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Kinetic and Equilibrium Aspects of Floc Coagulation. II. Slow Mixing Criteria

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Abstract

The effect of slow mixing on colloid flocculation is examined within the framework of a modified Gouy-Chapman model, and the forces between the floc particles compared with the viscous drag forces due to velocity gradients. The effects of ionic strength and floc zeta potential are examined. It is found that the viscous drag forces are too small to force together floc particles having surface potentials greater than about 14.5 mV; no potential barrier occurs at surface potentials below about 14 mV. The strongly attractive but short-range dispersion force is apparently greatly weakened by the irregularities of the surfaces of the floc particles.

INTRODUCTION

The technique of colloid removal from water and wastewater by flocculation has been in use for many years; it is also utilized in the recovery of metals from leachates. The basic factors affecting flocculation include zeta potential of the floc (which may be profoundly affected by pH or the presence of specifically adsorbed ions), particle size, ionic strength, presence of protective colloids or colloids having a charge of opposite sign, and the characteristics of the slow mixing step (*1–21*).

We recently applied the techniques of statistical mechanics and diffusion theory in a potential field to examine the effects of zeta potential, temperature, particle size, and ionic strength on the rate of coagulation of floc particles and the conditions under which these systems are stable or metastable (*22*). In this work we used a modification of the methods described by Verwey and Overbeek (*23*) and by Sonntag and Strenge (*24*) to calculate interaction potentials between the colloidal particles. Our approach was restricted to quiescent suspensions, however, and it is common practice to carry out coagulation with a slow mixing step to assist in floc formation (*4*, *11–13*).

We wished to determine the extent to which the forces on the particles due to velocity gradients resulting from this slow mix might be effective in causing the coagulation of floc particles between which repulsive electrical forces exist. In quiescent solutions, random thermal motions may cause the coalescence of particles which repel each other provided that the energy barrier is no greater than a few kT (perikinetic flocculation). As the particles coalesce and the floc size grows, however, the magnitude of the energy barrier increases roughly proportional to the area of the consolidated floc particle, so that thermal coalescence of repelling particles must eventually cease. The viscous drag forces on particles in a velocity gradient increase with increasing particle size, however, so one might hope that slow mixing could bring about the coagulation of particles into settleable flocs even at zeta potentials for which the particles repel each other.

In the absence of such repulsive potentials, the usual considerations of orthokinetic flocculation would apply (*25–27*); the slow mix merely increases the rate at which particles collide, and no considerations of the magnitude of the viscous drag forces relative to particle–particle repulsions arise.

ANALYSIS

We consider the forces exerted on two floc particles suspended in a stirred solution of electrolyte. We define:

$\mathbf{r}_i$ = vector drawn from origin to particle i, $i = 1, 2$
$\mathbf{u}(\mathbf{r}, t)$ = velocity of fluid at the point $\mathbf{r}$ at time t
$\mathbf{v}_i(\mathbf{r}_i, t)$ = velocity of particle i at time t
a = particle radius

η = fluid viscosity
m = particle mass
$r_{12} = |\mathbf{r}_2 - \mathbf{r}_1|$
$e_{12} = (\mathbf{r}_2 - \mathbf{r}_1)/r_{12}$
$f(r_{12})$ = floc–floc interaction force

The equations of motion of the two particles are given by

$$m\dot{\mathbf{v}}_1 = 6\pi\eta a[\mathbf{u}(\mathbf{r}_1, t) - \mathbf{v}_1(\mathbf{r}_1, t)] + \mathbf{e}_{12}f(r_{12}) \tag{1}$$

$$m\dot{\mathbf{v}}_2 = 6\pi\eta a[\mathbf{u}(\mathbf{r}_2, t) - \mathbf{v}_2(\mathbf{r}_2, t)] - \mathbf{e}_{12}f(r_{12}) \tag{2}$$

Subtracting Eq. (2) from Eq. (1) yields

$$m\ddot{\mathbf{r}}_{12} = 6\pi\eta a[\mathbf{u}(\mathbf{r}_1, t) - \mathbf{u}(\mathbf{r}_2, t)] - 6\pi\eta\dot{\mathbf{r}}_{12}(\mathbf{r}_1, \mathbf{r}_2, t) + 2\mathbf{e}_{12}f(r_{12}) \tag{3}$$

The first term on the right-hand side of Eq. (3) gives the effect of velocity gradients in the fluid in bringing the particles together or forcing them apart. We wish to estimate the magnitude of this term. We do so by expanding $\mathbf{u}(\mathbf{r}_2, t)$ about the point $(\mathbf{r}_1, t)$ in a Taylor series and keeping terms through those linear in $\mathbf{r}_{12}$:

$$\mathbf{u}(\mathbf{r}_2, t) = \mathbf{u}(\mathbf{r}_1, t) + \mathbf{i}(\nabla u_x)\cdot\mathbf{r}_{12} + \mathbf{j}(\nabla u_y)\cdot\mathbf{r}_{12} + \mathbf{k}(\nabla u_z)\cdot\mathbf{r}_{12} \tag{4}$$

On substituting this result in Eq. (3), we obtain

$$m\ddot{\mathbf{r}}_{12} = -6\pi\eta a[\mathbf{i}(\nabla u_x)\cdot\ + \mathbf{j}(\nabla u_y)\cdot\ + \mathbf{k}(\nabla u_z)\cdot]\mathbf{r}_{12} - 6\pi\eta a\dot{\mathbf{r}}_{12} - 2\mathbf{e}_{12}f(r_{12}) \tag{5}$$

We wish to compare the relative magnitudes of the first and third terms; the viscous force should be large enough to force the particles to surmount any potential barrier associated with $f(r_{12})$, but not so large as to tear the particles apart after the barrier has been surmounted and the particles have collided and stuck together.

The magnitude of the viscous force is given by

$$\begin{aligned} f_v &= 6\pi\eta a[(\nabla u_x\Delta x)^2 + (\nabla u_y\Delta y)^2 + (\nabla u_z\Delta z)^2]^{1/2} \\ \Delta x &= x_2 - x_1, \text{ etc.} \end{aligned} \tag{6}$$

We assume, since the particles must be quite close together to interact, and their relative positions random, that we can replace Δx^2, Δy^2, and Δz^2 by $\frac{1}{3}(2a)^2$. Substitution in Eq. (6) then yields

$$f_v = \frac{12\pi\eta a^2}{\sqrt{3}}[(\nabla u_x)^2 + (\nabla u_y)^2 + (\nabla u_z)^2]^{1/2} \tag{7}$$

$$= 4\sqrt{3}\pi\eta a^2\left[\sum_{i=x}^{z}\sum_{j=x}^{z}\left(\frac{\partial u_i}{\partial x_j}\right)^2\right]^{1/2} \tag{8}$$

We next wish to relate Eq. (8) to the power dissipation per unit volume in the fluid. The power dissipation per unit volume is given by (*25*)

$$\Phi = \eta\left[2\sum_{i=x}^{z}\left(\frac{\partial u_i}{\partial x_i}\right)^2 + (\nabla\times\mathbf{u})^2 - \frac{2}{3}(\nabla\cdot\mathbf{u})^2\right] \tag{9}$$

(Since our fluid is incompressible, $\nabla\cdot\mathbf{u} = 0$.) On expanding out Eq. (9), one obtains

$$\begin{aligned}\Phi = \eta\Bigg\{2\left[\left(\frac{\partial u_x}{\partial x}\right)^2 + \left(\frac{\partial u_y}{\partial y}\right)^2 + \left(\frac{\partial u_z}{\partial z}\right)^2\right] + \left(\frac{\partial u_z}{\partial y}\right)^2 + \left(\frac{\partial u_y}{\partial z}\right)^2 + \left(\frac{\partial u_z}{\partial x}\right)^2 \\ + \left(\frac{\partial u_x}{\partial z}\right)^2 + \left(\frac{\partial u_y}{\partial x}\right)^2 + \left(\frac{\partial u_x}{\partial y}\right)^2 - 2\left[\frac{\partial u_z}{\partial y}\frac{\partial u_y}{\partial z} + \frac{\partial u_z}{\partial x}\frac{\partial u_x}{\partial z} + \frac{\partial u_y}{\partial x}\frac{\partial u_x}{\partial y}\right]\Bigg\}\end{aligned} \tag{10}$$

We are interested in an average value of this power dissipation function; this should result in the dropping out of all the cross terms (the last set of terms) in Eq. (10), giving

$$\begin{aligned}\Phi = \eta\Bigg\{2\left[\left(\frac{\partial u_x}{\partial x}\right)^2 + \left(\frac{\partial u_y}{\partial y}\right)^2 + \left(\frac{\partial u_z}{\partial z}\right)^2\right] + \left(\frac{\partial u_z}{\partial y}\right)^2 + \left(\frac{\partial u_y}{\partial z}\right)^2 + \left(\frac{\partial u_z}{\partial x}\right)^2 \\ + \left(\frac{\partial u_x}{\partial z}\right)^2 + \left(\frac{\partial u_y}{\partial x}\right)^2 + \left(\frac{\partial u_x}{\partial y}\right)^2\Bigg\}\end{aligned} \tag{11}$$

Let us compare this to the double sum which appears in Eq. (8),

$$\begin{aligned}S = \left(\frac{\partial u_x}{\partial x}\right)^2 + \left(\frac{\partial u_y}{\partial y}\right)^2 + \left(\frac{\partial u_z}{\partial z}\right)^2 + \left(\frac{\partial u_x}{\partial y}\right)^2 + \left(\frac{\partial u_y}{\partial x}\right)^2 + \left(\frac{\partial u_x}{\partial z}\right)^2 \\ + \left(\frac{\partial u_z}{\partial x}\right)^2 + \left(\frac{\partial u_y}{\partial z}\right)^2 + \left(\frac{\partial u_z}{\partial y}\right)^2\end{aligned} \tag{12}$$

Evidently

$$S \le \frac{1}{\eta}\Phi \le 2S \tag{13}$$

We note that Φ contains the equivalent of 12 squared terms, while S contains only 9, so assume that

$$S = \frac{3}{4\eta}\Phi \tag{14}$$

We substitute Eq. (14) for the sum in Eq. (8) to get for the average viscous force

$$f_v = 6\pi a^2(\eta\Phi)^{1/2} = 18.85a^2(\eta\Phi)^{1/2} \tag{15}$$

An alternative approach is to relate Eq. (7) to the speed of the stirrer in the flocculation tank. We assume that the stirring is done by long cylindrical bars of radius R and moving at velocity V'. We shall estimate ∇u by calculating the skin friction drag per unit length on a stirrer bar, using this to calculate an effective boundary layer thickness δ in the vicinity of the bar in which the liquid is in shear, and then setting $|\nabla u| = V'/\delta$. We assume that we are in the laminar flow region. Schlichting (*28*) gives the following formula for the skin friction drag on an infinitely long circular cylinder:

$$\frac{\tau_0}{\frac{1}{2}\rho V^2}\left[\frac{V'R\rho}{\eta}\right]^{1/2} = 6.973\theta - 2.732\theta^3 + 0.292\theta^5 - 0.0183\theta^7 + 0.000043\theta^9 - 0.00115\theta^{11} \tag{16}$$

where

τ_0 = shear stress at cylinder boundary
ρ = fluid density
V' = free stream velocity of liquid relative to the stirring bar
R = radius of cylinder
$R\theta$ = distance from stagnation point measured along the surface of the cylinder; see Fig. 1

Schlichting further notes that the boundary layer separation occurs at $\theta = 108.8°$ (1.899 radians).

The total drag force due to skin friction is then given by

$$\begin{aligned} F_s &= 2R\int_0^{1.899} \tau_0 \, d\theta \\ &= V'^{3/2}(\eta R\rho)^{1/2}\int_0^{1.899}\sum_{n=0}^{5} c_{2n+1}\theta^{2n+1}\,d\theta \\ &= 5.568V'^{3/2}(\eta R\rho)^{1/2} \end{aligned} \tag{17}$$

where the c_{2n+1} are given in Eq. (16).

The drag per unit length of bar due to skin friction is distributed over an area per unit length of bar of 2 × 1.899R. We therefore calculate a

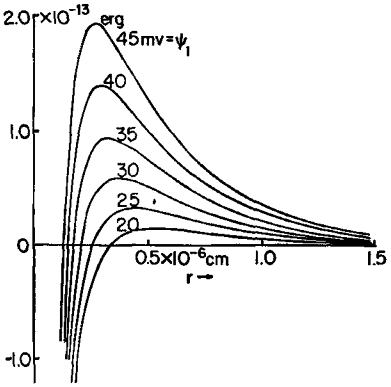

FIG. 1. Effect of ψ_1 on $V(r)$. $c_\infty = 10^{-5}$, $c_{\max} = 10^{-3}$ mole/cc, $T = 298\,^\circ\mathrm{K}$, $a = 50$ Å, $A_D = 5 \times 10^{-13}$ erg. $kT = 3.11 \times 10^{-14}$ erg.

mean boundary layer thickness δ from

$$F_s = \eta \frac{V'}{\delta} \times 3.798R \tag{18}$$

from which

$$\frac{V'}{\delta} = \frac{F_s}{3.798R\eta} \tag{19}$$

We then approximate the sum in Eq. (8) by $(V/\delta)^2$, so that the viscous force is given by

$$f_v = \frac{4v3\pi}{3.798}\frac{a^2F_s}{R} \tag{20}$$

Substituting for F_s from Eq. (17) then yields

$$f_v = 31.91a^2\left(\frac{\eta\rho}{R}\right)V'^{3/2} \tag{21}$$

as the relationship between the speed of a mixer bar and the viscous force tending to bring together or separate two particles nearly in contact with each other.

Let us next examine the force between the floc particles $f(r_{12})$ due to their interaction potential. From our earlier work (*22*, *29*), this force can be obtained from the following potential energy function:

$$V(r_{12}) = V_D(r_{12} - 2a) + V_E(r_{12} - 2a) \tag{22}$$

where V_D represents the Van der Waals attractive potential due to dispersion forces and is given by (*24*)

$$V_D(x) = \frac{-A_D}{48\pi}\left[\frac{1}{\frac{1}{4}x^2} + \frac{1}{(\frac{1}{2}x + 2a)^2} - \frac{1}{(\frac{1}{2}x + a)^2}\right]4a^2 \tag{23}$$

$$A_D \cong 5 \times 10^{-13}\,\text{erg}$$

The term V_E is due to the repulsive electrical force between the floc particles, and is given by (*29*)

$$V_E(x) = -Da^2C^{1/2}\int_{\psi_m(x)}^{\psi_1} \log\left[\frac{(1 + B\cosh w)^2}{(1 + B)(1 + B\cosh w_m)}\right] \cdot \left\{\log\left[\frac{1 + B\cosh w}{1 + B\cosh w_m}\right]\right\}^{-1/2} d\psi - V_E(\infty) \tag{24}$$

$$V_E(\infty) = \frac{-Da^2C^{1/2}}{\pi}\int_0^{\psi_1} 2\left\{\log\left[\frac{1 + B\cosh w}{1 + B}\right]\right\}^{1/2} d\psi \tag{25}$$

$C = 2A/\beta ezB$
$A = 8zec_\infty/(1 - 2c_\infty/c_{\max})D$
$B = 2c_\infty/(c_{\max} - 2c_\infty)$
$\beta = 1/kT$
$w = \beta ez\psi$
$w_m = \beta ez\psi_m(x)$
$|z|$ = |charge| of ions in the electrolyte solution in which the floc particles are dispersed (a 1–1 electrolyte)
e = electronic charge
c_∞ = electrolyte concentration, "molecules" per cm^3 ($6.023 \times 10^{23} \times$ mole/cm^3)
$c_{\max}$ = maximum possible electrolyte concentration, "molecules" per cm^3 (corrects for the finite volumes of the hydrated ions)
D = dielectric constant of the solution, approximately 78
$\psi_m(x)$ = electric potential midway between two floc particles separated by a distance x, calculated as indicated below
ψ = electric potential at a point ξ cm from Particle 1 when the two floc particles are x cm apart.

We calculate $\psi_m(x)$ from the relationship

$$\frac{d\psi}{d\xi} = \left[C\log\frac{1 + B\cosh\beta ez\psi}{1 + B\cosh\beta ez\psi_m}\right]^{1/2} \tag{26}$$

the first integral of the Poisson-Boltzmann equation appropriate for this system. Here $x/2 < \xi < x$, $(d\psi/d\xi)(x/2) = 0$, and $\psi(x) = \psi_1$, the surface potential of the floc particles; we use the numerical method outlined earlier (*29*).

Both of the terms in Eq. (22) are now defined, and we are in position to compute $V(r_{12})$ and the attractive force between the two particles,

$$f(r_{12}) = \frac{dV(r_{12})}{dr_{12}} \tag{27}$$

If the interaction potential exhibits no maximum, then evidently the slow mix has no repulsive force to overcome, and we only require that f_v be less than the maximum slope of $V(r_{12})$ in order that the floc, forming spontaneously on diffusion or mixing-induced collisions, not be torn to pieces once the particles have coagulated. Coagulation would occur spontaneously even in the absence of any mixing, albeit more slowly as the floc increased in size, the average distance between particles increased, and the diffusion constant decreased.

If the interaction potential exhibits a maximum which is only of the order of kT, roughly the thermal kinetic energy of the particles, the random thermal motions of the particles are sufficient to permit the potential barrier to be surmounted, and coagulation is again spontaneous in the absence of mixing until the growth of the barrier height with increasing floc size prevents further coalescence (*22*). Again we must require that $f_v < \max\,[f(r_{12})]$ in order that the flocculated particles not be separated by turbulence.

The most interesting case is that for which the potential barrier is large compared to kT. In a quiescent suspension this leads to a metastable

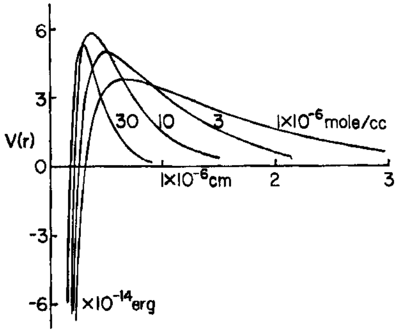

FIG. 2. Effect of c_∞ on $V(r)$. $\psi_1 = 30$ mV, other parameters as in Fig. 1.

colloid, as shown earlier (*22*). If, however, max $[f(r_{12})] > |\min [f(r_{12})]|$, one would expect there to exist a range of mixing power input Φ (or stirrer bar velocity V') for which the magnitude of the viscous force, f_v, is large enough to cause the particles to collide, permitting coalescence, but is not large enough to separate the particles once they have coalesced. The plots of particle–particle interaction potentials shown in Figs. 1 and 2, and the plots of max $[f(r_{12})]$ and $|\min [f(r_{12})]|$ in Figs. 3 and 4 give one some encouragement to hope that the slow mix step could be optimized. Thus, for colloidal suspensions in which the zeta potential, ionic strength, or other factors make coagulation difficult, one might hope that careful

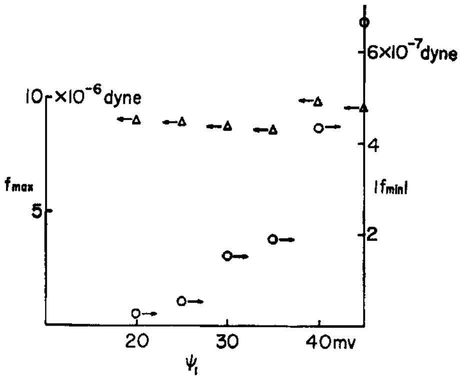

Fig. 3. Effect of ψ_1 on $f_{\max}$, $|f_{\min}|$. $c_\infty = 10^{-5}$, $c_{\max} = 10^{-3}$ mole/cc, $T = 298\,°\mathrm{K}$, $a = 50$ Å, $A_D = 5 \times 10^{-13}$ erg.

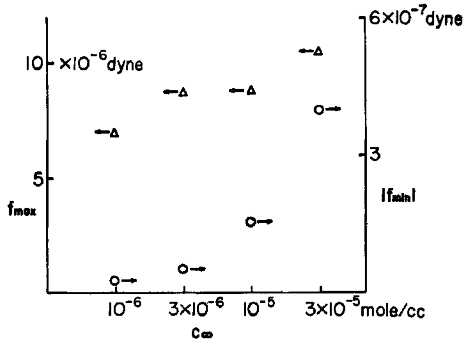

Fig. 4. Effect of c_∞ on $f_{\max}$, $|f_{\min}|$. $\psi_1 = 30$ mV, other parameters as in Fig. 3.

choice of power input in the slow mix step would result in significantly improved performance of the flocculator.

We note that we must have

$$|\min[f(r_{12})]| < f_v < \max[f(r_{12})] \tag{28}$$

On solving Eq. (15) for the mixing power input per unit volume, we get

$$\Phi = 2.87 \times 10^{-7} \frac{f_v^2}{a^4 \eta} \text{ horsepower/m}^3 \tag{29}$$

On solving Eq. (21) for the stirrer bar speed, we get

$$V' = 0.0994 \left(\frac{f_v^2 R}{a^4 \eta \rho} \right)^{1/3} \tag{30}$$

Let us consider floc particles having a surface potential of 20 mV in a solution of 0.01 mole/l ionic strength; for such particles we find that $|\min f(r_{12})|/a^2 = 1.0 \times 10^5$ dyne/cm^2. The viscosity is approximately 0.01 poise, so from Eq. (29), $\Phi = 2.9 \times 10^5$ horsepower/m^3, and from Eq. (30), assuming $R = 0.5$ cm, $V' = 789$ cm/sec. The value of Φ is preposterously large, and even the criterion based on the details of the stirring

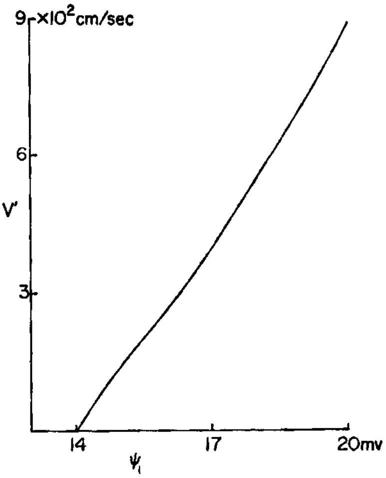

FIG. 5. Minimum stirring bar velocity necessary to bring about coagulation; effect of ψ_1. Here the repulsive force between the floc particles is approximated as $f_r = V_{\max}\kappa$. Parameters as in Fig. 1. κ^{-1} is the Debye length, given by $\kappa^2 = (8\pi e^2/DkT)c$ for 1-1 electrolyte.

process, Eq. (30), yields a value for the velocity of the stirrer bar sufficiently large that one is forced to conclude that flocculation of such colloids is not feasible.

In Fig. 5 we plot minimum stirrer bar velocity versus surface potential for particles suspended in a solution of 0.01 mole/l ionic strength. It is apparent that only for particle surface potentials in the range of 14 to 14.5 mV is slow stirring capable of inducing flocculation. One concludes that the effectiveness of slow mixing in facilitating flocculation is due almost exclusively to increased collision frequencies between nonrepelling floc particles. The viscous drag forces are too small to force particles together against any but the smallest repulsive forces unless the stirring velocity is extremely large.

One perplexing point concerns the inability of the strong short-range attractive force to hold coagulated floc particles together, even in the face of extremely vigorous stirring. The range of this force is seen from Figs. 2 and 3 to be of the order of 20 to 40 Å; surface roughness and irregularities may well be sufficient to prevent adjacent surfaces of two particles from approaching this close except over a small fraction of their common area, which would result in drastic weakening of these short-range forces.

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