Self-Assembly of Charge-Containing Copolymers at the Liquid–Liquid Interface

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In this study we demonstrate that amphiphilic polymers adsorbed at the liquid-liquid interface self-assemble through the hydrophilic/charged groups and exhibit a rich variety of shapes as the charge and hydrophilic fractions change. Using interfacial tension experiments, we have shown that an increase in charge leads to a significantly enhanced adsorption at the chloroform-water interface in both the homopolymer and copolymer cases. Molecular dynamics simulations suggest that this is due to an increased concentration of amphiphiles and the corresponding aggregate morphology at the chloroform-water interface.

Significance and Impact

Quantitatively understanding the self-assembly of amphiphilic macromolecules at liquid–liquid interfaces is a fundamental scientific concern due to its relevance to a broad range of applications including bottom-up nanopatterning, protein encapsulation, oil recovery, drug delivery, and other technologies. Elucidating the mechanisms that drive assembly of amphiphilic macromolecules at liquid–liquid interfaces is challenging due to the combination of hydrophobic, hydrophilic, and Coulomb interactions, which require consideration of the dielectric mismatch, solvation effects, ionic correlations, and entropic factors.



Figure: (a) Schematic setup for measuring the interfacial tension. The drop deformation is indicative of changes in the surface tension. The interfacial tension is calculated by fitting the drop profile and determining the shape parameter B0 and radius of curvature R0, which are related to the interfacial tension, γ , via eq 1.

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