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Self-assembly of block copolymers of two negatively charged polyelectrolytes and multivalent cations

Polyelectrolytes are widely used in industrial processes as anti-scaling agents, for waste water treatment or as rheology modifier in cement. They also serve as model system for natural polyelectrolytes such as proteins. The specific interaction of the charged polymer chain with oppositely charged species such as surfactants or ions is the basis for many applications in household and industry. Previous works on two different negatively charged polyelectrolytes showed that the interaction with multivalent cations such as Ca^{2+} , Sr^{2+} or Ba^{2+} strongly depends on the chemical nature of the anionic moiety.[1,2] In the case of polyacrylic acid (PAA) phase separation occurs with Ca^{2+} , Sr^{2+} and Ba^{2+} , whereas polystyrene sulfonate (PSS) only shows a phase separation with Ba^{2+} . We use this cation selectivity to self-assemble a block copolymer (PAA-*b*-PSS) consisting of polyacrylic acid and polystyrene sulfonate into well-defined micelles. The structure of these micelles was studied by a combination of light, small-angle neutron and X-ray scattering. Internal and external contrast variation in SANS by using different $\text{H}_2\text{O}/\text{D}_2\text{O}$ ratios as well as partly deuterated polymers provides detailed information about the polymer structure. On the other hand, SAXS gives information about the location of the multivalent cations. The block copolymer micelles show multi-responsive behavior towards pH, temperature or salt concentration. The present work demonstrates that the system can be switched between lower (LCST) and upper critical solution temperature (UCST) by changing from Ca^{2+} to Ba^{2+} . By using Sr^{2+} as a counter ion, we were able to combine LCST and UCST behaviour and create a double thermoresponsive system. Isothermal titration calorimetry complements our scattering data and shows that the temperature behaviour is a subtle balance between the release of counter ions and binding enthalpy. Understanding these interactions allows us to design new responsive and schizophrenic materials.

1. M. Hansch et al., *J. Chem. Phys.* (2018), 114906.
2. S. Lages et al., *Macromolecules* 42 (2009), 4288-4299.

Preferred topic

Conformation of polymers in solvents

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