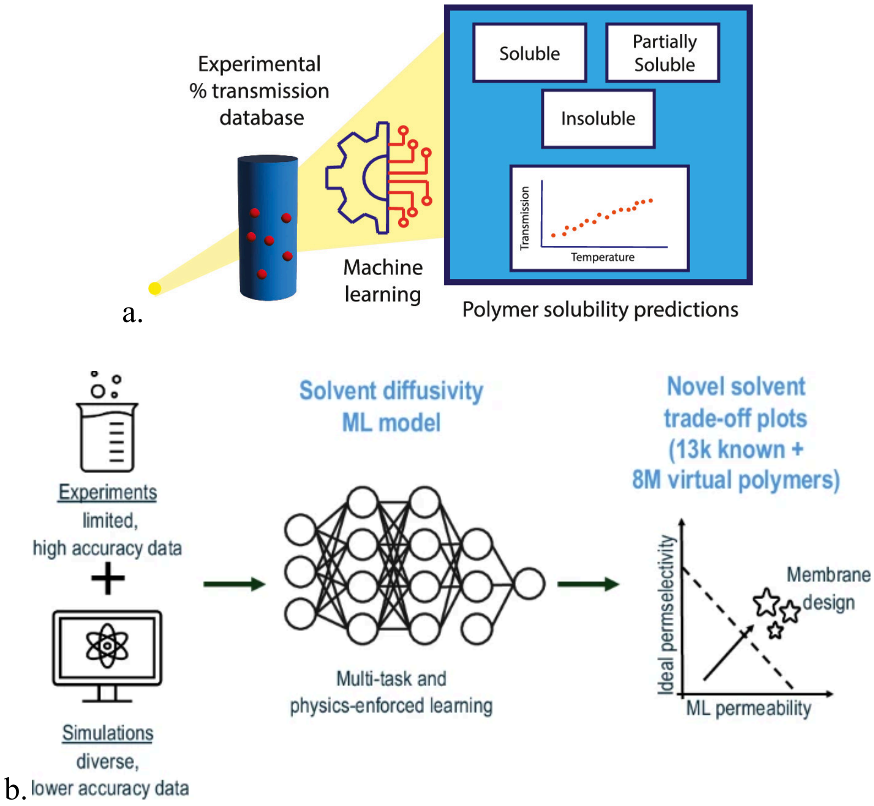

Figure 7. a: ML models to facilitate polymer solubility predicting (Figure from reference[178] with no change under a CC BY 4.0 license[8]); b: ML-driven membrane design (Figure from reference[179] with no change under a CC BY NC-ND 4.0 license[180])

Some recent examples of the use of AI/ML for the design, synthesis, and testing (solubility) are as follows:

AI/ML-driven computational modeling, HTE, and autonomous labs will facilitate the rapid development of water-soluble polymers as sustainable, high-performance materials.[162] High-quality polymer data and ML help untangle intricate relationships in polymer design and complexation behavior.[181] It is also key to novel functional and sustainable polymers.[182] Smart polymer characterization can be augmented by deep learning.[183] A prime example is applying ML-driven models for predicting polymer solubility across a wide range of solvents, concentrations, and temperatures.[178] Polymer solubility is critical for new formulations and solution processing, while ML models and robust data analytics significantly accelerate the estimation of solubility parameters and optimize formulation development (Figure 7).[178,184,185]

ML-driven inverse design and identification of novel PECs and PEMs for membranes and separation applications can be an immediate goal,[186] as well as ustainable and superior performance in polyelectrolyte and PEM membranes suitable for organic binary solvent diffusivity and separations.[179] ML-methods for predicting homopolymer and copolymer solubility can unlock a large range of possibilities for polymer composition design.[187]

The convergence of interpretable AI algorithms, statistical techniques, Bayesian optimization, theoretical modeling, and autonomous experiments was summarized in a recent review, given that ion transport in solid electrolytes is critical for many applications including batteries.[188] LLM-curated datasets of polymer electrolytes containing their ion-transport characteristics derived from MD simulations were provided for the *de novo* design of polymer electrolytes via GPT- and diffusion-based generative models.[189] Chemistry-informed ML was employed to improve forecasting accuracy for solid polymer electrolyte (SPE) ionic conductivity with the use of LLMs, optimized chemical descriptors for ion transport.[190] An accelerated screening of polyelectrolytes for optimal ion transport was conducted using a GNN and RL, trained based on short- and long-time-scale MD data.[191] An ML-screening protocol was implemented to ionic liquids (ILs) with high ionic conductivities and wide electrochemical windows, indicating their suitability for high-energy-density lithium metal batteries (LMBs).[192] ML approach was also used to explore new applications, optimization protocols, and fundamental insights in polymer and polyelectrolyte brush-grafted systems.[193]

Polyethylenedioxythiophene (PEDOT), a well-studied conducting polymer often complexed with polystyrene sulfonate (PEDOT:PSS) or other smaller anions, can be optimized for ion and electrical conductivity or mobility; for example, ML-driven MD simulations with density functional theory (DFT) and convolutional neural networks (CNNs) were adopted to comprehend multiscale and hierarchical transport processes.[194] The Polybot was introduced as an automated material laboratory, or self-driving lab, that autonomously explores pathways towards high-conductivity, high-quality PEDOT films with Bayesian optimization (Figure 8).[195] Automation and ML for expedited polymer analysis and advancement in general will be the favored path for high-throughput experimentation in the future.[196]

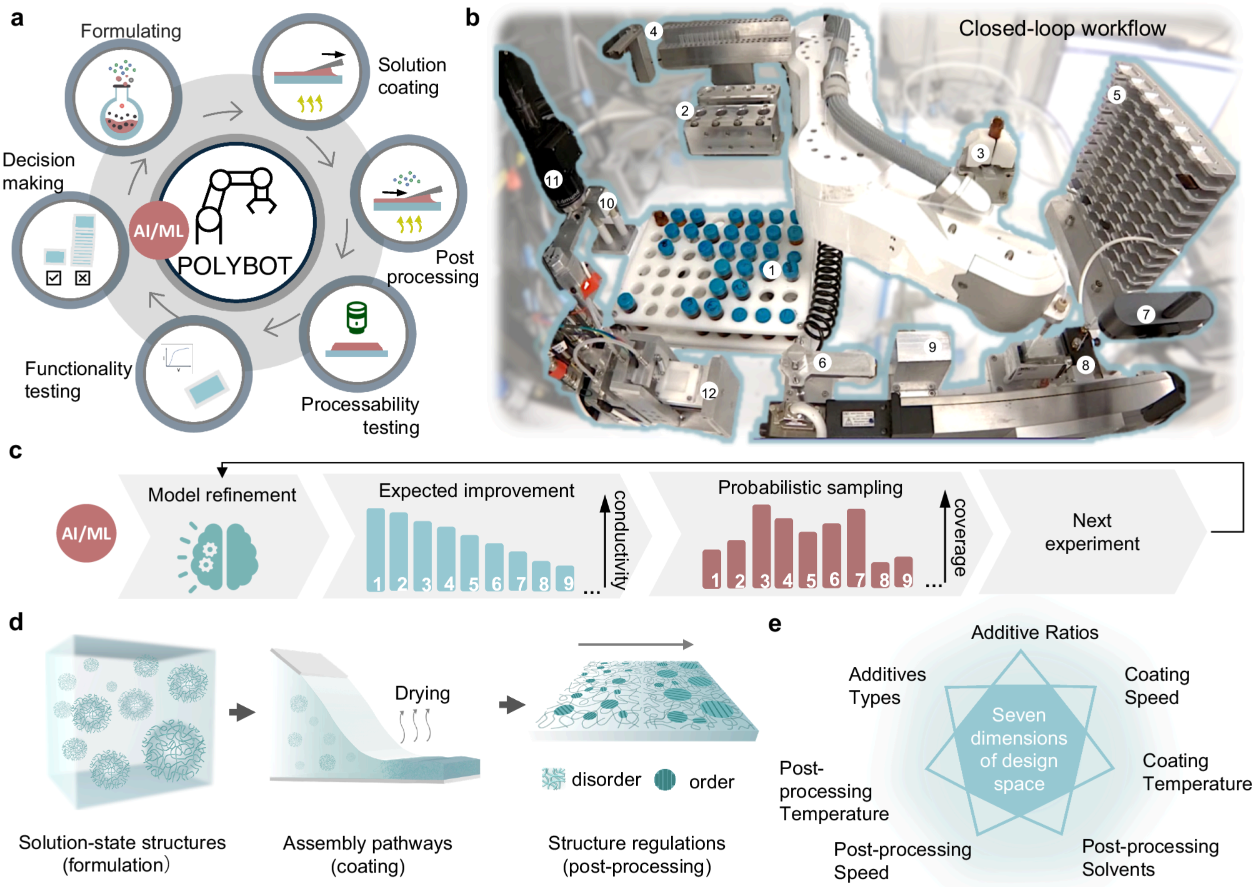

Figure 8. a Schematic illustrating the consecutive steps in the autonomous experimental workflow. b Image of the modular automated platform which includes (1) solution storage rack, (2) solution heating and mixing module, (3) capping and uncapping system, (4) pipette rack, (5) substrate rack, (6) substrate gripper, (7) imaging station, (8) blade-coating station, (9) blade cleaning station, (10) annealing station, (11) thickness characterization station, (12) electrical characterization station. c Schematic of the iterative multi-objective optimization strategy based on advanced learning algorithms enhanced by probabilistic sampling, strategically exploring undersampled areas of the search space and exploited available data to produce thin films with superior processability and conductivity. d Complex assembly pathways of electronic polymers from solution to thin films. e The total searches space to optimize the conductivity of the PEDOT:PSS. For this seven-variable problem, the full design space has 933,120 distinct data points. (Figure and caption from reference[195] with no change under a CC BY license 4.0[8])

4.Continuous Flow Reaction Chemistry, Synthesis, and Self-driving Laboratories

Although continuous flow chemistry (CFC) is used for scale-up and as an upgrade from micro- or milli-fluidics, it also enables polymer chemists to control, explore, and advance reactions that are difficult to do in batch.[197–199] These factors include flow rate or residence time, isothermal temperature control, and more uniform reagent (monomer or initiator) delivery. CFC enables control of kinetics, identification of side reactions or by-products, and understanding of selectivity. A continuous reaction under flow will allow incremental adjustments *in silico*, and online instrumentation permits systematic observations in units or space series.

Thus, it is possible to investigate the effects of temperature, pressure, stoichiometry, and concentration (volume) factors while maintaining consistent hydrodynamics or flow behavior. For LLMs, it enables the use of trained or curated datasets, with more online instrumentation for monitoring and generating more data. With the help of AI/ML, it can reduce the number of experiments and accelerate scale-up optimization. It does not have to replace batch reactions or experimentation, but can complement mapping the reaction space or a multi-step procedure. These are essential for scale-up and enable more AI/ML-driven discovery science.

Continuous-flow chemistry (CFC), separations, and analysis are methods for optimizing chemistry through unit operations and under more isothermal conditions.[197] Although previous focus on microfluidics has been demonstrated, it can also be achieved with bench-scale microreactors (via high surface area).[200] It can additionally be transformed into a high-throughput experiment platform for scalable chemical engineering unit operations.[201] The scale-up of kinetic and equilibrium-driven reactions can be better controlled by time and flow rate. Specific variants of the CFC experiment can be carried out as follows:

a) plug flow-homogeneous solutions reactions,
b) packed-bed heterogeneous reactions,[198,202]
c) control of complex flow behavior (laminar, turbulent flow, etc.),
d) gas reagents or monomers in multiphase reaction conditions,
e) safe reactions with flow reservoir and channel design,
f) photochemical or electrochemical catalyzed reactions, and
g) multi-step synthesis and purification or sequential addition of new reagents or reactor catalysts.[203]

Futhermore, for discovery and scaling up of water-soluble and polyelectrolyte materials, the following are some important possibilities: a) new mechanisms and concepts for polymerization: MW control, polydispersity, copolymerization, b) use of macromonomers, macroinitiators, polymer modification or polymer analogous reactions (PAR), c) new microstructures in polyelectrolyte dendrimer, macrocycles, catenation, knotty polymers, etc., d) microparticle and nanoparticle synthesis and PEC complexation studies, e) particle SIP and core-shell reactions e) emulsion reactions, and f) controlled micellization and core-shell self-assembly including Janus particle formation.

Notably, some of the reported advantages for polymerization reactions include:
a) controlled MW, polydispersity, microstructure, and sequence-specific polymerization, all of which are difficult to achieve in batch or bulk reactions.[204–206]
b) use of polymer-supported reactants, reagents, initiators, catalysts, and/or Scavengers in CFC is an important step.[207,208]
c) synthesis of diverse polymer materials, including nanoparticles, particles, beads, capsules, and fibers,[209]
d) new methodology development for living polymerization, copolymerization, different initiators and catalysts - for scale-up reaction engineering.[210–215]

Examples of our own CFC work in polymers and grafted particle polymers include the following: a) efficient RAFT polymerization process with ethanol/water and free radical (AIBN) initiator [216]. b) preparation of block copolymer–grafted silica microparticles in water/ethanol [217]. c) hyper-branched polymer control using photo-induced electron transfer RAFT (PET-RAFT).[218] d) hyper-branched poly[2-(dimethylamino)ethylmethacrylate] or PDMAEMA-functionalized silica microparticles by ATRP [219]. Additionally, PET-RAFT polymerizations under flow chemistry were reviewed, along with other surface-initiated reactions.[220]

CFC chemistry and microreactors for polymerization will enable high-throughput, precision synthesis and reaction-engineering. Control measures will be important and should be demonstrated for scale-up. The use of AI/ML-driven materials design and corresponding reaction routes, together with a high-throughput and automated SDL, will significantly speed up scientific discovery and optimize polymer synthesis.[221–224] The modular CFC will be a mechatronically and robotically designed configuration that can be coupled with high-throughput, online characterization.[225]

A plug-and-play environment was previously demonstrated for scalable synthesis.[226] In Figure 9, new reactor architectures and analytical techniques are now possible based on sound design engineering principles[227], enabling Bayesian optimization (BO) implementation and multi-objective optimization.[228] The ecosystem for data analytics and the development of better LLMs and agentic AI tasks will further produce higher-quality, standardized data and automate discovery processes in polymer materials.[229–231] It is evident that CFC will facilitate establishing AI/ML-driven workflows for high-throughput experimentation.[232] By constructing AI/ML-driven SDLs, it will be possible to codify chemical synthesis and materials discovery within the broader AI in science and data ecosystem.[141,233,234]

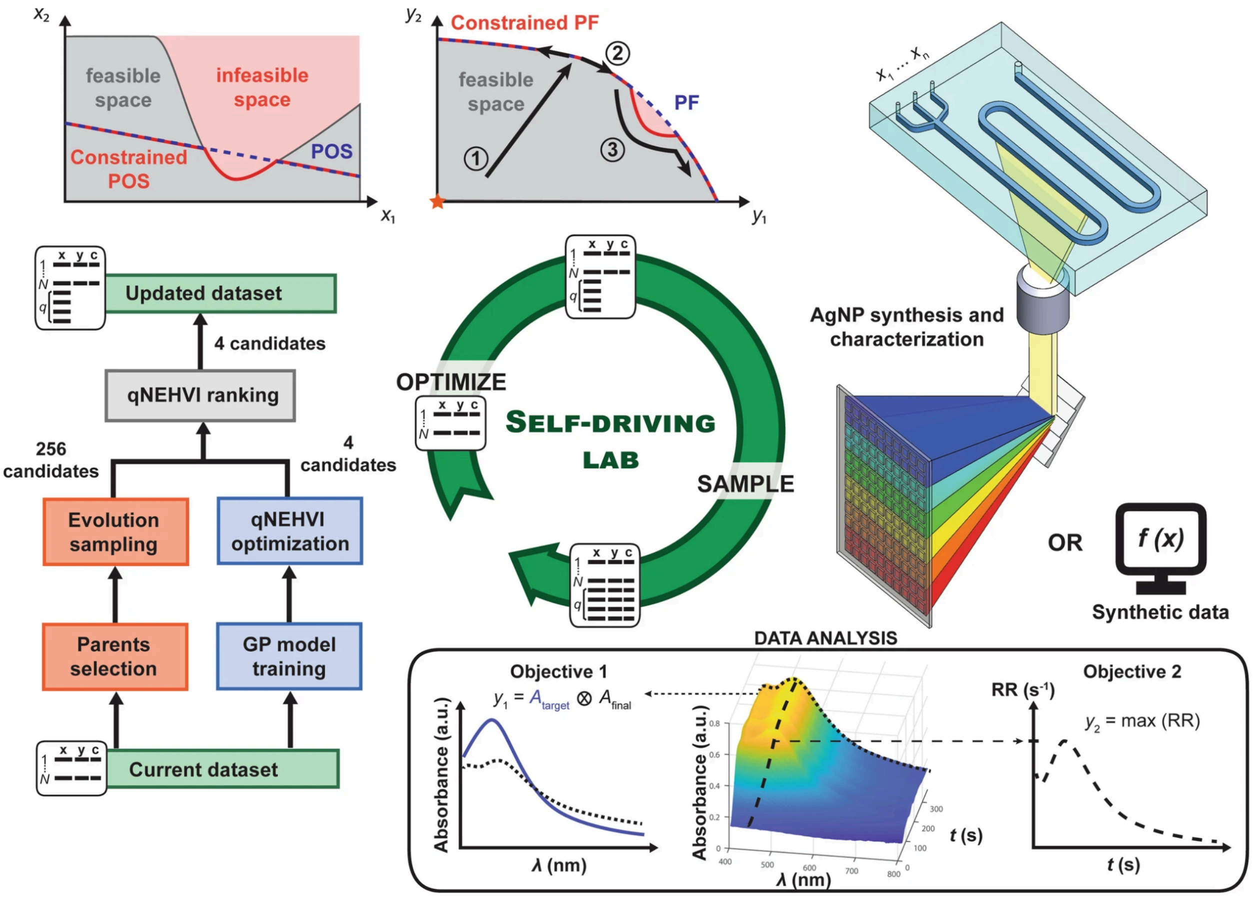

Figure 9. The illustration on the top left of the figure shows the unconstrained (blue dashed line) and constrained (red solid line) Pareto Optimal Set (POS) in a constrained decision space, and its respective projection as the Pareto Front (PF) in the objective space. EGBO algorithm (left) combines an evolutionary algorithm (orange) and a qNEHVI-BO (blue) working in parallel to suggest 4 optimal candidates for the cMOOP. The optimizer's goal is to (1) efficiently reach the PF, (2) uniformly explore the PF and (3) avoid infeasible domains near the PF (top left). The candidates are then sampled on a hyperspectral HTE platform optimizing AgNP synthesis (right) and further analysis is done to derive the objective values (bottom) before a new EGBO iteration. (Figure and caption from reference[228] with no change under a CC BY license 4.0[8])

Futhoremore, the automation and high-throughput of SDL will enable AI/ML-optimized design and simulation to obtain more data and validate it with empirical results and actual experiments.[235–237] This system will be an HTE platform,[227] that can also bridge the knowledge gap on reaction, hydrodynamics, flow, and transport behavior in multiple-phase reactors.[238] ML can eventually be augmented with RL, which can help make decisions at each step and predict future responses, ideal for multi-step synthesis in SDL.[239]Deep reinforcement learning (DRL) can be used as an effective multi-variate self-optimization strategy.[157]

5.3D Printing Polyelectrolytes and water-soluble polymers

3D Printing, or additive manufacturing (AM), has transitioned from a niche prototyping tool to a pillar of advanced manufacturing.[240–242] Unlike subtractive methods that carve away material, AM builds components layer-by-layer, enabling geometric complexities—such as internal lattices and organic topologies—that were previously difficult or even impossible to manufacture. Currently, the industry is increasingly defined by "Industrial Scale AM," where high-volume systems are integrated with AI-driven path optimization and real-time sensor monitoring to ensure "born-qualified" parts that meet rigorous aerospace and medical standards.[242–245] Table 2 compare a few common 3D printing techniques.

The most ubiquitous method remains Fused Deposition Modeling (FDM), where thermoplastic filament materials are melted, extruded through a nozzle, and quenched.[246,247] While traditionally used for low-cost modeling, advanced FDM now utilizes high-performance polymers like PEEK and carbon-fiber composites for functional parts. A related but more specialized technique is Direct Ink Writing (DIW).[248–250] Unlike FDM, which relies on heat, DIW extrudes "inks" (such as ceramics, hydrogels, or conductive pastes) that maintain their shape via shear-thinning properties and are later cured, making it a cornerstone for bioprinting and flexible electronics. For applications requiring extreme precision and smooth surface finishes, Stereolithography (SLA) is the gold standard, using a UV laser to selectively cure layers of liquid photopolymer resin.[251,252] While original SLA systems traced parts point-by-point, newer variants like Digital Light Processing (DLP) and Masked Stereolithography (MSLA) use projectors or LCD screens to flash an entire layer at once, significantly increasing throughput.[253] These methods are essential in the dental and jewelry industries, though the resulting parts are often more brittle than their thermoplastic counterparts. Industrial-grade durability is typically achieved through Selective Laser Sintering (SLS).[254] These systems use a laser to diffuse and fuse powdered materials (nylon for SLS). A major advantage of Powder Bed Fusion (PBF) is that the surrounding unsintered powder acts as a natural support, allowing for complex, interlocking geometries without the need for sacrificial structures. In the current manufacturing landscape, PBF is the preferred method for producing end-use engine components and customized orthopedic implants.

Table 2. Comparison of a few common 3D printing techniques.[240,242,245,255]

Method	**Material Type**	**Key Advantage**	**Common Use**
FDM	Thermoplastics	Low cost, wide material range	Functional prototypes
DIW	Pastes/Hydrogels	Prints non-plastics	Bioprinting, electronics
SLA/DLP	Liquid Resins	High resolution, smooth finish	Dental models, Jewelry
SLS	Polymer Powder	No supports needed, durable	End-use plastic parts
Metal laser sintering	Metal Powder	High strength, heat resistance	Aerospace, medical implants

Polyelectrolytes and PECs are polymers and materials that can be reacted, cross-linked or complexed. Their material properties not only include solubility but also insolubility as coacervates. The most processible form of PECs as viscoelastic materials is based on solution-based plastics or saloplastics.[90] Detailed methods on their preparation, solubilization, extrusion, forming, etc., have been described elsewhere.[256,257] Another important method for processing polyelectrolytes, natural polymers, and other water-soluble polymers is by 3D printing.[258] The incorporation of these polymers as soluble components in SLA crosslinkable photoresins, additives in binder jetting, and in direct ink writing (DIW) methods is widely adopted. Reports on polyelectrolyte, natural polymers, PEC 3D printing involved photoresin solubilization [259–262] and DIW 3D printing of materials in matrix solution or viscous media.[263–266]

Direct DIW 3D printing in ambient and air conditions has been reported for hydrogels.[267–269] Potential applications include biomimetic constructs, soft actuators, scaffolds for cell proliferation, ion-exchange materials, and sensors, as reported in the examples mentioned. Our group first reported 3D printing of PECs directly in air, via saloplastics preparations of the coacervates, subsequent printiing via DIW, and flushing with water to immediately condense the coacervate PEC,[258] in order to overcome rheology constraints. To print PECs in air without using water-filling methods, various silica nanoparticle compositions were prepared with the PEC to enhance the rheology for 3D printing.[270]

PECs were incorporated into photocurable hydrogel resins and fabricated via SLA 3D printing, resulting in tunable swelling behavior and controlled release properties (Figure 10).[271] For example, methacrylate and methacrylamide-based resins, such as PEGMA-co-(PNIPAM crosslinked with PEGDA), were used to showcase hydrogel 3D printing through digital light processing (DLP) in aqueous media.[272]

3D printing nanocomposite resins of natural polymers and nanofibers was established with cellulose and chitin-based materials and nanomaterials, for which we first reported on the 3D Printing of photocurable cellulose nanocrystal composites via stereolithography or SLA.[251] A mechanically enhanced 3D-printed photocurable nanocomposite incorporating Chitin nanowhiskers was demonstrated by SLA, exhibiting superior thermo-mechanical properties with just 0.5 wt% nano-additive.[273] Without the use of nanowhiskers, chitin can be 3D printed as surfactant-complexed polymers to generate a toughened polymethacrylate composites at high levels of additive concentrations. [274] High-strength SLA-printed nanocomposites were reported from photoresins containing graphene oxide, which show metastability and potential to be enhanced by annealing; for example, 3D printing of biocompatible polyurethane/poly(lactic acid)/graphene oxide nanocomposites showed improved and anisotropic thermo-mechanical properties.[275]

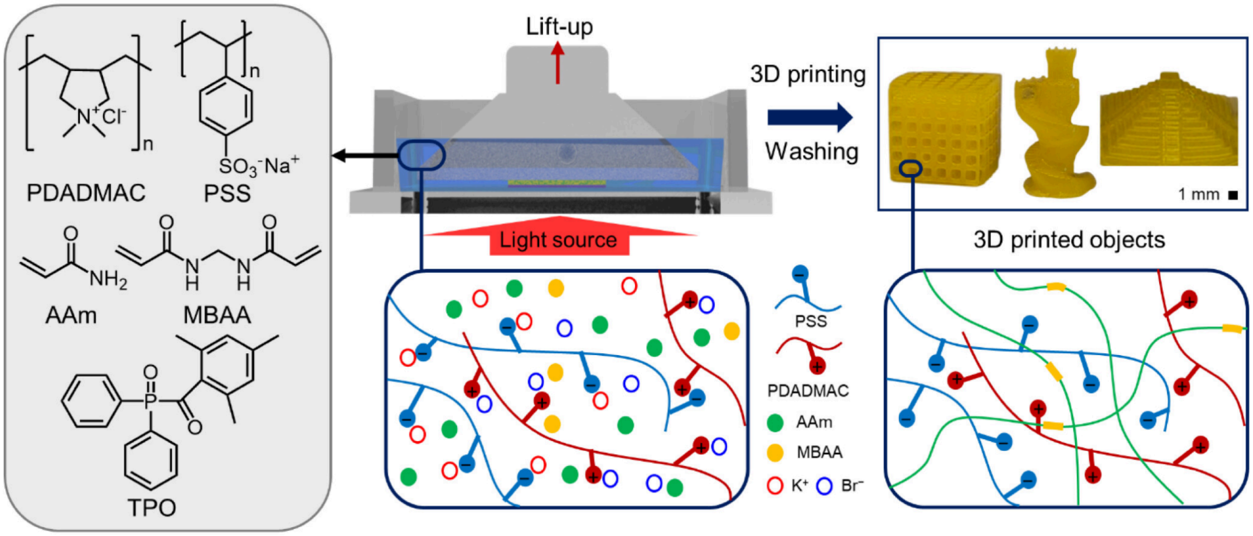

Figure 10. Schematic illustration of the MSLA 3D printing process using a photocurable hydrogel resin containing PEC. (Figure and caption from reference[271] with no change. Copyright © 2025 The Authors. Published by American Chemical Society)

Optimized 3D-printed hydrogels for tissue engineering[276] and 3D printing biomedically relevant polymer materials are summarized, along with their biocompatibility.[277] Both reviews highlighted the importance of water-soluble, biocompatible, polyelectrolytes and natural polymers applications via 3D printing. AI/ML can help explore parameter spaces for optimized 3D printing involving these polymers.[243] Not only can AI/ML optimize synthesis, but also it can improve the 3D printing process as well as automate feedback and decision-making.[278] 3D printers equipped with in-feedback-loop sensors and auto-correcting algorithms can eliminate residual stresses before they form a layer.[279] Real-time techniques are essential for defect detection, as they minimize defects or entrapped residual stresses and allow process parameters to be programmed directly in the printer before and during the printing process.[280,281] Furthermore, simulation and ML methods have the potential to augment the design and testing of sensor materials and composites, particularly via ML-driven finite element analysis (FEA).[282,283]

6.Conclusions and Future Work

AI and ML are set to transform many material classes, including water-soluble polymers, by building on fundamental research through ML-driven modeling and simulation. While natural polymers possess unique, source-dependent microstructures, generative AI can help predict their native compositions and supramolecular structures to improve harvesting and purification. Furthermore, ML-optimized LLMs can generate new (co)polymer compositions, though empirical validation and standardized testing remain necessary to address environmental and life-cycle challenges.

Polyelectrolytes and PEMs will see expanded use in batteries, membranes, packaging, and advanced manufacturing, with scaled production offering significant commercial opportunities. Beyond simple prediction, a "continuous learn and redesign" strategy using active redesigns, RL, and autonomous SDL will make previously challenging materials feasible. By moving toward creative design, researchers can re-examine discarded synthesis routes with shorter timelines. Ultimately, advanced manufacturing and 3D printing will enable new form factors and applications in membranes and biomedical engineering, a value proposition clear to both academia and industry.

Acknowledgement

This work was supported by the Center for Nanophase Materials Sciences (CNMS), which is a US Department of Energy, Office of Science User Facility at Oak Ridge National Laboratory, and Laboratory Directed R&D (ORNL INTERSECT).

Conflicts of Interest and Declarations

The author declares no conflict of interest. Also, an archived version of the manuscript was submitted as a non-peer-reviewed preprint in an archiving portal.

Author Contributions

RCA planned the layout. RCA and JC all contributed to the overall design, literature review, revision, and writing.

Reference:

(1) *Introduction to polymers: View as single page | OpenLearn*. https://www.open.edu/openlearn/science-maths-technology/chemistry/introduction-polymers/content-section-2.3.3/?printable=1 (accessed 2026-03-11).
(2) Front Matter. In *Introduction to Physical Polymer Science*; John Wiley & Sons, Ltd, 2005; pp i–xxx. https://doi.org/10.1002/0471757128.fmatter.
(3) Silva, A. C. Q.; Silvestre, A. J. D.; Vilela, C.; Freire, C. S. R. Natural Polymers-Based Materials: A Contribution to a Greener Future. *Molecules* **2022**, *27* (1), 94. https://doi.org/10.3390/molecules27010094.

(4) Amara, A. A. A. F. Natural Polymer Types and Applications. In *Biomolecules from Natural Sources*; John Wiley & Sons, Ltd, 2022; pp 31–81. https://doi.org/10.1002/9781119769620.ch2.
(5) Yan, C.; Li, G. The Rise of Machine Learning in Polymer Discovery. *Adv. Intell. Syst.* **2023**, *5* (4), 2200243. https://doi.org/10.1002/aisy.202200243.
(6) Irimia-Vladu, M.; Sariciftci, N. S. Natural Polymers for Emerging Technological Applications: Cellulose, Lignin, Shellac and Silk. *Polym. Int.* **2025**, *74* (2), 71–86. https://doi.org/10.1002/pi.6697.
(7) Benalaya, I.; Alves, G.; Lopes, J.; Silva, L. R. A Review of Natural Polysaccharides: Sources, Characteristics, Properties, Food, and Pharmaceutical Applications. *Int. J. Mol. Sci.* **2024**, *25* (2), 1322. https://doi.org/10.3390/ijms25021322.
(8) *Deed - Attribution 4.0 International - Creative Commons*. https://creativecommons.org/licenses/by/4.0/ (accessed 2025-03-19).
(9) Sharma, S.; Bhende, M. An Overview: Non-Toxic and Eco-Friendly Polysaccharides—Its Classification, Properties, and Diverse Applications. *Polym. Bull.* **2024**, *81* (14), 12383–12429. https://doi.org/10.1007/s00289-024-05307-9.
(10) Advincula, R.; Chen, J. Advanced Manufacturing with Renewable and Bio-Based Materials: AI/ML Workflows and Process Optimization. arXiv January 15, 2026. https://doi.org/10.48550/arXiv.2601.10382.
(11) Kamran, F.; Afshar, H.; Shahi, F. Emerging Frontiers in Natural Polymers: Sustainable Innovations for Smart Agriculture and Next-Generation Food Systems. *Polym.-Plast. Technol. Mater.* **2025**, *64* (15), 2283–2332. https://doi.org/10.1080/25740881.2025.2543436.
(12) Hsu, Y.-I. Development of Functional Degradable Materials by Precise Crosslinking Design of Biobased Polymers. *Polym. J.* **2025**, *57* (10), 1095–1105. https://doi.org/10.1038/s41428-025-01051-7.
(13) Shahbazi, M.; Jäger, H. Current Status in the Utilization of Biobased Polymers for 3D Printing Process: A Systematic Review of the Materials, Processes, and Challenges. *ACS Appl. Bio Mater.* **2021**, *4* (1), 325–369. https://doi.org/10.1021/acsabm.0c01379.
(14) Xie, Y.; Gao, S.; Zhang, D.; Wang, C.; Chu, F. Bio-Based Polymeric Materials Synthesized from Renewable Resources: A Mini-Review. *Resour. Chem. Mater.* **2023**, *2* (3), 223–230. https://doi.org/10.1016/j.recm.2023.05.001.
(15) Islam, M. S.; Elahee, G. M. F.; Fang, Y.; Yu, X. (Bill); Advincula, R. C.; Cao, C. (Chase). Polylactic Acid (PLA)-Based Multifunctional and Biodegradable Nanocomposites and Their Applications. *Compos. Part B Eng.* **2025**, *306*, 112842. https://doi.org/10.1016/j.compositesb.2025.112842.
(16) Cao, P.-F.; Mangadlao, J. D.; de Leon, A.; Su, Z.; Advincula, R. C. Catenated Poly(ε-Caprolactone) and Poly(l-Lactide) via Ring-Expansion Strategy. *Macromolecules* **2015**, *48* (12), 3825–3833. https://doi.org/10.1021/acs.macromol.5b00470.
(17) Li, J.; Tian, X.; Hua, T.; Fu, J.; Koo, M.; Chan, W.; Poon, T. Chitosan Natural Polymer Material for Improving Antibacterial Properties of Textiles. *ACS Appl. Bio Mater.* **2021**, *4* (5), 4014–4038. https://doi.org/10.1021/acsabm.1c00078.
(18) Grange, C.; Aigle, A.; Ehrlich, V.; Salazar Ariza, J. F.; Brichart, T.; Da Cruz-Boisson, F.; David, L.; Lux, F.; Tillement, O. Design of a Water-Soluble Chitosan-Based Polymer with Antioxidant

and Chelating Properties for Labile Iron Extraction. *Sci. Rep.* **2023**, *13* (1), 7920. https://doi.org/10.1038/s41598-023-34251-3.

(19) Grande, C. D.; Mangadlao, J.; Fan, J.; De Leon, A.; Delgado-Ospina, J.; Rojas, J. G.; Rodrigues, D. F.; Advincula, R. Chitosan Cross-Linked Graphene Oxide Nanocomposite Films with Antimicrobial Activity for Application in Food Industry. *Macromol. Symp.* **2017**, *374* (1), 1600114. https://doi.org/10.1002/masy.201600114.

(20) Beleño Acosta, B.; Advincula, R. C.; Grande-Tovar, C. D. Chitosan-Based Scaffolds for the Treatment of Myocardial Infarction: A Systematic Review. *Molecules* **2023**, *28* (4), 1920. https://doi.org/10.3390/molecules28041920.

(21) Osti, N. C.; Remy, R. A.; Ehrhardt, D. A.; Hensley, D. K.; Chen, J.; Mamontov, E.; Advincula, R.; Nguyen, N. A. Sustainable Shape Memory Elastomers with Reduced Melt Viscosity and Enhanced Stiffness. *ACS Appl. Polym. Mater.* **2025**, *7* (4), 2554–2566. https://doi.org/10.1021/acsapm.4c03810.

(22) Osti, N. C.; Remy, R. A.; Mamontov, E.; Advincula, R.; Nguyen, N. A. Designing a Dynamic Material via Interlocking Kraft Lignin with Ultrahigh Molecular Weight Poly(Ethylene Oxide). *ACS Appl. Polym. Mater.* **2024**, *6* (19), 11877–11888. https://doi.org/10.1021/acsapm.4c01971.

(23) Kennedy, S. M.; Wilson, L. A.; M, J. A.; D, R.; Rb, J. R.; S, B. Natural Fiber Filaments Transforming the Future of Sustainable 3D Printing. *MethodsX* **2025**, *14*, 103385. https://doi.org/10.1016/j.mex.2025.103385.

(24) Greiner, A.; Wendorff, J. H. Electrospinning: A Fascinating Method for the Preparation of Ultrathin Fibers. *Angew. Chem. Int. Ed.* **2007**, *46* (30), 5670–5703. https://doi.org/10.1002/anie.200604646.

(25) Lin, K.; Wang, Z. Multiscale Mechanics and Molecular Dynamics Simulations of the Durability of Fiber-Reinforced Polymer Composites. *Commun. Mater.* **2023**, *4* (1), 66. https://doi.org/10.1038/s43246-023-00391-2.

(26) Compression and Injection Molding Techniques for Natural Fiber Composites. In *Natural Fibre Composites*; Woodhead Publishing, 2014; pp 216–232. https://doi.org/10.1533/9780857099228.2.216.

(27) da Silva, Í. G. M.; Lucas, E. F.; Advincula, R. On the Use of an Agro Waste, Miscanthus x. Giganteus, as Filtrate Reducer for Water-Based Drilling Fluids. *J. Dispers. Sci. Technol.* **2022**, *43* (5), 776–785. https://doi.org/10.1080/01932691.2020.1845195.

(28) Resende, J. F.; Paulino, I. M. R.; Bergamasco, R.; Vieira, M. F.; Vieira, A. M. S. Hydrogels Produced from Natural Polymers: A Review on Its Use and Employment in Water Treatment. *Braz. J. Chem. Eng.* **2023**, *40* (1), 23–38. https://doi.org/10.1007/s43153-022-00224-8.

(29) Dhara (Ganguly), M. Polymer Hydrogels: Classification and Recent Advances. *J. Macromol. Sci. Part A* **2024**, *61* (5), 265–288. https://doi.org/10.1080/10601325.2024.2328733.

(30) Yan, Y.-H.; Rong, L.-H.; Ge, J.; Tiu, B. D. B.; Cao, P.-F.; Advincula, R. C. Mussel-Inspired Hydrogel Composite with Multi-Stimuli Responsive Behavior. *Macromol. Mater. Eng.* **2019**, *304* (7), 1800720. https://doi.org/10.1002/mame.201800720.

(31) Tamošaitienė, J.; Parham, S.; Sarvari, H.; Chan, D. W. M.; Edwards, D. J. A Review of the Application of Synthetic and Natural Polymers as Construction and Building Materials for Achieving Sustainable Construction. *Buildings* **2024**, *14* (8), 2569. https://doi.org/10.3390/buildings14082569.

(32)Cruz, G.; Dizon, J. R. C.; Farzadnia, N.; Zhou, H.; Margarito, M.; Garcia, J. A.; Liza, F. P.; Advincula, R. C. Performance, Applications, and Sustainability of 3D-Printed Cement and Other Geomaterials. *MRS Commun.* **2023**, *13* (3), 385–399. https://doi.org/10.1557/s43579-023-00358-x.

(33)Bran Anleu, P.; Le Pape, Y.; Chen, Q.; Advincula, R.; Sun, X.-G.; Felder, J. B.; Meyer, H. M.; Post, B. K.; Lance, M. J.; Paranthaman, M. P. An Innovative Carbonated Cementitious Material and Its Printability and Carbon Mineralization Capacity. *Prog. Addit. Manuf.* **2024**, *9* (2), 435–444. https://doi.org/10.1007/s40964-023-00463-2.

(34)Clark, J. A.; Douglas, J. F. Hydration Contribution to the Solvation Free Energy of Water-Soluble Polymers. *J. Phys. Chem. B* **2025**, *129* (26), 6548–6560. https://doi.org/10.1021/acs.jpcb.5c01009.

(35)Kleemann, K.; Sander, M. Accounting for Molecular Weight Distribution Dynamics in the Environmental Fate Assessment of Water-Soluble Polymers. *CHIMIA* **2025**, *79* (4), 212–215. https://doi.org/10.2533/chimia.2025.212.

(36)Wang, D.; Zheng, Y.; Deng, Q.; Liu, X. Water-Soluble Synthetic Polymers: Their Environmental Emission Relevant Usage, Transport and Transformation, Persistence, and Toxicity. *Environ. Sci. Technol.* **2023**, *57* (16), 6387–6402. https://doi.org/10.1021/acs.est.2c09178.

(37)Jayakody, M. M.; Kaushani, K. G.; Vanniarachchy, M. P. G.; Wijesekara, I. Hydrocolloid and Water Soluble Polymers Used in the Food Industry and Their Functional Properties: A Review. *Polym. Bull.* **2023**, *80* (4), 3585–3610. https://doi.org/10.1007/s00289-022-04264-5.

(38)Kintzi, A.; Daturpalli, S.; Battagliarin, G.; Zumstein, M. Biodegradation of Water-Soluble Polymers by Wastewater Microorganisms: Challenging Laboratory Testing Protocols. *Environ. Sci. Technol.* **2024**, *58* (34), 15246–15256. https://doi.org/10.1021/acs.est.4c05808.

(39)Menzies, J.; Wilcox, A.; Casteel, K.; McDonough, K. Water Soluble Polymer Biodegradation Evaluation Using Standard and Experimental Methods. *Sci. Total Environ.* **2023**, *858*, 160006. https://doi.org/10.1016/j.scitotenv.2022.160006.

(40)Bento, C.; Katz, M.; Santos, M. M. M.; Afonso, C. A. M. Striving for Uniformity: A Review on Advances and Challenges To Achieve Uniform Polyethylene Glycol. *Org. Process Res. Dev.* **2024**, *28* (4), 860–890. https://doi.org/10.1021/acs.oprd.3c00428.

(41)Zumstein, M.; Battagliarin, G.; Kuenkel, A.; Sander, M. Environmental Biodegradation of Water-Soluble Polymers: Key Considerations and Ways Forward. *Acc. Chem. Res.* **2022**, *55* (16), 2163–2167. https://doi.org/10.1021/acs.accounts.2c00232.

(42)Alexy, P.; Káchová, D.; Kršiak, M.; Bakoš, D.; Šimková, B. Poly(Vinyl Alcohol) Stabilisation in Thermoplastic Processing. *Polym. Degrad. Stab.* **2002**, *78* (3), 413–421. https://doi.org/10.1016/S0141-3910(02)00177-5.

(43)Elgharbawy, A. S.; El Demerdash, A.-G. M.; Sadik, W. A.; Kasaby, M. A.; Lotfy, A. H.; Osman, A. I. Synthetic Degradable Polyvinyl Alcohol Polymer and Its Blends with Starch and Cellulose—A Comprehensive Overview. *Polymers* **2024**, *16* (10), 1356. https://doi.org/10.3390/polym16101356.

(44)Cai, A.; Chen, Y.; Zhang, X.; Zou, J.; Yang, J.; Mondal, A. K.; Wu, Y.; Tang, Z. Facile Strategy of Supramolecular Adhesives Based on Water-Soluble Polymers and Tannic Acid. *Polymer* **2024**, *308*, 127343. https://doi.org/10.1016/j.polymer.2024.127343.

(45)Vu, N. Q.; Le, T. M.; Ngo, A. N. B.; Nguyen, M. H. T.; Liao, Y.-C.; Tran, T. T. Engineering Functional PVA: A Comprehensive Review of Chemical Modifications and Prospective

Developments. *ACS Polym. Au* **2026**, *6* (1), 33–60. https://doi.org/10.1021/acspolymersau.5c00133.

(46) Elgharbawy, A. S.; El Demerdash, A.-G. M.; Sadik, W. A.; Kasaby, M. A.; Lotfy, A. H.; Osman, A. I. Enhancing the Biodegradability, Water Solubility, and Thermal Properties of Polyvinyl Alcohol through Natural Polymer Blending: An Approach toward Sustainable Polymer Applications. *Polymers* **2024**, *16* (15), 2141. https://doi.org/10.3390/polym16152141.

(47) Mallakpour, S.; Tabesh, F.; Hussain, C. M. A New Trend of Using Poly(Vinyl Alcohol) in 3D and 4D Printing Technologies: Process and Applications. *Adv. Colloid Interface Sci.* **2022**, *301*, 102605. https://doi.org/10.1016/j.cis.2022.102605.

(48) Couți, N.; Porfire, A.; Iovanov, R.; Crișan, A. G.; Iurian, S.; Casian, T.; Tomuță, I. Polyvinyl Alcohol, a Versatile Excipient for Pharmaceutical 3D Printing. *Polymers* **2024**, *16* (4), 517. https://doi.org/10.3390/polym16040517.

(49) Wilts, E. M.; Long, T. E. Sustainable Additive Manufacturing: Predicting Binder Jettability of Water-Soluble, Biodegradable and Recyclable Polymers. *Polym. Int.* **2021**, *70* (7), 958–963. https://doi.org/10.1002/pi.6108.

(50) Finster, R.; Sankaran, P.; Bihar, E. Computational and AI-Driven Design of Hydrogels for Bioelectronic Applications. *Adv. Electron. Mater.* **2025**, *11* (17), 2400763. https://doi.org/10.1002/aelm.202400763.

(51) McBride, R. J.; Miller, J. F.; Blanazs, A.; Hähnle, H.-J.; Armes, S. P. Synthesis of High Molecular Weight Water-Soluble Polymers as Low-Viscosity Latex Particles by RAFT Aqueous Dispersion Polymerization in Highly Salty Media. *Macromolecules* **2022**, *55* (17), 7380–7391. https://doi.org/10.1021/acs.macromol.2c01071.

(52) McBride, R. J.; Geneste, E.; Xie, A.; Ryan, A. J.; Miller, J. F.; Blanazs, A.; Rösch, C.; Armes, S. P. Low-Viscosity Route to High-Molecular-Weight Water-Soluble Polymers: Exploiting the Salt Sensitivity of Poly(N-Acryloylmorpholine). *Macromolecules* **2024**, *57* (5), 2432–2445. https://doi.org/10.1021/acs.macromol.3c02616.

(53) Azhdari, S.; Gröschel, A. H.; Wurm, F. R. Biodegradable and Water-Soluble Polymers in Block Copolymer Architectures. *Polym. Degrad. Stab.* **2026**, *243*, 111749. https://doi.org/10.1016/j.polymdegradstab.2025.111749.

(54) Quandt, J.; Gumerov, R. A.; Kratzenberg, T.; Hohenschutz, M.; Kulczycki, D.; Richtering, W.; Potemkin, I. I.; Rodriguez-Emmenegger, C. Synthesis of Water-Soluble, Highly Branched Arborescent Poly(Acrylate)s: A Colloid-Macromolecule Chimera. *Polym. Chem.* **2025**, *16* (17), 2007–2022. https://doi.org/10.1039/D5PY00104H.

(55) Berninger, T.; Dietz, N.; González López, Ó. Water-Soluble Polymers in Agriculture: Xanthan Gum as Eco-Friendly Alternative to Synthetics. *Microb. Biotechnol.* **2021**, *14* (5), 1881–1896. https://doi.org/10.1111/1751-7915.13867.

(56) Mirzaie Yegane, M.; Boukany, P. E.; Zitha, P. Fundamentals and Recent Progress in the Flow of Water-Soluble Polymers in Porous Media for Enhanced Oil Recovery. *Energies* **2022**, *15* (22), 8575. https://doi.org/10.3390/en15228575.

(57) Yihang, Z. Application of Water-Soluble Polymer Inhibitor in Metal Corrosion Protection: Progress and Challenges. *Front. Energy Res.* **2022**, *10*. https://doi.org/10.3389/fenrg.2022.997107.

(58)Delgado, M. Z.; Aranda, F. L.; Hernandez-Tenorio, F.; Garrido-Miranda, K. A.; Meléndrez, M. F.; Palacio, D. A. Polyelectrolytes for Environmental, Agricultural, and Medical Applications. *Polymers* **2024**, *16* (10), 1434. https://doi.org/10.3390/polym16101434.

(59)Durmaz, E. N.; Sahin, S.; Virga, E.; de Beer, S.; de Smet, L. C. P. M.; de Vos, W. M. Polyelectrolytes as Building Blocks for Next-Generation Membranes with Advanced Functionalities. *ACS Appl. Polym. Mater.* **2021**, *3* (9), 4347–4374. https://doi.org/10.1021/acsapm.1c00654.

(60)Xu, W.; Ledin, P. A.; Shevchenko, V. V.; Tsukruk, V. V. Architecture, Assembly, and Emerging Applications of Branched Functional Polyelectrolytes and Poly(Ionic Liquid)s. *ACS Appl. Mater. Interfaces* **2015**, *7* (23), 12570–12596. https://doi.org/10.1021/acsami.5b01833.

(61)Dobrynin, A. V.; Rubinstein, M. Theory of Polyelectrolytes in Solutions and at Surfaces. *Prog. Polym. Sci.* **2005**, *30* (11), 1049–1118. https://doi.org/10.1016/j.progpolymsci.2005.07.006.

(62)Ting, J. M.; Wu, H.; Herzog-Arbeitman, A.; Srivastava, S.; Tirrell, M. V. Synthesis and Assembly of Designer Styrenic Diblock Polyelectrolytes. *ACS Macro Lett.* **2018**, *7* (6), 726–733. https://doi.org/10.1021/acsmacrolett.8b00346.

(63)Park, M.-K.; Advincula, R. C. The Layer-by-Layer Assemblies of Polyelectrolytes and Nanomaterials as Films and Particle Coatings. In *Functional Polymer Films*; John Wiley & Sons, Ltd, 2011; pp 73–112. https://doi.org/10.1002/9783527638482.ch3.

(64)Petrila, L.-M.; Bucatariu, F.; Mihai, M.; Teodosiu, C. Polyelectrolyte Multilayers: An Overview on Fabrication, Properties, and Biomedical and Environmental Applications. *Materials* **2021**, *14* (15), 4152. https://doi.org/10.3390/ma14154152.

(65)Laschewsky, A. Structures and Synthesis of Zwitterionic Polymers. *Polymers* **2014**, *6* (5), 1544–1601. https://doi.org/10.3390/polym6051544.

(66)*Deed - Attribution 3.0 Unported - Creative Commons*. https://creativecommons.org/licenses/by/3.0/ (accessed 2026-01-15).

(67)Morshed, M. Study of the Separation by Organic Solvent Nanofiltration of Diluted Solutes Using Commercial, Dense and Porous Membranes and Their Derivatives by Deposition of Polyelectrolyte Nanolayers. phdthesis, Université de Lorraine, 2019. https://hal.univ-lorraine.fr/tel-02351825 (accessed 2026-02-17).

(68)Muthukumar, M. 50th Anniversary Perspective: A Perspective on Polyelectrolyte Solutions. *Macromolecules* **2017**, *50* (24), 9528–9560. https://doi.org/10.1021/acs.macromol.7b01929.

(69)Lytle, T. K.; Chang, L.-W.; Markiewicz, N.; Perry, S. L.; Sing, C. E. Designing Electrostatic Interactions via Polyelectrolyte Monomer Sequence. *ACS Cent. Sci.* **2019**, *5* (4), 709–718. https://doi.org/10.1021/acscentsci.9b00087.

(70)Reznik, C.; Estillore, N.; Advincula, R. C.; Landes, C. F. Single Molecule Spectroscopy Reveals Heterogeneous Transport Mechanisms for Molecular Ions in a Polyelectrolyte Polymer Brush. *J. Phys. Chem. B* **2009**, *113* (44), 14611–14618. https://doi.org/10.1021/jp906487j.

(71)Reznik, C.; Darugar, Q.; Wheat, A.; Fulghum, T.; Advincula, R. C.; Landes, C. F. Single Ion Diffusive Transport within a Poly(Styrene Sulfonate) Polymer Brush Matrix Probed by Fluorescence Correlation Spectroscopy. *J. Phys. Chem. B* **2008**, *112* (35), 10890–10897. https://doi.org/10.1021/jp803718p.

(72)Angelopoulou, P. P.; Kearney, L. T.; Keum, J. K.; Collins, L.; Kumar, R.; Sakellariou, G.; Advincula, R. C.; Mays, J. W.; Hong, K. High-χ Diblock Copolymers Containing

Poly(Vinylpyridine-N-Oxide) Segments. *J. Mater. Chem. A* **2023**, *11* (18), 9846–9858. https://doi.org/10.1039/D3TA01386C.

(73)Ji, H.; Sakellariou, G.; Advincula, R. C.; Smith, G. D.; Kilbey II, S. M.; Dadmun, M. D.; Mays, J. W. Synthesis and Characterization of Well-Defined [Polystyrene-b-Poly(2-Vinylpyridine)]n Star-Block Copolymers with Poly(2-Vinylpyridine) Corona Blocks. *J. Polym. Sci. Part Polym. Chem.* **2007**, *45* (17), 3949–3955. https://doi.org/10.1002/pola.22148.

(74)Obeid, R.; Park, J.-Y.; Advincula, R. C.; Winnik, F. M. Temperature-Dependent Interfacial Properties of Hydrophobically End-Modified Poly(2-Isopropyl-2-Oxazoline)s Assemblies at the Air/Water Interface and on Solid Substrates. *J. Colloid Interface Sci.* **2009**, *340* (2), 142–152. https://doi.org/10.1016/j.jcis.2009.06.060.

(75)Ju, H.; Yin, Z.; Demchuk, Z.; Bocharova, V.; Gainaru, C.; Laub, J. A.; Vogiatzis, K.; Advincula, R.; Chen, J.; Cao, P.-F. Constructing Crown Ether-Based Supramolecular Adhesives with Ambient-Temperature Applicable and Durable Adhesion. *Adv. Funct. Mater.* **2024**, *34* (37), 2402165. https://doi.org/10.1002/adfm.202402165.

(76)Xu, Q.; Tao, S.; Jiang, Q.; Jiang, D. Ion Conduction in Polyelectrolyte Covalent Organic Frameworks. *J. Am. Chem. Soc.* **2018**, *140* (24), 7429–7432. https://doi.org/10.1021/jacs.8b03814.

(77)Jin, S.; Shan, X.; Tian, J.; Li, Y.; Ding, H.; Advincula, R.; Tian, M.; Cao, P.-F. High-Entropy Gel Polymer Electrolyte for Wide-Temperature Operatable Li-Metal Batteries. *Adv. Funct. Mater.* **2025**, *35* (45), 2500440. https://doi.org/10.1002/adfm.202500440.

(78)Christakopoulos, P.; Wang, H.; Bonnesen, P. V.; Keum, J. K.; Wang, Y.; Hong, K.; Lokitz, B. S.; Doucet, M.; Browning, J. F.; Advincula, R. C.; Kumar, R. Characterizing Hygroscopic Films of Polyzwitterions in Electric Fields Using Neutron and X-Ray Reflectometries: Electrostriction or Mass Loss? *ACS Appl. Mater. Interfaces* **2024**, *16* (36), 48307–48319. https://doi.org/10.1021/acsami.4c09938.

(79)Liu, Z.; Keum, J. K.; Li, T.; Chen, J.; Hong, K.; Wang, Y.; Sumpter, B. G.; Advincula, R.; Kumar, R. Anti-Polyelectrolyte and Polyelectrolyte Effects on Conformations of Polyzwitterionic Chains in Dilute Aqueous Solutions. *PNAS Nexus* **2023**, *2* (7), pgad204. https://doi.org/10.1093/pnasnexus/pgad204.

(80)Youk, J. H.; Locklin, J.; Xia, C.; Park, M.-K.; Advincula, R. Preparation of Gold Nanoparticles from a Polyelectrolyte Complex Solution of Terthiophene Amphiphiles. *Langmuir* **2001**, *17* (15), 4681–4683. https://doi.org/10.1021/la010438s.

(81)Park, M.-K.; Sakellariou, G.; Pispas, S.; Hadjichristidis, N.; Advincula, R. On the Quantitative Adsorption Behavior of Multi-Zwitterionic End-Functionalized Polymers onto Gold Surfaces. *Colloids Surf. Physicochem. Eng. Asp.* **2008**, *326* (3), 115–121. https://doi.org/10.1016/j.colsurfa.2008.05.034.

(82)Ayoub, A.; Lucia, L. A. Sustainable Cationic Polyelectrolytes from Agri-Forestry Biomass: Conventional Chemistry to AI-Optimized Reactive Extrusion. *Sustainability* **2025**, *17* (22), 10060. https://doi.org/10.3390/su172210060.

(83)Fu, X.; Zhang, Y.; Jia, X.; Wang, Y.; Chen, T. Research Progress on Typical Quaternary Ammonium Salt Polymers. *Molecules* **2022**, *27* (4), 1267. https://doi.org/10.3390/molecules27041267.

(84)Ajjarapu, V. M. K. Enhancing Industrial and Municipal Water Treatment Efficiency Using Anionic and Cationic Polyacrylamide Polymers. *Am. J. Polym. Sci.* **2025**, *14* (1), 7–16.

(85)Tiu, B. D. B.; Advincula, R. C. Polymeric Corrosion Inhibitors for the Oil and Gas Industry: Design Principles and Mechanism. *React. Funct. Polym.* **2015**, *95*, 25–45. https://doi.org/10.1016/j.reactfunctpolym.2015.08.006.

(86)Fisher, R. S.; Liao, J.; Ahn, S. Y.; Modi, N.; Kidane, A. K.; Obermeyer, A. C. Engineering Protein–Polyelectrolyte Interactions for Cellular Applications. *Annu. Rev. Chem. Biomol. Eng.* **2025**, *16* (Volume 16, 2025), 119–145. https://doi.org/10.1146/annurev-chembioeng-100722-105929.

(87)Daniels, C. R.; Reznik, C.; Kilmer, R.; Felipe, M. J.; Tria, M. C. R.; Kourentzi, K.; Chen, W.-H.; Advincula, R. C.; Willson, R. C.; Landes, C. F. Permeability of Anti-Fouling PEGylated Surfaces Probed by Fluorescence Correlation Spectroscopy. *Colloids Surf. B Biointerfaces* **2011**, *88* (1), 31–38. https://doi.org/10.1016/j.colsurfb.2011.05.044.

(88)Li, J.; Li, L.; Brink, H. A.; Allegri, G.; Lindhoud, S. Polyelectrolyte Complex-Based Materials for Separations: Progress, Challenges and Opportunities. *Mater. Horiz.* **2025**, *12* (14), 4998–5030. https://doi.org/10.1039/D4MH01840K.

(89)Li, L.; Rumyantsev, A. M.; Srivastava, S.; Meng, S.; de Pablo, J. J.; Tirrell, M. V. Effect of Solvent Quality on the Phase Behavior of Polyelectrolyte Complexes. *Macromolecules* **2021**, *54* (1), 105–114. https://doi.org/10.1021/acs.macromol.0c01000.

(90)Schaaf, P.; Schlenoff, J. B. Saloplastics: Processing Compact Polyelectrolyte Complexes. *Adv. Mater.* **2015**, *27* (15), 2420–2432. https://doi.org/10.1002/adma.201500176.

(91)Keum, J. K.; Christakopoulos, P.; Liu, Z.; Li, T.; Chen, J.; Williams, A.; Hensley, D. K.; Hong, K.; Wang, Y.; Advincula, R.; Kumar, R. Chemical Specificity in Polyzwitterion-Polyelectrolyte Coacervates: Polycations vs Polyanions. *Eur. Polym. J.* **2024**, *215*, 113177. https://doi.org/10.1016/j.eurpolymj.2024.113177.

(92)Putro, J. N.; Lunardi, V. B.; Soetaredjo, F. E.; Yuliana, M.; Santoso, S. P.; Wenten, I. G.; Ismadji, S. A Review of Gum Hydrocolloid Polyelectrolyte Complexes (PEC) for Biomedical Applications: Their Properties and Drug Delivery Studies. *Processes* **2021**, *9* (10), 1796. https://doi.org/10.3390/pr9101796.

(93)Bediako, J. K.; Mouele, E. S. M.; El Ouardi, Y.; Repo, E. Saloplastics and the Polyelectrolyte Complex Continuum: Advances, Challenges and Prospects. *Chem. Eng. J.* **2023**, *462*, 142322. https://doi.org/10.1016/j.cej.2023.142322.

(94)Sukhishvili, S. A.; Kharlampieva, E.; Izumrudov, V. Where Polyelectrolyte Multilayers and Polyelectrolyte Complexes Meet. *Macromolecules* **2006**, *39* (26), 8873–8881. https://doi.org/10.1021/ma061617p.

(95)Sciortino, F.; Mir, S. H.; Pakdel, A.; Oruganti, A.; Abe, H.; Witecka, A.; Shri, D. N. A.; Rydzek, G.; Ariga, K. Saloplastics as Multiresponsive Ion Exchange Reservoirs and Catalyst Supports. *J. Mater. Chem. A* **2020**, *8* (34), 17713–17724. https://doi.org/10.1039/D0TA05901C.

(96)Reurink, D. M.; Willott, J. D.; Roesink, H. D. W.; de Vos, W. M. Role of Polycation and Cross-Linking in Polyelectrolyte Multilayer Membranes. *ACS Appl. Polym. Mater.* **2020**, *2* (11), 5278–5289. https://doi.org/10.1021/acsapm.0c00992.

(97)Park, M.-K.; Deng, S.; Advincula, R. C. Sustained Release Control via Photo-Cross-Linking of Polyelectrolyte Layer-by-Layer Hollow Capsules. *Langmuir* **2005**, *21* (12), 5272–5277. https://doi.org/10.1021/la047008x.

(98)Ghostine, R. A.; Markarian, M. Z.; Schlenoff, J. B. Asymmetric Growth in Polyelectrolyte Multilayers. *J. Am. Chem. Soc.* **2013**, *135* (20), 7636–7646. https://doi.org/10.1021/ja401318m.

(99)Müller, M.; Meier-Haack, J.; Schwarz, S.; Buchhammer, H. M.; Eichhorn, K.-J.; Janke, A.; Keßler, B.; Nagel, J.; Oelmann, M.; Reihs, T.; Lunkwitz, K. Polyelectrolyte Multilayers and Their Interactions. *J. Adhes.* **2004**, *80* (6), 521–547. https://doi.org/10.1080/00218460490477189.

(100) Séon, L.; Lavalle, P.; Schaaf, P.; Boulmedais, F. Polyelectrolyte Multilayers: A Versatile Tool for Preparing Antimicrobial Coatings. *Langmuir* **2015**, *31* (47), 12856–12872. https://doi.org/10.1021/acs.langmuir.5b02768.

(101) Boddohi, S.; Killingsworth, C. E.; Kipper, M. J. Polyelectrolyte Multilayer Assembly as a Function of pH and Ionic Strength Using the Polysaccharides Chitosan and Heparin. *Biomacromolecules* **2008**, *9* (7), 2021–2028. https://doi.org/10.1021/bm8002573.

(102) *Deed - Attribution-NonCommercial-NoDerivatives 4.0 International - Creative Commons*. https://creativecommons.org/licenses/by-nc-nd/4.0/ (accessed 2025-10-04).

(103) Baba, A.; Kaneko, F.; Advincula, R. C. Polyelectrolyte Adsorption Processes Characterized in Situ Using the Quartz Crystal Microbalance Technique: Alternate Adsorption Properties in Ultrathin Polymer Films. *Colloids Surf. Physicochem. Eng. Asp.* **2000**, *173* (1), 39–49. https://doi.org/10.1016/S0927-7757(00)00579-3.

(104) Advincula, R.; Aust, E.; Meyer, W.; Knoll, W. In Situ Investigations of Polymer Self-Assembly Solution Adsorption by Surface Plasmon Spectroscopy. *Langmuir* **1996**, *12* (15), 3536–3540. https://doi.org/10.1021/la9601622.

(105) Tauzin, L. J.; Shuang, B.; Kisley, L.; Mansur, A. P.; Chen, J.; de Leon, A.; Advincula, R. C.; Landes, C. F. Charge-Dependent Transport Switching of Single Molecular Ions in a Weak Polyelectrolyte Multilayer. *Langmuir* **2014**, *30* (28), 8391–8399. https://doi.org/10.1021/la5012007.

(106) Park, M.-K.; Deng, S.; Advincula, R. C. pH-Sensitive Bipolar Ion-Permselective Ultrathin Films. *J. Am. Chem. Soc.* **2004**, *126* (42), 13723–13731. https://doi.org/10.1021/ja0484707.

(107) Park, M.-K.; Advincula, R. C. In-Plane Photoalignment of Liquid Crystals by Azobenzene−Polyelectrolyte Layer-by-Layer Ultrathin Films. *Langmuir* **2002**, *18* (11), 4532–4535. https://doi.org/10.1021/la015771a.

(108) Advincula, R. C.; Fells, E.; Park, M. Molecularly Ordered Low Molecular Weight Azobenzene Dyes and Polycation Alternate Multilayer Films: Aggregation, Layer Order, and Photoalignment. *Chem. Mater.* **2001**, *13* (9), 2870–2878. https://doi.org/10.1021/cm0101172.

(109) Dante, S.; Advincula, R.; Frank, C. W.; Stroeve, P. Photoisomerization of Polyionic Layer-by-Layer Films Containing Azobenzene. *Langmuir* **1999**, *15* (1), 193–201. https://doi.org/10.1021/la980497e.

(110) Kato, K.; Kawashima, J.; Baba, A.; Shinbo, K.; Kaneko, F.; Advincula, R. C. Attenuated Total Reflection and Emission Due to Surface Plasmon Excitation of Layer-by-Layer Ultrathin Films Containing Azo-Dye. *Thin Solid Films* **2003**, *438–439*, 101–107. https://doi.org/10.1016/S0040-6090(03)00780-6.

(111) Shinbo, K.; Baba, A.; Kaneko, F.; Kato, T.; Kato, K.; Advincula, R. C.; Knoll, W. In Situ Investigations on the Preparations of Layer-by-Layer Films Containing Azobenzene and

Applications for LC Display Devices. *Mater. Sci. Eng. C* **2002**, *22* (2), 319–325. https://doi.org/10.1016/S0928-4931(02)00214-X.

(112) Shinbo, K.; Ishikawa, J.; Baba, A.; Kaneko, F.; Kato, K.; Advincula, R. C. Alignments of Nematic Liquid Crystal Molecules on Azo-Dye-Containing Alternate Self-Assembled Films Investigated Using Attenuated Total Reflection Method. *Jpn. J. Appl. Phys.* **2002**, *41* (4S), 2753. https://doi.org/10.1143/JJAP.41.2753.

(113) Ishikawa, J.; Baba, A.; Kaneko, F.; Shinbo, K.; Kato, K.; Advincula, R. C. Photo-Induced in-Plane Alignment of LC Molecules on Layer-by-Layer Self-Assembled Films Containing Azo Dyes Evaluated by Attenuated Total Reflection Measurements. *Colloids Surf. Physicochem. Eng. Asp.* **2002**, *198–200*, 917–922. https://doi.org/10.1016/S0927-7757(01)01020-2.

(114) Tiu, B. D. B.; Kernan, D. L.; Tiu, S. B.; Wen, A. M.; Zheng, Y.; Pokorski, J. K.; Advincula, R. C.; Steinmetz, N. F. Electrostatic Layer-by-Layer Construction of Fibrous TMV Biofilms. *Nanoscale* **2017**, *9* (4), 1580–1590. https://doi.org/10.1039/C6NR06266K.

(115) de Leon, A.; Barnes, A. C.; Thomas, P.; O'Donnell, J.; Zorman, C. A.; Advincula, R. C. Transfer Printing of Self-Folding Polymer–Metal Bilayer Particles. *ACS Appl. Mater. Interfaces* **2014**, *6* (24), 22695–22700. https://doi.org/10.1021/am5068172.

(116) Frau, A. F.; Lane, T. J.; Schlather, A. E.; Park, J. Y.; Advincula, R. C. Modulating Electrochemical Activity in Polyaniline/Titanium Oxide Hybrid Nanostructured Ultrathin Films. *Ind. Eng. Chem. Res.* **2011**, *50* (9), 5532–5542. https://doi.org/10.1021/ie101805a.

(117) Estillore, N.; Advincula, R. C. Surface-Initiated Polymerization and Layer-by-Layer Films. In *Multilayer Thin Films*; John Wiley & Sons, Ltd, 2012; pp 437–454. https://doi.org/10.1002/9783527646746.ch18.

(118) Estillore, N. C.; Advincula, R. C. Free-Standing Films of Semifluorinated Block Copolymer Brushes from Layer-by-Layer Polyelectrolyte Macroinitiators. *Macromol. Chem. Phys.* **2011**, *212* (15), 1552–1566. https://doi.org/10.1002/macp.201100066.

(119) Estillore, N. C.; Advincula, R. C. Stimuli-Responsive Binary Mixed Polymer Brushes and Free-Standing Films by LbL-SIP. *Langmuir* **2011**, *27* (10), 5997–6008. https://doi.org/10.1021/la200089x.

(120) Estillore, N. C.; Park, J. Y.; Advincula, R. C. Langmuir−Schaefer (LS) Macroinitiator Film Control on the Grafting of a Thermosensitive Polymer Brush via Surface Initiated-ATRP. *Macromolecules* **2010**, *43* (16), 6588–6598. https://doi.org/10.1021/ma100726z.

(121) Fulghum, T. M.; Estillore, N. C.; Vo, C.-D.; Armes, S. P.; Advincula, R. C. Stimuli-Responsive Polymer Ultrathin Films with a Binary Architecture: Combined Layer-by-Layer Polyelectrolyte and Surface-Initiated Polymerization Approach. *Macromolecules* **2008**, *41* (2), 429–435. https://doi.org/10.1021/ma0717136.

(122) Waenkaew, P.; Phanichphant, S.; Advincula, R. C. Electropolymerization of Layer-by-Layer Precursor Polymer Films. *Polym. Adv. Technol.* **2011**, *22* (5), 753–758. https://doi.org/10.1002/pat.1886.

(123) Huang, C.; Jiang, G.; Advincula, R. Electrochemical Cross-Linking and Patterning of Nanostructured Polyelectrolyte−Carbazole Precursor Ultrathin Films. *Macromolecules* **2008**, *41* (13), 4661–4670. https://doi.org/10.1021/ma800268n.

(124) Baba, A.; Jiang, G.; Park, K.-M.; Park, J.-Y.; Shin, H.-K.; Advincula, R. Electro-Nanopatterning of Surface Relief Gratings on Azobenzene Layer-by-Layer Ultrathin

Films by Current-Sensing Atomic Force Microscopy. *J. Phys. Chem. B* **2006**, *110* (35), 17309–17314. https://doi.org/10.1021/jp063955y.

(125) Jiang, G.; Baba, A.; Ikarashi, H.; Xu, R.; Locklin, J.; Kashif, K. R.; Shinbo, K.; Kato, K.; Kaneko, F.; Advincula, R. Signal Enhancement and Tuning of Surface Plasmon Resonance in Au Nanoparticle/Polyelectrolyte Ultrathin Films. *J. Phys. Chem. C* **2007**, *111* (50), 18687–18694. https://doi.org/10.1021/jp075986e.

(126) Baba, A.; Tian, S.; Stefani, F.; Xia, C.; Wang, Z.; Advincula, R. C.; Johannsmann, D.; Knoll, W. Electropolymerization and Doping/Dedoping Properties of Polyaniline Thin Films as Studied by Electrochemical-Surface Plasmon Spectroscopy and by the Quartz Crystal Microbalance. *J. Electroanal. Chem.* **2004**, *562* (1), 95–103. https://doi.org/10.1016/j.jelechem.2003.08.012.

(127) Baba, A.; Park, M.-K.; Advincula, R. C.; Knoll, W. Simultaneous Surface Plasmon Optical and Electrochemical Investigation of Layer-by-Layer Self-Assembled Conducting Ultrathin Polymer Films. *Langmuir* **2002**, *18* (12), 4648–4652. https://doi.org/10.1021/la011078b.

(128) Tian, S.; Baba, A.; Liu, J.; Wang, Z.; Knoll, W.; Park, M.-K.; Advincula, R. Electroactivity of Polyaniline Multilayer Films in Neutral Solution and Their Electrocatalyzed Oxidation of β-Nicotinamide Adenine Dinucleotide. *Adv. Funct. Mater.* **2003**, *13* (6), 473–479. https://doi.org/10.1002/adfm.200304320.

(129) Jiang, G.; Baba, A.; Advincula, R. Nanopatterning and Fabrication of Memory Devices from Layer-by-Layer Poly(3,4-Ethylenedioxythiophene)−Poly(Styrene Sulfonate) Ultrathin Films. *Langmuir* **2007**, *23* (2), 817–825. https://doi.org/10.1021/la061817u.

(130) Baba, A.; Knoll, W.; Advincula, R. Simultaneous in Situ Electrochemical, Surface Plasmon Optical, and Atomic Force Microscopy Measurements: Investigation of Conjugated Polymer Electropolymerization. *Rev. Sci. Instrum.* **2006**, *77* (6), 064101. https://doi.org/10.1063/1.2204587.

(131) Park, M.-K.; Onishi, K.; Locklin, J.; Caruso, F.; Advincula, R. C. Self-Assembly and Characterization of Polyaniline and Sulfonated Polystyrene Multilayer-Coated Colloidal Particles and Hollow Shells. *Langmuir* **2003**, *19* (20), 8550–8554. https://doi.org/10.1021/la034827t.

(132) Park, M.-K.; Xia, C.; Advincula, R. C.; Schütz, P.; Caruso, F. Cross-Linked, Luminescent Spherical Colloidal and Hollow-Shell Particles. *Langmuir* **2001**, *17* (24), 7670–7674. https://doi.org/10.1021/la011006k.

(133) Fulghum, T. M.; Patton, D. L.; Advincula, R. C. Fuzzy Ternary Particle Systems by Surface-Initiated Atom Transfer Radical Polymerization from Layer-by-Layer Colloidal Core−Shell Macroinitiator Particles. *Langmuir* **2006**, *22* (20), 8397–8402. https://doi.org/10.1021/la0601509.

(134) Aggour, K. S.; Gupta, V. K.; Ruscitto, D.; Ajdelsztajn, L.; Bian, X.; Brosnan, K. H.; Chennimalai Kumar, N.; Dheeradhada, V.; Hanlon, T.; Iyer, N.; Karandikar, J.; Li, P.; Moitra, A.; Reimann, J.; Robinson, D. M.; Santamaria-Pang, A.; Shen, C.; Soare, M. A.; Sun, C.; Suzuki, A.; Venkataramana, R.; Vinciquerra, J. Artificial Intelligence/Machine Learning in Manufacturing and Inspection: A GE Perspective. *MRS Bull.* **2019**, *44* (7), 545–558. https://doi.org/10.1557/mrs.2019.157.

(135) Adir, O.; Poley, M.; Chen, G.; Froim, S.; Krinsky, N.; Shklover, J.; Shainsky-Roitman, J.; Lammers, T.; Schroeder, A. Integrating Artificial Intelligence and Nanotechnology for

Precision Cancer Medicine. *Adv. Mater.* **2020**, *32* (13), 1901989. https://doi.org/10.1002/adma.201901989.

(136) Chou, W.-C.; Canchola, A.; Zhang, F.; Lin, Z. Machine Learning and Artificial Intelligence in Nanomedicine. *WIREs Nanomedicine Nanobiotechnology* **2025**, *17* (4), e70027. https://doi.org/10.1002/wnan.70027.

(137) Handoko, A. D.; Made, R. I. Artificial Intelligence and Generative Models for Materials Discovery -- A Review. arXiv August 5, 2025. https://doi.org/10.48550/arXiv.2508.03278.

(138) Choudhary, K.; Kelley, M. L. ChemNLP: A Natural Language-Processing-Based Library for Materials Chemistry Text Data. *J. Phys. Chem. C* **2023**, *127* (35), 17545–17555. https://doi.org/10.1021/acs.jpcc.3c03106.

(139) Karanikolas, N. N.; Manga, E.; Samaridi, N.; Stergiopoulos, V.; Tousidou, E.; Vassilakopoulos, M. Strengths and Weaknesses of LLM-Based and Rule-Based NLP Technologies and Their Potential Synergies. *Electronics* **2025**, *14* (15), 3064. https://doi.org/10.3390/electronics14153064.

(140) Badillo, S.; Banfai, B.; Birzele, F.; Davydov, I. I.; Hutchinson, L.; Kam-Thong, T.; Siebourg-Polster, J.; Steiert, B.; Zhang, J. D. An Introduction to Machine Learning. *Clin. Pharmacol. Ther.* **2020**, *107* (4), 871–885. https://doi.org/10.1002/cpt.1796.

(141) Chen, J.; Yuan, Y.; Ziabari, A. K.; Xu, X.; Zhang, H.; Christakopoulos, P.; Bonnesen, P. V.; Ivanov, I. N.; Ganesh, P.; Wang, C.; Jaimes, K. P.; Yang, G.; Kumar, R.; Sumpter, B. G.; Advincula, R. AI for Manufacturing and Healthcare: A Chemistry and Engineering Perspective. arXiv May 2, 2024. https://doi.org/10.48550/arXiv.2405.01520.

(142) Yang, J.; Hu, X.; Feng, L.; Liu, Z.; Murtazt, A.; Qin, W.; Zhou, M.; Liu, J.; Bi, Y.; Qian, J.; Zhang, W. AI-Enabled Portable E-Nose Regression Predicting Harmful Molecules in a Gas Mixture. *ACS Sens.* **2024**, *9* (6), 2925–2934. https://doi.org/10.1021/acssensors.4c00050.

(143) Scheichl, R.; Klopffer, M.-H.; Benjelloun-Dabaghi, Z.; Flaconnèche, B. Permeation of Gases in Polymers: Parameter Identification and Nonlinear Regression Analysis. *J. Membr. Sci.* **2005**, *254* (1), 275–293. https://doi.org/10.1016/j.memsci.2005.01.019.

(144) Andrews, H. B.; Sadergaski, L. R.; Cary, S. K. Pursuit of the Ultimate Regression Model for Samarium(III), Europium(III), and LiCl Using Laser-Induced Fluorescence, Design of Experiments, and a Genetic Algorithm for Feature Selection. *ACS Omega* **2023**, *8* (2), 2281–2290. https://doi.org/10.1021/acsomega.2c06610.

(145) Ziatdinov, M.; Liu, Y.; Kelley, K.; Vasudevan, R.; Kalinin, S. V. Bayesian Active Learning for Scanning Probe Microscopy: From Gaussian Processes to Hypothesis Learning. *ACS Nano* **2022**, *16* (9), 13492–13512. https://doi.org/10.1021/acsnano.2c05303.

(146) Raissi, M.; Perdikaris, P.; Karniadakis, G. E. Physics-Informed Neural Networks: A Deep Learning Framework for Solving Forward and Inverse Problems Involving Nonlinear Partial Differential Equations. *J. Comput. Phys.* **2019**, *378*, 686–707. https://doi.org/10.1016/j.jcp.2018.10.045.

(147) Spining, M. T.; Darsey, J. A.; Sumpter, B. G.; Nold, D. W. Opening Up the Black Box of Artificial Neural Networks. *J. Chem. Educ.* **1994**, *71* (5), 406. https://doi.org/10.1021/ed071p406.

(148) Sumpter, B. G.; Noid, D. W. Neural Networks and Graph Theory as Computational Tools for Predicting Polymer Properties. *Macromol. Theory Simul.* **1994**, *3* (2), 363–378. https://doi.org/10.1002/mats.1994.040030207.

(149) Jolliffe, I. T.; Cadima, J. Principal Component Analysis: A Review and Recent Developments. *Philos. Transact. A Math. Phys. Eng. Sci.* **2016**, *374* (2065), 20150202. https://doi.org/10.1098/rsta.2015.0202.

(150) Chiong, K. X.; Shum, M. Random Projection Estimation of Discrete-Choice Models with Large Choice Sets. *Manag. Sci.* **2019**, *65* (1), 256–271. https://doi.org/10.1287/mnsc.2017.2928.

(151) Hout, M. C.; Papesh, M. H.; Goldinger, S. D. Multidimensional Scaling. *WIREs Cogn. Sci.* **2013**, *4* (1), 93–103. https://doi.org/10.1002/wcs.1203.

(152) Chen, J.; Christakopoulos, P.; Chen, K. D.; Ivanov, I. N.; Advincula, R. Agentic AI and Machine Learning for Accelerated Materials Discovery and Applications. arXiv January 15, 2026. https://doi.org/10.48550/arXiv.2601.09027.

(153) Inizan, T. J.; Yang, S.; Kaplan, A.; Lin, Y.; Yin, J.; Mirzaei, S.; Abdelgaid, M.; Alawadhi, A. H.; Cho, K.; Zheng, Z.; Cubuk, E. D.; Borgs, C.; Chayes, J. T.; Persson, K. A.; Yaghi, O. M. System of Agentic AI for the Discovery of Metal-Organic Frameworks. arXiv April 18, 2025. https://doi.org/10.48550/arXiv.2504.14110.

(154) Peng, J.; Liu, C.; Luo, Y.; Dandapat, K. Accelerating Multimetallic Catalyst Discovery with Robotics and Agentic AI. ChemRxiv November 6, 2025. https://doi.org/10.26434/chemrxiv-2025-13n3f-v2.

(155) Xin, H.; Kitchin, J. R.; Kulik, H. J. Towards Agentic Science for Advancing Scientific Discovery. *Nat. Mach. Intell.* **2025**, *7* (9), 1373–1375. https://doi.org/10.1038/s42256-025-01110-x.

(156) Xu, M.; Kumar, A.; LeBeau, J. M. Towards Augmented Microscopy with Reinforcement Learning-Enhanced Workflows. *Microsc. Microanal.* **2022**, *28* (6), 1952–1960. https://doi.org/10.1017/S1431927622012193.

(157) Yewale, A.; Yang, Y.; Nazemifard, N.; Papageorgiou, C. D.; Rielly, C. D.; Benyahia, B. Deep Reinforcement Learning-Based Self-Optimization of Flow Chemistry. *ACS Eng. Au* **2025**, *5* (3), 247–266. https://doi.org/10.1021/acsengineeringau.5c00004.

(158) Kiumarsi, B.; Vamvoudakis, K. G.; Modares, H.; Lewis, F. L. Optimal and Autonomous Control Using Reinforcement Learning: A Survey. *IEEE Trans. Neural Netw. Learn. Syst.* **2018**, *29* (6), 2042–2062. https://doi.org/10.1109/TNNLS.2017.2773458.

(159) Kim, M.; Ha, M. Y.; Jung, W.-B.; Yoon, J.; Shin, E.; Kim, I.; Lee, W. B.; Kim, Y.; Jung, H. Searching for an Optimal Multi-Metallic Alloy Catalyst by Active Learning Combined with Experiments (Adv. Mater. 19/2022). *Adv. Mater.* **2022**, *34* (19), 2270147. https://doi.org/10.1002/adma.202270147.

(160) Slautin, B. N.; Liu, Y.; Funakubo, H.; Vasudevan, R. K.; Ziatdinov, M.; Kalinin, S. V. Bayesian Conavigation: Dynamic Designing of the Material Digital Twins via Active Learning. *ACS Nano* **2024**, *18* (36), 24898–24908. https://doi.org/10.1021/acsnano.4c05368.

(161) Liu, Y.; Kelley, K. P.; Vasudevan, R. K.; Funakubo, H.; Ziatdinov, M. A.; Kalinin, S. V. Experimental Discovery of Structure–Property Relationships in Ferroelectric Materials via Active Learning. *Nat. Mach. Intell.* **2022**, *4* (4), 341–350. https://doi.org/10.1038/s42256-022-00460-0.

(162) Yue, T.; He, J.; Li, Y. Machine-Learning-Assisted Molecular Design of Innovative Polymers. *Acc. Mater. Res.* **2025**, *6* (8), 1033–1045. https://doi.org/10.1021/accountsmr.5c00151.

(163) Ferji, K. Basic Concepts and Tools of Artificial Intelligence in Polymer Science. *Polym. Chem.* **2025**, *16* (21), 2457–2470. https://doi.org/10.1039/D5PY00148J.

(164) Ishii, M.; Ito, T.; Sado, H.; Kuwajima, I. NIMS Polymer Database PoLyInfo (I): An Overarching View of Half a Million Data Points. *Sci. Technol. Adv. Mater. Methods* **2024**, *4* (1), 2354649. https://doi.org/10.1080/27660400.2024.2354649.

(165) Ma, R.; Luo, T. PI1M: A Benchmark Database for Polymer Informatics. *J. Chem. Inf. Model.* **2020**, *60* (10), 4684–4690. https://doi.org/10.1021/acs.jcim.0c00726.

(166) *Khazana*. https://khazana.gatech.edu/ (accessed 2026-03-11).

(167) *CAMPUS Plastics Database*. Default. https://altair.com/campus-plastics (accessed 2026-03-11).

(168) *Crow Polymer Science. Polymer Properties Database. - References - Scientific Research Publishing*. https://www.scirp.org/reference/referencespapers?referenceid=3200210 (accessed 2026-03-11).

(169) He, R.; Cao, J.; Tan, T. Generative Artificial Intelligence: A Historical Perspective. *Natl. Sci. Rev.* **2025**, *12* (5), nwaf050. https://doi.org/10.1093/nsr/nwaf050.

(170) Goodfellow, I. J.; Pouget-Abadie, J.; Mirza, M.; Xu, B.; Warde-Farley, D.; Ozair, S.; Courville, A.; Bengio, Y. Generative Adversarial Networks. arXiv June 10, 2014. https://doi.org/10.48550/arXiv.1406.2661.

(171) Choudhary, K.; DeCost, B.; Chen, C.; Jain, A.; Tavazza, F.; Cohn, R.; Park, C. W.; Choudhary, A.; Agrawal, A.; Billinge, S. J. L.; Holm, E.; Ong, S. P.; Wolverton, C. Recent Advances and Applications of Deep Learning Methods in Materials Science. *Npj Comput. Mater.* **2022**, *8* (1), 1–26. https://doi.org/10.1038/s41524-022-00734-6.

(172) Debus, B.; Parastar, H.; Harrington, P.; Kirsanov, D. Deep Learning in Analytical Chemistry. *TrAC Trends Anal. Chem.* **2021**, *145*, 116459. https://doi.org/10.1016/j.trac.2021.116459.

(173) Weininger, D. SMILES, a Chemical Language and Information System. 1. Introduction to Methodology and Encoding Rules. *J. Chem. Inf. Comput. Sci.* **1988**, *28* (1), 31–36. https://doi.org/10.1021/ci00057a005.

(174) *RDKit*. https://www.rdkit.org/ (accessed 2026-01-27).

(175) *Why modred? — modred 2.1.0 documentation*. https://modred.readthedocs.io/en/stable/whymodred.html (accessed 2026-03-11).

(176) *Dragon 6 user's manual*. https://www.talete.mi.it/help/dragon_help/index.html?dragon_molecular_descriptors.htm (accessed 2026-03-11).

(177) Deagen, M. E.; Dalle-Cort, B.; Rebello, N. J.; Lin, T.-S.; Walsh, D. J.; Olsen, B. D. Machine Translation between BigSMILES Line Notation and Chemical Structure Diagrams. *Macromolecules* **2024**, *57* (1), 42–53. https://doi.org/10.1021/acs.macromol.3c01378.

(178) Amrihesari, M.; Kern, J.; Present, H.; Moreno Briceno, S.; Ramprasad, R.; Brettmann, B. Machine Learning Models for Predicting Polymer Solubility in Solvents across Concentrations and Temperatures. *J. Phys. Chem. B* **2024**, *128* (51), 12786–12797. https://doi.org/10.1021/acs.jpcb.4c06500.

(179) Nistane, J.; Datta, R.; Lee, Y. J.; Sahu, H.; Jang, S. S.; Lively, R.; Ramprasad, R. Polymer Design for Solvent Separations by Integrating Simulations, Experiments and Known Physics via Machine Learning. *Npj Comput. Mater.* **2025**, *11* (1), 187. https://doi.org/10.1038/s41524-025-01681-8.

(180) *Deed - Attribution-NonCommercial-NoDerivatives 4.0 International - Creative Commons*. https://creativecommons.org/licenses/by-nc-nd/4.0/ (accessed 2025-03-19).

(181) Gao, L.; Lin, J.; Wang, L.; Du, L. Machine Learning-Assisted Design of Advanced Polymeric Materials. *Acc. Mater. Res.* **2024**, *5* (5), 571–584. https://doi.org/10.1021/accountsmr.3c00288.

(182) Tran, H.; Gurnani, R.; Kim, C.; Pilania, G.; Kwon, H.-K.; Lively, R. P.; Ramprasad, R. Design of Functional and Sustainable Polymers Assisted by Artificial Intelligence. *Nat. Rev. Mater.* **2024**, *9* (12), 866–886. https://doi.org/10.1038/s41578-024-00708-8.

(183) van den Hurk, R. S.; Pirok, B. W. J.; Bos, T. S. The Role of Artificial Intelligence and Machine Learning in Polymer Characterization: Emerging Trends and Perspectives. *Chromatographia* **2025**, *88* (5), 357–363. https://doi.org/10.1007/s10337-025-04406-7.

(184) Hassan, R.; Kazemi, M. R. Data-Driven Frameworks to Robustly Predict Solubility Parameter of Diverse Polymers. *Sci. Rep.* **2025**, *15* (1), 31157. https://doi.org/10.1038/s41598-025-12758-1.

(185) Hasegawa, S.; Sawada, T.; Serizawa, T. Identification of Water-Soluble Polymers through Machine Learning of Fluorescence Signals from Multiple Peptide Sensors. *ACS Appl. Bio Mater.* **2023**, *6* (11), 4598–4602. https://doi.org/10.1021/acsabm.3c00736.

(186) Dangayach, R.; Jeong, N.; Demirel, E.; Uzal, N.; Fung, V.; Chen, Y. Machine Learning-Aided Inverse Design and Discovery of Novel Polymeric Materials for Membrane Separation. *Environ. Sci. Technol.* **2025**, *59* (2), 993–1012. https://doi.org/10.1021/acs.est.4c08298.

(187) Stubbs, C. D.; Kim, Y.; Quinn, E. C.; Pérez-Soto, R.; Chen, E. Y.-X.; Kim, S. Predicting Homopolymer and Copolymer Solubility through Machine Learning. *Digit. Discov.* **2025**, *4* (2), 424–437. https://doi.org/10.1039/D4DD00290C.

(188) Wang, Q.; Sato, R.; García-Méndez, R.; Jang, W.; Ou, P.; Soon, A.; Zhao, J.; Wang, X.; Orimo, S.; Cheng, E. J. AI Agents for Solid Electrolytes: Opportunities, Challenges, and Future Directions. *AI Agent* **2025**, *1* (1), N/A-N/A. https://doi.org/10.20517/aiagent.2025.10.

(189) Yang, Z.; Ye, W.; Lei, X.; Schweigert, D.; Kwon, H.-K.; Khajeh, A. De Novo Design of Polymer Electrolytes Using GPT-Based and Diffusion-Based Generative Models. *Npj Comput. Mater.* **2024**, *10* (1), 296. https://doi.org/10.1038/s41524-024-01470-9.

(190) Bradford, G.; Lopez, J.; Ruza, J.; Stolberg, M. A.; Osterude, R.; Johnson, J. A.; Gomez-Bombarelli, R.; Shao-Horn, Y. Chemistry-Informed Machine Learning for Polymer Electrolyte Discovery. *ACS Cent. Sci.* **2023**, *9* (2), 206–216. https://doi.org/10.1021/acscentsci.2c01123.

(191) Xie, T.; France-Lanord, A.; Wang, Y.; Lopez, J.; Stolberg, M. A.; Hill, M.; Leverick, G. M.; Gomez-Bombarelli, R.; Johnson, J. A.; Shao-Horn, Y.; Grossman, J. C. Accelerating Amorphous Polymer Electrolyte Screening by Learning to Reduce Errors in Molecular Dynamics Simulated Properties. *Nat. Commun.* **2022**, *13* (1), 3415. https://doi.org/10.1038/s41467-022-30994-1.

(192) Li, K.; Wang, J.; Song, Y.; Wang, Y. Machine Learning-Guided Discovery of Ionic Polymer Electrolytes for Lithium Metal Batteries. *Nat. Commun.* **2023**, *14* (1), 2789. https://doi.org/10.1038/s41467-023-38493-7.

(193) Ishraaq, R.; Das, S. Machine Learning for Probing Polymer and Polyelectrolyte Brushes. *Trends Chem.* **2025**, *0* (0). https://doi.org/10.1016/j.trechm.2025.10.011.

(194) Zahabi, N.; Petsagkourakis, I.; Rolland, N.; Beikmohammadi, A.; Liu, X.; Fahlman, M.; Pavlopoulou, E.; Zozoulenko, I. Deciphering Conductivity in PEDOT Guided by Machine Learning: From Solvent Baths to Charge Paths. *Phys. Rev. Mater.* **2025**, *9* (10), 105402. https://doi.org/10.1103/5h7d-yvsd.

(195) Wang, C.; Kim, Y.-J.; Vriza, A.; Batra, R.; Baskaran, A.; Shan, N.; Li, N.; Darancet, P.; Ward, L.; Liu, Y.; Chan, M. K. Y.; Sankaranarayanan, S. K. R. S.; Fry, H. C.; Miller, C. S.; Chan, H.; Xu, J. Autonomous Platform for Solution Processing of Electronic Polymers. *Nat. Commun.* **2025**, *16* (1), 1498. https://doi.org/10.1038/s41467-024-55655-3.

(196) Beaucage, P. A.; Sutherland, D. R.; Martin, T. B. Automation and Machine Learning for Accelerated Polymer Characterization and Development: Past, Potential, and a Path Forward. *Macromolecules* **2024**, *57* (18), 8661–8670. https://doi.org/10.1021/acs.macromol.4c01410.

(197) Jas, G.; Kirschning, A. Continuous Flow Techniques in Organic Synthesis. *Chem. – Eur. J.* **2003**, *9* (23), 5708–5723. https://doi.org/10.1002/chem.200305212.

(198) Greenway, G. M.; Haswell, S. J.; Morgan, D. O.; Skelton, V.; Styring, P. The Use of a Novel Microreactor for High Throughput Continuous Flow Organic Synthesis. *Sens. Actuators B Chem.* **2000**, *63* (3), 153–158. https://doi.org/10.1016/S0925-4005(00)00352-X.

(199) Alfano, A. I.; García-Lacuna, J.; Griffiths, O. M.; Ley, S. V.; Baumann, M. Continuous Flow Synthesis Enabling Reaction Discovery. *Chem. Sci.* **2024**, *15* (13), 4618–4630. https://doi.org/10.1039/D3SC06808K.

(200) Shields, G. C. Computational Approaches for the Design of Peptides with Anti-Breast Cancer Properties. *Future Med. Chem.* **2009**, *1* (1), 201–212. https://doi.org/10.4155/fmc.09.13.

(201) Kobayashi, S. Flow “Fine” Synthesis: High Yielding and Selective Organic Synthesis by Flow Methods. *Chem. – Asian J.* **2016**, *11* (4), 425–436. https://doi.org/10.1002/asia.201500916.

(202) Kirschning, A.; Solodenko, W.; Mennecke, K. Combining Enabling Techniques in Organic Synthesis: Continuous Flow Processes with Heterogenized Catalysts. *Chem. – Eur. J.* **2006**, *12* (23), 5972–5990. https://doi.org/10.1002/chem.200600236.

(203) Britton, J.; L. Raston, C. Multi-Step Continuous-Flow Synthesis. *Chem. Soc. Rev.* **2017**, *46* (5), 1250–1271. https://doi.org/10.1039/C6CS00830E.

(204) Wilms, D.; Klos, J.; Frey, H. Microstructured Reactors for Polymer Synthesis: A Renaissance of Continuous Flow Processes for Tailor-Made Macromolecules? *Macromol. Chem. Phys.* **2008**, *209* (4), 343–356. https://doi.org/10.1002/macp.200700588.

(205) Zaquen, N.; Rubens, M.; Corrigan, N.; Xu, J.; Zetterlund, P. B.; Boyer, C.; Junkers, T. Polymer Synthesis in Continuous Flow Reactors. *Prog. Polym. Sci.* **2020**, *107*, 101256. https://doi.org/10.1016/j.progpolymsci.2020.101256.

(206) Reis, M. H.; Davidson, C. L. G.; Leibfarth, F. A. Continuous-Flow Chemistry for the Determination of Comonomer Reactivity Ratios. *Polym. Chem.* **2018**, *9* (13), 1728–1734. https://doi.org/10.1039/C7PY01938F.

(207) Hodge, P. Synthesis of Organic Compounds Using Polymer-Supported Reagents, Catalysts, and/or Scavengers in Benchtop Flow Systems. *Ind. Eng. Chem. Res.* **2005**, *44* (23), 8542–8553. https://doi.org/10.1021/ie040285e.

(208) Phan, N. T. S.; Khan, J.; Styring, P. Polymer-Supported Palladium Catalysed Suzuki–Miyaura Reactions in Batch and a Mini-Continuous Flow Reactor System. *Tetrahedron* **2005**, *61* (51), 12065–12073. https://doi.org/10.1016/j.tet.2005.07.109.

(209) Steinbacher, J. L.; McQuade, D. T. Polymer Chemistry in Flow: New Polymers, Beads, Capsules, and Fibers. *J. Polym. Sci. Part Polym. Chem.* **2006**, *44* (22), 6505–6533. https://doi.org/10.1002/pola.21630.

(210) Reis, M. H.; Leibfarth, F. A.; Pitet, L. M. Polymerizations in Continuous Flow: Recent Advances in the Synthesis of Diverse Polymeric Materials. *ACS Macro Lett.* **2020**, *9* (1), 123–133. https://doi.org/10.1021/acsmacrolett.9b00933.

(211) *Copper-mediated controlled radical polymerization in continuous flow processes: Synergy between polymer reaction engineering and innovative chemistry - Chan - 2013 - Journal of Polymer Science Part A: Polymer Chemistry - Wiley Online Library*. https://onlinelibrary.wiley.com/doi/10.1002/pola.26711 (accessed 2026-01-17).

(212) Melker, A.; Fors, B. P.; Hawker, C. J.; Poelma, J. E. Continuous Flow Synthesis of Poly(Methyl Methacrylate) via a Light-Mediated Controlled Radical Polymerization. *J. Polym. Sci. Part Polym. Chem.* **2015**, *53* (23), 2693–2698. https://doi.org/10.1002/pola.27765.

(213) Su, Y.; Song, Y.; Xiang, L. Continuous-Flow Microreactors for Polymer Synthesis: Engineering Principles and Applications. In *Accounts on Sustainable Flow Chemistry*; Noël, T., Luque, R., Eds.; Springer International Publishing: Cham, 2020; pp 147–190. https://doi.org/10.1007/978-3-030-36572-1_5.

(214) Reis, M. H.; Varner, T. P.; Leibfarth, F. A. The Influence of Residence Time Distribution on Continuous-Flow Polymerization. *Macromolecules* **2019**, *52* (9), 3551–3557. https://doi.org/10.1021/acs.macromol.9b00454.

(215) Hornung, C. H.; Guerrero-Sanchez, C.; Brasholz, M.; Saubern, S.; Chiefari, J.; Moad, G.; Rizzardo, E.; Thang, S. H. Controlled RAFT Polymerization in a Continuous Flow Microreactor. *Org. Process Res. Dev.* **2011**, *15* (3), 593–601. https://doi.org/10.1021/op1003314.

(216) Ye, P.; Cao, P.-F.; Su, Z.; Advincula, R. Highly Efficient Reversible Addition–Fragmentation Chain-Transfer Polymerization in Ethanol/Water via Flow Chemistry. *Polym. Int.* **2017**, *66* (9), 1252–1258. https://doi.org/10.1002/pi.5374.

(217) Ye, P.; Cao, P.-F.; Chen, Q.; Advincula, R. Continuous Flow Fabrication of Block Copolymer–Grafted Silica Micro-Particles in Environmentally Friendly Water/Ethanol Media. *Macromol. Mater. Eng.* **2019**, *304* (2), 1800451. https://doi.org/10.1002/mame.201800451.

(218) Rong, L.-H.; Cheng, X.; Ge, J.; Caldona, E. B.; Advincula, R. C. Synthesis of Hyperbranched Polymers via PET-RAFT Self-Condensing Vinyl Polymerization in a Flow Reactor. *Macromol. Chem. Phys.* **2022**, *223* (1), 2100342. https://doi.org/10.1002/macp.202100342.

(219) Ge, J.; Yin, Y.; Caldona, E. B.; Advincula, R. C. Hyperbranched PDMAEMA-Functionalized SiO2 Microparticles: ATRP Polymerization and Grafting in a Continuous Flow Reactor. *MRS Commun.* **2022**, *12* (6), 1147–1153. https://doi.org/10.1557/s43579-022-00263-9.

(220) Rong, L.-H.; Caldona, E. B.; Advincula, R. C. PET-RAFT Polymerization under Flow Chemistry and Surface-Initiated Reactions. *Polym. Int.* **2023**, *72* (2), 145–157. https://doi.org/10.1002/pi.6475.

(221) Desai, S.; Addamane, S.; Tsao, J. Y.; Brener, I.; Swiler, L. P.; Dingreville, R.; Iyer, P. P. AutoSciLab: A Self-Driving Laboratory For Interpretable Scientific Discovery. arXiv December 16, 2024. https://doi.org/10.48550/arXiv.2412.12347.

(222) Orouji, N.; Bennett, J. A.; Canty, R. B.; Qi, L.; Sun, S.; Majumdar, P.; Liu, C.; López, N.; Schweitzer, N. M.; Kitchin, J. R.; Xin, H.; Abolhasani, M. Autonomous Catalysis Research with Human–AI–Robot Collaboration. *Nat. Catal.* **2025**, *8* (11), 1135–1145. https://doi.org/10.1038/s41929-025-01430-6.

(223) Low, A. K. Y.; Cheng, J. J. W.; Hippalgaonkar, K.; Ng, L. W. T. Self-Driving Laboratories: Translating Materials Science from Laboratory to Factory. *ACS Omega* **2025**, *10* (28), 29902–29908. https://doi.org/10.1021/acsomega.5c02197.

(224) Leong, S. X.; Griesbach, C. E.; Zhang, R.; Darvish, K.; Zhao, Y.; Mandal, A.; Zou, Y.; Hao, H.; Bernales, V.; Aspuru-Guzik, A. Steering towards Safe Self-Driving Laboratories. *Nat. Rev. Chem.* **2025**, *9* (10), 707–722. https://doi.org/10.1038/s41570-025-00747-x.

(225) Coley, C. W.; Thomas, D. A.; Lummiss, J. A. M.; Jaworski, J. N.; Breen, C. P.; Schultz, V.; Hart, T.; Fishman, J. S.; Rogers, L.; Gao, H.; Hicklin, R. W.; Plehiers, P. P.; Byington, J.; Piotti, J. S.; Green, W. H.; Hart, A. J.; Jamison, T. F.; Jensen, K. F. A Robotic Platform for Flow Synthesis of Organic Compounds Informed by AI Planning. *Science* **2019**, *365* (6453), eaax1566. https://doi.org/10.1126/science.aax1566.

(226) Leonard, K. C.; Hasan, F.; Sneddon, H. F.; You, F. Can Artificial Intelligence and Machine Learning Be Used to Accelerate Sustainable Chemistry and Engineering? *ACS Sustain. Chem. Eng.* **2021**, *9* (18), 6126–6129. https://doi.org/10.1021/acssuschemeng.1c02741.

(227) Ali, R. S. A. E.; Meng, J.; Jiang, X. Synergy of Machine Learning and High-Throughput Experimentation: A Road Toward Autonomous Synthesis. *Chem. – Asian J.* **2025**, *20* (20), e00825. https://doi.org/10.1002/asia.202500825.

(228) Low, A. K. Y.; Mekki-Berrada, F.; Gupta, A.; Ostudin, A.; Xie, J.; Vissol-Gaudin, E.; Lim, Y.-F.; Li, Q.; Ong, Y. S.; Khan, S. A.; Hippalgaonkar, K. Evolution-Guided Bayesian Optimization for Constrained Multi-Objective Optimization in Self-Driving Labs. *Npj Comput. Mater.* **2024**, *10* (1), 104. https://doi.org/10.1038/s41524-024-01274-x.

(229) Gridach, M.; Nanavati, J.; Abidine, K. Z. E.; Mendes, L.; Mack, C. Agentic AI for Scientific Discovery: A Survey of Progress, Challenges, and Future Directions. arXiv March 12, 2025. https://doi.org/10.48550/arXiv.2503.08979.

(230) *A Survey of AI for Materials Science: Foundation Models, LLM Agents, Datasets, and Tools*. https://arxiv.org/html/2506.20743v1 (accessed 2026-01-13).

(231) Werheid, J.; Melnychuk, O.; Zhou, H.; Huber, M.; Rippe, C.; Joosten, D.; Keskin, Z.; Wittstamm, M.; Subramani, S.; Drescher, B.; Göppert, A.; Abdelrazeq, A.; Schmitt, R. H. Designing an LLM-Based Copilot for Manufacturing Equipment Selection. *Manuf. Lett.* **2025**, *46*, 123–127. https://doi.org/10.1016/j.mfglet.2025.10.017.

(232) Lyall-Brookes, G.; Padgham, A. C.; Slater, A. G. Flow Chemistry as a Tool for High Throughput Experimentation. *Digit. Discov.* **2025**, *4* (9), 2364–2400. https://doi.org/10.1039/D5DD00129C.

(233) Mukhin, N.; Jha, P.; Abolhasani, M. The Role of Flow Chemistry in Self-Driving Labs. *Matter* **2025**, *8* (7). https://doi.org/10.1016/j.matt.2025.102205.

(234) Advincula, R.; Ivanov, I.; Vasudevan, R.; Kumar, R.; Christakopoulos, P.; Tsakanika, M.; Chen, J.; Michael Carillo, J.; Zhu, Q.; Sumpter, B. Harnessing Data and Control with AI/ML-Driven Polymerization and Copolymerization. *Faraday Discuss.* **2026**. https://doi.org/10.1039/D5FD00066A.

(235) Bonciolini, S.; Pulcinella, A.; Noël, T. Tech-Enhanced Synthesis: Exploring the Synergy between Organic Chemistry and Technology. *J. Am. Chem. Soc.* **2025**, *147* (32), 28523–28545. https://doi.org/10.1021/jacs.5c10303.

(236) Abolhasani, M.; Kumacheva, E. The Rise of Self-Driving Labs in Chemical and Materials Sciences. *Nat. Synth.* **2023**, *2* (6), 483–492. https://doi.org/10.1038/s44160-022-00231-0.

(237) Tom, G.; Schmid, S. P.; Baird, S. G.; Cao, Y.; Darvish, K.; Hao, H.; Lo, S.; Pablo-García, S.; Rajaonson, E. M.; Skreta, M.; Yoshikawa, N.; Corapi, S.; Akkoc, G. D.; Strieth-Kalthoff, F.; Seifrid, M.; Aspuru-Guzik, A. Self-Driving Laboratories for Chemistry and Materials Science. *Chem. Rev.* **2024**, *124* (16), 9633–9732. https://doi.org/10.1021/acs.chemrev.4c00055.

(238) Zhu, L.-T.; Chen, X.-Z.; Ouyang, B.; Yan, W.-C.; Lei, H.; Chen, Z.; Luo, Z.-H. Review of Machine Learning for Hydrodynamics, Transport, and Reactions in Multiphase Flows and Reactors. *Ind. Eng. Chem. Res.* **2022**, *61* (28), 9901–9949. https://doi.org/10.1021/acs.iecr.2c01036.

(239) Pijper, B.; Abdiaj, I.; Mazuela, J.; Linares, M. L.; Gómez, J. E.; Rodriguez, R.; Arquero, B. C.; Palao, E.; Cañellas, S.; Alcázar, J. Multistep and Multivectorial Assembly Line Library Synthesis in Flow. *Chem Catal.* **2024**, *4* (10). https://doi.org/10.1016/j.checat.2024.101118.

(240) Ali, Md. A.; Hu, C.; Yttri, E. A.; Panat, R. Recent Advances in 3D Printing of Biomedical Sensing Devices. *Adv. Funct. Mater.* **2022**, *32* (9). https://doi.org/10.1002/adfm.202107671.

(241) Goh, G. L.; Zhang, H.; Chong, T. H.; Yeong, W. Y. 3D Printing of Multilayered and Multimaterial Electronics: A Review. *Adv. Electron. Mater.* **2021**, *7* (10), 2100445. https://doi.org/10.1002/aelm.202100445.

(242) Chen, J.; Yuan, Y.; Wang, Q.; Wang, H.; Advincula, R. C. Bridging Additive Manufacturing and Electronics Printing in the Age of AI. *Nanomaterials* **2025**, *15* (11), 843. https://doi.org/10.3390/nano15110843.

(243) Goh, G. D.; Sing, S. L.; Yeong, W. Y. A Review on Machine Learning in 3D Printing: Applications, Potential, and Challenges. *Artif. Intell. Rev.* **2021**, *54* (1), 63–94. https://doi.org/10.1007/s10462-020-09876-9.

(244) Hassan, M.; Misra, M.; Taylor, G. W.; Mohanty, A. K. A Review of AI for Optimization of 3D Printing of Sustainable Polymers and Composites. *Compos. Part C Open Access* **2024**, *15*, 100513. https://doi.org/10.1016/j.jcomc.2024.100513.

(245) Kalkal, A.; Kumar, S.; Kumar, P.; Pradhan, R.; Willander, M.; Packirisamy, G.; Kumar, S.; Malhotra, B. D. Recent Advances in 3D Printing Technologies for Wearable (Bio)Sensors. *Addit. Manuf.* **2021**, *46*, 102088. https://doi.org/10.1016/j.addma.2021.102088.

(246) Mazzanti, V.; Malagutti, L.; Mollica, F. FDM 3D Printing of Polymers Containing Natural Fillers: A Review of Their Mechanical Properties. *Polymers* **2019**, *11* (7), 1094. https://doi.org/10.3390/polym11071094.

(247) Mohamed, O. A.; Masood, S. H.; Bhowmik, J. L. Modeling, Analysis, and Optimization of Dimensional Accuracy of FDM-Fabricated Parts Using Definitive Screening Design and Deep Learning Feedforward Artificial Neural Network. *Adv. Manuf.* **2021**, *9* (1), 115–129. https://doi.org/10.1007/s40436-020-00336-9.

(248) Elahee, G. M. F.; Cheng, X.; Rong, L.; Xu, M.-W.; Souza, L.; Advincula, R. C.; Maia, J. On the Cogent Formulation of an Elastomeric Silicone Ink Material for Direct Ink Write (DIW) 3D Printing. *Polym. Eng. Sci.* **2024**, *64* (6), 2476–2490. https://doi.org/10.1002/pen.26703.

(249) Elahee, G. M. F.; Cheng, X.; De Souza, L. R.; Caldona, E. B.; Maia, J.; Advincula, R. C. Optimization in 3D Printable Silicone DIW Ink Formulations: Gelling Behavior Control and the Use of Carbon Nanotubes. *J. Appl. Polym. Sci. n/a* (n/a), e70442. https://doi.org/10.1002/app.70442.

(250) Garcia, V. J.; Fazley Elahee, G. M.; Collera, A. K.; Thornton, T.; Cheng, X.; Rohan, S.; Howard, E. L.; Espera, A. H.; Advincula, R. C. On the Direct Ink Write (DIW) 3D Printing of Styrene-Butadiene Rubber (SBR)-Based Adhesive Sealant. *MRS Commun.* **2023**, *13* (6), 1266–1274. https://doi.org/10.1557/s43579-023-00436-0.

(251) Palaganas, N. B.; Mangadlao, J. D.; de Leon, A. C. C.; Palaganas, J. O.; Pangilinan, K. D.; Lee, Y. J.; Advincula, R. C. 3D Printing of Photocurable Cellulose Nanocrystal Composite for Fabrication of Complex Architectures via Stereolithography. *ACS Appl. Mater. Interfaces* **2017**, *9* (39), 34314–34324. https://doi.org/10.1021/acsami.7b09223.

(252) Manapat, J. Z.; Chen, Q.; Ye, P.; Advincula, R. C. 3D Printing of Polymer Nanocomposites via Stereolithography. *Macromol. Mater. Eng.* **2017**, *302* (9), 1600553. https://doi.org/10.1002/mame.201600553.

(253) *DLP 3D Printing Explained: A Complete Guide for Professionals*. https://www.stratasys.com/en/resources/blog/digital-light-processing-dlp-3d-printing-explained/ (accessed 2025-05-22).

(254) Espera Jr., A. H.; Valino, A. D.; Palaganas, J. O.; Souza, L.; Chen, Q.; Advincula, R. C. 3D Printing of a Robust Polyamide-12-Carbon Black Composite via Selective Laser Sintering: Thermal and Electrical Conductivity. *Macromol. Mater. Eng.* **2019**, *304* (4), 1800718. https://doi.org/10.1002/mame.201800718.

(255) Ngo, T. D.; Kashani, A.; Imbalzano, G.; Nguyen, K. T. Q.; Hui, D. Additive Manufacturing (3D Printing): A Review of Materials, Methods, Applications and Challenges. *Compos. Part B Eng.* **2018**, *143*, 172–196. https://doi.org/10.1016/j.compositesb.2018.02.012.

(256) Porcel, C. H.; Schlenoff, J. B. Compact Polyelectrolyte Complexes: "Saloplastic" Candidates for Biomaterials. *Biomacromolecules* **2009**, *10* (11), 2968–2975. https://doi.org/10.1021/bm900373c.

(257) Rydzek, G.; Pakdel, A.; Witecka, A.; Awang Shri, D. N.; Gaudière, F.; Nicolosi, V.; Mokarian-Tabari, P.; Schaaf, P.; Boulmedais, F.; Ariga, K. pH-Responsive Saloplastics Based on Weak Polyelectrolytes: From Molecular Processes to Material Scale Properties. *Macromolecules* **2018**, *51* (12), 4424–4434. https://doi.org/10.1021/acs.macromol.8b00609.

(258) Jurago, A. A.; Viers, R. A.; Nguyen, A. T.; Ribeiro, E. L.; Espera, A. H.; Caldona, E. B.; Advincula, R. C. On the 3D Printing of Polyelectrolyte Complexes: A Novel Approach to Overcome Rheology Constraints. *MRS Commun.* **2023**, *13* (5), 862–870. https://doi.org/10.1557/s43579-023-00415-5.

(259) Pruthi, V.; Chen, Y.-F.; Chen, J.-T.; Théato, P. Azobenzene-Enhanced 3D Printed Complex Hydrogels: Unlocking Light and pH Responsiveness in Polyelectrolyte-Based Networks. *ACS Appl. Polym. Mater.* **2025**, *7* (22), 15809–15820. https://doi.org/10.1021/acsapm.5c03207.

(260) Lee, K.; Mata, J.; Bobrin, V. A.; Kundu, D.; Peterson, V. K.; Corrigan, N.; Boyer, C. 3D Printing Highly Efficient Ion-Exchange Materials via a Polyelectrolyte Microphase Separation Strategy. *Small Sci.* **2024**, *4* (5), 2400019. https://doi.org/10.1002/smsc.202400019.

(261) Li, C.; Cheng, J.; He, Y.; He, X.; Xu, Z.; Ge, Q.; Yang, C. Polyelectrolyte Elastomer-Based Ionotronic Sensors with Multi-Mode Sensing Capabilities via Multi-Material 3D Printing. *Nat. Commun.* **2023**, *14* (1), 4853. https://doi.org/10.1038/s41467-023-40583-5.

(262) Pruthi, V.; Hirschberg, V.; Théato, P. 3D Printable Polyelectrolyte Complex-Integrated Interpenetrating Network Hydrogels with Customizable Mechanical Strength and pH-Responsiveness. *Adv. Mater. Technol.* **2024**, *9* (13), 2400416. https://doi.org/10.1002/admt.202400416.

(263) Jin, Z.; Seong, H.-G.; Srivastava, S.; McGlasson, A.; Emrick, T.; Muthukumar, M.; Russell, T. P. 3D Printing of Aqueous Two-Phase Systems with Linear and Bottlebrush Polyelectrolytes. *Angew. Chem. Int. Ed.* **2024**, *63* (25), e202404382. https://doi.org/10.1002/anie.202404382.

(264) Zhu, F.; Cheng, L.; Yin, J.; Wu, Z. L.; Qian, J.; Fu, J.; Zheng, Q. 3D Printing of Ultratough Polyion Complex Hydrogels. *ACS Appl. Mater. Interfaces* **2016**, *8* (45), 31304–31310. https://doi.org/10.1021/acsami.6b09881.

(265) Deng, X.; Qi, C.; Meng, S.; Dong, H.; Wang, T.; Liu, Z.; Kong, T. All-Aqueous Embedded 3D Printing for Freeform Fabrication of Biomimetic 3D Constructs. *Adv. Mater.* **2024**, *36* (50), 2406825. https://doi.org/10.1002/adma.202406825.

(266) Wang, H.; Zhou, X.; Wang, J.; Zhang, X.; Zhu, M.; Wang, H. Fabrication of Channeled Scaffolds through Polyelectrolyte Complex (PEC) Printed Sacrificial Templates for Tissue Formation. *Bioact. Mater.* **2022**, *17*, 261–275. https://doi.org/10.1016/j.bioactmat.2022.01.030.

(267) Liu, Q.; Li, Q.; Xu, S.; Zheng, Q.; Cao, X. Preparation and Properties of 3D Printed Alginate–Chitosan Polyion Complex Hydrogels for Tissue Engineering. *Polymers* **2018**, *10* (6), 664. https://doi.org/10.3390/polym10060664.

(268) Singh, A. K.; Pramanik, K. Fabrication and Investigation of Physicochemical and Biological Properties of 3D Printed Sodium Alginate-Chitosan Blend Polyelectrolyte Complex Scaffold for Bone Tissue Engineering Application. *J. Appl. Polym. Sci.* **2023**, *140* (12), e53642. https://doi.org/10.1002/app.53642.

(269) Zolfagharian, A.; Kouzani, A. Z.; Maheepala, M.; Yang Khoo, S.; Kaynak, A. Bending Control of a 3D Printed Polyelectrolyte Soft Actuator with Uncertain Model. *Sens. Actuators Phys.* **2019**, *288*, 134–143. https://doi.org/10.1016/j.sna.2019.01.027.

(270) Nguyen, A.; Jurago, A. A.; Viers, R. A.; Patten, C.; Chen, Q.; Caldona, E. B.; Advincula, R. C. 3D-Printing Formulated Polyelectrolyte Complexes (PECs) in Air: Silica Compositions in Rheological Optimization for Layering. *MRS Commun.* **2023**, *13* (6), 1326–1334. https://doi.org/10.1557/s43579-023-00457-9.

(271) Yang, J.; Wang, Y.; Angelopoulou, P. P.; Keum, J.; Park, J.; Chen, J.; Advincula, R. C. 3D Printing of Polyelectrolyte Complex-Integrated Photocurable Hydrogel Resins. *ACS Appl. Polym. Mater.* **2025**, *7* (23), 15917–15927. https://doi.org/10.1021/acsapm.5c03018.

(272) Castillo, A. E.; Trejo-Maldonado, M.; Lara-Ceniceros, T. E.; Ramírez-Soria, E. H.; Torres-Lubián, J. R.; Garza-Navarro, M. A.; Advincula, R. C.; Bonilla-Cruz, J. 3D-Printed PEGMA-Co-(PNIPAM Crosslinked with PEGDA) Hydrogels by Digital Light Processing in Aqueous Media. *MRS Commun.* **2025**, *15* (3), 320–330. https://doi.org/10.1557/s43579-025-00732-x.

(273) Maalihan, R. D.; Chen, Q.; Tamura, H.; Sta. Agueda, J. R. H.; Pajarito, B. B.; Caldona, E. B.; Advincula, R. C. Mechanically and Thermally Enhanced 3D-Printed Photocurable Polymer

Nanocomposites Containing Functionalized Chitin Nanowhiskers by Stereolithography. *ACS Appl. Polym. Mater.* **2022**, *4* (4), 2513–2526. https://doi.org/10.1021/acsapm.1c01816.

(274) Maalihan, R. D.; Chen, Q.; Agueda, J. R. H. Sta.; Pajarito, B. B.; Tamura, H.; Advincula, R. C. On the Use of Surfactant-Complexed Chitosan for Toughening 3D Printed Polymethacrylate Composites. *Macromol. Mater. Eng.* **2021**, *306* (1), 2000448. https://doi.org/10.1002/mame.202000448.

(275) Chen, Q.; Mangadlao, J. D.; Wallat, J.; De Leon, A.; Pokorski, J. K.; Advincula, R. C. 3D Printing Biocompatible Polyurethane/Poly(Lactic Acid)/Graphene Oxide Nanocomposites: Anisotropic Properties. *ACS Appl. Mater. Interfaces* **2017**, *9* (4), 4015–4023. https://doi.org/10.1021/acsami.6b11793.

(276) Advincula, R. C.; Dizon, J. R. C.; Caldona, E. B.; Viers, R. A.; Siacor, F. D. C.; Maalihan, R. D.; Espera, A. H. On the Progress of 3D-Printed Hydrogels for Tissue Engineering. *MRS Commun.* **2021**, *11* (5), 539–553. https://doi.org/10.1557/s43579-021-00069-1.

(277) Sta. Agueda, J. R. H.; Chen, Q.; Maalihan, R. D.; Ren, J.; da Silva, Í. G. M.; Dugos, N. P.; Caldona, E. B.; Advincula, R. C. 3D Printing of Biomedically Relevant Polymer Materials and Biocompatibility. *MRS Commun.* **2021**, *11* (2), 197–212. https://doi.org/10.1557/s43579-021-00038-8.

(278) Brion, D. A. J.; Pattinson, S. W. Generalisable 3D Printing Error Detection and Correction via Multi-Head Neural Networks. *Nat. Commun.* **2022**, *13* (1), 4654. https://doi.org/10.1038/s41467-022-31985-y.

(279) Zhang, H.; Yu, Y.; Liu, X.; Liang, N.; Kim, Y.; Shin, D.; Moon, S. K.; Choi, J. P. Autonomous Printing Process Optimisation and In-Situ Anomaly Detection in Fused Deposition Modelling Using an Integrated Data-Driven Approach. *Virtual Phys. Prototyp.* **2025**, *20* (1), e2545523. https://doi.org/10.1080/17452759.2025.2545523.

(280) Bin Abu Sofian, A. D. A.; Lim, H. R.; Chew, K. W.; Show, P. L. Advancing 3D Printing through Integration of Machine Learning with Algae-Based Biopolymers. *ChemBioEng Rev.* **2024**, *11* (2), 406–425. https://doi.org/10.1002/cben.202300054.

(281) Choi, W.; Advincula, R. C.; Wu, H. F.; Jiang, Y. Artificial Intelligence and Machine Learning in the Design and Additive Manufacturing of Responsive Composites. *MRS Commun.* **2023**, *13* (5), 714–724. https://doi.org/10.1557/s43579-023-00473-9.

(282) Rupom, R. H.; Islam, M. N.; Demchuk, Z.; Advincula, R.; Dahotre, N. B.; Jiang, Y.; Choi, W. Tailoring Piezoelectricity of 3D Printing PVDF-MoS2 Nanocomposite via In Situ Induced Shear Stress. *ACS Appl. Nano Mater.* **2024**, *7* (19), 22714–22722. https://doi.org/10.1021/acsanm.4c03695.

(283) Islam, Md. N.; Rupom, R. H.; Adhikari, P. R.; Demchuk, Z.; Popov, I.; Sokolov, A. P.; Wu, H. F.; Advincula, R. C.; Dahotre, N.; Jiang, Y.; Choi, W. Boosting Piezoelectricity by 3D Printing PVDF-MoS2 Composite as a Conformal and High-Sensitivity Piezoelectric Sensor. *Adv. Funct. Mater.* **2023**, *33* (42), 2302946. https://doi.org/10.1002/adfm.202302946.

Last update: 3.11.2026