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Gel Phase Formation in Dilute Triblock Copolyelectrolyte Complexes SAMANVAYA SRIVASTAVA, MARAT ANDREEV, IME, The University of Chicago, VIVEK PRABHU, NIST, JUAN DE PABLO, MATTHEW TIRRELL, IME, The University of Chicago — Assembly of oppositely charged triblock copolyelectrolytes into phase-separated gels at extremely low polymer concentrations (<1 % by mass) has been observed in scattering experiments and molecular dynamics simulations. In contrast to uncharged, amphiphilic block copolymers that form discrete micelles at low concentrations and enter a phase of strongly interacting micelles in a gradual manner with increasing polymer concentrations, the formation of a dilute phase of individual micelles is prevented in polyelectrolyte complexation-driven assemblies of triblock copolyelectrolytes. Gel phases form and phase separate almost instantaneously upon solvation of the copolymers. Furthermore, molecular models of self-assembly demonstrate the presence of oligo-chain aggregates in early stages of triblock copolyelectrolyte assembly, at experimentally unobservable polymer concentrations. Our discoveries not only contribute to our fundamental understanding of the structure and pathways of complexation driven assemblies, but also raise intriguing prospects for formation of gel structures at extraordinarily low concentrations, with applications in tissue engineering, agriculture, water purification and theranostics.

Samanvaya Srivastava
Univ of Chicago

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