THE INFLUENCE OF SURFACTANTS ON THE DRAINAGE AND RUPTURE OF MOBILE LIQUID FILMS BETWEEN DROPS: A PARAMETRIC NUMERICAL STUDY

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ABSTRACT

Film drainage and rupture during coalescence between two drops approaching each other under a constant force in the presence of insoluble surfactants affected by the van der Waals forces is studied numerically. The mathematical problem is based on the coupled equations of flow in each phase and the convection-diffusion equation governing the surfactant distribution at the interface as well as the related Marangoni effects. The latter are subject to the boundary conditions in the limit of gentle interactions (small-deformations) for which the drops are nearly spherical, except in the near-contact region, where a flattened thin film forms. The finite difference method is used to discretize the lubrication equation in the gap and the interfacial diffusion-convection equation while the boundary integral method is employed to solve the flow in the drops. In this work, a parametric study is carried out by a numerical simulation for Peclet numbers (Pe) ranging from $10^2$ to $10^4$, initial dimensionless surfactant concentrations ($\Gamma^*_0$) from 0 to 10 and dimensionless Hamaker parameter ($A^*$) from $10^{-7}$ to 10. The results indicate that the critical film thickness ($h_c$) is strongly dependent on the combination of the three parameters which determines the predominance of van der Waals forces or those of Marangoni.

Keywords: film drainage and rupture, drops, insoluble surfactant.

INTRODUCTION

The understanding of coalescence phenomena is important for many liquid-liquid systems. Coalescence or its prevention is an essential element in the preparation and stability of emulsions and foams, liquid-liquid extractions, emulsification, separation, multiphase transportation and other technical problems referring to liquid-liquid dispersions [1]. The design of nuclear reactors, chemical reactors, boiling and condensation equipment are some examples of such applications. Most of the time the liquid-liquid dispersions contain surfactants that can strongly influence the hydrodynamics and the interactions between the drops. For pure systems, numerical studies of the coupled processes of drop deformation and film drainage under the action of a constant interaction force with and without van der Waals forces have been carried out by many authors [2 - 8]. The results show that the drop begins to flatten near the axis of symmetry whereas the film thinning rate is larger near the
edges of the flattened regions. As time progresses, the interface forms a dimple and the film thinning process slows down. The case of constant approach velocity of the drops has also been studied in the partially mobile case, with and without van der Waals forces [9 - 10]. The presence of surfactants or mass transfer can have a significant influence on the coalescence of drops. The effect of a surfactant or interphase mass transfer on film drainage arises from the extra “Marangoni” forces generated by gradients of the interfacial tension, associated with variations of the solute concentration over the interface [1, 10, 11 - 12]. Yeo et al. [10] investigated numerically the drainage of the film between two drops approaching one another at constant velocity in the presence of an insoluble surfactant. Chesters & Bazhlekov [13] have presented several calculations of the film drainage between drops colliding at a constant force in the presence of an insoluble surfactant. The case of tangentially mobile interfaces has been discussed but only for purely convective surfactant distribution and in the absence of van der Waals forces. More recently, Alexandrova [14] has investigated numerically the drainage of the film between two drops approaching one another at a constant force in the presence of an insoluble surfactant. In the absence of a surfactant, the numerical results have been validated against the numerical studies of Rother et al. [4] and Bazhlekov et al. [5]. The results have also confirmed the long-time asymptotic equations of Nemer et al. [7]. In this study, which is an extension of the work of Alexandrova [14], a further detailed parametric study is carried out using a numerical simulation. Particular interest is focused on how the Peclet number, the van der Waals forces and the surfactant concentration affect the film drainage and rupture. Our study is limited to “tangentially-mobile” interfaces, for which the time scale for near contact motion is determined by the drop phase viscosity [2]. In this case, the results apply to most liquid-liquid systems with the exclusion of the extreme viscosity ratios (fully mobile interfaces, immobile interfaces). We solve the mathematical problem consisting of the coupled equations of flow in each phase and the convection-diffusion equation governing the surfactant distribution at the interface as well as the related Marangoni effects. The latter are subjected to the boundary conditions. The following part describes the equations governing the film drainage and rupture in the presence of surface-active species together with the relevant scal-

FORMULATION OF THE PROBLEM

Two drops of deformable interfaces subjected to a constant interaction force are considered in the presence of van der Waals intermolecular forces and insoluble surfactants. Both the dispersed phase and the continuous phase are assumed immiscible, Newtonian and incompressible. The study examines the class of gentle, axi-symmetrical interactions, for which the equations governing film drainage assume a universal form in the pure liquid case [3 - 5, 14]. By “gentle” it is meant that the film radius is much smaller than that of the drop, i.e.

\[
a/R << 1, \tag{1}
\]

where \(a\) stand for the film radius (Fig. 1). It is related to the interaction force \(F\) by:

\[
F = \pi a^2 \frac{2\sigma_0}{R} \tag{2}
\]

Fig. 1. Schematic representation of two coalescing drops.
where \( \sigma_0 \) is a characteristic value of the interfacial tension. The governing equations are identical for unequal and equal drops provided the equivalent radius, \( R_{eq} \), is used:

\[
\frac{1}{R_{eq}} = \frac{1}{2} \left( \frac{1}{R_1} + \frac{1}{R_2} \right)
\]

(3)

The other restrictions referring to the drainage hydrodynamics, which have a broad range of applicability [3 - 5] are as follows: (i) the inertial forces are disregarded; (ii) the plug flow in the film is approximated; (iii) the viscous normal stresses are disregarded; (iv) the body forces due to gravity are disregarded. In addition, the restrictions concerning the surfactant refer to: (v) a low surfactant concentration; (vi) an insoluble surfactant in both phases.

In the small surfactant concentration range considered in this study the magnitude of the surface tension is linearly related to the surfactant surface concentration \( \Gamma \).

Here, we use the expression given by Stone & Leal [25]:

\[
\sigma_0 - \sigma = \Gamma R_g T
\]

(4)

where \( \sigma_0 \) and \( \sigma \) are the values of the surface tension in the absence and in the presence of a surfactant respectively, \( R_g \) is the gas constant, while \( T \) is the absolute temperature.

**GOVERNING HYDRODYNAMIC EQUATIONS**

On the ground of the preceding approximations, the continuity and Stokes equations result in thin film equations coupling the lubrication flow in the gap region to the flow within the drops [13, 14].

Following Saboni et al. [3], Chesters & Bazhlekov [13] and Alexandrova [14], we introduce dimensionless variables in order to reduce the numbers of parameters that occur explicitly. Accordingly, the dimensionless variables (radial coordinate \( r^* \), film thickness \( h^* \), time \( t^* \), pressure \( p^* \), tangential stress \( \tau^* \), tangential interfacial velocity \( u^* \) and surfactant concentration \( \Gamma^* \) ) are related to the dimensionless ones by the relations:

\[
\begin{align*}
    r^* &= \frac{r}{a' R_{eq}}; \quad h^* &= \frac{h}{a'^2 R_{eq}}; \quad t^* = \frac{t \sigma_0 a'}{\mu R_{eq}}; \\
p^* &= \frac{p R_{eq}}{\sigma_0}; \quad \tau^* &= \frac{\tau R_{eq}}{\sigma_0 a'}; \quad u^* = \frac{u \mu}{\sigma_0 a'^2}; \quad \Gamma^* = \frac{\Gamma R_g T}{\sigma_0 a'^2}
\end{align*}
\]

(5)

where \( a' \) is a dimensionless film radius defined as \( a' = a / R_{eq} \).

Thus, the equations governing the drainage contain three dimensionless parameters given as:

\[
A^* = \frac{A}{4 \pi \sigma_0 R_{eq}^2 \sigma^*}; \quad \Gamma_0^* = \frac{\Gamma R_g T}{\sigma_0 a'^2}; \quad Pe = \frac{\sigma_0 R_{eq} a'^2}{D \mu}
\]

The film thickness and the tangential stress are given respectively by:

\[
\frac{\partial h^*}{\partial t^*} = - \frac{1}{r^*} \left( \frac{r^* h^* u^*}{r^*} \right)
\]

(6)

\[
\tau^* = - \frac{h^* \partial p^*}{2 \partial r^*}
\]

(7)

The small-slope restriction (Eq. (1)) implies that the interface may be approximated as a plane using the drop circulation scale. As the inertial forces are negligibly small (Stokes flow), the boundary integral theory provides a relation between the interface velocity and the shear stress exerted on the interface by the dispersed phase:

\[
\Phi(r^*, \rho^*) = \int_0^{\pi} \Phi(\rho^* \theta) r^* d\rho^*
\]

(8)

There

\[
\Phi(r^*, \rho^*) = \frac{1}{2 \pi} \frac{1}{\sqrt{r^* - 1}} \int_{\rho^*}^{\rho^*} \frac{\cos \theta d\theta}{\sqrt{1 - k^2 - \cos \theta}}
\]

(9)

is an elliptic-type Green’s function kernel having an integrable (logarithmic) singularity at \( \rho^* = 1 \) (k = 1), where:

\[
k^2 = \frac{2r^* \rho^*}{r^* - \rho^*}
\]

(10)

The first interface condition, i.e. the velocity continuity, has been tacitly incorporated in Eq (6). The second condition requires the balance between the tangential stresses exerted by the adjacent phases on the interface and the Marangoni stress component due to the interfacial tension gradient:

\[
\tau^*_d = \tau^* + \frac{\partial \Gamma^*}{\partial r^*}
\]

(11)

where \( \tau_d \) and \( \tau \) are the tangential stresses acting on
the interface by the drop and the film.

The third interface condition refers to the normal stress jump associated with the interfacial tension:

\[
p^* = \frac{1}{2} \frac{\partial^2 h^*}{\partial r^2} + \frac{1}{r^* \partial r^*} + \frac{2A^*}{3h^3} \tag{12}
\]

where \(A\) is the Hamaker parameter due to the presence of van der Waals interactions [3, 4, 9].

The force balance is described by:

\[
\int_0^{r_{\text{large}}} \left( p^* - \frac{2A^*}{3h^3} \right) r^* dr^* = 1 \tag{13}
\]

The initial film thickness condition refers to spherical drops at a large separation \((h_0^*)\), i.e.:

\[
h^* (r^*, t^* = 0) = h_0^* + r^*^2 \tag{14}
\]

EQUATIONS GOVERNING THE SURFACANT TRANSPORT

The surfactant concentration along the interface is evaluated by solving the surface convective-diffusion equation:

\[
\frac{\partial \Gamma^*}{\partial t^*} + \frac{1}{r^*} \frac{\partial \left( r^* \Gamma^* \mu^* \right)}{\partial r^*} - \frac{1}{Pe_s} \frac{\partial}{\partial r^*} \left( r^* \frac{\partial \Gamma^*}{\partial r^*} \right) = 0 \tag{15}
\]

The boundary condition imposed across the axis of symmetry is that of no flux at \(r = 0\):

\[
\frac{\partial \Gamma^*}{\partial r^*} \bigg|_{r^* = 0} = 0 \tag{16}
\]

The boundary condition imposed far from the film region is that of a uniform surfactant distribution:

\[
\frac{\partial \Gamma^*}{\partial r^*} \bigg|_{r^* = r_{\text{large}}} = 0 \tag{17}
\]

Initially, the insoluble surfactant of a concentration \(\Gamma_0^{*}\) is uniformly distributed on the interfaces:

\[
\Gamma^* (r^*, t^* = 0) = \Gamma_0^* \tag{18}
\]

The coupled equations of flow in each phase and the convection–diffusion equation for the surfactant at the interface are solved using finite difference approximations [14]. Given the thickness and the surfactant concentration at time-step \(n\), the thickness and the surfactant concentration at \(n+1\) are respectively derived from the continuity equation (Eq. (6)) and the diffusion-convection equation (Eq. (15)). Then the pressure is obtained from the normal stress (Eq. (12)). The calculated pressure is used in Eq. (7) to find the shear stress exerted by the film on the drop. Eq. (11) is used to find the shear stress exerted on the film by the drop, which, via the boundary integral method (Eq. (8) and Eq. (9)), gives the velocity at the interface.

RESULTS

This section presents the results of a parametric numerical study in which the deformation, drainage and rupture of the film have been carried out for a range of values of the dimensionless surfactant concentration, Peclet number, and dimensionless Hamaker parameter. Simulations are made using values of the Peclet number and the initial surfactant concentration not only encountered in practice but also consistent with the model assumptions. The product, \(\mu_sD_s\), is typically of the order of \(10^{-12}\) N, \(\sigma_0\) is also of the same order in most systems, i.e. \(\sigma_0 = 2.5x10^{-2}\) N/m, while the radius, \(R\), is usually of the order of 1 mm. The value of \(Pe_s\) is thus primarily determined by \(a'\). Since the highest value of \(a'\) compatible with the requirements of Eq.(1) is about 0.1, \(Pe_s\) becomes equal to \(2.5 \times 10^4\). In view of \(R_G = 8.314\) J/mol K and \(T=293\) K, the value of the initial surfactant concentration,

\[
\Gamma_0^* = \frac{\Gamma R_G T}{\sigma_0 a'^*} \tag{19}
\]

used in the present simulations corresponds to \(\Gamma_0^* \sim 10^{-7} \Gamma_0\). That is, numerical computations have been carried out for Peclet numbers ranging from \(10^2\) to \(10^4\), initial dimensionless surfactant concentrations varying from 0 to 10 and dimensionless Hamaker parameter, \(A^*\), having values from \(10^{-7}\) to 10. Hereafter all simulations are carried out with an initial separation between the two drops taken equal to 4.

In order to validate the numerical model, we consider initially film drainage and film rupture in the absence of a surfactant. This problem has already been treated [3 - 5, 14]. Fig. 2 displays the decrease of the minimum film thickness \((h_{\text{min}}^*)\) with \(t^*\) for different values of the dimensionless Hamaker parameter \((A^*)\). In the absence of van der Waals attraction \((A^*=0)\), the evolutions of \(h_{\text{min}}^* (t^*)\) show fast decrease followed by an asymptotic behavior. The presence of van der Waals interactions...
forces results in an additional force on the film which becomes significant once the film is sufficiently thin. In case the film thickness becomes small enough for van der Walls term domination, the drainage accelerates locally and the zero film thickness is attained in finite time, \( t^* \). According to Saboni et al. [3] and Chesters & Bazhlekov [13], the effective critical film rupture thickness, \( h^*_c \), may be defined as the value of \( h^*_{\text{min}} \) in the absence of van der Walls forces (\( A^* = 0 \)). The time rupture and the critical film thickness are illustrated in Fig. 2 by the dashed lines for \( A^* = 10^6 \). The obtained results show that the critical rupture thickness decreases with decrease of the van der Waals forces and that the film rupture time decreases with increase of these same forces. The present results are in good agreement with those obtained by Rother et al. [4].

In order to follow the time dependence of the surfactant effect on the film drainage, the dimensionless film thickness is presented as a function of the dimensionless radial position in Fig. 3 for different dimensionless forces.
times ($t^* = 5, 10, 50, 100, 500$ and $1000$). Fig. 3a shows the process of film formation which involves flattening followed by development of dimples in surfactant absence. The evolution of the film thickness profiles for initial surfactant concentration ($\Gamma^*_0 = 1$) and three Peclet numbers ($Pe = 100, Pe = 1000$ and $Pe = 10000$) is illustrated in Figs. 3b - 3d. It can be easily juxtaposed to the reference case (Fig. 3a). The results show that the surfactant effect overcomes the hydrodynamic resistance of the film pressure. Hence, the film thins faster at the centre but not in the downstream regions. Indeed, Fig. 3c shows that in case $Pe = 1000$, unlike the case of reference, $h^*_{\text{min}}$ is located not at $r^* = 1$ but at $r^* = 0$. This in turn involves the dimple removal. In case $Pe = 10000$ (Fig. 3d), one can make the same observations concerning the suppression of the dimple. Fig. 4 displays the dimensionless interfacial surfactant gradient distribution $\frac{\partial \Gamma^*}{\partial r^*}$ for an initial surfactant concentration $\Gamma^*_0 = 1$ and two Peclet numbers: (a) $Pe = 10^2$, (b) $Pe = 10^4$. It may be noted that the interfacial surfactant gradient decreases rapidly for $Pe = 10^2$. The decrease is much smaller for $Pe = 10^4$. In the latter case, the Marangoni stress component is still important and implies a very different drainage than in the surfactant absence (Fig. 3a), (Fig. 3d).

Fig. 5 presents the evolution of the minimal film thickness as a function of time for different values of $A^*$, a fixed value of Peclet number ($Pe = 10000$) and a fixed initial surfactant concentration ($\Gamma^*_0 = 0.25$). Fig. 6 presents the evolution of the minimal film thickness as a function of time for different values of $A^*$, a fixed value of Peclet number ($Pe = 10000$) and two initial surfactant concentrations ($\Gamma^*_0 = 1$ and $\Gamma^*_0 = 2$). Figs. 5 and 6 indicate that the critical film thickness, $h^*_{\text{min}}$, is greatly dependent on the surfactant amount on the interfaces. Fig. 5 shows that the profiles of the minimal film thickness are slightly similar to those in surfactant absence (Fig. 2). This means that the van der Waals forces predominate when compared to the Marangoni one in case of low concentrations. The power balance can be reversed at higher concentrations. Thus Fig. 6a shows that the profiles of the minimum film thickness in case of $A^* < 10^{-3}$ cross with referring to $A^* = 0$. The film rupture does not occur because the van der Waals forces fail to counter those of Marangoni. This phenomenon is even more pronounced at higher concentrations (Fig. 6b).

Fig. 7 presents the evolution of the minimal gap thickness ($h^*_{\text{min}}$) as a function of time for different values of Peclet number, a fixed Hamaker parameter ($A^* = 10^{-3}$) and two initial surfactant concentrations ($\Gamma^*_0 = 1, \Gamma^*_0 = 2$). Fig. 7 shows that the Peclet number increase implies a sharp increase of the coalescence time. By eliminating the surfactant, the flow in the film causes a depletion in the surfactant concentration near $r^* = 0$. This decrease continues until the concentration difference between

![Fig. 4. Variation of interfacial surfactant gradient, $\frac{\partial \Gamma^*}{\partial r^*}$, as function of dimensionless radial position, $r^*$, and dimensionless time, $t^*$, (a) $\Gamma^*_0 = 1$, $Pe = 10^2$; (b) $\Gamma^*_0 = 1$, $Pe = 10^4$.](image-url)
those at the center and the periphery of the film becomes large enough so that the Marangoni stresses arise leading to a reverse flow. The latter will carry the surfactant into the surface active-depleted area. Stopping of the drainage is observed when the two flows cancel. This is the case of a very high Peclet number for which \( h^*_{\text{min}} \) reaches a limiting value which indicates that the drainage has completely stopped. For a low Peclet number the drainage stop is only temporary, which is particularly the case for \( \text{Pe} = 100 \).

Fig. 8 shows the evolution of the film thickness profiles for a fixed value of the Hamaker parameter \( A^* = 10^{-3} \) in absence of a surfactant (Fig. 8a) and in presence of a surfactant (Figs. 8b - 8d) for three Peclet numbers (\( \text{Pe} = 100, \text{Pe} = 1000 \) and \( \text{Pe} = 10000 \)) and a fixed initial surfactant concentration \( \Gamma^*_{0} = 1 \). The process of film formation which involves flattening followed by the development of dimples in absence of a surfactant is illustrated in Fig. 8a. The comparison with the drainage behavior in the absence of van der Waals forces (Fig. 3a) reveals similar behavior up to \( t^* \) around 10, which is then followed by an intense acceleration of the local thinning rate. For a relatively low Peclet number (\( \text{Pe} = 100 \)) the film thinning occurs rapidly, while the rupture takes place in the nose region unlike the case in absence of van der Waals, where the flattening is followed by dimples development (Fig. 3b). A lack of dimple is observed at \( \text{Pe} = 1000 \) (Fig. 8c). It is worth noting that no van der Waals forces are acting (Fig. 4c). In this case the rupture of the film proceeds much later than in the case referring to \( \text{Pe} = 100 \). Fig. 8d shows that the drainage of the film is much slower for \( \text{Pe} = 10000 \) than that at lower Peclet numbers (Figs. 8b and 8c) and besides no film rupture occurs.
Fig. 7. Dimensionless minimum film thickness, $h_{\text{min}}^*$, versus dimensionless time, $t^*$, for different values of Peclet number, a fixed value of the Hamaker parameter $A^* = 10^{-3}$ and a fixed initial surfactant concentration, a) $\Gamma_0 = 1$, b) $\Gamma_0 = 2$.

Fig. 8. Dimensionless film thickness, $h^*$, as function of dimensionless radial position, $r^*$, and dimensionless time, $t^*$, for a fixed value of the Hamaker parameter $A^* = 10^{-3}$: (a) in the absence of surfactant, (b) $\Gamma_0 = 1$, $Pe = 10^2$, (c) $\Gamma_0 = 1$, $Pe = 10^3$, (d) $\Gamma_0 = 1$, $Pe = 10^4$. 
We now focus on the combined effect of an insoluble surfactant and van der Waals forces on the film rupture at a high Peclet number. In this case the film drainage and rupture may be strongly affected. The calculated values of the critical film thickness are presented in Table 1 for different values of the Hamaker parameter $A^*$ ($10^{-7} \leq A^* \leq 10$), for different initial surfactant concentration ($0 \leq \Gamma_0^* \leq 8$), and a fixed Peclet number, $Pe = 10000$. Table 1 shows that the critical film thickness increases with the Hamaker parameter increase. It is also evident that the van der Waals forces are always predominant involving film rupture at relatively low concentrations ($\Gamma_0^* \leq 0.25$). For relatively very high concentrations ($\Gamma_0^* \geq 8$), the Marangoni forces dominate and coalescence is no longer possible. For intermediate cases, combinations of the Hamaker parameter and the initial surfactant concentration determine the predominance of the van der Waals forces or those of Marangoni.

### CONCLUSIONS

A numerical study has been carried out to investigate the effects of a surfactant on the coalescence in liquid–liquid dispersions. This paper presents the results of a parametric numerical study in which the film drainage and rupture are computed over the ranges of $100 < Pe < 10000$, $10^{-7} < A^* < 10$ and $0 \leq \Gamma_0^* \leq 10$. A retardation of the coalescence process is found for intermediate Peclet number, while the dimple formation is suppressed and the initial drainage rates decrease significantly at a very high Peclet number. The results show a strong dependence of the film drainage and rupture on the values of the Peclet numbers, the initial surfactant concentration and the Hamaker parameter. The critical film thickness, $h^*_c$, is strongly dependent on the combination of those three parameters as it determines the predominance of the van der Waals forces or those of Marangoni. While the present results are encouraging vis a vis, the modeling of the coalescence processes in systems containing a surfactant, it should be emphasized that the theoretical and computational framework developed concerns an insoluble surfactant. The study of soluble surfactants present in two phases does not present any particular problems. But it requires the introduction of two additional parameters related to the diffusion-convection equation in each phase. Such a study has been started and will be presented in a forthcoming article.

### Acknowledgements

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### NOTATIONS

- $a$ film radius [m]
- $a'$ dimensionless film radius, $a' = a/R_{eq}$ [-]
- $A$ Hamaker parameter [-]
- $D$ molecular diffusivity [m$^2$/s]
- $F$ interaction force exerted by one drop on another [N]

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Table 1. The critical film thickness, $h^*_c$, as a function of $A^*$ and different initial surfactant concentration for $Pe=10000$. The symbol - means that no coalescence is predicted.
REFERENCES