

BREAKUP AND AGGREGATION IN TURBULENTLY STIRRED VESSELS

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Abstract. The breakup and aggregation of small solid particle aggregates in heterogeneous flows is modeled through a multizonal population balance model. Breakup and aggregation are described through comprehensive rate expressions that take into account the local properties of the turbulent flow. Two approaches are pursued to define the zones: in the first approach the zones are predefined and computational fluid dynamics is used to compute the zone properties. In the second approach the zone properties are directly fitted to experimental data. Model results compare well with experiments for the aggregation of a polystyrene latex in a stirred tank even when only two zones are used.

Key words: Breakage, aggregation, population balance equation, zone model, macromixing

1 INTRODUCTION

Breakup of sparsely suspended solid aggregates due to vigorous agitation is an important process in a variety of industrial and natural processes, e. g., processing of polymeric colloids and minerals, crystallization, dispersion of powders, flocculation of suspended solids in natural waters, etc. In this work the breakup of small solid particle aggregates during aggregation in a turbulent flow is investigated. For this, a multizonal population balance equation (PBE) model is developed that couples both aggregation and breakup kinetics with the macromixing pattern in the processing unit.

PBEs are well established tools for describing particulate processes, and its application for aggregation processes in agitated flows has received considerably attention during the last years [1–4]. However, the focus of these studies was mainly on the interplay between aggregation and breakage whereas the detailed mechanisms that drive aggregate breakup have received relatively little attention. In particular, in most of these studies breakup is described through a lumped rate expression, i. e., a power law, in combination with some simple rules for fragment formation, i. e., binary breakage. This not only gives little insight into the physics of breakup (i. e., the parameters of the power law expression lack a clear physical meaning), but also, it adds little to the understanding of the role of fluid flow and turbulence in the breakup process. The latter is however essential for up-scaling or transfer of aggregation processes.

Recently, Bäbler *et al.* [5] developed a physically sound model for the rate of breakup of aggregates in a turbulent flow. The flow was assumed to be statistically homogenous

and a multifractal model was used to describe the statistics of the turbulent fluctuations. However, recalling that the flow in an agitated vessel is highly heterogeneous on the large scale we have that Bähler *et al.*'s model applies only locally. Thus, in the present work the model of Bähler *et al.* is combined with a zone model to account for large scale heterogeneity and macromixing in the vessel. In this frame the flow domain is divided into a small number of interconnected zones where in each zone the flow and the concentration field are assumed to be homogeneous [6]. A PBE that accounts for aggregation, breakage and exchange with other zones is then solved for each zone. Two approaches were pursued to formulate the zone model. In the first approach, we used pre-defined zones and computational fluid dynamics (CFD) was used to compute turbulent quantities and volume fluxes for each zone. In the second approach we defined the zones empirically and fitted the model parameters directly to experimental data obtained from an aggregation experiment in a stirred tank [7].

2 MODEL DEVELOPMENT

In what follows, the aggregates are assumed to be small and of similar density than the suspending fluid such that the aggregate Reynolds number $Re_p = R\Gamma^2/\nu \ll 1$ and the aggregate Stokes number $St_p = \Gamma t_p \ll 1$. In these equations, R is the characteristic radius of an aggregate, Γ is the characteristic velocity gradient of the fluid flow, and $t_p = (2\rho_a/\rho_f + 1)R_g^2/9\nu$ is the aggregate relaxation time, i. e., the time it takes for an aggregate to accommodate its velocity to the velocity of the surrounding fluid flow. $Re_p \ll 1$ implies that hydrodynamic forces acting on the aggregate are controlled by viscous fluid motions, whereas $St_p \ll 1$ implies that the aggregate has negligible inertia and that it closely follows the fluid flow. As a consequence of this the governing breakup mechanism of the aggregates is due to viscous stresses acting on the aggregate. Breakup due to collisions or impacts is negligible in the present framework because small aggregates with negligible inertia are subject to a strong retardation when approaching each other [8] or when approaching a wall. Last, note that when the flow is in a liquid (for which $\rho_a/\rho_f \sim \mathcal{O}(1)$) and turbulent (for which $\Gamma \approx (\varepsilon/\nu)^{1/2}$) the two conditions given above reduce to $R \ll \eta$ where $\eta = (\nu^3/\varepsilon)^{1/4}$ is the Kolmogorov length scale of the flow. Further, the aggregates are assumed to exhibit a porous fractal structure which is expressed through:

$$x = k_g(R_g/R_p)^{d_f} \quad (1)$$

where R_g is the radius of gyration of an aggregate consisting of x primary particles, k_g is a prefactor taken as $k_g = 1$, and d_f is the fractal dimension.

Before turning to the model equations a few words on averaging the flow properties are required. Next to the actual values that are fluctuating in both time and space we have three levels of averaging: First, we have ensemble averages that hereafter are denoted by $\langle \cdot \rangle$. For the statistically steady flow considered here the ensemble averages depend only on spatial coordinates, and it are these quantities that are typically provided by CFD models, e. g., RANS or LES. Although these quantities vary substantially with location in, e. g., a stirred tank, and thus exhibit also a probability density distribution (PDF), we have to note that they do not represent the fine scale turbulent structure, i. e., the local fluctuations. The latter are believed to be essential for breakup processes [9, 10] and in [5] a multifractal model is formulated to describe these fluctuations. Second, we have zone averages, i. e., volume averages over the zone volume, that approximate to local the ensemble averages. The zones are ideally defined such that they enclose a region where the ensemble averages exhibit only small variations [6], thus allowing for a reduction of

Table 1: Integral operators for aggregation and breakage.

$$\mathcal{L}_a^{(k)} \{c_k(x, t)\} = \frac{1}{2} \int_0^x k_a^{(k)}(x-y, y) c_k(x-y, t) c_k(y, t) dy - c_k(x, t) \int_0^\infty k_a^{(k)}(x, y) c_k(y, t) dy$$

$$\mathcal{L}_b^{(k)} \{c_k(x, t)\} = -k_b^{(k)}(x) c_k(x, t) + \int_x^\infty g^{(k)}(x, y) k_b^{(k)}(y) c_k(y, t) dy$$

the size of problem but keeping resolved the large scale heterogeneity of the flow. Zone averages are hereafter denoted by $\langle \cdot \rangle_k$ where the index k refers to the zone. On the third level we have volume averages over the entire flow domain which are denoted by $[\cdot]_{av}$.

2.1 Multizone population balance model

The governing equation of our model is a PBE that describes the evolution of the cluster mass distribution (CMD). Let $c(x, t)dx$ denote the number of cluster per unit volume with masses $\in (x, x + dx)$, where $c(x, t)$ is made dimensionless by dividing it by the total number of primary particles per unit volume, C_0 . The dimensionless PBE that describes the change in the CMD in zone k , $c_k(x, t)$, reads as

$$\frac{\partial c_k(x, t)}{\partial t} = \frac{1}{\varphi_k} \sum_{l=1}^m \left(q_{kl} c_l(x, t) - q_{lk} c_k(x, t) \right) + \phi \mathcal{L}_a^{(k)} + \mathcal{L}_b^{(k)}. \quad (2)$$

The first term on the rhs of Eq. (2) accounts for the exchange with other zones. Thereby, q_{kl} is the (dimensionless) volume flux from zone l to zone k , and φ_k is the relative volume of zone k . The second and the third term on the rhs of Eq. (2) account for aggregation and breakage within zone k where the integral operators $\mathcal{L}_a^{(k)}$ and $\mathcal{L}_b^{(k)}$ describing these two processes are given in Tab. 1. In these equations $k_a^{(k)}(x, y)$ is the aggregation rate function, $k_b^{(k)}(x)$ is the breakage rate function, and $g^{(k)}(x, y)$ is the fragment mass distribution function. The latter specifies the number of fragments of mass $\in (x, x + dx)$ resulting from the breakup of a cluster of mass y to be equal to $g^{(k)}(x, y)dx$.

The dimensionless form of Eq. (2) was obtained from the corresponding dimensional equation by dividing the latter by GC_0 , where $G = ([\varepsilon]_{av}/\nu)^{1/2}$ is a characteristic shear rate of the processing vessel (note that G represents an improper average of the shear rate in the stirred tank, i. e., $G \neq [G]_{av}$). Accordingly, t reads as (dimensional time) $\times G$, $q_{kl} = Q_{kl}/VG$, and $\phi = \frac{4\pi}{3} R_p^3 C_0$ is the solid volume fraction.

The multizonal PBE model consist of Eq. (2) formulated for all the m zones and the aggregation and breakage terms given in Tab. 1. To complete the model appropriate expressions for the rate functions appearing in Tab. 1 as well as the definition of the zone network is required. These two points are addressed next.

2.2 Rate expressions for breakup and aggregation

Bäbler *et al.* [5] developed a model for the rate of breakup in a homogeneous turbulent flow. It was assumed that an aggregate breaks up instantaneously (i. e., with a characteristic time that is shorter than the fastest time scale of the turbulent flow) once the

Table 2: Dimensionless rate expressions for aggregation and breakage.

$$k_b^{(k)}(x) = k_{b0}\gamma_k \left[\int_{\varepsilon_{cr}(x)}^{\infty} p_k(\varepsilon) (\varepsilon/\langle\varepsilon\rangle_k)^{1/2} d\varepsilon \right] \left[\int_0^{\varepsilon_{cr}(x)} p_k(\varepsilon) d\varepsilon \right]^{-1}$$

$$g^{(k)}(x, y) = \left[\int_{\varepsilon_{cr}(y)}^{\varepsilon_{cr}(x)} \varepsilon^{1/2} p_k(\varepsilon) \frac{2y}{[x_{cr}(\varepsilon)]^2} d\varepsilon \right] \left[\int_{\varepsilon_{cr}(y)}^{\infty} \varepsilon^{1/2} p_k(\varepsilon) d\varepsilon \right]^{-1}, \quad x \leq y$$

$$k_a^{(k)}(x, y) = k'_{a0}\gamma_k f_k(x, y) \left(x^{1/d_f} + y^{1/d_f} \right)^3 + \frac{3}{Pe} \left(x^{-1/d_f} + y^{-1/d_f} \right) \left(x^{1/d_f} + y^{1/d_f} \right)$$

$$k_{a0'} = 3k_{a0}/4\pi, \quad \gamma_k = G_k/[G]_{av}, \quad Pe = 6\pi\mu[G]_{av}R_p^3/kT$$

hydrodynamic stress acting on it exceeds a critical value. The latter was assumed to be determined solely by the aggregate and its micro-mechanical properties. For aggregates below the Kolmogorov length scale the hydrodynamic stress is proportional to the local velocity gradient of the fluid flow that is determined by the local energy dissipation rate and the viscosity. The former is thereby known to exhibit strong and intermittent fluctuations even in a flow that is homogeneous on the large scale. The rate of breakage is then given by the frequency at which the local energy dissipation rate exceeds the critical value (corresponding to the critical stress). Working out this principle led to a breakage rate function, that in dimensional form reads as

$$K_B(x) = k_{b0} \left[\int_{\varepsilon_{cr}(x)}^{\infty} p(\varepsilon; R_\lambda)/\tau(\varepsilon) d\varepsilon \right] \left[\int_0^{\varepsilon_{cr}(x)} p(\varepsilon; R_\lambda) d\varepsilon \right]^{-1} \quad (3)$$

where k_{b0} is a prefactor taken as $k_{b0} = 1$, and $\varepsilon_{cr}(x)$ is the critical energy dissipation rate above which an aggregate of mass x breaks up. The functions in the arguments of the integrals represent the statistics of the turbulent flow: $p(\varepsilon; R_\lambda)$ is the PDF of the *fluctuating* energy dissipation rate, $\tau(\varepsilon)$ is the characteristic time of a turbulent event of magnitude ε , and R_λ is the Reynolds number based on the Taylor length scale that equates as $R_\lambda = (\frac{20}{3}k^2/\langle\varepsilon\rangle\nu)^{1/2}$. For the multifractal model used to describe $p(\varepsilon; R_\lambda)$ reference to [5] should be made. The timescale $\tau(\varepsilon)$ is approximated through the local Kolmogorov time scale, $\tau(\varepsilon) \sim (\varepsilon/\nu)^{1/2}$ [5, 9] where the factor of proportionality goes into the prefactor k_{b0} . Last, the critical energy dissipation rate above which an aggregate of mass x breaks up is expressed through a power law [11], conveniently expressed through

$$x_{cr} = C_s \varepsilon^{-q} \quad (4)$$

where C_s and q are parameters that depend on the bonds between the primary particles and the aggregate structure, respectively.

In a similar way, Bähler *et al.* [5] derived an expression for the fragment distribution function $g(x, y)$. For this, it was assumed that all fragments that result from a breakup event caused by a turbulent structure of magnitude ε have to be smaller than the maximum attainable size in that particular flow structure, which is given by Eq. (4). The resulting expression is given in Tab. 2 together with the rate functions in dimensionless form.

Table 3: Three zone model from CFD.

(a) Zone averaged flow properties					(b) Volume fluxes		
Zone	ϕ_i	$\langle \varepsilon \rangle_i / [\varepsilon]_{av}$	$k_i / [k]_{av}$	$R_{\lambda,i} / R_{\lambda,0}$			
1	0.0096	19	4.9	1.26	$Q_{kl} = ND^3 \begin{bmatrix} 0 & 0 & 0.197 \\ 0.197 & 0 & 0.364 \\ 0 & 0.561 & 0 \end{bmatrix}$		
2	0.0484	6.9	3.9	2.20			
B	0.9420	0.51	0.81	1.28			

It is worth to point out that both the breakage rate function given in Eq. (3) as well as the fragment distribution function given in Tab. 2 are built of three constituting parts, i. e., the PDF of the local energy dissipation rate, $p(\varepsilon; R_\lambda)$, the characteristic eddy timescale, $\tau(\varepsilon)$, and the aggregate strength function $\varepsilon_{cr}(x)$ (or its inverse $x_{cr}(\varepsilon)$). Provided that the flow properties $\langle \varepsilon \rangle$ and k are given the breakage model used here contains therefore next to the prefactor k_{b0} (that is fixed to unity) only the two parameters defining the aggregate strength, i. e., q and C_s . These two parameters are used as fitting parameters.

Regarding aggregation, particles in the colloidal and micrometer size range are brought into close contact due to their relative motion in the fluid flow and due to their Brownian motion. Both these mechanisms lead to different aggregation behavior expressed through different rate functions. As derived theoretically in [12] and recently confirmed experimentally in [13] it occurs that for fully destabilized particles, i. e., particles with negligible repulsion, the effective aggregation rate can be expressed as a linear combination of these two mechanisms. Accordingly, the aggregation rate function reads as

$$K_A^{(k)}(x, y) = k_{a0} G_k f(x, y) \left(R_g(x) + R_g(y) \right)^3 + \frac{2kT}{3\mu} \left(\frac{1}{R_g(x)} + \frac{1}{R_g(y)} \right) \left(R_g(x) + R_g(y) \right) \quad (5)$$

where k_{a0} is a prefactor, $f(x, y)$ is the aggregation efficiency, and $G_k \equiv (\langle \varepsilon \rangle_k / \nu)^{1/2}$. For the aggregation efficiency, the model of Bähler [14] is used according to which $k_{a0} \approx 0.36$. It has been shown recently that this model accurately describes aggregation in turbulent flows [13].

Dimensionless forms of both the aggregation model and the breakage model are given in Tab. 2 where $R_g(x)$ appearing in Eq. (5) is substituted for the fractal scaling relation, Eq. (1). Accordingly, the aggregation rate function contains solely the fractal dimension d_f that cannot be determined from the process conditions. For the polymeric aggregates used to validate the model, d_f was found to be $d_f \approx 2.6$ [7].

2.3 Zone definition

Two approaches are pursued to define the zones within the stirred tank studied in this work. In the first approach the zones are predefined according to the flow map of Bourne and Yu [15]. There, the volume of the stirred tank is divided into (1) an impeller swept zone, (2) a discharge zone, and (3) several zones to describe the circulation pattern, see [15]. In the present work, only the first two of these zones are used whereas the remaining zones are treated as bulk flow. This is motivated by the fact that the energy is mainly dissipated in the first two zones.

Having defined the zones, the properties of the turbulent flow and the volume fluxes between the zones were computed through CFD applying standard k - ε model. Calculations at several stirring speeds indicate that the macro-flow pattern becomes independent

of the stirring speed once the flow is fully turbulent. Tab. 3 gives the zone properties of the three zone model computed from CFD. With this method the most turbulent region of our stirred tank assumes $\langle \varepsilon \rangle = 19[\varepsilon]_{av}$. This is roughly an order of magnitude below the highest values for $\langle \varepsilon \rangle$ found in CFD that, however, are only assumed within a few cells of the CFD grid, that is to say, within a very small volume. To explore the influence of these large values of $\langle \varepsilon \rangle$ we applied a second method for defining the zone. Thereby, the reactor volume was arbitrarily divided into two zones where we assumed $\langle \varepsilon \rangle_1/[\varepsilon]_{av} = 50$ and $k_i/[k]_{av} = 33$ and that 99% of the energy are dissipated in zone 1.

3 RESULTS

The multizonal PBE model developed in this work is used to interpret the aggregation experiments reported in [7]. In this study a fully destabilized colloidal polystyrene latex with a primary particle size of $R_p = 0.405 \mu\text{m}$ was aggregated in a baffled stirred tank equipped with a Rushton impeller. A method was applied to measure the steady state CMD as a function of the solid volume fraction of the suspension through continuous dilution [16]. At steady state, aggregation and breakage balance each other and it is this data to which the model is applied in this work, i. e., hereafter, only stationary solutions to Eq. (2) are considered. It reveals that in particular variations in the solid volume fraction present a severe test to the model: due to the different order in concentration of aggregation and breakage, i. e., second and first order, respectively, and the macromixing between regions that either pronounce breakage (impeller region) or aggregation (bulk zone) changing the solid volume fraction implies a complicated interplay between linear and non-linear phenomenon.

In [7] static light scattering was used for measuring two integral properties of the CMD. These were the mean radius of gyration defined as

$$\langle R_g^2 \rangle = \left[\int_0^\infty c(x,t) x^z R_g(x)^2 dz \right] \left[\int_0^\infty c(x,t) x^z dz \right]^{-1} \quad (6)$$

where $R_g(x)$ is the radius of gyration of gyration of an aggregate of mass x that for $x > 3$ is given by Eq. (1); for smaller aggregates the values given in [14] where used. Further, it was a mean aggregate mass given by the zero angle scattering intensity:

$$I_0 = \left[\int_0^\infty c(x,t) x^z dz \right] \left[\int_0^\infty c(x,t) x dz \right]^{-1} \quad (7)$$

In Eqs. (6) and (7) z is a light scattering exponent that in the limit of vanishingly small primary particles and vanishingly tenuous aggregates assumes $z = 2$. For the compact aggregates and the large primary particles considered here z is expected to be substantially smaller than 2; its actual value is determined by fitting it to the experiments. In the present model, Eqs. (6) and (7) are applied to compute $\langle R_g^2 \rangle$ and I_0 within each zone. Volume averaged quantities are then compared with the experiments.

Fig. 1 shows the mean radius of gyration (Fig. 1a) and the zero angle intensity (Fig. 1b) at steady state as a function of the solid volume fraction at a stirring speed of 200 rpm (circles) and 600 rpm (triangles). Symbols represent experimental data whereas the dashed line represents calculations assuming the stirred tank is homogeneous (single zone model). For these calculations, the volume averaged flow properties reported in [7] where used. Model parameters were $q = 0.55$, $C_s = 8 \cdot 10^3 (\text{m}^2 \text{s}^{-3})^q$, and $z = 1.55$. The solid lines represent calculations from the three zone model with the flow properties

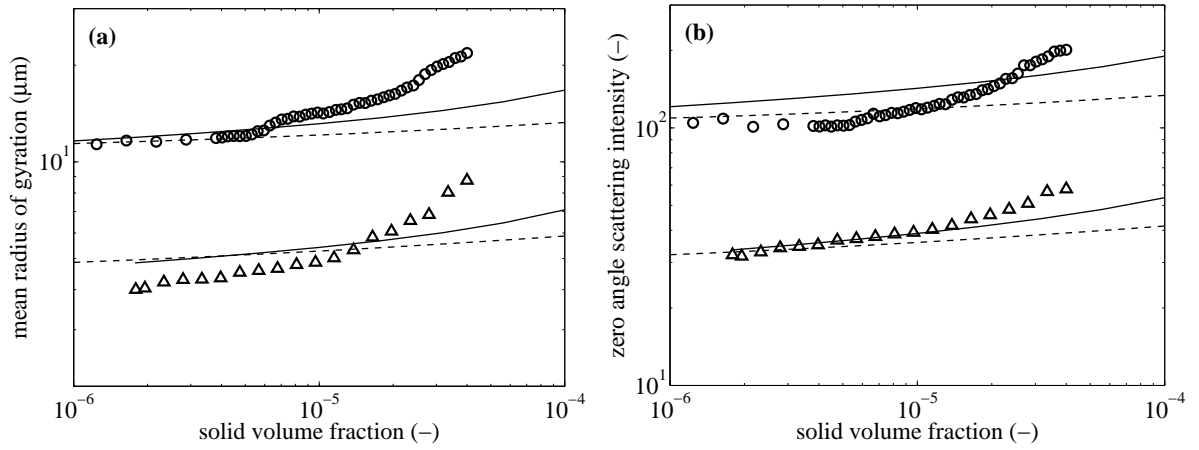


Figure 1: Mean radius of gyration (a) and zero angle scattering intensity (b) as a function of the solid volume fraction for 200 rpm (circles) and 600 rpm (triangles). Solid lines show the three zone model whereas dashed lines show calculations assuming a homogeneous flow (single zone model).

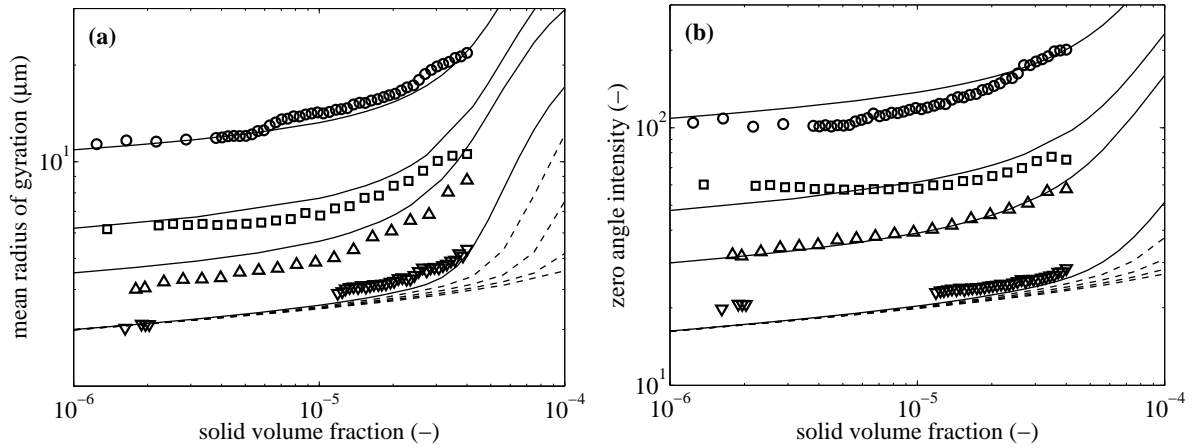


Figure 2: Mean radius of gyration (a) and zero angle scattering intensity (b) as a function of the solid volume fraction for 200 rpm (circles), 400 rpm (squares), 600 rpm (triangles up), and 1000 rpm (triangles down). Lines show calculations from the two zone model where the dashed line refer to (starting from below) to reducing the macromixing between the two zones. Solid lines refer to the final result with optimized parameters.

listed in Tab. 3. Model parameters in this case were $q = 0.60$, $C_s = 4 \cdot 10^5 (\text{m}^2 \text{s}^{-3})^q$, and $z = 1.56$. It is seen that the model in either version captures the general trend of the experimental data. In particular the variation in the stirring is well described (which is supported by experiments at even higher stirring rate not shown here). However, the increase in the aggregate size with increasing the solid volume fraction observed for both measured quantities is poorly predicted by the model. However, the difference between the single zone model (dashed lines) and the three zone model (solid lines) indicate that this increase is due to macromixing between the more vigorous flow regions and the less vigorous regions. An ensemble of aggregates that is circulating between these regions can grow while it is in the less vigorous bulk region whereas it undergoes breakage while in the vigorous impeller region. At a larger solid volume fraction the aggregation rate is higher and thus the aggregates grow larger while in the bulk region. The non-linearity of the aggregation and breakage rate lead then to the observed behavior of the aggregate size observed in the experiments.

This interpretation receives further support when studying the two zone model where the volume flux between the zone is a fitting parameter. Fig. 2 shows the results from these calculations. Model parameters are $q = 0.58$, $C_s = 1.3 \cdot 10^5 (\text{m}^2 \text{s}^{-3})^q$, and $z = 1.55$. Regarding the volume flux between the two zones, it was found that reducing Q_{12} ($=Q_{21}$) leads to a stronger increase of the aggregate size with increasing solid volume fraction. This is illustrated for the experiment at 1000 rpm (triangles down) where the dashed lines show calculation where Q_{12}/ND^3 decreases from 0.077 (lowest dashed curve) to 0.029 (most upper dashed curve). Accordingly, the solid lines refer to calculations with a relative low volume flux, i. e., $Q_{12}/ND^3 = 0.008$; only for the experiment at 1000 rpm (triangles down) a slightly larger volume flux of 0.020 was used.

To conclude, our analysis shows that the increase in aggregate size with increasing solid volume fraction in an aggregation-breakage process is controlled by macromixing. Aggregates that circulate between the intense and calm regions of the flow undergo either breakage or aggregation, respectively where increasing ϕ pronounces the latter. Predicting the macromixing from geometric flow maps where the zones are defined based on the reactor internals (impeller, baffles) is insufficient. Future work therefore has to address a method to identify the small volumes where the flow is very intense and their exchange rate with the bulk flow.

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