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Injectable hydrogels from thermoresponsive tri- and tetrablock terpolymers investigated using scattering methods

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Lower critical solution temperature (LCST) polymers have attracted great interest for 3D bioprinting, as they can form a runny solution at room temperature, but a hydrogel at body temperature. In block copolymers featuring LCST blocks, the mechanical properties in the gel state strongly depend on the architecture of the polymer. Here we address an ABC triblock terpolymer and a BABC tetrablock terpolymer consisting of the hydrophilic OEGMA (A), the hydrophobic BuMA (B), and the thermoresponsive DEGMA (C).

The results from dynamic light scattering (DLS) on dilute solutions indicate that the hydrodynamic radii of the micelles formed by both, ABC and BABC, increase strongly above 25 ℃, and the solutions feature a cloud point, i.e. aggregation of the micelles sets in. By synchrotron small-angle X-ray scattering and neutron scattering (SAXS/SANS), we found that the triblock terpolymers ABC form spherical core-shell micelles, that transform into cylinders at high temperatures, and then form compact large aggregates upon further heating. In contrast, the core-shell micelles formed by tetrablock terpolymers BABC stay spherical and form small aggregates at higher temperatures. Particularly, at high concentration, BABC transforms into an elastic two-compartment network upon further heating. Hence, the additional hydrophobic block in BABC results in a different type of gel formation.

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