ORIGINAL ARTICLE

A bio-inspired flexible fiber array with an open radial geometry for highly efficient liquid transfer

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Generally, highly efficient liquid transfer is difficult to achieve in open fibrous systems, as compared with traditional closed systems, because capillary coalescence frequently occurs when fiber arrays interact with a liquid. Dandelion pappi, which are flexible fiber arrays with open radial geometries, exhibit the unique ability to capture and hold large amounts of water steadily. Drawing inspirations from this natural system, we developed an artificial flexible glass fiber array with a radial geometry that is capable of holding large volumes of liquid and controllably transferring the liquid onto a substrate. The highly efficient liquid transfer of the open fibrous array can be attributed to the synergistic effects of the flexibility of fibers that radially arranged with a big open angle that enables a distinct elastic deformation of the fibers and the consequent formation of a large space within the fiber array for liquid storage, and the highly adhesive property of fibers that helps the pinning of three-phase contact line on the fibers. This unique open system may provide a novel strategy for highly efficient and controllable liquid transfer.

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INTRODUCTION

Fibrous structures are widely distributed and abundant in nature, providing versatile functions such as thermal insulation, motion assistant and light reflectivity that aid in the survival of living organisms and help them to better adapt to the local environment.^{1–5} Among the many functions imparted by fibrous structures, their wettability at both the micro- and macro-scale is very important because it can facilitate or even determine the manner in which organisms interact with the surrounding environment.^{4–6} As our recent studies revealed, the spider silk with unique periodic spindle-knot and joints can harvest water from humid air,⁴ certain cactus species with conical spines can efficiently collect fog⁵ and certain fish with anisotropic hook-like spines exhibit self-cleaning under water.⁷ It has also been suggested that the nanowire-structured bristles of gecko feet enable high adhesion to solid and liquid drops arising from the wetting-induced structure rebuilding of the nanowire array.^{8,9} Such natural phenomena have inspired materials scientists to more closely examine the wetting behavior of fibrous media,^{10–13} resulting in the development of numerous bio-inspired artificial fibrous systems with special wettability properties. To date, most endeavors have focused on developing either anisotropic fibers for water collection^{4,5} and pumping¹⁴ or a vertically aligned fiber array for controllable morphologies through capillarity-induced self-assembly.^{12,15} It has been demonstrated that fibrous arrays can realize versatile dynamic wetting superior to traditional closed systems (such as tubes and channels).^{16–18} For example, artificial spider silk made from

hydrophilic poly(ethylene glycol) and hydrophobic polystyrene can harvest water from humid air,¹⁹ and polymer conical needle arrays can separate micro-sized oil droplets from water.²⁰ Moreover, open fibrous arrays enjoy the advantages of easy preparation, low cost and low hydrodynamic resistance.^{14,19} However, manipulating liquid in open fibrous systems is rarely reported and remains a significant challenge,²¹ because capillary coalescence is frequently encountered when fiber array interacted with a liquid.²²

Here, we describe the special dynamic wetting behavior of dandelion pappi. A large mass of water can be encapsulated by an open array of pappi arranged in a radial geometry with a large open angle. We demonstrated that the flexibility and highly adhesive ability of the pappus fiber, associated with the hydrophilic annular pulvinus, played a crucial role in efficiently encapsulating liquid in such an open fiber array. The highly adhesive pappus and hydrophilic annular pulvinus impart a large adhesive force to water that helps to pin the three-phase contact line during interaction with liquid. The elasticity of the pappi arranged with large open angles enables a distinct elastic deformation under the joint forces of surface tension and hydrostatic pressure during interaction with liquid. Thus, a large space is generated within the deformed pappi for liquid storage. Inspired by these findings, we developed an artificial flexible fiber array with an open radial geometry that mimics the structural features of dandelion pappi and the highly efficient liquid transfer was realized. We envision that such an open system may provide a novel strategy for highly efficient and controllable liquid transfer.

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EXPERIMENTAL PROCEDURES

Materials

Dandelion seeds were picked fresh from the grassland of Beihang University (39.98° N, 116.34° E). Flexible glass fibers with a diameter of $\sim 20\ \mu\text{m}$ and a bending stiffness $B = 5.6 \times 10^{-10}\ \text{Nm}^{-2}$ were purchased from Taishan Fiberglass, Inc. (Shandong, China). Hydrophilic glue-modified acrylate adhesive was purchased from Tianjin Weldmaster Technological Development Co., Ltd. (Tianjin, China).

Procedures

Characterization of the material morphology and structure. Optical images were acquired with a camera (Canon EOS40D, Tokyo, Japan). The microstructures of the dandelion seeds and the artificial conical fibers were investigated by scanning electron microscopy (JEOL, JSM-6700F, Tokyo, Japan) at an accelerating voltage of 3.0 kV.

Measurement of the adhesive force of the fibers. The adhesive force measurement of the pappus to water was performed using a high-sensitivity micro-electromechanical balance system (DataPhysics DCTA 11, Filderstadt, Germany). A water droplet ($1\ \mu\text{l}$) was suspended on a copper cap connected to the microbalance. A single pappus was then moved toward the water droplet at a speed of $0.02\ \text{mm s}^{-1}$ until it contacted the water droplet. The shape changes of the water droplet and the force variations were recorded by a digital video camera (WV-CP280/CH, Panasonic, Osaka, Japan) and the microbalance, respectively.

Preparation of an open radial conical glass fiber array. Flexible conical glass fibers were prepared by a standard electrochemical method. First, commercial flexible glass fibers were rinsed with deionized water and dried with nitrogen. Next, the clean fibers were held vertically close to a container filled with hydrofluoric acid on a programmed lift table. By moving the lift table up and down at a $0.5\ \text{mm s}^{-1}$ speed, the glass fibers were etched to achieve conical shapes. The flexible conical glass fibers of the same diameter and length were then fixed radially and uniformly onto the edge of a rigid polymer rod to fabricate a dandelion-inspired artificial open radial system.

Process of water entrapment. The water-entrapping properties of the dandelion and the dandelion-inspired artificial system were evaluated using a DataPhysics OCA20 system with a moving platform and a CCD camera. The process was recorded by the CCD camera.

RESULTS AND DISCUSSION

Water encapsulation by dandelion pappi

Taraxacum officinale, often simply called dandelion, is a common composite perennial of the *Asteraceae* family that can be found growing in many temperate and moist regions of the world.²³ The capitulum of the dandelion contains hundreds of seeds forming a ball of fluff (Figure 1a) when the dandelion seeds are ripe. The main part of each seed unit is the flying appendant of white pappus that is connected with the achene by a beak. The most attractive feature of the dandelion is that the seeds are easily detached from the mother plant by wind after maturity to parachute the seeds elsewhere for the start of the next generation.²⁴ Interestingly, we observed another intriguing but less-studied feature in that a large water droplet can be encapsulated by the pappi of the dandelion seed after it rains (Figure 1b). As shown in Figure 1b, the dandelion seed was capable of capturing a large dew droplet within the pappi. Even for downward-oriented pappi, the droplets are readily held in place with little distortion in their spherical shape, as indicated by the arrows in Figure 1b. This phenomenon strongly suggested that the open system of dandelion seed pappi could hold a relatively large volume of water that could potentially be used to facilitate its growth and maturation.²⁵

To explore the unique ability of dandelion pappi to encapsulate water, a single dandelion seed was evaluated and inversely fixed on the holder of a motorized precision translation stage (OCA20, DataPhysics) capable of smooth movement at a speed of $v = 1.7\ \text{mm s}^{-1}$ toward a static water surface (Figure 1c₁). A camera was used to record this process. When the pappi of the dandelion seed were pressed downwards into the water, they did not penetrate the air–water interface until they were vertically moved to a position $\sim 0.4\ \text{cm}$ below the surface of the water (Figure 1c₂). During their interaction with the water surface (Supplementary Figure S1), the pappi bent upwards, producing a water dimple (Figure 1c₂). This behavior can be ascribed to the hydrophobicity of the pappus (Supplementary Figure S2) and the surface tension of the water between the neighboring pappi, as has been suggested for *Nymphoides* flowers.²⁶ Surprisingly, the hydrophobic pappi held a relatively large water droplet after detachment from the water surface (Figure 1c₄). After pulling the pappi vertically upwards, the liquid adhering to the pappi was also drawn upwards, forming a water column between the pappi and the water interface (Figure 1c₃). After the pappi were fully detached from the air–water interface, there was a large water droplet that was cooperatively encapsulated by the numerous deformed pappi (Figure 1c₄). Quantitatively, a considerable mass of liquid (up to 96 times the weight of the pappi) could be captured, demonstrating that the open dandelion pappi array is capable of highly efficient liquid storage.

Structure of the dandelion pappus

The dandelion pappi system is a typical macroscopic open radial fibrous array, where hundreds of fibrous pappi with hierarchical anisotropic microstructures are radially arranged on the annular pulvinus (center plate) (Figures 2a and c). The top-view image of a dandelion seed in Figure 2c illustrates that the pappi are radially arranged on the pulvinus at the top of the beak. Each dandelion seed contains $\sim 113 \pm 11$ pappi that are oriented in a radial pattern with an open angle, 2β , of $\sim 160^\circ$ to 180° . To investigate their structures in detail, we used scanning electron microscope to observe individual pappus and the pulvinus (Figures 2d–f). The main body of the pappus has a tapered architecture with the diameter successively increasing from an average of $\sim 10\ \mu\text{m}$ at its tip to an average of $\sim 40\ \mu\text{m}$ at the butt end (Figure 2e and Supplementary Figure S3). Considering that a single pappus is $\sim 0.5\ \text{cm}$ in length, the single-celled cellulose fiber pappus²⁷ has a high aspect ratio of 300 ± 100 , making it mechanically flexible (Supplementary Figure S4). Moreover, many elaborate microspines and aligned microgrooves are evident on each pappus, forming a unique hierarchical structure that provides many anchoring points for pinning the three-phase contact line.²⁸ We also noticed that the pulvinus had a sucker-like structure with a protuberant closed circle in the middle (Figure 2f₁). This structure favors pinning of the liquid because of the close contact with the water²⁹ in conjunction with the radially distributed rough-textured geometry on its surrounding areas (Figures 2f₂ and f₃).

Physical mechanism for highly efficient water capture

It is proposed that the special wettability of the pappus and the annular pulvinus significantly contributes to the highly efficient liquid capture/encapsulation of dandelion pappi. For a single pappus with a diameter of $\sim 20\ \mu\text{m}$, there was high adhesion to water (Figure 3a and Supplementary Figures S5 and S6). As suggested in Figure 3a, a water droplet ($1\ \mu\text{l}$) can tightly adhere to a single pappus and become deformed when detached from the pappus, with a small amount of

Figure 1 Water droplet encapsulation by an open flexible dandelion pappi system. (a) The white fluff ball formed after dandelion seed maturity. (b) Many water droplets are held by the dandelion pappi in all directions after it rains, even for seeds with downward-oriented pappi, as indicated by arrows. (c) A relatively large water droplet was encapsulated by the pappi through the dip-withdrawal process. The pappi gradually approached the water interface (c_1) and made a dimple (c_2) when pressed into the water, while a water column (c_3) was adhered to the pappi and encapsulated into a large water droplet (c_4). The weight of the water droplet was ~ 96 times the weight of the pappi.

Figure 2 Morphology and structure of the dandelion seed unit. (a) Optical photo of a side view of a dandelion seed unit, consisting of the pappus-beak-achene with $\sim 113 \pm 11$ pappi that are 5 mm in length. (b) The cartoon of the dandelion seed unit indicates that the open angle between each pappus, noted as 2β , is $\sim 160^\circ$ to 180° . (c) Optical photo of a top view of a dandelion seed unit, with the pappi radially arranged on top of the beak to form an open radial system. (d) Scanning electron microscope (SEM) image of a single pappus revealing a conical structure with a length of ~ 0.5 cm. (e) Magnified SEM images that taken from three different positions, the tip (e_1), the middle (e_2) and the bottom (e_3) of the pappus. (f) SEM images of the sucker-like structure of the pulvinus (f_1) with a protuberant closed circle in the middle (f_2) and a radial distributed roughly textured surface on the surrounding area (f_3).

residuum on the pappus. The measured adhesive force of water on a single pappus is larger than $9.75 \pm 0.9 \mu\text{N}$ when evaluated using a water droplet of $1 \mu\text{l}$ (Supplementary Figure S6). Moreover, the

pulvinus of the dandelion is hydrophilic, such that water can easily adhere and become pinned on it (Figure 3b). Taken together, the pappi and the pulvinus both display high adhesion to water that is

Figure 3 Mechanism for highly efficient water capture. (a) Adhesion force measurement of a water droplet (1 μ l) on a pappus. The $\Delta F = 9.75 \pm 0.9 \mu$ N stretch force is smaller than the adhesive force of the pappus to water, indicating that the pappus is highly adhesive to water. The successive images of a water droplet interacting with a pappus are denoted by (a₁–a₃). Residual water (a₄) remained on the pappus after water droplet detachment. (b) Digital images of the pulvinus before (b₁) and after (b₂) interaction with water. The water becomes tightly pinned to the pulvinus, indicating the high adhesiveness of the pulvinus to water. (c) A two-dimensional (2D) model describing the deformation of the pappus because of interaction of the elasticity of the pappus and the adhesion force to the water arising from the surface tension and the hydrostatic pressure. (d) Two radially arranged glass fibers with the open angle of 176° interacting with water to encapsulate a relatively large water membrane. After interacting with the water interface (d₁ and d₂), the radial glass fibers adhered a water membrane (d₃) when drawn upwards, and totally encapsulated the large water membrane (d₄) when left the water interface.

favorable for the pinning the three-phase contact line when the structure is detached from the air–water interface.

When capturing water, the water adhering to the pulvinus is drawn upward, forming a water column when the dandelion pappi are pulled off the water surface. Through the combined effect of the pappus bending deformation, a relatively large water droplet can be encapsulated by the open radial fibers. In this procedure, the elasticity of the pappus allows it to deform under the joint forces of the surface tension and the hydrostatic pressure (Figure 3c). For a flexible dandelion pappi, the elastic bending force (F_e) that is generated from the deformation of the fiber can be expressed as:

$$F_e = \frac{2\theta EI}{l^2} \quad (1)$$

where θ is the local angle between the x axis and the fiber, l is the horizontal displacement of the deformed fiber, E is the Young's modulus of the fiber and $I = \pi d^4/64$ is the second moment of the area of the section of the fiber with diameter d . The adhesion force (F_a) is equal to surface tension force (F_γ) that can be expressed as:

$$F_a = F_\gamma = -2\gamma ds \quad (2)$$

per unit length, where γ is the water surface tension and s is the arc length along the fiber spanning from 0 to l . The hydrostatic pressure can be expressed as:

$$F_h = -\rho g(Y+H)n \quad (3)$$

where $n = -\sin\theta e_x + \cos\theta e_y$ is the unitary vector normal to the upper face.³⁰ For a point (A'), when immediately withdrawn from the water

interface, the F_γ and F_h are constant, whereas F_e decreases with an increase in l , suggesting that the elastic deformation of the fibers increases to form a larger space between the fibers for liquid storage. The tips of the pappi then aggregate once the pappi leave the water interface to encapsulate the water droplet. From this analysis, we can conclude that the flexibility of the radially arranged pappi with a large open angle, associated with its high adhesiveness to water, allow the dandelion pappi to efficiently capture water. Another favorable driving force is the Laplace pressure from the conical structures of the pappi that can further hold the encapsulated water within the dandelion pappi.

To understand the dynamic water capture process, a model of 'two radially arranged flexible glass fibers' was constructed, where a pair of conical flexible glass fibers was fixed to a rigid polymer rod using hydrophilic glue at one side and arranged in plane with an open angle, 2β , of $\sim 176^\circ$ (Figure 3d₁). A dip-withdrawal experiment was then carried out to model the water-encapsulation phenomenon of the flexible dandelion pappi (Figures 3d₁–d₄). It is noted that the open angle of the flexible glass fibers decreased as the fibers were withdrawn from the water. Simultaneously, a water column was pinned on the fibers and moved with them during withdrawal from the air–water interface. Thus, the model system exhibited essentially the same behavior as the natural dandelion pappi. Here, the glass fiber exhibited a rather high adhesion to water, resulting in a high adhesive force larger than $22.5 \pm 0.9 \mu$ N (Supplementary Figure S7a). The rigid polymer (Supplementary Figure S7b1) that was coated using a layer of hydrophilic glue is a typical hydrophilic surface with a water contact

Figure 4 Parallel fibers with small open angles (approaching to 0°) encapsulate less liquid. (a) The interacting with water for two parallel glass fibers with the open angle of $\sim 0^\circ$, where a very small water membrane is formed. a_1 – a_2 is the process of the parallel glass fibers approaching and contacting the water interface. a_3 – a_4 is the process of the glass fibers aggregating together, forming a small water membrane when detaching from the water interface. (b) The corresponding model for two parallel glass fibers with the open angle of $\sim 0^\circ$.

Figure 5 Highly efficient liquid transfer by the artificial open radial fiber array with an open angle approaching 180° . Optical photos of (A) a top view and (B) a side view of an artificial glass fiber array with 40 glass fibers. (C) Low- and (D) high-magnification scanning electron microscope (SEM) images of a single conical glass fiber revealing its conical shape. (E) The artificial glass fiber array encapsulating a relatively large water droplet after the dip-withdrawal process. When approaching (e_1) and dipping into water, the artificial fiber array bent forming a dimple (e_2), and encapsulated the water column that adhered to the fibers (e_3) into a large water droplet when withdrawn from the water bath (e_4). (F) The relationship of the glass fiber number, length of fibers and water droplet weight. Optical photos of a water droplet encapsulated by 20 glass fibers (G) and by 80 glass fibers (H). For a better observation, the water was colored into black.

angle of $68.7 \pm 1.1^\circ$ (Supplementary Figure S7b2). Resembling that of natural pappi system, the high adhesiveness of glass fibers and the polymer were beneficial for pinning the three-phase contact line during withdrawal from the water surface. The role of the adhesiveness of the fibers in water capture is further evidenced by the fact that ethanol with a much smaller surface tension of 22.7 mN m^{-1} compared with water (72.8 mN m^{-1}) cannot be encapsulated within the fibers (Supplementary Figure S8). As mentioned above, F_a is equal to F_γ that decreases dramatically with decreasing surface tension. Thus, the adhesive force that is generated from the surface tension

becomes insufficient to bend the fiber, and deformation of the fiber cannot occur. As a result, ethanol cannot be encapsulated by dandelion pappi (Supplementary Figure S8).

The large open angle of the glass fibers enables maximum fiber bending by virtue of the fiber elasticity, resulting in the formation of a large space between the fibers where liquid can be encapsulated. The elastic force can be dynamically balanced by the joint forces of the surface tension and the hydrostatic pressure (Figure 3c). Here, the open angle of the radial fiber array is directly related to the elastic bending deformation of the fibers that plays a crucial role in the

highly efficient water capture process. To explore the role of the open angle, we conducted another experiment in which pairs of flexible glass fibers were arranged in a vertically parallel manner, such that the open angle, 2β , was zero (Figure 4a₁). A different situation occurred when these fibers were withdrawn from the air–water interface. Capillary coalescence occurred, allowing the fiber array to hold only a small volume of water (Figures 4a₂–a₄). This indicates that changing the open angle of the fibers can vastly impair their ability to capture liquid. Because the open angle between the glass fibers is so small that the elastic deformation corresponding to the bending moment is also extremely small, only a very narrow space can be formed between the fibers. Thus, when withdrawn from the liquid, the fiber array cannot form a water column and instead the fibers coalesce, allowing only a limited amount of liquid to be encapsulated by the parallel glass fibers (Figures 4a₁–a₄). The small open angle between glass fibers easily induces elastocapillary coalescence, causing aggregation of the glass fibers (Supplementary Figure S9).²² Thus, the large open angle between the flexible fibers generates a beneficial elastic curvature, corresponding to the large space, that can hold liquid, making it possible to capture liquid with high efficiency.

Highly efficient liquid transfer by an artificial glass fiber array with an open radial geometry

Inspired by this finding, we fabricated an open radial fiber array to mimic the geometry of the dandelion pappi in which flexible glass fibers were fixed radially and uniformly onto the edge of one side of a rigid polymer rod (Figures 5A and B) by hydrophilic glue. Moreover, the tips of the flexible glass fibers were chemically etched into a conical shape (Figures 5C and D) in hydrofluoric acid. Using this artificial open radial fiber array, we achieved highly efficient liquid retention with the weight of the water encapsulated as high as ~ 117 times that of the fibers. As suggested by Figure 5E, the radial glass fibers deformed upon interacting with the water. A water column with a large mass adhered to the fiber array during the withdrawal process. Consequently, a large volume of water was encapsulated in the fiber array after detachment from the air–water interface. This also demonstrates the crucial roles of the radial arrangement of fibers and their adhesive properties for holding and transferring water efficiently.

For the artificial fiber array, we evaluated the effect of the number and the length of flexible glass fibers on the liquid capturing ability, as summarized in Figure 5F. For a fixed length of flexible glass fibers, our bio-inspired system could encapsulate more liquid as the number of individual fibers increased. For the system with same number of flexible glass fibers, the amount of captured water increased with increasing fiber length up to a certain length, where longer fibers decreased the amount of water captured. We assume that the critical length, L_{crit} , is the optimal length in our bio-inspired radial fiber array that shows a maximum water capturing ability. For $L < L_{\text{crit}}$ such that the length of the fibers is short, the fibers cannot completely encapsulate the water droplet because of their impaired elasticity. Consequently, a relatively small water droplet was captured by the fiber array. For glass fibers beyond the critical length, $L > L_{\text{crit}}$, the cooperative effect of aggregation caused by elastocapillary imbibition, as well as by gravity, also leads to the capture of relatively smaller droplets of water. Here, the critical length of ~ 0.5 , 0.9 and 1.0 cm was given for the radial fiber array composed of 20, 40 and 80 fibers, respectively, making the aspect ratio similar to that of natural pappi. Only when the tips of the fibers aggregate together at the critical length can the fibers display maximum deformation to capture the largest volumes of water.

CONCLUSION

Here, we revealed a novel open geometry fiber system (that is, dandelion pappi) that enables the encapsulation of a large volume of water. We propose that the radially arranged flexible pappi (fibers) with large open angles and the high adhesion properties of the pappi and the pulvini play a key role in the liquid manipulation ability of this open fibrous system. Theoretical analysis demonstrated that large volumes of water can be steadily held within this topologically structured open fibrous array by the cooperative effect of the elasticity of the pappi that is responsible for balancing the joint forces of the surface tension and hydrostatic pressure of the liquid, and the high adhesiveness of the fibers that pins the three-phase contact line. Inspired by this finding, we designed an artificial radial fiber array to mimic the structural features of dandelion pappi. We observed identical liquid handling abilities between the artificial system and the dandelion pappi. Liquid was captured by the artificial radial array with a remarkably high efficiency. Moreover, the liquid retention could be tuned by varying the number of fibers and their lengths. Furthermore, these bio-inspired fiber arrays can perform controllable liquid transfer, as demonstrated by multiple rounds of transfer of the liquid onto a substrate in a controllable manner (Supplementary Figure S10). We envision that this open system may provide a novel strategy for highly efficient liquid transfer in a well-controlled manner.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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