



Short Communication

Stability of thin emulsion film between two oil phases with a viscoelastic liquid–liquid interface

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ARTICLE INFO

Article history:

Received 12 May 2008

Accepted 29 October 2008

Available online 7 November 2008

Keywords:

Film stability

Viscoelastic interface

Surface dilatational viscosity

Surface dilatational elasticity

Surface shear viscosity

Surface shear elasticity

Perturbation growth coefficient

Thin emulsion film

Maxwell model

Viscoelasticity

Linear stability

Liquid–liquid interface

ABSTRACT

The viscoelastic properties of adsorbed protein layer in food emulsions and foams are important in providing stability to such systems. Linear stability analysis for a protein stabilized aqueous film sandwiched between two semi-infinite oil phases with a viscoelastic liquid–liquid interface is presented. The interfacial dilatational and shear viscoelastic properties are described by Maxwell models. The aqueous film is found to be more stable for smaller values of dilatational (shear) relaxation times and larger values of interfacial dilatational (shear) viscosities. The asymptotic values of maximum growth coefficient for very large and very small values of interfacial dilatational (shear) viscosities were found to be independent of relaxation times and correspond to those for immobile and fully mobile liquid–liquid interfaces respectively. The aqueous film is shown to be more stable for larger viscosities of the oil phase with the maximum growth coefficient approaching zero as the ratio of viscosities of oil and aqueous phases approach very large values and an asymptotic value corresponding to that for a foam film for very small viscosity ratios.

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1. Introduction

Thin liquid films are encountered in many food colloidal systems such as foams, emulsions, dispersions etc. Some of the examples of such food systems are whipped toppings, salad dressing, mayonnaise, ice cream etc. The stability of such systems depends on the stability of thin film separating gas bubbles, emulsion droplets and dispersed particles. Food emulsifiers and proteins are usually employed to provide stability to such colloidal systems. In foams and emulsions, liquid from thin films drains due to capillary forces as a result of surface/interfacial tension and radius of curvature of film interface. The capillary pressure, responsible for film drainage, is counterbalanced by intermolecular van der Waals, steric, electrostatic, hydration and depletion interactions whenever the film thickness becomes of the order of a few nanometers so that the film eventually reaches a mechanical equilibrium. These intermolecular interactions are mainly influenced by the emulsifiers and proteins that are employed in such systems. Interfacial tension and repulsive interactions between the two faces of the film will tend to attenuate any imposed perturbations whereas the van der Waals interactions will tend to enhance them thereby re-

sulting in their growth. The film is deemed stable if the imposed perturbations decay whereas the growth of the perturbations will lead to eventual film rupture. Extensive investigations [1–9] on linear stability analysis of equilibrium thin films have been carried out to evaluate the growth of perturbations and the timescale of resulting film rupture. Recent studies have extended the analysis of rupture of thin film on solid [10,11] as well as foam film [12] due to imposed random mechanical perturbations. The effect of these intermolecular interactions is usually accounted for through a disjoining pressure [13] in the normal stress boundary conditions. It has been shown that the imposed perturbations grow whenever this disjoining pressure gradient (with respect to film thickness) is positive [3]. The rupture time of an equilibrium film can be evaluated as the time at which the amplitude of growing imposed perturbation equals one half the film thickness. Previous studies have investigated the effects of density variations [14] and hydrophobic interactions [15] on film stability. Rupture of non-Newtonian [16,17] as well as viscoelastic [18,19] thin films have been investigated. The analysis has been extended to rupture of a draining film due to imposed thermal [13,20] as well as random mechanical [21] perturbations. In food systems, proteins are extensively employed to provide stability to emulsions and foams by modifying the interparticle forces and, more importantly, by providing improved interfacial rheological properties to the oil–water and air–water interfaces. Surface rheological measurements have

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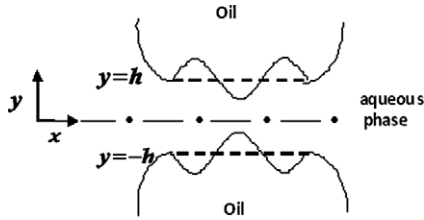


Fig. 1. Schematic of a thin emulsion film between two semi-infinite oil phases.

shown that adsorbed films of disordered proteins (such as α_s1 -casein, β -casein) have a low surface shear viscosity whereas films of globular proteins (such as α -lactalbumin, β -lactoglobulin) are highly viscous [22–25]. The viscoelastic properties of adsorbed protein film are important for stabilizing oil droplet and gas bubbles against coalescence in emulsion and foam systems [26,27]. In a previous study [28], we have investigated the stability of a thin film on a solid surface with a viscoelastic air–liquid interface by linear stability analysis. In this study, we extend this analysis to an aqueous thin film sandwiched between two semi-infinite oil phases accounting for the viscoelastic nature of oil–water interface. The stability analysis gives information on the dependence of maximum growth coefficient and film rupture time on interfacial viscoelastic properties.

2. Governing equations

Consider a thin protein stabilized aqueous film of thickness $2h$ sandwiched between two semi-infinite oil phases. The schematic of the film is shown in Fig. 1. The film is of length L in the x direction and can be considered semi infinite in the other direction. The plane of symmetry is the origin of y direction. Therefore, $y = \pm h$ refer to the top and bottom oil–water interfaces respectively. At $t = 0$, an asymmetric periodic perturbation $f_0 \sin(kx)$ of wavenumber k is imposed on the two faces of the film. The imposition of this perturbation will result in a flow within the film which is also influenced by Marangoni flow. If the perturbation grows, the film will eventually rupture; otherwise the film will be stable. The equations to describe the flow due to imposed disturbance are given by.

2.1. Aqueous film

The continuity equation is

$$\frac{\partial v_x}{\partial x} + \frac{\partial v_y}{\partial y} = 0. \quad (1)$$

Assuming quasi-steady state, the equations of motion are

$$-\frac{\partial p}{\partial x} + \mu \left(\frac{\partial^2 v_x}{\partial x^2} + \frac{\partial^2 v_x}{\partial y^2} \right) = 0, \quad (2)$$

$$-\frac{\partial p}{\partial y} + \mu \left(\frac{\partial^2 v_y}{\partial x^2} + \frac{\partial^2 v_y}{\partial y^2} \right) = 0. \quad (3)$$

The validity of the assumption of quasi-steady state is discussed elsewhere [11]. Since the motion in the liquid film is due to the imposed perturbation, it is reasonable to assume that the film thickness $h(x, t)$, pressure $p(x, y, t)$, velocity in x direction $v_x(x, y, t)$, and velocity in y direction $v_y(x, y, t)$ also have oscillations of the same frequency as the imposed perturbation, i.e.,

$$f(x, t) = f_0 \exp(ikx + \beta t), \quad (4)$$

$$p(x, y, t) = p_{ss} + p'(y) \exp(ikx + \beta t), \quad (5)$$

$$v_x(x, y, t) = v'_x(y) \exp(ikx + \beta t), \quad (6)$$

$$v_y(x, y, t) = v'_y(y) \exp(ikx + \beta t), \quad (7)$$

where p_{ss} is the pressure in the Plateau border and β is the growth coefficient. If β is positive, then the amplitude of perturbation will keep increasing resulting in rupture of the film, otherwise the perturbation decreases and disappears eventually.

Symmetry condition at the midpoint gives,

$$y = 0, \quad \frac{\partial v_x(x, 0, t)}{\partial y} = 0, \quad (8)$$

$$y = 0, \quad \frac{\partial v_y(x, 0, t)}{\partial y} = 0. \quad (9)$$

At the oil–water interface, the kinematic condition gives,

$$y = h, \quad v_y(x, h, t) = \frac{\partial f}{\partial t}. \quad (10)$$

It is to be noted that the surfactant (protein) is soluble only in the aqueous phase. Equation of continuity for the surfactant in the aqueous phase for quasi steady state yields,

$$\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} = 0 \quad (11)$$

with the boundary conditions,

$$x = 0, \quad c = c_0,$$

$$x = L, \quad c = c_0, \quad (12)$$

$$y = 0, \quad \frac{\partial c}{\partial y} = 0. \quad (13)$$

The surfactant flux to the interface can be decomposed into (i) diffusive flux j_{diff} to the subsurface and (ii) adsorptive flux j_{ads} from the subsurface to the interface. These are given by,

$$j_{diff} = -D \frac{\partial c}{\partial y} \quad \text{at } y = y_{sub}, \quad (14)$$

where y_{sub} refers to the location of the subsurface and

$$j_{ads} = -k_{ads}(\Gamma - \Gamma_0(c_{sub})). \quad (15)$$

In the above equation, k_{ads} is the adsorption rate constant, Γ is the surface concentration of surfactant and $\Gamma_0(c_{sub})$ is the equilibrium surface concentration corresponding to the subsurface concentration c_{sub} . The above equation is written for small deviation from the equilibrium [13]. When the energy barrier to adsorption is small (much less than kT), every surfactant molecule is immediately adsorbed upon its arrival at the subsurface. In this case, the surfactant transport to the interface is governed by Eq. (14) [13]. Also, for small subsurface thickness, $y_{sub} \approx h$.

Surfactant balance at the liquid–liquid interface yields,

$$-D \frac{\partial c}{\partial y} \Big|_{y=h} = \frac{\partial}{\partial x} (\Gamma v_x^0) - D_s \frac{\partial^2 \Gamma}{\partial x^2} + \frac{\partial \Gamma}{\partial t}, \quad (16)$$

where D and Γ are the diffusion coefficient and surface concentration of surfactant respectively in the aqueous phase, v_x^0 is the interfacial velocity of the film and D_s is the surface diffusion coefficient. It is to be noted that the surfactant is not soluble in the oil phase. Therefore, oil phase need not be included in the above conservation equation for the surfactant, Eq. (16) can be rewritten as,

$$-D \frac{\partial c}{\partial y} \Big|_{y=h} = \frac{\partial}{\partial x} (\Gamma v_x^0) - D_s \frac{\partial^2 \Gamma}{\partial x^2} + \left(\frac{\partial \Gamma}{\partial c} \right) \frac{\partial c}{\partial t}. \quad (17)$$

For Langmuir adsorption isotherm,

$$\Gamma_0 = \frac{\Gamma_m Kc}{1 + Kc}, \quad (18)$$

where K and Γ_m are constants. Equation (17) can be non-dimensionalized to yield,

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