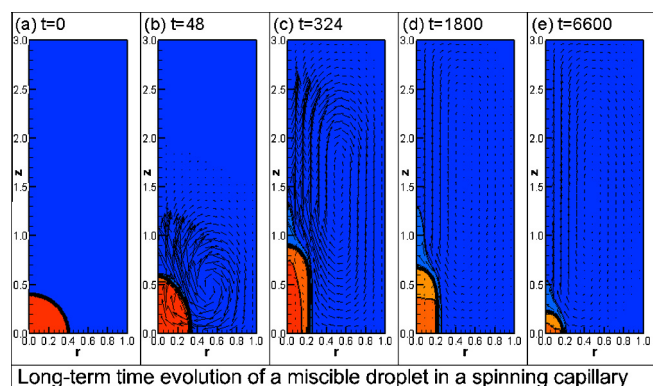


Regular Article

Phase-field modelling of a miscible system in spinning droplet tensiometer

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GRAPHICAL ABSTRACT



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ABSTRACT

The spinning drop tensiometry is used for measurements of surface tension coefficients, especially, when interfaces are characterised by low and ultra-low interfacial stresses. A droplet of lighter liquid is introduced into a rotating capillary that was initially saturated with another heavier liquid. The tube is subject to axial rotation that results in droplet's elongation along the tube's axis. The equilibrium shape of the droplet is used to determine the surface tension coefficient. In this work, the evolution of a slowly miscible droplet introduced into a spinning capillary is investigated. This technique is frequently employed for studies of the dynamics of miscible systems, even despite the fact that a strict equilibrium is never achieved in a mixture of fully miscible liquids. The numerical modelling of a miscible droplet is fulfilled on the basis of the phase-field (Cahn-Hilliard) approach. The numerical results are compared against the experimental data pursuing two objectives: (i) to verify the use of the phase-field approach as a consistent physics-based approach capable of accurate tracking of the short- and long-term evolution of miscible systems, and (ii) to estimate the values of the phenomenological parameters introduced within the phase-field approach, so making this approach a practical tool for modelling of thermohydrodynamic changes in miscible systems within various configurations.

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1. Introduction

In a tube with two closed ends filled with two immiscible liquids and then rotated about its axis, the centrifugal force separates the liquids so that a lighter one tends to occupy the inner part of

Nomenclature

$\bar{C}; C_{cr}; C = \bar{C} - C_{cr}$	concentration; value of concentration at the critical point; concentration with shifted reference point	$C_0; C_{in}$	equilibrium concentration profile for a flat interface (21); initial concentration profile (24)
$\bar{\rho}; \rho_{cr}; \rho = \bar{\rho} - \rho_{cr}$	density; density at the critical point; density with shifted reference point	$\delta; \delta_0; \delta_{in}$	interface thickness; thickness of an equilibrium flat interface (21); initial thickness of an interface
ρ_1 and $\rho_2; \phi$	densities of pure solvent and solute; density contrast (2)	r and $z; \vec{e}_r$	radial and axial coordinates; the unit vector in the radial direction
$f; f_0$	specific free energy; classical part of the specific free energy (3)	$\vec{u} = (u_r, u_z)$	vector of velocity
ϵ	capillary constant (3)	Π	pressure field
$\mu; \mu_0$	chemical potential; classical part of the chemical potential	ω and ψ	vorticity and streamfunction (20)
a and b	phenomenological parameters determining the thermodynamic state of a mixture	$L_*; \tau_*; \nu_*; p_*; \mu_*$	typical length and time, velocity, pressure and chemical potential (9)
α	mobility constant	$Pe; Ca; Re; Gr$	Peclet (10), capillary (11), Reynolds (12), and Grashof (13) numbers
Ω	angular velocity of the tube's rotation	A	non-dimensional parameter determining the thermodynamic state of a mixture
$R; H$	radius of the tube; half-length of the tube scaled in unites of R	$\sigma; \sigma_0; \sigma_V; \sigma_{CH}$	surface tension coefficient; surface tension coefficient for a flat interface (23); surface tension coefficient calculated from the Vonnegut formula (25); surface tension coefficient calculated from the structure of the solute/solvent interface (22)
$a_r; a_z; r_0; a_{r,eq}$	non-dimensional radial and axial dimensions of the droplet (measured in units of R); radius of an initial (spherical) droplet; droplet's radial dimension in the equilibrium state (for an immiscible droplet)	$m; m_0$	rate of change of the droplet's radial dimension; a constant (26)

the tube, while the heavier liquid occupies the outer part. The surface tension associated with the liquid/liquid interface aims to restore the spherical shape of the droplet. In a state of mechanical equilibrium, the droplet has an ellipsoidal shape defined by the balance of the centrifugal and surface tension forces. The equilibrium shape of the droplet is used to determine the value of the surface tension coefficient for the liquid/liquid interface.

Vonnegut [1] derived a simple formula to relate the surface tension coefficient to the droplet's dimensions. His formula assumes that the droplet is strongly elongated so it can be roughly represented as a cylinder of radius $a_r R$ and length $2a_z R$ (here R is the tube's radius, and a_r and a_z are the non-dimensional radial and axial dimensions of the droplet), with two hemispherical ends. The surface tension coefficient is then given by,

$$\sigma = \frac{1}{4}(\rho_1 - \rho_2)\Omega^2(a_r R)^3, \quad (1)$$

where ρ_1 and ρ_2 are the densities of two liquids in a mixture, and Ω is the angular velocity. Expression (1) is valid for sufficiently elongated droplets, when $a_z > 2a_r$, which is generally true for higher speeds of the tube's rotation.

This idea was employed to build a number of spinning drop apparatuses, see e.g. [2,3]. Further improvements to the design of a spinning drop tensiometer were suggested in a number of other studies. For instance, the case of lower rotation speeds, when buoyancy effects may become essential are examined in [4–6]. Inertial oscillations of the liquid/liquid interfaces were studied in [5]. Criteria to avoid the adhesion of a droplet to the tube walls were provided by Princen et al. [7], and, on the contrary, the spinning droplet technique was modified for measurements of the contact angles at the liquid/liquid/solid contact in another work by Princen and Vaidya [8]. In [9], a tube of square cross section was used, which permitted interpretation of the experimental data without knowledge of the refraction index of a denser liquid (this was normally needed to take into account the apparent magnification of the droplet).

The main advantages of the spinning drop method are as follows, (i) the force that determines the droplet's shape, i.e. the centrifugal force, can be varied at will, in contrast to the techniques

based on the action of the gravity force (pendant drop, sessile drop), (ii) the technique can be successfully employed for measurements of ultra-small values of interfacial stress, and (iii) the shape of the droplet is stable under 'normal' experimental conditions. There are several shortcomings as well. The main difficulty was found to be a sensitivity of a mechanical equilibrium of the droplet to e.g. alignment of the spinning tube, warming up of the apparatus, etc. For instance, Chan et al. [10] reported that the measured surface tension coefficient was a function of the rotation rate in their experiments. The reason of such dependence remained unexplained, with likely explanation that the liquid/liquid system did not reach the equilibrium state.

In the current work we deal with the surface tension introduced for an interface between two miscible liquids [11]. The surface tension stems from the difference in intermolecular interactions in different liquids at different sides of the interface. For a slowly miscible interface, such a difference in intermolecular forces would still exist for prolong time periods, resulting in e.g. a spherical shape of a honey droplet that is immersed in tea (despite the fact that honey and water are fully miscible). The need to introduce the surface tension for miscible interfaces was first pointed out by Korteweg [12] and van der Waals [13], and later by Zeldovich [14], Joseph [15] and others. The interfacial stresses for miscible systems are low and the interfacial diffusion makes the surface tension process-dependent. Nevertheless, the existence of the surface tension at miscible interfaces was confirmed in a number of experiments [16–21]. Such measurements were possible, as the changes in the surface tension were slower than the time needed for the measurements.

The use of the spinning drop tensiometry for measurements of the surface tension between two miscible liquids was first reported in [22,23]. Later, similar measurements were undertaken by Pojman's group [19,20], who found that the Vonnegut formula holds for long time periods after the liquids reach a thermal equilibrium. The time-changes in the surface tension upon dissolution of the droplet were reported. It was also found that the surface tension was independent of the rotation rate for the speeds over 6000 rpm, and it was independent of the initial droplet volume. It was also observed, that the droplet's interface remains sharp for the entire duration of the dissolution process.

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