Rate of Particle Depletion via Coagulation in Isotropic Turbulence

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Abstract

We revisit the turbulent-coagulation fluctuation model originally proposed by Koch & Pope (2002) and extend it to regimes of elevated particle concentration and high Taylor-scale Reynolds number (Re_λ). By retaining first-order fluctuations in both singlet concentration and shear rate—modeled as coupled Ornstein–Uhlenbeck processes—we derive a closed-form analytical expression for the rate ratio, incorporating the full shear–concentration correlation. Our model reduces to the Koch–Pope result in the dilute limit but reveals a power-law enhancement in collision rates at higher Re_λ and particle volume fractions due to non-Gaussian concentration intermittency. These analytical predictions are validated by stochastic simulations supported by DNS, which recover the classical Koch–Pope behavior at low concentrations.

1. Introduction

Turbulence-induced particle aggregation governs everything from raindrop formation in clouds and spray atomization in industry to the growth and removal of aerosols from the atmosphere, and gas bubbles coalescence. It is then necessary to study the singlet depletion to n-plet. A seminal treatment by Koch & Pope (2002) formulated the singlet-population dynamics via a Smoluchowski-type equation

$$\frac{dc_0}{dt} = k\Gamma\, c_0^2 \qquad (1)$$

where c_0 is the mean concentration of singlets, K the baseline collision kernel for laminar encounter, and the aggregation-rate ratio : $r = \frac{\langle c^2\,\Gamma\rangle}{\langle c^2\rangle\langle\Gamma\rangle}$ accounts for turbulent fluctuations in both shear rate Γ and concentration c. By modeling $ln\left(\frac{\Gamma}{\langle\Gamma\rangle}\right)$ as coupled Ornstein–Uhlenbeck processes —and assuming homogeneous, isotropic turbulence with log-normal shear statistics—they derived a closed-form expression for r valid in the dilute, low–Re_λ limit. The purpose of this note to explore the possibility of extending their model to perhaps higher concentrations. Toward that direction we first used a stochastic simulation, we compared the results with that of DNS type simulation. The limitations associated with this kind of simulation are then explored. We then propose a model that extends the limits of analytical results obtained in Koch and Pope (2002) used a valuable tool to support the stochastic simulation.

2. Stochastic Simulation Methodology

Our numerical procedure mirrors that of Koch & Pope (2002), employing the same DNS-derived shear histories to drive the packet model. A stochastic model was used in turbulent flow by representing the fluid as a sequence of small "packets," each of which carries a fixed shear-fluctuation history and lives for a small fraction of the integral-eddy turnover time, T_E.

Fluid packets are considered much smaller than integral scale yet larger than Kolmogorov dissipation length scale. In each packet a shear field characterised by two independent Ornstein–Uhlenbeck processes—parameterized to match the small- and large-scale turbulence time scales—are sampled by an Euler–Maruyama integrator on a uniform time grid. Their sum, exponentiated pointwise, yields a fluctuating shear-field $\Gamma(t)$ that drives collision rates. The particle concentration c(t) is stepped forward under three effects: Coagulation sink proportional to $\Gamma(t)\, c^2$; linear relaxation toward the target mean c0; and Source injection at strength s0 chosen so that the long-time ensemble average of c remains constant c0. This invoke our dynamic equilibrium condition for packets.

A two-stage (trapezoidal) update ensures second-order accuracy in time. We run N independent packets on the order of 100, 000, each with a fresh random-number seed, and discard an initial spin-up period (typically a few packet lifetime) before collecting statistics. To limit memory and cost, we employ an on-the-fly reservoir sampler that picks a fixed number of time points per packet from the quasi-steady window. We also found 10,000 packets would give a good statistic. In the outer loop, we sweep a small, warm-started list of candidate source strengths s0 for each desired mean concentration c0. For each (c0, s0) we compute the percent-error $|\langle c\rangle - c0|/c0$ and stop as soon as it falls below 1%. This yields a converged pair (c0, s0) whose packet-averaged coagulation-rate ratio r and normalised variance of concentration are then recorded.
By sampling each packet after its initial spin-up (a fraction of mixing time, depending on the Reynolds number and concentration).

Koch and Pope (2002) employed a stochastic-packet modeling framework and successfully captured the transient dip and subsequent recovery of the coagulation-rate ratio, r(t), consistent with trends observed in direct numerical simulations (DNS). Their predicted final rate-ratio versus mean volume-fraction curve closely aligns with DNS data, with discrepancies remaining within a few percent over time (see their Figure 3). Notably, the DNS-derived r-c trajectory exhibits a distinct U-shaped curvature, where the minimum marks the characteristic dipping time. This same U-shape behavior has been experimentally reported in studies of cellulose fiber aggregation and is also evident in our DNS-style simulations (see Figure 1a,b) [PhD Thesis]. Furthermore, results from the Gamma-field simulations—corroborated by DNS—reveal that even under nearly constant turbulent intensity, the transient dip in coagulation rate arises intrinsically from the aggregation dynamics themselves.

The stochastic packet model replaces costly DNS turbulence modeling with two correlated 1D stochastic processes per packet, dramatically reducing computational expense while retaining the key physics of packet-to-packet shear and concentration fluctuations. Nevertheless, the simulation result by itself could not be accurate unless the dipping times on DNS simulations are given. That is the problem we challenged when running our simulations, since we wanted to avoid the DNS type calculations to figure the dipping time. In order to improve the accuracy of simulations without being calibrated through DNS data, we propose a model to incorporate the effect of $Re_\lambda C_0$ on choosing proper time scales in the simulation. But before presenting the simulations results, first some dynamical aspects of aggregation process is presented leading to a more generalised fluctuation model in aggregation, we then compare the simulations results with the models developed.

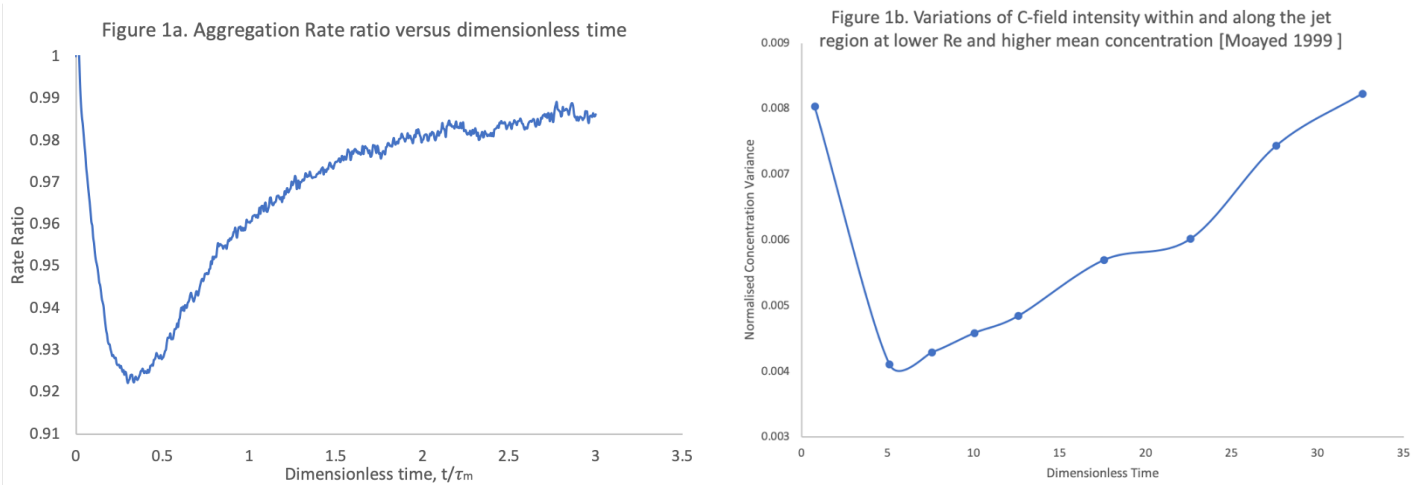

Figure 1a (left)*: DNS type simulations show a U-shape behaviour;* ***Figure 1b (right)****: The same U-shape experimentally observed though in a different flow situation (see Moayed 1999).*

3. Dynamics of Singlet Concentration Fluctuations

Time evolution of the local singlet concentration $c_0(\boldsymbol{x}, t)$ studies here includes terms for coagulation, turbulent mixing, and a uniform source S. The exact singlet balance is

$$\dot{c_0} = -k\Gamma c_0^2 - \frac{c_0 - \langle c_0 \rangle}{\tau_m} + S. \qquad (2)$$

Defining standard Raynolds averaging for Γ and c_0 fields, yields

$$\frac{dc'_0}{dt} + \frac{\acute{c}_0}{\tau} = -k\bar{c}_0^2\Gamma' \qquad \ldots\ldots(3)$$

Where

$$\tau = \frac{\tau_m}{1 + q_\lambda \bar{c}_0}.; and \;\; q_\lambda = 2k\bar{\Gamma}\tau_m \ldots\ldots\ldots\ldots\ldots\ldots\ldots(4)$$

Using the averaging procedure of equation (3) , and introducing a double correlation function for Γ - filed gives a close form for the correlation $\overline{\Gamma'\acute{c}_0}$ (equation14 in KP 2002): $\overline{\Gamma'\acute{c}_0} = -k\bar{c}_0^2 \int_0^\infty e^{\frac{-t}{\tau}} \overline{\Gamma'(s+t)\Gamma'(s)}\, dt$. The equation is similar but now we added an effective mixing time scale that takes into account the effect of mean concentration. Therefore, we get the double correlation function similar to equation 16 (KP 2002). The fact that logarithm of the shear rate in a isotropic turbulence is normally distributed (Pope), would lead them to consider to model the behaviour of $\ln\Gamma$ via two stationary random variables described by a Weiner's process ; ψ varies over the Kolmogorov time scale and χ over the integral time scale. This is an Ornstein–Uhlenbeck (OU) stochastic process for modelling fluctuations in Γ. Actually, they showed that mathematically, there is relationship between the first moment $\overline{\Gamma'\acute{c}_0}$ and concentration variance.

$$\frac{\overline{c_0'^2}}{\bar{c}_0^{\,2}} = -(k\,\tau)\overline{\Gamma' c_0'} \qquad (5)$$

Where the only difference is now that here τ is replaced with the mixing time length τ_m. To further expand the accuracy of the model proposed by KP for extremely dilute condition, we note that the rate of aggregation (or singlets depletion) ratio, may be defined in a more general way. Form equation (1) it follows $r = -\frac{d\bar{c}_0}{dt} = k\bar{\Gamma}\bar{c}_0^{\,2} + 2k\,\bar{c}_0\,\overline{\Gamma' c_0'} + k\,\bar{\Gamma}\,\overline{c_0'^2} + k\,\overline{\Gamma' c_0'^2}$; for a dilute and weak fluctuations in c, this reduces to KP rate as: $r_{kp} = -\frac{d\bar{c}_0}{dt} = k\bar{\Gamma}\bar{c}_0^{\,2} + 2k\,\bar{c}_0\,\overline{\Gamma' c_0'}$. For more concentrated fields, one may similarly write take into account the contributions of higher order scales as:

$$r_l = \frac{r}{k\,\bar{\Gamma}\,\bar{c}_0^{\,2}} = 1 + \frac{2\overline{\Gamma' c_0'}}{\bar{\Gamma}\,\bar{c}_0} + \frac{\overline{c_0'^2}}{\bar{c}_0^{\,2}} + \frac{\overline{\Gamma' c_0'^2}}{\bar{\Gamma}\,\bar{c}_0^{\,2}} + \frac{\overline{\Gamma' c_0'^3}}{\bar{\Gamma}\,\bar{c}_0^{\,3}} + \cdots \qquad \ldots\ldots(6)$$

Physically as concentration increases the contribution of higher moments of concentration distribution becomes more relevant, the first two terms are the contributors of mean and variance, where as the third and the fourth terms are the contributions of skewness and kurtosis of the concentration distribution.

Now we are dealing with the turbulence closure problem. The more terms we introduce the more equations needed to be formed and solved. we propose a geometric cascading model that incorporates higher-order concentration fluctuations into the dynamic equation for the aggregation rate. The concentration fluctuations are assumed to follow a hierarchical cascade driven by interactions across turbulent eddies, with each scale contributing to local enhancement of collision probability. The first-order correction emerges naturally from the linear correlation $\langle c'\Gamma'\rangle$, but higher-order terms, such as $\langle c'^2\Gamma'\rangle$ and $\langle c'^3\Gamma'\rangle$, require modeling the nonlinear coupling between concentration variance and shear fluctuations.

Physically, this approach reflects the non-uniform migration of particles under turbulent straining (moving preferentially toward less strained regions) to be entrained and encounter clusters, generating local zones of amplified aggregation. Mathematically, we express this by extending the dynamic equation for $\langle c\rangle$ with additive terms involving higher-order correlations like $\langle c'^n\Gamma'\rangle$, approximated using a slaved log-normal model of concentration. In this model we start with the log-normal ansatz for the concentration field: $c = \bar{c}e^Y, Y = N\left(-\frac{1}{2}\sigma_Y^2, \sigma_Y^2\right)$ and define the mixed moments as $\langle c'^n\Gamma'\rangle = \bar{c}^n\langle(e^Y-1)^n\Gamma'\rangle$. When we retain only the leading factor in the binomial expansion and invoking slaving approximation we get:

$$\langle c'^n\Gamma'\rangle = e^{(n-1)\mu_0}\,\bar{c}^{(n-1)}\,\langle\Gamma' c'\rangle \qquad (7)$$

By incorporating the analytical form of $\langle\Gamma' c'\rangle$, which is essentially the same form as equation (16, KP-2002) but with the replacement of effective time scale equation (4), and equation (9) into equation (7), we get:

$$r_l = 1 - 2\beta_c\,z + \beta_c\sum_{i=2}^{n} z^i, \qquad \ldots\ldots(8)$$

Where $z = \frac{(\epsilon\, q_\lambda\, \overline{c_0})}{2(1+q_\lambda \overline{c_0})}$; $q_\lambda = 2k\left(\frac{3}{20}\right)^{\frac{1}{2}}\left(\frac{\bar{\Gamma}}{\sqrt{\bar{\Gamma}^2}}\right) Re_\lambda$, and $\epsilon = e^{\mu_0}$ is the intermittency factor signifying the degree of contribution to the rate ratio driven from higher moments of concentration distribution. It depends on the number of contributors, n. Note that in this equation the definition for β_c is now different than the coefficient β defined in equation (15, KP-2002), altered by the definition of τ. Figure (2) compares the two β's for a very dilute condition. As can been seen the curves are nearly identical. This is a striking result. From this figure we confirm the prediction of our model beta with that of supported by DNS at extremely dilute suspension, which is equivalent to consider only the first linear term in above equation for the rate ratio.

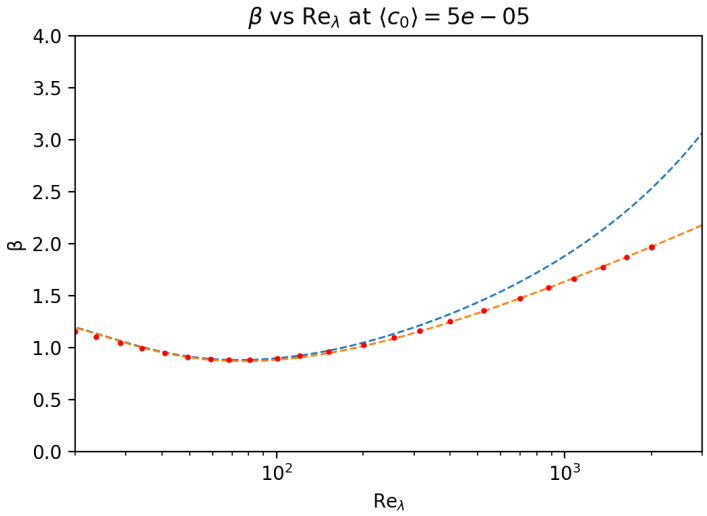

Figure 2: *Variations of beta with Reynolds number at two dilute concentrations; Dots are taken from KP 2001 DNS supported data, the dashed curve is the current model at nearly zero concentration and the upper curve is the prediction od the model at slightly higher concentration.*

For an extreme case n>>1 and $Re_\lambda \overline{c_0} \gg 1$; we find $r_l^\infty \sim 0.76\ at$ at $\epsilon = 1$ larger than the rate predicted by segregation model at Re=500; $r_{segg} = 0.739$; nevertheless considering a model up to n=4; would yield a closer value at $\epsilon = 0.99$; $r_l^\infty \sim 0.745$.
Similarly we can find the normalised variance of singlet concentration from the following equation:

$$\frac{\overline{c_0'^2}}{\overline{c_0}^2} = \beta_c \sum_{i=2}^{n} z^i = \frac{\beta_c\, z^2(1-z^n)}{1-z} \ldots\ldots\ldots\ldots (9)$$

Our comparisons with stochastic simulation confirms that Equations (8-9) better fit DNS supported data at higher concentrations, capturing the upward curvature in the aggregation rate ratio not explained by second-order model. However, the model's shortcoming lies in its assumption to slave all moments to a single correlation function at the limit of very dilute suspension, which may not hold true for systems that particles have effect on turbulence. Nevertheless, for coagulating systems we are dealing with sizes of very small particles, possibly even smaller than Kolmogorov length scale. It also provides a systematic way to include the

effect of turbulent intermittency through an expansion in log-normal moments tied to the intermittency coefficient e^{μ_0} .

4. Results and Discussions

Koch and Pope (2002) used DNS data at R_λ=500 to extract time-resolved shear-rate fields along fluid trajectories. They performed simulations on 10,000 independent fluid packets, each evolving its concentration based on the DNS shear rate Γ(t) using a population balance equation similar to equation 2.

To validate equations (8) and (9), we ran simulations of the rate ratio and normalized variance at Reynolds numbers, $Re_\lambda = 24, 125, 250, and\ 500$. As noted above, the stochastic model's initial "burn-in" time or transient time depends on both the particle concentration and R_λ, and decreases as $R_\lambda C_0$ increase. Considering adopting calibration curve Figure 2 for extremely low concentration, we therefore shoot our simulations at very low concentration to be consistent with this DNS supported curve. Using these adjustments, we then ran guided simulations of the rate ratio at the same R_λ. As Figure 4 illustrates, the partial unguided simulations deviate markedly from the expected behavior at very low concentrations; adopting the correct burn-in time scale brings them into much better agreement with the guided curve.

Comparisons between the rate ratios between the simulations and the model (see Figure 3 and 4), revealed that the only most likely contributors to the rate of aggregation are up to the order 4th term, i.e skewness and kurtosis of particle concentration at higher Reynolds number, in contrast at lower Reynolds number, the rate may be affected up to order 3, i.e skewness, good enough to describe almost whole spectrum.
As can be seen the agreement between the models proposed with that of simulations for both $\frac{\overline{c_0'^2}}{\overline{c_0}^2}$, and r_l are excellent, nevertheless the present model predicts an equilibrium for variance of about 0.122; at extremely large mean concentration, shown in Figure (3). This value is less than that of predicted by a segregation model (KP)of about 0.162. Meaning that our model slightly under predicts the variance at extremely large concentration. The possible short coming is the net effect of higher order fluctuations may increase the variance. This translates to the fact that there may be other non-linear sources rather than just the one used in equation (8) contributing to the total variance of singlet concentration.

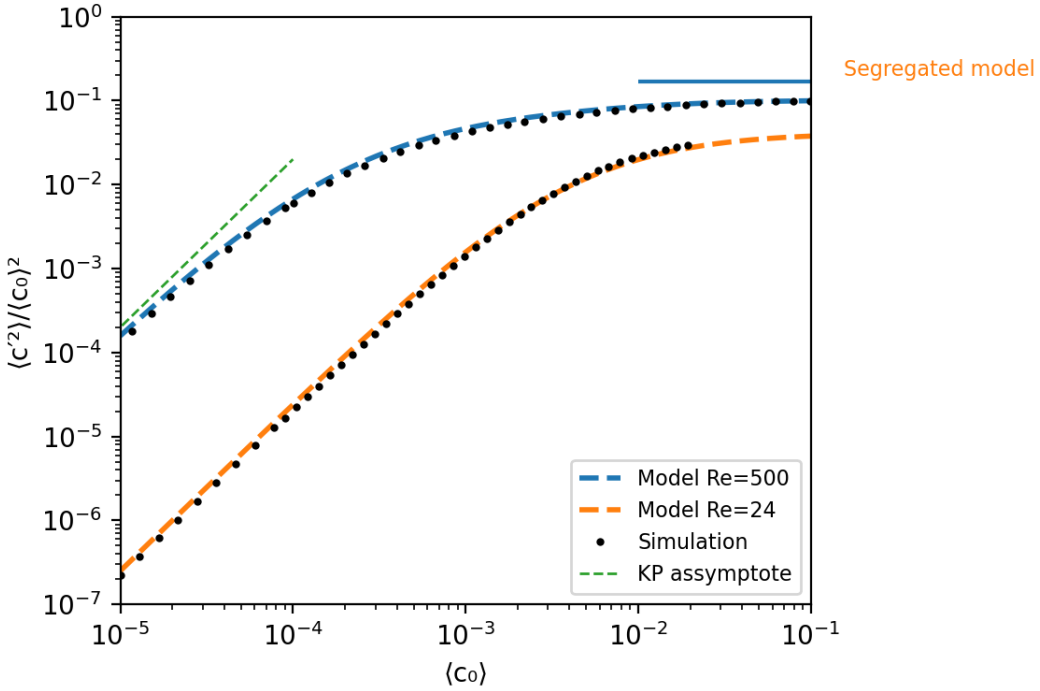

Figure 3: *Comparison between the proposed model (Equation 9) and stochastic simulations for the normalized variance of volumetric concentration, $\langle c^2 \rangle / \langle c0 \rangle^2$, as a function of mean concentration $\langle c0 \rangle$. Results are shown for two Reynolds numbers, Re_λ=24 and Re_λ=500. At higher Reynolds number, higher-order moments of the concentration distribution (e.g., skewness and kurtosis) contribute significantly to the variance. In contrast, at lower Re_λ, the variance is primarily governed by the skewness alone. The KP asymptotic model is shown for reference. The segregated model curve provides an upper bound representing maximal variance.*

5. Conclusions

We have developed and validated an extended turbulent-coagulation model that retains first-order fluctuations in both shear rate and particle concentration—modeled as coupled Ornstein–Uhlenbeck processes—and derived a closed-form expression for the global aggregation–rate ratio, r, and normalised concentration variance valid from the dilute limit well into elevated volume fractions and high Taylor-Reynolds numbers. Key findings include:

1. Although it is often regarded as an introductory model describing the early stages of aggregation, it nonetheless captures key physical features of turbulent coagulation. Notably, this work demonstrates that the characteristic U-shaped behavior observed in the unsteady r-c curve arises directly from coagulation dynamics, even when the turbulent intensity remains constant.
2. Unified formulation**:** In the dilute limit our expression recovers the classic Koch & Pope result, while at higher concentrations non-Gaussian intermittency produces a pronounced, power-law enhancement of collision rates.
3. Role of higher moments**:** Simulation and analytical comparison across Re_λ=24–500 show that skewness and kurtosis (3rd– and 4th-order concentration moments) suffice to

capture the upward curvature of r; beyond these, additional terms yield diminishing returns.

4. Accurate variance prediction: Our model reproduces the normalized concentration variance across a broad parameter space, predicting the correct asymptotic equilibrium level at very high mean concentrations.

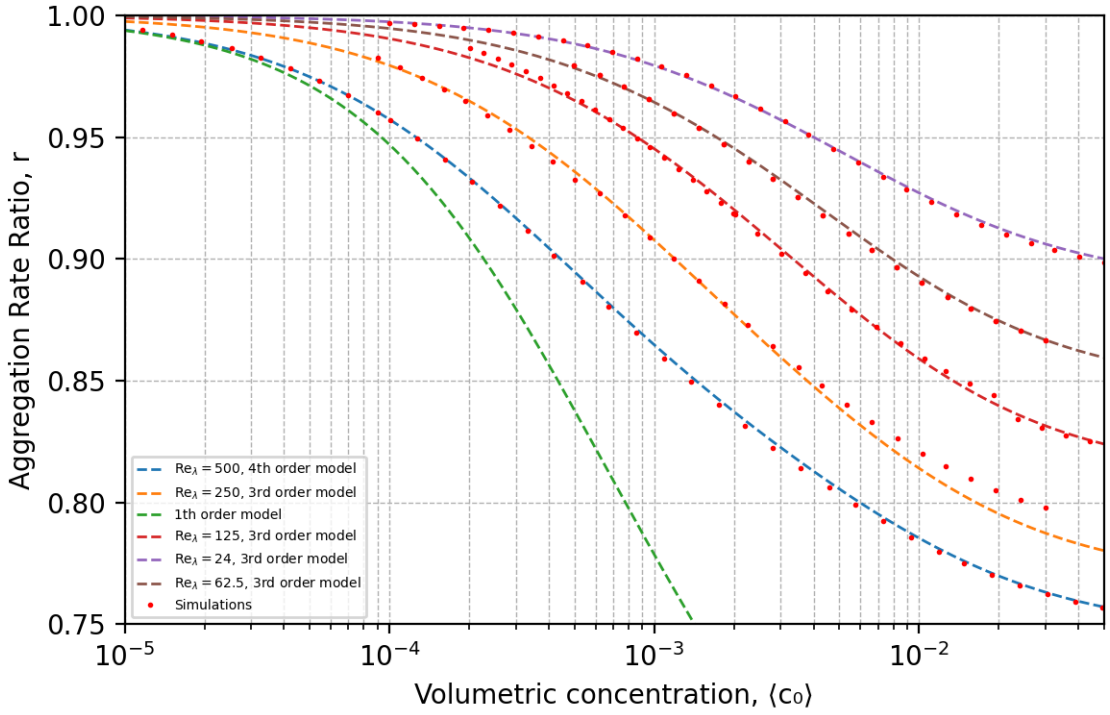

Figure 4: *Comparison between the analytical model (Equation 8) and stochastic simulations for the aggregation rate ratio r as a function of volumetric concentration ⟨c0⟩. Simulations are shown for various Reynolds numbers (Re_λ=24 to 500). The third-order model generally agrees*

Potential Application: Electrolysis-Driven Hydrogen Production

The extended coagulation solver can be integrated into CFD simulations of water–electrolysis reactors, where gas-bubble nucleation, coalescence, and detachment critically affect mass-transfer efficiency and product purity. By coupling our rate–ratio and variance predictions to local shear and concentration fields around electrode surfaces, one can:

- Predict bubble-size distributions as functions of current density and turbulent mixing.
- Optimize electrode spacing, flow-channel geometry, and operating conditions to maximize hydrogen yield.
- Reduce parasitic coalescence losses and control bubble-induced voltage fluctuations.

Beyond electrolysers, this framework applies broadly to aerosol dynamics in fuel cells, crystallization in stirred reactors, and flocculation processes in water treatment—wherever turbulence-driven aggregation dictates performance.

well with the simulation data, except at Re_λ=250, where deviations emerge at higher concentrations. At Re_λ=500, higher-order contributions (up to fourth order) become significant and necessary for accurate prediction.

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