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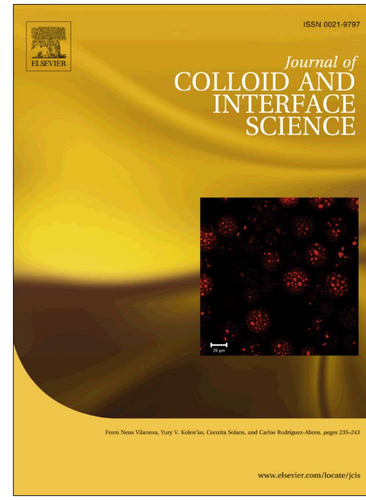

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Modeling the evaporation of sessile multi-component droplets

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Abstract

We extended a mathematical model for the drying of sessile droplets, based on the lubrication approximation, to binary mixture droplets. This extension is relevant for e.g. inkjet printing applications, where ink consisting of several components are used. The extension involves the generalization of an established vapor diffusion-limited evaporation model to multi-component mixtures. The different volatilities of the liquid components generate a composition gradient at the liquid-air interface. The model takes the composition-dependence of the mass density, viscosity, surface tension, mutual diffusion coefficient and thermodynamic activities into account. This leads to a variety of effects ranging from solutal Marangoni flow over deviations from the typical spherical cap shape to an entrapped residual amount of the more volatile component at later stages of the drying. These aspects are discussed in detail on the basis of the numerical results for water-glycerol and water-ethanol droplets. The results show good agreement with experimental findings. Finally, the accuracy of the lubrication approximation is assessed by comparison with a finite element method.

Keywords: droplets, lubrication approximation, multi-component flow, evaporation, Marangoni flow

1. Introduction

Because of the widespread range of applications, ranging from inkjet printing [1] and spray cooling [2] to the deposition of protein molecules on microarray slides [3] and more, the evaporation of sessile droplets in ambient gas has aroused significant interest in the latest decades. Although this represents a ubiquitous and classical phenomenon, the underlying physics, comprising the evaporation rate, the flow in the droplet and the dynamics at the contact line, is rather complex and still subject of ongoing research.

A substantial contribution to the theoretical understanding of the drying of droplets was achieved by Deegan et al. [4, 5] and Popov [6]. They found that the evaporation rate is essentially controlled by the diffusion of vapor from the droplet surface to the ambient, which results in a non-homogeneous evaporation rate with a singularity at the contact line. Based on this model, a variety of numerical and analytical investigations were carried out [7–14].

While the temperature-driven Marangoni flow or the presence of solute particles and their deposition to the substrate have been taken into account in some of these studies, they were confined to droplets consisting of a pure fluid. In applications like inkjet printing, however, the ink is typically a mixture of a solvent and various co-solvents, as well as surfactants and possibly colloidal solute particles. Experimentally, it has been shown that the evaporation of a binary mixture droplet can exhibit interesting non-monotonic behavior of the contact angle [15–19], initial condensation of water from the surrounding air [20] and presumably an entrapped residual amount of the more volatile component [21, 22]. Furthermore, Christy et al. [23] and Bennacer & Sefiane [24] revealed by particle image velocimetry that evaporating water-ethanol droplets undergo flow transitions: initially, in regime I, the preferential evaporation of ethanol triggers a solutal Marangoni flow that drives a complicated non-axisymmetric flow with multiple vortices in the droplet. After that, they observed a rapid transition with an intense velocity peak (regime II) followed by the usual capillary flow to the contact line as in the case of pure droplets (regime III). Recently, Zhong & Duan showed that the duration of the transition regime II can also last for rather long time, where they observed a nearly axisymmetric Marangoni flow [25].

Binary mixture droplets can also be combined with surfactants and surface-absorbed polymers to create homogeneous deposition patterns [26]. Beyond that, Cira et al. have recently shown that neighboring binary mixture droplets can interact through the vapor phase which allows the assembly of autonomous fluidic machines [27].

The present study attempts to reveal for the first time the underlying physical phenomena in a single axisymmetric binary mixture droplet by mathematical modeling and subsequent comparison with experiments. A mathematical model for the evaporation of sessile binary mixture droplets faces two challenges. On the one hand, the established diffusion-limited evaporation model has to be generalized to mixtures. As already mentioned by Guéna et