

## Rapid Communication

# Biomimetic Strategies for the Spinning of Artificial Silk Fibers

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**Abstract**

Spider silks possess unrivalled toughness but recombinant spider silk fibers can hardly obtain comparable mechanical properties. While for silkworm silks, the Regenerated Silk Fibers (RSF) can be even more superior than their counterparts. Of various factors, a suitable spinning method is of significance for the fiber behaviors. Wet-spinning and microfluidic approaches are extensively investigated for the fabrication of artificial silk fibers and it becomes a tendency to make a biomimetic design inspired from the natural spinning. In this work, efforts have been spared on the discussion of biomimetic strategies applied in the current spinning process and two potential designs have also been put forward.

**Keywords:** Spider silks; Wet spinning; Microfluidic chip; Biomimetic method; Artificial fibers

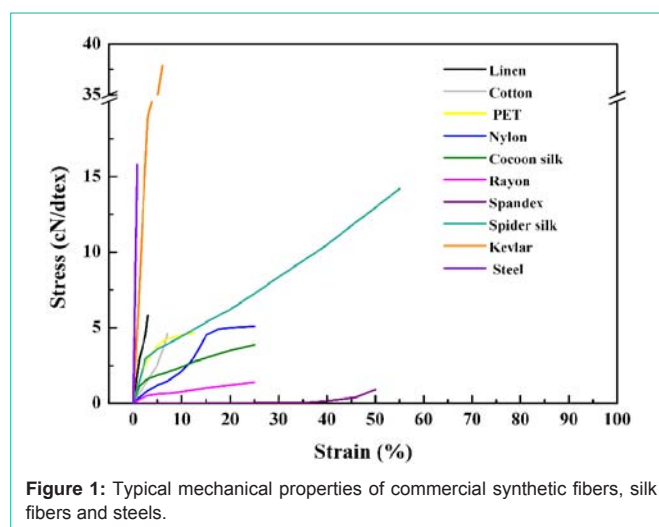
**Introduction**

Impressively, spider silks exhibit remarkable toughness outperforming almost all commercial synthetic fibers (including Kevlar) and even steels (Figure 1). However up to date, it remains a big challenge to fabricate artificial silk fibers with comparable mechanical properties of their counterparts [1]. It is generally recognized that it is most likely to reveal this goal by designing a bio-inspired spinning method to replicate the natural spinning process. Herein, strategies have been provided for the biomimetic wet-spinning and microfluidic technologies.

Even though the current systems are able to produce artificial silk fibres with comparable toughness and extensibility, their strength and stiffness is yet to meet the golden standard of natural fibres. This demands further studies in unveiling the mystery behind natural spinning process to develop a scalable technology that will widen the opportunities of artificial silk fibres in high performance sector.

**Biomimetic Wet-spinning Method**

The quest to produce artificial silk fibers with mechanical properties equivalent to natural silk has increased attention towards bioinspired wet spinning approaches. Madurga et al., [2] demonstrated that straining flow spinning process utilized a capillary with coaxial jet to induce shear forces in the dope solution, which was extruded in a mild acidic dehydrating bath to mimic the low pH spinning duct environment of spiders and silkworms. Recent studies show that the focus of biomimetic spinning involves the production of native-like aqueous spinning dopes for biomimetic wet spinning process. Spider glands store silk proteins at a concentration range from 30–50% (w/v) [3-4]. Andersson et al., [4] synthesized a chimeric recombinant silk protein engineered terminal regions which were aqueous soluble and could be concentrated up to 50%. Subsequently, the highly concentrated dope was used to spin fibre using a mild acidic bath that mimicked the lock and trigger mechanism of native spidroin. Interestingly, the unstretched fibres produced using this



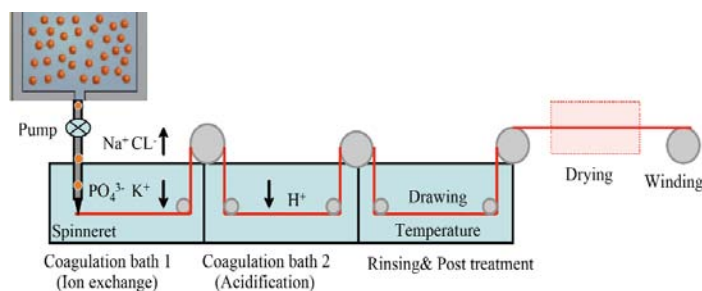
**Figure 1:** Typical mechanical properties of commercial synthetic fibers, silk fibers and steels.

method showed excellent toughness than any other recombinant silk fiber reported to date.

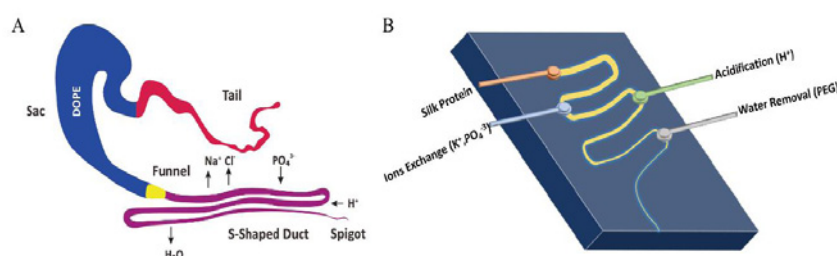
As shown in (Figure 2), a bio-inspired wet-spinning facility is put forward by mimicking the natural spinning process with ions exchange, acidification and elongation. Normally, most current wet-spinning facilities contain one coagulation bath with alcoholic solvent and another bath for post-drawing treatment. Here, totally three baths are provided and ions are added in the same order as the spider silk gland.

**Biomimetic Microfluidic Method**

Both wet-spinning and microfluidic approaches are capable of mimicking the ions exchange and acidification, but the latter method shows greater potential on the generation of elongation and shear stress. Additionally, the microfluidics can be well-designed with a number of dependent silk-gland-like channels, allowing the flows observed and tuned in a controlled manner.



**Figure 2:** Wet spinning apparatus bio-inspired from the natural spinning.



**Figure 3:** (A) Schematic image of the major ampullate gland; (B) microfluidic device biomimicking S-shaped tapered duct of major ampullate gland.

Michelle et al., [5] have reported a microfluidic device inspired by the silkworm silk gland. With this unique channel structure, it is feasible to predict and tune the mechanical properties as well as diameters of the RSF. Furthermore, this advanced technology has also been used to study the molecular transition of fibroins during fiber formation. Similarly, a gradient channel made for a microfluidic chip was designed to mimic the natural spinning apparatus [6]. A bio-inspired microfluidics can also be used as concentrators to regulate the concentrations of aqueous silk solution [7].

To investigate the assembly mechanism of the recombinant spider silk proteins, the effects of pH, ions and the structure of the microfluidic device are discussed to present a model for the silk aggregation and early step of fiber assembly [8]. This microfluidic is capable of changing the ionic and pH conditions by designing different channels. In addition, three types of modules are studied for the specific microfluidic chip to provide various flow behaviors. Thereof, the elongational flow is applied by narrowing the channel width. In good agreement with the model for silk formation, colloidal aggregates are a prerequisite for fiber formation and shear is required to induce the spherical spheres into fibers. Edward et al., [9] has prepared a microfluidic system consisting of a digital, programmable flow control that mimics the spider silk-spinning process. Through this system, artificial spindle-knots and joints are generated, and the water-droplets are observed on the fiber in a way like on the spider silk. Distinguishing from the complicated channels designed in the microfluidic chips, a microfluidic device with dual laminar mobile phases is presented to investigate the spider fibroins with/without the pH-switching N-terminal domain [10].

Microfluidic systems facilitate development of spinning devices that can offer potential imitation of spider silk spinning process. In microfluidic spinning, the flow channel can be designed similar to the tapering duct of the spider's major ampullate gland which

can impart shear and laminar flow as seen in natural silk spinning (Figure 3). Additionally, tapering duct flanked with three channels to mimic natural physicochemical environment with ions exchange, acidification and water removal. S-shaped tapering duct in our design will produce different velocity fields that induce distinct shear and extensional flow field of spidroin. The proposed design is expected to enhance the alignment of protein molecules that can subsequently improve the final fiber properties.

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