Capillary phenomena in miscible fluids

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Abstract

Interfacial tension between immiscible fluids is a well-defined, well-known quantity occurring in a wide range of phenomena. By contrast, this quantity is neither easily defined nor fully understood for miscible fluids. Following the work of Korteweg in 1901 [1], an "effective interfacial tension" (EIT) is supposed to exist between miscible fluids, which stems from spatial gradients of concentration or density in a multifluid system. Nevertheless, the existence of these so-called "Korteweg stresses" is still debated [2-3], since they are hardly detectable in simple miscible liquids, where diffusion rapidly smears out interfaces. In this talk I will give an overview on the recent results obtained in our lab showing that

capillary forces do exist at the boundary between miscible liquids.

First, I will report on the measurement of non-equilibrium interfacial tension of polymers and hard sphere suspensions in contact with their own solvent [3]. By visualizing fingering instability (VF) in radial Hele-Shaw geometry, appearing when the solvent displaces the suspensions, we have measured interfacial tensions in function of the volume fraction of the suspended colloids or polymers, showing that the internal degrees of freedom of the particles drive the low volume fraction behavior. Our results support the existence of a positive tension between miscible fluids, confirm the quadratic scaling predicted by Korteweg [1] for long linear and crosslinked polymers and show a positive rapidly growing tension for hard sphere suspensions up to maximum packing, whose description necessitates a theoretical framework going beyond the classic square gradient model [1,2].

More recently, we have performed a series of spinning drop tensiometry (SDT) experiments. In SDT, a drop of one fluid is injected in a background fluid contained in a cylindrical capillary. The capillary is rapidly spun, and one follows the evolution of the drop shape by video imaging. We have modified a commercial apparatus and use fluorescent drops in order to retrieve the full three-dimensional concentration profile of the drop, allowing us to follow the dynamics of the interface with unprecedented detail.

We have first investigated the model case of immiscible molecular fluids, for which we find that the time evolution of the drop shape agrees nicely with theoretical predictions [4] that where not tested so far. These results constitute a solid starting point for SDT on miscible fluids. We have found that the shape of a miscible drop continuously evolves, in contrast to the saturation effect reported in previous investigations that explored a smaller temporal range. By varying systematically the composition of the fluids, we show that the shape of miscible drops during elongation is not only determined by the density and viscosity contrast with respect to the background fluid, but also by the chemistry of the fluids. This crucial

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result rules out the possibility that the drop dynamics are purely dictated by hydrodynamics and strongly hints at the existence of an effective interfacial tension.

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