Physical Aspects of Polymer Science
8–10 September 2015
Manchester Institute of Biotechnology
Manchester, UK
http://paps15.iopconfs.org

Tuesday 8 September, Session 1, 14:35 – 14:55

Investigating the protein gelation mechanism underlying natural silk fibre spinning

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Silks are fibres spun by arthropods, such as spiders and many lepidoptera larvae (i.e. caterpillars). The impressive mechanical properties of silks from orb weaving spiders are well known [1]; even more remarkable, though, is the means by which they are spun. Silk fibres are produced 'on demand' and relatively quickly, by extruding a proteinaceous feedstock, which is prepared and stored inside the animal in specialised organs (silk glands) [2]. In fact, this spinning process distinguishes silks from other animal fibres such as wool or hair, which are grown continuously, but much more slowly.

Thus for silks, natural fibre formation occurs at ambient temperature, by a flow-induced phase-change, with minimal change in chemical composition [3]. This is in stark contrast to industrial fibre spinning processes, which typically involve considerable changes in temperature (i.e. melt spinning) or chemical composition (i.e. solution spinning).

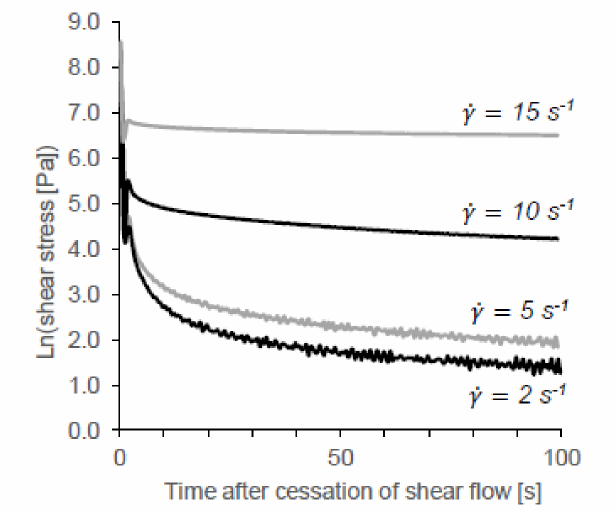

Figure 1: Stress relaxation observed following shear flow (γ = 100) at the shear rates shown.

Rheology provides a powerful method for studying silk feedstocks [4,5]. The work reported here used a combination of stress relaxation and oscillatory measurements to follow changes in molecular dynamics of silk feedstocks from the domesticated silkworm (*Bombyx mori)* during gelation [5]. It was found that some shear stress persisted for a considerable time (> 100 s) following the cessation of relatively slow shear flow; moreover, the relaxation became considerably slower, with dramatic increases in stress persistence following short periods of faster flow (Fig. 1).

We suggest that this is consistent with flow-induced changes in chain conformation leading to entropically-driven de-solvation. Somewhat surprisingly, this hypothesis also provides a link between flow-induced gelation and similar changes initiated by heating or freezing, as we shall discuss.

[1] Vollrath F, Porter D, Holland C. The science of silks. MRS Bull 2013;38:73–80
[2] Vollrath F, Porter D, Holland C. There are many more lessons still to be learned from spider silks. Soft Matter 2011;7
[3] Holland C, Vollrath F, Ryan AJ, Mykhaylyk OO. Silk and Synthetic Polymers: Reconciling 100 Degrees of Separation. Adv Mater 2012;24:105–9
[4] Holland CA, Terry AE, Porter D, Vollrath F. Comparing the rheology of native spider and silkworm spinning dope. Nat Mater 2006;5:870–4
[5] Laity PR, Gilks SE, Holland C. Rheological behaviour of native silk feedstocks. Polymer 2015;67:28–39