Structure and Rheology of Polyelectrolyte Complex Coacervates

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Abstract:
Polyelectrolyte complexes are highly tunable materials that span from low-viscosity liquids (coacervates) to high-modulus solids with high water content, making them attractive as surface coating, membrane purification and bioadhesive materials. However, most of their properties and their effects with salt, pH, polymer ratio and temperature have only been qualitatively described. Here, we present a scattering investigation of the structure and chain conformations, and rheological properties of polyelectrolyte complex (PEC) coacervates comprising model polyelectrolytes. Systematic studies using small-angle X-ray scattering (SAXS) of the structure and chain behavior in liquid PEC coacervates revealed a physical description of these materials as strongly screened semidilute solutions of polyelectrolytes comprising oppositely charged chains. At the same time, solid PECs were found to be composed of hydrogen-bonding driven stiff ladder-like structures with large correlation lengths. While the liquid complexes behaved akin to semidilute polyelectrolyte solutions upon addition of salt, the solids were largely unaffected by it. Terminal relaxations of the chains in PEC coacervates were explored by rheology measurements. Excellent superposition of the dynamic moduli data was achieved by a time-salt superposition, although with the shift factors varying more strongly than previously reported with increasing salt concentration.