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Soft Fibrous Structures in Nature as Liquid Catcher

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ABSTRACT Past decades have witnessed the explosive growth of interest in the field of bio-inspired materials, of which the structures and properties can be well utilized for industrial and bioengineering applications. Among these structures, the natural fibrous structures propose diverse strategies to adapt to their environment, offering inspirations for versatile applications, especially droplet manipulation. With various well-adapted soft structures and materials, these fibrous structures show good control over their interaction with liquids (e.g., water), providing a database full of effective solutions to these droplet-related scientific and technical problems (e.g., colloidal synthesis, single-cell gene sequencing, drug delivery and solution synthesis). In this review, the current achievements in water collection by multiple fibrous structures are highlighted; the structures, basic models, bio-inspired structures and their applications are presented; and the current challenges and future prospects for the development of bio-inspired fibrous structures are discussed.

KEY WORDS Soft fibrous structure, Micro- and nanostructure, Bio-inspired materials, Surface tension

1. Introduction

Hard and soft structural composites in nature have been providing inspirations to material science for decades, yielding an impressive collection of vital progress. Due to their important roles in biological and other engineering systems, soft structural materials have undergone a fast development to keep up with the growing attention and application prospects. Among these interesting fields, droplet manipulation (e.g., generation, transfer and release [1–3]) is of particular interest and great technological importance in biological analysis and chemical engineering [4, 5].

Inspired by nature, researchers developed novel approaches and fabricated advanced artificial structures for droplet transfer and liquid separation. Versatile dynamic wetting behaviors exist in fibrous

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[⋆] Corresponding author. E-mail: fengxu@mail.xjtu.edu.cn systems. For example, in the early morning, dewdrops can be found glistening on spider webs. The unstable dew settled on the threads breaks up spontaneously into droplets [6]. Similar scenes prevail in nature, *e.g.*, the collection of water droplets on conical cactus spines from the air in desert [7], water droplets hanging on the pappi of dandelion seed after the rain or in foggy days [8], etc. The dynamic liquid-moving process on/in fibrous systems is controlled by structural gradient, chemical gradient, elasticity of a single fiber, and so on. Due to the Laplace pressure generated by structural (e.g., droplet curvature radius gradient) or chemical (e.g., inhomogeneous surface energy) gradient, the liquid moves to the low-curvature or more wettable area along the fibers [9]. The bending deformation of the fibers can be balanced by the surface tension of liquids. In a more complex situation, multiple fibers are integrated into clusters or arrays, and the wetting behaviors are affected by the properties of individual fibers as well as the performance of adjacent fibers [10].

In this paper, we begin by introducing three fibrous structures in nature (i.e., spider silk, cactus spine and dandelion seed) and their liquid catching behaviors. Then, we discuss their intriguing mechanisms on droplet motion and liquid catching. In the end, the artificial materials and their typical applications are presented.

2. Three Soft Fibrous Structures in Nature as Liquid Catcher

Jiang's research group first reported the phenomenon that wetted spider silk can directionally collect water based on its special wettability [6]. The multi-level structure (i.e., periodical spindle-knots) observed by optical microscope plays a pivotal role in the water collecting behavior (Fig. 1A(b)). A series of nanofibril puffs on dry hydrophilic silk fibers will shrink into spindles when exposed to humid air, and the nanofibrils are then stretched and the knots are separated from one another, generating the 'joints'. Water droplets gradually condense on the spindle-knots and joints of silk fibers in a random way. It was observed that the micrometer-size drops move to the nearest spindle-knots when reaching the critical size, and coalesce into larger water droplets.

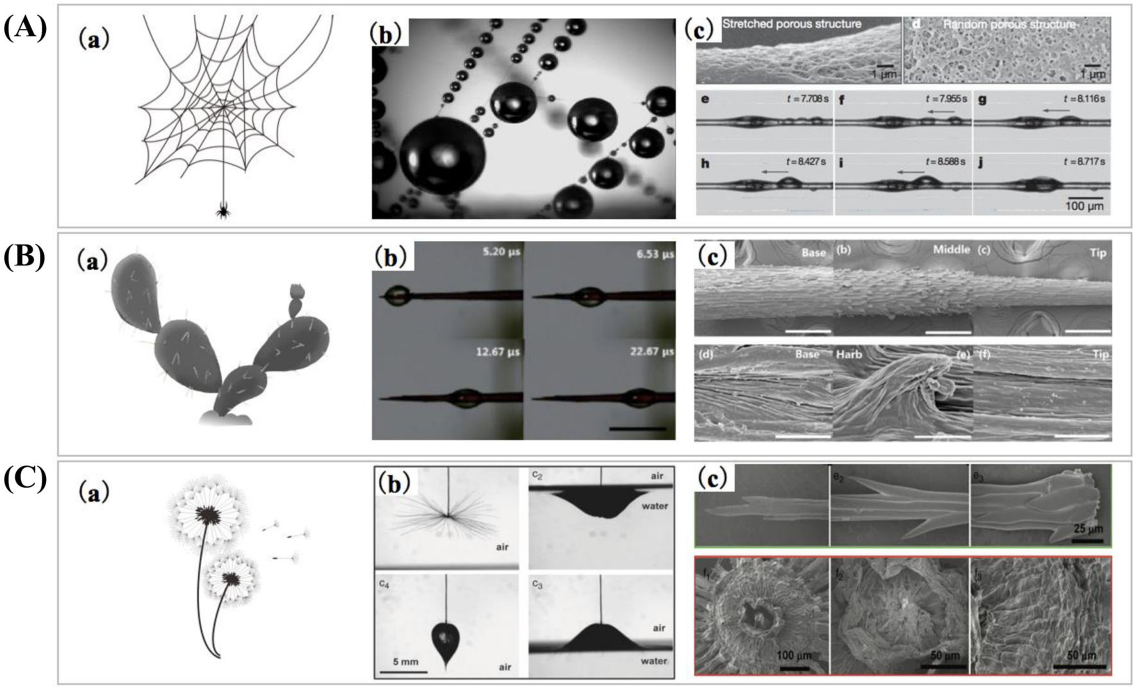

Fig. 1. Natural fiber systems on liquid handling. **A** Spider silk. (a) spider web in nature (scale bar: 100 μm), (b) droplet suspension on spider silk [6], (c) microstructure of spider silk [11]. **B** (a) cactus, (b) droplet on cactus spines (scale bar: 100 μm), (c) microstructure of conical cactus spine [12]. **C** (a) dandelion, (b) catching water behavior (scale bar: 5 mm), (c) microstructure of dandelion seed [8]

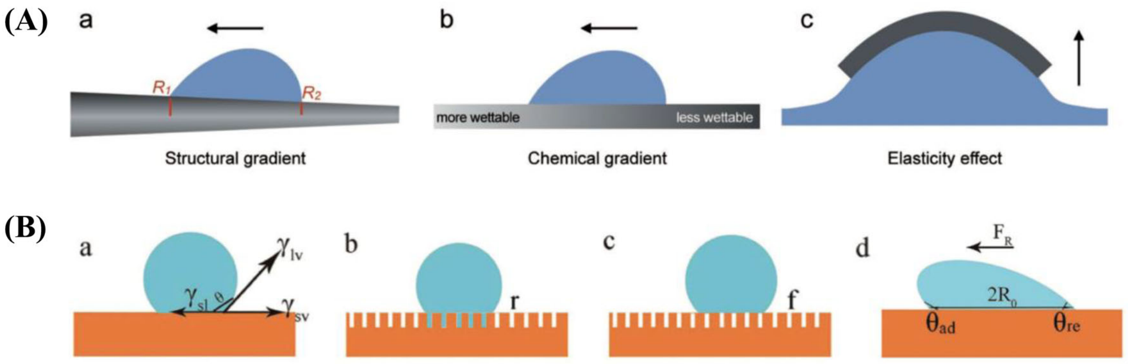

Fig. 2. Driving forces and wetting models of a droplet on solid surface. **A** Driving forces [17]. (a) structural gradient (e.g., conical shape), (b) chemical gradient and (c) elasticity. **B** Wetting models [18]: (a) Young's model, (b) Wenzel's model, (c) Cassie's model and (d) contact angle hysteresis

Similarly, the cacti in desert also have a very efficient method to catch water from air with their conical spines (Fig. 1B(b)), which have functionally equivalent structures as spider silk [7, 12]. The clusters of conical spines with gradient grooves are well distributed on the surface of the cactus (Fig. 1B(c)), helping to generate surface free energy gradient and Laplace pressure difference to facilitate water collection. There are many other creatures in nature having similar conical or needle structures that can also catch water, such as Cynodon dactylon [13], the trichome surface of Sarracenia [14] and scorpion setae [15].

In addition to the above-mentioned single-fiber structures, one dandelion seed contains ∼ 90 white feathery pappi (Fig. 1C(a)), which are also known to be capable of collecting rain droplets [8]. Highly efficient liquid transfer has been observed in such open fibrous systems capable of capturing and holding water steadily (Fig. 1C(b)). Unlike spider silk or conical cactus spines utilizing characteristic structures on a single fiber, dandelion seed achieves liquid collection capability by the fibrous array distributed with a large open angle. The large elastic deformation of the fibers ensures a large space for liquid storage. This is a more controllable and proactive approach for liquid capture compared with those based on liquid condensation from the humid environment or droplet movement induced by the pressure gradient of the surface.

3. Basic Models

The water collection mechanism is based on the wetting performance, which is determined by the contact angle. Many models have been proposed to determine the contact angle, such as Young's contact model, Wenzel's model, Cassie's model, the surface energy model and the Laplace pressure gradient model (Fig. 2A). The Young's contact angle for a physically smooth and chemically homogeneous surface is

$$\gamma_{\mathrm{sg}} - \gamma_{\mathrm{sl}} = \gamma_{\mathrm{lg}} \cos\theta \tag{1}$$

where γ_{sg}, γ_{sl} and γ_{lg} are surface tensions between different phases, respectively (s presents solid, l presents liquid and g presents gas); and θ is Young's contact angle (Fig. 2B(a)).

Wenzel's model and Cassie's model further consider the effects of roughness and heterogeneity (Fig. 2B). Wenzel's model describes the case that the liquid completely penetrates into the roughness grooves of the surface, which is

$$\cos\theta^{*} = r\cos\theta \tag{2}$$

where r is the roughness of the surface and θ^{*} is the contact angle on the rough surface.

While Cassie's model describes the case that liquid droplets rest on the roughness grooves of the surface with air trapped underneath, which is

$$\cos\theta^{*} = f_s\left(\cos\theta + 1\right) - 1 \tag{3}$$

where f_{s} is the ratio of the liquid–solid contact area to the entire contact area.

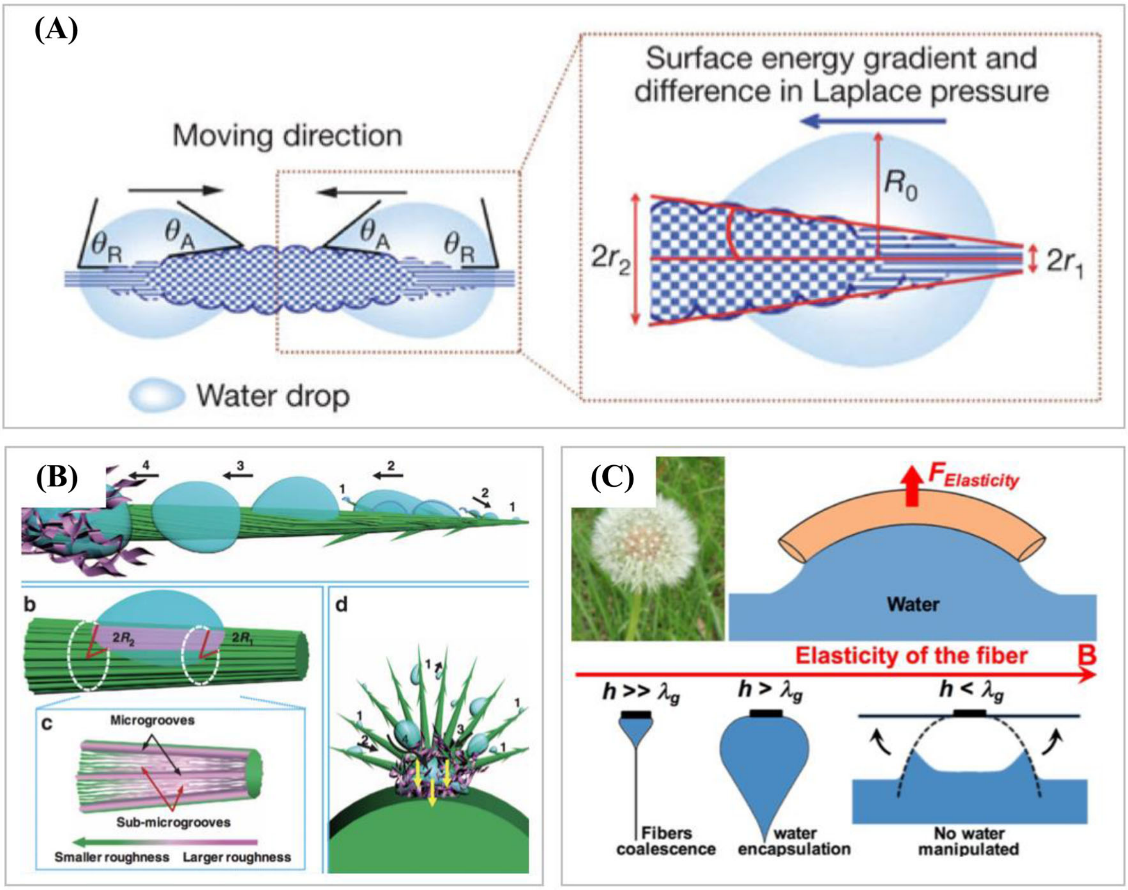

Fig. 3. Mechanical models. **A** Spider silk [6]. **B** Conical cactus spine [30]. **C** Dandelion seed [8]

The droplet tends to move to the zone with lower surface energy, that is, more wettable surface. The surface energy gradient causes difference in Laplace pressure, which could also lead to the movement of droplet toward the zone with lower curvature. The Laplace pressure is:

$$\Delta P = \frac{2\gamma_{\mathrm{lg}}}{R} \tag{4}$$

where R is the radius of the surface.

The water collection mechanisms of spider silk and cactus spines are similar. The surface energy gradient or Laplace pressure difference is generated on such a surface, and a drop can move due to contact angle hysteresis. For spider silk, the Laplace pressure difference and surface free energy gradient between knots and joints are mainly caused by the periodic spindle-knot structure on spider silk [6]. The random distribution of nanofibrils on spindle-knot results in better hydrophilicity of the spindle-knot than of the joints with aligned nanofibrils. The spindle-knot thus has a higher surface energy and drives water toward itself due to the surface energy gradient. This force caused by the difference of surface roughness can be expressed as [16]:

$$F = \int_{L_j}^{L_k} \gamma_{\mathrm{lg}} \left(\cos\theta_A - \cos\theta_R\right) \mathrm{d}l \tag{5}$$

where θ_A and θ_R are advancing and receding contact angle, respectively; L_j and L_k are the lengths of the joint and the spindle-knot, respectively. On the other hand, the geometric gradient could accelerate the collection process by deforming droplets and generating forces (Fig. 2A). The curvature difference between the spindle-knot and the joint will generate a Laplace pressure as [9]:

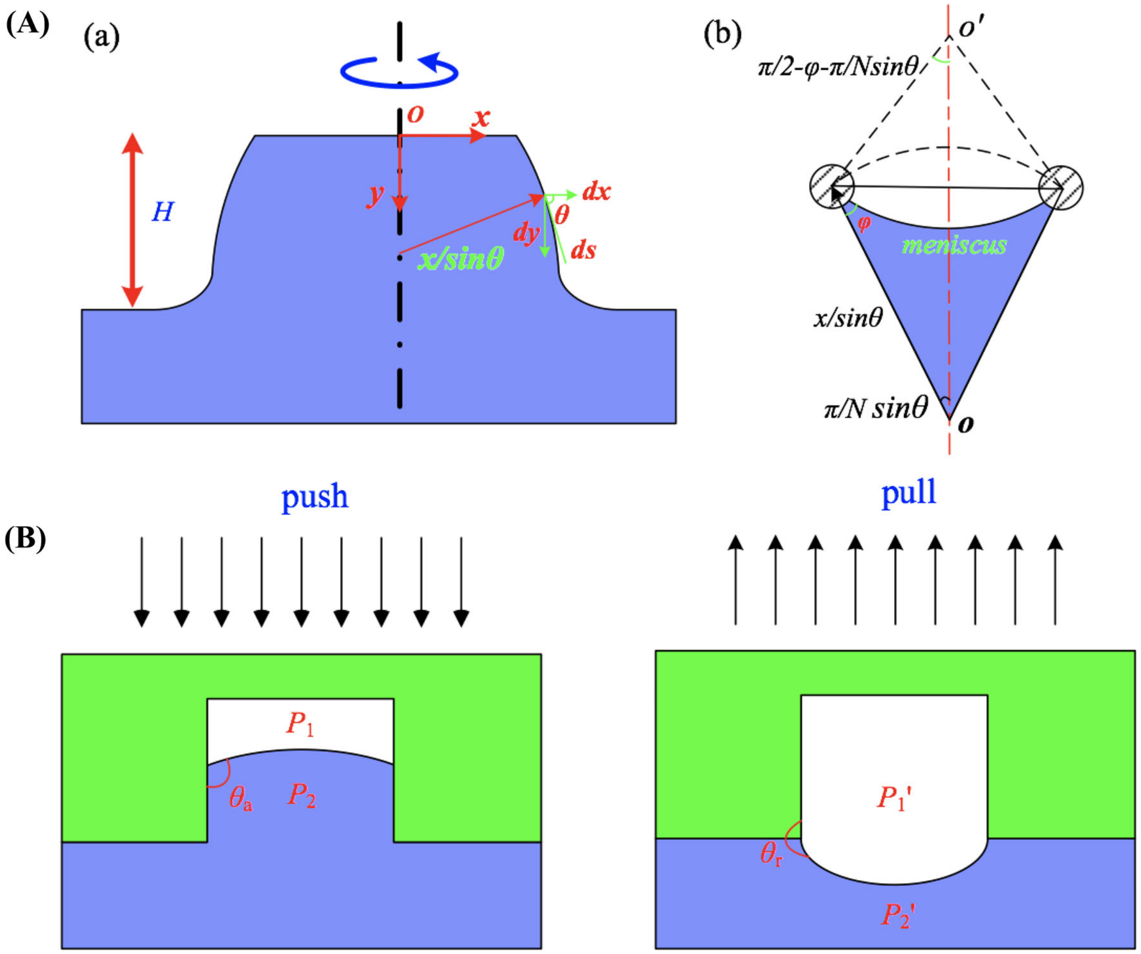

Fig. 4. Schematic diagram of dandelion pappi. **A** Geometry and coordinate system of dandelion pappi. (a) Side view: the top flat part is the pulvinus of the dandelion, (b) bird's eye view of two adjacent pappi. **B** The wetting of pulvinus under pushing and pulling. θa stands for advancing contact angle, and θr for receding contact angle

$$\Delta P = -\int_{r_1}^{r_2} \frac{2\gamma_{\mathrm{lg}}}{(r+R_0)^2} \sin\beta \mathrm{d}z \tag{6}$$

where R_0 is the radius of droplet; β is the half apex-angle of spindle-knot; r_1 and r_2 are the local radii of the joint and the spindle-knot, respectively.

Cactus spines with aligned structures also generate Laplace pressure on droplets due to the conical shape based on Eq. (6). The spine with smaller radius has a larger Laplace pressure according to Eq. (4), driving the movement of droplet from the tip to the base (Fig. 2B). And the grooves on tip and base have different roughnesses, generating a surface free energy gradient as well.

The capture mechanism of dandelion pappi as a collective fibrous structure is different from that of spider silk and cactus spines. The large bending deformation of dandelion pappi results from a joint effect of several forces including elastic bending, surface tension, hydrostatic pressure and adhesive force [8], providing space for liquid storage (Fig. 2C). But there still lacked more in-depth explanation. The theoretically predicted stability of liquid in a model brush can be adapted to a dandelion seed [10]. Han et al. first quantified the catching capacity of a real dandelion both experimentally and theoretically. The mathematical model uncovers that the collective wetting behavior is affected by both the properties of each single fiber and the group geometry. The liquid storage of a dandelion seed could be tuned by varying the number of fibers and their lengths. The success in droplet capture is the result of bending energy and surface energy. Particularly, the number and distribution of fibers play important roles in determining the state of wetting and capturing capacity, illustrating a cooperative-like behavior of fibrous materials. The catching capacity can be represented by the captured volume, which can be obtained by the simulation of a single fiber deformation and integration. When the length increases up to ∼6 mm (the average length of the pappi), the tip would "zip up"; that is, the fiber tips will stick together and the captured volume will no longer increase, indicating that the natural length of the dandelion seed may be optimal for adsorbing liquid.

Numerical models have been proposed to calculate the liquid volume grabbed by dandelion. For one pappus, its deformation is governed by the following equation (Fig. 4A)

$$\frac{1}{\rho} = \frac{\mathrm{d}\theta}{\mathrm{d}s} = \frac{M}{EI} \tag{7}$$

$$I = \frac{\pi d^4}{64} \tag{8}$$

The pappus bears two forces, i.e., the hydrostatic pressure and surface tension. The hydrostatic pressure can be neglected because the projection area exposed to the liquid is small owing to the large aspect ratio of pappus. The surface tension can be derived as follows:

$$q_s\left(x\right) = 2\gamma_{\mathrm{lg}}\cos\theta \tag{9}$$

The boundary conditions can be obtained as: at $s = 0$, the pappus is clamped ($\theta = 0$); and at $s = s_0$, it is free-end ($\frac{\mathrm{d}\theta}{\mathrm{d}s} = 0$), where s_0 is the total length of the pappus.

Similarly, Reis et al. [19] discovered that water can be captured by petal-like structures, as inspired by flowers. The water-grabbing behaviors of dandelion and flower are both under the effects of surface tension and hydrostatic pressure, but the difference is which effect plays a primary role. The hydrostatic pressure dominates the grabbing process of flower due to the large action area of wide petals, in which surface tension can be ignored. For water grabbing by dandelion, the hydrostatic pressure is negligible since the action area is relatively small, and surface tension dominates the process.

The spikes on a pappus are critical to the wetting of fibers when pushing the pappus into water, helping to generate the transition from the Cassie wetting state to the Wenzel wetting state, under which the adhesive force is enhanced. If the pulvinus is pre-wetted, the water can fill (or partially fill) the cavity. Then, the contact line would be pinned at the corner due to the geometric discontinuity when it is pulled (Fig. 4B). If the liquid is oil, then the pushing part is not necessary, as it would get wetted naturally.

4. Bio-inspired Materials and Their Applications

The discoveries of these three soft structures help in the design of multifunctional materials benefiting many different fields. Inspired by the spider silk, the artificial spider silk has been fabricated, on which the water droplet can move directionally under a misty environment (Fig. 3A) [6]. The main idea on fabricating artificial spider silk is to generate wettability gradient (e.g., chemical composition, roughness gradient) or geometry gradient (e.g., curvature). Many methods have been developed including dip coating (materials like nylon) [11, 20], fluid coating [21], microfluidic technology [11, 20–22] and electrospinning (materials like poly(methyl methacrylate)) [23–25] (Fig. 4A). The artificial intelligence nanofibers have also been fabricated to respond to external stimuli by introducing other physical responsive molecules, such as temperature, light and humidity [22, 23, 26]. By introducing magnetic nanoparticles, the graphene microfiber could adsorb oil from a water/oil mixture due to its hydrophobic surface properties and be easily retrieved (Fig. 4A(c)) [27]. These results put up with approaches to move microscale droplets, contributing to the design of highly efficient devices for water/fog collection, cell culture, microfluidics and other biology devices or systems [10, 19, 26–29].

The key for artificial cactus bio-inspired designs is to obtain multiscale needle-like structures for water transportation from the tip to the base. Single and arrays of artificial cactus spines were fabricated using copper needles capable of collecting water from humid air (Fig. 3C) [7, 12, 31]. Besides, magnetically responsive flexible conical arrays were fabricated, which could spontaneously and continuously capture fog from the humid environment (Fig. 4B(c)) [32]. Moreover, an oleophilic

Table 1. **Characterizations of three fibrous structures**

	Scale	Wettability	Collection efficiency	References
Natural spider silk	Dry: puff: 130.8 ± 11.1 μm; joint: 41.6 ± 8.3 μm; nanofibrils: 20–30 nm	Hydrophobic	First condense: 0.156 s	[6]
	Wetted: spindle-knot: 21.0 ± 2.7 μm; joint: 5.9 ± 1.2 μm		Move directionally: 0.702–0.796 s	
Artificial spider silk	Spindle-knot: 43.7 ± 5.4 mm; joint: 13.5 ± 0.7 mm	Polymers (e.g., PMMA, PS) from 56.7° to 92.7°	First condense: 7.708 s	[11]
	Height of $\sim$34.9 μm; length of $\sim$128.5 μm		Move directionally: 7.955–8.717 s	[47]
	Length: 1.5 mm		200–1000 μm/s.	[43]
	Spindle-knot: $\sim$41.9 μm in height, $\sim$128.2 μm in width		$\sim$0.8 nL of water, it needs about 7 s, 3 s and 2 s under low ($\sim$25 cm/s), middle ($\sim$75 cm/s) and high ($\sim$100 cm/s) fog flow velocities, respectively.	
Natural cactus spine	Base's distance: $\sim$7 to 23 mm; length: $\sim$800 to 2,500 mm; diameter in the middle portion: $\sim$30 to 65 mm	Hydrophobic	$\sim$20 to 30 cm/s	[7]
	Width of microgroove: $\sim$6.8 mm (base) to $\sim$4.3 mm (tip)			
Artificial cactus spine	Length of $\sim$2 cm	Superhydrophobic $\sim$155°	$\sim$29.9 cm/s	[30]
	Height: $\sim$3 mm; diameter (base): $\sim$770 μm		Array containing 144 spines /cm^2: 0.2 mL/h	[33]
	Base's distance: $\sim$680 μm.		Single: 1.41 μL/h	[45]
			180 artificial spines: 0.87 mL/15 min	
			Single: 0.3 μL/min	
Natural dandelion	Pappus length: 4.65–5.90 mm; pappus diameter: $10-40$ μm; spike length: 19.9 ± 2.3 μm; orientation angle: 36.7 ± 3.3°	130 ± 5.6° for water	Length of 8mm: $\sim$10 μL	[8]
	80–120 fibers per seed	43.3 ± 1.8° for oil	Length of 4 mm: $\sim$6 μL	[34]
Artificial dandelion	$\sim$5 mm	Glass fiber (hydrophilicity)	1.2×10^{-13} Nm^2 : 4.3 mg	[46]
			1.1×10^{-10} Nm^2: 126.9 mg	

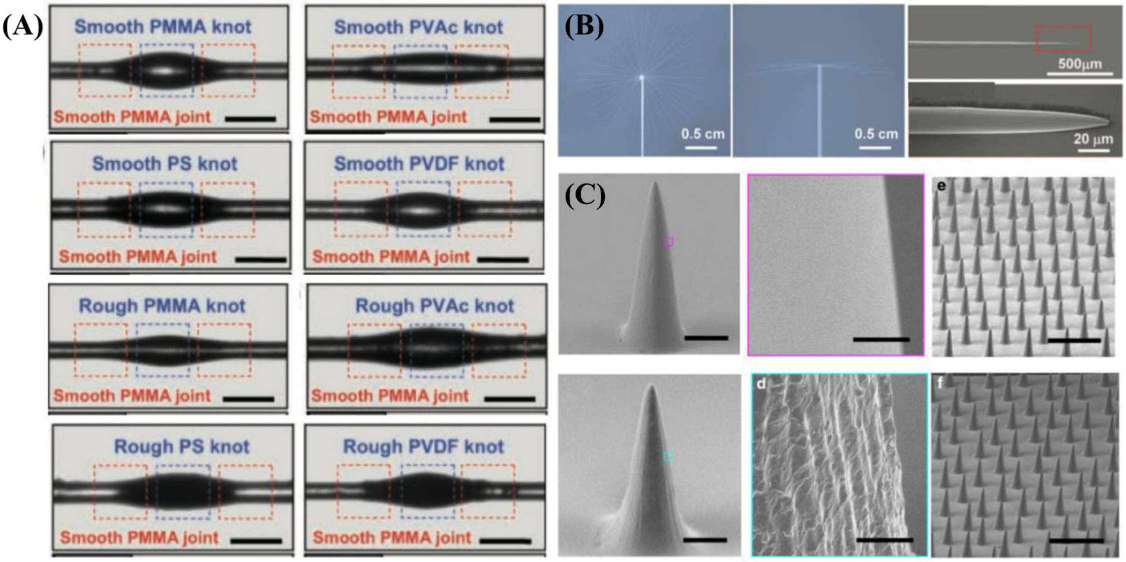

Fig. 5. Artificial bio-inspired materials. **A** Spider silk (scale bar: 50 µm) [11]. **B** Dandelion seed [8]. **C** Conical cactus spine (scale bar: 100 µm, 5 µm and 500 µm from left to right) [30]

PDMS conical needle array was fabricated for separating ∼ 10-µm oil droplets from water, showing great potential in both water collection and water/oil separation [12]. In many engineering applications, oil/water mixtures with micron-sized oil droplets are more convenient and efficient than two separated phases. These findings would be helpful for designing novel smart devices and propose new approaches in microfluidics, oil/water separation and water storage in drought regions [30, 33] (Fig. 4B).

Natural fibrous system, such as dandelion pappus, provides a novel strategy for catching liquid in a highly proactive and well-controlled way. Jiang's group has also shown the possibility of fabricating pappus structure using glass fibers which presented stable performance of water catching (Fig. 3B) [12]. It is found that the pappus can manipulate both aqueous and oil droplets with its hydrophilic/oleophilic structure (contact angle of ∼ 130° with water and ∼ 43° with mineral oil) [34]. The wetting/separation process can be achieved by downward pressing operations (Fig. 4C(b)). The dewetting/release process can be triggered by immersing the pappus with captured water (with higher surface tension) into oil (with lower surface tension) (Fig. 4C(c)). This presents great potentials in a broad range of applications, including oil/water separation and pump-free, digital liquid handling, especially for small volumes of high-viscosity liquids (Fig. 4).

Fibers with gradient structure can be fabricated by the Rayleigh instability method [35–37], electrodynamic method [38, 39] and microfluidic method [40, 41]. If water collection velocities are ∼ 30 µm/s for natural spider silk and ∼ 12 µm/s for cactus spine, the velocities for artificial structure under the same size are ∼ 21 µm/s and ∼ 3 µm/s, respectively. In general, the artificial fibers with larger structure size have higher efficiency in collection than the smaller ones (Table 1). Different materials (e.g., temperature-responsive hydrogel [26]) or coating methods [21, 42] help to further regulate the wettability gradient of artificial fibers. Under the same conditions, arrays [34] and fiber network [43] have been used to achieve larger scale of collection (Figs. 5, 6).

In terms of scale and structure complexity, it is still challenging to achieve bionic fabrication precisely. For example, microgrooves on the cactus spine enhance the surface roughness and induce larger Laplace pressure gradient, driving droplets to move along the spine. The in-depth understanding of microscale mechanism will help to improve the performances of artificial structures and further expand their application prospects.

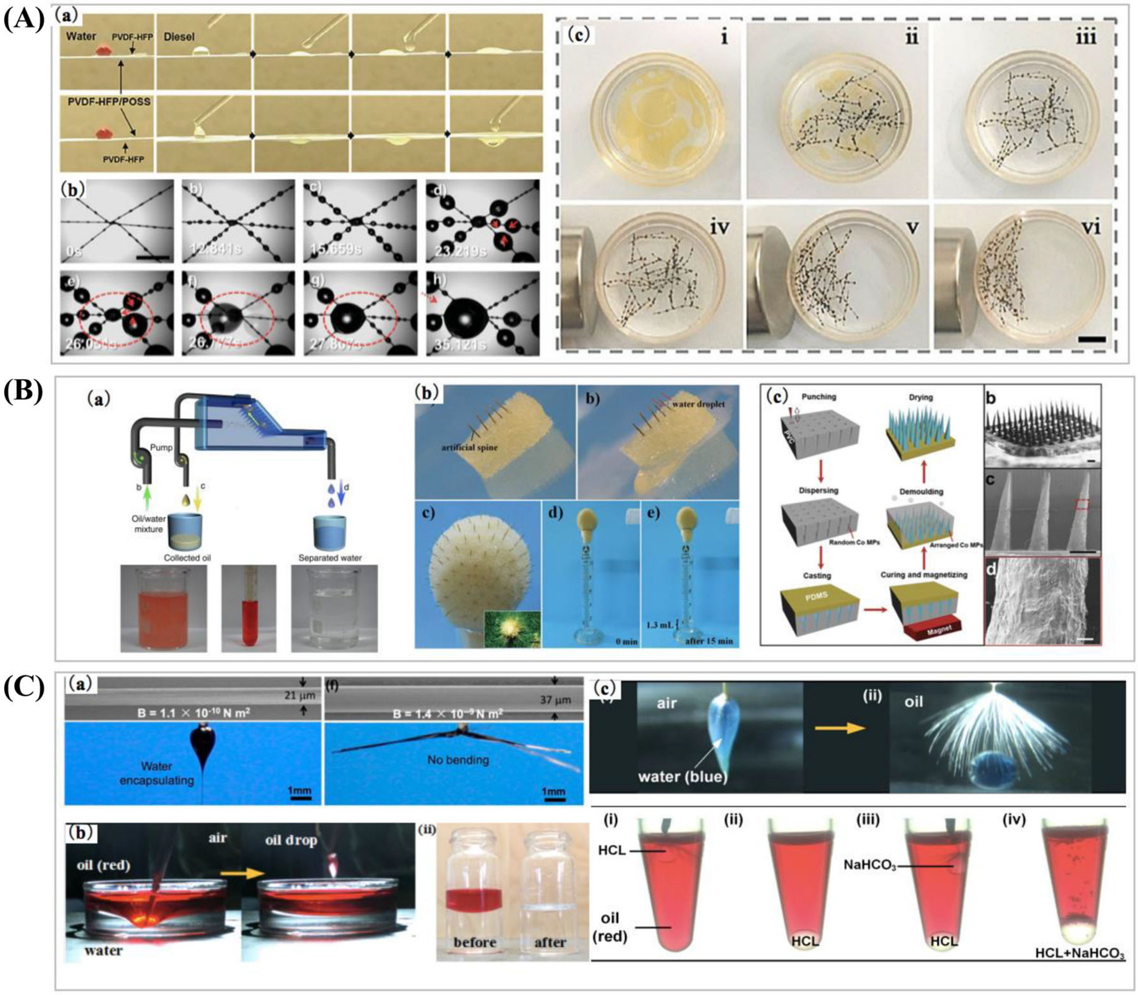

Fig. 6. Applications. **A** Artificial spider silks are used in (a) droplet motion (scale bar: 1 µm) [44], (b) water collection (scale bar: 1 mm) [43] and (c) oil absorption (scale bar: 1 cm) [27]. **B** Artificial cactus spines are used in (a) oil/water separation [12], (b) water/fog collection [45] and (c) magnetic collection (scale bar: b and c, 1 mm; d, 50 µm) [32]. **C** Dandelion pappi are used in (a) catching water (scale bar: 1 mm) [46], (b) oil/water separation [34] and (c) liquid release [34]

5. Conclusions and Outlook

Nature has been the source of inspirations for centuries, providing thousands of vivid cases. Many reviews have presented examples of bio-inspired surfaces, but far less attention has been devoted to soft fibrous structures in nature. In this short review, we summarize the structures, basic models, bio-inspired structures and applications of three soft fibrous structures, i.e., spider silk, cactus spine and dandelion pappi. Although fibers have various advantages for liquid manipulation, research on natural and synthetic fibers still has a long way to go. The precisely biomimic fabrication of both structure and wetting characteristic remains an open challenge. Lacking of in-depth understanding of structure and microscale mechanism limits the optimization work to further improved functions. Most studies are limited to the performance of single fiber, which, however, does not apply to large-scale applications. Integrated fibers (e.g., fiber web and dandelion pappus structure) show more potential to achieve various applications with high efficiency, but the mechanism of integrated effect of fibers is still ambiguous. It is likely to be achieved by using fiber materials with deeper understanding of the wettability mechanism and specific surface treatment. These phenomena have implications for the bio-inspired applications of natural structures in droplet manipulation and other potential fields.

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