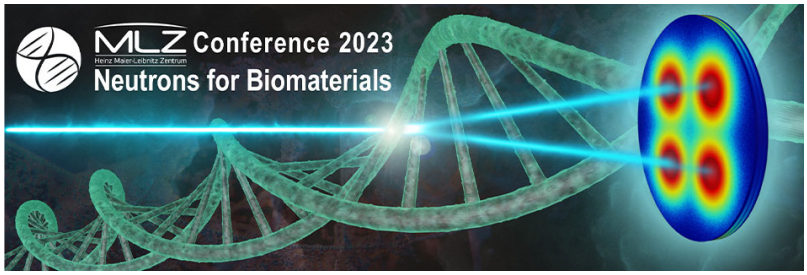

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Injectable hydrogels from thermoresponsive tri- and tetrablock terpolymers

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Lower critical solution temperature (LCST) polymers have attracted great interest for 3D bioprinting, as they are water-soluble and form solutions at room temperature, while they form a hydrogel at body temperature [1]. The hydrogel properties depend strongly on the architecture of polymers and their concentration. Poly(ethylene glycol) (PEG) based thermoresponsive polymers are particularly promising, because PEG is a hydrophilic, biocompatible and FDA-approved polymer. Here, we address an ABC triblock terpolymer and a BABC tetrablock terpolymer consisting of the hydrophilic oligo(ethylene glycol) methyl ether methacrylate with an average Mn of 300 g/mol (OEGMA, A), hydrophobic *n*-butyl methacrylate (BuMA, B) and thermoresponsive di(ethylene glycol) methyl ether methacrylate (DEGMA, C). Dynamic light scattering (DLS) on dilute micellar solutions shows that for both, ABC and BABC, the hydrodynamic radii of the micelles increase strongly above 25 ℃, and the solutions feature a cloud point at 31-34 ℃. By small-angle neutron scattering (SANS) on 15 wt% polymer solutions, the structural changes at the origin of the gel formation were identified: While the triblock terpolymers ABC form spherical core-shell micelles, that transform into cylinders at high temperatures, the spherical core-shell micelles formed by the BABC tetrablock terpolymers remain spherical and form loose aggregates at higher temperatures, that become more compact upon further heating. Thus, for the two architectures, the hydrogel formation proceeds via vastly different mechanisms.

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