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Stereoregularity Drives Precipitation in Polyelectrolyte Complex Formation¹ MATTHEW TIRRELL, SARAH PERRY, LORRAINE LEON, MATTHEW KADE, DIMITRIS PRIFTIS, University of Chicago, KATIE BLACK, University of California, Berkeley, KYLE HOFFMAN, JONATHAN WHITMER, JIAN QIN, JUAN DE PABLO, University of Chicago — This study investigates the effect of stereoregularity on the formation of polypeptide-based complex formation and assembly into micelles, hydrogels and ordered phases. We demonstrate that fluid complex coacervate formation (rather than solid complex precipitation) between oppositely charged polypeptides requires at least one racemic partner in order to disrupt backbone hydrogen bonding networks and prevent the hydrophobic collapse of the polymers into compact, fibrillar secondary structures. Computer simulations bear this out and enable visualization of the molecular structure of the complexes. The ability to choose between conditions of fluid phase formation and solid phase formation is a useful tool in developing new self-assembled materials based on polyelectrolyte complex formation.

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