

Drainage and critical thickness of foam films from aqueous solutions of mixed nonionic surfactants

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Abstract

The thinning and the critical thickness (of rupture or “black spots” formation) of foam films from aqueous solutions of mixed nonionic surfactants are studied under varied experimental conditions, as a function of film radius (0.05–0.15 mm), surfactant concentration (0.01–1.0 CMC) and ionic strength (0.001–0.1 M NaCl). The experimental values of the drainage coefficient (α), determined from the film thickness versus time dependences, were used to calculate the theoretical values of the film critical thickness.

The real velocity of film thinning is a major factor in the process of reaching the state of kinetic instability when approaching the critical thickness (Scheludko’s criterion). The classical equation used to describe the film thinning rate, proposed and named by Scheludko (1955) “Reynolds Law”, is applicable for small film radii ($r < 0.05$ mm). At larger radii the velocity of thinning follows the equation of Manev et al. [E.D. Manev, R. Tsekov, B. Radoev, J. Colloid Interf. Sci. 18 (1997) 769], which takes into account the effect of the film thickness local non-homogeneity.

The studied stabilizing surfactants include *n*-dodecyl- β -D-maltoside (β -C12G2) and hexaethyleneglycol monododecyl ether (C12E6). Along with confirming the dependences following from the theories of the critical thickness [B. Radoev, A. Scheludko, E.D. Manev, J. Colloid Interf. Sci. 95 (1983) 254] and film thinning [E.D. Manev, R. Tsekov, B. Radoev, J. Colloid Interf. Sci. 18 (1997) 769], the results of the present investigation established also certain deviations for films stabilized with mixed surfactants (β -C12G2 + C12E6). The effectiveness of the empirical equation, employing the drainage coefficient (α) to describe the film thinning, is emphatically proven here.

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1. Introduction

The stability of the disperse systems is an essential topic in surface and colloid science, for their behaviour is of paramount importance in chemical science and practice, as well as in the adjacent fields of biology and biotechnology, medicine, pharmacology, environmental science, etc. In all these domains of knowledge and technology it is often necessary to have means of control over the colloidal stability. Studying the hydrodynamics and stability of the thin liquid films that build the disperse sys-

tems is an efficient tool for this task. A number of comprehensive reviews dedicated to the subject have been released during the past decades, among which those of Mysels et al. [1], Scheludko [2], Ivanov [3] and Exerowa and Kruglyakov [4].

The correct description of the process of spontaneous self-destruction of the thin liquid films at the so-called ‘critical thickness’ is essential for the understanding of their kinetic and thermodynamic stability. The critical thickness and the factors that govern this phenomenon have been the subject of many theoretical and experimental investigations [5–21]. The kinetic stability of thin liquid films and the role of their drainage behaviour have been reviewed recently by Manev and Nguyen [22,23]. It well known that the presence of a surfactant is a necessary condition for the formation of stable foam; it is the surfactant adsorption at the water–air interface that provides the

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films with the ability to resist the local thinning during the process of drainage. The concentration of the surfactant as well as its molecular structure strongly influences the film stability.

In practice, single (individual) surfactants are seldom used as stabilizers of colloidal systems. Hence, investigations of films stabilized by surfactant mixtures are of importance. However, only a few such studies of films from solutions of well-defined surfactant mixtures are so far available in the literature. To our knowledge, the only foam film studies with surfactant mixtures are those in which technical grade surfactants were used [24–26].

The present study is dealing with mixtures of nonionic surfactants accentuating on mixtures of *n*-dodecyl- β -D-maltoside (β -C₁₂G₂) and hexaethyleneglycol monododecyl ether (C₁₂E₆). Together with a preceding study of film drainage [27], they constitute parts of a comprehensive European project, funded by the European Commission, on the interaction forces, adsorption properties, surface rheology, and bulk properties of these mixtures. The reason for studying mixtures of ethylene oxide (C_{*i*}E_{*j*}) and sugar (C_{*n*}G_{*m*}) based surfactants is that these surfactants – in spite of both being nonionic – behave quite differently. For example, the hydration number of these two surfactants and the flexibility of their head groups are different (while a maltoside unit behaves like a hard disc, the ethylene oxide units is more like a short polymer chains). Thus, they have different pH sensitivity: the ethylene oxide unit responds to pH-variations by changing its hydration degree (or its conformation) while the hydration degree of sugar unit is pH insensitive, etc. [28–31].

Taking into account the differences between ethylene oxide (C_{*i*}E_{*j*}) and sugar surfactants (C_{*n*}G_{*m*}) one can deduce that the films stabilized by mixture of these surfactants shall behave differently, depending on the surfactant composition of the mixture. It is of interest to understand whether the ethylene oxide or the sugar-based surfactant dominates the behaviour of the mixture when they are in compatible ratios, or when one of the surfactants prevails in quantity (e.g., C_{*n*}G_{*m*} with traces of C_{*i*}E_{*j*}, or vice versa) affect the overall properties of the system.

The aim of the presented study is to reach the answers to such questions by investigating the characteristics of foam films stabilized by the individual β -C₁₂G₂ and C₁₂E₆ surfactants and their mixtures. The drainage of foam films was studied as a function of the electrolyte (0.001–0.1 M NaCl) and the total surfactant concentration (0.1–1.0 CMC), respectively.

It is established theoretically that the ‘critical thickness’, that is, the thickness manifesting the end of existence of the thinning film, is related to the rate of film drainage. The latter is determined by the film size and the interactions in the film, as well as the surface tension and viscosity of the parent solution. That is why the critical thicknesses of all investigated films were measured interferometrically under well-defined conditions, necessary to match the theoretical investigations. This permits to compare the obtained data to the values calculated through the Radoev–Scheludko–Manev (RSM) equation [11], while employing different expressions for the rate of film thinning, as the so-called “Reynolds Law” (Re) and the Manev–Tsekov–Radoev (MTR) formula.

2. Theory

2.1. Film drainage

2.1.1. Reynolds equation

For the rate of thinning (*V*) of a microscopic horizontal circular film (of radius *r*) with plane-parallel and tangentially immobile surfaces, Scheludko [2] has proposed the expression:

$$V_{\text{Re}} = -\frac{dh}{dt} = \frac{2h^3 \Delta P}{3\mu r^2} \quad (1)$$

where V_{Re} is the rate of film thinning; *h* is the film thickness; μ is the dynamic viscosity; *r* is the film radius; $\Delta P = P_c - \Pi$ is the driving pressure of film thinning (P_c is capillary pressure; Π is disjoining pressure).

Eq. (1) is analogous to the expression derived by Reynolds for the velocity of mutual approach of two parallel rigid disks, separated by liquid layer. Scheludko [2] has named Eq. (1) ‘Reynolds Law’ or ‘Reynolds Equation’; respectively, the rate of thinning calculated from Eq. (1) is called “Reynolds velocity” (V_{Re}). Many experimental studies have shown that Eq. (1) is satisfactorily applicable only for circular liquid films of small diameter (below 0.1 mm).

2.1.2. Manev–Tsekov–Radoev formula

The experiment [12,13] has shown that the deviations in film thinning rate from the classical Reynolds equation are due to thickness non-homogeneities, which strongly increase with the film size and persist during its evolution up to the critical thickness. Manev, Tsekov and Radoev [11] have assumed that in the process of thinning larger films disintegrate into an ensemble of several smaller centers with accelerated drainage. As a result of the quantitative description of the thickness non-homogeneity, a new formula for the rate of film thinning as function of film size was put forward:

$$V_{\text{MTR}} = \frac{1}{6\mu} \sqrt[5]{\frac{h^{12}(P_\sigma - \Pi)^8}{4\sigma^3 r^4}} \quad (2)$$

where V_{MTR} is the velocity of film thinning and σ is the surface tension of the liquid phase.

According to Eq. (2), the rate of thinning (*V*) is proportional to $r_f^{-4/5}$ and therefore its dependence on film size is much weaker than that theoretically predicted by Eq. (1) ($V \sim r_f^2$). Besides, according to Eq. (2) the rate of thinning does not depend on the driving pressure ΔP in the mode required by the Reynolds equation and includes an explicit dependence on surface tension, in addition to the implicit dependence through the capillary pressure.

2.2. Critical thickness of thin liquid films

Scheludko [5] was the first to establish the necessary condition for film rupture (*Scheludko’s Criterion*). According to it the liquid film reaches upon thinning the state of kinetic instability where, due to spontaneously growing thermal fluctuations (surface waves), collapse of the film occurs at its critical thickness.

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