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Combined gravitational and thermocapillary interactions of spherical drops with incompressible surfactant

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A R T I C L E I N F O

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Collision efficiencies are calculated by a trajectory analysis for two contaminated spherical drops under the combined influence of buoyancy and a constant temperature gradient at low Reynolds number and with negligible thermal convection in the limit of nearly uniform surfactant coverage. As in the case of clean drops, a region in the parameter space exists where collisions are forbidden when the driving forces are opposed. However, because of the increased effect of thermocapillary repulsion when surfactant is present, coalescence can be inhibited even when the driving forces are aligned in the same direction. In addition to trajectories leading to coalescence and separation of the drops, closed trajectories are also observed. At parameter values where the asymmetric mobility function is zero, retrograde motion can occur, where the angle between vertical and the drops' line of centers decreases as the drops come into contact. This retrograde motion requires alteration to the closed form expression for the collision efficiency. The effect of incompressible surfactant on dilute dispersions of two physical systems is also considered. Populations dynamics simulations for both driving forces aligned in the same direction with van der Waals attraction indicate that the volume-averaged radius begins to level out and then continues to grow because the collision efficiency passes through a minimum but is not identically zero.

1. Introduction

In the absence of significant gravity, a temperature gradient can be used to drive the motion of drops and bubbles [1]. However, in order to study motion due to an applied temperature gradient on earth, the presence of gravity must be accounted for, or Marangoni-induced flow can be completely masked. In addition to this practical limitation, droplet interactions in combined gravitational and thermocapillary motion have been studied because of their importance in materials processing. In particular, when the alignment of gravity and the temperature gradient is antiparallel, coalescence rates can be greatly reduced [2,3], a result of potential benefit in the production of liquid-phase-miscibility-gap materials and immiscible metal alloys [4]. Other processes where both driving forces can be important include glassmelting [5,6], crystal growth [7], where bubbles may be a significant contaminant [8], preparation for space experiments under normal gravity [9,10] and many space operations in extraterrestrial environments [11].

Because of the unavoidable presence of gravity, earth-bound experiments [12–15] on thermocapillary migration, from the first results up to the present time, have included buoyant effects. With respect to a macrophysical situation, early theory, involving population dynamics, predicted the behavior of a dilute dispersion of small, spherical drops or bubbles when both gravity and thermocapillary forces were present but with hydrodynamic interactions neglected, i.e. the collision efficiency was set equal to unity [2]. Subsequently, the microphysical problem of determining collision efficiencies for two spherical drops in either Marangoni or buoyancy motion was solved, and the relative importance of the competing

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effects was compared by considering each separately [16]. The combined action of gravity and a vertical temperature gradient was later studied for two spherical bubbles [17] and drops [3]. A significant result of the drop study, which included population dynamics to follow the drop-size distribution, was that coalescence can be greatly retarded under certain conditions when the temperature gradient and gravity vector are oppositely aligned. In this collision-forbidden region of parameter space, droplet distributions become monodisperse because further growth is prohibited beyond a critical drop size.

In this work, we consider the combined influence of buoyancy and thermocapillarity on interactions of two spherical drops with negligible inertia and thermal convection in the presence of bulk insoluble, non-ionic surfactant. As in the case of gravity, surfactants represent an almost unavoidable complication [18]. For instance, anomalies in past thermocapillary experiments have been explained in terms of the accidental presence of surface-active agents [14,19]. The limit investigated here is that of an incompressible surfactant film [20,21]. The concept of incompressible surfactant is similar to that of incompressible flow. In the latter case, a large pressure change leads to a small change in density. In the former case, an infinitesimal change in surfactant concentration corresponds to a finite variation in interfacial tension. Thus, the limit of incompressible surfactant is the same as that of nearly uniform surfactant surface coverage. As shown by scaling arguments [22], incompressible surfactant has physical importance, especially for small drops.

Nearly uniform surfactant coverage has been studied in binary interactions under a variety of flows, including Brownian motion and linear flows [22,23], flotation of a bubble covered with incompressible surfactant and a much smaller particle [24], gravitational interactions [25], and thermocapillary motion [26]. An important result of the investigation into thermocapillary interactions of two contaminated drops in the limit of nearly uniform surfactant coverage concerned the importance of thermocapillary repulsion. For clean drops [27], thermocapillary repulsion, where the smaller drop moves faster than the larger one due to disturbances in the temperature field, occurs only at large drop-to-medium thermal conductivity ratios and very small separations. Moreover, in the absence of surfactant, thermal repulsion is relatively weak. However, when surfactant is present [26], thermal repulsion is observed for all cases except that of non-conducting drops. Moreover, the effect is much more pronounced even at large separations because the velocities of the contaminated drops are very close in magnitude.

Herein, we investigate the limit of nearly uniform surfactant coverage in the case of two spherical drops in combined gravitational and thermocapillary motion. The goal of the work is to extend earlier results for clean drops [3] and determine the effect of surfactants on the collision-forbidden region of the parameter space. Section 2 contains assumptions in the model, problem formulation and method of solution. Section 3 provides results and discussion and Section 4 is concluding remarks.

2. Problem statement and method of solution

Fig. 1 portrays two spherical drops moving under the combined action of gravity and a temperature gradient that is constant in magnitude. The driving forces g and ∇T may be aligned in the same or opposite directions. Flow conditions are such that inertia, thermal convection, and Brownian motion are negligible. In other words, the Reynolds number Re and Marangoni number Ma are small, while the Péclet number Pe is large. Moreover, in order for deformation to be insignificant, the capillary number Ca must be small. Three dimensionless parameters arise naturally from the physical properties of the drop liquid and surrounding unbounded matrix: $k = a_1/a_2$ is the ratio of the smaller drop radius to the larger drop radius and must be less than unity for relative motion to occur; $\hat{\mu}=\mu_d/\mu_e$ is the ratio of the drop viscosity to that of the surrounding liquid; and $\hat{k}=k_d/k_e$ is the ratio of the drop thermal conductivity to that of the matrix.

Standard definitions such as $Re=\rho_e V_2^{(0)} a_2/\mu_e$ and $Ma = a_2 V_{T,2}^{(0)}/D_T$ for the larger drop can be applied, where $V_2^{(0)}=V_{G,2}^{(0)} \pm V_{T,2}^{(0)}$, with $V_{G,2}^{(0)}$ and $V_{T,2}^{(0)}$ being the velocities due to gravity and the temperature gradient alone and are defined in Eqs. (3) and (4). The sign used in combining the gravitational and thermocapillary velocity components depends on whether the driving forces are parallel or antiparallel. The matrix liquid density and thermal diffusivity are ρ_e and D_T, respectively.

The drops are covered with a bulk-insoluble, non-ionic surfactant under conditions such that the film is incompressible. That is, the deviation in the surfactant surface concentration is small. When surfactant is incompressible,

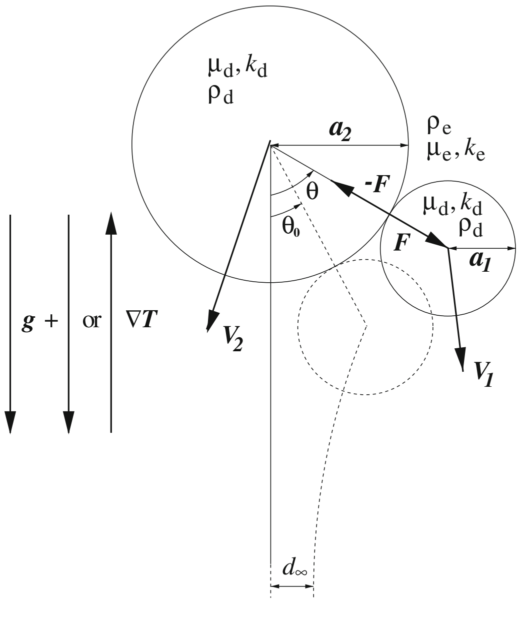

Fig. 1. Definition sketch for two drops covered with incompressible surfactant interacting due to the combined influence of gravity g and an applied temperature gradient ∇T.