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Mechanism of Thermal Crystallization in Silk Fibroin[1] XIAO HU, DAVID KAPLAN, PEGGY CEBE, Tufts University — We investigated the formation of beta pleated-sheets in *B. mori* silk fibroin films cast from water solution. Differential scanning calorimetry (DSC), its temperature-modulated variant (TMDSC), and the time-resolved techniques of Fourier Transform infrared spectroscopy and X-ray scattering, were used to monitor the detailed structural changes of silk fibroin during heating and isothermal crystallization above the glass transition temperature, Tg. Results show that bound water molecules inside the film play a very important role for the transformation from non-crystalline fibroin to crystalline beta sheet containing fibroin. Silk fibroin forms a new tighter structure with loss of intermolecular bound water during heating, which promotes the formation of the beta-sheets crystals above Tg. In addition, quick heating of the silk fibroin causes a water-induced glass transition, which results in a temporary water-silk structure where the bound water acts as a plasticizer for this polymeric system. This study will lead to a deeper understanding of the formation of beta pleated-sheets during the crystallization process in silk fibroin, with implications for the crystallization of the naturally occurring silk.

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