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Polyelectrolyte Complex Hydrogels: Self-assembly and the Influence of Charged and Neutral Blocks SAMANVAYA SRIVASTAVA, DAVID GOLDFELD, ADAM LEVI, JUN MAO, Univ of Chicago, WEI CHEN, Argonne National Laboratory, MATTHEW TIRRELL, Univ of Chicago — Polyelectrolyte complexes (PEC) form when oppositely charged polyelectrolyte chains spontaneously associate and phase separate in aqueous mediums. Bulk phase separation of the PECs can be evaded by combining one or both of the polyelectrolytes with a neutral polymer, thus engineering pathways for self-assembled PEC micelles and hydrogels. The PEC domains in these assemblies can encapsulate therapeutics as well as genetic materials and thus have tremendous potential in drug delivery and tissue engineering applications. We will present insights on the equilibrium structure and self-assembly kinetics of PEC hydrogels with large-scale ordering of the nanoscale PEC domains through detailed structure characterization and rheology studies of self-assembled materials comprising of functionalized polyallyl glycidyl ethers (PAGE) connected to either single poly(ethylene glycol) (PEG) chain to form diblock copolymers or as functionalized end-groups on a triblock copolymer with a PEG midblock. The effect of key parameters such as polymer concentration, polymer block lengths, salt, ionic strength, and degree of charge mismatch on the equilibrium materials properties will be discussed, with a special emphasis on the structure-defining role of the charged blocks and the structure-directing role of neutral blocks. Additionally, interesting similarities, and differences between structures and dynamics of hydrogels comprising diblock and corresponding triblock polyelectrolytes, respectively, will be discussed.

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