

## The effect of spinning forces on spider silk properties

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### Summary

**A new forced silking procedure has been developed that allows measurement of the low forces involved in the silking process and, subsequently, retrieval and tensile testing of the samples spun at the measured silking forces. A strong correlation between silking force and tensile behaviour of spider silk has been established. Fibres spun at high silking force – compared with the conventional yield stress – are stiff and show stress–strain curves previously found in forcibly silked fibres. By contrast, fibres spun at low and very low silking forces are more compliant, and their tensile behaviour corresponds to that of fibres naturally spun by the spider or to fibres subjected**

**to maximum supercontraction, respectively. It has also been found that samples retrieved from processes with significant variations in the silking force are largely variable in terms of force–displacement curves, although reproducibility improves if force is re-scaled into stress. Fibres retrieved from processes with constant silking force show similar tensile properties both in terms of force–displacement and stress–strain curves.**

Key words: spider silk, forced silking, silking force, *Argiope trifasciata*, tensile test.

### Introduction

Millions of years of evolution have endowed spider silk fibres, especially silk produced by the major ampullate gland, with a range of properties that are striking when compared with other natural or man-made materials (Kaplan et al., 1991; Elices et al., 2004). These properties are the consequence of a complex microstructure and a sophisticated processing procedure whose details are slowly being uncovered (Kaplan et al., 1994; Vollrath and Knight, 2001; Jin and Kaplan, 2003; Gatesy et al., 2001). In this context, the production of artificial fibres based on natural silks (Lazaris et al., 2002) is an extremely attractive and highly challenging objective that has been recognized as a basic milestone in the emerging field of biomimetics (Elices, 2000).

Tensile tests – in natural as well as in artificial silk fibres – have shown a wide variability, as recognised in some of the earliest works (Work, 1976; Griffiths and Salinatri, 1980). The variability of natural silk has been interpreted in biological terms as a contribution to the capacity of survival of the spider (Madsen et al., 1999), and efforts were made to establish a correlation between silking conditions and the tensile properties. When testing fibres *naturally spun* by spiders it was found that, even after a careful analysis of the web elements to be tested (Work, 1976; Denny, 1976) and with the use of monofilaments instead of the whole thread retrieved from the web (Pérez-Rigueiro et al., 2001), the tensile properties of the silk collected from the web show an intrinsic large variability, even among fibres from a single web. Fibres obtained from the

safety line show similar variability (Garrido et al., 2002a). Fig. 1 illustrates the range of variation of naturally spun (NS) fibres. *Forced silking* allows silk to be collected in significant quantities by a process that shares significant features with the spinning of artificial silk. Forced silking consists of pulling the fibre from the spider's spinneret, usually by winding it on a rotating mandrel (Work and Emerson, 1982). Despite the control exerted on the silking parameters, it has been reported that forcibly silked (FS) fibres also show a significant variability in their tensile properties (Work, 1976; Cunniff et al., 1994; Madsen et al., 1999). Reproducibility has been found to improve significantly if precautions are taken such as re-scaling the force into stress and discarding the samples obtained at the beginning of the reeling process or when the reeling has proceeded for too long (Guinea et al., 2005). FS fibres are usually much stiffer than natural ones, as is shown in Fig. 1.

A number of studies have investigated the influence of the silking parameters on the tensile properties of the silk fibres, considering the silking process as a basic source of the observed variability; spider silk fibres have been reeled at different silking speeds (Guess and Viney, 1998), from anaesthetized specimens (Madsen and Vollrath, 2000) or from specimens with varying degrees of starvation (Madsen et al., 1999). However, the attempts to unravel the processing mechanisms that allow the spider to modify the properties of silk have made only marginal advances.

The possible influence of the silking force on the properties of spider silk has been considered only recently (Ortlepp and Gosline, 2004). It was found that spiders possess a friction brake that allows them to control the tension applied to their silks when drawn and that the forces exerted by the friction brake differ between natural and forced silking. On the basis of these observations, we have developed a procedure that allows us to measure the silking force exerted by the spider during forced silking, and we have analysed the retrieved fibres by the methodology of characterization of the tensile properties published elsewhere (Guinea et al., 2005).

We found that the tensile behaviour of spider silk fibres can, as suggested by Ortlepp and Gosline (2004), be traced back to the silking force during the reeling process, there being a strong correlation between the mean value of the silking force and the intrinsic tensile properties (i.e. stress–strain curves) of FS fibres. Interestingly, forced silking processes that proceeded under low silking force – i.e. loads much lower than the conventional yield stress – have yielded fibres with tensile properties similar either to NS or to maximum supercontracted fibres tested in air (Pérez-Rigueiro et al., 2003). These findings, to the authors' knowledge, are the first report of spider silk fibres obtained by forced silking that are similar to the NS or even to the supercontracted fibres.

## Materials and methods

### *Immobilisation of the spider*

The immobilisation technique has been described elsewhere (Guinea et al., 2005). Briefly, the spider is placed in a self-zipping plastic bag, suited to its size, and a small hole is perforated in the bag, allowing access to the spinnerets. The bag is fixed with pins to a Styrofoam surface, and the stress exerted by the taut bag is enough to immobilise the spider. After immobilisation, the initial length of silk was obtained by making contact on the area of the spinnerets with a piece of adhesive paper and pulling gently. This procedure does not require anaesthetising the spider at any step of the process.

### *Monitored force silking process*

The immobilized spider was placed upside down on the base of an Instron 4411 testing machine (Canton, MA, USA), and the initial length of silk was attached to a load cell HBM Q-11 (Darmstadt, Germany; resolution  $\pm 5$  mg) fixed to the crosshead of the machine (Fig. 2). Forced silking was done by displacing the crosshead of the silking machine at constant speed while simultaneously measuring the silking force. In this research, all forced silking processes proceeded either at  $10 \text{ mm s}^{-1}$  or at  $1 \text{ mm s}^{-1}$ . Approximately 1 m of silk fibre was obtained from each silking process.

No effort was made to separate the initial two-filament fibre (known as a bave) into its constituent monofilaments (known as brins), so all the tests were performed on baves. The force corresponding to the spinning of a given segment of the fibre was obtained from the silking force *vs* time curve. Since the crosshead of the testing machine moved at constant speed, the

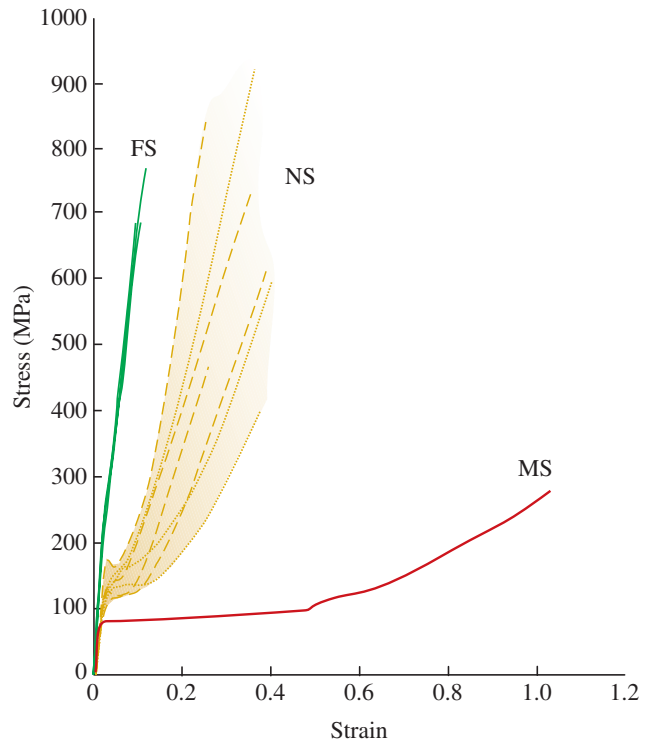


Fig. 1. Typical stress–strain curves of forcibly silked (FS), naturally spun (NS) and maximum supercontracted fibres (MS) tested in air.

time – from the start of silking – at which a given section was spun was calculated as the distance from the fibre tip divided by the crosshead speed. By this method, the spinning force throughout a number of silking processes was determined, as plotted in Figs 3A, 4A, 5A, 6A.

### *Tensile testing*

Samples with a gauge length of 20 mm were glued on cardboard frames (Pérez-Rigueiro et al., 1998). Tensile tests were performed in an Instron 4411 testing machine at a constant crosshead speed and an average strain rate of  $2 \times 10^{-4} \text{ s}^{-1}$ . The load applied to the sample was measured with a balance (AND 1200 G; A&D Instruments, Oxford, UK; resolution  $\pm 10$  mg) attached to the lower end of the sample. The crosshead displacement was taken as a direct measurement of the sample deformation, since the compliance of silk has been estimated as 1000 times greater than that of the equipment. The tests were performed in air under nominal conditions of  $20^\circ\text{C}$  and 40% relative humidity.

### *Geometry characterisation*

Both ends adjacent to the length of the fibre to be tested were secured and retrieved before tensile testing to determine the fibre's cross-sectional area. Samples were sputtered with gold and examined in a JEOL 6300 scanning electron microscope (Tokyo, Japan; observation conditions  $V=10 \text{ kV}$ ,  $I=0.06 \text{ nA}$ ; Pérez-Rigueiro et al., 1998). Brin diameters were measured from each micrograph, and the mean value of the diameters

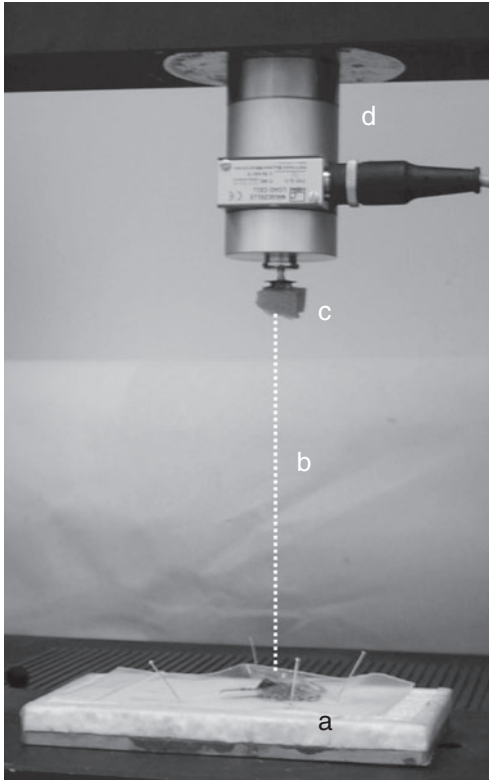


Fig. 2. Experimental set-up of the monitored forced silking process. The immobilized spider (a) is placed upside down, and the tip of the fibre (b) is fixed with a magnet (c) to the load cell (d). The silking process proceeds by displacing the crosshead of the testing machine at constant speed.

corresponding to both ends of a given fibre was used to compute the cross-sectional area of the fibre, assuming a circular cross-section (Pérez-Rigueiro et al., 2001).

This research was done with silk from the orb-web weaving spider, *Argiope trifasciata* Forskål, specifically with fibres spun by the major ampullate gland, which are used by the spider for the dragline and for web frames and radii.

## Results and discussion

### Forced silking at high loads

Fig. 3A shows the measured force during a typical reeling process at constant speed, in this example at  $10 \text{ mm s}^{-1}$ . When the silking force was plotted *versus* the position along the fibre, a region where the silking force remains almost constant –  $\sim 7 \text{ mN}$ , on average – was found. Fig. 3A shows the six samples taken from this region for tensile tests. Fig. 3B shows the results of the six tensile tests. The plots of tensile force *versus* displacement are very repetitive and reproducible. Notice that the average force during silking was higher than the conventional yielding force ( $\sim 5 \text{ mN}$ ), even in sample 5. Fig. 3C shows the tensile tests re-scaled after measuring the fibre diameter. Plots of stress *versus* strain are again repetitive and reproducible. It was found that the fibre diameter was

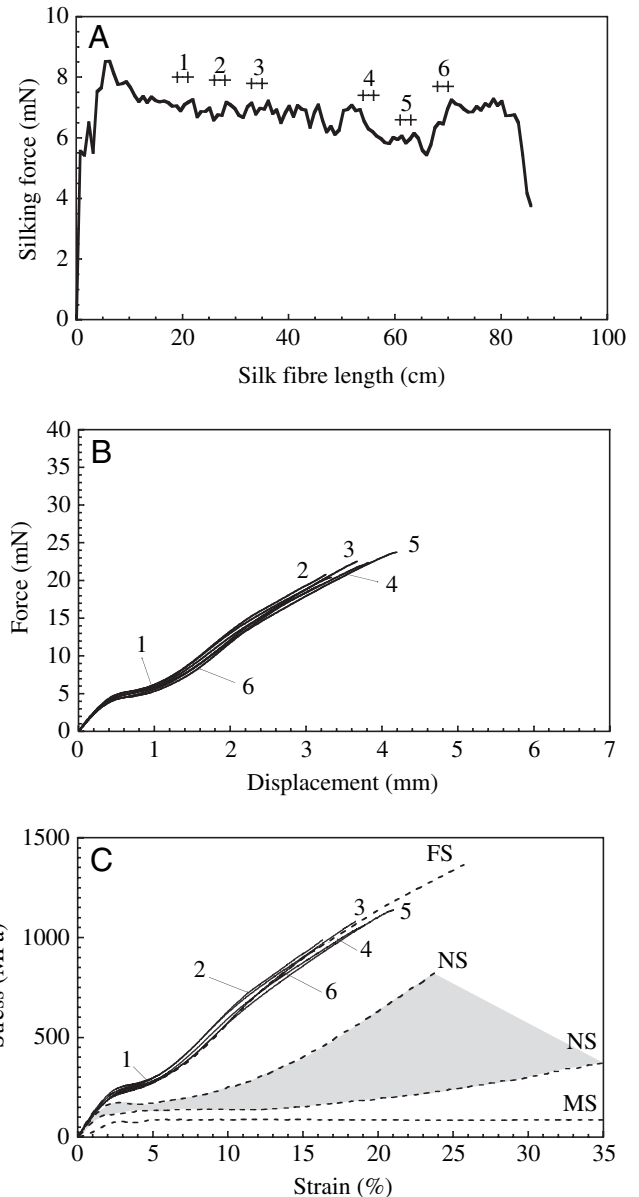


Fig. 3. Silking force and tensile properties of fibres retrieved from a forced silking process characterized by high and constant silking force. (A) Silking force *vs* position along the silk fibre obtained from a silking process at  $10 \text{ mm s}^{-1}$  reeling speed. The numbered intervals identify the samples shown in B and C. (B) Force–displacement curves of the silk samples obtained in the forced silking process shown in A. Base length=20 mm. For A and B, nominal reeling/test conditions were  $T=20^\circ\text{C}$ , relative humidity=40%. (C) Stress–strain curves obtained by re-scaling the force–displacement curves in B using the individual cross-section of each sample. In B and C, samples are identified by a number, indicating the reeling interval in which they were spun, as labelled in A. NS, naturally spun fibres; FS, forcibly silked fibres; MS, maximum supercontracted fibres.

almost constant ( $\sim 3.7 \mu\text{m}$ ). The kind of fibre obtained in this process was of type FS, as shown in Fig. 1.

If the silking force is no longer constant during the reeling process, but the average value is still high, the results remain

similar to the above, although some variability appears. These findings are shown in Fig. 4. Fig. 4A shows a plot of the silking force, which is seen to change substantially in each of the six samples. Here, the reeling speed was  $1 \text{ mm s}^{-1}$ . Tensile tests

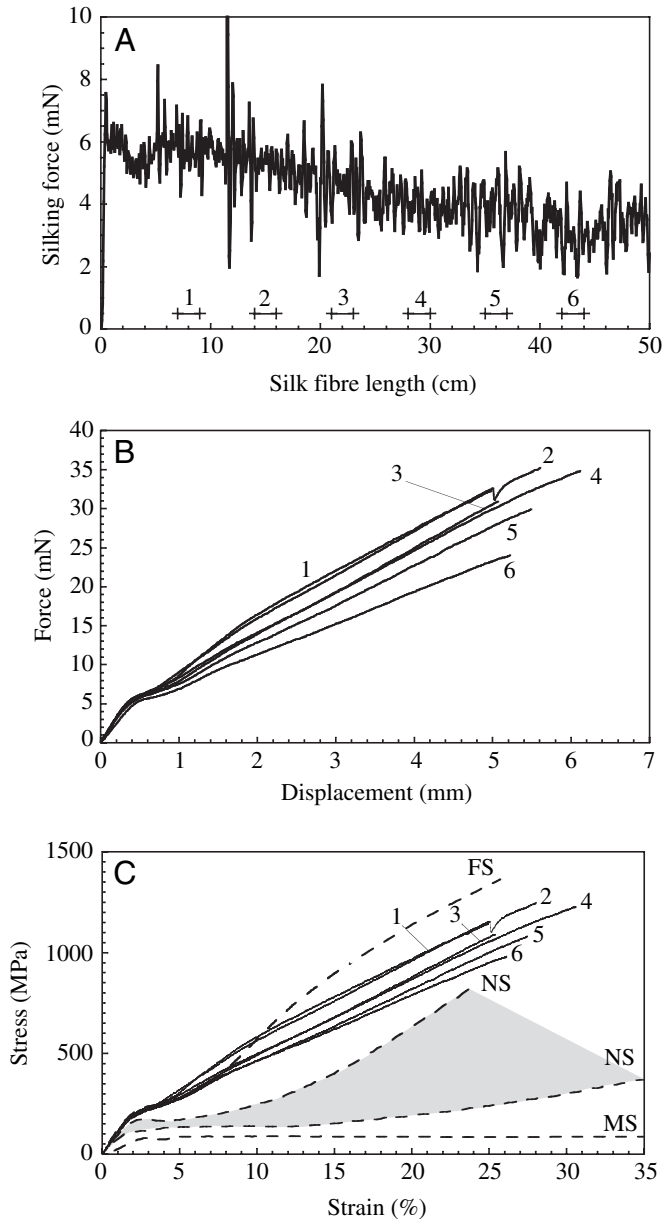


Fig. 4. Silking force and tensile properties of fibres retrieved from a forced silking process characterized by high and variable silking force. (A) Silking force vs position along the silk fibre obtained from a silking process at  $1 \text{ mm s}^{-1}$  reeling speed. The numbered intervals identify the samples shown in B and C. (B) Force–displacement curves of the silk samples obtained in the forced silking process shown in A. Base length=20 mm. For A and B, nominal reeling/test conditions were  $T=20^\circ\text{C}$ , relative humidity=40%. (C) Stress–strain curves obtained by re-scaling the force–displacement curves in B using the individual cross-section of each sample. In B and C, samples are identified by a number, indicating the reeling interval in which they were spun, as labelled in A. NS, naturally spun fibres; FS, forcibly silked fibres; MS, maximum supercontracted fibres.

are shown in Fig. 4B; after the initial region – below the yielding point – dispersion is apparent. This improves somewhat when tests are drawn as stress–strain curves, as seen in Fig. 4C, but some variability remains. Here, the diameters of the samples differ, in contrast to those of samples obtained at constant silking force, even though the fibre is still of type FS (Fig. 1).

#### Forced silking at low loads

The usual reeling processes normally provide silking forces around the yielding limit,  $\sigma_y$ , as shown in Figs 3A and 4A. However, lower forces were measured sporadically; silking forces well below the yielding limit (between  $0.3\sigma_y$  and  $0.1\sigma_y$ ) were measured in 10% of tests performed at a reeling speed of  $1 \text{ mm s}^{-1}$ , as discussed below.

Fig. 5A shows the force measured along the fibre of a forced silking process at low load. Four samples were tensile tested and the results are plotted in Fig. 5B. The silking forces are much lower than the yield limit ( $\sim 3 \text{ mN}$ ). When plotted as stress–strain curves (Fig. 5C), the results come within the domain of fibres naturally spun (NS) by the spider. This is a completely different finding from previously (see Figs 3C, 4C), where fibres were of type FS, were stiffer, had low values of maximum strain and were well outside the realm of NS fibres.

Results obtained with silking forces well below the conventional yielding limit ( $<0.1 \text{ mN}$ ) are shown in Fig. 6A. Three samples were tensile tested; the force–displacement curves are shown in Fig. 6B, and the corresponding stress–strain curves in Fig. 6C. Again, another interesting result was found; at such low silking forces, the curves are much more compliant than the NS fibres, they are below the NS region and are similar to those of the maximum supercontracted fibres (MS) tested in air (Pérez-Rigueiro et al., 2003). As for the data shown in Fig. 3 for almost constant silking forces, the force–displacement curves here were reproducible, a behaviour that improves when stress–strain curves are considered.

#### Modulation of the tensile behaviour of spider silk through the silking forces

Table 1 outlines the results presented above and compares the mean silking force ( $F_s$ ) in each process with the spider's weight ( $W$ ) and the values of yielding force ( $F_y$ ) and breaking force ( $F_u$ ). The absolute values of the silking force span a range of two orders of magnitude ( $F_s=0.06\text{--}6.5 \text{ mN}$ ). Since the absolute values are likely to depend on the spider's size, the results have been re-scaled by the spider's weight to show that a similar range of values is found ( $F_s/W=0.01\text{--}0.96$ ). Classification of the silking process according to the silking force has proceeded by comparing the silking force and the tensile parameters of the fibre. The comparison of the silking force and the breaking force is an intuitive way of describing the stresses to which the fibres are subjected during processing. However, breaking force is affected by large variations (Garrido et al., 2002b) related to the low Weibull modulus of



spider silk [Weibull modulus is a statistical property of fibres (Chou, 1992) that measures the spread of the tensile strength of different samples of a given material; a low Weibull

modulus indicates large spread in the tensile strength]; consequently, the value of the yield force has been favoured as the classification parameter. The intervals spanned by both

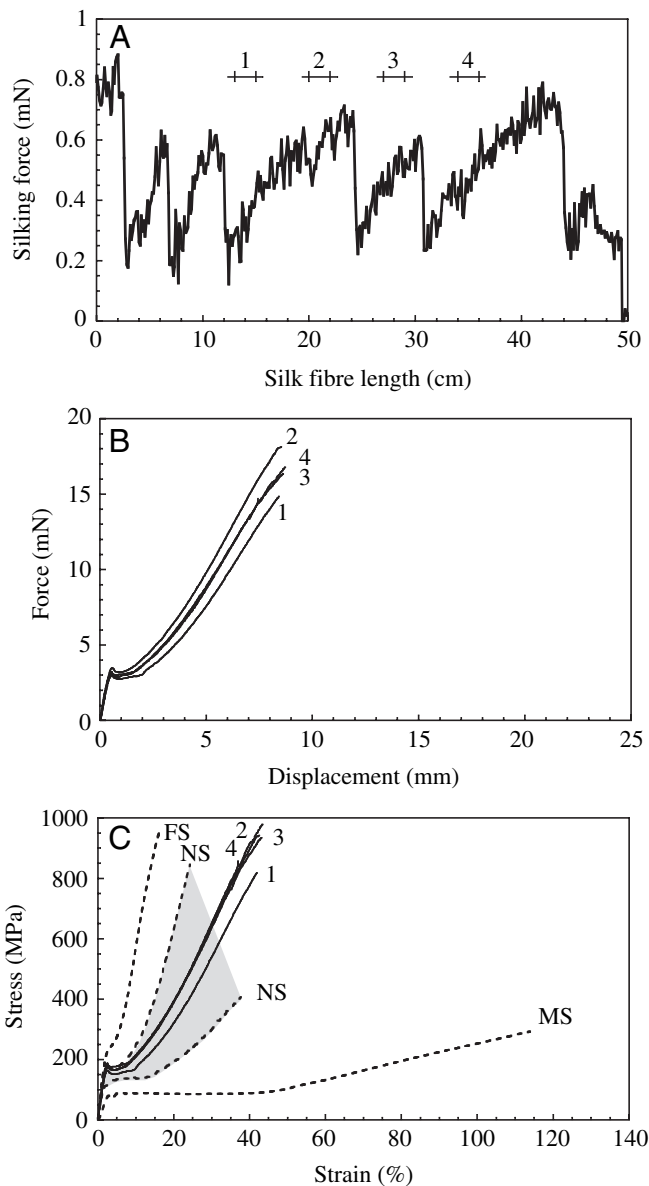


Fig. 5. Silking force and tensile properties of fibres retrieved from a forced silking process characterized by low silking force. (A) Silking force vs position along the silk fibre obtained from a silking process at  $1 \text{ mm s}^{-1}$  reeling speed. The numbered intervals identify the samples shown in B and C. (B) Force–displacement curves of the silk samples obtained in the forced silking process shown in A. Base length=20 mm. For A and B, nominal reeling/test conditions were  $T=20^\circ\text{C}$ , relative humidity=40%. (C) Stress–strain curves obtained by re-scaling the force–displacement curves in B using the individual cross-section of each sample. The upper and lower limits of the range of tensile properties exhibited by naturally spun (NS) fibres, and the stress–strain plots of maximum supercontracted (MS) fibres and typical forcibly silked (FS) fibres are plotted as dotted lines to allow comparison. In B and C, samples are identified by a number, indicating the reeling interval in which they were spun, as labelled in A.

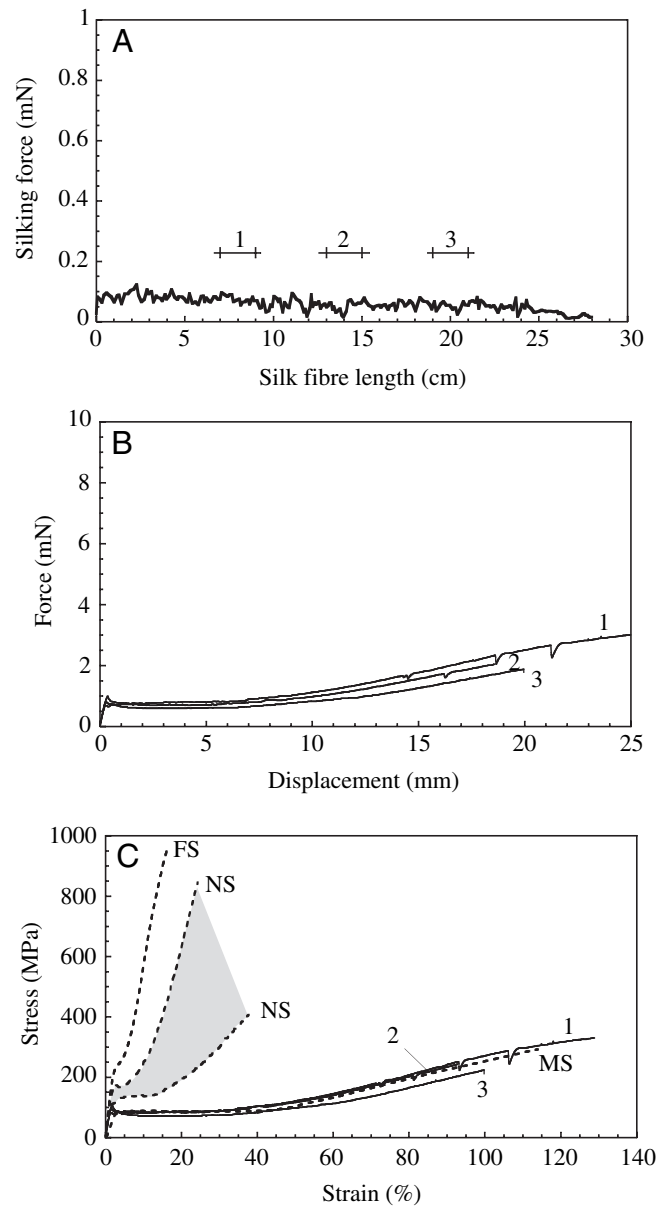


Fig. 6. Silking force and tensile properties of fibres retrieved from a forced silking process characterized by very low silking force. (A) Silking force vs position along the silk fibre obtained from a silking process at  $1 \text{ mm s}^{-1}$  reeling speed. The numbered intervals identify the samples shown in B and C. (B) Force–displacement curves of the silk samples obtained in the forced silking process shown in A. Base length=20 mm. For A and B, nominal reeling/test conditions were  $T=20^\circ\text{C}$ , relative humidity=40%. (C) Stress–strain curves obtained by re-scaling the force–displacement curves in B using the individual cross-section of each sample. The upper and lower limits of the range of tensile properties exhibited by naturally spun (NS) fibres, and the stress–strain plots of maximum supercontracted (MS) fibres and typical forcibly silked (FS) fibres are plotted as dotted lines to allow comparison. In B and C, samples are identified by a number, indicating the reeling interval in which they were spun, as labelled in A.

Table 1. Comparison of the spider's weight and the tensile properties of spider silk fibres with the silking force

Data source	$W$	$F_s$	$F_s/W$	$F_y$	$F_s/F_y$	$F_u$	$F_s/F_u$
Fig. 3	6.7	6.5±0.5	0.96	4.6±0.1	1.47	21.5±0.6	0.32
Fig. 4	5.9	4.5±1.0	0.76	5.7±0.1	0.78	31±2	0.14
Fig. 5	5.6	0.5±0.2	0.09	3.2±0.1	0.15	16.5±0.7	0.03
Fig. 6	4.9	0.06±0.02	0.01	0.84±0.07	0.07	2.3±0.3	0.02

$W$ , spider's weight (mN);  $F_s$ , mean silking force (mN);  $F_y$ , yielding force (mN) measured from the force–displacement curves;  $F_u$ , force at breaking (mN) measured from the force–displacement curves.  $F_s$ ,  $F_y$  and  $F_u$  are expressed as means ± s.d.

ratios,  $F_s/F_y$  and  $F_s/F_u$ , are comparable and narrower than the interval spanned by the  $F_s/W$  parameter.

The results obtained with *A. trifasciata* are consistent with previously published results on garden spider *Araneus diadematus* (Ortlepp and Gosline, 2004). It has been found that spiders during vertical descents can exert forces on the silk fibres even up to  $2.2\times$  the spider's weight. This value is comparable with the silking force measured in the high silking force processes, which lead to fibres with FS properties. At the other extreme of the silking force range, it has been estimated that spiders in free fall spin silk at forces of approximately  $0.1\times$  the spider's weight. These results are consistent with the data presented in Fig. 5, since fibres spun at  $0.09\times$  body weight show the characteristics of NS fibres. Combining the results on free-falling spiders (Ortlepp and Gosline, 2004) with those on undisturbed climbing spiders (Garrido et al., 2002) can account for an intriguing property of the latter. It has been found that fibres spun during undisturbed climbing represent the lower limit of the stress–strain curves retrieved either from the web or from the safety line. The results obtained from free-falling spiders suggest that these fibres could be spun at the lowest silking force available to the spider.

In this context, Fig. 6, which shows fibres with tensile features characteristic of MS fibres, would represent an anomalous behaviour of the spider. The extremely low forces involved in this silking process could be related to changes in the anatomy or physiology of the spinning process as a consequence of the advanced age of the spinning spider (the specimen died a few days after the silking process). However, Fig. 6 indicates that, even under the minimum silking force, the tensile properties of the fibres are within the previously established range of stress–strain curves described for *A. trifasciata* spiders (Pérez-Rigueiro et al., 2003).

The identification of the influence of protein sequence and processing on the final properties of spider silk fibres has prompted an intense debate, driven by the need to determine the essential features of spider silk in order to synthesize artificial silk fibres. The observation that a reduced number of motifs appears conserved in the protein sequence of spiders belonging to widely different groups (Gatesy et al., 2001) suggests that the protein sequence plays an essential role in the properties of spider silk. Our results suggest that, although the basic properties of the silk fibre can be determined by the

protein sequence, its tensile behaviour can be modulated through the silking force in a range that spans maximum supercontracted fibres and forcibly silked fibres and includes the naturally spun fibres produced during web building and as a safety line.

### Conclusions

(1) The forced silking technique described here allows us to measure the silking force in each silk segment along the fibre length and, additionally, retrieve the sample to determine its tensile properties. A correlation between silking force and tensile properties was verified. The experimental procedure is complicated due to the small forces involved in spinning (forces as low as 0.1 mN have been measured) and by the control exerted by the spider on the spinning process, which excludes the possibility of performing a silking process at a constant spinning force.

(2) The silking force greatly influences the type of silk fibre. When the force is high, i.e. around the conventional yield limit, the fibres are stiffer than those naturally spun by spiders. As the load decreases, fibre stiffness decreases, and forcibly silked fibres come to resemble the naturally spun ones. At very low loads, the fibre is even more compliant, and its behaviour is similar to that of the supercontracted fibre, tested in air. In addition, our results open a new and promising scenario for forced silking and its application in the spinning of artificial silk, since this method can be used to obtain all kinds of fibre and not only stiff FS ones.

(3) When tensile tests on samples taken from the same fibre are compared, variability was found to decrease if stress–strain curves are considered instead of force–displacement curves. In this respect, the diameter of fibres reeled at constant force was more homogeneous than that of fibres reeled at variable force.

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