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Doctoral Dissertation

Interfacial Fluctuation and Fluid Convection in Meniscus Splitting of Polysaccharide Dispersions

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Abstract

Splitting patterns in nature, such as geometric patterns of Salt Lake, wrinkles patterns of hydrogels, and Rayleigh–Bénard patterns in heated fluids, are not randomly generated, but rather are self-organizing in nonequilibrium systems. Closely related to the dissipative structures, the evolution from disorder to ordered patterning through continuous exchange of energy and matter in open systems. This thesis focuses on the meniscus splitting patterns in drying polysaccharides dispersion systems and aims at exploring the kinetic mechanism both on the interfacial behavior and inter-fluid behaviors during the evaporation process, specifically. In Chapter 2, the recognition of spatial finiteness in "meniscus splitting phenomena" in aqueous polymer dispersions is demonstrated during water evaporation. By providing heat energy to polymer dispersions in a Hele-Shaw cell, an interface fluctuation with concentration unevenness is induced to split the evaporative interface. The results of the quasi-natural experiments revealed that the nonequilibrium drying period for repositioning polymer clusters allows for considerable changes in Reynolds number in a low range ($<10^{-6}$) to form multiple nuclei, like the life in living organisms. In Chapter 3, by studying the internal flow during nonequilibrium process, the evolution of unique mass flows as Marangoni circulations is demonstrated during the growth of splitting pattern. The confluences of multiple circulations are distinctly aligned one-to-one with the nucleation positions. This convection patterns periodically integrates the polymers in a quasi-two-dimensional space. By studying the Marangoni convection in different capillary environments, the disappearance of convection implies a different role of capillary forces and surface tension. In Chapter 4, we investigate how evaporation-induced convection influence interfacial fluctuations and nucleation behavior in meniscus splitting. Using pectin dispersion, we compared the synchronous and asynchronous nucleation process. Through introducing the asymmetry bottoms (flat, step-like, sloped) to break convective symmetry, confirmed the influence of spatial symmetry on convective nucleation. Furthermore, we compare pectin and xanthan gum dispersions, which exhibit dramatically different flow behaviors under evaporation: Marangoni circulations in the former, and almost no internal flow in the latter. From the emergence of convection to interfacial fluctuations and flocculation-induced nucleation, the hydrodynamic mechanism in meniscus splitting offers new insights into the evaporation induced self-assembly of polymer dispersion/colloidal. This has significant implications for understanding pattern formation in nature, particularly interfacial structures in biopolymer and soft matter systems spanning micro to macroscopic scales.

Keywords: Dissipative structures, Fluctuations, Polysaccharides, Skin layer, Marangoni effect, Capillary effect.