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Self-assembly Morphology and Crystallinity Control of Di-block Copolymer Inspired by Spider Silk[1] WENWEN HUANG, SREEVIDHYA KRISHNAJI, DAVID KAPLAN, PEGGY CEBE, Tufts University — To obtain a fuller understanding of the origin of self-assembly behavior, and thus be able to control the morphology of biomaterials with well defined amino acid sequences for tissue regeneration and drug delivery, we created a family of synthetic silk-based block copolymers inspired by the genetic sequences found in spider dragline, HABn and HBAn (n=1,2,3,6), where B = hydrophilic block, A = hydrophobic block, and H is a histidine tag. We assessed the secondary structure of water cast films by Fourier transform infrared spectroscopy (FTIR). The crystallinity was determined by Fourier self-deconvolution of amide I spectra and confirmed by wide angle X-ray diffraction (WAXD). Results indicate that we can control the self-assembled morphology and the crystallinity by varying the block length, and a minimum of 3 A-blocks are required to form beta sheet crystalline regions in water-cast spider silk block copolymers. The morphology and crystallinity can also be tuned by annealing. Thermal properties of water cast films and films annealed at 120 C were determined by differential scanning calorimetry and thermogravimetry. The sample films were also treated with 1,1,1,3,3,3-Hexafluoro-2-propanol (HFIP) to obtain wholly amorphous samples, and crystallized by exposure to methanol. Using scanning and transmission electron microscopies, we observe that fibrillar networks and hollow micelles are formed in water cast and methanol cast samples, but not in samples cast from HFIP.

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Wenwen Huang
Tufts University

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