Dispersion by Laminar Flow

Dispersion was studied by subjecting clusters of spherical particles (20–400 μm) to laminar flow fields. The clusters were: cohesionless, consisting of particles suspended in the same liquid as the bulk medium and cohesive for which a different liquid was used to make the suspension. Both qualitative and quantitative results are presented.

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SCOPE

In many industrial processes, such as the pulping of wood fibers and the dispersing of fillers in polymers, mixing of solid particles in a liquid phase must be effected. These particles initially cluster together in flocs or aggregates and are subjected to a flow field which separates the particles and eventually forms a homogeneous suspension. Such mixing was studied using model systems consisting of solid spherical particles suspended in a high viscosity fluid (silicone oil) at large volume fractions (0.6). This suspension was formed into small spherical particle clusters which were dispersed in various well-defined laminar flows, e.g., simple shearing and extensional, of the same silicone fluid. Such clusters were non-cohesive in the sense that the particles were large enough (>20 μm) that the attractive forces between them were insignificant and since the same fluid was used for the suspending and bulk media, no surface tension forces were present. The experiments were performed using a four-roller and a Couette device with the dispersion being recorded using a video system. The influence of flow kinematics, particle size, initial cluster size and particle concentration was examined.

Mixing of cohesive clusters was also investigated in the same types of flows and bulk medium. These clusters were formed by suspending spherical particles in water to which a surface active agent had been added. Because of the finite surface tension between the water solution and the silicone oil the dispersion process is fundamentally different than in the case of the cohesive clusters and qualitative features of this process were demonstrated.

CONCLUSIONS AND SIGNIFICANCE

For cohesionless clusters of spherical particles, the experimental results indicate that a simple model, valid for short times, which predicts that the non-dimensional cluster radius is linearly related to a non-dimensional time (Eq. 4) adequately describes the dispersion process. Interpreting the slope of the data thus plotted as a "dispersion rate", it was found that this rate was significantly influenced by the flow type with the extensional flow having the greatest rate, all other parameters being equal. This rate was also strongly dependent upon the ratio of the particle radius to the initial cluster radius, with changes of two-thirds in this parameter changing the dispersion rate by a factor of 50 in some cases.

In the presence of interfacial forces between the cluster and the bulk phase and under the same flow conditions as used for the cohesionless clusters, break-up was much more difficult to obtain except when the interfacial tension was low. Generally, the clusters behaved as immiscible liquid drops whose viscosities were governed by the particle concentration. At high concentration, the clusters were deformed slightly by the hydrodynamic forces, but, no break-up was observed. As the concentration was decreased, the deformation would increase until conditions were reached (depending upon surface tension, extensional rate, etc.) under which the cluster would break-up. The debris from this process consisted of smaller particle clusters containing one or more individual particles. Every such cluster was completely encased in the suspending medium with no particles being separated from the suspending fluid. For an individual particle detached from the parent cluster, the volume of the sheath of suspending fluid surrounding it was often such as to increase the particle concentration of the main cluster after detachment. Thus, the break-up process could be self-stabilizing. In addition, it was found that the suspending fluid alone could be depleted in the main cluster by the detachment of individual drops. This also increases the particle concentration in the cluster and inhibits dispersion.

This work provides valuable background information to other research aimed at examining the mixing of fiber flocs in turbulent flow fields which have large extensional or shearing components. Current theoretical and experimental work on mixing of colloidal systems can use the theoretical model to predict *a prior* parameters that appear in their theories permitting more effort to be focused upon understanding those phenomena unique to colloidal systems. The approach used here can also serve as a basis for studying the mixing of particulate fillers in polymer fluids.

INTRODUCTION

The role of mixing in many industrial processes has long been recognized and over the last several years, many fundamental studies of it have been performed. In turbulent flows (Uhl and Gray, 1966; Brodkey, 1973), such investigations examine how the flow field affects the process of dispersion, carefully distinguishing between fine scale mixing and diffusion. The former, being effected solely by the flow field, results in a substance comprised of two or more materials which, at some scale, are separate (Brodkey, 1973). The latter consists of the smoothing out of local variations in composition at the smallest length scales.

Studies of mixing play an important role in the design of dispersed phase reactors (Valentas et al. 1966) for which mathematical models can be developed that require a knowledge of the dynamics of the breaking and coalescence of liquid drops (Acrivos and Lo, 1978; Barthes-Biesel and Acrivos, 1973; Grace, 1971; Rallison and Acrivos, 1978; Rumscheidt and Mason, 1961; Torza et al., 1972). Turbulent mixing of colloidal particles has been the subject of recent research especially in the area of flocculation and deflocculation phenomena (Firth, 1976; Firth and Hunter, 1976; van de Ven and Hunter, 1976; van de Ven and Mason, 1976, 1977). This fun-

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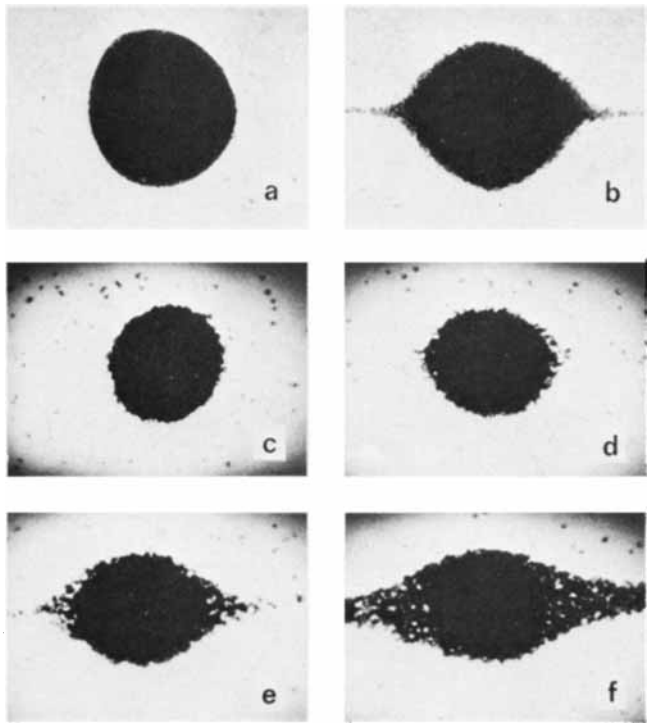

Figure 1. Break-up of clusters in pure shear (1a and 1b) and simple shear (1c–1f) flows. Figure 1a: Cluster is approximately spherical at short times. Figure 1b: As time progresses, spheres migrate along the surface away from its vertical plane of symmetry to its horizontal one. There, particles accumulate forming nodes from which individual particles detach, and are convected away. Figure 1c: In simple shear, at short times cluster behaves as a rigid sphere rotating at the local angular velocity of a fluid element. Figure 1d: In the first stage of break-up particles detach from the surface and rotate in the closed streamline region near the cluster. The process continues (Figure 1e) with more particles detaching until the entire closed streamline region is filled (Figure 1f).

damental research was used by Spielman (1978) to develop a theory of mixing in agitated vessels. Solid-liquid dispersion is also important for understanding mixing of solid particles, such as mineral tailings and chopped glass fibers, in highly viscous liquids especially polymer melts [Kim and Han, 1976]. Here, the flow is always laminar however since the liquid is non-Newtonian, new problems are introduced.

In systems having three phases, a solid phase with a sheath of liquid A surrounding it immersed in a second, immiscible liquid B, mixing means creating a suspension in which the solid and the sheath liquid coexist but are physically separate. Many such three phase systems exist in materials which are processed industrially, such as Athabasca tar sands (Beckowitz and Speight, 1975) and filled polymer melts with a coupling agent [Han et al., 1981].

The purpose of this paper is to examine dispersion phenomena in model systems. In section (B) we discuss a simple mathematical model to describe experiments on dispersion processes similar to those reported in short note by Kao and Mason (1975). In those experiments, well-defined flow fields were used to disperse aggregates of spherical particles suspended in a fluid which was the same as that used to make the suspension (cohesionless clusters). Here (section D.1) we extend their results by examining the effect of flow strength, particle size and concentration on the dispersion process. Several experiments are reported in section (D.2) that qualitatively examine dispersion in three phase systems: suspensions of solid spheres in liquid A dispersed in liquid B (cohesive clusters). The interfacial tension between liquid A/liquid B is a critical factor in these experiments and the effectiveness of several surface active agents is discussed.

MODEL

If a material, A, is placed in a quiescent fluid, B, and the two substances are immiscible, both phases will remain separate. But, it is possible that agitation of the bulk phase (B) will cause break-up or mixing of phase A in B to occur. In general, developing a model capable to describe this dispersion under all flow conditions is difficult, however, by simplifying the types of materials and flow fields being considered, it is possible to derive equations which predict qualitative features of the phenomena.

TABLE 1

Material	Density g/cm³	Average Radius μm
Lucite	1.15	55
Polystyrene	1.0	29
Polystyrene	1.0	85
Polystyrene	1.0	200

For the system considered here phase A consists of a concentrated suspension molded into a spherical shape. This suspension is prepared by placing solid spherical particles, having radii greater than 20 μm, in the same liquid as that used for phase B. The break-up of the clusters of particles is effected in well-defined laminar flow fields, called linear two-dimensional flows, which have the form (Kao et al., 1977)

$$U_1 = 0 \qquad U_2 = \lambda G X_3 \qquad U_3 = G X_2; \qquad -1 \le \lambda \le 1 \quad (1)$$

where G is the "strength" of the flow and λ is a dimensionless parameter which delineates the particular type of flow under consideration. For example, $\lambda = -1$ is pure rotational flow, $\lambda = 0$ is simple shear flow and $\lambda = 1$ is pure shear flow.

In order to have a better understanding of the phenomena being discussed, Figures 1a and 1b show the progression of the dispersion process in pure shear flow. Initially (Figure 1a), the cluster is spherical, having radius R_o, and all particles are attached to the cluster. As time, t, progresses (Figure 1b) particles are carried along the surface by the incoming flow, away from the top and bottom of the cluster reaching the horizontal where the flow is away from the cluster. Here small nodes form from which individual particles detach and convect away. The net result of this process is that the average radius, $R = R(t)$, continuously decreases with time [Kao and Mason, 1975] until total disintegration occurs. As can be seen from Figures 1a and 1b, during much of the experiment, except for the small region near the nodes, the aggregate remains essentially spherical. At the latter stages of break-up, it assumes an elongated shape.

For the flows having non-zero vorticity, cluster disintegration proceeds somewhat differently, as illustrated in Figures 1c–1f for the case of simple shear flow ($\lambda = 0$). Initially (Figure 1c), the aggregate is spherical, rotates with the local angular velocity of the fluid and behaves as a rigid sphere in simple shear flow (Kao et al., 1977). As the experiment progresses (Figures 1d and 1e), particles detach from the cluster and move near to it in closed orbits, a result of the closed streamlines known to exist near the solid sphere. By this detachment process, the closed streamline region eventually becomes filled with particles (Figure 1f). Then, either hydrodynamic particle-particle interactions or a further decrease in the size of the cluster causes particles in the closed streamline region to move across the limiting streamline separating this region from the

TABLE 2. SURFACTANTS USED IN EXPERIMENTS ON CLUSTER BREAK-UP

Name	Suppliers
Lithium Sterate	Matheson, Coleman and Bell
Polyoxyl (40) Sterate	Atlas Chemical Industries
OP-10 (Polyoxyethylene Octylo-phenyl ether)	Nippon Surfactant Co. Ltd.
Lux Dishwashing Detergent	Lever Detergents Ltd.
TRS-1080	Witco Chemical
Petrostep 465	Stepan

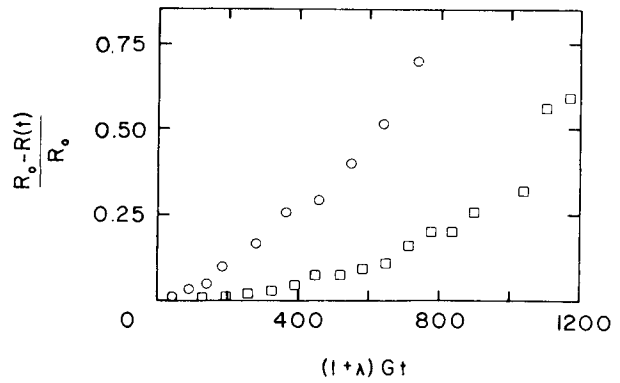

Figure 2. Effect of flow type on the dispersion of cohesionless clusters. The circles (○) are for pure shear flow ($\lambda = 1$), the squares (□), for simple shear flow ($\lambda = 0$). The rate of dispersion is decreased by the presence of vorticity in the simple shear flow.

open streamline region where particles are convected away from the cluster.

A simple model of the process described above was proposed by Kao and Mason (1975) who assumed that the rate at which particles leave the surface of the cluster is proportional to the surface area of the cluster. Using the same starting point it is possible to obtain several conclusions about the disintegration process from this model which were not found previously.

Calling $N = N(t)$ the surface-average number of particles which have detached from the cluster since the initiation of an experiment, the model described above is formulated as

$$\frac{dNR^2}{dt} \propto \left(\frac{R}{a}\right)^2 \tag{2}$$

where a is the radius of an individual sphere from which the cluster is formed. Since $a^3\,dNR^2/dt$ is proportional to the rate of decrease of the volume of the aggregate, dR^3/dt, the relationship 2 becomes

$$\frac{dR/R_0}{dt} = -\hat{k}\left(\frac{u_{i,j}}{(1+\lambda)G}, c, \frac{a}{R_o}\right)\frac{a}{R_o}. \tag{3}$$

Here, $\hat{k}$ is the proportionality constant which gives the influence of the local velocity gradient, $u_{i,j}$, and volume fraction of particles, c, on the dispersion process. For Eq. 3 to hold, the cluster must remain spherical so that its overall dimensions are characterized only by the radius, R. Based upon the appearance of the clusters shown in Figure 1, it is considered that this assumption is valid during the initial stages of an experiment. Also, to obtain Eq. 3, buoyancy and inertial effects are neglected.

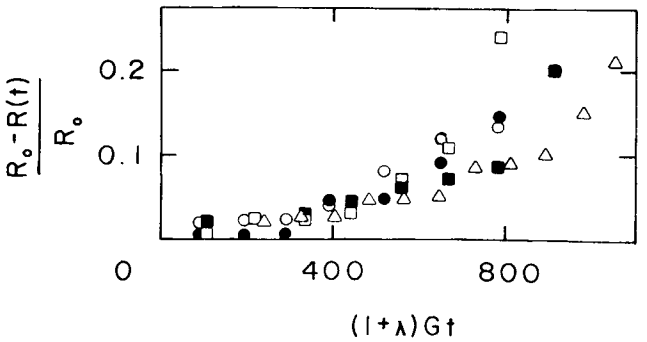

Figure 3. Effect of varying the flow strength, G, on the dispersion of cohesionless clusters in simple shear flows ($\lambda = 0$). Volume fraction and radii of particles and initial radius of the cluster are fixed. The symbols correspond to flow strengths as follows: (○), (●)-G = 1.3 s^{-1}; (□), (■)-G = 3.7 s^{-1}; (△)-G = 0.54 s^{-1}. To within experimental error, changing the flow strength is not found to modify the dispersion process.

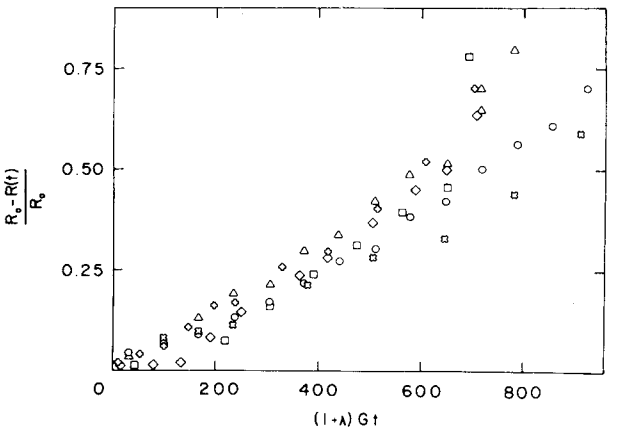

Figure 4. Effect of varying the flow strength, G, on the dispersion of cohesionless clusters in pure shear flows ($\lambda = 1$). Volume fraction and radii of particles and initial radius of cluster are fixed. The symbols correspond to flow strengths as follows: (○)-G = 0.11 s^{-1}; (□)-G = 0.29 s^{-1}; (△)-G = 0.17 s^{-1}; (✣)-G = 0.45 s^{-1}; (⊠)-G = 0.68 s^{-1}. To within experimental error, changing the flow strength is not found to modify the dispersion process.

Equation 3 can be solved to give

$$\frac{R_o - R(t)}{R_o} = k\left(\frac{u_{i,j}}{(1+\lambda)G}, c, \frac{a}{R_o}\right)\frac{a}{R_o}(1+\lambda)Gt \tag{4}$$

where k is a dimensionless constant. (In this model, k is a constant to be determined experimentally, although a mechanistic approach to this problem should yield explicit predictions of k.) Based upon Eq. 4, several experimentally verifiable predictions can be made which will be discussed in the *Results* section.

EXPERIMENTAL PROCEDURE

A four-roller device in which the speed and direction of each roller could be varied independently (Giesekus, 1962; Kao and Mason, 1975) was used to generate all linear two-dimensional flows except simple shear for which a Couette apparatus described by Bartok and Mason (1975) was used.

The cohesionless clusters were formed from suspensions in which the liquids were Dow 200 series silicone oils having viscosities of 10 N·s/m^2 and 20 N·s/m^2. The characteristics of the spherical particles used in this study are given in Table 1.

The cohesive clusters were made by suspending polystyrene spheres in a water solution. The solutions were produced using distilled water and a variety of surface active materials to modify the interfacial tension (Table 2). All experiments were performed in pure shear flows ($\lambda = 1$) effected in a 10 N·s/m^2 silicone oil.

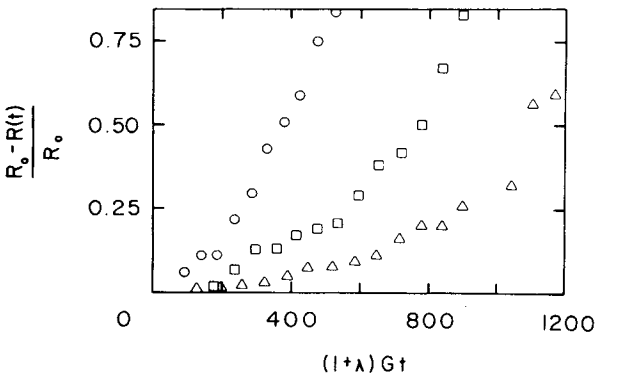

Figure 5. Effect of varying the ratio of the particle radius, a, to the initial cluster radius, R_o, on the dispersion process for simple shear flows ($\lambda = 0$). Volume fraction of particles and flow strength are both held fixed. The symbols correspond to values of a/R_o as follows: (○) a/R_o = 0.0275; (□) a/R_o = 0.02; (△) a/R_o = 0.0165. Increasing a/R_o (e.g., by decreasing the initial radius) is found to strongly influence cluster break-up. A 2/3 increase in a/R_o increases the dispersion rate by a factor of 60.

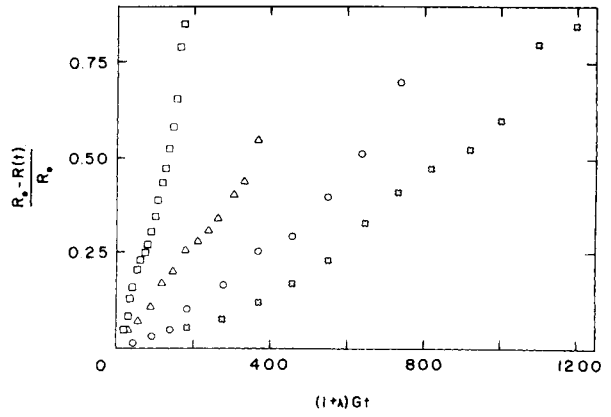

Figure 6. Effect of varying the ratio of the particle radius, a, to the initial cluster radius, R_o, on the dispersion process for pure shear flows ($\lambda = 1$). Symbols correspond to values of a/R_o as follows: (□) $-a/R_o = 0.027$; (△) $-a/R_o = 0.023$; (○) $-a/R_o = 0.018$; (⊠) $-a/R_o = 0.014$. Increasing a/R_o is found to strongly influence cluster break-up.

A Sony television camera combined with a Panasonic Time Lapse Video Tape Recorder NV-8030 was used to record the break-up of the clusters. In most experiments, the contrast between the cluster and the surrounding medium was increased by bright field illumination. However, direct observation of individual particles was facilitated with a dark field implemented by illuminating the cluster from the side with a Ziess KL150B fiber optics light source and two separate fiber bundles. Recorded signals were played back and the radius of cluster was measured directly from the television screen. These measured radii were related to the actual radii of the cluster by calibrating the camera-lens system for the various magnifications that were used.

RESULTS AND DISCUSSION

Cohesionless Clusters

The analysis presented in section B suggests several experiments. One of these has been previously reported by Kao and Mason (1975) who showed the dependence of the break-up process on the flow parameter λ for $0 \leq \lambda \leq 1$. They found that $R_o^3 - R^3/R_o^3$ is linear with $(1 + \lambda)Gt$. Similar data have been obtained here in the

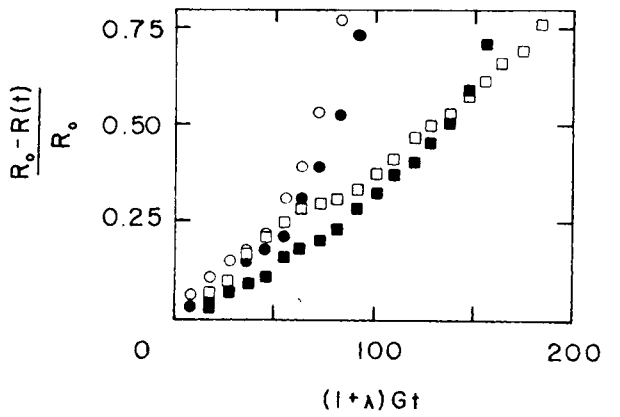

Figure 7. Effect of varying the particle radius, a, and the initial cluster radius, R_o, while keeping a/R_o fixed at 0.0285. Data were obtained using pure shear flow ($\lambda = 1$) at $G = 0.5\ s^{-1}$. Filled and unfilled circles represent two sets of data for 200 μm polystyrene spheres and squares, two sets for 55 μm PMMA spheres. For small values of $(1 + \lambda)Gt$, the curves are linear and coincide to within experimental error. At larger $(1 + \lambda)Gt$, the curves diverge reflecting the large deformation from sphericity observed for the clusters composed of 200 μm spheres.

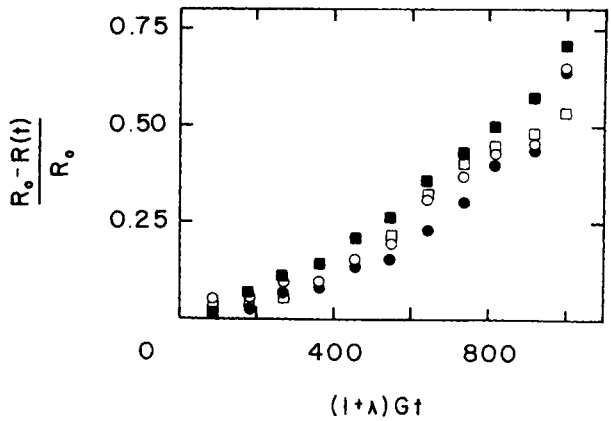

Figure 8. Effect of varying the particle radius, a, and the initial cluster radius, R_o, while keeping a/R_o fixed at 0.014. Data were obtained using simple shear ($\lambda = 0$) flow at $0.5\ s^{-1}$. Filled and unfilled circles represent two sets of data for 85 μm polystyrene spheres and squares, two-sets for 55 μm PMMA spheres. Up to $(1 + \lambda)Gt = 600$ the curves coincide and are linear to within experimental error. At large non-dimensional times, the linear behavior is no longer found, and the curves diverge.

case of simple shear ($\lambda = 0$) and pure shear ($\lambda = 1$) flows and also for flows having $-1 \leq \lambda < 0$. The results for $\lambda = 0$ and $\lambda = 1$ flows were obtained with c, R_o and a fixed and are plotted in the form suggested by Eq. 4, viz., $R_o - R/R_o$ vs. $(1 + \lambda)Gt$ in Figure 2. There is a linear region of the curves for small values of $(1 + \lambda)Gt$ and the slope of this region is a function of λ. For larger values of $(1 + \lambda)Gt$, the curve is non-linear implying that the assumptions which are implicit in the derivation of Eq. 4 are no longer valid.

For flows having $\lambda < 0$, the spherical cluster behaves differently than those having $\lambda \geq 0$. Theoretically, for a neutrally buoyant liquid droplet or solid sphere in a fluid undergoing a flow having $\lambda < 0$ there are no open streamlines (Kao et al., 1977). For cluster break-up, this implies that while particles can "detach" from the surface, they cannot be convected away from the parent cluster. It was found experimentally that in $\lambda < 0$ flows, particles move away from the parent cluster where there is a high number density of spheres to regions where the number density is lower. This process creates a situation in which the boundary of the cluster does not remain well-defined and although the number of particles in the parent cluster decreases with time, this decrease cannot be determined by the methods being used in these experiments.

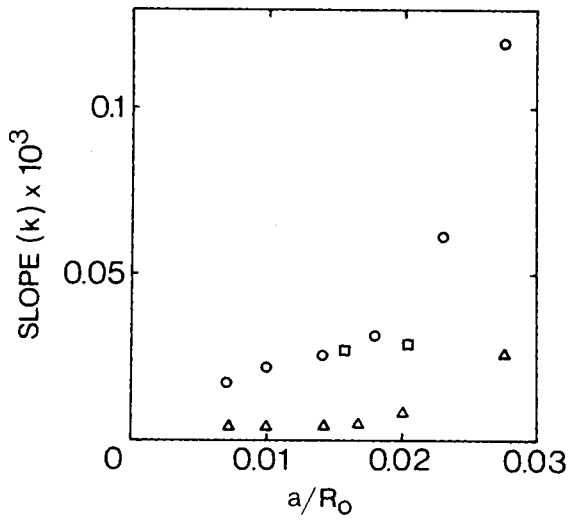

Figure 9. Dependence of the rate of dispersion, k, on the ratio of the particle radius, a, to the initial cluster radius, R_o in pure shear flows ($\lambda = 1$) ((○) and (□)) and simple shear flows ($\lambda = 0$) (△). The data are independent of shear-rate and the volume fraction is 0.6 for the circles and triangles and 0.65 for the squares.

Figure 10. Periodic extension and compression of particle cluster in simple shear flow ($\lambda = 0$). The volume fraction of particles is 0.5. The same fluid as the bulk medium is used to make the suspension from which the cluster is formed causing the surface tension between the cluster and the bulk to be zero. The cluster thus behaves as a drop with zero surface tension but of higher viscosity than the surrounding medium, initially spherical (Figure 10a), first extended (Figure 10b), then compressed back to its spherical shape (Figure 10c).

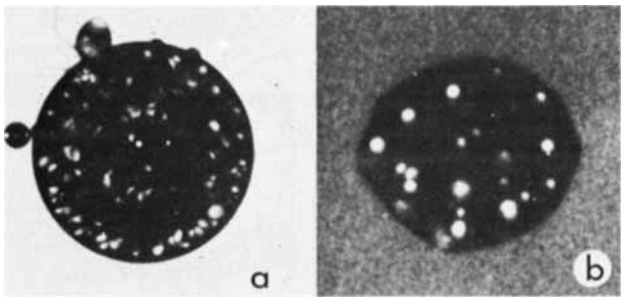

Figure 11. Particle clusters formed using an aqueous surfactant solution in a quiescent bulk medium of 10 N · s/m² silicone oil. At low particle concentrations, volume fraction below 0.55, a thin film of surfactant solution surrounds the cluster which assumes a spherical shape (figure 11a). At higher particle concentrations, individual spheres protrude into the silicone oil.

In the case of flows having $0 \leq \lambda \leq 1$, Eq. 4 adequately describes the break-up process at short times (Figure 2). Based upon Eq. 4, several other predictions can be made which can be verified by experiment. For example, keeping a, c, λ, and R_o fixed, Eq. 4 predicts that the dispersion curves should be independent of G. This was examined for $\lambda = 0$ and $\lambda = 1$ flows and the results are shown in Figures 3 and 4. For $\lambda = 0$, (Figure 3) data were obtained at three different shear-rates; and for $G = 1.3$ and $3.7\ s^{-1}$, two sets of data were obtained to determine the reproducibility of the results. The data superimpose up to values of $(1 + \lambda)Gt$ of 700 and are roughly linear with $(1 + \lambda)Gt$. The breakdown of the linear relationship and the superposition of the data at larger values of $(1 + \lambda)Gt$ indicate that Eq. 4 is no longer valid.

Similar observations can be made for pure shear flows ($\lambda = 1$), (Figure 4). Here, up to $(1 + \lambda)Gt = 600$ the data obtained at five different values of G superimpose and are linear with $(1 + \lambda)Gt$.

The other parameters which determine the form of the dispersion curve include the particle radius, a, and the initial cluster radius, R_o. The dependency upon a and R_o was examined by keeping c, λ and G fixed and varying a/R_o, both by varying the particle size (using the various particles described in Table 1) keeping R_o fixed and by varying R_o keeping the particle size fixed. Experiments were performed using $\lambda = 0$ and $\lambda = 1$ flows and the results are shown in Figures 5 and 6, respectively. For $\lambda = 0$, a 2/3 increase in a/R_o (i.e., a decrease in the initial radius by 40%) strongly affects the break-up process with the slope of the linear region of the curve increasing by a factor of 60. This same effect is found using $\lambda = 1$ flows (Figure 6), where a decrease in the cluster radius by 50% increases the slope by a factor of 10.

From these experiments it was concluded that the dispersion process depended upon the ratio a/R_o in a systematic way and that if a/R_o were fixed while a and R_o were varied, similar dispersion curves would result. These results agree with Eq. 4 which predicts that in the linear region of the $(R_o - R)/R_o$ vs. $(1 + \lambda)Gt$ curve, the dispersion process depends only on the ratio a/R_o.

This prediction was further tested using all particles described in Table 1 and varying R_o so as to maintain the ratio a/R_o fixed. Two values of a/R_o were used and the experiments were performed in a $\lambda = 1$ flow at fixed G. The results for $a/R_o = 0.0285$ are shown in Figure 7. The curves coincide to within experimental error for all values of $(1 + \lambda)Gt$ less than 60. Beyond this, the cluster composed of 85 μm spheres experienced large deformations from the initially spherical shape, and the final stages of break-up consisted of a long extension of the cluster rather than the detachment of single spheres. Similar observations can be made in the case where $a/R_o = 0.014$ (Figure 8), the main difference being that the dispersion curves coincide up to a value of $(1 + \lambda)Gt$ of 800.

Using the results of all the experiments in which the effects of particle size and initial radius were examined, the dependency of the constant k (Eq. 4) upon the parameter a/R_o can be ascertained. This is shown in Figure 9 for $\lambda = 0$ and $\lambda = 1$ flows. In both cases, if a/R_o is small, there is a linear relationship between k and a/R_o. For values of a/R_o greater than 0.018, there is a stronger dependency of k upon a/R_o. Over the entire range of a/R_o, the value of k obtained in $\lambda = 1$ flows is 3–4 times greater than that found in $\lambda = 0$ flows. These observations relate only to the initial stages of break-up and no attempt has been made to deal with the final stages when effects due to the deformation of the cluster, uneven distribution of particles or a small particle to cluster diameter ratio may become important.

Experiments were also performed in which the volume fraction

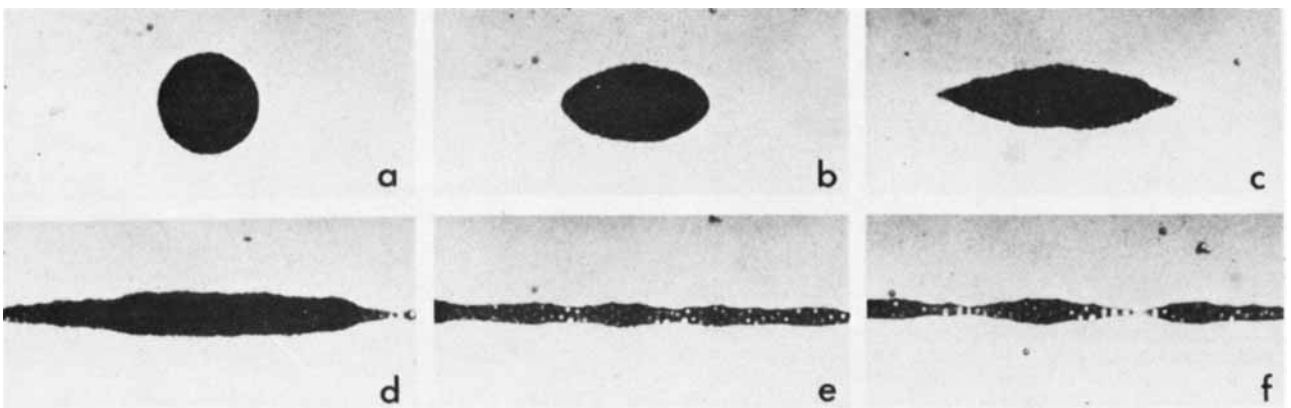

Figure 12. Deformation and break-up of a cluster of spherical particles in an aqueous surfactant (TRS 1080) solution in pure shear flow effected using 10 N·s/m² silicone oil. The cluster is initially spherical (Figure 12a), deforming into an ellipsoidal shape (Figure 12b) and then being pulled into a cylinder (Figures 12c and 12d). The diameter of the cylinder decreases and necking occurs along its length (Figure 12e). The narrow regions, one particle wide and four or five long, eventually rupture (Figure 12f), creating small clusters.

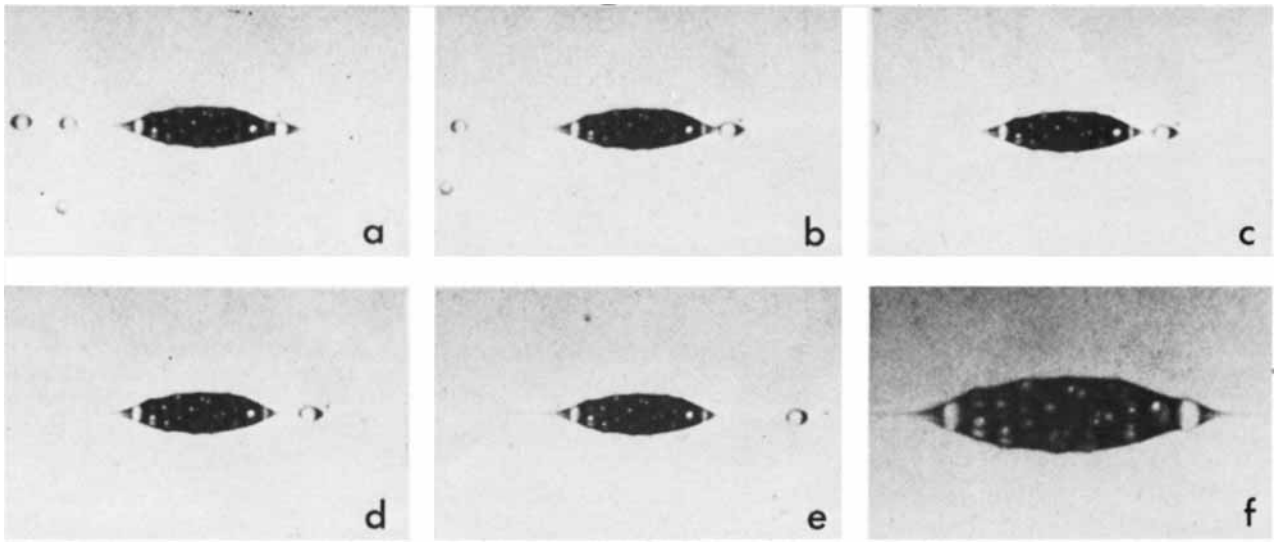

Figure 13. Detachment of a single particle from the parent cluster. The flow and suspension are the same as those used in Figure 12. The detaching particle is initially positioned at the right node of the cluster (Figure 13a). As the particle moves away from the cluster (Figures 13b and 13c) it remains connected to it by a thin thread of solution which also surrounds the particle. This thread becomes unstable and breaks (Figure 13d) allowing the sheathed particle to be convected away (Figure 13e). A close-up of the final, stable configuration (Figure 13f) of the cluster shows the liquid threads extending from its nodes.

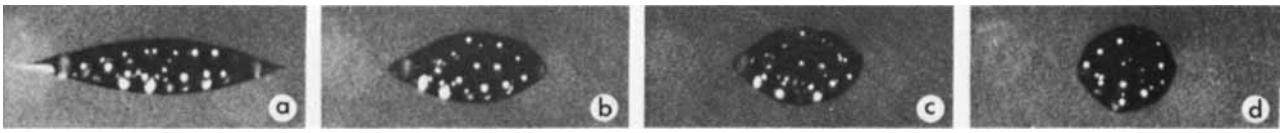

Figure 14. Recoil of a deformed cluster upon cessation of flow. The flow and suspension are the same as those used in Figures 12. After partial break-up, a stable unit is formed (Figure 14a) which has a higher volume fraction of particles than its parent cluster due to depletion of the liquid caused by its being stripped away at the nodes. Upon stopping the flow, the cluster is brought back to a spherical shape by surface tension (Figures 14b–14d) but in the unstressed state, it is observed to have individual particles protruding into the bulk medium (Figure 14d), which was not the case with its parent (Figure 11a).

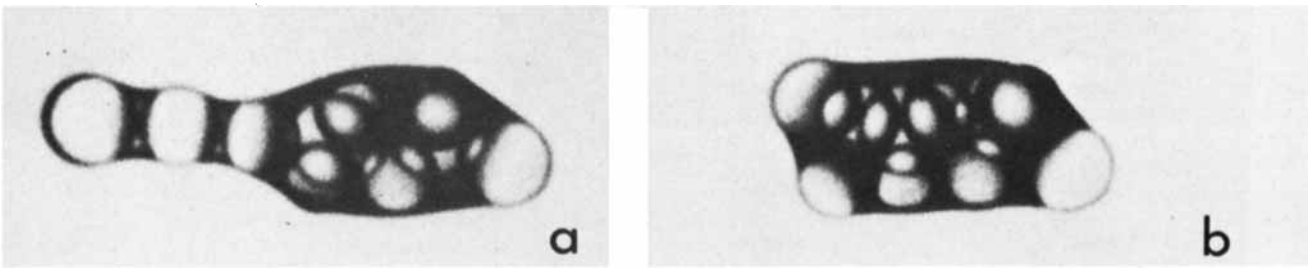

Figure 15. Recoil of a deformed cluster, highly depleted of its aqueous phase, upon cessation of flow. The flow and suspension are the same as those used in Figure 12. Under stress produced by flow, a linear chain of particles connected by bridges formed on the left side of the cluster (Figure 15a). When the flow is stopped, this chain is no longer stable, and surface tension attempts to reform a spherical array, but there is an inadequate amount of aqueous phase to allow sufficient mobility of the particles.

of particles, c, was varied. In all of the experiments thus far discussed, $c = 0.6$ was used. This was found to produce a sufficiently viscous suspension to permit molding the cluster into a spherical shape. Decreasing the particle concentration below 0.55 the clusters behaved like a liquid drop with zero surface tension. In simple shear flows ($\lambda = 0$), the cluster rotated as before, and simultaneously underwent a periodic extension and compression (Figure 10), similar to that observed by Torza et al. [1972] for liquid drops with no surface tension in simple shear flows. In pure shear ($\lambda = 1$) flows, the cluster continuously deformed into a thread eventually "rupturing". Since the clusters were not spherical during these experiments, even at short times, it was not possible to obtain meaningful measurements of R as a function of time. Two experiments in which the volume fraction of particles beyond 0.6, indicated that the rate of dispersion might be somewhat decreased below that found for $c = 0.55$ (Figure 9).

Finally, some insight into the mechanism of the dispersion process is obtained by considering the cluster as a solid sphere and examining the trajectory of a fluid particle near its surface. This motion is similar to that experienced by a spherical particle near the surface of the cluster. The governing equations are derived from the work of Kao et al. (1977) by expanding their results to obtain a solution valid near the sphere. The velocity component parallel to the surface of the sphere is found to be $0\,(a/R_o)$ while that normal to the surface is $0\,(a/R_o)^2$. Roughly speaking, this implies that k should be linear in a/R_o if the shearing motion acting on the aggregate causes the detachment and not the normal component of motion. According to the data presented in Figure 9 it appears that the tangential motion is effecting the dispersion process.

Cohesive Clusters

As mentioned in the Introduction, in many dispersion processes interfacial forces between the material to be dispersed and the bulk medium exist. Therefore, experiments with cohesive clusters were

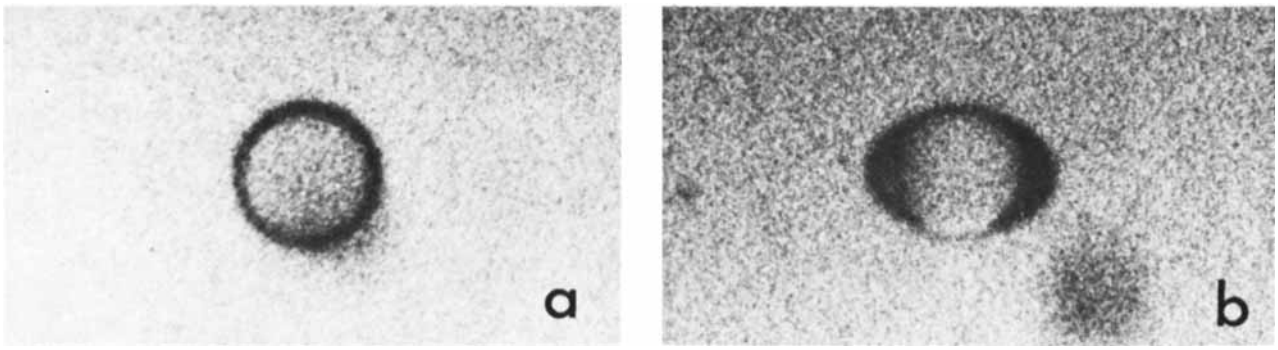

Figure 16. Deformation of liquid sheath surrounding a spherical particle in pure shear flow. The sheath is an aqueous surfactant (TRS 1080) solution immiscible in the bulk phase (10 N · s/m^2 silicone oil). When no flow is present, the sheathed particle appears to be a spherical, solid particle (Figure 16a). The presence of the sheath becomes evident when subjected to the stresses generated by the flow (Figure 16b). This configuration is stable and in pure shear flow the sheath could not be removed from the particle for values of flow strength up to 1 s^{-1}.

performed to understand qualitatively the effect of this interfacial tension on the laminar mixing process.

Markedly different behavior of the cluster was observed when the volume fraction of particles was above and below 0.55. For $c < 0.55$, a continuous layer of the water solution was observed at the suspension/bulk interface (Figure 11a), and, the interfacial forces caused the suspension to assume a spherical shape. At the higher concentrations, some particles protruded into the bulk medium (Figure 11b), and the cluster did not assume a spherical shape.

In the absence of surface active agents, the flow caused no discernable change in the configuration of the cluster at the highest particle concentrations. As the concentration was decreased, starting the flow caused some rearrangement in the internal structure of the cluster which eventually assumed a stable, ellipsoidal shape. At lower concentrations, the cluster deformed into a long cylinder which pinched at regular intervals along its length and broke into small clusters of particles. At all concentrations no individual particles were detached from the main cluster.

These observations might have been anticipated by considering the suspension as a liquid droplet the viscosity of which can be varied, without changing the surface tension, by changing the particle concentration. For liquid drops, the viscosity ratio of the two phases (Λ = droplet viscosity/bulk viscosity) is one of two parameters that determines drop deformation (Cox, 1969), the other being the surface tension. For Λ large (high particle concentration), there is little deformation, As Λ is decreased (by decreasing particle concentration), the deformation increases, pinching occurs and smaller droplets are formed (Acrivos and Lo, 1978; Rumsheidt and Mason, 1961).

The analogy between the behavior of the clusters and that of liquid droplets is extended by recalling that the drop deformation is a function of both viscosity ratio and a surface tension parameter (Cox, 1969). The effect of surface tension was examined using several surface active agents (Table 2) at concentrations above the critical micelle concentration. The results obtained using the first 4 surfactants listed in Table 2 were similar to those obtained when no surfactant was used, implying that these surfactants do not substantially change the surface tension at the water/oil interface.

Using the two surface active agents, TRS 1080 and Petrostep 465 listed in Table 2, which are known to produce very low surface tensions at the oil/water interface [Cash et al., 1977], several interesting phenomena were observed. At low particle concentration the cluster continuously deformed into a thread (Figure 12) having regions one particle wide and several particles long (Figures 12e and 12f). These particle-wide regions reached a critical length at which pinching occurred causing smaller clusters to form as in the cases discussed above. For suspensions having c roughly 0.55, upon starting the flow the cluster deformed with some particles detaching from it either one-by-one or in small groups (Figure 13). After this initial break-up, the cluster either stabilized and formed an ellipsoidal shape (Figure 13f) or proceeded to disintegrate by "rupturing." This allowed particles to be freely pulled away from the parent cluster similar to the break-up shown in Figure 12. Either of these two scenarios was possible for a given cluster. This was proved by stopping the flow after a cluster had reached a stable configuration, then restarting it and observing the "rupture" type of break-up. At very high particle concentrations, ($c > 0.6$) no break-up was found with the cluster behaving similar to the high concentration suspension made using water or the first four surfactants in Table 2.

At the micro-level other interesting features of the dispersion process were observed using the TRS 1080 and Petrostep 465 surfactants. For particle concentrations sufficiently low to observe cluster deformation, a thin thread of liquid extends from the cluster (Figure 13f) at the two points on its surface which experiences the maximum tensile stresses. Initially this thread extends far into the bulk phase causing a depletion of liquid in the cluster. This causes an effective increase in the volume fraction of particles in that region where a two-phase system exists, and is therefore a stabilizing influence. If these threads break-up, there is a decrease in the total volume of solution and when the flow is stopped, (Figure 14) a cluster which initially had a sheath of solution surrounding it will have spheres protruding through the surface (Figure 14d) reflecting the increase in volume fraction. An extreme case of solution depletion is shown in Figure 15. Under flow conditions, individual particles extended from the left side of the cluster, upon stopping the flow, (Figure 15b) there was not sufficient solution even for a nearly spherical configuration to be assumed.

Detachment of single particles from a cluster with surface tension is a fundamentally different process than the case discussed in the last section. In Figure 13a a particle on the right side of the cluster is positioned to commence detaching. It separates from the parent cluster remaining connected by a thread of fluid (Figure 13b). As the particle moves farther away (Figure 13c), the thread becomes smaller, finally rupturing (Figure 13d) with a sheath of solution remaining on the single particle, which is convected away (Figure 13e). [This detachment mechanism, due to the instability of an extending thread of liquid, has been examined by Mikami and Mason (1975).] A closer view of such a single particle surrounded by a water sheath in a quiescent fluid and in pure shear flow is given in Figures 16a and 16b, respectively. The flow causes the sheath to deform due to the stresses acting on it, however, for values of G up to 1 s^{-1}, it was not possible to separate the water from the sphere.

For these water/surfactant systems the deformation due to flow was a function of the initial radius of the cluster. If the cluster was sufficiently small, little deformation and no threads of fluid were observed. Following the analogy between the suspension and a liquid drop, this behavior is expected since not only does the surface tension govern the drop deformation, but also the drop radius (Cox, 1969).

NOTATION

a = particle radius
c = volume fraction

G = flow strength
k = proportionality constant (Eq. 3)
N = total number of detached particles per unit surface area
R_o, R = initial and time dependent cluster radius
t = time
X_1, X_2, X_3 = rectangular Cartesian coordinates
U_1, U_2, U_3 = components of undisturbed velocity field
u_i = components of velocity field with cluster present

Greek Letters

Λ = viscosity ratio
λ = dimensionless parameter delineating the flow type (Eq. 1)

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