Thermocapillary migration of liquid droplets in a temperature gradient in a density matched system

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Abstract. An experimental investigation of thermocapillary flow in droplets of a vegetable oil (partially hydrogenated soybean oil) immersed in silicone oil was conducted in a test cell with a heated top wall and a cooled bottom wall. The liquids are nearly immiscible and have equal densities at a temperature below the room temperature, thus providing a simulation of low-gravity conditions by reducing the buoyancy forces. The interfacial tension between the two oils was measured in the temperature range 20–50 °C using a capillary tube and \( \frac{d \sigma}{dT} \) was determined to be negative. Droplets ranging in sizes from 3 mm to 1 cm diameter were injected into the silicone oil. The vertical temperature profile in the bulk liquid (silicone oil) produces temperature variations along the interface which induce variations in the interfacial tension. The flow inside the droplet driven by the resulting interfacial shear stresses was observed using a laser light-sheet flow visualization technique. The flow direction is consistent with the sign of \( \frac{d \sigma}{dT} \). The observed maximum surface velocities are compared to the theoretical predictions of Young et al. (1959).

For short times after injection, the droplets were driven by this flow towards the hot wall above the matched-density temperature until the droplets reached a point where the forces due to the flow and buoyancy were equal. After longer times, the droplets moved to the cold side due to suspected density changes caused by mass transfer from the droplets to the silicone oil. This was confirmed by tests under isothermal conditions, where it was observed that droplets of all sizes fell to the cold bottom eventually.

Thus, even though the thermocapillary flow inside the droplets persisted for long times in spite of the mass transfer, the migration of droplets towards the hot side was eventually affected by uncontrolled buoyancy forces resulting from density changes due to mass transfer. While additional liquids are being tried, it is suggested from the present experience that reduced gravity experiments will probably be necessary to provide unambiguous data for the migration of droplets.

1 Introduction

The investigation of the migration of droplets in liquids due to thermocapillary flow is significant not only in enhancing the understanding of such flows but also has direct application to material sciences and chemical engineering. It has been suggested that the low gravity environment available in space orbit can be exploited to provide advancements in materials processing of composites formed from phases with a miscibility gap (Walter, 1986; Predel et al., 1987). Space manufacturing endeavors in general involve complex, non-equilibrium systems oftentimes with multiple interfacial regions providing the potential for significant surface tension driven flows which could affect product quality. Thus, it is imperative to develop an understanding of such effects through a coordinated earth-space research effort. The ground-based experimental study reported here was conducted to support such an effort in one special area – thermocapillary migration of immiscible droplets/bubbles in a continuous phase liquid.

The migration of bubbles, or droplets, is an important aspect of many engineering sciences and space manufacturing issues. It is a prime candidate for many studies as it is without complications due to contact lines. In material science processes as in glass manufacturing, etc., gas bubbles can be formed from the byproducts of chemical reactions or gas trapped in the interstices of the raw material. In a 1-g environment the migration of the bubbles due to buoyancy effects can be utilized as a separation mechanism. However, in a microgravity environment of space other forces must be utilized for the separation. Young et al. (1959) pointed out that a fluid droplet placed in a thermal gradient environment will experience a force due to the induced capillary flow. The interfacial tension will typically be minimum at the warm pole of the droplet, and will increase toward the cooler pole. The resulting tangential stress will drag the neighboring continuous phase fluid toward the cooler pole, and the reaction on the droplet will therefore be in the opposite direction. If the droplet is free to move, it will migrate toward warmer regions as a consequence, and hence provide a separation mechanism sensitive to the magnitude and direction of the bulk temperature gradient. After the pioneering work of Young et al. (1959), the Marangoni migration of bubbles in liquids was subject to many experimental investigations (Meritt and Subramanian, 1988; Hardy, 1979; Thompson, 1979; Nähle et al., 1987). The investigations of the thermocapillary migration of a droplet in a liquid are more recent and less have been performed. Lacy et al. (1982) observed the
migration of ethylsalicylate drops in diethylene glycol. Chifu et al. (1983) observed the flow on the surface of a droplet (freely suspended in a liquid) by injecting a small amount of a dyed surfactant into the interface. The velocity of the spreading surfactant front was measured and compared with the values resulting from their theoretical model. By also using a two component system with a miscibility gap (methanol/cyclohexane) and cooling it down below the temperature where cyclohexane separates, they observed a thermocapillary migration of the separated droplets in the direction of the warmer region. The thermocapillary migration of water droplets in butyl benzoate was investigated by Deltitzsch et al. (1984) and Hahnel et al. (1989). The two components of the liquid system that they used were immiscible and have the same density at about 32 °C. Water droplets injected into a stably stratified layer of butyl benzoate sorted out according to their diameters in a manner that smaller droplets migrated to higher temperatures than the larger ones. The final droplet positions was determined by the balance of the thermocapillary (proportional to the surface area) and buoyancy forces (proportional to the volume). Siekmann et al. (1986) also used water droplets in silicone oil in a microgravity environment (DI shuttle mission), but did not observe thermocapillary migration. Barton and Subramanian (1989), also observed thermocapillary migration in the liquid systems used by Lacy et al.

In order to continue to increase understanding of these phenomena it was decided that an important step would be to elucidate the flow inside and on the interface of a droplet, and its migration as well as its equilibrium position in a vertical temperature gradient. The visualization of the flow pattern would be an added confirmation that the mechanism for the migration is a result of the surface induced thermocapillary flow (in the opposite direction to the migration) and not due to any bulk induced motion (like buoyancy induced flow, for example, which would be in the same direction). Since conflicting results have been obtained using water systems, it was decided to use an easily available matched-density (at 15 °C) liquid-liquid system consisting of vegetable oil as the droplet liquid and 5 CS. silicone oil as the continuous phase liquid.

2 Theoretical considerations

To our knowledge, there is no theoretical treatment of thermocapillary migration of droplets for finite Marangoni numbers (resulting from thermocapillary motion) or Peclet numbers (from combined buoyancy and thermocapillary motion). The values of these parameters in the current experiments are displayed in Table 2 for silicone oil. The Peclet number is based on the measured surface velocity. Both parameters are greater than one. Thus, there is no theory to which the experimental results can be precisely compared. One choice for the purpose of comparison of experimental results is to refer to the classical linear theory of Young et al. (1959) as a guideline. They obtained the migration velocity $V$ of the droplets as

$$V = \frac{D^2 g(\rho - \rho_0)(\mu + \mu')}{6 \mu (2 \mu + 3 \mu')} \frac{dT}{dz} \frac{Dk}{(2 \mu + 3 \rho')(2k + k')}$$  \hspace{1cm} (1)$$

where $D$ is the droplet diameter, $g$ the gravitational acceleration, $\frac{dT}{dz}$ the temperature gradient at infinity, $\frac{d\sigma}{dT}$ the thermal interfacial tension coefficient, $\mu$ and $\mu'$ the dynamic viscosities and $k$ and $k'$ the thermal conductivities of the continuous phase fluid and the droplet liquid respectively. In the above equation and in what follows, a prime (') designates the property in the droplet phase. The migration velocity is the sum of two terms, the first representing the velocity of a droplet due to buoyancy forces and the second term describing the velocity induced by Marangoni forces. When the droplet reaches a stationary position, Marangoni and gravity forces balance each other and $V = 0$.

Substituting the linear approximations $\sigma = \sigma_0 [1 - \beta(T_e - T_m)]$, $\sigma' = \sigma_0 [1 - \beta'(T_e - T_m)]$, (subscript m denotes matched-density (conditions) into the above equation, one can write:

$$T_e - T_m = K \frac{dT}{dz}$$  \hspace{1cm} (2)$$

where

$$K = \frac{6 \mu \left( \frac{d\sigma}{dT} \frac{d\sigma}{dT} \right)}{\sigma_0 \theta (\mu + \rho') \left( 2 \frac{1}{k} + \frac{k'}{k} \right)} = \frac{1}{D} \left( \frac{d\sigma}{dT} \right) \left( \frac{d\sigma}{dT} \right)$$

is a material constant. Therefore the temperature differences from the matched density temperature ($T_e - T_m$) at which the droplet is stationary ($T_e$) is a function of two controllable variables, namely $\frac{1}{D}$, and $\frac{d\sigma}{dT}$. For constant $\frac{d\sigma}{dT}$, small droplets would move farther than large ones. The deflection increases with increasing $\frac{d\sigma}{dT}$. In the present study, experiments were performed to measure ($T_e - T_m$) with $D$ and $\frac{d\sigma}{dT}$ as controlled variables.

The maximum velocity on the interface according to the theory of Young et al. occurs at the equator of the droplet, and is expressed as follows:

$$V_{max} = \frac{D}{(\mu + \mu') \left( 2 \frac{1}{k} + \frac{k'}{k} \right)}$$  \hspace{1cm} (3)$$

This has been used for a comparison to our measured interfacial velocities on the droplet equator.