Toughness-enhancing metastructure in the recluse spider’s looped ribbon silk†

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In nature’s toughest materials, sacrificial bonds and hidden lengths play a key role in dissipating energy. Here, we show that the recluse spider (Loxosceles genus) spins its 50 nm-thin silk ribbons into sacrificial micro-loops, providing inspiration for the design of toughened uniaxial metamaterials. Previous attempts to incorporate sacrificial metastructure in cylindrical fibers have either failed to enhance toughness or required prohibitively complex manufacturing. In the recluse system, the loops of the ribbon-like strand are anchored by silk-to-silk bonds that do not compromise the fiber’s ultimate strength upon release and require no additional adhesive. The silk’s thin ribbon morphology facilitates the formation of these strong yet sacrificial bonds and reduces the risk of failure due to bending. Modeling and an experimental proof of concept are used to demonstrate that a looped ribbon metastructure can considerably enhance a material’s toughness.

Sacrificial bonds and hidden lengths are key elements of nature’s toughest materials. In molecules like titin1 and spidroin,2,3 substantial energy is dissipated through work against entropic forces after the breaking of weak bonds releases hidden length.4,5 Hidden length also plays an important role at higher hierarchical levels in materials like nacre,5 bone,6 and spider webs,7–10 where the arrangement of diverse components into sacrificial elements results in outstanding combinations of stiffness, strength, and toughness.11

Recent efforts to implement hidden length in uniaxial fibres for enhanced toughness have shown promising results,12–14 yet significant challenges remain. Pugno et al. introduced knots into synthetic fibers14 and silkworm silk12 to yield substantial toughness gains, and Passieux et al. found that extruding a synthetic polymer dye into a meandering path yielded a tougher strand.13 However, incorporating the necessary slipknots into fibres requires a complicated fabrication process,12,14 and when Passieux et al. fabricated looped fibres, they displayed decreased strength that undermined any potential toughness gains.13 Premature failure was unavoidable in these looped fibres due to cusps and defects that formed as the loops unraveled.13

Here, we show that the recluse genus of spiders (Loxosceles) spins its ribbon silk into a looped metastructure that can enhance a fibre’s toughness. Loxosceles has been known to produce a uniquely flattened silk strand that is 7–10 μm wide and only 50–70 nm thick, with the spider’s web previously described as a disorganized sheet or cobweb.15–17 However, a close investigation of the recluse web and spinning process revealed that Loxosceles uses an intricate motion of its specialized spinnerets to fashion the extruded silk ribbons into serial loops, with each loop held in place by a silk-to-silk self-adhesive junction (Fig. 1a–d). The loop junctions act as sacrificial bonds, as they can open above a certain tensile force without rupturing the ribbon. The loops also appear necessary to stabilize the silk’s...
S3, ESI† spinning mechanism to produce its silk (Fig. 1k–n and Videos S2, 0). The anterior lateral spinnerets (ALS, green), posterior median spinnerets (PMS, magenta), posterior lateral spinnerets (PLS, blue), and colulus (C, a vestigial structure). (f) Flattened major ampullate spigot. (g) Posterior spinnerets poised to interweave. (h and i) PLS plate-like seta. (j) PMS tapered seta. (k–n) High-speed video frames of the Loxosceles spinning motion and (k′–n′) accompanying schematics, with only the spider’s right ALS active (Video S3, ESI†). The time stamp of each stage is shown in the top-right corner, with the time required to complete the displayed spinning stroke indicated in parentheses in (k).

ribbon-like morphology; when forcibly extracted into straight strands, the silk collapsed into narrow, rolled cylinders (Fig. 1d). Spun naturally, the looped silk accumulates into bales that the spider deposits as it travels its lair, forming a disorganized web of silk clumps strung between extended supporting lines (Fig. 1a, b and Video S1, ESI†).

Loxosceles employs a complex and, to our knowledge, unique spinning mechanism to produce its silk (Fig. 1k–n and Videos S2, S3, ESI†). High-speed video revealed that two looped ribbons are simultaneously produced by a coordinated motion of three spinneret pairs: the anterior lateral spinnerets (ALSs), posterior median spinnerets (PMSs), and posterior lateral spinnerets (PLSs) (Fig. 1e and e′). Loops are formed by a sewing motion of each ALS coupled with a clamping motion of the same-side PLS and opposite-side PMS. On each stroke, a length of silk is first extruded (Fig. 1k and k′) from the flattened major ampullate spigot found at the apex of each ALS (Fig. 1f).15,16,18 The ALS then pivots to meet the posterior spinnerets, with the same-side PMS holding the strand in place (Fig. 1l and l′). The resulting loop is clamped by an interweaving of the same-side PLS and opposite-side PMS (Fig. 1g, m and m′). The setae (hairs) covering these posterior spinnerets are notable for their distinctive shape and surface morphology, which appear to facilitate fibre clamping. The plate-like PLS setae (Fig. 1h and i) and tapered PMS setae (Fig. 1j) interweave (Video S3, ESI†), seemingly to encourage ribbon-to-ribbon bonding. Also, all setae feature distally directed nodules tapering to 100–150 nm at their ends (Fig. 1i and j). We suggest these nodules facilitate a secure clamp by preventing slippage towards a seta’s base, and they enable a smooth release by minimizing adhesion onto the rough, nodular surface.19 Finally, the anterior spinneret performs its upstroke, the posterior spinnerets execute a slight posterior shift to make room for another loop, and the same-side PMS releases its hold (Fig. 1n and n′). The spinning process repeats at 10–15 Hz, with the two ALSs oscillating at a 180 degree phase difference to produce two looped strands.

To assess the mechanical behaviour of the Loxosceles silk metastructure, we conducted tensile tests of looped and non-looped strands (Fig. 2). In agreement with past studies of uniaxial fibres with hidden length stored via sacrificial bonding,1,4 stress peaks of sizeable height and width were observed in looped stress–strain curves (Fig. 2a). The termination of each peak reflects a loop opening event (asterisk, Fig. 2a), which releases

![Fig. 1](image1.png)

**Fig. 1** Loxosceles silk loops and spinning mechanism. (a) A restrained Loxosceles specimen spins a bale of looped silk (inset). (b) Optical microscopy image of a looped strand. (c) SEM image of a loop junction. (d) SEM image of forcibly extracted, rolled-up Loxosceles ribbon. (e) False-coloured SEM and (e′) accompanying schematic of the Loxosceles spinnerets, showing anterior lateral spinnerets (ALS, green), posterior median spinnerets (PMS, magenta), posterior lateral spinnerets (PLS, blue), and colulus (C, a vestigial structure). (f) Flattened major ampullate spigot. (g) Posterior spinnerets poised to interweave. (h and i) PLS plate-like seta. (j) PMS tapered seta. (k–n) High-speed video frames of the Loxosceles spinning motion and (k′–n′) accompanying schematics, with only the spider’s right ALS active (Video S3, ESI†). The time stamp of each stage is shown in the top-right corner, with the time required to complete the displayed spinning stroke indicated in parentheses in (k).

![Fig. 2](image2.png)

**Fig. 2** Loxosceles silk tensile tests. (a) Representative engineering stress–strain curves for non-looped (red) and looped (blue) recluse strands. (a′) Non-looped strand schematic, where \( L_0 \) is the initial length. (a″) Loop length schematic, where \( L_0 \) is the initially loaded length of the strand and \( L_s \) is the length of a single loop. (b) Ultimate strength \( \sigma_u \) and (c) effective toughness \( W \) of recluse silk. Data was collected from 8 individuals, with values averaged from roughly 3 looped and 3 non-looped strands per individual. Left frames (white background): non-looped (red) and looped (blue) paired data for each individual, connected with black lines. Right frames (grey background): difference between looped and non-looped data for each individual (circles), mean difference (horizontal bar), zero difference (red dotted line), and 95% confidence interval (CI, vertical bar). No significant difference in \( \sigma_u \) was detected (\( P = 0.53 \), two-sided t-test), with the entire CI falling within the 25% zone of relative equivalence (b, black dotted lines), while the toughness \( W \) for looped samples was found to be significantly less than for non-looped (\( P < 0.001 \), two-sided t-test) because some loops failed to open.
hidden length into the system and thus relaxes the fibre. Further extension is required to exhaust the released length before stress is again encountered and the next stress peak is initiated. By subjecting the strand to this successive strain and relaxation, i.e. “strain cycling”, an increase of the total tensile energy of the system is possible.

In previous attempts at producing looped fibres with enhanced toughness, potential gains were completely negated by a ≈50% reduction in the tensile strength $\sigma_u$ of looped strands relative to their non-looped equivalents.\(^\dagger\) Since these fibres featured cylindrical profiles, cusps formed after the loops opened, leading to stress concentration and premature failure. In addition, the loop junctions in this system were thermally bonded, inducing defects upon loop opening.

*Loxosceles* silk overcomes the limitations observed in previously reported looped fibres: looped ribbons from eight individuals did not display a significant reduction in strength compared to non-looped silk (Fig. 2b). We suggest the ribbon’s thinness confers a degree of flexibility that prevents the formation of cusps as its loops unravel, while the silk-to-silk adhesion via non-covalent bonds\(^16,17\) allows the loop junctions to release without introducing defects. Notably, the silk loop junctions are of considerable strength relative to the fibre due to the strand’s ribbon morphology: the strand-to-strand contact area for a ribbon is vastly greater than for a cylinder. These advantages over the cylindrical looped fibre system—prevention of cusps, defect-free loop unrolling, and strong loop junction bonds—are made possible by the thin ribbon morphology of *Loxosceles* silk.

To explore the potential of a looped ribbon structure in terms of toughness enhancement, we modelled looped fibres and considered their effective specific toughness $W$, i.e. energy divided by total mass:\(^14\)

$$W = \frac{1}{m} \int_0^{r_{\text{max}}} Fdx = \frac{A L_0}{\rho A (NL + L_o)} \int_0^{r_{\text{max}}} \sigma \, dr = \frac{1}{r \rho} \int_0^{r_{\text{max}}} \sigma \, dr$$

where $F$ is tensile force, $x$ is fibre extension, $A$ is cross-sectional area, $\varepsilon$ is engineering strain, $\sigma$ is engineering stress, $\rho$ is mass density, $L_0$ is the initially loaded strand length (Fig. 2a’ and a’’), $N$ is the number of loops, $L_i$ is the length of a single loop, and $\lambda = (L_0 + NL)/L_o \geq 1$ is the ratio of total strand length to $L_o$. In the case of a non-looped fibre, $L_i = 0$, $\lambda = 1$, and $W = \rho^{-1} \int_0^{r_{\text{max}}} \sigma \, dr$ is simply the area underneath the stress–strain curve divided by the mass density. For a looped fibre, $L_i > 0$, and thus $\lambda > 1$, accounting for the additional mass in the loops. In our models of looped fibres, we compared the effective toughness of a looped strand, $W_{L}$, to that of a non-looped strand, $W_{n}$, by calculating the normalized gain in toughness, $\phi = (W_{L} - W_{n})/W_{n}$. We modelled ideal elastic (Fig. 3a–c) and plastic (Fig. 3d–f) looped fibres (see Supplementary text, ESI\(^\dagger\)), with the plastic fibre assumed to be strain-hardening to reflect the behaviour of many materials,\(^20\) including silk.\(^21\)

Our models show that significant toughness gains are possible if looping does not diminish the fibre strength: a greater than 1000% toughness increase was predicted in some cases (Fig. 3c). Introducing more loops always leads to a greater enhancement, while increasing loop size has less of an effect above a certain value (Fig. 3c and f). In cases where loops are too small to allow the fibre to fully relax after a loop unrolls (Fig. 3b and e), they introduce “pseudo-ductility,” allowing the material to continuously elongate beyond its bulk extensibility. This is akin to the mechanism employed by nacre, in which microstructure confers increased toughness to an initially brittle material.\(^11\) Since the underlying toughening mechanism relies on repetitive strain cycling, an elastic phenomenon, the effect is most pronounced in elastic materials (Fig. 3c). Energy absorption through plastic deformation is not amplified, and thus plastic materials exhibit less impressive relative toughness enhancements (Fig. 3f).

To evaluate the toughness of looped silk, we first measured the total strand length (and thus mass) prior to testing by inspecting the number and size of loops via optical microscopy. Tensile tests revealed that the maximum extensibility of looped strands was much lower than expected (Table S1, ESI\(^\dagger\)), meaning that not all loops opened. Because their mass was still counted, the effective specific toughness of looped strands decreased (Fig. 2c). If only energy added by strain cycling is considered, which discounts the unopened loop weight, we found an average improvement of 21% (Fig. S1, ESI\(^\dagger\)). This demonstrates the toughness enhancements possible in principle with the looped *Loxosceles* silk system—even for a small number of loops. The failure of all loops to open was potentially due to imperfections in our sample preparation or testing parameters; e.g. tensile loading rates and extrusion speeds during forcible silk pulling may not

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have matched those experienced in nature. Importantly, *Loxosceles* silk functions not only to bear tensile loads, but also to capture prey through entanglement and adhesion to its flat, conformable surface. As we showed, the ribbon morphology that enables this adhesive function was observed only when the silk was naturally looped; when forcibly pulled, the silk collapsed into a curled cylinder (Fig. 1d). Considering the role that the looping plays in this additional functionality, it is likely that the looped ribbons are co-optimized for several distinct properties.

To demonstrate that the toughness gains predicted by our models can be realized, we fabricated looped strands of tape inspired by *Loxosceles* silk that successfully released all hidden length before fracture and displayed no decrease in strength after loop unravelling. We chose strapping tape as our proof-of-concept material (Fig. 4a–c) for its elastic behaviour, ribbon morphology, and high resistance to torsional tearing due to its fibrillar composition. When a single loop of normalized size $\alpha \approx 1.5$ was introduced, no significant decrease in strength was detected (Fig. S2a, ESI†) and toughness was significantly increased (Fig. 4c); the mean toughness gain of $30\%$ was in good agreement with the $22\%$ gain predicted by the elastic model. Far greater gains are predicted in systems with more loops (Fig. 3c).

Other mechanisms of enhancing toughness in uniaxial fibres are possible; for instance, in the case of knotted fibres studied by Pugno et al.,$^{12,14}$ energy is dissipated via friction. We propose another metastructure for increasing toughness in a ribbon: a self-adhering fold (Fig. 4d–f). In this case, work is performed by progressively separating the two adhered interfaces, effectively investing the surface energy of these materials. Folded masking tape showed a pronounced toughness increase: a single fold of size $\alpha \approx 0.5$ yielded a slight reduction in strength (Fig. S2b, ESI†) but $251\%$ enhancement in mean toughness (Fig. 4f).

Notably, the energy dissipation mechanism in a looped ribbon is distinctly different from that employed in either a knotted or folded fibre. In a looped ribbon, the toughness enhancement is not due to friction or adhesion. While strong adhesion is required to achieve high loop opening forces, the associated energies are negligibly small due to the relatively small contact area of the loop joints. In a manner similar to what is observed in a sacrificially bonded molecule,$^4$ the additional energy dissipation is due to repeated straining of the fibre—a mechanism that can significantly enhance toughness, even for materials already featuring outstanding structural properties.

**Conclusions**

In summary, the recluse spider uses an extraordinary spinneret choreography to spin its silk into loops at a rate of 10–15 loops per second. The resulting looped strand owes its functionality to the silk’s ribbon morphology, which (a) retains the strand’s ultimate strength by avoiding the formation of cusps and corresponding stress concentrations during extension, and (b) facilitates the formation of strong sacrificial bonds at the ribbon-to-ribbon contact area that do not produce defects upon bond release. The sacrificial loop junctions and hidden loop length effectively introduce pseudo-ductility into the fibre’s tensile response. We also showed through modelling and a proof of concept that the inclusion of hidden length via sacrificially bonded loops can increase toughness and extensibility of an elastic filamentous material with negligible inherent ductility. By tuning the number and size of loops, the strength of the sacrificial loop bond, and the cross-section of the ribbon, tensile properties in such looped 1D macroscopic metamaterials can be tailored over a wide range and transcend the bulk properties of the fibre material. Thus, the recluse’s unique spinning process and resulting looped ribbon inspire the design of uniaxial metamaterials with tunable and predictable tensile behaviour and superior toughness.

**Experimental methods**

**Spider care & silk extraction**

Chilean recluse spider (*Loxosceles laeta*) specimens were housed individually in cylindrical capsules and fed a weekly diet of crickets. Individuals were randomly chosen for silk sampling; if the individual produced silk during the reeling process, it was included in the study. Silk for tensile testing was collected by anesthetizing a spider with CO$_2$, restraining it with needles and cotton strips, and waiting for the spider to resuscitate. Occasionally, the spider revived but did not spin silk, but in most cases, the spider’s spinnerets became active about one minute after anesthetization. If silk was produced, it was teased from the spinnerets using a needle, deposited onto a mandrel with spaced collection bars, and reeled at $3 \text{ mm s}^{-1}$, a speed that allowed the spider to form loops; this silk was designated “looped.” Once a sufficient amount of looped silk was collected,
the reeling speed was increased to 10 mm s$^{-1}$, a standard reeling speed$^{22}$ that was sufficiently fast to prevent the formation of loops. The resulting straightened strands were designated “non-looped.”

**Optical and electron microscopy**

Optical imagery of looped silk (Fig. 1b) was captured with an IX71 inverted optical microscope (Olympus). SEM images of silk (Fig. 1c and d) were acquired by applying silk to carbon tape, coating with AuPd, and imaging at 5 kV using a S4700 SEM (Hitachi). SEM images of *Loxosceles* spinnerets and setae (Fig. 1e–j) were acquired by dehydrating an adult female *Loxosceles* in 70% ethanol, critically point drying with a PVT-3B (Sandri), coating with AuPd, and imaging at 5 kV using a S4700 SEM (Hitachi). False-colouring of SEM images was conducted using Gimp (www.gimp.org).

**Video of spinning mechanism**

Natural spinning behaviour was captured by opening a spider’s capsule and filming at 60 fps with a Canon DSLR camera (Video S1, ESI†). High-speed video of restrained spinning was filmed at 1000 fps using a v1610 high-speed camera (Phantom) affixed to a SMZ 800 stereo microscope (Nikon). Spinneret activity was captured by anesthetizing and restraining a *Loxosceles* specimen with CO$_2$, then filming as the specimen revived. The spinning process was captured from an angled view with all spinnerets active (Video S2, ESI†) and with only the right ALS and associated posterior spinnerets active (Video S3, ESI†). False-colouring and high-pass filtering of high speed video images (Fig. 1k–n) was conducted using Gimp.

**Silk tensile testing**

Silk samples from 8 spiders were tested, with approximately 3 looped and 3 non-looped strands sampled and averaged for each individual to account for natural sample variability and testing inconsistencies.$^{23}$ Some variation in sampling per individual occurred due to sample loss during handling and testing (Table S1, ESI†). To conduct tensile testing of silk samples, each strand was first applied across a 5 mm gap in a card stock “C”-frame by bringing the frame into contact with the suspended silk with a micromanipulator. 24 hour epoxy (ACE) was applied to the silk at the frame edges and the silk was cut away from the mandrel using a heated wire to avoid applying tension. Card stock squares were precisely applied to each epoxy drop to ensure consistent adhesion up to the gap edges. Each sample was inspected using a stereo microscope to determine the exact gap width $L_0$ and, in the case of looped samples, to count the number of loops $N$. Tensile testing was performed using a UT150 tensile tester (Keysight) with a 5 N load cell. After each frame was clamped in place, the reinforcing edge of the frame was cut away to leave the silk freely suspended. If required, the arms of the tensile tester were laterally adjusted to ensure proper vertical alignment of the strand. Samples were tested at 1 mm min$^{-1}$. Statistical details, silk cross-sectional area measurement, and loop length measurement can be found in the ESI.$^\dagger$

**Tape tensile testing**

For tensile tests of looped tape (Fig. 4a–c), we employed heavy-duty strapping tape (Shurtech) with a width of 24.2 mm and thickness of 0.130 mm. This tape features a polypropylene film reinforced with fiberglass fibres and coated on one side with a rubber-based adhesive. We found that reducing the tape’s width to 6–14 mm best facilitated the formation of strong loop junctions. Loop lengths were calculated to be $x = 1.54 \pm 0.19$ from the strain values at the point of reloading after loop opening (Table S2, ESI†). An 810 Material Testing System (MTS) with a 25 kN load cell was used for testing. Strands with a folded morphology (Fig. 4d–f) were fabricated using standard label tape (Fisherbrand) with dimensions $25.4 \text{ mm} \times 0.123 \text{ mm}$; the length hidden in the fold was $x = 0.497 \pm 0.003$ (Table S3, ESI†). Tensile testing on these folded fibres was conducted using a 5848 MicroFester (Instron) with a 1 kN load cell. Initial strand lengths and tape widths were measured using precision calipers (Carrera). Tape thicknesses were tested using an MDC-1™ PJ Digimatic Micrometer (Mitutoyo). Statistical test details can be found in the ESI.$^\dagger$

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**Notes and references**