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Temperature-dependent Phase Behavior of the Thermoresponsive Polymer Poly(N-isopropylmethacrylamide) in Aqueous Solution

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Compared to the widely-investigated poly(N-isopropylacrylamide) (PNIPAM), poly(N-isopropylmethacrylamide) (PNIPMAM) has a higher phase transition temperature (43 °C instead of 32 °C). PNIPMAM has a similar chemical structure as PNIPAM, but the additional methyl groups on its backbone may lead to steric hindrance and weaker intramolecular interactions. To understand how these effects affect the thermal and structural behavior of PNIPMAM aqueous solutions, we investigate the phase behavior of PNIPMAM in D₂O 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 °C above TCP. The methyl groups in PNIPMAM give rise to a more compact local chain conformation than in PNIPAM. Moreover, physical crosslinks and loosely packed large-scale inhomogeneities and physical crosslinks appear already in the one-phase state. We assign these differences to enhanced attractive intermolecular interactions resulting from the hydrophobic methyl groups. In the two-phase state, PNIPMAM mesoglobules are larger and more hydrated than PNIPAM mesoglobules. This is attributed to the steric hindrance caused by the methyl groups, which weaken the intrapolymer interactions. Thus, the methyl groups in PNIPMAM chains play a crucial role in the thermal and structural behavior around the phase transition.

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