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Influence of the architecture on the micellar structures from thermo- and pH-responsive triblock terpolymers

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Self-assembled thermoresponsive hydrogels are attractive candidates for 3D bioprinting and tissue engineering. Here, we present a novel dual-stimuli system that features PDMAEMA, a weak cationic polyelectrolyte with pKa ~7.5 that is thermoresponsive when uncharged. It is combined in a triblock terpolymer with a hydrophobic polymethyl methacrylate block (PMMA, B) and a poly(N-(2-methacryloyloxyethyl) pyrrolidone) block (PNMEP, C), a hitherto uninvestigated polymer with a pKa of 5.2. The present study addresses the effect of the sequence of the two responsive blocks, namely BAC and BCA, on the temperature and pH-dependent micellar structures in dilute aqueous solution. Using dynamic light scattering and synchrotron small-angle X-ray scattering, we found that the micellar sizes and aggregation behaviour depend strongly on pH, temperature and the block sequence: At pH 6, both architectures show only little size change with increasing temperature. In contrast, at pH 8 and 25 ℃, the hydrodynamic radius of the micelles formed by the BCA triblock terpolymer (24.5 nm) is larger than the one of the BAC (18.2 nm). Moreover, an increase in temperature results in the presence of large clusters of BCA micelles at 32 ℃ and above, while BAC does not exhibit any shape and size change up to 43 ℃. Hence, the micellar structures depend strongly on the pH-value and temperature, and the same can be expected for the corresponding micellar hydrogels.

Primary authors: ZHENG, Feifei (TUM); Ms ROMÁN QUINTERO, Montserrat; XU, Wenqi (Technical University of Munich, TUM School of Natural Sciences, Soft Matter Physics Group); ZHANG, Peiran (TUM); Dr SOLOVIOV, Dmytro (EMBL); Ms WANG, Lezhi (Imperial College London); GEORGIOU, Theoni K.; PAPADAKIS, Christine (Technische Universität München, Physik-Department, Fachgebiet Physik weicher Materie)

Presenter: ZHENG, Feifei (TUM)

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