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New insights into the self-assembly of amphiphilic poly(ethylene glycol-b-caprolactone) diblock copolymers in aqueous solution

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The ability to self-assemble into nanostructures is a fundamental phenomenon in many living and non-living systems. The design of polymeric systems that assemble into hierarchically structured nanomaterials requires careful consideration of the microstructure and molecular interactions. For many applications, such as micellar drug delivery systems, precise control over the self-assembly process is required. However, the relationship between molecular structural characteristics of block polymers and their micellar self-assembly mechanisms varies with different block types. In this study, the effect of polymer molecular weight and copolymer block ratio on the micellization of poly(ethylene glycol-b-caprolactone) (PEG-b-PCL) block copolymers was investigated. The stealth properties of PEG and biodegradable nature of PEG-b-PCL make it a suitable choice for biomedical applications, including tissue engineering and drug delivery. Nuclear magnetic resonance (NMR) and dynamic light scattering (DLS) were used to measure the diffusion of block copolymers in water, from which the hydrodynamic diameters and dispersity of the polymer aggregates were determined; three aggregation scenarios were inferred from the data, including unimers (no self-assembly), large metastable aggregates, and monodisperse micelles. Small-angle x-ray scattering (SAXS) from polymer solutions provided morphological information on the shape of the micelles and their relationship to the polymers' microstructure. The PEG molecular weight and PCL:PEG ratio was the primary factor affecting micelle shape. A clear transition from unimers to large aggregates to cylindrical and ellipsoid micelles was observed as the PEG molecular weight and PCL:PEG ratio increased, with an increase in the micelle hydrodynamic radii. We therefore propose a self-assembly phase diagram for the PEG-b-PCL system in aqueous media by combining NMR, DLS and SAXS data. Block copolymer composition with larger PEG molecular weights and larger PEG-b-PCL block ratios formed more monodisperse micelles, whereas copolymer compositions with smaller PEG molecular weights and smaller PEG-b-PCL block ratios formed large metastable aggregates.

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