Introduction

Orb-weaving spiders are able to spin up to 7 different types of silks.[1] The aim of this project was to increase our knowledge on the structural properties of silks and in particular to study the formation of silk in-situ. This is analogous to synthetic polymer extrusion [2] but for fibre diameters of a few micrometers and for a living biological object.

Specimen

Most experiments were performed on *Nephila senigalensis* and *Nephila eudulis* spiders, which are reared in the Oxford Zoology department.[3] Complementary experiments were, however, performed on other spider species such as *Cyrtophora*. We generally investigated major ampullate silk extrusion.[1] It was, however, possible to measure in some cases major and minor ampullate silk extrusion from the same specimen.

Methods

Spinning set-up. An in-situ spinning set-up was developed for the experiments.[4-6] (Fig.1)

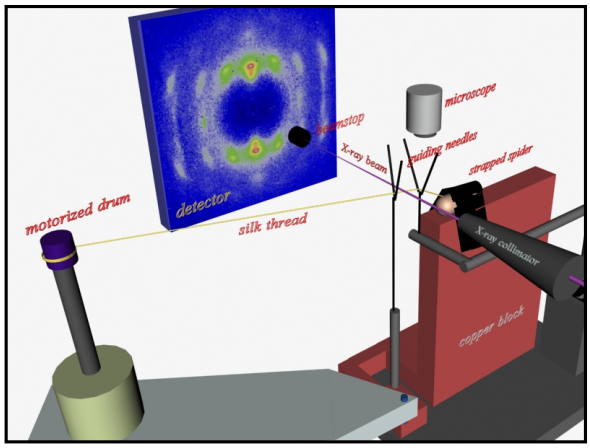

Fig.1 Schematic design of spinning-setup.[6]

The spider was fixed on a copper-support, which could be thermosthatized by a circulating liquid. The spider was initially lightly anesthetized under CO_2 and strapped down to the support structure. This and the subsequent X-ray diffraction experiments left the spiders unharmed. A motor drew the silk fibre (or thread) to a drum. The fibre was guided across two forks to the drum, which allowed stabilizing the fibre for microdiffraction experiments. We found that the fibre was not stable enough in position in the immediate vicinity of the spigots. To cope with this problem we motorized the fork closest to the spigot and placed the fork close to the spigots so that the fibre became taut. It was thus possible to record patterns within 1-2 mm from the spigots exit.

Variation of external parameters during silk extraction.

(i) a motorized drum allowed selecting drawing speeds up to about 100 mm s^{-1}.

(ii) the body temperature was changed between about 13^0C and 38^0C by modulating the temperature of the copper support. The temperature was measured by a thermocouple on the back of the spider.

(iii) the influence of CO_2 on the spinning process was studied by placing a mask over the head of the spider and flushing it with CO_2 gas.

Wide-angle scattering (WAXS). Experiments were usually done with a 10 micron beam at 13 keV using the scanning setup of ID13.[7] The fibres had diameters of about 5 microns or smaller. In order to cope with slight positional drifts we step-scanned the fibre through the beam in vertical direction. This allowed capturing at least one good data point. A 16-bit MAR CCD was used in general in order to cope with the weak signals. In a few cases a faster 12-bit Photonic Science detector was used. Recording times per pattern were between 30 sec and 3 minutes depending on the fibre crystallinity and thickness.
Small-angle scattering (SAXS). The SAXS set-up has been described elsewhere.[8] We performed combined SAXS/WAXS experiments for a sample-to-detector distance of 150-200 mm. This allowed recording peaks up to d≈15 nm.

Selected results
Fibre formation. As shown in Fig.2, the fibre pattern has already formed in the immediate vicinity of the spigots (position 0).[5] It was initially believed that the additional diffuse scattering was due to an increased amount of amorphous material. More recent data suggest, however, that the diffuse ring is rather due to water co-extracted with the solid fibre.[6]

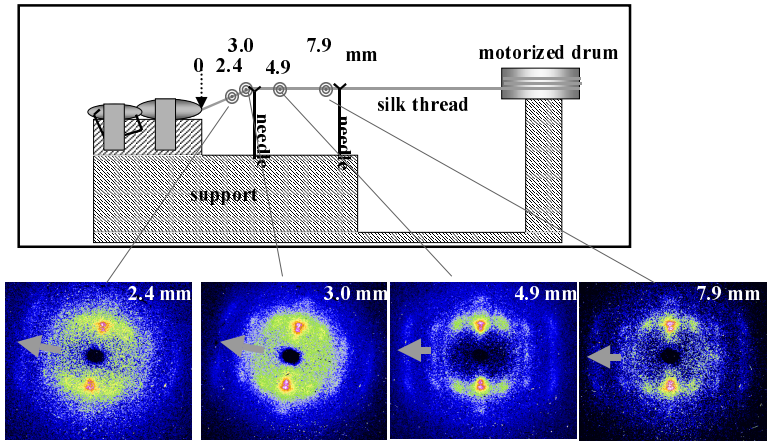

Fig.2 Schematic design of spinning set-up and diffraction patterns recorded at selected distances from the spigots.[5] The positions of the probing beam are depicted by circles.

Post-drawing stage. An increase in extrusion speed improves the alignment of crystalline domains in the fibre. This can be shown by analyzing the variation of Herman's orientation function[9] with drawing speed. (Fig.3)

Post-processing stage. At high extrusion speeds, water is co-extracted with the fibre while the water is resorbed at web-building drawing speeds in the spinning organ.[6,10] (Fig.4) SAXS/WAXS data show that this water enters the amorphous part of the fibre as the scattering contrast of the fibrillar system is modified. The crystalline fraction of the fibrillar

system is, however, not affected as the Bragg peaks are not modified. Water has a plastifying effect on the fibre and reduces its mechanical strength through a breaking of the hydrogen-bonded network.[11] It has therefore been suggested that the tarsal claws act as a post-processing unit during fast extraction of major ampullate silk (dragline formation).[6]

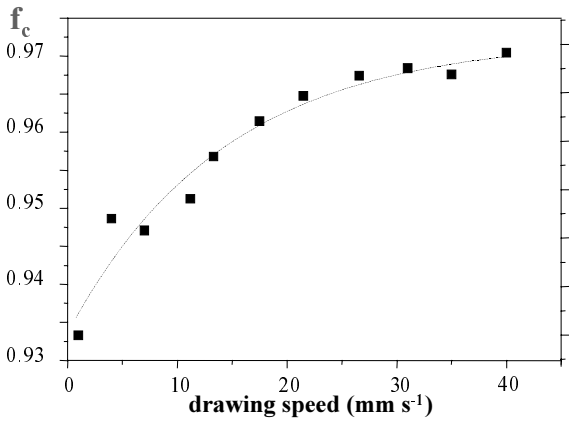

Fig.3 Variation of Herman's orientation function (f_c) with drawing speed.[5]

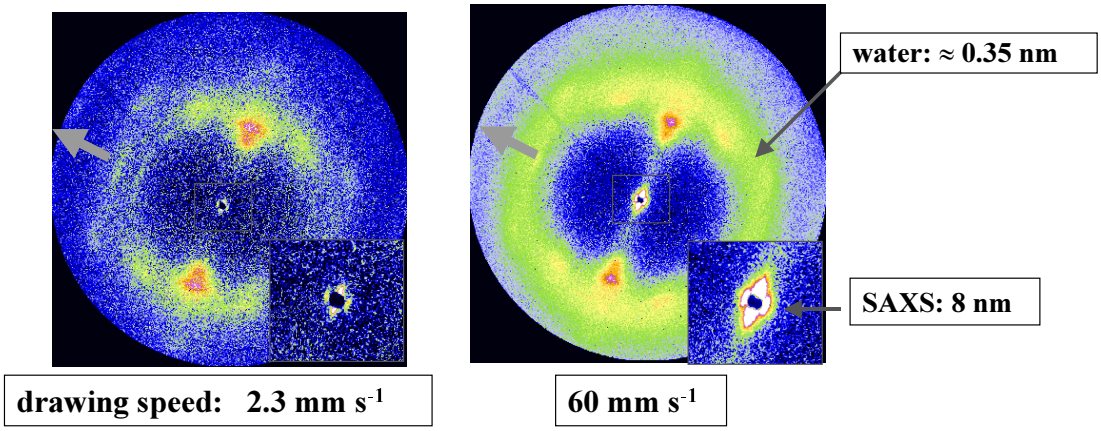

Fig.4 SAXS/WAXS patterns recorded a low (2.3 mms^{-1}) and high (60 mms^{-1}) drawing speeds.[6] The fibre axis is indicated by an arrow. A strong ring due to amorphous water appears at the high drawing speed. The SAXS pattern shows an 8 nm peak due to a semicrystalline fibrillar morphology. The equatorial streak is also related to the fibrillar system.[6,12] The absorption of water by the random polymer network is indicated by the increase in SAXS intensity, which is analogous to cellulose fibres.[13]

References

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