

Stretchy spider silks can be springs or rubber

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It's stronger than steel and nylon, and more extensible than Kevlar. So what is this super-tough material? Spider silk; and learning how to spin it is one of the materials industries' Holy Grails. John Gosline has been fascinated by spider silks and their remarkable toughness for most of his scientific career.

He explains that if we're to learn how to manufacture spider silk, we have to understand the relationship between the components and the spun fibre's mechanical properties; which is why he is focusing on major ampullate silk, one of the many silks that spiders spin.

According to Gosline, spiders use major ampullate silk for draglines and to build the frame and radial structures in webs, all of which have to deform and absorb enormous amounts of energy without fracturing. Comparing the amino acid sequences of major ampullate silk proteins from *Araneus diadematus* and *Nephila clavipes*, Gosline realised that the sequences differed on one count; *Araneus* silk is relatively rich in the amino acid proline, while proline levels in *Nephila* silk are very low.

Curious to know how the presence of proline affects the silks, Gosline and his student, Ken Savage, set about comparing the silks' mechanical properties to find out how the amino acid affects spider silk toughness.

However, obtaining consistent spider silk samples is a problem. Gosline explains that spiders adjust the way they manufacture their silks depending on their circumstances, so he and Savage left the spiders roaming free so that the strands of dragline silk that they dropped were

as uniform as possible. Having established a reliable silk supply, Savage set about testing the silks' mechanical properties. Gently stretching the dry silk while measuring the force on it, the team quickly realised that the silks behaved almost identically; the presence of proline had little or no effect on dry silk.

However, when Savage began investigating the hydrated silk it was a completely different story. For a start, the wet *Araneus* silk shrank and swelled much more than the proline deficient *Nephila* silk. Savage also tested the silk's stiffness, and found that the *Nephila* silk was almost ten times stiffer than the *Araneus* silk. Finally, knowing that regions of the silk proteins stack to form microscopic crystals in a fibre, Savage measured the fibre's birefringence to see how the two silks compared and if the organisation of the proteins in the silk fibre changed when they were damp. The proteins in the *Nephila* silk were always more organised than the proteins in the *Araneus* silk, regardless of whether they were wet or dry. And as Savage stretched the silks, the degree of organisation in the hydrated *Nephila* silk increased much more than the *Araneus* silk.

Gosline realised that the different mechanical properties could be accounted for by the silk proteins' amino acid composition. According to Gosline, proline amino acids are famed for breaking up the organised three-dimensional structures that protein chains fold into, so protein structures with high proline content would be poorly organised in comparison to proteins with little or no proline. *Araneus* silk contains 16% proline, found mostly in linker regions between the protein's crystalline structures, which would make the linkers flexible and randomly arranged. Gosline realised that if this was the case, the hydrated silk might behave like an elastic band.

Nephila silk, on the other hand, has a very low proline content in the linker regions, allowing the linkers to form a relatively well organised

crystalline structure and behave more like a stiff spring. Gosline and Savage decided to investigate both silks' stretchiness to see if they were more rubber-like or spring-like.

Stretching samples of the hydrated silks, Savage gently raised and lowered the temperature from 30 to 10°C while carefully measuring the minute forces required to maintain the extension. For Nephila silk the force remained essentially constant as the temperature changed, a clear indication of spring-like elasticity. However, for the proline-rich Araneus silk the force varied in direct proportion to the temperature, behaving like a rubber-band. So proline-rich spider silks extend like floppy rubber bands, while spider silks with low proline levels behave more like rigid springs.

Having found that proline amino acids have a dramatic effect on the mechanical behaviour of hydrated spider silks, Gosline and Savage are keen to find out why the behaviour of the dry silks is almost indistinguishable and what the functional significance is of the different proline contents.

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