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Flow-induced beta-sheet formation in silk fibroin solutions

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Silk fibres from the Bombyx Mori silkworm are well known for their impressive strength and toughness. They have mechanical properties that are comparable to the best synthetic fibres available, and consequently have numerous applications as structural materials in biomedicine and engineering. Despite their importance as high performance materials, a comprehensive understanding of the how the sequence of the structural protein, fibroin, impacts the fibre formation during the spinning process is still lacking. In this talk, we present experimental studies of reconstituted silk fibroin proteins and supporting computer simulations of silk-mimetic peptide fragments that suggest two important mechanisms in the fibre formation process: (1) tyrosine residues in the fibroin sequence template the inter-molecular association chains in solution, and (2) flow-induced chain tension acts to nucleate the formation of inter-molecular beta sheet content in an orientational dependent manner. Moreover, we show that covalent cross-linking of tyrosine side chains results in spontaneous beta-sheet formation upon removal of water, even in the absence of shear, while non-crosslinked samples require post treatment to induce similar levels of crystallinity. The implications of these findings for ex vivo fabrication of high-strength silk fibres will be discussed.

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