

Development of the Cluster-Size Distribution in Flowing Suspensions

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The complexities involved in modeling the simultaneous formation and disintegration of clusters or aggregates in suspensions are well documented in the literature. It is known that breakup can occur in conjunction with aggregation once the suspension is mechanically stirred or undergoes continuous flow, even during sedimentation. Works related to the microscopic nature of aggregation and breakup in such cases have typically centered on detailed analyses of the motion of individual particles or clusters as they approach other particles or clusters to form larger ones—a phenomenon observed in sedimentation, shear, and turbulent flows.

Macroscopically, the problem constitutes an overall study of the development of the cluster-size distribution by simultaneous coagulation and breakup. Typical analytical approaches to such have been through the population balance equation (for example, Elminyaw et al., 1991) and statistical means (Cohen, 1990, 1991a). While the generality of the former method must be appreciated, it is recognized that obtaining solutions requires inserting certain empirical parameters related to the rates of breakup and coagulation, in addition to other unknowns, into the kernels of the integro-differential equation. On the other hand, the latter approach assumes a purely random process, which allows it to bypass the need for these assumptions. One, however, would expect this approach to become valid when certain interactions, such as DLVO (based on a theory by Derjaguin, Landau, Verwey, and Overbeek), are overwhelmed—for example, by exposing the suspension to rapid mechanical mixing, as discussed briefly in Cohen (1992) and/or when the particles are relatively massive.

There exist many publications dealing with the development of aggregates in closed systems. Upon incorporating inflow and outflow, which would subsequently classify the system as open or continuous flow, significant complications in predicting the cluster-size distributions arise. Once again, typical analytical methods have utilized the population balance equation, an example of which was used by McGrady and Ziff (1988) for the special case of pure fragmentation.

Clearly, all processes involving simultaneous formation and disintegration of clusters possess, at any given time, both sto-

chastic and “orderly” characteristics. If these processes were totally random, then for practical purposes, a single population balance equation, having a universal set of coefficients and kernels, would suffice to solve all problems of this nature. Nevertheless, knowing that the two behaviors (order and disorder) can co-exist, it would be useful to draw a fine line differentiating between the two, since only then the mechanisms at work might be identifiable. As of yet, except for a handful of investigations that provide lengthy descriptions based on visual observations, none has aimed at achieving this goal. A quantitative approach, therefore, could present an important step toward a better understanding of the fundamentals involved in the simultaneous coagulation and breakup of clusters.

This work focuses on two objectives. First, it is to prove that the statistical model proposed by Cohen (1990, 1991a), which is based on closed (or batch) systems, can be extended to open-flow systems, provided that the coagulation-breakup process is random. Second, it is to relate the theoretical predictions with the experimental data obtained from some open-flow experiments to demonstrate the co-existence of stochastic and orderly behaviors, as well as to quantitatively pinpoint and explore their regimes of manifestation.

Problem Formulation

The details of the *evolution* of the cluster-size distribution by simultaneous and random coagulation and breakup in batch systems are discussed by Cohen (1991a). Of importance is the result that

$$p(i) = \frac{1}{e^\mu - 1} \frac{\mu^i}{i!} \quad (1)$$

where i is the aggregate size, $p(i)$ is the size-distribution probability defined as:

$$p(i) \equiv \frac{N_i}{N} \quad (2a)$$

and N_i and N , respectively, are the *total* number of clusters of size i , and the *total* number of clusters in the system—the two being related by:

$$N = \sum_i N_i \quad (2b)$$

Furthermore, μ is the distribution-controlling parameter, which is obtainable from:

$$\frac{N_o}{N} = \frac{\mu}{1 - e^{-\mu}} \quad (3)$$

Finally, N_o , which is given by

$$N_o = \sum_i i N_i \quad (4)$$

is the *total* number of primary particles in the batch system. Since the above are derived for closed systems, N_o remains constant, while N_i and N vary as the cluster-size distribution evolves over time.

It should be pointed out that in addition to the size-distribution probability defined in Eq. 2a, the weight distribution probability, $W(i)$, which is equivalent to the probability of finding a particle in a cluster of size i , is expressible by:

$$W(i) \equiv \frac{i N_i}{N_o} \quad (5)$$

Upon combining Eqs. 1–5, it is easy to prove that:

$$W(i) = \frac{1 - e^{-\mu}}{e^{\mu} - 1} \frac{\mu^{i-1}}{(i-1)!} = e^{-\mu} \frac{\mu^{i-1}}{(i-1)!} \quad (6)$$

Hence, to calculate the instantaneous distribution according to this model, one must: (i) know N_o ; (ii) choose some value of N representing the total number of clusters at any instant in time prior to reaching steady state; (iii) obtain μ from Eq. 3; and (iv) substitute it into Eq. 1 or 6, whichever is being sought. Simply stated, Eqs. 1 and 6 represent the *most probable* distribution probabilities at any instant in time in a random coagulation-breakup process, given N and N_o .

Concerning the predicted distribution functions presented above, an important observation is that they are characterized by the single parameter μ , which itself is a function of only the ratio N/N_o (see Eq. 3). Thus, if we let \forall be the total volume of the closed system, μ then becomes a function of the ratio c/c_o where c and c_o are the number *concentrations*. These are given by:

$$c \equiv N/\forall \quad (7a)$$

and

$$c_o \equiv N_o/\forall \quad (7b)$$

Now, owing to the fact that the ratio c/c_o has no explicit dependence on \forall , it is an “intensive” or “point” property,

independent of system size and configuration. In view of this, the distribution characteristic, μ , which can be evaluated from

$$\frac{c_o}{c} = \frac{\mu}{1 - e^{-\mu}} \quad (8)$$

(obtained upon combining Eqs. 3, 7a, and 7b) is also an intensive property of the local region defined by the concentration ratio, c/c_o .

Upon applying these arguments to a suspension of particles undergoing continuous flow or sedimentation, it is easy to visualize that at any instant in time, the ratio c/c_o has some measurable value ($0 < c/c_o \leq 1$) at any small region of the system. It, therefore, follows that μ can be determined *locally* using Eq. 8, from which the local and instantaneous distribution probabilities as given by Eqs. 1 and 6, can be obtained provided, of course, that the coagulation-breakup process is random. Obviously, for nonrandom processes, the above equations are not expected to hold.

Of interest, also, is that the local concentration ratio, c/c_o , has all the effects of flow rate (related to stirring intensity) and solids volume fraction manifested in it. Hence, one needs not to worry about the details of the flow field and the rate of change of the size distributions due to different solids volume fractions and/or flow conditions. Rather, knowing only the instantaneous local value of c/c_o , the distribution probabilities can be calculated locally.

Finally, comparison of the predictions of the random model with actual experiments can help assess the degree of randomness (or order) in the data. What follows next is the application of the proposed model to expose and identify orderly and chaotic behaviors in two unrelated systems of large particles, one which is continuous flow and the other, sedimentation.

Comparison with Existing Continuous Flow and Sedimentation Experiments

As mentioned above, a model for random coagulation and breakup of clusters in stirred systems, which is the simplest case when both occur simultaneously, has already been proposed (Cohen, 1990, 1991a). Inherent in the assumption of randomness is that the model may be more suitable to situations where certain interactions, such as DLVO and/or viscous, are negligible in comparison to other physical forces, for instance, inertia and shear mixing. In turn, DLVO interactions can become significant when agitation is at the thermal level, acquiring energy comparable to κT , where κ is Boltzmann's constant and T is the absolute temperature (Cohen, 1992), whereas viscous interactions dominate when the intensity of turbulence (on the length scale of the particle size) is weak.

Experimental measurements of cluster-size distributions have typically taken into account very small particles. Although the literature contains many works dealing with micron- and sub-micron-size particles undergoing thermal-induced aggregation and breakup in batch systems (Cahill et al., 1987; Pefferkorn and Stoll, 1990), few have experimented in great detail with cluster-size distributions in continuous flow and sedimenting systems.

In addition, a few existing studies of the development of the cluster-size distributions employing large particles in open systems are not only limited in number, but also confined to being

Table 1. Letter designations for the different experiments of Graham and Bird (G&B) (1984) and Chen et al. (1991).

Reference	Reynolds no., Re^*	Solids holdup, ϕ^{**}	Letter designation	Concentration ratio, c/c_o^\dagger	μ^\ddagger
Exp. 1 of G&B (1984)	0.0110	0.01	A	0.822	0.406
Exp. 2 of G&B (1984)	0.0272	0.01	B	0.840	0.359
Exp. 3 of G&B (1984)	0.0421	0.01	C	0.861	0.308
Fig. 5a of Chen et al. (1991)	3.16	0.0056 (0.0054)	D	0.942	0.120
Fig. 5b of Chen et al. (1991)	3.16	0.0094 (0.039)	E	0.934	0.139
Fig. 5c of Chen et al. (1991)	3.16	0.019 (0.039)	F	0.830	0.384
Fig. 5d of Chen et al. (1991)	3.16	0.028 (0.065)	G	0.788	0.498
Fig. 6a of Chen et al. (1991)	0.662	0.019	H	0.899	0.218
Fig. 6b of Chen et al. (1991)	1.0	0.019	I	0.864	0.300
Fig. 6c of Chen et al. (1991)	3.16	0.019	J	0.832	0.381
Fig. 6d of Chen et al. (1991)	5.8	0.019	K	0.905	0.203
Fig. 6e of Chen et al. (1991)	10.9	0.019	L	0.922	0.163

*In Chen et al., $Re = 2aU_t/\nu$, whereas in G&B, $Re = \dot{\gamma}a^2/\nu$.

**Solids volume fractions in parentheses are the three-dimensional values extrapolated by Chen et al. from 2-D.

†The concentration ratios, c/c_o , were obtained directly from the experimental plots of $W(i)$ vs. i using Eq. 10.

‡Values of μ were calculated from Eq. 8 using the experimental concentration ratios, c/c_o .

predominantly experimental. This is probably because there is a need for such data in the literature, and the conventional analytical methods for dealing with problems involving simultaneous aggregation and breakup have generally been too complicated. Nonetheless, some experimental data do exist (for example, Graham and Bird, 1984; Chen et al., 1991).

Regarding these two works, it is important to bear in mind that they are macroscopically very different. For instance, Graham and Bird (1984) have looked at simultaneous coagulation and breakup in continuous shear flow, whereas Chen et al. (1991) have studied the phenomenon more or less from the standpoint of two-dimensional sedimentation. The two works, however, are similar in that they have implemented particles with sizes ranging from $a = 3.2$ mm to 1.25 mm (where a is the particle radius) as used by Graham and Bird and Chen et al., respectively. These are considerably larger than the sub-micron- and micron-sized ones that have been typically utilized so that Brownian effects for all practical purposes are non-existent.

The relevance of these two to this work lies in the detailed gathering of data (by photographic techniques) at some local region in the flow field and their representation as weight distribution probabilities, $W(i)$, as defined in Eq. 5 and in the sentence preceding it. The experimental results of Chen et al. (1991) are available in Figures 5a–5d and 6a–6e of that reference, while those of Graham and Bird (1984) are provided in Table 1 of that work.

We should emphasize, however, that although Table 1 of Graham and Bird (1984) includes a total of six experiments, numbered 1–6, only experiments 1–3 will be of use here. The reason is that these experiments (1–3) utilize a *dilute* dispersion of 100% opaque spheres, thereby the corresponding data presented are directly applicable to our definition for $W(i)$, which, as mentioned above, is conveniently given by Eq. 5. Another useful note is that Reynolds number, Re , in Graham and Bird (1984) is defined as $\dot{\gamma}a^2/\nu$, where $\dot{\gamma}$ is the shear rate and ν is

the kinematic viscosity of the continuous phase, while $Re = 2U_t a/\nu$ in Chen et al. (1991), where U_t is the terminal velocity of a particle.

Data Analysis

By virtue of Eqs. 2b, 7a and 7b, the concentration ratio, c/c_o , is expressible by:

$$\frac{c}{c_o} = \frac{\sum_i c_i}{c_o} \quad (9)$$

Therefore, using Eq. 5, this can be written as:

$$\frac{c}{c_o} = \sum_i \frac{W(i)}{i} \quad (10)$$

Hence, given the experimental weight distribution data, $W(i)$ vs. i , belonging to a local region, the concentration ratio, c/c_o , associated with that region can be calculated from Eq. 10.

Once the experimental c/c_o is known, μ can be computed from Eq. 8, and upon substituting it into Eq. 6, the theoretical weight distribution based on the random model can be obtained. Ultimately, this can be related with experiment. It is important to note that, in this way, comparison of the random model with experiments involves *no* empirical or adjustable parameters, whatsoever.

Using Eq. 9, the values of c/c_o were extracted from experiments 1–3 of Graham and Bird (1984), and Figures 5a–5d and 6a–6e of Chen et al. (1991), all of which are in terms of $W(i)$ vs. i . Subsequently, the corresponding values of μ were calculated from Eq. 8, and the results in Table 1 of this work, where each experiment has been designated by an alphabetic letter.

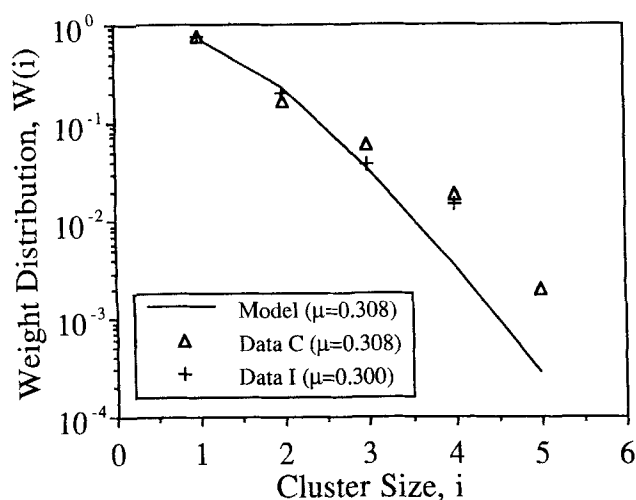


Figure 1. Comparison of random model with experimental data C and I.

It is evident from Table 1 that conditions such as Reynolds number and solids volume fraction affect the concentration ratio, c/c_o , and, therefore, μ . In this work, how c/c_o , and subsequently μ , are influenced by the experimental conditions is not of concern. Rather, any given region can be characterized by a certain measured concentration ratio, c/c_o , which has built into it all the local manifestations of these variables. Clearly, the simplicity that arises from such an analysis (that is, probability distributions being dependent only on the local concentration ratio, c/c_o , while c/c_o depends on the system variables) offers the advantage that results of different systems and experiments can be compared more easily.

A log-linear plot of $W(i)$ vs. i , comparing the theoretical prediction of the stochastic model with some arbitrarily chosen data, say, experimental data C and I (corresponding, respectively, to experiment 3 of Graham and Bird and Figure 6b of Chen et al., see Table 1 of this work for letter designations),

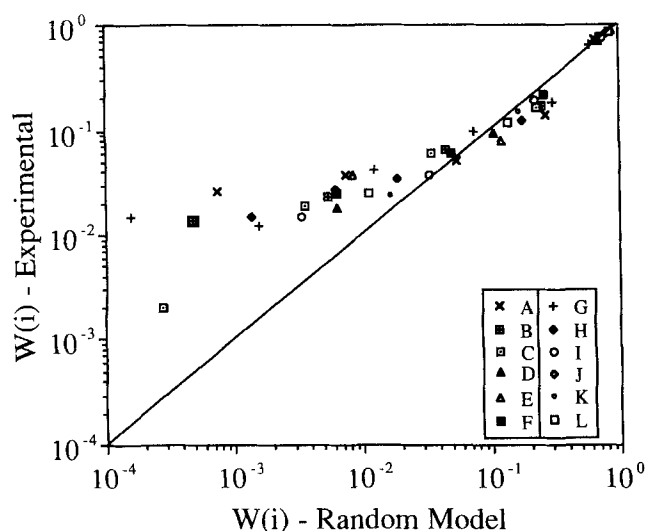


Figure 2. Overall comparison of data with random model.

Note that the deviation from randomness occurs at $W(i) \approx 0.05$, indicating a more orderly behavior.

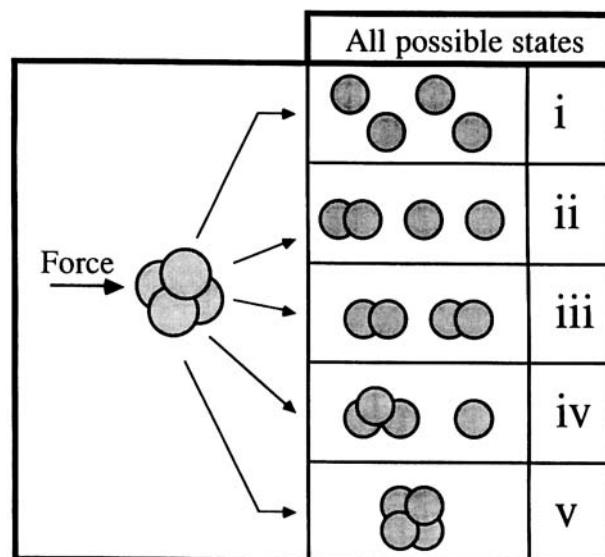


Figure 3. All available or possible states upon breakup of a cluster of size $i=4$.

is provided in Figure 1. Because the theoretical curves for $\mu=0.300$ and 0.308 lie indistinguishably close to each other, only one line is depicted. The agreement appears to be very good for cluster sizes up to and including $i=3$, but deviations start to show at $i \geq 4$. Since the behavior in Figure 1 is typical when comparing the model to the remainder of the data in the two references, no other similar plots have been included here in the interest of space. Instead, all data have been combined into one graph, which is shown in Figure 2, where the experimental $W(i)$ is plotted against the theoretical $W(i)$ over several orders of magnitude.

Here we observe that deviations from randomness begin to appear at $W(i) \approx 0.05$. This corresponds to cluster sizes of 4 and greater, as shown in Figure 1. At this time, one can conclude within reason that clusters of sizes 1, 2 and 3 form and disintegrate in, more or less, a random fashion, while clusters of size 4 and greater exhibit some degree of order (since they deviate from the expected stochastic behavior). Interestingly, in both systems, which are totally unrelated, the line distinguishing between chaos and more orderly behavior lies at $i=4$ [or $W(i) \approx 0.05$].

While currently we are not in a position to conclude with confidence as to what is causing the formation and disintegration of clusters of size $i=4$ and greater to deviate from randomness, we speculate that the fluid mechanics are playing a major role. To explain, Figure 3 displays *all* states available upon breakup of a single cluster (in this case, size $i=4$) subjected to some force (or impact) of a given magnitude. We note that there are a total of five states possible subsequent to applying the force. For convenience, these are numbered (i)–(v). State i is complete shattering, whereas states ii–v signify partial or no fragmentation. Note that state v also suggests that the force was not enough to break the cluster.

Now, a stochastic assumption requires that *all* possible or available states (all 5 states for $i=4$, as shown in Figure 3) be equally accessible upon fragmentation. In reality, however, subjecting a cluster of particles to a limited force, be it shear or pressure fluctuation, does not assure equal accessibility to

all these states. This is because the magnitude of the force may not be sufficient to shatter the cluster into smaller fragments (such as states i and ii in Figure 3), whereas achieving all other states that comprise larger fragments might be possible. Consequently, total randomness would require forces of sufficient magnitudes to access all fragmented states with equal probabilities, which means that as the cluster size increases, the number of available states increases, and so should the force. An argument similar to the above has also been used to model the breakup of liquid drops (Cohen, 1991b).

Likewise, for coagulation, which again is representable by Figure 3, but with the direction of the arrows (pointing to the states) reversed, randomness implies that a cluster of any size ($i=4$ for the case shown in Figure 3) must be equally attainable starting from any initial fragmented state (it is important to note that a cluster of size i can also form by the breakup of a larger cluster). In this case, the effects opposing coagulation, as the particles come together, are brought about by the viscous interactions (see, for example, Happel and Brenner, 1965), which must be counteracted by high-intensity turbulence. Therefore, the more the number of fragments in the initial state, the more the turbulence intensity needed to overcome these viscous interactions for coagulation to take place successfully.

In short, it is the multiplicity of the available states, coupled with the fact that not all are equally accessible (because of limited hydrodynamic forces) that makes deviations from the stochastic behavior more likely to occur in larger clusters. As for monomers, the presence of only one available state offers the least leeway for any deviation (related to multiple states) from the predicted behavior. Perhaps this is why, at least for the data examined here, the agreement between model and experiment is so good as $i=1$ (see Figure 1, and also the cluster of points, representing monomers, nearest to the top-right-hand corner in Figure 2). More specifically, maximum deviations between experiments and the model at $i=1$ reach only about 6%, and these occur at the highest values of μ (lower c/c_o) belonging to data A and G. The remaining data, nonetheless, show significantly smaller differences.

Upon returning to Figure 2 and looking at data A, B, and C of Graham and Bird (1984), which correspond to Reynolds numbers 0.0110, 0.0272, and 0.0421, respectively, we note that the experiments approach the random model prediction as the Reynolds number is increased. This implies that randomness in coagulation and breakup is indeed approached as stirring rate is increased, a consequence of which might be that high-intensity turbulence in the length scale of the particles could ultimately lead to the predicted stochastic behavior—a conclusion that follows directly from the arguments presented above. Evidently, even though the relatively low rates of sedimentation and shear in the two sets of data examined here are not sufficient to impose a stochastic effect on larger clusters, indications are that they do tend to break and form smaller clusters at random.

Comparison of the Random Model with the Theory of Coagulation by Binary Collisions

In compliance with the nomenclature adopted here, initiation and development of the cluster-size distribution by binary

coagulation, based on Smoluchowski's theory, can be summarized as:

$$W(i) \equiv \frac{ic_i}{c_o} = i \left(\frac{c}{c_o} \right)^2 \left(1 - \frac{c}{c_o} \right)^{i-1} \quad (11)$$

where

$$\frac{c}{c_o} = \frac{1}{1 + t/t_c} \quad (12)$$

In the above, t is time and t_c is the characteristic time of coagulation, which is expressible by $3\hat{\mu}/4\kappa Tc_o$ ($\hat{\mu}$ is the viscosity) if the process is Brownian. Equation 12 also implies that at $t=0$, $c/c_o=1$, which denotes a fully dispersed initial condition. Moreover, Eqs. 11 and 12 are applicable to any similar process, not necessarily Brownian, provided that it is governed by the binary collision mechanism having some characteristic coagulation time, t_c .

Figure 4 compares the results of the binary collision theory with those of the random model. Also included are the experimental points, which will be discussed shortly.

Regarding the two models, they approach one another in the limit $\mu \ll 1$, which corresponds to $c/c_o \rightarrow 1$ (see Eq. 8). This indicates that the onset of coagulation, if one starts with a fully dispersed initial state ($c/c_o=1$, $\mu=0$), is sparked in a random manner. The reason for this is that since all particles at time $t=0$ are identical, in a homogeneous system they are all subjected to identical conditions. Hence, the number of dimers forming at $t=0^+$ must certainly be governed stochastically. Nevertheless, with further coagulation and decreasing c/c_o (increasing μ), deviations become larger (compare $\mu=0.120$ and 0.498 with the diagonal line in Figure 4) as the constraints imposed by binary coagulation with no breakup begin to dominate.

Concerning the data in Figure 4, we find that they also tend to diverge strongly from the binary collision model. Interestingly, however, they seem to lie closer to this theory than to the random model (Figure 2). This is surprising because the

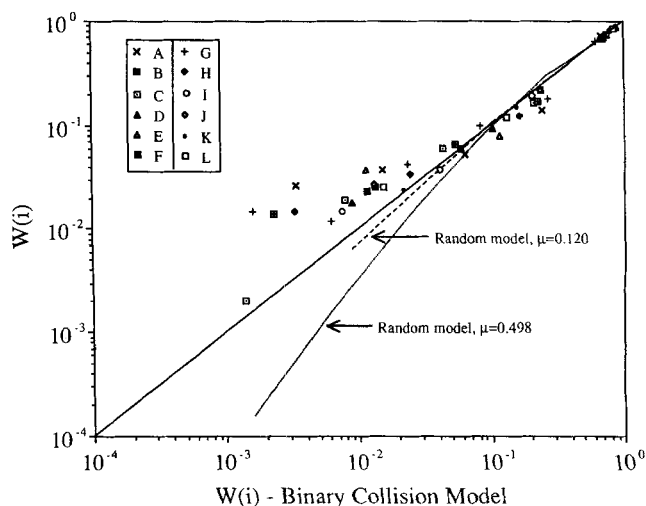


Figure 4. Comparison of binary collision theory with random model and experimental data.

binary collision theory disregards any cluster dissociation, whereas visual observations have confirmed its presence in these experiments. A probable explanation for the better agreement is that the overall blend of order and disorder in the two experiments may, on average, cause the distributions to mimic more closely the patterns predicted by coagulation due to binary collisions.

Finally, as for the time scale of transition from chaos to the behavior expected from the binary collision model, we can designate a certain small μ , say 0.2, based on Figure 4, to provide us with this information. By virtue of Eq. 8, therefore, we obtain $c/c_0 \approx 0.91$, which corresponds to $t/t_c \approx 0.1$ upon using Eq. 12. Hence, if coagulation by binary collisions were to begin in a fully dispersed system, the time it takes to deviate from the initial chaotic behavior and to achieve the characteristics unique to the binary-collision theory should be on the order of $0.1t_c$.

Discussion and Conclusion

Analytical modeling of the formation and disintegration of clusters of particles in flowing suspensions has always been and remains to be extremely complicated. One can attest to this by simply following the step-by-step description of the dynamics of the process (Chen et al., 1991). In general, the overall behavior results from a combination of both randomness and order. However, descriptions based on visual observations alone not only fail to provide any information on which it is dominant, but also do not differentiate between the two.

In this work, through a model that predicts the evolution of cluster sizes by random coagulation and breakup in suspensions, some data related to continuous flow systems and sedimentation have been examined with the hope of achieving these tasks. We should mention, however, that extension of the model from batch to open-flow systems was possible because the predicted distribution probabilities are intensive properties, which allow them to be compared with samples obtained locally or regionally, such as by photography.

Comparison of the data with predictions of the model points clearly toward the co-existence of chaos and order. More specifically, it indicates that, for the set of data examined here, deviations from random behavior occur at lower values of the weight distribution probability, $W(i)$, corresponding to larger clusters. This implies that smaller clusters are more likely to be influenced by stochastic effects, whereas larger ones are not. As to the cause of these deviations, our speculation, based on reasons discussed earlier, is that limited stirring intensities cannot access equally all the available states involved in cluster formation and disintegration.

The model was also compared briefly with the theory of coagulation by binary collisions. Here we found that the two approach each other in the limit $\mu \ll 1$, which suggests that, beginning with a fully dispersed initial condition, coagulation due to binary collision is limited by random behavior.

In conclusion, the work presented here proposes a method for distinguishing between randomness and order in a given system, and to pinpoint their respective regions. The outcome of this could, therefore, imply that the population balance equation, which remains to be the tool of choice because of its great flexibility, should probably be inspected more closely to account for both stochastic and orderly effects when both coagulation and breakup are involved.

Acknowledgment

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