Stabilization of Thin Liquid Films by Repulsive

van der Waals Force

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Appendix I

Stefan-Reynolds Flat Disk Model – Stokes flow with at least one interface is no-slip

In the Stefan-Reynolds flat disk model^{A1,A2} the approach of the bubble towards the meniscus is modeled as a flat disk of radius *a* approaching an infinitely large flat surface in a parallel orientation. The separation, h(t), between the disk and the flat surface varies with time, *t*. This model is applicable is the tangentially immobile or no-slip hydrodynamic boundary condition holds on at least one of the surfaces.

We define a coordinate system with the unit vector, \hat{z} , of the z-axis to point out of the flat surface towards the disk. If the disk moves with speed dh/dt, the hydrodynamic force arising from drainage of the intervening fluid between the disk and the flat surface is

$$F_{hy} = -\frac{3\pi\mu a^4}{2\beta h^3} \frac{dh}{dt} \hat{z}$$
(A1)

where the constant, β takes on value depending on the hydrodynamic boundary conditions on the disk and on flat surface. We have

 $\beta = 1$ no-slip on both surfaces,

 $\beta = 4$ no-slip on one surface & slip on the other

To obtain an upper limit of the drainage time, we can take $\beta = 1$.

The van der Waals force between the disk and the flat surface has the form

$$F_{vdw} = -\pi a^2 \frac{A}{6\pi h^3} \hat{z} \tag{A2}$$

The Hamaker constant A > 0 for an attractive interaction that will cause film rupture or coalescence.

The constant buoyancy force on the rising bubble has the form

$$F_{buoy} = -\frac{4\pi}{3}\rho g R^3 \ \hat{z} \equiv -F_g \ \hat{z}$$
(A4)

,where $F_g > 0$ as defined.

Applying the force balance condition $F_{hy} + F_{vdw} + F_{buoy} = 0$, we obtain

 $\langle \cdot \cdot \cdot \rangle$

$$-\frac{3\pi\mu a^4}{2\beta h^3}\frac{dh}{dt} - \pi a^2 \frac{A}{6\pi h^3} - F_g = 0$$
(A5)

This equation can be readily integrated to give, in dimensionless form

$$T = \frac{1}{6\alpha^3} \left\{ log\left[\left(\frac{1+\alpha}{H+\alpha} \right)^3 \left(\frac{H^3+\alpha^3}{1+\alpha^3} \right) \right] + 2\sqrt{3} \left[arctan\left(\frac{2-\alpha}{\sqrt{3}\alpha} \right) - arctan\left(\frac{2H-\alpha}{\sqrt{3}\alpha} \right) \right] \right\}$$

where the scaled parameters with initial separation $h_o = h(t=0)$ are

$$F_g \equiv \frac{4\pi}{3} \rho g R^3$$

$$\alpha^3 \equiv \frac{a^2 A}{6h_o^3 F_g}$$

$$\tau \equiv \frac{3\pi\mu a^4}{2\beta h_o^2 F_g}$$

$$H \equiv \frac{h(t)}{h_o}$$

$$T \equiv \frac{t}{\tau}$$

For an attractive van der Waals interaction with Hamaker constant, A > 0, we have $\alpha > 0$.

The dimensionless coalescence time, T_{coales} is found by putting $H(t) = h(t)/h_o = 0$

$$T_{coales} = \frac{1}{6\alpha^3} \left\{ log\left[\frac{(1+\alpha)^3}{1+\alpha^3} \right] + 2\sqrt{3} \left[\arctan\left(\frac{2-\alpha}{\sqrt{3}\alpha} \right) + \arctan\left(\frac{1}{\sqrt{3}} \right) \right] \right\}$$

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(A7)

(A6)

Appendix II

Potential flow with BOTH interfaces obey the slip boundary condition

In this case, we can no longer use Stokes flow model that assumes viscosity effect dominates film drainage. With the slip boundary conditions on both surfaces, drainage is determined by inertia. Using the model developed for the equation of motion of a non-deforming spherical bubble of radius, R, approaching a flat surface or two non-deforming spherical bubbles approaching each other^{A3,A4}

$$\frac{d}{dt}\left(M\frac{dh}{dt}\right) - \frac{1}{2}M\left(\frac{dh}{dt}\right)^2 = -\frac{4\pi}{3}\rho gR^3 \tag{A8}$$

where M is the position dependent effective mass of the bubble given by

$$M = C(h) \frac{4\pi}{3} \rho g R^3 \tag{A9}$$

and C(h) is the effective mass coefficient the decreases from 0.8 at h = 0 to 0.5 at $h = \infty$, and dC/dh is finite. C(h) does not depend on the radius.

Since

$$\frac{dM}{dt} = \frac{dM}{dh}\frac{dh}{dt}$$
(A10)

the equation of motion becomes

$$M\frac{d^2h}{dt^2} + \frac{1}{2}\frac{dM}{dh}\left(\frac{dh}{dt}\right)^2 = -\frac{4\pi}{3}\rho gR^3$$
(A11)

and after substituting for the effective mass M

$$C\frac{d^{2}h}{dt^{2}} + \frac{1}{2}\frac{dC}{dh}\left(\frac{dh}{dt}\right)^{2} = -g$$
(A12)

Thus the coalescence time is *independent* of the bubble radius and scales as

$$T_{coales} \sim \sqrt{\frac{h_o}{g}}$$
 (A13)

The small magnitude is comparable to the observed cases in which the coalescence time is independent of the bubble radius.

Appendix III

Disjoining pressure between two deformable interfaces



Figure A1 (a) Pressure values in the Bulk liquid, P_{Bulk} , in the Upper liquid, P_{Upper} , and in the bubble, $P(\infty)$, when the bubble is far from the meniscus. (b) Pressure values when the bubble is separated from the meniscus by a film of thickness, *h* and its Laplace pressure is P(h) as a result of deformations.

When the bubble is far from the meniscus as depicted in Fig. A1 (a), the mean radius of curvature of the bubble is $R(\infty)$ and that of the meniscus is $R_m(\infty)$. Consideration of the Young-Laplace equation across the bubble interface gives

$$P(\infty) = P_{Bulk} + \frac{2\gamma_b}{R(\infty)}$$
(A14)

and across the meniscus gives

$$P_{Upper} = P_{Bulk} - \frac{2\gamma_m}{R_m(\infty)} \tag{A15}$$

When the bubble is close to the meniscus, a film of bulk liquid of thickness, h, forms between the meniscus and the bubble as depicted in Fig. A1 (b). The mean radius of curvature of the bubble is R(h) and that of the meniscus is $R_m(h)$. Now consideration of the Young-Laplace equation across the bubble interface gives

$$P(h) = P_{Bulk} + \Pi(h) + \frac{2\gamma_b}{R(h)}$$
(A16)

and across the meniscus gives

$$P_{Upper} = P_{Bulk} + \Pi(h) - \frac{2\gamma_m}{R_m(h)}$$
(A17)

From eqn (A15) and (A17) we deduce that

$$\Pi(h) = 2\gamma_m \left(\frac{1}{R_m(h)} - \frac{1}{R_m(\infty)}\right)$$
(A18)

so that for a repulsive disjoining pressure $\Pi(h) > 0$, the bubble will deform the meniscus resulting in a smaller mean radius of curvature of the meniscus: $R_m(h) < R_m(\infty)$. Similarly from eqn (A14) and (A16), and assuming P(h) and $P(\infty)$ are not too different, we get

$$\Pi(h) = 2\gamma_b \left(\frac{1}{R(\infty)} - \frac{1}{R(h)}\right) \tag{A19}$$

so that a repulsive disjoining pressure $\Pi(h) > 0$ means the bubble will have a larger mean radius of curvature at the top pole: $R(h) > R(\infty)$.

The size of the film at equilibrium will be given by balancing the buoyancy force against integrals of pressures acting over the surface of the bubble, A_{bubble} . This can be represented approximately by

$$[P_{Bulk} + \Pi(h)]A_{film} - P_{Bulk}[A_{bubble} - A_{film}] = \frac{4\pi}{3}\rho_b R(\infty)^3 g$$
(A20)

where the first term on the LHS is the total downward force exerted on the bubble by the total pressure in the film of area, A_{film} , the second term on the LHS is the total upward force exerted on the bulk fluid pressure along the bottom surface of the bubble. The RHS is the buoyancy force on the bubble in the bulk liquid of density ρ_b .

A complete solution of the stability of this problem then requires explicit forms of the disjoining pressure, $\Pi(h)$, and expressions for the radii of curvature in differential form and then solving the coupled differential equations.

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