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Separation of Solids at Liquid–Liquid Interface: Theory

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Abstract

The interrelationship of the forces acting on a solid suspended at a liquid–liquid interface was studied. An analysis is presented of the dependence of the maximum specific gravity of the solid, at which it will remain suspended, on the physical and physicochemical parameters of the system. The conditions for sustained suspension and the changes required for this purpose in some of the above parameters were determined. The mathematical relationships obtained yielded a theoretical model for separation of solids heavier than either of the liquids.

INTRODUCTION

Ore separation by flotation utilizes surface effects in aqueous solutions of surface-active agents.

This paper analyzes a new variant of the conventional technique realized at the interface of two immiscible liquid phases, with one fraction of the vertically-fed solids—which in this case are heavier than either of the liquids—remaining suspended (gravity overcome by surface tension), and the others settling.

A particular case of the problem was studied by Gaudin (*1*), who assumed that the suspended solid is cylindrical in shape. Barnea, in his own paper (*2*) and in collaboration with Mizrahi (*3*) and with Orell (*4*), presented a set of equations for a similar case involving a spherical solid,

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without reference to the separation process. Further works, such as those of Miller (*5*), Mitzmager and Mizrahi (*6*), and Mellgren and Shergold (*7*, *8*), also considered the light liquid/solid/heavy liquid system, but with emphasis on the flocculation aspect. Hartland and Robinson (*9–12*) dealt with still another aspect—that of the dynamic equilibrium of the interface during passage of the solid through it (including the drainage effect in the film between it and the heavier liquid)—and properly concluded that this dynamic state is identical with static equilibrium with a light-phase contact angle of 180°, i.e., perfect wetting. Princen and Goddard (*13*) considered a similar system in the context of the effect of mineral oil on the surface properties of binary surfactant systems; they plotted force vs displacement curves and derived an expression for the force acting on a horizontal cylinder passing through the interface.

The present paper analyzes the forces in the system in question and attempts to determine the conditions required for sustained suspension of the solid at the interface, including the dependence of the suspension on the geometrical and physicochemical parameters of the system, and the changes required in some of them for this purpose. The findings are eventually utilized in constructing a theoretical model for the separation process.

THEORETICAL

The system in question comprises three phases with distinct interfaces: liquid phases 1 and 2 (immiscible; 2 heavier than 1) and the solid phase s, concentrated between the other two and assumed to be spherical (so as to obviate the shape factor and ensure freedom from energy concentrations).

Physical and Physicochemical Forces in System

The forces are shown schematically in Fig. 1. The physical forces (due to gravity, F_s, and to buoyancy, F_1 and F_2) are represented by their directions of action. The physicochemical forces are due to the interface tensions—$F_{\gamma_{12}}$, $F_{\gamma_{1s}}$, and $F_{\gamma_{2s}}$.

The forces are functions of the radius of the suspended particle. The three physical forces vary linearly with its volume, i.e., as the cube of the radius. By contrast, $F_{\gamma_{12}}$ is a linear function of the radius, and in these circumstances its contribution is more noticeable the smaller the radius.

Interrelationship of Parameters

The equilibrium of the system is represented by

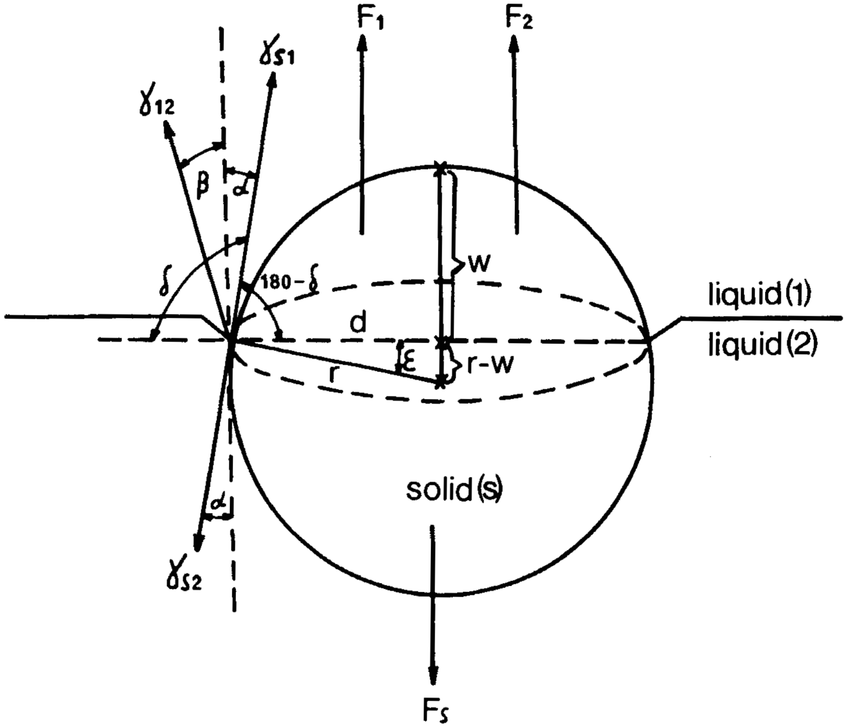

FIG. 1. Solid sphere suspended at liquid–liquid interface. (Dashed ellipse: projection of common circle).

$$[(\gamma_{s1} - \gamma_{s2}) \cos \alpha + \gamma_{12} \cos \beta]l = g[V_{\text{total}}\rho_s - V_1\rho_1 - V_2\rho_2] \qquad (1)$$

where l is circumference of the circle common to all three phases, namely

$$l = 2\pi d = 2\pi r \sin \delta \qquad (2)$$

also

$$\cos \alpha = \sin \delta$$

By the Young-Dupré equation,

$$\gamma_{s1} - \gamma_{s2} = \gamma_{12} \cos \theta$$

Substituting in Eq. (1), we have

$$[\gamma_{12} \cos \alpha \cos \theta + \gamma_{12} \cos \beta]2\pi r \sin \delta = g[V_{\text{total}}\rho_s - V_1\rho_1 - V_2\rho_2] \qquad (3)$$

and, substituting the geometrical values for the three volumes, namely

$$V_1 = \frac{\pi r^3}{3}[2 + 3\cos\delta - \cos^3\delta] \tag{4}$$

$$V_2 = \frac{\pi r^3}{3}[2 - 3\cos\delta + \cos^3\delta] \tag{5}$$

$$V_{\text{total}} = \frac{4\pi r^3}{3} \tag{6}$$

we finally obtain

$$\gamma_{12} = \frac{gr^2}{6\sin\delta(\sin\delta\cos\theta + \cos\beta)}[4\rho_s - \rho_1(2 + 3\cos\delta - \cos^3\delta) - \rho_2(2 - 3\cos\delta + \cos^3\delta)] \tag{7}$$

Equation (7) is the basic equation for the case of a partially-wetted spherical solid at the interface of two liquids.

Angle δ vs w

Figure 1 shows that $\cos\delta = -(r - w)/r$:

$$\Delta\delta = \frac{\partial\delta}{\partial y}\frac{\partial y}{\partial w}\Delta w \tag{8}$$

Denoting $\cos\delta$ by y, we have

$$\frac{\partial\delta}{\partial y} = \frac{1}{\sqrt{1 - y^2}}, \qquad \frac{\partial y}{\partial w} = \frac{1}{r}$$

Substituting the above in Eq. (8) we find:

$$\Delta\delta = \frac{1}{r}\frac{1}{\sqrt{w(2r - w)}}\Delta w \tag{9}$$

Angle β vs w

Denoting $\cos\beta$ by y, Eq. (7) yields

$$y = \cos\beta = \frac{gr^2}{6\gamma_{12}\sin\delta}[4\rho_s - \cos^3\delta(\rho_2 - \rho_1) + 3\cos\delta(\rho_2 - \rho_1) - 2(\rho_2 + \rho_1)] - \sin\delta\cos\theta$$

$$\Delta\beta = \frac{\partial\beta}{\partial y}\frac{\partial y}{\partial\delta}\Delta\delta$$

$$\frac{\partial\beta}{\partial y} = \frac{1}{\sqrt{1-y^2}} \tag{10}$$

and [for the evaluation of the partial derivative $\partial\theta/\partial\delta$, see Eq. (24)]

$$\frac{\partial y}{\partial g} = \frac{gr^2}{6\gamma_{12}}\left\{\frac{1}{\sin\delta}[3\cos^2\delta\sin\delta(\rho_2-\rho_1) - 3\sin\delta(\rho_2-\rho_1)]\right.$$
$$\left.-\frac{\cos\delta}{\sin^2\delta}[4\rho_s - \cos^3\delta(\rho_2-\rho_1) + 3\cos\delta(\rho_2-\rho_1) - 2(\rho_2-\rho_1)]\right\}$$
$$-\cos\delta\cos\theta + \sin\delta\sin\theta\frac{\partial\theta}{\partial\delta}$$

and again, substituting the above in Eq. (10), we find

$$\Delta\beta = \frac{\partial y}{\partial\delta}\frac{1}{\sqrt{1-\cos^2\beta}}\frac{1}{\sqrt{w(2r-w)}}\Delta w \tag{11}$$

Force K vs w

Denoting by K the algebraic sum of the three physical forces (which equals the force due to γ_{12}) in Eq. (1), namely

$$K = g(V_{\text{total}}\rho_s - V_1\rho_1 - V_2\rho_2)$$

and substituting the geometrical values for the three volumes, we have

$$K = \frac{\pi}{3}g[-w^3(\rho_2-\rho_1) + 3rw^2(\rho_2-\rho_1) - 4r^3(\rho_2-\rho_s)] \tag{12}$$

whence

$$\frac{dK}{dw} = \frac{\pi}{3}g[-3w^2(\rho_2-\rho_1) + 6rw(\rho_2-\rho_1)]$$

Substituting the above in the identity

$$\Delta K = \frac{dK}{dw}\Delta w \tag{13}$$

we find

$$\Delta K = g\pi w(\rho_2-\rho_1)(2r-w)\Delta w \tag{14}$$

With the physical force changed by ΔK, with a resulting vertical deformation Δw of the sphere, Eq. (3) will read

$$2\pi r\gamma_{12}\sin(\delta+\Delta\delta)[\sin(\delta+\Delta\delta)\cos\theta+\cos(\beta+\Delta\beta)]=K+\Delta K \qquad (15)$$

Maximum Specific Gravity of Solid vs Degree of Wetting

The maximum specific gravity at which a solid will remain suspended at the interface is a direct function of its degree of wetting by the two liquids. By Eq. (7) we have

$$\rho_s=\frac{6\gamma_{12}\sin\delta(\sin\delta\cos\theta+\cos\beta)}{4gr^2}+\frac{\rho_1}{4}(2+3\cos\delta-\cos^3\delta)$$
$$+\frac{\rho_2}{4}(2-3\cos\delta+\cos^3\delta)$$

At the moment of disruption of the suspension equilibrium ($\rho_s=\rho_{s\,\max}$) we have $\partial\rho_{s\,\max}/\partial\theta=0$, so that differentiating Eq. (7) with respect to θ we obtain

$$\frac{\partial\rho_{s\,\max}}{\partial\theta}=0=\frac{3\gamma_{12}}{2gr^2}\left[2\sin\delta\cos\delta\cos\theta\frac{\partial\delta}{\partial\theta}-\sin^2\delta\sin\theta+\cos\delta\cos\beta\frac{\partial\delta}{\partial\theta}\right.$$
$$\left.-\sin\delta\sin\beta\frac{\partial\beta}{\partial g}\right]+\frac{\partial\delta}{\partial\theta}[-3\sin\delta+3\cos^2\delta\sin\delta]\frac{\rho_1}{4}$$
$$+\frac{\partial\delta}{\partial\theta}[3\sin\delta-3\cos^2\delta\sin\delta]\frac{\rho_2}{4} \qquad (16)$$

With the contact angle measured in phase 1, Fig. 1 shows that $\theta=\alpha+\beta$, where $\beta=90+\theta-\delta$ and $\partial\beta/\partial\theta=1-(\partial\delta/\partial\theta)$.

Substituting the above in Eq. (16), we find

$$\frac{\partial\rho_{s\,\max}}{\partial\theta}=\frac{3\gamma_{12}}{2gr^2}\left[2\sin\delta\cos\delta\cos\theta\frac{\partial\delta}{\partial\theta}-\sin^2\delta\sin\theta+\cos\delta\cos\beta\frac{\partial\delta}{\partial\theta}\right]$$
$$\left.-\sin\delta\sin\beta+\sin\delta\sin\beta\frac{\partial\delta}{\partial\theta}\right]$$
$$+\frac{\partial\delta}{\partial\theta}[-3\sin\delta+3\cos^2\delta\sin\delta]\frac{\rho_1}{4}$$
$$+\frac{\partial\delta}{\partial\theta}[3\sin\delta-3\cos^2\delta\sin\delta]\frac{\rho_2}{4} \qquad (17)$$

For evaluating $\partial\delta/\partial\theta$, we denote by P the trigonometric product in the denominator in Eq. (7):

$$P = \sin\delta(\sin\delta\cos\theta + \cos\beta)$$

$$= \frac{gr^2}{6\gamma_{12}}[4\rho_s - \rho_1(2 + 3\cos\delta - \cos^3\delta)$$

$$- \rho_2(2 - 3\cos\delta + \cos^3\delta)] \tag{18}$$

$$\frac{\partial\delta}{\partial\theta} = \frac{\partial\delta}{\partial P}\frac{\partial P}{\partial\theta} \tag{19}$$

Differentiating Eq. (18) with respect to θ, we find

$$\frac{\partial P}{\partial\theta} = 2\sin\delta\cos\delta\cos\theta\frac{\partial\delta}{\partial\theta} - \sin^2\delta\sin\theta + \cos\delta\cos\beta\frac{\partial\delta}{\partial\theta}$$

$$+ \sin\delta\sin\beta\frac{\partial\beta}{\partial\theta} \tag{20}$$

Recalling that $\beta = 90 + \theta - \delta$, and substituting $\sin\beta$, $\cos\beta$, and $\partial\beta/\partial\theta$, we find

$$\frac{\partial P}{\partial\theta} = \frac{\partial\delta}{\partial\theta}[4\sin\delta\cos\delta\cos\theta + \sin^2\delta\sin\theta - \cos^2\delta\sin\theta]$$

$$- [2\sin^2\delta\sin\theta + \sin\delta\cos\delta\cos\theta] \tag{21}$$

Similarly, differentiating Eq. (18) with respect to δ, we find

$$\frac{\partial P}{\partial\delta} = \frac{-gr^2}{6\gamma_{12}}[\rho_1(-3\sin\delta + 3\sin\delta\cos^2\delta) + \rho_2(3\sin\delta - 3\sin\delta\cos^2\delta)] \tag{22}$$

or

$$\frac{\partial P}{\partial\delta} = \frac{-gr^2}{2\gamma_{12}}\sin^3\delta(\rho_2 - \rho_1) \tag{23}$$

and finally, substituting both derivatives in Eq. (19), we find

$$\frac{\partial\delta}{\partial\theta} = \frac{2\gamma_{12}[2\sin^2\delta\sin\theta + \sin\delta\cos\delta\cos\theta]}{gr^2\sin^3\delta(\rho_2-\rho_1)+2\gamma_{12}[4\sin\delta\cos\delta\cos\theta-\sin\theta\cos^2\delta+\sin^2\delta\sin\theta]} \tag{24}$$

Maximum Specific Gravity of Solid for Different Parameter Combinations

Under constant pressure and temperature conditions, the free parameters of the system are δ, θ, r, γ_{12}, ρ_s, ρ_1, and ρ_2. Their influence on $\rho_{s\,\max}$ is examined below, assuming, as the first step, that the liquids (i.e., ρ_1 and ρ_2) are given.

Figures 2–4 show the dependence of $\rho_{s\,\max}$ on r, θ, and δ in the case of distilled water and tetrabromoethane (TBE), i.e., $\rho_1 = 1.00$ g/cm^3, $\rho_2 = 2.93$ g/cm^3, and $\gamma_{12} = 38.8$ dyne/cm for the following parameter combinations: in Fig. 2, $\rho_{s\,\max}$ vs θ with r as parameter (δ constant); in Fig. 3, $\rho_{s\,\max}$ vs δ with r as parameter (θ constant); and in Fig. 4, $\rho_{s\,\max}$ vs δ with θ as parameter (r constant).

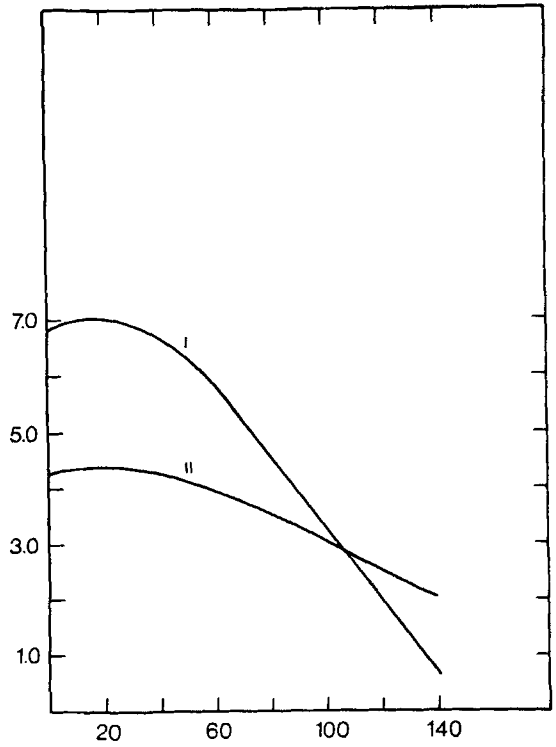

FIG. 2. $\rho_{s\,\max}$ vs contact angle θ for different radii of solids. I: $r = 0.150$ cm; $\delta = 120°$. II: $r = 0.250$ cm; $\delta = 120°$. Abscissa: Contact angle θ (degrees). Ordinate: $\rho_{s\,\max}$(g/cm^3).

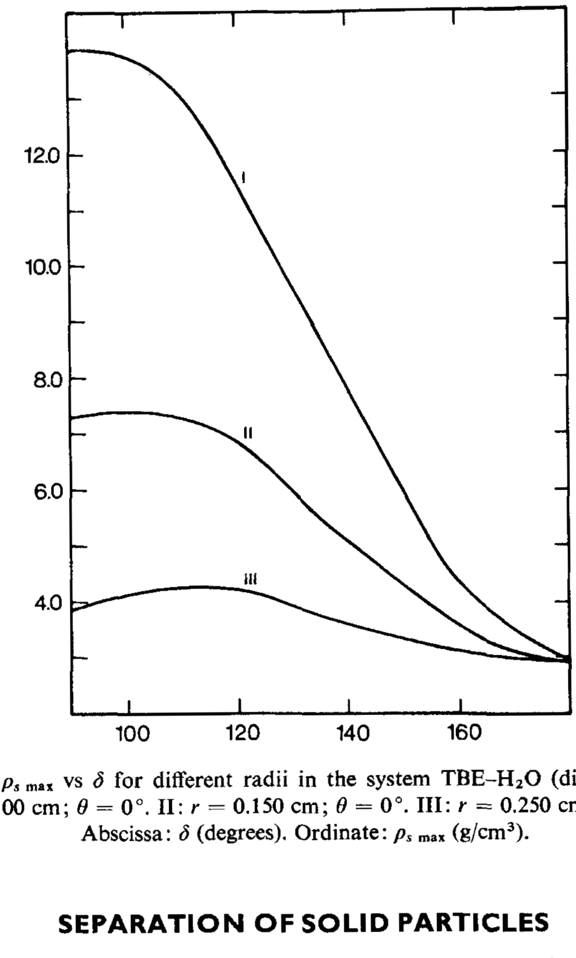

FIG. 3. $\rho_{s\,\max}$ vs δ for different radii in the system TBE–H_2O (distilled). I: $r = 0.100$ cm; $\theta = 0°$. II: $r = 0.150$ cm; $\theta = 0°$. III: $r = 0.250$ cm; $\theta = 0°$. Abscissa: δ (degrees). Ordinate: $\rho_{s\,\max}$ (g/cm^3).

SEPARATION OF SOLID PARTICLES

The system under consideration may be used for two variants of separation: (a) identical solids of different sizes (classification), and (b) different solids of either the same size or different sizes (concentration). Denoting the materials to be separated by the subscripts A and B, we have by Eq. (7)

$$\Delta\rho_{s\,\max} = \rho_{s\,\max\,A} - \rho_{s\,\max\,B} = \frac{3}{2}\frac{\gamma_{12}}{gr^2}\sin\delta_A(2\sin\delta_A\cos\theta_A - \cos\delta_A\sin\theta_A)$$
$$+ \frac{1}{4}\rho_1(2 + 3\cos\delta_A - \cos^3\delta_A) + \frac{1}{4}\rho_2(2 - 3\cos\delta_A + \cos^3\delta_A)$$

$$-\left[\frac{3}{2}\frac{\gamma_{12}}{gr^2}\sin\delta_B(2\sin\delta_B\cos\theta_B-\cos\delta_B\sin\theta_B)\right.$$
$$\left.+\frac{1}{4}\rho_1(2+3\cos\delta_B-\cos^3\delta_B)+\frac{1}{4}\rho_2(2-3\cos\delta_B+\cos^3\delta_B)\right] \tag{25}$$

Identical Solids

If A and B are identical in respect to ρ, θ, and δ, the above equation becomes

$$\Delta\rho_{s\ \max}=\frac{3}{2}\frac{\gamma_{12}}{g}\sin\delta(2\sin\delta\cos\theta-\cos\delta\sin\theta)\left(\frac{1}{r_A{}^2}-\frac{1}{r_B{}^2}\right) \tag{26}$$

This variant is illustrated in Fig. 3. For example, for $\delta = 120°$, $r_A =$

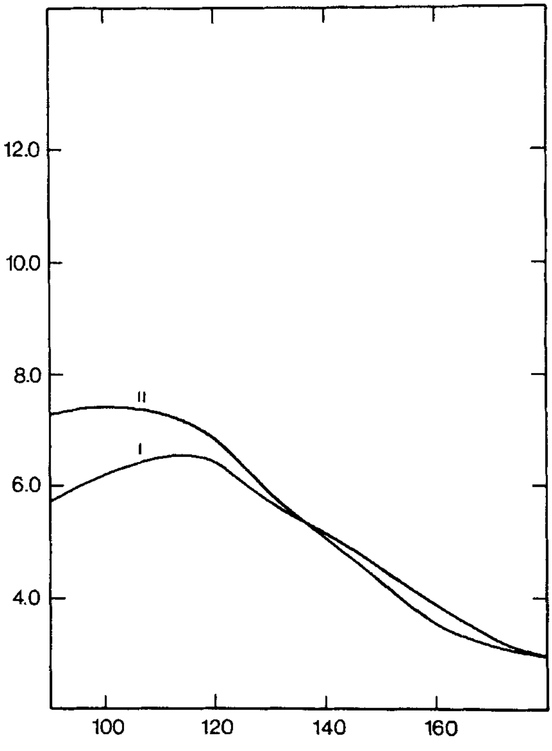

FIG. 4. $\rho_{s\ \max}$ vs δ for different contact angles θ. I: $r = 0.150$ cm; $\theta = 45°$. II: $r = 0.150$ cm; $\theta = 0°$. Abscissa: δ (degrees). Ordinate: $\rho_{s\ \max}$ (g/cm^3).

0.100 cm, $r_B = 0.250$ cm, and $\theta = 0$, and the maximum specific gravities for suspension are $\rho_{s\ \max\ A} = 11.5$ g/cm^3 and $\rho_{s\ \max\ B} = 4.2$ g/cm^3. In other words, between these limits all the larger particles will settle and all the smaller ones will remain suspended at the interface.

Nonidentical Solids

For solids differing in respect of specific gravity, surface properties, etc., three cases may be distinguished:

Different θ, identical r *and δ*

By Eq. (7)

$$\Delta\rho_{s\ \max} = \frac{3}{2}\frac{\gamma_{12}}{gr^2}\sin\delta[2\sin\delta(\cos\theta_B - \cos\theta_A) - \cos\delta(\sin\theta_B - \sin\theta_A)] \tag{27}$$

This case is illustrated in Fig. 4. For example, for r = 0.150 cm, $\delta = 100°$, $\theta_A = 45°$, and $\theta_B = 0$ (no wetting), we have $\rho_{s\ \max\ A} = 6.2$ g/cm^3 and $\rho_{s\ \max\ B} = 7.3$ g/cm^3. Separation is possible here when $\rho_{s\ \max\ A} > 6.2$ and $\rho_{s\ \max\ B} < 7.3$ (A settling, B suspended), or $\rho_{s\ \max\ A} < 6.2$ and $\rho_{s\ \max\ B} > 7.3$ (A suspended, B settling).

Different r *and δ, identical θ*

$$\begin{aligned}\Delta\rho_{s\ \max} = \frac{3}{2}\frac{\gamma_{12}}{g{r_A}^2{r_B}^2}\Bigg[&[2\cos\theta({r_B}^2\sin^2 A - {r_A}^2\sin^2 B) \\ &- \frac{\sin\theta}{2}({r_B}^2\sin 2\delta_A - {r_A}^2\sin 2\delta_B)]\Bigg] \\ &+ \frac{1}{4}(\rho_2 - \rho_1)[(\cos^3\delta_A - \cos^3\delta_B) - 3(\cos\delta_A - \cos\delta_B)]\end{aligned} \tag{28}$$

This case is illustrated in Fig. 3. For example, for $r_A = 0.250$ cm, $\delta_A = 120°$; $r_B = 0.100$ cm, and $\delta_B = 110°$, Curve III yields $\rho_{s\ \max\ A} = 4.2$ g/cm^3 and Curve I yields $\rho_{s\ \max\ B} = 13.0$ g/cm^3. Separation is possible when $\rho_{s\ \max\ A} > 4.2$ and $\rho_{s\ \max\ B} < 13.0$ (A settling, B suspended).

Different r, *identical θ and δ*

Equation (26) is also valid in this case, which is illustrated in Fig. 2. For example, for $r_A = 0.150$ cm, $r_B = 0.250$ cm, and $\theta = 30°$, we have

$\rho_{s\ \max\ A} = 6.9$ g/cm^3 and $\rho_{s\ \max\ B} = 4.3$ g/cm^3. Separation is possible when $\rho_{s\ \max\ A} > 6.9$ and $\rho_{s\ \max\ B} < 4.3$ (B suspended, A settling) or when $\rho_{s\ \max\ A} < 6.9$ and $\rho_{s\ \max\ B} > 4.3$ (A suspended, B settling).

DISCUSSION AND CONCLUSIONS

(1). Diverse alternatives exist for varying the parameters so as to adapt the system for each combination of solids. If, for example, both solids behave identically (suspension or settling), the contact angle of either (or both) may be increased or reduced as necessary by means of an adsorbed surfactant. Alternatively, ρ_1 or ρ_2, or both, can be changed; such a change is likely to affect the interface tension γ_{12}.

(2). Figure 2 (and even more so Fig. 3) illustrates the dependence of $\rho_{s\ \max}$ on r. Small-size solids can be suspended even if much heavier than liquid 2, a point of special significance in treatment of heavy ores.

(3). It is reasonable to assume that the poorer the wetting ability of the heavy liquid phase, the higher the specific gravity limit for suspension. This is confirmed in Fig. 2, where the main trend is decrease of $\rho_{s\ \max}$ with increasing θ—moderate up to $\theta = 30$ to $35°$ and steep afterwards. [As for the slight initial increase in $\rho_{s\ \max}$ at very low θ (more pronounced in Curve I, which refers to small r), it is not clear whether it is accidental or systematic.]

(4). Equation (7) was mainly used for determining the maximum specific gravity of the solid for suspension at the interface. Conversely, it is possible to suspend a known solid and thus determine the interfacial tension of the liquid phase. This is useful where the conventional method for interfacial-tension measurement fail to yield satisfactory accuracy.

SYMBOLS

d	radius of common circle of three phases
F_1	buoyancy of solid s in liquid 1
F_2	buoyancy of solid s in liquid 2
F_s	weight of solid s
F_{12}	force due to interfacial tension between liquid phases
g	gravity acceleration
l	circumference of common circle
r	radius of solid s
V_1	volume of spherical segment in liquid 1
V_2	volume of spherical segment in liquid 2
V_{total}	volume of sphere

w	altitude of spherical segment in liquid 1.

Greek symbols

α	angle between vector γ_{s1} and vertical
β	angle between vector γ_{s2} and vertical
δ	angle between vector γ_{s1} and plane of common circle
γ	interfacial tension
ρ	specific gravity
$\rho_{s\,\max}$	maximum specific gravity at which a solid will remain suspended
θ	thermodynamic contact angle at three-phase interface

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