ABSTRACT

Film drainage between two drops under a constant interaction force is studied numerically using computational fluid dynamic technique. The interaction and deformation of drops in axisymmetric configuration are considered without assumptions about the dimensions of the film radius. Arbitrary Lagrangian–Eulerian (ALE) formulation including Stokes equations in both, dispersed and continuous phases, is used to allow interface tracking. The model is used to investigate the time-dependent deformation of the drop interface and the drainage of the film between drops. Comparisons with numerical results from existing theoretical lubrication models, with small dimple approximate, have been performed, and a good agreement was found. ALE simulations were also used to explore near-contact drop dynamics under finite-deformation conditions i.e. when the film radius is of the same order of magnitude of drop radius. The numerical results show that film drainage between the two drops depends strongly on the interface deformation as well as on the dispersed to continuous-phase viscosity ratio (\( \lambda \)).

Keywords: film drainage, coalescence, drops with finite-deformations.

INTRODUCTION

Understanding the coalescence phenomena between two drops 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, separation, multiphase transportation and other technical problems concerned with liquid-liquid dispersions. In multiphase flows, the evolution of the dispersed phase volume fraction and final drop-size distribution depends on the prevalence of drop coalescence or break-up. Kamp et al. [1] provide a detailed analysis of the effectiveness of coalescence and influencing parameters. As pointed out by Kamp et al. [1] among others, coalescence investigations are still an area of active research. Following Yiantsios and Davis [2 - 3], many studies of two-drop interactions assume small-deformation conditions (drops are nearly spherical away from the near contact region) and use the lubrication theory. Numerical solutions of the equations governing film drainage have been obtained for both constant-force and constant-velocity interactions in the presence or in the absence of van der Waals forces [4 - 9]. In the limit of small-deformation conditions, numerical solutions were also obtained for film drainage in the presence of mass transfer or surfactants [10 - 13]. Nemer et al. [14] presented a long-time asymptotic analysis for drops with tangentially mobile interfaces under the action of a body force in a quiescent fluid in the limit of small-deformation conditions. The predictions of their theory are quantitatively confirmed by their thin-film simulations. For drops with larger deformation conditions, many studies [11, 15 - 18] used Boundary Integral Method (BIM) for which only a discretization of the interfaces is
necessary. BIM method, despite its efficiency and its low computational cost, has some limitations. According to Nemer et al. [14], BIM simulations are unable to resolve the long-time asymptotic behavior of the film drainage dynamics. An additional problem was pointed by Jansen and Anderson [15] concerning BIM simulations for non-unity viscosity ratios (λ ≠ 1). Another drawback of this method is that the formulation only exists for Newtonian Stokes regime and there are no perspective to include inertia terms or mass transfer effects. We have thus to resort to other methods. An alternative approach that considers the continuity and momentum equations, which fundamentally describe the fluid dynamics can overcome the deficiencies of the BIM method and thin films models. Such approach can be achieved through an implementation of a Computational Fluid Dynamic (CFD) method. This method is used in the present study in which we use Arbitrary Lagrangian–Eulerian (ALE) formulation to allow interface tracking. ALE is based on the coupling of Eulerian and Lagrangian formulations. This hybrid approach combines the best features of both the Lagrangian and Eulerian descriptions while minimizing their disadvantages. For more details on ALE method we refer to Donea et al. [19].

In this work, we investigated the drainage and deformation of two drops in interaction under the action of a constant force. Unlike the lubrication theory based among others on the assumption of a small interface deformation, no simplifying assumptions were used for the solution of equations. The Stokes equations inside and outside the drops are solved in ALE formulation with moving mesh implanted in Comsol 4.2. Film drainage will be studied and the results obtained will be compared with results from thin films models and analytical solutions derived from asymptotic theory. Of particular interest to this study are the wider range in which the assumptions made in thin film descriptions are valid and the influence of drops deformations and viscosity ratio on the film drainage.

**Governing equations**

We consider the axisymmetric near contact of two, initially undeformed, spherical drops with radii \( R_1 \) and \( R_2 \), viscosity \( \mu \), and interfacial tension \( \sigma \), subjected to an external constant force \( F \) in a quiescent fluid with viscosity \( \mu_s \) (Fig. 1). The drops are pushed towards each others along their line of centres by equal and opposite contact forces \( F/2 \) and \(-F/2\). Both, the continuous and dispersed phases are Newtonian and incompressible but no restrictions on the viscosity ratio are retained. The velocity and pressure fields are obtained by solving the transient Stokes equations:

\[
\nabla \cdot \mathbf{u}_i = 0 \tag{1}
\]

\[
\mu_i \frac{\partial \mathbf{u}_i}{\partial t} = -\nabla p_i + \nabla \cdot \left( \mu_i \left( \nabla \mathbf{u}_i + \nabla^T \mathbf{u}_i \right) \right) + \mathbf{F}_v \tag{2}
\]

where \( \mathbf{u}_i \) is the vector velocity in each phase, \( \rho_i, \mu_i \) are density and viscosity of each phase, \( p_i \) is the pressure (\( i =1, 2 \)) and \( \mathbf{F}_v \) is the volume force (N/m\(^3\)).

The boundary conditions are given as follows:

- The normal component of the interfacial stress is given by:

\[
(p_1 - p_2) \left[ \mu_2 \left( \nabla \mathbf{u}_2 \right) \cdot \mathbf{n} - \mu_1 \left( \nabla \mathbf{u}_1 \right) \cdot \mathbf{n} \right] \cdot \mathbf{n} - \sigma \nabla \cdot \mathbf{n} = 0 \tag{3}
\]

where \( \mathbf{n} \) is the unit normal to interface, \( \sigma \) is the interfacial tension and \( \nabla \cdot \mathbf{n} \) is the local curvature of the interface, with \( \nabla \) the surface gradient operator.

- The continuity of tangential stresses is expressed as:

\[
\left[ \mu_2 \left( \nabla \mathbf{u}_2 \right) \cdot \mathbf{n} - \mu_1 \left( \nabla \mathbf{u}_1 \right) \cdot \mathbf{n} \right] \cdot \mathbf{t} = 0 \tag{4}
\]

where \( \mathbf{t} \) is the unit tangential to interface.

- The continuity of the velocity fields is given by:

\[
\nabla \cdot \mathbf{u}_i = 0 \tag{5}
\]

**Fig. 1.** Schematic representation of two coalescing drops.
\[ \mathbf{u}_i = \mathbf{u}_2 \]  
(5)

- The no-slip boundary condition is imposed far from the drops:
\[ \mathbf{u}_2 = 0 \]  
(6)

In our analysis, we adopt a dimensionless variables similar to those introduced by Nemer et al. (2004). The variables are nondimensionalized by choosing the equivalent drop radius \( R_{eq} \) as the length scale and \( \sigma / \mu \) as the velocity scale. Accordingly, the scales for time and pressure are, respectively, \( R_{eq} / \mu / \sigma \) and \( \rho R_{eq} / \sigma \):
\[ \begin{align*}
  \tau' &= \frac{\tau}{R_{eq}^2}, \\
  \zeta' &= \frac{\zeta}{R_{eq}}, \\
  \mu' &= \frac{\mu}{\sigma}, \\
  \rho' &= \frac{\rho R_{eq}}{\sigma}.
\end{align*} \]  
(7)

According to this dimensionless formulation, the flow inside and outside the drops is characterized by two dimensionless parameters, the dimensionless force \( F' \), and the viscosity ratio \( \lambda \):
\[ \begin{align*}
  F' &= \frac{F}{\sigma R_{eq}} \\
  \lambda &= \frac{\mu_1}{\mu_2}.
\end{align*} \]  
(8)

In order to facilitate further analysis and comparison with thin film theory, we introduce the deformation parameter \( \alpha' \) which is related to the dimensionless force by:
\[ \alpha' = \sqrt{\frac{F'}{2\pi}} \]  
(9)

Under small-deformation conditions i.e. quasi spherical drops, \( \alpha' \) will tend towards dimple radius, \( a' \), as predicted by small deformation theory.

**Numerical procedure**

Since only head-on collisions are studied, the governing equations have been solved in an axisymmetric system of coordinates in order to optimize the computational time. The governing equations have been solved with Comsol 4.2 using finite element method. In order to track interfaces Arbitrary-Lagrangian-Eulerian (ALE) method is used. Getting the shape and the interface deformation with the moving mesh ALE method is based on the mesh deformation in every time step which is governed by a mesh smoothing function. In Comsol 4.2, different mesh smoothing functions are implemented: Winslow, Laplace and hyperelastic. After testing these different methods, it was observed that the Winslow smoothing method gives better results compared to the other methods, especially in regions where interfaces are highly stretched. The time-dependent problem was handled by an implicit backward second order method. The Newton iterative scheme was used for solving nonlinear algebraic equations at each time integration step. During Newton iterations, systems of linear algebraic equations were solved by direct PARDISO solver. The relative and absolute tolerances were set to \( 10^{-6} \) and \( 10^{-7} \), respectively to control the error at each integration step. The computational domain comprising both the continuous and the dispersed phases, is meshed using triangular elements forming a grid structure with very fine cells close to the contact zone due to relatively steep gradients in this region and it was made progressively coarse far from the interface. Therefore in order to find the most appropriate length of the computational domain, many simulations with different lengths were carried out. The minimum dimensions of the computational domain, for which the solutions are almost unaffected, were determined to be about three drop radii in the \( r \) and \( z \) directions. In order to determine at what number of element the solution of the governing partial differential equations (PDEs) becomes grid independent, a grid independence study was conducted on the finite element mesh. The objective was to find the grid which would offer the best compromise between numerical solutions accuracy and computational cost. Five numerical grids with different number of computational cells were used: grid systems with approximately 45000 elements, 100000 elements, 200000 elements, 400000 elements and 700000 elements of which more than 50 % are located in the dimple region which depends on the \( \alpha' \) value. The influence of the different mesh grids on the prediction of the minimal film thickness for a typical case (\( \alpha' = 0.3 \) and \( \lambda = 1 \)) is presented in Table 1. An examination of this table shows that the numerical results seem to have stabilized with a relative error less than 1.5 \% between the two grids \( G_4 \) and \( G_5 \). Hence the grid \( G_4 \) was used for all computations with values of the deformation parameter up to 0.3 (\( \alpha' \leq 0.3 \)). However, grids with higher number of elements were used for larger deformation parameter. For instance, for \( \alpha' = 0.8 \), a computational grid with approximately 800000 elements were used. It should be noted that unlike the lubrication theory, the film thickness, \( h \), does not appear in the governing equations and therefore can not be calculated directly. In fact, the numerical procedure allows to calculate out the positions.
RESULTS AND DISCUSSION

To validate our model we compared the obtained numerical results with those from asymptotic theories and of simple film-drainage models. In order to compare our results with those of Nemer et al. [14], we introduce the dimensionless variables used by these authors, designated with bars and which are related to those used here as follows:

\[ \overline{h} = \frac{h'}{2\alpha'^2}, \quad \overline{t} = 2t'\alpha' \]  

(10)

The evolutions of the dimensionless minimum film thickness \( \overline{h}_m \) and dimensionless centre film thickness \( \overline{h}_0 \) are shown in Fig. 2. Initially, the profiles show a fast decrease. At large time, both thicknesses follow a trend that is similar to the long-term asymptotes found by Nemer et al. [14] and given by:

\[ \overline{h}_m = 0.4876 \overline{t}^{-4/5} \]  
[11]

\[ \overline{h}_0 = 1.007 \overline{t}^{-3/5} \]

Next, we compared our results with those of Bazhlekov et al. [7] and Alexandrova et al. [12], by introducing the dimensionless variables used by these authors, designated with asterisks and which are related to those used here as follows:

\[ h^* = \frac{h'}{\alpha'^2}, \quad t^* = t'\alpha' \]  

(12)

Fig. 3 shows the evolution of the minimum thickness, as function of time for \( \alpha' = 0.1 \) and \( \lambda = 0.1, 1 \) and 100. Very good agreement for predicting of film thinning rate is observed between the present results and those of Bazhlekov et al. [7] and Alexandrova et al. [12].

In order to follow the effect of deformation and viscosity ratio on the drainage of the film over time, simulations were conducted for different values of the parameter \( \alpha' \) (\( \alpha' = 0.1, 0.2, 0.3, 0.5 \) and 0.8) and for different values of viscosity ratio (\( \lambda = 0.1, 1, 10 \) and 100). The numerical results will be presented in form of film shapes at multiple times, as well as the evolution in time of the film thickness at the centerline, \( h_{cm} \), the minimal film thickness, \( h_m \), and the film radius \( r_m \).

Fig. 4 shows the process of film formation for different viscosity ratio \( \lambda \) and different values of the deformation parameter \( \alpha' \). In all studied cases, the drops have initially the same spherical shape (convex interfaces). As the interfaces moves closer to each other, they become flattened, and further approaching leads to a dimple (concave shape). The formation of the dimple is due to the slow motion of the central region of the interfaces compared to the approaching speed of the rim. The time evolution of the film radius is presented in Fig. 5 for a fixed...
Fig. 4. Time evolution of the film thickness: a) \( \lambda = 0.1 \) and \( \alpha' = 0.2 \), b) \( \lambda = 1 \) and \( \alpha' = 0.8 \), c) \( \lambda = 10 \) and \( \alpha' = 0.3 \) and d) \( \lambda = 100 \) and \( \alpha' = 0.5 \).

Fig. 5. Dimple radius as function of time and \( \alpha' \) for a fixed viscosity ratio \( \lambda = 1 \).

Fig. 6. Final drop film radius as function of \( \alpha' \).
viscosity ratio $\lambda = 1$ and different values of $\alpha'$. In all cases, the initial dimple radius, $r'_m$, is zero and increases with time reaching a steady value. The film radius grows faster when the contact force is larger.

Fig. 6 shows the calculated final film radius as function of the deformation parameter $\alpha'$ for two viscosity ratio ($\lambda = 0.1$ and 100). Our results are presented together with those of small interaction predictions. The numerical results show that the final dimple radiuses for $\alpha' = 0.10$, 0.20, 0.3, 0.5 and 0.8 are respectively $r'_m = 0.10$, 0.195, 0.29, 0.46 and 0.67. From these results, it is clear that for weak deformations, the results coincide exactly with the predictions of the Derjaguin and Kussakov theory [20], which states that the dimple radiuses must be equal to the value of $\alpha'$. For higher values of $\alpha'$, the dimple radius is slightly smaller, since small deformation theory loses its validity for these cases. Fig. 6 shows also that final drop film radius is not influenced by the viscosity ratio $\lambda$.

The obtained drop shapes are presented in Fig. 7. Results illustrate that the drop deformation increases when $\alpha'$ increase. The figure shows also that the assumption of spherical drop shape outside the film region is valid only for weak values of $\alpha'$.

Fig. 8 presents the minimal film thickness as a function of the time for different values of $\alpha'$, and different viscosity ratio $\lambda$. Even though the drainage rate depends on both $\lambda$ and $\alpha'$, in all cases, the evolutions of $h_{min}$ ini-
tially shows a fast decrease which is more pronounced for drops with large contact force. On the contrary, for long time drainage after the film is formed, it is observed that the drainage is facilitated for drops with small deformations. This is quite expected because, at the beginning of the drainage, that is when the two drops are far enough apart, they behave like two indeformable drops and the contact force serves essentially to bring the two drops closer to each other, hence a higher drainage rate when the force is greater. In contrast, when the drops are quite close to each other, the increase in the interaction force increases the film radius and decreases the rate of thinning, which is why coalescence is favored by gentle collisions.

CONCLUSIONS

A numerical study has been carried out to investigate the effects of interface deformation and viscosity ratio on the film drainage between two drops. Arbitrary Lagrangian–Eulerian (ALE) formulation including Stokes equations in both, dispersed and continuous phases, is used to allow interface tracking. For interacting drops with small deformations, the numerical results are consistent with theoretical predictions based on simple film drainage models and with those from asymptotic analysis.

ALE simulations were also used to explore near-contact drop dynamics under finite-deformation conditions. The numerical results show that film drainage between the two drops depends strongly on the interface deformation as well as on the dispersed to continuous-phase viscosity ratio. The numerical results also show that, for a fixed viscosity ratio, by increasing interaction force drainage rate increases in the earlier stage of drainage while it decreases after the film is formed. The results from this study demonstrate the ability of the ALE approach to extend the results of previous simple drainage models to a wider range of parameters. The use of this numerical method should enable further improvements of the models to include van der Waals forces, inter-phase mass transfer or surfactants diffusion.

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