Bioinspired Superwetting Open Microfluidics: From Concepts, Phenomena to Applications

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Microfluidics and bioinspired superwetting materials, as two crucial branches of scientific research, are entering their golden age of development. As an emerging interdisciplinary subject of these two fields, bioinspired superwetting open microfluidics is triggering technological revolutions in many disciplines, including rapid medical diagnosis, biochemical analysis, liquid manipulation, 3D printing, etc. However, this new research area has yet to attract extensive attention. So, a timely review is necessary to organize the development process, summarize current achievements, and discuss the challenges or chances for the ongoing scientific trend. In this review, the evolution from closed to open microfluidics is combed first. Then, three typical bioinspired superwetting systems are introduced emphatically. Based on this, the bioinspired superwetting open microfluidics is divided into different categories according to the bionic objects as the focus of this study. Taking natural phenomena as the entry point, the research from the underlying mechanism to the application is systematically discussed and summarized. Several emerging applications are also mentioned. Finally, some views on major problems, existing challenges, and developing trends are briefly put forward in this field to guide future research.

1. Introduction

Driven by the desire for miniaturization and planarization of biochemistry devices for analysis, detection, and separation, microfluidic technology has experienced explosive development

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in the past few decades.^[1-8] The rise and vigorous growth of microfluidic technology enable the precise control and operation of microliter or less liquid. Microfluidics-based lab-on-a-chip and micrototal analysis systems (µTAS) have therefore been established. At the same time, microfluidics has led to disruptive changes in other areas of science and technology, such as microdiagnostics, chemical analysis, micro-nano manufacturing, artificial tissue, cell detection, and so on.^[6,9–14] In the study of microfluidics, the choice of pipeline material and the control of fluid are the two most concerning issues. For material selection, researchers have developed a series of microfluidic systems using different materials.^[1] Inorganic material is first used to build microfluidic systems because of their excellent stability, strong resistance to various solvents, and good thermal conductivity. The traditional glass capillary tube can be considered a preliminary microfluidic device.

Glass-based microfluidics is the most common choice, but their fragility, difficulty in constructing complex pathways, and weak ability to regulate surface chemistry and structure are unavoidable.^[15-18] Organic materials can solve these problems to some extent.^[19-23] The preparation of microfluidic channels with organic materials is easier to obtain complex pathways, which can also be modified and functionalized better. However, these advantages are achieved at the expense of the material's light transmittance, mechanical properties, and chemical stability. More importantly, organic polymer microfluidic devices often strongly interfere with chemical detection and analysis. Ideally, inorganic-organic hybrid materials can provide an effective way to overcome these challenges. And currently, this is becoming the most promising selection of microfluidic materials.^[1,24] Recently, liquid has been utilized for constructing microfluidic channels to reduce transport resistance.^[25-30] However, fluid control is also a problem for the liquid channels.

Due to the scale effect, the fluid properties from the microscopic perspective are entirely different from the macroscopic view. It can be divided into three phenomena: the significant surface/interface effect, the advantage of viscous force over inertial force, and the efficient mass and heat transfer performance.^[6,31,32] These three points are like a double-edged sword, giving microfluidics some application potential and bringing some inevitable defects. In terms of fluid transport in closed microfluidics, the disadvantages are mainly manifested in three



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Figure 1. From a) closed microfluidic technology to b) open microfluidic technology. 2D/3D open microfluidic technology can effectively solve the three significant challenges of the closed system by breaking up the constraints of closed pipelines. The advantages of open microfluidic technology are based on lower fluid transport resistance, better real-time operation ability, and lower preparation costs. Reproduced with permission.^[56] Copyright 2012, RSC; Reproduced with permission.^[42] Copyright 2008, PNAS.

aspects. First, a closed pipeline brings considerable flow resistance, so the microfluidic system needs to be equipped with enough pump devices to drive the liquid transport. Second, the nonoperationality caused by the closed pipeline restricts the real-time control of microfluidics, significantly reducing flexibility. Third, the closed pipeline is not only complex to prepare but also expensive, limited by curvature, space density, and many other factors (Figure 1a).^[16,22,33] With the continuous progress of science and technology, and microfluidic technology is developing in the direction of intelligence, integration, and portability. One promising solution to these problems is to break the restrictions of closed pipelines and push microfluidic technology into open space. Open microfluidics has become a new research focus since the start of this century,[21,34-39] and paper-based open microfluidics is the typical representative in its primary stage. Paper-based microfluidics technology^[40-44] based on porous substrates has the excellent rapid diagnosis and microchemical analysis advantages. But due to the lack of a solid wall, it is difficult to restrict the movement path of microfluidics with only help from the substrate edge. In addition, it is challenging to introduce a pumping device to provide extra driving force and control to the open system. Therefore, developing new liquid-solid interaction modes is necessary to realize the high degree of control in an open microfluidic system. Fortunately, the rapid development of bioinspired superwetting surfaces has ignited hope.^[45-49] A series of bioinspired superwetting surfaces, such as superhydrophobic, superhydrophilic, and superlubricated surfaces,^[50,51] can be potentially applied to the open microfluidic system to overcome the current constraints. With the synergistic effect of microstructure and wettability, liquids exhibit unique static or dynamic behaviors on these bioinspired superwetting surfaces.^[52-55] Taking advantage of the unique properties of the bioinspired superwetting surfaces, the concept of bioinspired superwetting open microfluidics emerged at the right moment, and many excellent new creations have been reported and presented. However, this new-born concept has not been widely appreciated, and even many relevant researchers have not established a clear understanding of it.^[10] So, it is exceptionally essential to sort out, classify and summarize the current research works of bioinspired open microfluidics. A timely review can facilitate spreading this new concept and help guide the next research step.

In this review, we first briefly combed the evolution from closed microfluidics to open microfluidics. Next, the roles of various superwetting systems in open microfluidic technology are summarized. Significantly, according to different bionic objects, typical bioinspired superwetting open microfluidics have been summarized from phenomena, mechanism to application. Finally, we put forward our views on existing problems, future challenges, and developing trends in this field to guide further research.

2. Traditional Paper-Based Open Microfluidics

Paper-based microfluidics,^[56-59] as the most typical pioneer in open microfluidics, was born as a novel diagnostic platform in George Whiteside's group in 2007.^[43] It was initially used as a microfluidic paper-based analytical device (µPAD) with several advantages over traditional methods, including low-cost, biocompatibility, no-pump sources, and safety. Its appearance was based on society's need for rapid diagnosis technology of diseases. Due to the wanton pollution and destruction of the environment, a variety of new epidemic diseases keep emerging and sweeping the world. Since the beginning of the 21st century, several disease disasters have caused severe trauma to human life, such as the severe acute respiratory syndrome (SARS) in 2003, the H1N1 influenza epidemic in 2009, the Ebola virus in 2014, and the COVID-19 in 2020. The rapid spread of these diseases is mainly due to the lack of fast, effective, sensitive, versatile, and inexpensive diagnostic devices. In this background, paper-based open microfluidics has excellent potential and has experienced rapid development in the last decade (Figure 1b). In the first work, Whiteside's group described a simple method for patterning paper bounded by hydrophobic polymer lines or "walls" to provide spatial control of biological fluids in millimeter-sized channels. This patterned paper possessed the simultaneous detection of glucose and protein in trace urine. In 2008, Li et al. reported a new plasma treatment method to prepare patterned paper-based microfluidic.^[60] This study not only simplified the preparation process but also made it possible to introduce some functional elements to the patterns. Simple preparation methods include wax, inkjet, photolithography, and paper cutting.^[61–64] In addition to rapid diagnosis, μPAD was also used for other detection, such as environmental science. In 2010, Nie et al. first proposed for electrochemical detection of heavy metal ions in liquids such as Pd and Zn ions. These early pioneering works have laid a good foundation for research in this field.^[65]

At the same time, related theoretical research is also deepening.^[66–68] Researchers are concerned with the flow issue of liquids in paper-based porous materials. A significant challenge in creating paper-based open microfluidics devices is achieving robust control of fluid transport. Fluids transport in paper-based porous materials, essentially, is a passive process driven by capillary pressure, $P_{\text{capillary}} = (2)\cos\theta/R$.^[52,69] Here, γ and θ are liquid surface tension and contact angle on solid, respectively, and *R* is the pore radius. Fluids flow rate (*Q*) can be described by Hagen–Poiseuille equation, $Q = \frac{\pi R^4 P_{\text{capillary}}}{8\eta L}$, where η is liquid dynamic viscosity, and *L* is the liquid column length.^[41,56] *L* and time (*t*) can be connected by the Lucas-Washburn equation, which is a classic model shown as follow

$$L = \left(\frac{\gamma R \cos \theta}{2\eta}\right)^{\frac{1}{2}} \tag{1}$$

In addition, Darcy's law, $Q = -\frac{\kappa A}{\mu L} P_{\text{capillary}}$, also can be used for fluid transport in paper-based materials with variational pore sizes, where *A* is the cross-sectional area of the pore, and κ is the permeability of the paper. This shows that capillary force drives fluid transport into the pores, but the viscous force is the resistance force. For a long time, researchers have tried to indirectly control the liquid flow by adjusting physical parameters such as pore diameter, thickness, and width of paper-based materials.^[64,67] As mentioned earlier, this is not an effective, robust control approach. Subsequent researchers have developed 3D paper-based open microfluidic devices to enhance their controllability further and expand their applications. These excellent studies have greatly enriched the field, but complex preparation is a disadvantage.^[67,70]

In recent years, the study of the superwetting surface has undergone significant development. Inspired by nature, some new liquid–solid interaction modes, such as superhydrophobic/ superhydrophilic/superlubricant, have been gradually discovered (**Figure 2**).^[50,54,55,71] Liquids can be wholly spread out on the superhydrophilic surface but well restricted on the superhydrophobic surface. Therefore, the superwetting treatment of substrates was recognized as a good strategy for liquid regulation in open systems. Elsharkawy et al. presented a facile way to fabricate low-cost surface bio-microfluidic devices using a standard household printer on superhydrophobic paper.^[72] Using this

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methodology, regions of varying wettability were achieved by changing the intensity of ink to tune the sliding angles of 10 μ L water droplets in the range from 13° to 40°. Li et al. introduced more extreme wettability states into paper-based open microfluidic devices in 2016.^[73] Localization, manipulation, and transport of all high and low-surface-tension liquids became a reality. It is worth noting that some of the applications proposed in this article, for example, continuous oil– water separation, liquid–liquid extraction, and open-channel microfluidic emulsification, are very meaningful.

However, there needs to be systematic research on superwetting paper-based open microfluidics. Slow driving speed, low selectivity, and material limitation are unavoidable problems of paper-based open microfluidics. It is challenging to build highly controllable open microfluidic devices relying solely on paperbased porous materials. Fortunately, the study of bioinspired superwetting surfaces has injected much new energy into open microfluidics. It allows the research of open microfluidics to break the bound of paper-based materials and provide new possibilities for other materials. However, it raises a valuable question worthy of our deep consideration: can the paper-based open microfluidic treatment methods be applied to the preparation of bioinspired superwetting open microfluidics? Due to the natural hydrophilicity of paper, paper-based microfluidic treatment mainly focuses on constructing hydrophobic/superhydrophobic patterns. Common methods include wax printing. hydrophobic inkjet printing/etching, photolithography, chemical vapor deposition, cutting, etc. These methods benefit from the flexibility or small thickness of the paper.

Therefore, applying it to the preparation of biomimetic open microfluidics is challenging. In terms of structure, bioinspired superwetting open microfluidics have more complex micronano rough structures, for example, lotus leaves' micro-nano mastoid structure, Nepenthes' microcavity with a sharp arcshaped edge, and spider silk's spindle-knots.^[35,37,50,51] These complex 3D structures are difficult to be treated comprehensively by the above inkjet printing/etching, photolithography method because of the shielding of the convex shape. It is more difficult to selectively modify in small areas (microscale), for example, inside microgrooves, so the above treatment methods are difficult to work in small spaces; In terms of materials, wax printing, and hydrophobic inkjet printing/etching are very suitable for paper with good flexibility. However, bioinspired superwetting open microfluidics are generally rigid. So these methods can't be copied directly. In addition, bioinspired superwetting open microfluidics are more focused on superhydrophilic treatment, so the above methods can be helpful (such as photolithography or chemical vapor deposition). The use of photolithography to construct highly bioinspired structures on the microscale is also one of the common technologies in the field of bioinspired superwetting open microfluidics. Chemical vapor deposition is also a mature technology for hydrophobic treatment (fluoridation) of materials with complex 3D bionic structures. It is worth mentioning that commercial plasma treatment is a good option, but it is a challenge to operate in specific tiny areas as well.^[54,57] Therefore, the treatment and construction of bioinspired superwetting open microfluidics will be one of the focuses of the following sections. In general, paper-based microfluidics opens the door to new research fields





Figure 2. Three bioinspired superwetting systems of open microfluidics. a) Superhydrophobic lotus leaf with the high WCA (>150°). b) Superhydrophilic fish scales with the low WCA (\approx 0°). c) Superlubricant pitcher plant with the low sliding friction force. d) Schematic diagram of Superhydrophobic/superhydrophilic open microfluidic channels. The superhydrophobic wall acts as a domain-limiting function. Fluidics is transported as a continuous liquid flow on the superhydrophilic path. e) Schematic diagram of superlubricant/slipper open microfluidic channels. The transport form of open microfluidics on the superlubricated path is discontinuous microdroplets. Reproduced with permission.^[91] Copyright 2009, Wiley-VCH. Reproduced with permission.^[01] Copyright 2018, ACS.

of microfluidics, and the study of bioinspired superwetting surfaces builds a new world behind the door. But the construction and perfection of this new world still need our best efforts.

3. Bioinspired Superwetting Systems

Lotus leaf's unique superhydrophobic nature has long been discovered by human beings. Water droplets can remain perfectly spherical on the surface and roll freely.^[74–76] For a long time, researchers have tried to reveal the mechanism behind this mysterious natural phenomenon. Still, due to the limitations of the equipment, a perfect answer was not given until 1997. With the popularization of scanning electron microscopy, researchers observed that lotus leaves have micron dendrites on their surface, which are covered with a hydrophobic wax layer.^[77] Furthermore, in 2002, Jiang's group proposed that the dual-scale micro-nano composite structure on the lotus leaf surface was the fundamental reason for its excellent superhydrophobicity (Figure 2a).^[78] It can confine air between coarse, multiscale micro-nano structures, forming an air cushion

between liquid and solid. Therefore, multiscale rough structure and low surface energy are two criteria to realize a superhydrophobic surface. In addition to lotus leaves, many species with unique superhydrophobic surfaces in nature, such as anisotropic rice leaves/butterfly wings, highly adherent superhydrophobic rose petals, and water strider legs.^[79-82] These creatures have inspired the design and preparation of artificial superhydrophobic surfaces. With the development in recent decades, many artificial superhydrophobic surfaces/materials have been reported, and a series of new applications have been developed, including self-cleaning, anticorrosion, oil-water separation, etc. However, because of their weak mechanical properties, superhydrophobic materials are not widely used daily.^[83] To our main topic, superhydrophobic surfaces will play an important role in open microfluidics. Water is difficult to spread on the superhydrophobic surface but will be restricted to a particular area.^[84] Therefore, superhydrophobic characters are often used to construct boundaries, or "walls" in open microfluidics to obtain open microfluidics channels (Figure 2d). The second but more important role of superhydrophobic surfaces in open microfluidics is as a platform for liquid manipulation.^[85-87] Using the

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suitable rolling property of the droplet on the superhydrophobic surface, the open space microdroplet transportation, merging and bouncing can be realized. This microdroplet manipulation mode differs from the traditional open microfluidic mode but has gradually become essential. Generally speaking, how to build a high-precision superhydrophobic wall and improve the mechanical properties of the superhydrophobic working platform are the most critical problems in this field.

The superhydrophilic surface is fundamental for the transport ability of liquids in open space. Superhydrophilic surfaces are more common in nature. However, the research on superhydrophilic biomimetic materials started late.^[88–90] The primary reason is that researchers have not found the unique application of superhydrophilic materials for a long time. Fortunately, with the discovery of some superhydrophilic surfaces with unique properties, the study of superhydrophilic materials in open microfluidics has entered a new stage. In 2009, Liu et al. found that superhydrophilic fish scales could keep clean underwater because of their excellent superoleophobicity, which originates from the water-phase hierarchical micro-/nanostructures structures (Figure 2b).^[91] Because of the competition of surface tension, the superhydrophilic water film on the surface can hinder the spread of oil droplets in the underwater environment which provides a new strategy for us to construct the barrier/wall of microfluidics channels under the liquid. Note that underliquid microfluidics can also be considered generalized open microfluidics because of the absence of closed solid tubes. This not only greatly enriched the category of open microfluidics but also promoted the birth of new applications. In 2010, Zheng's group discovered the ability of superhydrophilic spider silk to collect fog.^[92] A unique spindle-knots structure results in a surface energy gradient and a difference in Laplace pressure to achieve continuous directional water condensation. A similar fog collection ability was also found in Opuntia microdasys (cactus) in 2012.^[93] These groundbreaking researches provide a new idea for transporting microdroplets on the 2D open fibers as a new form of open microfluidics transport model. Another classical creature surface with a continuous directional water transport ability is the peristome surface of Nepenthes alata.^[94,95] With the help of superhydrophilic surfaces and unique surface microstructure, microdrop can unidirectional transport on their peristome surface. It is expected to provide new solutions for open microfluidic pumpless transportation.

Nepenthes pitcher plant offers more than insights on liquiddriven models for scientists. Even earlier, it was found that the peristome of the Nepenthes pitcher plant has a unique lubrication property, which helps it capture insects sliding into the interior to get nutrients (Figure 2c). In 2004, the scientific significance of this particular lubrication property was revealed for the first time by Bohn and co-workers.^[95] They found that the thin liquid film bound by the texted structure on the plant peristome surface is the root cause of the extremely low friction resistance on the surface. Therefore, a new method of constructing low-friction and superlubrication surfaces is born. Since then, several research groups have carried out related research work. Aizenberg's group demonstrated an artificial bioinspired slippery liquid-infused porous surface (SLIPS) in 2011.^[96] Varanasi et al. proposed one kind of bioinspired lubricant-impregnated surfaces in 2013.^[97] Similar to the superhydrophobic surfaces mentioned above, superlubrication surfaces with low friction resistance provide a new strategy for constructing open microfluidic channels or platforms (Figure 2e). But it needs to be emphasized that the difference between the superhydrophilic path and the superlubricated path is also very clear. Microfluidics on the superhydrophilic path is usually transported in continuous liquid flow, which depends on passive Laplace pressure difference, capillary force, wettability gradient, etc. It requires constant replenishment of external liquid to maintain its transport. When a liquid passes through the hydrophilic path, the path will be changed into a wet state. However, liquid transport on the superlubricated path is discontinuous microdroplets. Therefore, it is necessary to set the inclination angle or give the initial velocity of the microdroplet with direction to maintain its transport. So, a single microdroplet that meets the conditions can transport continuously. When the microdroplet passes through, the properties of the superlubricated path will not change and the following microdroplets can continue to pass through. This property of the superlubricated path can eliminate the problems of weak mechanical strength and complex preparation of the superhydrophobic surface and has good universality, tolerance, and self-healing abilities. More importantly, the superlubricated surface breaks the limitation of hydrophilic and hydrophobic, which is easy to realize the manipulation of more low-surface energy liquids.^[37] This greatly expanded the scope of application of open microfluidics and generated some new applications, the most important of which is patterned open microfluidics.^[27,30,98] Combination of the superlubricated surfaces with other unique wettability surfaces, such as (super) hydrophilic/hydrophobic or adhesive, in precise micropatterns is relatively easy to implement, leading to entirely new applications including water collection, bioadhesion control, and bioaccumulation.^[87,99-101] Building open microfluidic systems using superlubricated surfaces is a newly emerging research area that requires more effort to explore in the future (Figure 3).

4. Bioinspired Superwetting Open Microfluidics

This section will start with the underlying mechanism of various bioinspired superwetting phenomena and focus on the representative research work in the past few years. Here, we first conducted an objective and qualitative comparison and summary of microfluidics, paper-based open microfluidics and bioinspired superwetting open microfluidics to establish an overall understanding as shown in **Table 1**. In this section, our focus will be the preparation of materials, the exploration of mechanisms, and the expansion of applications. We choose a few inspiring biological examples, including pitcher plants, spider silk, cactus, lizards, etc. This review cannot contain all the research works, but it will summarize the challenges and point the way forward for different research directions.

4.1. Nepenthes-Inspired Open Microfluidics

The unique structure of *Nepenthes* has intrigued researchers for a long time (**Figure 4**a).^[95] However, it is only in recent years that its superlubricating properties have attracted wide





Figure 3. Increase of research interest (number of papers) on the topic of "microfluidics," "open microfluidics," "superhydrophobic," "superhydrophobic," and "superlubricant/slippery" materials. The deadline for statistics is Jan. 2023.

attention (Figure 4b).^[96] Aizenberg's team proposed that three conditions must be satisfied to prepare the bioinspired slippery liquid-infused porous surfaces (SLIPS): 1) The porous substrate where the lubricating liquid can be soaked by capillary action; 2) The substrate must be wetted by the lubricating liquid rather than by the liquid being transported; 3) The lubricating liquid and the liquid being transported are immiscible. The second condition requires the relation between the surface energy of the liquid and the solid to meet two inequality equations shown as follows

$$\Delta E_1 = R \left(\gamma_B \cos \theta_B - \gamma_A \cos \theta_A \right) - \gamma_{AB} > 0 \tag{2}$$

$$\Delta E_2 = R \left(\gamma_B \cos \theta_B - \gamma_A \cos \theta_A \right) + \gamma_A - \gamma_A > 0 \tag{3}$$

Here, E_1 and E_2 are the total interfacial energies of lubricating fluid with and without the transported immiscible liquid. γ_A and γ_B are the surface tensions for the transported and lubricating liquid. γ_{AB} is the interfacial tension at the liquid-liquid interface. θ_A and θ_B are the contact angles of the transported liquid and the lubricating liquid with the solid substrate. R is the roughness factor. To ensure the substrate is wetted preferentially by the lubricating liquid, one should have $\Delta E_1 = E_A - E_1 > 0$ and $\Delta E_2 = E_A - E_2 > 0$. E_A is the interfacial energy of substrate with the immiscible lubricating liquid. Bioinspired artificial SLIPS is generally used to control microdroplets on the open platform to take advantage of its low friction resistance characteristics. As a pioneer in the field, in 2016, Aizenberg's team came up with a bionic strategy based on SLIPS. It achieved a greater volume of water collection compared to other surfaces.^[103] Theoretically, the pitcher-plant-inspired surface can facilitate feedback between coalescence-driven growth and capillary-driven motion on the way down. In Figure 4b, they put out the "adaptive surfaces" concept.^[104] The slippery liquid in a nano porous elastic substrate can achieve a smooth and defect-free surface to roughen through a continuous range of topographies. In 2018, her group also introduced a ferrofluid-containing liquid-infused porous surface (FLIPS).^[105] Several interesting functions were demonstrated, especially microdroplets manipulation on FLIPS, such as release and coalescence of droplets.

The unidirectional water-transport ability of the slippery peristome of Nepenthes was demonstrated by Chen et al. in 2016.^[94] This discovery provides a new way to think about the driving mechanism of open microfluidics transport. A two-order hierarchy of parallel microgrooves on the peristome of Nepenthes is considered to be the main reason for the realization of unidirectional transportation. Anisotropic overlapping and archshaped microcavities with the tops of the arches pointing toward the outer side are distributed along the second-order microgrooves. Each closed microcavity has a sharp arc-shaped edge forming a gradient wedge angle side. In the inside direction, the spreading of water is prevented by the pinning effect of sharp corners. In the outside direction, the overlapping filling of microcavities produces continuous water transport.^[106] In the water filling a single microcavity process, the gradient Taylor rise, as enhanced capillary rise, results in rapid liquid flow due to the gradient wedge angle in the microcavity. The water spreads rapidly along the wall and finally meets in front of the microcavity.^[107]

After the intrinsic mechanism was revealed, researchers tried to prepare the bionic artificial pitcher plant structure by various methods to realize the spontaneous and directional microfluidic transport in open space (Figure 4c). After six years of development, some artificial preparation methods have been put forward successively. The first method proposed is the replica molding method. The poly(dimethylsiloxane) (PDMS) is poured on the surface of the peristome of Nepenthes directly.^[94] By two steps of replication, the artificial peristome was obtained. This method can obtain the artificial microstructure, which is highly consistent with the peristome structure. Still, it cannot be prepared in a large area, and the process is very complicated. Photolithography is also involved in the preparation process, which requires specific equipment and a relatively high cost. At present, the most widely accepted and used method is 3D printing. Researchers can use computer software to model the structure of the peristome and then choose the appropriate material to print directly. However, the accuracy of this method is limited by the state of the art of 3D printing technology. The obtained structure is often more significant than that of the peristome. The smaller system can be obtained by a two-step

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Table 1. A summary and comparison of some recent representative researches of closed microfluidics, paper-based open microfluidics, and bioinspired superwetting open microfluidics.*.

Туре	Common material	Fabrication methods	Posttreatment	Cost	Large scale production	Controllability	Ref.
Closed microfluidics	PE fiber, Nylon 6	Twist insertion, annealing	No	Low	Feasible	Low	[3]
	Amorphous silica nanopowders	Polymerization, thermal, debinding, sintering	No	High	Low probability due to complex preparation	Low	[16]
	Si wafers	Laser lithography, multiple baking	Fabrication of e-gates	High	Hard	High but complex	[18]
	PVDF	Electrospinning	Infuse low-energy liquids	Moderation	Feasible	Low	[29]
Paper-based open microfluidics	ITW Technicloth wipers, Double-sided carpet tape	Photolithography	Oxygen plasma	High	Hard	High	[42]
	Nitrocellulose membranes, PMMA	CO ₂ laser	No	low	Feasible	High	[56]
	Filter paper, alkyl ketene dimer	Dip and heated	Plasma	Low	Feasible	Low	[60]
	TiO ₂ nanoparticles, PTFE, Paraffin	Spraying	UV treatment	Moderation	Feasible	Low	[64]
	Conductive carbon ink, paper or polyester film	Photolithography or wax Printing	No	Moderation	Feasible	Low	[65]
Bioinspired superwetting open microfluidics	Resin	3D printing	No	Low	Feasible	High	[35]
	Resin	3D printing	Oxygen plasma	Moderation	Feasible	High	[37]
	Nylon fiber, PMMA/DMF-EtOH	Immersing and drawing out	No	Low	Feasible	Indeterminate	[92]
	PDMS	replica molding	Oxygen plasma	Low	Low	High	[94]
	Conical copper wire	Electrochemical corrosion	Incrementally submerged in 1-dodecanethiol	Moderation	Low	Indeterminate	[117]
	PDMS	Lithography	Oxygen plasma	High	Low	High	[119]

replication process, printing the antitemplate, and utilizing the shrinkage characteristics of the gel for replication. Dong's group has made outstanding contributions to this field. They fabricated a peristome-mimicking surface via a high-resolution 3D printing method.^[37] The detailed overflow-controlled unidirectional transportation mechanism was discovered by X-ray microscopy. The open microfluidic directional transport on the elevated spiral was achieved (Figure 4c1). Water flows upward and climbs along the curve without retraction at the rear side. Take advantage of this mechanism, they reached programmed pressure-controlled Inchworms-like liquid strip directional motion (Figure 4c₂).^[108] They reported a "plug-and-go" separation model of water-oil microdroplets in open space^[109] Microliter water-in-oil droplets were separated into pure water and oil droplets on curved peristome-mimicking surfaces without energy input. Furthermore, they also developed a new timedependent switchable open microfluidics.^[110] The synergistic action of the surface curvature and the surface microtextures was utilized to manipulate the switchable transport performance. Recent research has also proved that Nepenthes alata offers a remarkably integrated system on its peristome surface to harvest water continuously in a humid environment. Then Dong's group designed an artificial multicurvature water harvester taking advantage of the surface-gradient-induced Laplace pressure at the ratchet and concavity (Figure 4c₃).^[111] As an emerging research field, two problems deserve our attention. First, advanced 3D printing technology has been developed to produce more accurate biomimetic microstructures. The second is how to develop new applications of open microfluidics based on directional transport.

Most of these studies were inspired by the slippery Peristome of Nepenthes with Asymmetrical microstructures. In a recently published study, Dong's group proposed a new open microfluidic model inspired by the tendrils of pitcher plants.^[112] The researchers observed that Nepenthes could achieve fast water drainage in bulk through its metamorphosed leaf, combining 1D tendrils and 2D/3D board leaf structures in one system. It can quickly drain rainwater from the leaves through the tendrils, acting as a diversion to prevent rainwater from falling into the pitchers. The fundamental reason is that the tendril has a stripe-like cellular morphology along the longitude direction (Figure 5a).^[112] Therefore, a novel open-air microfluidic transport strategy with multimicrogrooves and cellular structures has been developed. Bioinspired microwick with a different number of microgrooves was prepared through the 3D printing method. The



NEPENTHES PITCHER



Figure 4. Peristome of *Nepenthes-inspired* open microfluidics. a) Nepenthes pitcher and peristome morphology. It has a special multiscale and ordered microgroove structure. b) Schematics showing the fabrication of a SLIPS by infiltrating a functionalized porous/textured solid with a low-surface energy, chemically inert liquid to form a physically smooth and chemically homogeneous lubricating film on the surface of the substrate. The picture below is schematic diagram of the mechanism of the "adaptive surfaces," and optical photos of dynamic droplet control experiment. c) The application of the Peristome of *Nepenthes* inspired open microfluidics in continuous directional transport. c₁) The open microfluidic directional transport on the 3D printed elevated spiral. c₂) Programmed pressure-controlled Inchworms-like liquid strip directional motion. c₃) Artificial multicurvature water harvester device. Reproduced with permission.^[95] Copyright 2004, PNAS. Reproduced with permission.^[96] Copyright 2011, Springer Nature Limited. Reproduced with permission.^[104] Copyright 2013, Springer Nature Limited. Reproduced with permission.^[108] Copyright 2020, Wiley-VCH. Reproduced under the terms of the CC-BY license.^[111] Copyright 2020, The Authors, published by PNAS.

surface microgrooves can confine the liquid to form a liquid film, thus inhibiting the Rayleigh–Plateau instability from forming large droplets and realizing ultraefficient transport of microfluidics in closed-open-closed space (Figure 5b). The experimental results show that water and ethanol transfer efficiency is up to 98% and 97% based on this open microfluidic transport mechanism. Furthermore, applications of open-air microfluidic, polydirectional dynamic guided transport, multisegment closed–open–closed microfluidic transport devices, and open-air direct writing 3D printing have been developed (Figure 5c). This study provides a brand-new approach to open microfluidic transport mode and has excellent potential for drug delivery, chemical synthesis, microfluidic chip, and additive manufacturing applications.

4.2. Desert Beetle-Inspired Open Microfluidics

Parker and Lawrence found an interesting phenomenon in the Namib Desert in 2001. Some beetles can collect drinking water from the fog-laden wind on their backs. They have shown that these large droplets form by the insect's bumpy surface, which consists of alternating hydrophobic, wax-coated, and hydrophilic, nonwaxy regions (**Figure 6**a).^[113] The droplet generation

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Figure 6. The surface microstructures, mechanisms, and process of microdroplet directional transport on a) desert beetle, b) spider silk, c) cactus spines, and d) *Sarracenia* trichome. Reproduced with permission.^[112] Copyright 2001, Springer Nature Limited. Reproduced with permission.^[93] Copyright 2012, Springer Nature Limited. Reproduced with permission.^[119] Copyright 2018, Springer Nature Limited.

system works by "growing" water droplets on hydrophilic seeding sites in peaks. Each attached droplet eventually reaches a size where its contact area covers the entire hydrophilic island. Beyond this size, the ratio of its mass to surface contact area increases rapidly until it overcomes the capillary forces attached to the surface. This 2D curvature-wetting synergy enables the directional transport of several droplets in an open space for artificial water collection.^[114] However, this bioinspired design severely limits microfluidics' transport speed and distance, so it has not been widely used.

4.3. Spider Silk/Cactus Spines-Inspired Open Microfluidics

The asymmetrical cone-shaped substrate induces directional transport of microdroplets on spider silk or cactus spines (Figure 6b). It, therefore, can be seen as 1D open microfluidics. The driving force arises from the gradient of Laplace pressure (Figure 6c).^[92,93,115] For example, the driving force (F_L) microdroplet on a size-matched nonstructured substrate can be expressed as follows

$$F_L \sim -\int_{R_S}^{R_L} \frac{2\gamma}{(R+R_0)} \sin \alpha dz \tag{4}$$

where γ is the surface tension, R_L and R_S are the radii of the substrate at two side of the microdroplet, respectively, R and R_0 are the radii of cone-shaped substrate and microdroplet, and α is half apex angle. In addition, the surface tension gradient

caused by the variable chemical composition of the 1D conical surface can also act as the driving force (F_C) of the microdroplet transport

$$F_C \sim \pi R_0 \gamma (\cos \theta_b - \cos \theta_a) \tag{5}$$

where θ_a and θ_b are the contact angles at two sides of the microdroplet, respectively. However, the microdroplet transport is subjected to resistance force, mainly caused by the contact angle hysteresis (friction resistance of superhydrophilic surface is very small). The contact angle hysteresis is the difference between the advancing contact angle (θ_{Adv}) and receding contact angle (θ_{Res}), which can be abstained by the equation

$$F_{R} \sim \pi R_{0} \gamma (\cos \theta_{Res} - \cos \theta_{Adv}) \tag{6}$$

Directional transports of microdroplets on spider silk and cactus spines have a similar driving mechanism, but the processes are different (Figure 6c). Specifically, the wet-rebuilt phenomena of spider silk can be found in forming periodic spindle knots and joint structures. The tiny droplets will first deposit on their surface, and the tiny drops can transport toward the spindle-knots position directionally to merge into microdroplets.^[92,93] The mentioned Laplace pressure and surface energy gradients dominate the result. As to cactus spines, microdroplet can move to the middle position with a larger curvature radius by Laplace pressure difference. In this process, the oriented barbs on its tip can supply anisotropic contact angle hysteresis to facilitate movement. The grooves in the middle position of

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spines cause the roughness gradient and wettability gradient. So, the microdroplet will move to the base and be absorbed.

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Based on understanding this transport principle, various spider silk-inspired, and cactus spine-inspired artificial open microfluidic water collection fiber or needle structures have been reported with excellent absorption and transport abilities. Bai et al. prepared a string of polymer spindle-knots on the nylon fiber due to the Rayleigh instability.^[116] Microwater drops can be transported by the cooperation of curvature gradient, chemical, and roughness gradients on this spider silkinspired fiber surface. This dip coating method to fabricate spindle structure is convenient with widely applied potential. In 2017, Wang's group fabricated microfibers with unique spindle cavity knots from the jet phase with gas bubbles.^[99] This cavitymicrofiber shows unique mechanical strength and long-term durability due to the design of the cavity, thus enabling an outstanding performance of water collection. The preparation of the artificial cactus spines-inspired water collector has to figure out how to get the microcone with a structure gradient. In 2013, Ju et al. prepared a conical copper wire (CCW) with increasing wettability from tip to base through the gradient electrochemical corrosion method.^[117] This gradient design guarantees a high drop growth rate on the hydrophobic tip and the quick transportation of the drops due to the Laplace pressure gradient, allowing efficient fog collection. Recently, Zheng's group proposed a dual-bionic strategy based on the principles of spider silk and cactus spines. Hydrophilic zinc oxide (ZnO) on the original 3D fiber web can capture fog droplets and generate coalesced droplets making droplets transport efficient due to the Laplace pressure difference. The water film on the fiber web can break up into droplets relating to Rayleigh instability.^[118]

In general, spider silk and cactus spines provide new insights into bioinspired open microfluidics' transport patterns and driving mechanisms. However, it has a lot of limitations. The most important two are the limited transport distance and the slow transport speed, which significantly restricts its range of use. Beyond fog collection, researchers have yet to find a better outlet for applications. Therefore, there is still much room for development in this research field.

4.4. Sarracenia Trichome-Inspired Open Microfluidics

As mentioned above, in the bioinspired models of spider silk/ cactus spines, the transport speed of open microfluidics is a considerable limitation. Achieving fast microfluidic transport on such an open conical structure is a hot issue. The research by Chen et al. on Sarracenia trichome brought light to solving this problem. An ultrafast water transport process on the surface of a Sarracenia trichome was discovered. Its transport velocity is about three orders faster than those measured in spider silk/cactus spines.^[119] The high speed of water transport is attributed to the unique hierarchical microchannel organization of the trichome (Figure 6d). Regarding mechanism, two types of ribs with different heights regularly distribute around the trichome cone, which constructs the unique open microfluid channel. The two adjacent high ribs form a large channel that contains a few low ribs that define smaller base channels. This structure gives rise to two continuous but distinct modes of water transport. Briefly, a rapid thin film of water is formed inside the base channels (Step I), followed by ultrafast water sliding on top of that thin film (Step II). However, preparing such a hierarchical microchannel structure is difficult, so there are few studies on bioinspired *Sarracenia* trichome open microfluidics. Although the structure improves the transmission speed of open microfluidic, the limitation of the transport distance is not well solved.

4.5. Lizard Skin-Inspired Open Microfluidics

The survival of plants and animals in arid areas depends on the availability of adequate water. In 2015, scientists discovered that Texas horned lizard could use their unique skin texture to suck water from the ground and deliver it to their mouths.^[120] The mechanism behind this phenomenon was revealed: the opening of periodically asymmetrical microchannels on the skin surface can provide sufficient periodical Laplace pressure for the fluid to drive its passive, continuous, and stable transport in a certain direction (Figure 7a).^[121,122] This phenomenon is also known as the liquid diode. Transport smaller amounts of water via capillary skin channels seems to be a selfadaptation arid habitat of lizards. This microchannel structure on the skin can be reduced to a periodic asymmetric saw-tooth shape model. The liquid in such microchannel will develop a meniscus at each accessible liquid-air interface, which provides a positive pressure difference ΔP across the liquid interface, which can be described by the Young-Laplace equation

$$\Delta P = \gamma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) \tag{7}$$

where γ is the surface tension, R_1 and R_2 , are the orthogonal radii of curvature. In this model, $R_2 \rightarrow \infty$, so, $\Delta P = \frac{\gamma}{R_1}$. The value of R_1 can be calculated by the geometrical relations of liquid and microchannel: $R_1 = d(x)(2\cos(\theta - \alpha))^{-1}$, where d(x) is the capillary width at position *x*. Equation (8) can be rewritten as

$$\Delta P = 2\gamma \cos(\theta - \alpha) d(x)^{-1}$$
(8)

A concave curvature of the liquid–air interface provides driven force for unidirectional transport along the direction of narrowing, whereas a convex curvature will retard the transport. Therefore, the front and back curvature difference can determine the liquid transport direction.

It is not difficult to prepare a microchannel with the asymmetric gradient on a 2D surface. Buchberger et al. reported a large-scale, fast, low-cost CO₂ laser engraving method to obtain the scale-like poly (methyl methacrylate) capillary channel network. The author made a comprehensive study on the influence of various factors on transportation performance (Figure 7b).^[122] They also accessed the fluid transport performance via distance, velocity, wetted area, and flow asymmetry aspects. This work is very instructive for future research, especially in the structural design of lizard skin-inspired open microfluidic devices. The fabrication of 2D bionic lizard skin microgrooves on flat substrates is not difficult. But offering enough power to open



LIZARD SKIN



Figure 7. Lizard skin inspired open microfluidics. a) Optical photograph of Lizard and open microfluidic transport on the Lizard skin. A colored water droplet is transported on the dorsal integument and the principle of "interconnection" for two saw-tooth-shaped capillary channels. b) Fabrication and the transport process of a Lizard skin inspired open microfluidics in poly(methylmethacrylate) (PMMA) plates by laser etching. c) Artificial Lizard skin-inspired open microfluidic surfaces with the asymmetric arc-shaped A-shaped island arrays prepared by 3D printing method. Dyed liquids can transport directionally on this Lizard skin inspired open microfluidics. Reproduced under the terms of the CC-BY license.^[120] Copyright 2015, The Authors, published by The Royal Society. Reproduced with permission.^[122] Copyright 2018, Elsevier. Reproduced with permission.^[35] Copyright 2018, ACS.

liquid transport to achieve long-distance and continuous unidirectional transport is hard. In order to solve this problem, Si et al. developed the 3D micro/macro dual-scale arrays via the 3D printing.^[35] Using the periodic Laplace pressure difference,



liquids can rapid, spontaneous, and continuous unidirectional transport along the sloping direction of arrays (Figure 7c). The authors systematically explored the relationship between liquid unidirectional transport performance, surface morphology parameters, and surface wettability. Significantly, they developed new applications such as liquid uphill running, microfluidics patterning, liquid shunting, and free combination transport in open space without external energy input. This study creates a new strategy of utilizing Laplace pressure difference to drive liquid and develops a new transport mode of bioinspired open microfluidics.

4.6. Araucaria Leaf-Inspired Open Microfluidics

Liquids' unique directional transport phenomena across animal and plant surfaces have led to a significant

development in open microfluidics. Researchers are also becoming more enthusiastic about exploring nature and making breakthroughs. Inspired by the double cantilever structure of the horizontal and longitudinal curvature of the Araucaria leaf, Wang's group reported a 3D-printed bioinspired superwetting open microfluidic system in which the liquid can choose its spreading direction independently (Figure 8a).^[123] The mechanism is that the submillimeter 3D capillary ratchets can induce asymmetric 3D solidliquid interface interactions to regulate the spread patterns of fluids with different surface tensions. The phenomenon of oil and water spreading in opposite directions without external field stimulation and morphology change has been reported for the first time and immediately triggered a strong response. This study differs from previous studies and provides intelligence for exploring the new application of open microfluidics.





Figure 8. Araucaria leaf and butterfly probosci's inspired open microfluidics. a) Araucaria leaf-inspired open submillimeter 3D capillary ratchets to regulate the spread direction of microfluidics with different surface tensions. b) Butterfly proboscis inspired open multilevel porous probe for detecting minute amounts of liquid. An electric field can operate the probe. Reproduced with permission.^[123] Copyright 2010, AAAS. Reproduced with permission.^[124] Copyright 2013, The Royal Society. Reproduced with permission.^[124] Copyright 2012, The Royal Society. Reproduced with permission.^[128] Copyright 2011, Royal Society of Chemistry.



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4.7. Butterfly Proboscis Inspired Open Microfluidics

The butterfly proboscis is the feeding device for the insects to acquire fluidic food from the environment. It was traditionally considered as a drinking straw, with the assumption that the insect depends only on the action of the sucking pump in its head to acquire liquids. However, a recent study revealed the nanopores and slits structure on the dorsal and ventral linkages along the length of the proboscis, changing the system from a closed tube to an open microfluidic system (Figure 8b).^[124] The hydrophobic-hydrophilic dichotomy on the outer surface of proboscis propels the liquid to converge at the hydrophilic pores and slits regions. Eventually, the gathered liquid is sucked into the microsize food canal under the suction force from the pump and capillary force from the pores and slits.^[125] With the coiling and uncoiling flexibility of proboscis, the butterfly uses such a natural open microfluidic device to acquire fluidic food from diverse sources.^[126] Besides acquiring microfluidics from the environment, the proboscis is also a device for butterflies to release saliva for assembly and repair the separated proboscis or cleaning after feeding.^[127] Inspired by the butterfly proboscis, Tsai et al. fabricated the multilevel porous polyvinylidene fluoride (PVDF) fiber bundles through electrospinning to probes for a minute amount. Taking advantage of the electroconductibility of PVDF, the flexible PVDF fiber bundles can be manipulated by an electric field to detect the micro drops precisely. And the multilevel porous structure provides fast absorption rates and good retention capacity.^[128] More unusual behaviors and abilities of butterfly proboscis to manipulate microfluidics are expected to be discovered, and more bioinspired open micro-fluidics is to develop accordingly shortly.

4.8. Spore Emission-Inspired Open Microfluidics

Many fungi in nature rely on spore emissions to reproduce. The mechanism behind it is microscopic and complex. Researchers generally believe that electrostatic repulsion and the release of surface tension dominate the process (**Figure 9**a).^[129] Inspired by this, Li et al. realized the manipulation of tiny droplets in space, including ballistic launching and bouncing on a superhydrophobic surface by using the electrostatic repulsive force (Figure 9a).^[85] Specifically, the static electricity emitter can ionize the surrounding air. The microdroplet can acquire the same polarity when the ions collide with the drop. Then, the negative charges rearrange and gather at the top leading edge of the water/air interface. There are several factors to affect the mean amounts gained by drops, including the size of drops and so on. The forces on the droplet detached from the substrate are analyzed

$$M\frac{d\nu}{dt} = -6\pi\eta R\nu + qE_1 - Mg\cos\alpha \tag{9}$$

where *M* and *q* are the mass and charge of microdroplet, respectively. η is the viscosity between the liquid and the air, ν is the velocity, and *R* is the droplet's radius. *g* is the gravity acceleration, α is the angle between the electrostatic line and



Figure 9. Three typical bioinspired examples and the liquid transport processes on the corresponding artificial bioinspired superwetting open microfluidics device. a) Spore emission, b) water strider, and c) butterfly wing-inspired open microfluidic systems. Reproduced with permission.^[129] Copyright 2009, The Company of Biologists Ltd. Reproduced with permission.^[85] Copyright 2018, Wiley-VCH. Reproduced with permission.^[132] Copyright 2013, PNAS. Reproduced with permission.^[133] Copyright 2019, ACS.

the vertical direction. A new antifreezing strategy is developed based on the electrostatic manipulation of liquid drops. The electrostatic acting time for a microliter supercooled drop jumping is too fast to ice.

In 2019, Dai et al. further improved how to manipulate microdroplets with static electricity.^[86] First of all, the authors realized the use of electrostatics to control the high-speed motion of microdroplets in any direction in the 2D superhydrophobic plane. Meanwhile, out-of-plane electrostatic charging can achieve controlled dynamic switching between the onset (moving) and offset (pinning) states. Based on this model, open-air microdroplets-merging behavior and directional chemical microreaction were achieved. Finally, a complex droplet motion, in a "snooker" style, was reported by adjusting the droplets' charge type and wetting state.

In the previous research on open microfluidic droplets, most methods rely on the ultralow adhesion force of the superhydrophobic surface. So, they have limitations in space and velocity. Recently, Sun et al. successfully realized the manipulation of microdroplets in 3D space.^[130] They demonstrate a high-velocity and ultralong transport of droplets driven by surface charge density (SCD) gradients printed on any substrates created by impacting water droplets. Interestingly, droplets with different radii can move upward on a vertically placed SCD gradient superamphiphobic surface. The transport velocity ν scales of microdroplets can be described as follows according to Newton's second law

$$\nu \sim (\varepsilon_r - 1)\varepsilon_0 k \tag{10}$$

where ε_r is the relative dielectric constant of the liquid, ε_0 is the dielectric constant of empty space, and k is the SCD gradient imposed on the moving microdroplet. It is worth mentioning that microdroplets as the car wheel for spontaneous transport of solid cargo on SCD gradient superamphiphobic surfaces are an excellent practical application.

Driving droplets by electrostatic interaction is an innovative method to open microfluidic droplets' manipulation. It leads to a significant breakthrough in motion speed and inspires many new applications. But it is still not a highly controllable strategy for now. The direction of droplet motion and the control of droplet velocity have certain randomness. However, a broad space remains to explore as an emerging research field.

4.9. Water Strider Inspired Open Microfluidics

The natural mechanism of a water strider's ability to walk quickly on water (Figure 9b) thanks to its micro-nano composite legs consisting of inclined tapered setae with quasi-helical nanogrooves has long been revealed.^[82] However, another interesting phenomenon is that the directional transport of a microdroplet within the water strider's striated tapered setae is not well known. The fundamental reason is the stiffness gradient caused by uneven thickness. The microdroplet in the soft elastic nano-micro scale setae substrate with stiffness gradient is subject to the competition between stiffness effect and capillary force. Under the rigid substrate's adhesion, the droplet's triple-phase contact line will deform into an asymmetric ridge shape, resulting in the gradient of the apparent contact angle. The apparent angle gradient can provide the driving force for the transport of the liquid. This process was studied in detail by Wang et al. in 2015.^[131] The unique textures of the water strider's leg were successively exploited as a water condenser, starting from self-penetration, sweeping effect, and individual tapered setae. The diagram shows that the transport direction of the microdroplets is opposite to that of the conical cactus spines. The expelled microdroplets coalesce with each other, creating a release of surface tension that further fuels the droplets' movement (Figure 9b). Although it provides a new driving mode for open microfluidics, it is not highly controllable. Another intriguing biological phenomenon is the durotaxis of cells on stiffness gradients substrate. The unresolved mechanisms are generally believed to involve active sensing and locomotion. Style et al. found that microdroplets undergo durotaxis on a 2D soft flat substrate with stiffness gradients. Although the contrary transport direction of microdroplets on the water strider, the driving force can also be attributed to the rise of the apparent contact angle. The durotaxis on the 2D soft flat substrate can be used to create large-scale open microfluidics patterns and microfabrication (Figure 9b).^[132]

4.10. Butterfly Wing-Inspired Open Microfluidics

In humid natural environments where temperatures vary greatly, water vapor can condense on the surface of superhydrophobic organisms' characters into tiny microdroplets on nanoscale nucleation sites. The droplets formed in this way are generally trapped in the microstructure of the biological surface, presenting a high adhesion Wenzel state with large contact hysteresis. But as the microdroplets grow, the neighboring microdroplets touch each other and coalesce into a larger droplet. During this process, a conversion from surface energy to kinetic energy occurs. The surface-to-kinetic energy conversion can act as the driving force to achieve microdroplets' self-propelled directional transport. The amount of the released energy and transport direction is relevant to solid surface morphology and the size of the microdroplets.^[133] Assuming two microdroplets with the same radius, R_0 , the released energy can be measured by Equation (11)

$$two\Delta E = \gamma \pi \left(2 - 2^{2/3} \right) R_0^2$$
 (11)

where γ is the surface tension of the liquid. The kinetic energy gained by the coalescence process of the microdroplet overcomes the adhesion between liquid and solid. For an ordinary superhydrophobic surface, the transport direction of the condensate microdroplet is perpendicular to the surface. For asymmetrical anisotropic superhydrophobic surfaces, microdroplets will be transported directionally. The anisotropic structure of butterfly wings was revealed as early as 2007 (Figure 9c).^[81] A droplet easily rolls off along the radially outward direction of the body's central axis. Still, it is prevented from rolling in the opposite direction. This directional adhesion phenomenon is caused by the unique overlapped scales with porous asymmetric ridges covering the butterfly wings. The asymmetric growth and coalescence of microdroplets on



the micro/nano ratchet-like structure of butterfly wings will result in its directed transport. There have been many reports about microdroplets coalescence on superhydrophobic surfaces, but researchers have focused more on energy conversion than directional control.^[134,135] In 2016, Liu et al. designed a microanisotropic superhydrophobic surface consisting of periodic isosceles triangle microarrays.^[136] This surface enables microdroplets to self-powered jumping along relatively long-distance and directional transport. A liquid bridge forms between two coalescing microdroplets, subsequently growing and impacting the substrate. During the retraction process, the coalesced microdroplet displays an asymmetric shape leading to asymmetric and unbalanced pinning force, resulting in a net lateral momentum of the coalesced microdroplet during the leaping process. The coalescence and jumping of microdroplets also commonly occur on other surfaces, such as cicada's wings or drain flies. How to apply this uncontrollably driven mode to open microfluidic systems is an important problem.

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4.11. Other Natural Phenomenon-Inspired Open Microfluidics

Besides the most representative natural creatures inspired open microfluidic concepts mentioned above, researchers never stop exploring the unrevealed unique and fascinating natural phenomenon. Fortunately, some remarkable discoveries have been made in recent years. In a recent new study, scientists have revealed that sharpshooter insects (Cicadellidae) can remove excreta (microdroplets) at up to 30 cm s⁻¹ in the open environment. The essence of this superpropulsion phenomenon is that these insects temporally tune the frequency of their anal stylus to the Rayleigh frequency of their surface tension-dominated elastic drops as a single-shot resonance mechanism. Although this novel research has not vet realized the bionic design of artificial open microfluidic devices, this energy-saving and efficient open microfluidic transport mode is bound to bring breakthroughs from self-cleaning structures and soft engines to generate ballistic motions (Figure 10a).^[102] In the vertical direction, Duoss's Group proposed a new bioinspired concept of cellular fluidics inspired by the water transport system under the effect of plant transpiration (Figure 10b).^[137] They realized the modeling of programmable fluid flow and a series of fluid processes, such as the deterministic control of multiphase flow, transport, and reaction processes. Microfluidic flows in these structures can be "programmed" by architectural designs of cell types, sizes, and relative densities, enabling engineering structures to be implemented in laboratory and industrial processes. Dong et al. proposed a new concept of hydrobot (Figure 10c), which realized the manipulation of microdroplets in 3D space.^[138] Using superhydrophilic iron beads as micro motors, the microdroplet can start and brake quickly and program movement in a 3D space orbit using solid-liquid adhesion. Inspired by the escape behavior of fish on a lotus leaf, they developed a new cargo delivery application based on microdroplet systems. These groundbreaking and novel works have shown the potential of open microfluidics in interdisciplinary applications.[34,139-141] The main studies in this section are summarized and compared in Table 2.

The applications of open microfluidics are more concentrated in the biochemistry detection and analysis field, and some professional, comprehensive reviews on this aspect have been reported.^[12,24,40,41,44] But for bioinspired superwetting open microfluidics, researchers focus more on their potential applications in fog collection and directional transport. We have introduced some of the representatives' works in front. In fact, according to our summary of the recent literature, several new applications are taking root and growing in this newborn field.

5.1. Superhydrophobic and Superhydrophilic Synergy

The most extensive and important roles of bionic superhydrophilic and superhydrophobic surfaces in microfluidics are to build efficient and energy-saving microfluidics channels via their synergistic effect. The superhydrophilic microchannel can be used as a capillary pump to provide power for the transport of microfluidics. In contrast, the superhydrophobic region can be used as an invisible wall to confine the flow range of microfluidics and plays a vital role in reducing resistance. This approach is first applied to closed-pace microfluidic transportation. In 2000, Handique et al. constructed a nanoliter measuring device by introducing a hydrophobic patch into a hydrophilic microchannel.^[45] In open microfluidics, the absence of closed pipelines can effectively reduce the resistance of liquid transport but also weaken the effect of capillary force. Therefore, most early studies can only realize the asymmetric spread of microdroplets with slow velocity and short distance.

Ghosh et al. achieved a large volume of repeated droplet transport via the combination of wedge-shaped patterns and photoinduced wettability transformation of TiO₂ in confined tracks on open-air devices, even when acting against gravity.^[47] The authors discussed the optimization between rapid transport rate and distance. They proposed attractive liquid transport models involving fluid metering, merging, and dispensing by patterning spaced parallel wedge tracks. To extend the transported liquid to the lowest surface energy, Morrissette et al. reported a nanocomposite coating comprised of fluorinated silica as filler, a perfluoroalkyl methacrylate copolymer as the binder, and fluorinated polyhedral oligomeric silsesquioxane to repel liquids with low surface tension.[48] Regions of the laser-treated coatings become the superoleophilic pattern. Low-surface-tension microfluidics can be pumpless transported in open air by harnessing forces arising from spatial confinement. The biggest problem facing the bionic superhydrophilic and superhydrophobic co-regulated open microfluidic system is realizing rapid and continuous transport over long distances without a pump. Periodic microscale structures would be a solution, but coordinating different wettability is just as necessary. In addition, introducing a superhydrophobic valve device into an open space to control microfluidics transport is also a significant challenge. The surfaces with changeable wettability under an external field stimulus would be good choices.



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Figure 10. Other nature-inspired open microfluidics. a) Sharpshooter insects exhibit ultrafast droplet excreta catapulting. b) Cellular fluidics inspired by the water transport system under the effect of plant transpiration. Microfluidic flows in these open structures can be "programmed." c) Inspired by the escape behavior of fish on a lotus leaf, the microdroplet as a hydrobot can be manipulated in a 3D space orbit. Reproduced with permission.^[102] Copyright 2023, Springer Nature Limited. Reproduced with permission.^[137] Copyright 2021, Springer Nature Limited. Reproduced with permission.^[138] Copyright 2021, Elsevier.

5.2. Under-Liquids Transport

The microfluidic transport under the liquid environment can also be classified as generalized open microfluidics because of the absence of a closed solid wall. It has tremendous application potential in liquid catalysis, chemical analysis, extraction, and energy storage. However, a big challenge is constructing spatially oriented invisible walls in a

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Table 2. A summar	y of artificial	bioinspired	superwetting	open microfluidics.
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Bionic object	Main materials	Fabrication method	Surface structure	Wettability	Complexity ^{a)}	Ref.
Nepenthes	Perfluorinated fluids, ordered epoxy-resin, Teflon	Liquid-infused coating	Ordered/random nanostructured	Slippery	***	[96]
	PDMS, porous Teflon, perfluorinated fluid		Nanoporous			[104]
	Ferrofluid, perfluoropolyether oil		Microchannel substrates			[105]
	PDMS	Replica molding	Microcavity with a sharp arc- shaped edge	Superhydrophilic	*	[94]
	Resin	3D printing, oxygen-plasma-treated	Arch-shaped microcavity		*	[37]
	PDMS, PVA	3D printing		Hydrophobic, superhydrophilic	**	[109]
	PVA	3D printing, Replica molding		Superhydrophilic		[111]
	Resin	3D printing	Microwick with microgrooves		*	[112]
Desert beetle	TiO ₂ particles, FAS	Spin-coating, chemical vapor deposition	Star-shaped patterns	Superhydrophilic, Superhydrophobic	**	[114]
Spider silk/cactus	PMMA, nylon fiber, PVA	Drawing	Spindle-knots	Superhydrophilic	*	[116]
spines	PEG, PVA	Capillary-based microfluidic	Cavity-microfiber	Hydrophilic	*	[99]
	Copper wires	Gradient chemical modification	Conical	Hydrophobic, hydrophilic	*	[117]
	ZnO	Hydrothermal growth	Nanocone, fiber network	Hydrophilic	**	[118]
Sarracenia trichome	PDMS	Llithography	Hierarchical microchannels	Superhydrophilic	**	[119]
Lizard skin	PMMA	CO ₂ laser	Capillary channels		*	[122]
	Resin	3D printing	Micro/macro dual-scale arrays	i	*	[35]
Araucaria leaf	Resin	3D printing	3D capillary ratchets	Hydrophilic	*	[123]
Butterfly proboscis	PVDF, PEO	Electrospinning	Nanopores	Hydrophilic	**	[128]
Spore emission	Tetraethoxysilane, soot	chemical vapor deposition	Re-entrant Nanofilaments	Superhydrophobic	**	[85]
	PDMS	Photolithography, Replica molding	Microscaled pyramids		**	[86]
	Tetraethoxysilane, PFOTS, soot	Chemical vapor deposition	Nanoparticles		***	[130]
Water strider	polyester-resi, silicone gel	Replica molding	Stiffness gradient	Hydrophobic	***	[132]
Butterfly wing	Silicon wafer, FAS-17	Lithography	Anisotropic micropatterns	Superhydrophobic	**	[136]
Plant transpiration	Resin	3D printing, plasma-treated	Unit-cell-based, 3D structures	Hydrophilic	*	[137]
Fish escape	SiO ₂ , TiO ₂	Coating	SiO ₂	Superhydrophobic, superhydrophilic	*	[138]

^{a)}The more stars, the more complex the fabrication process.

fluid environment to achieve the coupling of function and direction.

To realize the microfluidic transport under the liquid environment, two primary conditions must be satisfied: one is to construct a differential wetting surface under the liquid, and the other is to provide the asymmetric driving force by the microstructure of a substrate. For example, in 2016, Huang et al. reported a surface-tension-driven spontaneous pumpless transportation (SPT) of liquids.^[142] The SPT system can achieve various nonpolar organic liquids transport unidirectional underwater by extreme wettability patterns. The capillary force provided by the wedge-shaped superoleophilic track is the main driving force. Compared to the air environment, the open microfluidic transport using this wedgeshaped structure under the liquid environment would face more complex forces, such as buoyancy and liquid resistance. As a result, microfluidic transport under a liquid environment is challenging to have an excellent directional ability. Zhou et al. utilized the bioinspired peristome of pitcher plant structure, which has an exceptional directional ability in the air, to attempt directional liquid transport under the liquid. The directional microfluidic transport of oil in water and underwater is realized using the same structure with different materials (**Figure 11**a).^[98] With the help of buoyancy, 3D microfluidic transport, which is difficult to achieve in the air environment, was achieved under liquid conditions. The authors also proposed an exciting transport method for the closed path of the Mobius ring. This work helped inspire exploring the advantages of a liquid environment transport and designing a spatial liquid control.





Walsh et al. creatively proposed that under-liquid microfluidics can be achieved without the help of the morphology of the substrate. Still only by the difference in the surface tension of the two liquids (Figure 11b).^[26] They proposed a Freestyle Fluidics concept, and the aqueous circuits with any 2D shape were printed on superhydrophilic substrates in seconds. The liquid channel under the liquid was constructed utilizing the pinning effect of the three-phase contact line to maintain the shape and then covering the surface with nonvolatile low-surface tension liquid for protection and limitation. The droplets added to the microfluidic path were driven by Laplace force and hydrostatic pressure. The injection or reduction of liquid would not move the solid-liquid-liquid contact line and change the shape of the microfluidic path. This method can also be used for micropatterning under structural fluid, mainly for cell biology. Yu et al. used the directional movement of microfluidics in the air to control the microdroplets in oil flow.^[25] In addition to the differences in surface tension between liquids, the semipermeable membrane can also be grown by interfacial chemical reactions to construct a microchannel under liquids (Figure 11c). Feng et al. have made significant progress in this area.^[27] The nano clay-polymer surfactant was assembled between oil and water to form a semipermeable membrane as all-liquid microfluidic devices with bespoke properties. The formed semipermeable membrane channel can prompt underwater microfluidic directional transport and build complex chemical systems to perform the tasks, such as chemical separations and multistep chemical transformations (Figure 11d). Interestingly, by using this superfast liquid film-forming method, the authors realized reconfigurable 3D printing under the liquid as liquid microreactors with logical optical outputs. Underwater programmable 3D printing opens up new ideas for directional open microfluidic transport. The microdroplets can also be manipulated in the subaqueous environment.^[143] In 2020, through the amplification effect of the permanent magnet on electromagnetic, the programmable motion of microdrop containing magnetic nanoparticles under a magnetic field was realized.[144] The distribution, generation, filtration, and combination of droplets on the microfluidic platform systems were completed. The application of automatic biological detection in microfluidic logistics was also explored.

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5.3. Microdrop Fabrication

Spreading tiny microdroplets is one of the most critical applications of microfluidics.^[2,6,11,145,146] Homogeneous and highly dispersed droplets can be rapidly generated and manipulated using immiscible multiphase flows inside microchannels. After nearly twenty years of development, this field has matured. Some extensive applications have been developed, such as drug delivery, emulsification, nanomaterials fabrication, and bio(chemical) analysis. However, the microdroplets obtained by the traditional closed microfluidic technology can only exist in the liquid phase, which significantly limits the application of microfluidic droplets in open space.^[147,148] The development of open microfluidics brings the dawn to solve this problem. If taking the superhydrophobic region as the terminal of the open microfluidic channel, the liquid would aggregate at the boundary between the superhydrophilic and superhydrophobic regions and form microdroplets. In 2011, Ledesma-Aguilar et al. reported a new instability strategy to achieve the controlled generation of microdrops in the air.^[149] The actions of the capillarity, viscous dissipation, and surface wetting are responsible for the generated microdrop size and emission period. The liquid filament and film will suffer asymmetry force between hydrophilic and hydrophobic regions. The open hydrophobic region can reduce the friction to cause the entrainment of the film, which forces microfilament at sufficiently strong driving, leading to microdrop emission. The generation of microdroplets by using open microfluidics not only depends on the method of wetting regulation but also can be realized by adjusting the structure and morphology of the open surface. In 2019, Li et al. demonstrated an asymmetric ratchets array surface to regulate the drop dispensing process.^[148] Inspired by the anisotropic behavior of water droplets on the Phyllostachys pubescens leaf due to asymmetrically patterned needles on its grooved surface, the hydrophilic strip patterns were set as the start point, and the superhydrophobic microratchets as a control site. The microratchets' tilt angle can manipulate the final formed droplet to adjust the water bridge's break-up process between the pendulous drop on superhydrophobic region and the water strip on the superhydrophilic region.

Different from the above two methods, obtaining smaller droplets from the mother microdroplets is another preparation idea. Lee's group proposed creating dimples by vacuum suction to guide the movement of microdroplets on the superamphiphobic surface due to the gravitational force and asymmetrical droplet deformation (Figure 12a).^[87] The local dimple structure is formed by placing a moving vacuum tip under the soft and elastic substrate. The superhydrophilic array micropatterns were obtained on a superamphiphobic surface via plasma treatment. When the microdroplet was dragged past the superhydrophilic area due to ultrahigh adhesion force, a part of the droplet was pulled down to form a smaller tiny droplet. The microdroplets obtained by this method can be used in a singledroplet multiplex bioassay. The magnetic field can be applied to realize a similar droplet driven under the effect of the magnetic field.^[150] The magnetic PDMS dimples due to magnetic attraction caused the deformation of droplets. In this research, the authors realized the anti-gravity magnetic open microfluidic transport, microdrop stirring, and chemical microreactions. These two studies show that using the outfield to transport open microfluidics can lead to better controllability and higher motion capacity. Due to the effect of the Rayleigh–Plateau instability, when a liquid column is released from a closed microchannel into open air, it tends to form droplets with a smaller surface area.^[151,152] A series of new applications have been developed by regulating the properties and states of these in-air droplets. A new concept of in-air microfluidics is presented by Visser et al. Monodisperse emulsions, particles, and fibers can be obtained as a chip-free platform.

The fabrication rates are 10 to 100 times higher than traditional closed chip-based droplet microfluidics (Figure 12b).^[153] More importantly, this in-air microfluidics can be used to print 3D multiscale (bio)materials based on the partially solidified inflight of droplets. Lewis's team has been working on 3D printing for many years and has produced a series of revolutionary studies.^[154–156] In 2018, they devised a way for liquids 3D printing in open space via acoustophoretic (Figure 12c).^[157] An accurate, highly localized acoustophoretic force was generated to eject microliter-to-nanoliter volume droplets into the air. A wide range of liquids was applied in this study, from Newtonian fluids whose viscosities span four orders of magnitude to yield stress fluids. More recently, their group reported a direct bubble writing technology.^[158] Through ejecting the liquid shell-gas core droplets into the open air, polymer foams with local programming bubble size, volume fraction, and connectivity were rapidly prepared on the substrate due to the fast solidify ability to retain their overall shape. This kind of closed-open collaborative microfluidics has gradually become a new research trend. However, it always requires complex and high-precision instruments to achieve the preset goals, and the open space environment significantly impacts its final results. As a new member of open microfluidics, the challenges and opportunities coexist.

In general, the development of bioinspired superwetting open microfluidics has moved from traditional directional transport,^[35,37,101,109] water collection,^[159] droplet manipulation^[160] to more intelligent, microscopic, and comprehensive fields, such as microrobots,[18,161-164] microsensors, micronano manufacturing biological inspection, smart fabric and so on.^[165–169] This is an important time for researchers to pay more attention to this field and to strengthen the interaction with other areas. Therefore, it is important to emphasize that bioinspired superwetting open microfluidics needs to be extended to more niche open microfluidic devices, such as thread-based open microfluidic devices,^[170-172] thin film vortex fluidic devices (VFD),^[173-175] and so on. These fields belong to the category of open microfluidics. Still, the number of studies is relatively small, and almost no bioinspired concepts have been used by relevant researchers, resulting in a considerable research gap. For example, if we can apply the multiscale hierarchical microgroove structure of superhydrophilic Sarracenia trichomes to thread-based open microfluidics, it will undoubtedly have a positive impact on microfluidics transport speed in open space^[176-177]; If the inner wall of the VFD tube or jet feeds could be modified into superhydrophobicity or superhydrophilicity, the structure, properties, and production of the synthetic products would be affected differently, or the selectivity and success rate of the detection/diagnostic results would be increased.^[178-180] These are just simple ideas, but with the







Figure 12. Microdrop fabrication by bioinspired superwetting open microfluidics. a) Controlled movement of microdroplets from superhydrophilic array micropatterns on superamphiphobic surface to obtain tiny drops. b) The in-air chip-free microfluidic platform to form monodisperse emulsions and particles. c) Open liquids 3D printing in open space via acoustophoretic. Reproduced with permission.^[87] Copyright 2018, ACS. Reproduced under the terms of the CC-BY license.^[153] Copyright 2018, The Authors, published by AAAS. Reproduced under the terms of the CC-BY license.^[153] Copyright 2018, The Authors, published by AAAS.

future efforts of scientists, eventually, bioinspired superwetting open microfluidics will certainly be established as a subject with practical application value.

6. Summary and Outlook

In this review, the evolution from closed microfluidics to open microfluidics has been introduced briefly. Then, various natural superwetting systems, including superhydrophobic, superhydrophilic, or superlubricant/slippery, were introduced and the bioinspired open microfluidics based on different bionic objects were elaborated. There are profound mechanisms behind these unique biological phenomena. So, from the mechanism perspective, other bioinspired superwetting open microfluidic applications can be designed and realized. Some promising applications have emerged as an emerging research field but must be well-developed. In this regard, we also summarized it to shed some light on future development (**Figure 13**).

As a newborn concept, bioinspired superwetting open microfluidics has yet to be well-known to many researchers. There are three fundamental reasons for this result. First, the reported bioinspired superwetting open microfluidics models still need to be more controllable and cannot reach the desired goal in speed and distance. A standard solution to this challenge is to use external fields to assist in controlling microfluidics, such as electric field, magnetic field and light field, and so on. The input of field energy and the solid surface's chemical properties can effectively improve open microfluidics' controllability and speed. There are still problems of complexity and energy consumption in terms of the results, but this is one of the key directions for the future development of the research field. Second, close microfluidics can be actively controlled bidirectionally by external pumps. But it is difficult for open microfluidics to choose the flow direction independently due to its passive drive mechanism. The introduction of external fields is also a viable solution. The direction of liquid transport can be controlled by stimulating the change of physical structure or chemical properties of open microfluidics with the change of external field. It is also possible to achieve selective transport of liquids with different physical properties (such as surface tension, viscosity, temperature, etc.) through precise optimization



Figure 13. Current challenges and future development trends of the bioinspired superwetting open microfluidics.

of the microstructure of the devices. In the future, continuing to learn from nature to develop different open microfluidic active drive mechanisms (such as vibration) is also a very feasible way to solve this problem. Third, there is still conceptual disunity that bionics researchers often need to pay more attention to the significance of manipulating liquid based on superwetting surfaces in microfluidics. Scientists who study microfluidics rarely think in terms of wetting as well. As a result, these two broad concepts have yet to be universally unified. But these two areas have already been starting to collide and spark innovations. Bionics and microfluidics need to be integrated and popularized, which is the purpose and significance of our review.

So, how do we popularize the concept of bioinspired superwetting open microfluidics? First, we should return to nature and learn from nature. After billions of years of evolution, nature holds great secrets, but we know only a few. We should explore more biological systems with special wettability and reveal the underlying mechanisms and physicochemical essentials. The aim is to put forward more bioinspired models to expand the boundaries of the bioinspired open microfluidic field. Second, we should put forward more accurate bioinspired artificial materials preparation methods. The unique properties of living things are all based on the delicate structures that evolved over hundreds of millions of years. The current preparation methods need to accurately restore the biological system of the artificial materials, which leads to the gap between the performance of artificial materials and that of living organisms. Advanced 3D printing technology is a good choice, but improving its printing accuracy is an urgent problem to solve. The last and most important thing is the application issue. Many so-called bioinspired superwetting open microfluidics applications are still in the laboratory stage and need to be extended to real-life or industry. Biological models sometimes limit researchers' ideas, so they must make more changes beyond nature. Multidisciplinary approaches are the way to solve this bottleneck. We need ideas from other disciplines, such as medicine and agriculture, to stimulate a new round of innovation. In general, bioinspired superwetting open microfluidics requires our more effort to solve the existing challenge and promote it for all. There is no doubt that it will have a firenew and glorious future.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

bioinspired, liquid manipulation, open microfluidics, superhydrophilic, superhydrophobic

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