

Effect of ionic surfactants on drainage and equilibrium thickness of emulsion films

Stoyan I. Karakashev^a, Emil D. Manev^b, Roumen Tsekov^a, Anh V. Nguyen^{a,*}

^a Division of Chemical Engineering, School of Engineering, The University of Queensland, Brisbane, Queensland 4072, Australia

^b Department of Physical Chemistry, Sofia University, 1 J. Bourchier Ave, 1164 Sofia, Bulgaria

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Abstract

This paper presents new theoretical and experimental results that quantify the role of surfactant adsorption and the related interfacial tension changes and interfacial forces in the emulsion film drainage and equilibrium. The experimental results were obtained with plane-parallel microscopic films from aqueous sodium dodecyl sulphate solutions formed between two toluene droplets using an improved micro-interferometric technique. The comparison between the theory and the experimental data show that the emulsion film drainage and equilibrium are controlled by the DLVO interfacial forces. The effect of interfacial viscosity and interfacial tension gradient (the Marangoni number) on the film drainage is also significant.

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1. Introduction

Drainage of thin liquid foam and emulsion films is a major factor influencing the overall behaviour of disperse systems, in which such films are formed from the liquid of the continuous phase. Stability of such dispersions depends on the drainage behaviour of the liquid films and their resistance to coalescence of droplets or bubbles of the disperse phase. The thermodynamic properties and hydrodynamics of thin liquid films have been studied intensively over the past decades [1–4]. Foam and emulsion films possess a number of common characteristics but also exhibit differences. The interaction between the film interfaces, their shape and rheology are significant for kinetics and stability of individual liquid films. For both types of films the presence of a surfactant as a stabilizing component is a crucial factor. For emulsion films, it is known that the effect of the surfactants dissolved in the film on the film drainage is stronger than of those soluble in the disperse phase [5]. All these factors govern the behaviour of colloid systems in industrial applications and can

be used for optimizing the properties of foams, emulsions, and three-phase gas-oil-water mixtures. Therefore, the precise modelling of the key factors is a matter of scientific and industrial interests.

Scheludko [1] was the first to offer a theoretical description of liquid film drainage, applying the Stefan–Reynolds lubrication equation for the film drainage velocity. He assumed a planar film with tangentially immobile interfaces, thinning under the combined action of capillary and DLVO interfacial forces [6,7]. Further studies on film drainage focused on the effects of the interfacial mobility, film thickness non-homogeneity, and non-DLVO interfacial forces [8–14]. The influence of interfacial viscosity on emulsion film drainage was numerically studied by Wasan and co-workers [15]. An analytical drainage model accounting for interfacial viscosity was also obtained by Sharma and Ruckenstein [16] but the interfacial viscosity effect was completely overlooked for many years, until recently Karakashev and Nguyen [17] derived independently and validated an alternative analytical model. The modelling of electrostatic interactions underwent significant changes. The superposition approximation has already been replaced by the semi-numerical and asymptotic solutions of the Poisson–Boltzmann equation [18,19]. Depending on the ad-

* Corresponding author.

E-mail address: anh.nguyen@eng.uq.edu.au (A.V. Nguyen).

sorption layer at the film interfaces, the interfacial charge density can vary with the film thickness, i.e. the so-called charge regulation. Two particular models are well-known in the literature, namely, constant interfacial potential and constant interfacial charge density. The van der Waals disjoining pressure can be described by either the microscopic Hamaker or macroscopic Lifshitz theories. In symmetric thin liquid films the van der Waals disjoining pressure is always negative corresponding to attraction. The theory on non-DLVO interactions in thin liquid films also underwent significant developments. The known steric, structural and hydrophobic interactions are related with the physico-chemical properties of the dispersion phase and the medium, and can have significant impacts on drainage and stability of thin liquid films [20]. Among the other non-DLVO forces, the hydrophobic force is least understood at present although it has been investigated intensively over the last two decades [8,21]. Both the interfacial rheology and the interaction between the films interfaces are related to the adsorption layer. In the foregoing investigations [22] we studied how the interfacial rheology and interfacial forces jointly influence the drainage behaviour of aqueous foam films containing sodium dodecyl sulphate. Significant discrepancy between the experiment and theory was obtained. The analysis showed that the deviation of the theoretical kinetic curve from the experimental one is due to interfacial forces being not described well by the classical DLVO theory. It was therefore demanded to test the new theory on aqueous emulsion films and to compare the results to those obtained with the foam films.

The aim of this paper is to investigate the effect of surfactant adsorption and related interfacial properties on emulsion film drainage. Specifically, the paper examines the drainage pattern and interfacial force interactions in emulsion films produced from diluted aqueous solutions of sodium dodecyl sulphate (SDS) and the related properties of SDS adsorption at the film interfaces.

2. Theoretical background and modelling

For the toluene–water emulsion films, the surfactant is dissolved in water only [23]. Therefore, the surfactant adsorbs from the dispersion medium onto the planar film interfaces, controlling the Marangoni effect and the interfacial forces. Solving the Stokes equation and continuity equation in the lubrication approximation along with the film mass balance and tangential stress boundary equations yields a model for film drainage velocity, $-dh/dt$, described as

$$-\frac{dh}{dt} = \frac{2h^3}{3\mu R^2} \frac{P - \Pi}{f}, \quad (1)$$

where h is the film thickness, P and Π are the capillary and disjoining pressures, respectively, R is the film radius, and μ is the water (dynamic) viscosity. The correction factor f in Eq. (1) accounts for the deviation from the standard Stefan–Reynolds theory [1] for the film drainage velocity with plane-parallel, tangentially immobile interfaces. This factor deviating from unity

due to the interfacial properties can be described as

$$f = 1 - 32 \sum_{k=1}^{\infty} \frac{6/\lambda_k^4}{6 + \text{Bo}\lambda_k^2(h/R) + 2\text{Ma}}, \quad (2)$$

where λ_k is the k th root of the Bessel function of the first kind and zero order. In Eq. (2), $\text{Bo} = \mu_s/(\mu R)$ is the Boussinesq number, where the interfacial viscosity, μ_s , is equal to the sum of the interfacial shear and dilational viscosities. $\text{Ma} = -\Gamma(d\sigma/dc)/(\mu D)$ is the Marangoni number, where Γ is the interfacial excess of the adsorbed surfactant, D is the bulk diffusion coefficient and σ is the interfacial tension, which decreases with increasing the bulk surfactant concentration, c . Eqs. (1) and (2) can be established following the approach described in [17] but the final expression for the correction factor is simplified in this paper by keeping only the first order terms of h/R .

The similarity between Eqs. (1) and (2) for the toluene–water emulsion films and those for the air–water foam films can be justified as follows. Firstly, the previous studies [24] show that in the present case of surfactant soluble only in the film the effect of the surfactant on the flow in the toluene drop is not significant and the film virtually behaves like a foam film. Secondly, the scaling also shows that the normal stress due to the flow inside the toluene drop can be safely neglected because the gradient of the water velocity normal to the film interfaces is proportional to u/h , where u is the interfacial velocity and h is the film thickness, while the velocity gradient inside the toluene drop is proportional to u/R_d , where R_d is of the drop radius. Moreover, the viscosity of toluene (~ 0.00056 Pa s) is significantly lower than that of water. Therefore, the normal viscous stress at the film interfaces from the toluene drops is negligibly smaller than the stress from the film and can be safely neglected as in the case of air–water foam films, given that $h \ll R_d$ which is the case of our experimental system.

According to the DLVO theory the disjoining pressure in Eq. (1) is the sum of the van der Waals and electrostatic components. The van der Waals disjoining pressure can be neglected because the studied emulsion films are relatively thick. The electrostatic disjoining pressure Π_{el} can be predicted by solving the Poisson–Boltzmann equation employing appropriate boundary conditions at the film interfaces. Under the condition of constant interfacial potential, the numerical results of the non-linear Poisson–Boltzmann equation can be semi-analytically represented as [19]

$$\Pi_{\text{el}}(h) = 32c_{\text{el}}R_gT \tanh^2\left(\frac{y_0}{4}\right) \left\{ \frac{1}{1 + \cosh \kappa h} + g(y_0) \sinh^2 \frac{y_0}{4} \exp[-g(y_0)\kappa h] \right\}, \quad (3)$$

where R_g is the universal gas constant, c_{el} is the molar concentration of electrolytes in the solution (the SDS concentration in the present case), T is the absolute temperature. The Debye constant for a binary electrolyte of valence z is defined as $\kappa = (2c_{\text{el}}F^2z^2/\varepsilon\varepsilon_0R_gT)^{1/2}$, where ε_0 is the permittivity of vacuum. The dimensionless interfacial potential is defined as $y_0 = zF\psi_s/R_gT$, where F is the Faraday constant and ψ_s is the interfacial potential. For $|y_0| \leq 7$, function $g(y_0)$ is defined

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