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Amphiphilic Spider Silk-Like Block Copolymers with Tunable Physical Properties and Morphology for Biomedical Applications¹ WEN-WEN HUANG, SREEVIDHYA KRISHNAJI, DAVID KAPLAN, PEGGY CEBE, Tufts University — Silk-based materials are important candidates for biomedical applications because of their excellent biocompatibility and biodegradability. To generate silk amphiphilic biopolymers with potential use in guided tissue repair and drug delivery, a novel family of spider silk-like block copolymers was synthesized by recombinant DNA technology. Block copolymer thermal properties, structural conformations, protein-water interactions, and self-assembly morphologies were studied with respect to well controlled protein amino acid sequences. A theoretical model was used to predict the heat capacity of the protein and protein-water complex. Using thermal analysis, two glass transitions were observed: Tg1 is related to conformational changes caused by bound water removal, while Tg2 (>Tg1) is the glass transition of dry protein. Real-time infrared spectroscopy and X-ray diffraction confirmed that different secondary structural changes occur during the two Tg relaxations. Using scanning electron microscopy, fibrillar networks and hollow vesicles are observed, depending on protein block copolymer sequence. This study provides a deeper understanding of the relationship between protein physical properties and amino acid sequence, with implications for design of other protein-based materials.

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