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Physicochemical hydrodynamics of condensate-formation in evaporation-driven phase-separation

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Multicomponent droplets composed of partially-miscible constituents can spontaneously undergo liquid-liquid phase separation during evaporation. Crucially, the overall dynamics are dictated by the evaporation-driven hydrodynamics of the droplet as well as the inter-phase physicochemical interactions. The latter may lead to liquid-liquid phase separation, resulting in segregated phase-rich and phase-poor domains, and the formation of condensates. We illustrate this in figure 1, which depicts time (t)-lapsed fluorescence micrographs of the evaporation of a sessile droplet containing an aqueous ternary solution of polyethylene glycol (PEG), dextran (DEX; labeled red), and poly-L-lysine (PLL; labeled blue). As evaporation proceeds, spontaneous phase separation results in the emergence of phase-segregated lobes from the contact line (figures 1a-ii and 1a-iii), where DEX preferentially accumulates at the droplet periphery. Eventually condensates (figure 1a-iv) start appearing, resulting from the selectively binding of PLL to DEX via electrostatic interactions. Remarkably, the underlying complex physicochemical hydrodynamics leads to the spatial localization of the condensates along radially-arranged spokes that diverge from the center of the droplet. Moreover, we also observe an ordered size-segregation of the condensates along each radial spoke. Crucially, and perhaps quite puzzlingly, these liquid-like condensates do not exhibit coalescence. These patterns are qualitatively conserved across a wide range of concentrations in our experiments, albeit exhibiting a variation of the sizes and precise spatial locations of the condensates. Our work aims to shed light on the highly non-trivial interplay between electrostatic complexation, localized phase separation, and evaporation-driven hydrodynamics governing the dynamics of formation and transport of these condensates.

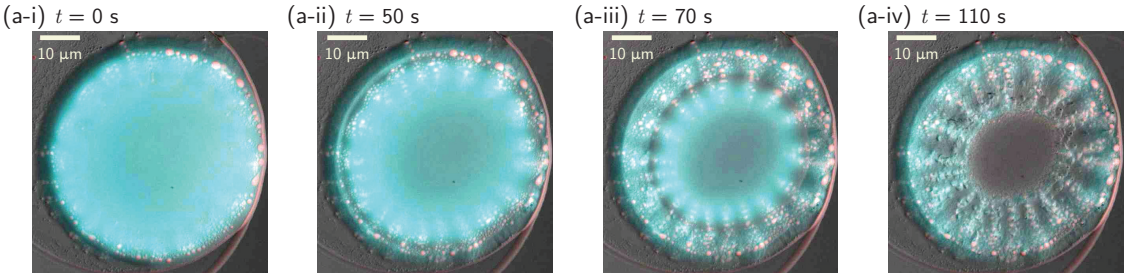

Figure 1: Time (t)-lapsed fluorescence micrographs of the evaporation of a droplet of a ternary aqueous solution containing polyethylene glycol, dextran (labeled red), and poly-L-lysine (labeled blue) leads to the formation of phase separated domains, and eventually condensates; the latter are spatially-arranged along radially-arranged spokes emanating from the center of the droplet.

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