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Complex Coacervation of Oppositely Charged Polyelectrolyte Mixtures: a Liquid-State Theory Study PENGFEI ZHANG, California Institute of Technology, NAYEF ALSAIFI, King Fahd University of Petroleum Minerals, JIANZHONG WU, University of California, Riverside, ZHEN-GANG WANG, California Institute of Technology — Complex coacervation refers to a phase separation of oppositely charged polyelectrolyte (PE) mixtures into a PE-poor supernatant phase and a PE-rich coacervate phase. This phenomenon arises from the delicate interplay among the mixing entropy, excluded volume, chain connectivity, and electrostatic interactions. Using a simple liquid-state (LS) theory that accounts for all of these effects, we study the phase behaviors of a mixture of oppositely charged PE solutions. In comparison with the prediction from the well-known Voorn-Overbeek (VO) theory, the LS theory predicts a much narrower phase-separated region for the stoichiometric mixture. Moreover, the LS theory predicts that salt concentration is higher in the supernatant phase than the coacervated phase, opposite to the VO prediction. The effect of non-stoichiometry on the phase behavior is also studied in detail. In particular, we find two different regimes of mixing ratio with respect to the partition of excess PE chains in the coexisting phases for the mixture without extra salt: in the weakly asymmetric regime, nearly all of the excess PE-chain are distributed in the coacervate phase, while in the strongly asymmetric regime, most of the excess PE chains are accumulated in the supernatant phase.

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