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Electrostatic Assembly of Polymers and Nanoparticles at Liquid-Liquid Interfaces.[1]

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The electrostatic attraction between charged solutes on opposite sides of the interface between immiscible liquids offers an efficient route to the self-assembly of two-dimensional films. As implemented by us, a hydrophobic polymer with amine end(s) or block(s) is presented in an oil phase, and a negatively charged nanoparticle is presented in an aqueous phase; both solutes are insoluble in the opposite phase but efficiently driven to the liquid-liquid interface by mutual electrostatically attraction to the solute in the opposite phase. Depending on experimental conditions (salt concentration, pH, solute concentrations, etc.), a continuous, nanoscopically thin composite film builds at the oil-water interface over the timescale of minutes, often accompanied by a dramatic reduction of interfacial tension akin to that observed for a surfactant. Film formation and properties by the new route will be discussed, as principally probed through pendant drop interfacial tensiometry and pendant drop interfacial rheometry. Components of model system are toluene-dissolved amine end-capped polystyrene and water-dispersed acid-treated carbon nanotubes or citrate-treated gold nanospheres. Film structures are complicated, as are crucial electrostatic interactions near the interface. With amine end-capped polystyrene partnered with acid-treated carbon nanotubes, high pH (above 5) and high polystyrene molecular weight (above 5000 g/mol) strongly hinder film formation. These films, which are liquid-like, show two viscoelastic relaxations, a fast relaxation (about 10 s) associated with polystyrene chain rearrangements (slightly impacted by carbon nanotube association) and a slow relaxation (about 20 min) associated with polystyrene adsorption/desorption; at intermediate times (or frequencies), the two-dimensional storage and loss moduli follow approximately the same power law dependences.

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