Head-on collision of two equal-sized drops with van der Waals forces

X. Jiang & A.J. James

Department of Aerospace Engineering and Mechanics, University of Minnesota, USA

Abstract

Drop collision is significant not only in natural processes, such as raindrop formation, but also in various engineering applications, such as chemical separation, spray coating, encapsulation, and drug delivery. In this study, head on collision of two equal-sized drops in a hyperbolic flow is investigated numerically. It is well known that van der Waals forces become important and govern coalescence when the thickness of the fluid film between the two drops falls into the range of several hundreds of Angstroms, 100-1000 Å. We have developed two new numerical methods to introduce non-retarded van der Waals forces into the Navier-Stokes equations. An axisymmetric volume of fluid (VOF) method is used to simulate the motion of a drop toward a symmetry plane where it interacts with, and possibly coalesces with, its mirror image. A hyperbolic flow imposed initially and at the domain boundaries drives the motion. In the absence of van der Waals forces, the VOF boundary condition on the symmetry plane determines whether the two drops coalesce or bounce. One method used to include van der Waals forces is to apply the force on the drop as the negative gradient of van der Waals potential between the drops. The other method computes the force in terms of a disjoining pressure in the film which depends on the film thickness. A cut-off is used as the thickness of the film approaches zero to avoid singularity. Comparisons of the results of the two methods at various values of the Hamaker constant are presented. A comparison to results without van der Waals forces is also reported. In future work mesh adaptivity will be used to better resolve the film between the two drops.
1 Introduction

Droplet collision has been of interest to researchers for decades. Recently, Orme [1] reviewed experimental studies on the collision behavior of water and fuel droplets. Different kinds of binary collision outcomes were discussed. Nobari and Jan [2] studied numerically the head-on collision of two equal-sized drops using a front tracking method. They caused the drops to either rebound or coalesce depending on whether the thin film between the two drops was removed artificially or not.

When two drops immersed in an immiscible fluid approach each other along the axis of their centers, a film forms between them and subsequently thins. When the thickness of the film falls into the range of several hundreds of Angstroms, 100-1000 Å, van der Waals forces become significant and often result in the rupture of the film. The literature in van-der-Waals-driven thin film rupture is rich, but most of studies are focused on the rupture of a free film [3], or a thin film on a horizontal solid plane [4]. Studies on thin film rupture in coalescence of drops are limited. Chen [5] set up a model to simulate the coalescence of two equal-sized drops in head-on collision with emphasis on drainage and rupture of the film between the two drops. Van der Waals forces were represented by the negative gradient of a scalar potential. Yiantsiros and Davis [6] examined the approach rate and deformation of two different-sized drops in head-on collision at small capillary number. They accounted for van der Waals forces as a disjoining pressure. Lubrication theory was applied in the thin film between the drops.

In this study, we present the numerical simulation of the collision of two equal-sized drops driven by a hyperbolic flow. The effect of van der Waals forces on the coalescence of the drops is examined. The van der Waals forces are accounted for by two different methods. In one method, a body force calculated from an interaction potential is introduced into the momentum equation [5, 7]. In the other method a disjoining pressure is used to represent van der Waals forces as in [4, 6]. The primary difference between our research and previous work is that the full Navier-Stokes equations, with modifications to represent van der Waals forces, are solved. In addition, we examine the whole coalescence process from the approach of the two drops to the formation of the single drop. We start with two drops separated by some distance and the film between the drops develops naturally.

2 Formulation

Two equal-sized spherical liquid drops are immersed in an axisymmetric extensional gas flow, as shown in figure 1. Driven by the suspending fluid, the drops will collide head-on and then may bounce apart or come into contact.

We use an axisymmetric volume of fluid (VOF) method [8] to simulate the collision process of the two drops. In this method, a volume fraction, $F$, is defined in each cell as the portion of the volume of the cell containing liquid. The interface is tracked through the evolution of the volume fraction. We use one set of equations that govern the motion in the whole domain. To do this, the surface tension is included in the momentum equation using the continuum surface force method [9].
Figure 1: The physical problem and the computational domain. The computational domain is comprised of 50x100 grid cells.

For incompressible, transient, interfacial flow, the dimensionless governing equations are the continuity equation, \( \nabla \cdot \vec{u} = 0 \), the Navier-Stokes equation, and the volume fraction convection equation:

\[
\rho \left( \frac{\partial \vec{u}}{\partial t} + \vec{u} \cdot \nabla \vec{u} \right) = -\nabla p + \frac{1}{Re} \nabla \cdot (2\mu D) + \frac{\kappa}{We} \nabla F + \vec{f}_v, \tag{1}
\]

\[
\frac{\partial F}{\partial t} + \vec{u} \cdot \nabla F = 0, \tag{2}
\]

where \( \vec{u} \) is the velocity, \( \rho \) is the density, \( \mu \) is the viscosity, \( p \) is the pressure, \( D \) is the rate of deformation tensor, \( \kappa \) is the interface curvature, and \( \vec{f}_v \) is the van der Waals force. Here we neglect gravity.

The length scale used in non-dimensionalization is the radius of the drop \( R \). The velocity scale is the strain rate of the hyperbolic flow, \( G \), times \( R \). Inertial time and pressure scales are used. The density and viscosity are scaled on the density of the drop \( \rho_d \) and the viscosity of the drop \( \mu_d \), respectively.

The non-dimensional parameters are the Reynolds number, \( Re = \rho_d G R^2 / \mu_d \), the Weber number, \( We = \rho_d G^2 R^3 / \sigma \), where \( \sigma \) is the surface tension coefficient, the density ratio, \( \alpha = \rho_o / \rho_d \), and the viscosity ratio, \( \beta = \mu_o / \mu_d \), where \( \rho_o \) and \( \mu_o \) are the density and viscosity of the surrounding fluid, respectively.

The flow field is solved by a projection method on a staggered grid [10]. The resulting pressure-Poisson equation is solved using an incomplete Cholesky conjugate gradient method [11]. The interface is reconstructed using a piece-wise linear approximation. Coalescence is considered to occur when any of the line segments intersect the symmetry plane. The code has been thoroughly tested and used to study the breakup of a vibrating drop by James et al [12].

To take advantage of symmetry of the geometry, we use only the upper right quadrant in figure 1 as the computational domain. Initially the two drops sit on the symmetry axis with their centers separated by four radii. The velocity of the flow field is initialized as \( \vec{u} = r\hat{r} - 2z\hat{z} \), where \( \hat{r} \) and \( \hat{z} \) are the unit vectors in the \( r \) and \( z \) directions, respectively. Based on the physics of the collision, two kinds of the volume fraction boundary conditions on the symmetry plane can be used: \( \partial F / \partial z = 0 \) and \( F = 0 \), which will lead to different collision outcomes. Details will be given in section 4.1.
3 Calculation of van der Waals forces

In the first method used to account for van der Waals forces, the interaction potential between the two drops is calculated. A body force which acts on the drops, and is calculated as the negative gradient of this potential energy, is introduced into the momentum equation. This method is based on the fact that two bodies of the same material in the medium of another material always attract each other.

Numerically the van der Waals force $f^v_i$ is calculated as described below. The potential energy between the two drops is calculated via a microscopic Hamaker procedure [7, 13]. We neglect the interaction with molecules of gas. The potential energy between two unit volumes is $-A^*r^6$, where the dimensionless Hamaker constant $A^*$ is related to the Hamaker constant $A^* = A^*/\rho_d R^5 G^2$, and $L$ is the distance between the two volumes. The potential energy per unit volume at cell $(m,n)$ in drop $i$ due to cell $(s,t)$ in the mirror drop $j$, $d\phi_{mn}$, is obtained by multiplying the volume fraction in cell $(s,t)$, $F_{st}$, and integrating over the cell volume, $d\phi_{mn} = \int_{cell(s,t)} -A^*F_{st}drdzd\theta$. For small grid spacings in the $r$ and $z$ directions, $\Delta r$ and $\Delta z$, $d\phi_{mn} \approx -Ar^z \Delta r \Delta z F_{st} \int_0^{2\pi} \frac{1}{L^2} d\theta$. The potential energy per unit volume at $(m,n)$ due to the entire drop $j$ is computed by summing over all cells in $j$. With $\int_0^{2\pi} \frac{1}{L^2} d\theta$ being evaluated analytically,

$$\phi_{mn} = -\frac{2A}{\pi} \Delta r \Delta z \sum_{s,t} \left\{ r^z F_{st} \frac{(H^2 + r^2 + r^2_m)^2 + 2r^2 r^2_m}{[(H^2 + r^2 + r^2_m)^2 - (2r^2 r^2_m)^2]^{3/2}} \right\}, \quad (3)$$

where $H$ is the perpendicular distance between the cell $(m,n)$ and $(s,t)$ in drop $j$, and $r_m$ and $r_s$ are the radial coordinates of cells $(m,n)$ and $(s,t)$, respectively.

The force per unit volume at $(m,n)$ is then $f^v = -F_{mn} \nabla \phi_{mn}$. It is weighted by the volume fraction in that cell, $F_{mn}$, to ensure that the force occurs only inside the drop.

In the other method, we incorporate the effect of van der Waals forces as a disjoining pressure in the thin film. The disjoining pressure can be introduced into the equation of motion [14]. For a plane-parallel film with a thickness of $h$, the disjoining pressure is given by $\Pi(h) = -\frac{A}{6\pi h^3}$ [6, 7, 13]. The gap between the two drops is not uniform, especially when the distance between the drops is large, but we assume that van der Waals forces are nearly negligible when the drops are far away. When the drops are close enough for van der Waals forces to be important, we assume that the thickness of the film changes gradually so that the slope of the interface is small. It is then appropriate to use the above equation to approximate the disjoining pressure. The van der Waals force reads $f^v = -\nabla \left( \frac{A}{6\pi h^3} \right)$. In the computation, the film thickness $h$ is defined only inside the film between the drops.

4 Results and discussion

In this section, we first present the results in the absence of van der Waals forces. The significance of the volume fraction boundary condition on the symmetry plane
is discussed. Then the results with van der Waals forces are reported. $\alpha$ and $\beta$ are both specified as 0.001. $Re = 1$, $We = 1$ are used in the calculations so that the effects of inertia, viscosity and surface tension are of equal importance.

### 4.1 Symmetry condition

When a symmetry condition is used, as in figure 2, the drop and its image merge automatically. For clarity the mirror images of the computational domain are also shown. The straight-line approximation in each interfacial grid cell is plotted. It is well known [5, 6] that as two drops approach each other the pressure between them builds up, leading to the formation of a dimple, as shown at $t=0.32$. At $t=0.38$, coalescence occurs at the rim of the dimple, resulting in a large surface tension force at the contact point. The contact area is then increased by surface tension, leaving a bubble trapped in the center. After the post-collision drop has reached its maximum deformation at $t=0.80$, it and the entrapped bubble relax to a rounder shape. Experimentally, it is seen that azimuthal instabilities lead to coalescence at a point, not along the entire ring that surrounds the dimple. However, azimuthal variations are not allowed in this simulation. This limitation also allows the formation of nonphysical toroidal bubbles in the simulation.

Details of the evolution of the film between the drops are shown in figure 3(a). This figure shows the half thickness of the film between the drops, $h/2$, which is calculated from the straight-line approximation. The interface translates downward with little deformation until $t=0.29$ when the interface flattens near the symmetry axis. After that a dimple forms and grows rapidly, the minimum thickness of the film decreases, and the film ruptures at the rim of the dimple at $t=0.38$. Figure 3(b) shows the pressure at the symmetry plane as a function of the radial position. From $t=0.27$ to $t=0.29$, there is a dramatic increase in pressure at the symmetry axis, giving rise to a large radial pressure gradient. Thus, it becomes harder and harder to squeeze out the fluid between the drops as the film becomes thinner and thinner. The thinning rate at the symmetry axis slows down under such a sudden increase of pressure, as can be seen in figure 3(a). The film at the symmetry axis thins quickly until a transition point at about $t=0.29$ when a dimple begins to form and then thins slowly. Comparison of figures 3(a) and 3(b) shows that as the dimple grows, the pressure peak shifts from the symmetry axis to the rim of the dimple with the peak value decreasing.

Figure 4 shows the rebound of two drops. The volume fraction on the symmetry plane is fixed at zero. Therefore the drop sees the boundary as a slip wall, but it cannot merge with its image. Impact of the drops creates a large pressure between them, so the drops rebound even though the applied velocity continues to force them toward each other. The pressure profiles appear quite similar to figure 3(b). Figure 5 gives detailed shapes of the film between the drops. Because of the boundary condition used, the dimpled film will not rupture, but flattens as the drops deform. As the drops rebound, the film resumes its dimpled shape until the drops are almost round.
358 Computational Methods in Multiphase Flow

Figure 2: Coalescence of two drops. Van der Waals forces are absent. $\partial F/\partial z = 0$ on the symmetry plane. $Re = 1$, $We = 1$, $\alpha = 0.001$, $\beta = 0.001$.

Figure 3: (a) Evolution of the film between two drops. (b) Symmetry plane pressure as a function of radial position. Van der Waals forces are absent. $\partial F/\partial z = 0$ on the symmetry plane. $Re = 1$, $We = 1$, $\alpha = 0.001$, $\beta = 0.001$.

Figure 4: Rebound of two drops. Van der Waals forces are absent. $F = 0$ on the symmetry plane. $Re = 1$, $We = 1$, $\alpha = 0.001$, $\beta = 0.001$.

4.2 Effect of van der Waals forces

In this section we focus on the effect of van der Waals forces on the rupture of the film. For this reason, $\partial F/\partial z = 0$ is used on the symmetry plane. The dimensionless Hamaker number, $A$, is varied to adjust the strength of van der Waals forces.
Figure 5: Evolution of the film between two drops. Van der Waals forces are absent. $F = 0$ on the symmetry plane. $Re = 1$, $We = 1$, $\alpha = 0.001$, $\beta = 0.001$.

4.2.1 Van der Waals force calculated from potential

Figure 6 shows the evolution of the film between the drops for three values of $A$. Two modes of rupture are observed for different values of $A$: 'rim rupture' [6] where the film ruptures at the rim of the dimple, as in figure 6(a) and (b); and 'nose rupture' [6] where the film ruptures at the symmetry axis, as in 6(c). At relatively small $A$, for example $A = 1$, a dimple is formed and grows. But compared with the dimple in figure 3(a), where $A = 0$, the radius of the dimple is smaller. The size of the dimple continues to decrease as $A$ increases. At $A = 10$ only a very small dimple is formed by the time of rupture. As $A$ is increased to 100, no dimple is formed at all. This suggests that under strong van der Waals forces, the drops approach each other at such a fast rate that there is no time for the dimple to form. Notice that at $A = 1$, there is an increase in film thickness at the axis of symmetry near the time of rupture. As the film evolves toward rupture, a large positive radial pressure gradient in the film is formed near the rim of the dimple, preventing the gas in the gap from flowing out and subsequently making the interface at the symmetry axis yield.

Figure 6. Evolution of the film between the drops with van der Waals forces calculated from potential. $Re = 1$, $We = 1$, $\alpha = 0.001$, $\beta = 0.001$. (a) $A = 1$, (b) $A = 10$, (c) $A = 100$. 
Figure 7. Half minimum thickness of the film as a function of time under different values of $A$. $Re = 1$, $We = 1$, $\alpha = 0.001$, $\beta = 0.001$.

Figure 7 shows the half minimum thickness of the film, $h_{min}/2$, as a function of time. It is obvious that the rupture time decreases as the strength of van der Waals forces grows. For small time, all the curves collapse with the curve $A = 0$, which implies that in this period of time van der Waals forces are weak. As $A$ increases, the corresponding curve deviates from the $A = 0$ curve sooner. It is also interesting to note the thinning rate of the film under different values of $A$. As mentioned in section 4.1, there is a transition point associated with the formation of the dimple, between the faster thinning in early time and the slower rate in later time, as shown in the curve $A = 0$. Under stronger van der Waals forces the film thins faster. At $A = 1$, after the transition point, the film thins at a faster rate than the case of $A = 0$. At $A = 10$, the small dimple formed seems to have little effect on the thinning rate. The film thins more rapidly and there is no dramatic change of thinning rate observed. When van der Waals forces are very strong, as $A = 100$, there is no dimple formed to slow down the thinning of the film, so the curve is smooth. Close to the rupture time, the slope of the curve is large, which reveals the rapid drainage near the rupture time due to van der Waals forces.

4.2.2 Van der Waals force calculated from disjoining pressure

Figure 8 shows the evolution of the film between the drops for different values of $A$. In the computation, a cutoff of $h = 10^{-4}$ is introduced to suppress the singularity as $h \to 0$. As in figure 6, rim rupture is observed for $A = 1$ and $A = 10$, and nose rupture for $A = 100$. Figure 9 compares the curves of half minimum film thickness as a function of time obtained by the two methods and that without van der Waals forces. The film between the two drops thins faster when the van der Waals force is calculated from the disjoining pressure. The form of the disjoining pressure is derived from the interaction potential between two semi-infinite liquid regions separated by a film of uniform thickness. In applying this to the interaction of two drops, the assumption that the regions are semi-infinite is valid as long as the drop thickness is relatively large, since the potential decreases as $L^{-6}$. The disjoining pressure includes the effect of a flat liquid region, but away from the current grid cell the film thickness is larger if the interface is convex, as it is near the contact point. Thus, under the assumption of the flat interface, the disjoining pressure over-predicts the attractive force, and hence the thinning rate.
Figure 8: Evolution of the film between the drops with van der Waals forces calculated from disjoining pressure. $Re = 1, We = 1, \alpha = 0.001, \beta = 0.001$. (a) $A = 1$, (b) $A = 10$, (c) $A = 100$.

Figure 9: Half minimum thickness of the film as a function of time. $Re = 1, We = 1, \alpha = 0.001, \beta = 0.001$. (a) $A = 1$, (b) $A = 10$, (c) $A = 100$. (1) Van der Waals force calculated from potential; (2) form disjoining pressure.

5 Conclusions

The simulations presented show that manipulation of the volume fraction boundary condition can be used to numerically control coalescence. In future work we will exploit this by coupling it to a small-scale model that will physically determine which boundary condition applies at any time.

The two methods presented to include van der Waals forces in simulations of drop collision result in qualitatively similar behavior, but there are small quantitative differences in the results. The disjoining pressure method is much less computationally intensive, but relies on the assumption that the interface slope is small. For the simulations presented here the difference in the coalescence time ranges from 5% when $A=1$ and the interface is relatively flat to 9% when $A=100$ and coalescence occurs before the interface has time to flatten. There is agreement between the two methods on the value of $A$ for transition from rim to nose rupture. Thus, the disjoining pressure method captures the main features of the evolution, but may not provide adequate accuracy under all conditions.

Van der Waals forces take effect over a length scale that is much smaller than the drop radius and result in rapid changes. Therefore, a fine grid is a necessity in
the near contact area. Whereas away from this region, a coarser grid is acceptable. In future work an adaptive mesh will be implemented in the current model so that the thin film between the two drops can be better resolved.

This work was supported by the Office of the Vice President for Research and Dean of the Graduate School at the University of Minnesota, and utilized the resources of the University of Minnesota Supercomputing Institute.

References