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The Structural and Thermal Behavior of the Thermoresponsive Polymer Poly(N-isopropylmethacrylamide) in Aqueous Solution

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Compared to the well-investigated poly(*N*-isopropylacrylamide) (PNIPAM), Poly(*N*-isopropylmethacrylamide) (PNIPMAM) has a higher phase transition temperature (43 ℃ instead of 32 ℃). This may be due to the presence of the additional methyl groups on the vinyl backbone, which lead to steric hindrance and weaken the intramolecular interactions. To understand how these effects influence the thermal and structural behavior of PNIPMAM aqueous solutions, we investigate the phase behavior of PNIPMAM in D_2O using turbidimetry, differential scanning calorimetry, Raman spectroscopy, small-angle and very small-angle neutron scattering (at KWS-1 and KWS-3 at MLZ). The PNIPMAM solutions undergo first macroscopic phase transition, but the PNIPMAM chains only dehydrate 2~3 ℃ above T_{CP}. The methyl groups in PNIPMAM lead to loosely packed large-scale inhomogeneities, and physical crosslinks already in the one-phase state. Besides, the local chain conformation of PNIPMAM is more compact than the one of PNIPAM, which is due to enhanced attractive intermolecular interactions originating from the hydrophobic moieties. In the two-phase state, PNIPMAM features larger and more hydrated mesoglobules than PNIPAM. This is due to the steric hindrance caused by the methyl groups, which weaken the intrapolymer interactions. We conclude that the methyl groups in PNIPMAM chains have a significant impact on the hydration behavior and the structural behavior around the phase transition.

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