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Gel Phase Formation in Dilute Triblock Copolyelectrolyte Complexes

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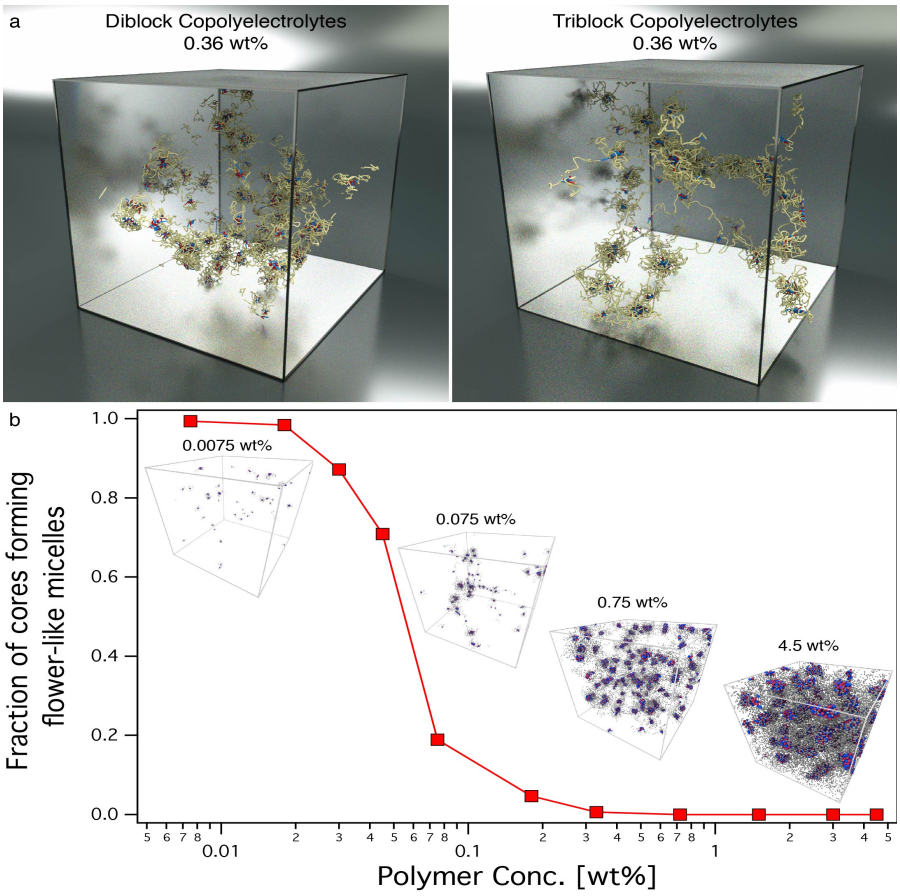

Caption: *MD Simulations reveal interconnected triblock networks. (a) Snapshots of the simulation box showing self-assembled structures comprising oppositely charged di-and tri-block copolyelectrolytes. The polycation, polyanion and neutral blocks are depicted by red, blue and grey colored beads, respectively. (b) Fraction of PEC cores forming isolated flower-like micelles as a function of polymer concentration. The fraction approaches a near zero value at ϕ = 0.36 wt %.*

Scientific Achievement

Assembly of oppositely charged triblock copolyelectrolytes into gel phases at extremely low polymer concentrations (< 1 wt%), accompanied with a phase separation of the gel phases, is reported using scattering experiments and molecular dynamics simulations. Unlike 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, and 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.

Significance

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.

Citation

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