Density-Driven Flows in Evaporating Binary Liquid Droplets

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Flow mechanisms and evaporation dynamics of a micro-particle laden sessile droplet deposited on a solid substrate have attracted a lot of interest for the past 20 years because of emerging applications in a wide range of industries such as inkjet printing, additive manufacturing and biomedical assays. Most research focuses on single component droplets, revealing an outward radial flow driven by bulk liquid transport towards a pinned contact line [1]. However, in many applications, droplets contain two or more liquid components of different volatilities, warranting a detailed understanding of the flow driving mechanisms and the resulting deposition patterns in these more complex solutions.

In this work, we report a pioneering development of rotatable optical coherence tomography to image the flow patterns in tilted evaporating binary liquid droplets deposited on substrates with contact angles between 20° and 100°. The evaporation of the droplets proceeds in three distinct stages: chaotic (Stage I), convective (Stage II) and outward radial flow (Stage III). Stage I is characterised by random and strongly circulating vortices, attributed to solutal Marangoni flows driven by the preferential evaporation of the more volatile component (ethanol) [2]. Stage III is characterised by an outward capillary flow of the less volatile single component droplet (water) [1]. Our measurements conclusively demonstrate that, in contrast to the accepted view and conventional calculations of the Marangoni and Rayleigh numbers, the convective flow (Stage II) is induced within the droplets by a difference in density between an outer shell and the bulk of the droplets due to the evaporation of one of the components. We also use gas chromatography to determine the time evolution of the concentration of the more volatile component within the droplet and confirm that a simple analysis of volume data provides the same information. Finally, we establish a flow phase diagram demonstrating the conditions under which the different stages occur.

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