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Clusters Formation during Sedimentation of Dilute Suspensions

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Abstract

Particle interactions in dilute monodispersed sedimenting suspensions of spherical particles are studied as a function of solid concentration. It is shown that in suspensions with solid concentrations below 0.83%, the interactions are too insignificant to effect the use of Stokes' law in sedimentation results. Beyond this concentration, however, a definite change in suspension behavior occurs, as particles come close enough to form clusters of varying sizes causing faster settling rates. Optimum clustering takes place around 4.5% solid concentration, corresponding to mean interspacing of 2.2 particle diameter within suspension and giving settling rates 1.58 times faster than the Stokes' velocity for a mean particle. Clusters start breaking beyond this concentration as the sedimentation becomes more hindered and the return upward flow of liquid becomes increasingly tortuous. The probability of clusters formation and their stability as a function of particle size, concentration, and the Reynolds number of suspensions are also investigated. The studies are further extended to demonstrate the effect of "immobile" liquid within the clusters in interpreting the sedimentation results.

INTRODUCTION

In a solid-liquid suspension of low concentration, each particle effectively falls as it would at infinite dilution. Upon increasing the concentration, however, the interactions of the particles are influenced by the presence of other particles so that the direct use of Stokes' law in interpreting the sedimentation parameters of suspensions becomes invalid (*1*).

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Particle–particle interaction in dilute suspensions has been studied by numerous workers, and their consistent behavior of faster settling rates at a critical solid concentration range of 0.1–3.0% by volume, apparently caused by particle clustering, has been observed unanimously (*2–5*).

Hall (*2*) observed that in dilute suspensions the interactions between two equisettling spheres traveling at 2.6 diameters apart would cause their settling velocity to be 1.2 times the Stokes' terminal velocity (V_s) of the single sphere. Kaye and Boardman (*3*), Johne (*4*), and Koglin (*5*), working on dilute suspensions with a concentration range of about 0.05–3.0% by volume, have observed an increase of up to 1.5 times the Stokes' terminal velocity in sedimentation rates of the settling particles. Above this concentration the settling rate of the suspension falls rapidly until, at higher concentrations, hindered settling is the dominant feature (*4*), when the suspension settles "en block" with an interface above which is a clear supernatant liquid.

Cluster formation in dilution was also observed by Slack (*6*) and Bhatty et al. (*7, 8*) who reported various formations of clusters with three, four, five, six, or more particles. Also, the effects of clusters formation in dilute suspensions have been mentioned by the above-named authors, and by Bhatty et al. (*8*) in calculating the mean particle radii, but the explanation of clusters formation in settling suspensions is still unresolved.

The present studies are therefore aimed at elucidating the formation, stability, and behavior of individual clusters in dilute suspensions as they settle under viscous or turbulent conditions. The effects of particle size on clustering and their consequences on particle size analysis are also discussed. The studies are extended to consider the presence of the "immobile" liquid incorporated in the clusters and the effect on the determination of mean particle size in the suspensions.

MATERIALS AND METHODS

The materials used were glass ballotinis (seven different grades A–G) and 75% aqueous glycerol. Their specifications are given in Table 1.

The glass ballotini were obtained from the English Glass Co., Leicester, UK. Microscopic observation showed that almost all the glass ballotinis were smooth and regular and virtually free of gas bubbles. About 80–90% of the ballotinis were completely spherical, and of the nonspherical ballotinis, less than 2% were completely irregular.

The generation of clusters and their settling behavior were observed in a 95-cm tall and 5.5-cm wide graduated glass column. The column was

TABLE 1

	Glass ballotini, grades						
	A	B	C	D	E	F	G
Nominal radii (mm)	0.650	0.450	0.350	0.325	0.275	0.238	0.138
Mean density, ρ_s (g/cm^3)	2.938						
	Glycerol (75% aqueous)						
Density, ρ_l (g/cm^3)	1.202	both at 24°C					
Viscosity, η (g · cm/s)	0.403	both at 24°C					

first filled with a thoroughly mixed 75% aqueous glycerol and left overnight to equilibrate. A uniform monolayer of dry and free-flowing glass ballotini was then spread out on top of the still glycerol miniscus. The layer was gently pricked with a sharp tipped rod to release clusters falling individually into the column. The size of clusters would depend upon the force of the tip and concentration of the ballotini layer. Clusters behavior, as they settled in the column, was observed by using a powerful lens.

The observed settling rates of clusters V_{so} (cm/s), with each grade of glass ballotini, after correcting for the wall effects (9), are given in Table 2. The combination of clusters formation for various grades of particles and their relative stability during settling in the dilute suspensions are also calculated and given in Table 3.

A separate experiment was carried out to observe the variation of the settling rates of particles with increasing solid concentration in the suspension. Glass ballotini of nominal diameter 0.1 cm, obtained by sieving, were used in the experiment, and the suspension medium was again 75% aqueous glycerol. The concentration of suspension was increased by increasing the mass of glass ballotini by known amounts while the suspension volume was kept constant at 180 cm^3. The results are presented in Fig. 1. Each reading was the mean of at least 15 separate observations.

The complete series of experiments was carried out in a constant room temperature thermostatted to within very close limits to 24°C.

DISCUSSION

The plot in Fig. 2, relating the settling rate of a suspension of glass ballotini to the concentration C (grams of ballotini in 180 cm^3 total

TABLE 2
Observed Settling Rates V_{so} (cm/s) and Reynolds Numbers (Re) for Clusters of Various Grades of Glass Ballotinis

Cluster size and formation		Grades of glass ballotinis and their radii (mm)						
		A 0.650	B 0.450	C 0.350	D 0.325	E 0.275	F 0.238	G 0.138
1 Single	V_{so}	3.49	2.06	1.01	0.98	0.72	0.57	0.19
	Re	1.27	0.58	0.20	0.19	0.12	0.08	0.016
2 Double	V_{so}	3.86	2.50	1.40	1.15	0.90	0.80	0.25
	Re	1.48	0.77	0.32	0.24	0.17	0.14	0.024
3 Trigonal	V_{so}	4.65	3.30	1.90	1.40	1.20	1.00	0.30
	Re	1.96	1.17	0.51	0.32	0.26	0.20	0.032
4 Tetrahedral	V_{so}	5.00	3.85	2.35	1.70	1.35	1.15	0.40
	Re	2.18	1.47	0.70	0.43	0.31	0.24	0.049
5 Double tetrahedral	V_{so}	5.45	4.35	2.60	1.95	1.65	1.40	0.45
	Re	2.48	1.77	0.82	0.57	0.41	0.32	0.059
6 Octahedral	V_{so}			3.00	2.35	1.85	1.50	0.50
	Re			1.01	0.70	0.49	0.36	0.69
8 Single cubic	V_{so}					2.25	1.75	0.58
	Re					0.66	0.45	0.086

Re = Reynolds no. measured by using V_{so} in the formula.

suspension), exhibits the onset of a sharp rise at about $C = 1.50$ (0.83% solid concentration). The calculated number of particles present in the suspension at this concentration is 1000, which corresponds to a mean interparticle (surface to surface) spacing of 0.47 cm or 4.7 particle diameters. The maxima of the curve is at $C = 8.5$ (4.72% solid concentration), at which the settling rate of the suspension is 1.58 times higher than the Stokes' terminal velocity for a single particle. The mean surface-to-surface interspacing at this point is approximately 2.2 particle diameters. On increasing the particle concentration further, the settling rate decreases. The right-hand limb of the curve shows an inflection at about $C = 75.0$, which corresponds to the breaks in sedimentation curves noted by Ramakrishna and Rao (*11*) and identified by Bhatty et al. (*7*) and Davies (*12*) as the onset of true hindered settling in which particles in a

TABLE 3
Relative Stability of Clusters Calculated on the Basis of Their Observed Behavior. S = Stable clusters. B = Broken clusters

Grades of ballotini:	A		B		C		D		E		F		G	
Specific surface, cm^2/g:	(15.7)		(22.7)		(28.2)		(31.4)		(37.1)		(43.0)		(74.26)	
Total clusters observed	24		45		39		40		36		35		49	
No. of stable clusters	4 (17%)		11 (24%)		15 (38%)		18 (45%)		18 (50%)		20 (57%)		29 (59%)	
No. of broken clusters	20 (83%)		34 (76%)		24 (62%)		22 (55%)		18 (50%)		15 (43%)		20 (41%)	
Size of cluster	S	B	S	B	S	B	S	B	S	B	S	B	S	B
2	4	10	8	19	7	11	7	9	6	6	7	3	10	7
3	—	6	2	8	5	5	6	7	5	6	5	3	9	6
4	—	3	1	4	2	2	4	2	4	2	4	3	5	3
5	—	1	—	3	1	3	1	2	2	2	2	3	2	2
6	—	—	—	—	—	3	—	2	1	1	1	2	2	2
8	—	—	—	—	—	—	—	—	—	1	1	1	1	1

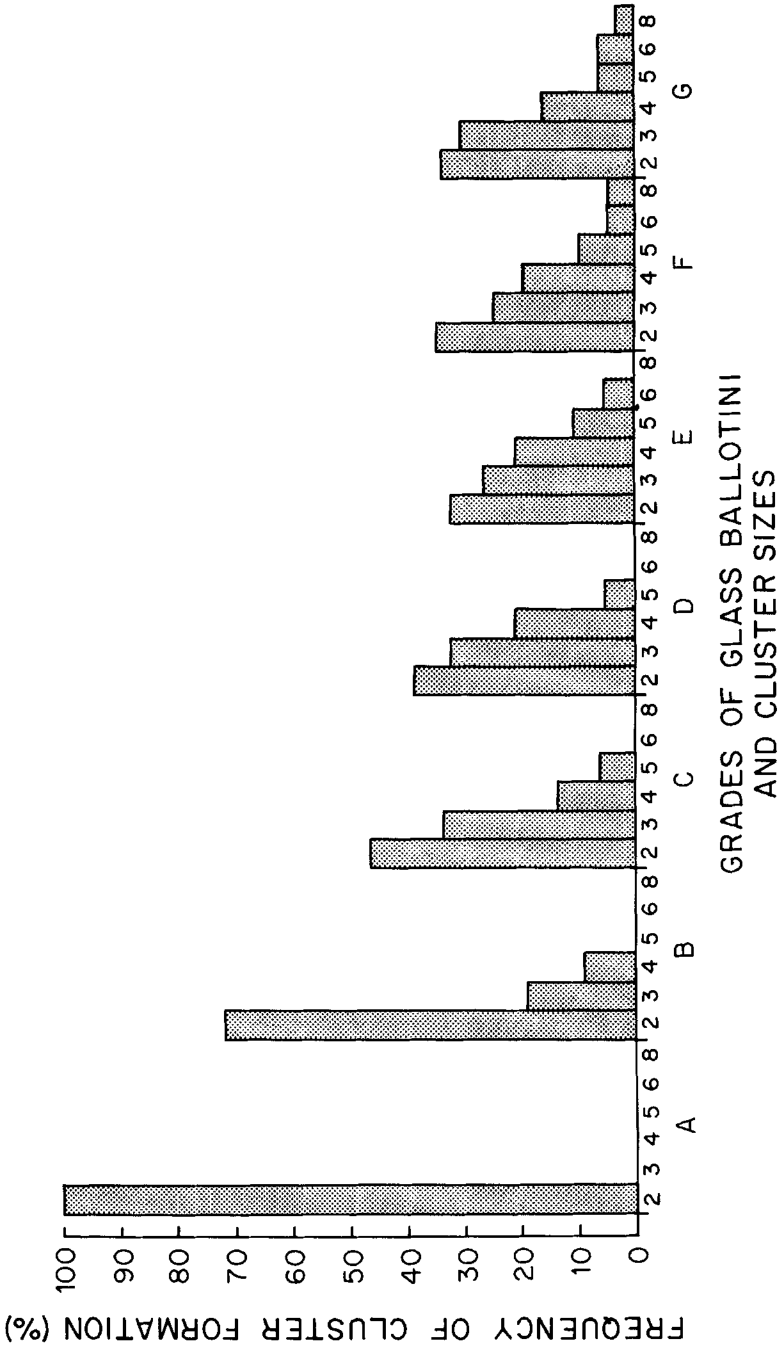

FIG. 1. Clusters formation as a function of glass ballotini grades.

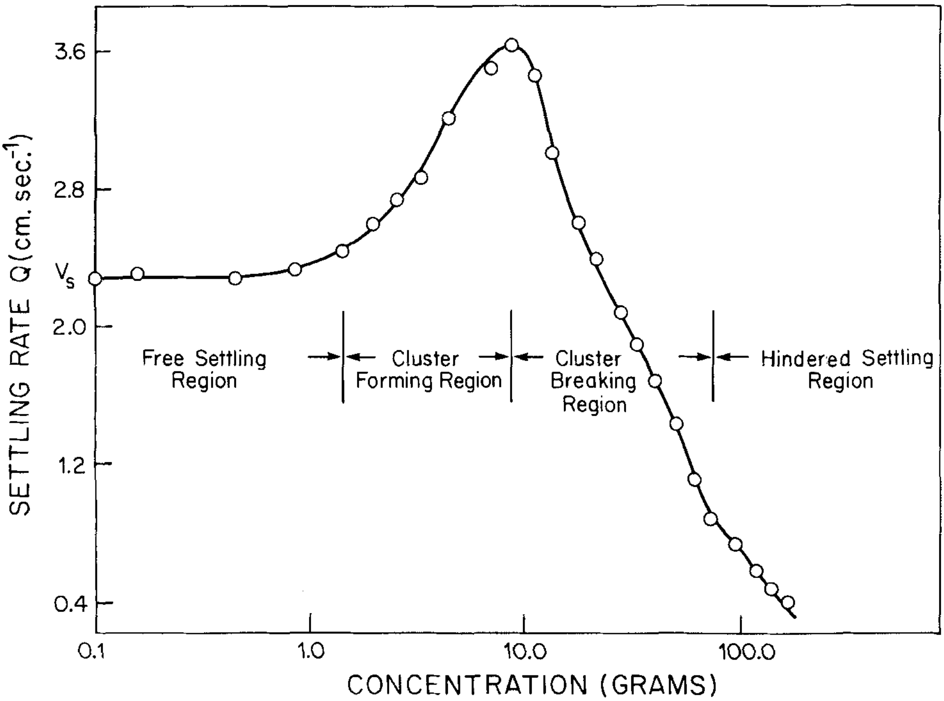

FIG. 2. Variation of the settling rate (Q, cm/s) with the concentration of glass ballotini suspensions.

suspension settle at a constant rate irrespective of their individual sizes. Figure 2 could, therefore, be divided into four distinct concentration ranges:

1) Up to $C = 1.5$ for free settling, when the particles are not influenced by each other and fall freely under Stokes' conditions.
2) From $C = 1.5$ to $C = 8.5$ for cluster formation and faster settling, resulting from a definite physical change in the suspensions.
3) From $C = 8.5$ to $C = 75.0$ for cluster breaking caused by the tortuous return fluid flow resulting in slower settling.
4) Beyond $C = 75.0$ for hindered settling, where uniformly diffused return flow of fluid occurs through the evenly dispersed cloud of particles.

The higher settling rate in the concentration region $C = 1.5$ to 8.5 may not be entirely due to true cluster formation, but could also be due to the interparticle behavior when settling separately but in close vicinity, as has been particularly mentioned by Hall (*2*).

Formation of Clusters—Visual Observations

Bigger particles settling at relatively high velocities show wakes behind them into which smaller particles easily fall, thus forming temporary clusters which can break apart if their relative velocities differ significantly.

Clusters of two particles (doublets) form more easily than the bigger ones, which is true for all size grades of ballotinis. Figure 1, for instance, shows that for all ballotini grades the frequency of forming doublets is more than that of the other cluster sizes. Doublets of equisized particles fall steadily so that the associated particles lie in the same horizontal plane. For particles of different sizes, however, the cluster movement becomes irregular in the beginning but more stable later so that the attached particles settle in two different planes.

A cluster of three (triplets) particles of the same size falls as a horizontal triangle of spheres. In the case of different particle sizes, a stable cluster would fall with the smaller particle staying at the top and the larger particle staying at the bottom.

A cluster of four (quadruplets) particles creates a tetrahedral configuration. In a stable tetrahedron with four equisized particles, the settling seems to be either apex-first or face-first. With clusters of different particle sizes the settling is irregular, often breaking into subclusters. Addition of another particle on any of the faces of the tetrahedron forms a cluster of five particles in the form of a triogonal bipyramid. In a stable position this cluster settles with the trigonal midplane horizontal.

Larger clusters do not form very often. However, with the smaller-sized ballotini (such as Grades E, F, and G), larger cluster formations have been observed as shown in Table 3. A cluster of six particles usually takes up an octahedral configuration (in contrast to Slack's observation of planar hexagonal), whereas a cluster of eight particles seems to form a simple cubic arrangement. A cluster bigger than this, although forming rarely, has a round shape.

Stable Clusters

Smaller particles have higher specific surfaces and may have a higher surface activity that can be associated to their higher degree of clustering.

From the results shown in Table 4 and Figs. 1 and 3, it is clear that the clusters made up of smaller particles, and hence of higher specific surfaces, are more stable. For instance, the relative stability of clusters with Grade G particles (radius 13.75 μm and specific surface 74.3 cm^2/g) is 59% compared to 17% for clusters made up of Grade A particles (radius 65 μm and specific surface 15.7 cm^2/g).

The stability of formation of a cluster is higher for clusters with fewer particles. Relative values of the stabilities of clusters are given in Table 3 and exhibited in Fig. 3.

Smaller clusters break down to individual particles whereas larger clusters break into subclusters. Apparently due to the upward thrust of fluid, some clusters become looser until each particle is sufficiently isolated to fall individually. Sometimes a larger cluster appears to lose its balance before breaking into subclusters due to its high settling velocity and turbulence during settling. The degree of turbulence has been estimated by calculating the Reynolds number, Re, from the observed

TABLE 4

Calculated Stokes Velocities of Clusters with and without Associated Liquids for Varying Sized Glass Ballotinis (V_{st} = Stokes' velocity without associated liquid, V_{stw} = Stokes' velocity with associated liquid)

Cluster size and formation		Grades of glass ballotinis						
		A	B	C	D	E	F	G
1 Single	V_{st}	3.97	1.90	1.15	0.99	0.71	0.53	0.18
2 Double	V_{st}	6.30	3.02	1.83	1.57	1.13	0.84	0.28
	V_{stw}	3.97	1.90	1.15	0.99	0.71	0.53	0.18
3 Trigonal	V_{st}	8.25	3.95	2.39	2.06	1.46	1.10	0.37
	V_{stw}	5.52	2.65	1.65	1.38	0.99	0.74	0.25
4 Single tetragonal	V_{st}	9.99	4.79	2.90	2.50	1.79	1.33	0.45
	V_{stw}	7.13	3.42	2.07	1.78	1.28	0.95	0.32
5 Double tetragonal	V_{st}	11.60	5.56	3.36	2.90	2.07	1.55	0.52
	V_{stw}	7.04	3.38	2.04	1.76	1.26	0.94	0.32
6 Octahedral	V_{st}	13.10	6.28	3.80	3.27	2.34	1.74	0.59
	V_{stw}	9.86	4.72	2.86	2.46	1.77	1.32	0.44
8 Single cubic	V_{st}	15.78	7.60	4.60	3.96	2.86	2.12	0.71
	V_{stw}	11.60	5.56	3.36	2.90	2.08	1.55	0.52

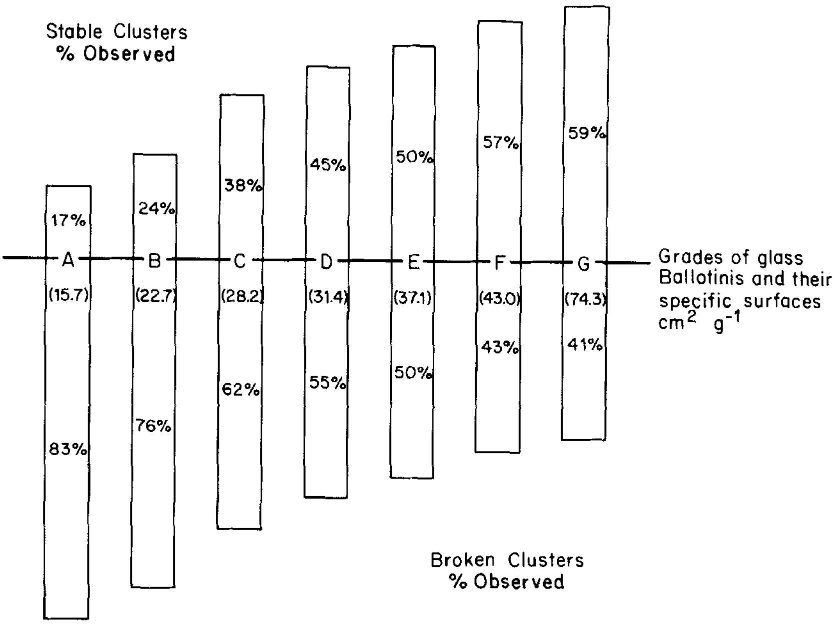

FIG. 3. Stable clusters vs broken clusters as a function of glass ballotini grades and their respective specific surfaces.

settling rates of individual clusters. The results are given in Table 2. Turbulence occurs at values of Re > 0.6 (*10*).

Reynolds numbers up to 0.6 correspond to the region of laminar flow. Above Re 0.6, flow is increasingly turbulent with increasing values of Re. Data by Richardson and Zaki (*11, 12*) show that the settling rate for a sphere is reduced to about $0.6V_s$ when Re rises to about 38. The most favorable circumstance for stable clustering is on the right-hand side of the heavy line in Table 2, where Re < 0.6. Some stable clusters, however, form at Re up to 1.48, for example, doublets of Grade A ballotinis, although their relative stability is very low due to the turbulent and tortuous conditions created by the larger and faster settling clusters. Values of Re > 1.48 given in Table 2 are for those clusters which break apart half way through settling. In a total suspension the degree of stable clustering observed at Re < 0.6 is therefore 52% compared to 22% at Re > 0.6.

The stable cluster formations observed in the nonlaminar flow region, i.e., at Re > 0.6, are presumably due to interparticle forces which are strong enough to resist the moments applied to them by the turbulent flow.

With smaller grades of ballotini, Grades F and G in particular, which have low Re values, clusters of eight particles or even more are observed.

"Immobile" Liquid and Particle Size Analysis

A particle settling in a fluid appears to carry a layer of immobile liquid around it (*13–17*). In the present investigation the falling particles in suspension fall into two classes; those that are "completely wetted" and those that are not. The latter are referred to here as "dry clusters." Completely wetted particles show a reduced tendency to stick together compared to the "dry cluster." These dry clusters are regular, and visual observation indicates that the particles are touching whereas in the completely wetted clusters the falling group is observed to consist of an aggregate in which not all the adjacent particles are touching.

The settling parameters of a cluster with or without any associated liquid are considered by taking into account the effect of variation in the bulk density of the settling cluster. A cluster of a certain number of particles without associated liquid is assumed to be equivalent to a single sphere of equal mass and density but of different diameter. Thus, for example, in a cluster of four equal spheres of radius r and density ρ_s, arranged in a tetrahedral formation, the calculation for the settling rate of an equivalent single sphere with radius R would be as follows. The volume of four spheres forming the cluster is $4[\frac{4}{3}\pi(r)^3]$, which, for the volume of a single sphere of radius R, becomes $\frac{4}{3}\pi[(4)^{1/3}r]^3$, giving $R = [(4)^{1/3}r]$.

Now a sphere with radius R and density ρ_s, when settled in a given fluid of known density ρ_l and viscosity η, will settle with a terminal velocity V_{st}:

$$V_{st} = \frac{2gR^2(\rho_s - \rho_l)}{9\eta} \tag{1}$$

The settling rate (V_{stw}) of a cluster with associated liquid is calculated by substituting in for a modified cluster density ρ_c when

$$\rho_c = v_l\rho_l + v_s\rho_s \tag{2}$$

with the values of ρ_l and ρ_s known, whereas v_l and v_s are calculated as follows.

A regular cluster is assumed with trapped liquid within its interstices. The sphere is then assumed to contain just the whole cluster. For instance, in a cluster of four spheres of radius r, in the tetrahedral form, the radius R of the containing sphere is $R = 2.225r$. Therefore, the volume of the containing sphere is $\frac{4}{3}\pi(2.225r)^3$, and the volume of the four spheres is $(4)\frac{4}{3}\pi(r)^3$.

The volume fraction of the liquid (v_l) in the containing sphere is

$$v_1 = \frac{\frac{4}{3}\pi(2.225r)^3 - (4)\frac{4}{3}\pi(r)^3}{\frac{4}{3}\pi(2.225)^3} \quad (3)$$

and the volume fraction of solid, v_s, is

$$v_s = (1 - v_1) \quad (4)$$

Therefore, the effective density of the cluster ρ_c is calculated by substituting the respective values of v_s and v_l in Eq. (2).

Substituting ρ_c in Stokes' equation, the value of V_{stw} for a cluster settling in a liquid of known density ρ_l and viscosity η is

$$V_{stw} = \frac{2gr^2(\rho_c - \rho_1)}{9\eta} \quad (5)$$

Similar calculations can be made for other cluster formations.

The amount of fluid attached to a cluster presumably varies with the size of a cluster. A larger cluster wtih a greater interstitial area will carry more liquid. The observed steady rates of fall of clusters as a function of cluster size are given in Table 2. Comparison of these with the theoretical rates for geometrically regular clusters, with or without associated liquid, can be used to give estimates of the velocities in the associated liquid. Table 4 gives the calculated theoretical values of these rates. The theoretical rate of fall for a given cluster without associated liquid is equal to that of a single sphere of the same mass and density and is calculated by using Stokes' law. The theoretical rate for a cluster with associated liquid is also obtained by using Stokes' law, assuming that the cluster is surrounded by a sphere of fluid just containing the cluster and then using the radius and mean density corresponding to this sphere. Note from Tables 2 and 4 that the observed settling rates for clusters fall between the theoretical rates for clusters with and without associated liquid.

Estimation of Cluster Size in a Particular Case

If the size of the glass ballotini used for the settling rate curve in Fig. 2 is assumed to be approximately equivalent to that of a Grade B ballotini (nominal diameter 0.09 cm), then the maximum settling rate, Q_{max}, of 3.62 cm/s, observed in Fig. 2, corresponds closely to that of observed clusters of three or four particles as shown in Fig. 4. The maximum rate referred

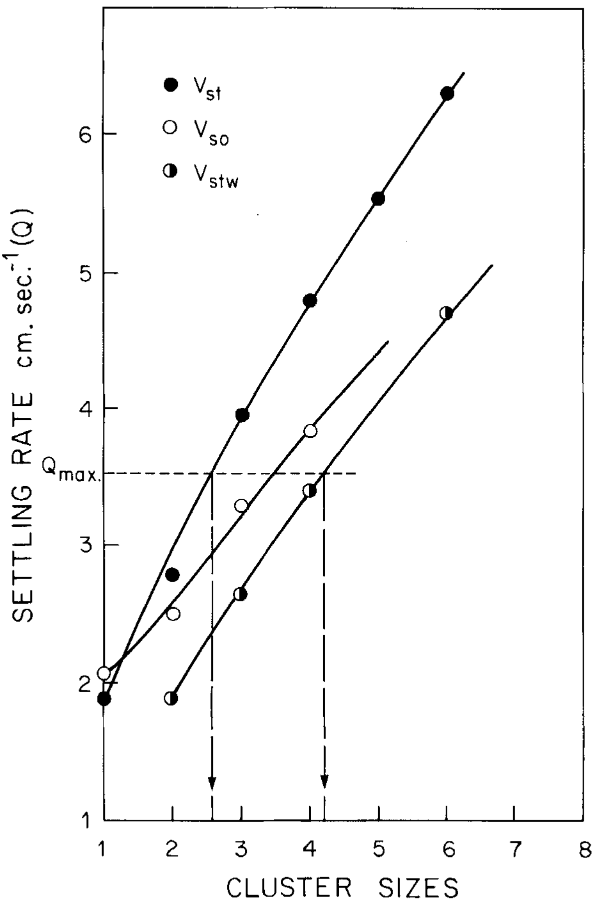

FIG. 4. Correspondence of the maximum settling rate (Q_{max}) of Grade B glass ballotini suspension to the size of cluster. The range between two arrows shows the optimum cluster sizes.

to here could therefore arise from the tendency at a given solid concentration for particles to cluster in threes and fours and then fall virtually unhindered by the presence of other particles. This is in agreement with the fact that the maxima occurred at $C = 8.50$, which corresponds to a liquid volume fraction of suspension = 0.984, well above the value of 0.90 reported by Kaye and Boardman (*3*) for well-developed hindered settling. The occurrence of the maximum settling rate at a liquid volume fraction of 0.984 is in very good agreement with Koglin's (*5*) corresponding value of 0.985, obtained for both monodisperse and polydisperse samples of spheres over a considerable range of cylinder to particle radius ratios.

Thus, as mentioned earlier, the maximum observed settling rate of 1.58 times the Stokes' settling velocity of a single particle for monosize suspensions occurs when the mean separation between particle surfaces is about 2.2 diameters. At this concentration the settling rate corresponds to the formation of clusters of three and four particles which have some associated "immobile" fluid incorporated in them, and which fall essentially unaffected by the presence of other particles. The particle-particle separation within the clusters is, however, much less than 2.2 diameters.

CONCLUSIONS

In a less concentrated suspension the settling particles group together to form clusters of varying sizes and formations, the stability of which largely depends on the size of the particles and of the clusters. Breakdown of cluster occurs beyond a certain solids concentration due to hydrodynamic demixing and the turbulence caused by increasing Reynolds numbers.

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