

Superhydrophobic “Pump”: Continuous and Spontaneous Antigravity Water Delivery

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Antigravity transportation of water, which is often observed in nature, is becoming a vital demand for advanced devices and new technology. Many studies have been devoted to the motion of a single droplet on a horizontal or inclined substrate under specific assistance. However, the self-propelled water motion, especially continuous antigravity water delivery, still remains a considerable challenge. Here, a novel self-ascending phenomenon driven only by the surface energy release of water droplets is found, and a superhydrophobic mesh to pump water up to a height of centimeter scale is designed. An integrated antigravity transportation system is also demonstrated to continuously and spontaneously pump water droplets without additional driving forces. The present novel finding and integrated devices should serve as a source of inspiration for the design of advanced materials and for the development of new technology with exciting applications in microfluidics, microdetectors, and intelligent systems.

by a suctorial proboscis of butterflies, and open-close motion of the shorebirds mouthpart with a capillary ratchet.^[1] In 1992, a self-propelling millimeter-scale uphill run of a water droplet was achieved on the functional silicon wafer with a wettability gradient,^[2] which can be attributed to the asymmetrical Laplace pressure. This pioneering work both inspired and stimulated research for the generation of droplet motion driven by wettability gradient,^[3] geometrical shape,^[4] and other external stimuli.^[5] Although a single water droplet has more priorities in self-ascending due to its tiny volume and considerable surface energy in comparison with water flow, the self-propelled water droplets motion, especially continuous and spontaneous antigravity water delivery, still remains a substantial challenge.

1. Introduction

“Water flows downward,” one of the most ubiquitous natural phenomena, implies a scientific principle, i.e., the gravitational potential energy of water can spontaneously convert into its kinetic energy. In nature, water can be continuously lifted against gravity only with external driving factors, such as the transpiration of giant plants, the negative pressure supplied

In the past decades, bio-inspired superhydrophobic materials have attracted a great deal of attention and provided an important number of sources in developing technology.^[6] The superhydrophobic surface is an optimal candidate to impart a nearly perfect spherical morphology to a water droplet with maximum surface energy. The surface energy (E_{SE}) of a spherical water droplet can be evaluated with Equation (1)

$$E_{SE} = \gamma_{\text{water}} \cdot 4\pi r^2 \quad (1)$$

where γ_{water} is the surface tension of water and r is the radius of a water droplet. For a water droplet with a radius of 0.5 mm, its surface energy is about 228 nJ. Under an ideal situation, if the released surface energy of a water droplet with a radius of 0.5 mm can be completely converted into the gravitational potential energy, the maximum ascending height (h) of water can reach over 40 mm calculated using Equation (2)

$$h = \frac{3E_{SE}}{4\pi r^3 \cdot \rho_{\text{water}} \cdot g} \quad (2)$$

where ρ_{water} is the density of water and g is the gravitational acceleration.^[7] When the spherical water droplet contacts a water column, its surface energy will be largely released. Additionally, the low contact angle hysteresis (CAH) of the water droplet on a superhydrophobic substrate^[8] will facilitate the maximum surface energy release. Therefore, the above theoretical prediction demonstrated that it is feasible to achieve

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the water self-ascent powered by the surface energy released upon coalescence between a spherical water droplet and a water column.

Here, we designed a superhydrophobic “pump” to achieve spontaneous antigravity water delivery using a superhydrophobic mesh without any external forces. The spherical water droplet and a thin water column were applied on the lower and upper sides of the superhydrophobic mesh, respectively. The spontaneous water ascent results from the surface energy released upon coalescence between the droplet and the water column. Utilizing the superhydrophobic “pump,” water droplet can be spontaneously uplifted to a centimeter height scale, forming a continuous water flow. This work should serve as a source of inspiration in designing advanced fluid delivery systems.

2. Results and Discussion

2.1. The Self-Ascending Phenomenon of a Single Droplet

To test the feasibility of the superhydrophobic “pump” and visualize the mergence process, a charge coupled device (CCD) was used to in situ record the capture process of a 5 μL water droplet located on a superhydrophobic substrate with a low CAH (Figure 1A and Movie S1, Supporting Information). When a water droplet just contacted the lower surface of the superhydrophobic “pump,” the droplet can actively penetrate through the mesh and spontaneously merge into the water column on the upper mesh surface without any external forces, resulting in self-propelled rise of the liquid level in the tube, similar to the action of an engineering pump powered by a motor. During the penetration and ascent process of the droplet, it was found that the three-phase contact line of “solid–liquid–gas” was not extended horizontally, which is different from the droplet absorption by a hydrophilic substrate such as a sponge. The penetration process of a spherical water droplet through the mesh into the upper water column of the superhydrophobic “pump” is schematically represented in Figure 1B. Once the

droplet started to merge with the upper water column, the rapid release of its surface energy can propel the antigravity delivery of water.

2.2. The Effects on the Water Self-Ascending Height

Wettability of solid materials is an important issue in surface chemistry,^[8,9] which also strongly affected the performances of the as-prepared superhydrophobic “pumps.” In this work, we fabricated a series of meshes with different wettability from superhydrophobicity to superhydrophilicity and investigated the effect of wettability on the “pump” performance through continuous water droplets supply. The as-prepared superhydrophobic mesh possessed typical micro/nanoscale roughness on the wire, whereas the surface of the hydrophobic and pristine meshes was relatively smooth (Figure S1, Supporting Information). The continuous droplets were provided by a hydrophobic syringe needle with an inner diameter of 100 μm , which can give a negligible adhesive of droplets as compared to the Laplace pressure (Figure 2A). It was found that surface wettability of the mesh in the “pumps” can affect maximum self-propelled height of the liquid level (Figure 2B,C and Movie S2, Supporting Information). When a water droplet with a radius of ≈ 0.75 mm (1–2 μL) was continuously applied, hydrophobic meshes with a contact angle (CA) of $150^\circ \pm 2^\circ$ and $137.5^\circ \pm 2.5^\circ$ can result in the maximum ascending liquid level with 13.6 ± 0.4 and 10.3 ± 0.3 mm in height, respectively. Once the liquid level reaches its maximum self-ascending height, the further applied droplet from the lower side of the “pump” will result in a descent of the liquid level in the tube. Due to the insufficient Laplace pressure from the lower droplet, the water will downward transport into the droplet and spill rapidly, when the droplet and the upper water column start to merge. However, after the liquid level collapses, the original maximum self-ascending height can be reversibly achieved by applying continuous droplets and the switchable process shows good reversibility (Figure 2D and Movie S4, Supporting Information). For the pristine mesh with a CA of $115^\circ \pm 1^\circ$, the ascending height is only 2.5 mm. Hydrophilic and superhydrophilic meshes cannot fulfill the water self-ascent and no water column can be supported on their upper sides. Although the measured contact angles might not be completely suitable for interpreting this process, the effect of the wettability on the maximum self-ascending height of water was proved.

The radius of the applied droplets apparently influenced the water ascending height of the “pumps” as well as the wettability of meshes. According to the Young–Laplace equation, $P_L = 2\gamma_{\text{water}}/r$,^[10] the upward Laplace pressure (P_L) is inversely proportional to the water droplet radius (r). The increase of the droplet radius results in the decrease of both the upward Laplace pressure and the maximum water ascending height. For the integrated “pump” using the superhydrophobic mesh (the pore size is 500 μm and

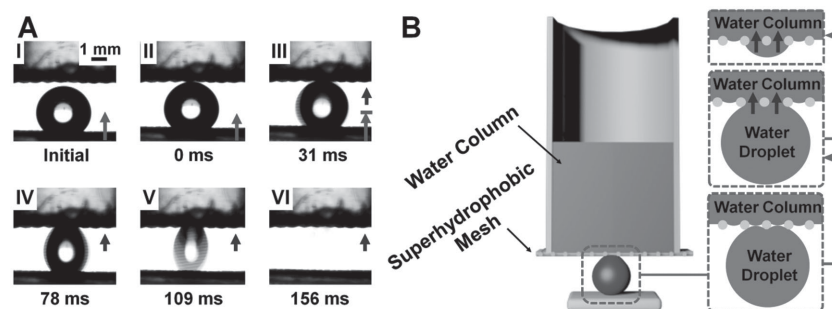


Figure 1. Superhydrophobic “pump” driven by the surface energy released upon coalescence between a droplet and a water column. A) A CCD detector in situ records a 5 μL water droplet captured, pumped, and uplifted by the superhydrophobic pump. Once the droplet is in contact with the lower side of the superhydrophobic pump, it actively penetrates through the mesh without any external forces, resulting in a self-ascent of the liquid level in the tube, similar to an engineering pump. The red and blue arrows indicated the motion of the platform and the self-ascending delivery of the applied droplet, respectively. B) Cross-sectional schematic of the superhydrophobic “pump” and the self-ascending process of the applied droplet. The surface energy release of the applied droplet results in the antigravity water delivery.

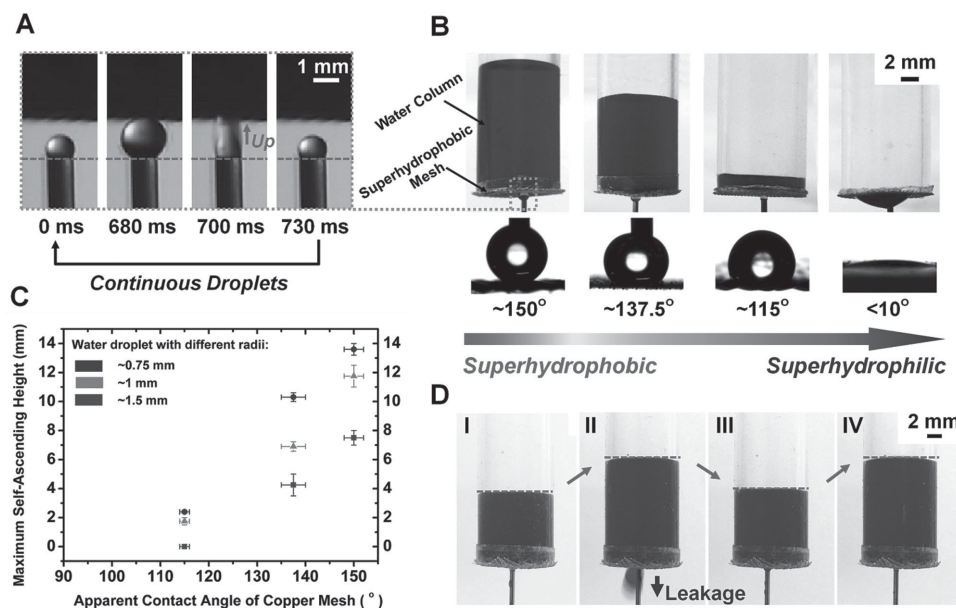


Figure 2. The maximum water self-ascending height of the “pump.” A) Visualized process of the continuous droplets capture achieved by the superhydrophobic “pump.” B) The effect of mesh wettability in the “pump” on the critical self-ascending height of the water column. The critical height of the antigravity water delivery (top) is increased with the mesh wettability (bottom) enhanced from superhydrophobicity to superhydrophilicity. C) Trend of the critical self-ascending height of the water level as a function of the water contact angle for the mesh in the integrated “pump” applied water droplets with different radii ≈ 0.75 mm (blue), 1 mm (olive), 1.5 mm (wine). D) For the superhydrophobic pump, after reaching the maximum self-ascending height of the liquid level (I), the further applied droplet will induce a descent of the liquid level (II and III). Under applying continuous droplets, the liquid level will revert to the original state (IV), exhibiting good reversibility.

the CA is about 150°), with the increase of the radius of applied water droplets from the minimum value ≈ 0.75 mm (1–2 μL), to ≈ 1 mm (3–5 μL), and ≈ 1.5 mm (10–15 μL), the resultant self-ascending height of water in the tube was decreased from about 14, to 12 and 7 mm, respectively (Figure 2C). Similar obvious trends were also observed for the integrated “pumps” using other hydrophobic mesh. In an extreme condition, the radius of a water droplet tends to infinity such as a water film, and the water film cannot be pumped and uplifted using the superhydrophobic “pump,” conversely, it will result in the collapse of the water column above the mesh (Figure S2 and Movie S5, Supporting Information).

In addition to the surface wettability of the mesh and the size of applied droplets, the effect of mesh size on the performance of the superhydrophobic “pump” was also investigated. The mesh-size strongly influences the initial condition of the droplet ascending and the maximum water support. When the pore size is lower than 200 μm , the water droplets cannot easily penetrate through the superhydrophobic mesh. As the pore size is as large as ≈ 1 mm, even the superhydrophobic mesh can only realize less than 10 mm water ascending height, arising from the low critical intrusion pressure calculated from Equation (3)^[7,11]

$$P_{\text{intrusion}} = \frac{4\gamma_{\text{water}} \cdot |\cos\theta_{\text{ca}}|}{D_{\text{pore}}} \quad (3)$$

where D_{pore} is the pore diameter of the mesh, θ_{ca} is the water contact angle of the hydrophobic mesh. In this work, the mesh with CA of 150° and with pore-size of 500 μm was applied to integrate the superhydrophobic “pump” (Figure S1, Supporting

Information). According to Equation (3), the critical intrusion pressure of this mesh is about 500 Pa, indicating the theoretic maximum height of the water column supported by such superhydrophobic mesh was about 50 mm ($h_{\text{max}} = P_{\text{intrusion}}/\rho_{\text{water}}g$). Similar to the estimation, the experimental maximum height of the water column supported by such superhydrophobic mesh was larger than 40 mm (Figure S9, Supporting Information). The results exhibited that the maximum water height supported by the superhydrophobic mesh is much larger than the maximum water self-ascending height via the superhydrophobic “pump.”

2.3. Investigation on the Mechanism of the Droplets Self-Ascending

Deeper understandings on the self-ascending mechanism of droplets would be useful for a rational design and the reproducible construction of the superhydrophobic “pump” as well as its practical application. When a droplet contacts a solid surface, it will transform from a sphere to an arch in accompany with the CA (θ_{ca}).^[7] Therefore, the θ_{ca} variation should be considered in Equation (2) (Figures S3–S7 and Equations (S1)–(S10), Supporting Information). According to Equation (S10) (Supporting Information), the self-ascending height is about 20 mm by considering θ_{ca} , which is obviously larger than the experimental data (about 13 mm). Due to the probable energy consumption such as the kinetic energy of droplets, the surface energy of the applied droplet cannot be completely converted into its gravitational potential energy. Therefore, the proposed model should be further optimized.

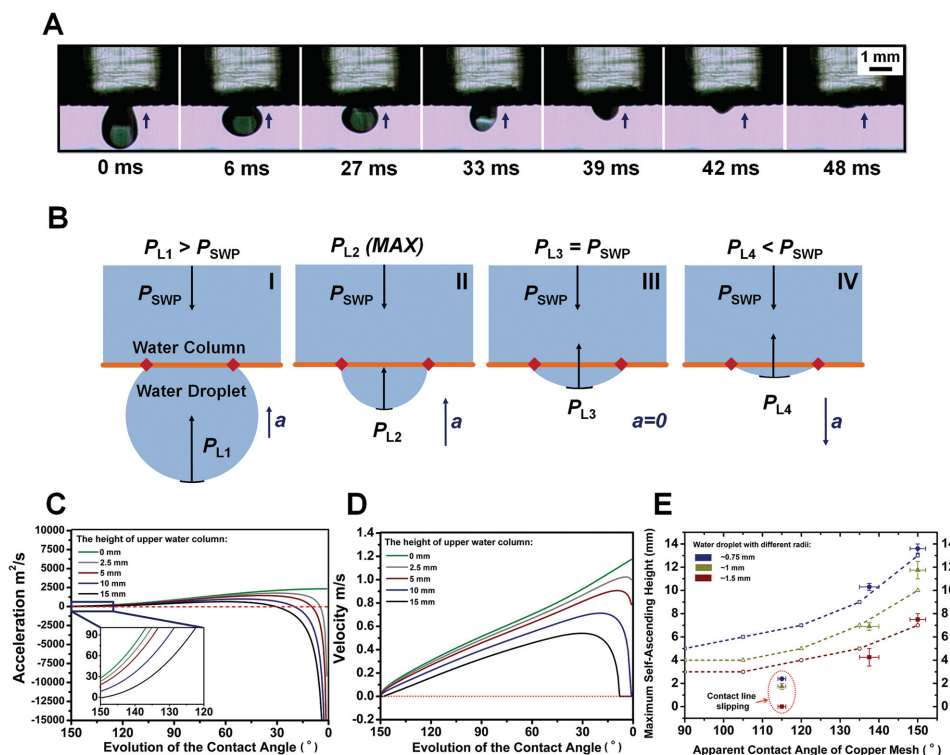


Figure 3. Proposed quasi-static model of the spontaneous antigravity water transportation for the present superhydrophobic “pump.” The water droplet is “pumped” by the difference between the Laplace pressure (P_L) resulted from its curved surface and the static water pressure (P_{SWP}) of the upper water column. A) A high speed video camera visualizes the penetration and self-ascension process of the droplet applied at the lower side of the integrated superhydrophobic pump. The result clearly shows that the solid–liquid–gas three-phase contact line can remain pinned during the self-ascending process of the applied droplet penetrating through the mesh. B) Four stages in the droplet-ascension process: (I) the initial condition of droplet-ascension; (II) achieving the maximum P_L ; (III) the equilibrium of P_L and P_{SWP} ; (IV) insufficient P_L for assisting the droplet-ascension. Under water columns with a known height (selectively exhibited 0, 2.5, 5, 10, and 15 mm in the figures), the variation of P_L directly determines the C) instantaneous acceleration and D) instantaneous velocity of the droplet with a radius of 1 mm. The acceleration and velocity of droplets were gradually increasing at beginning of droplet-ascension. Afterward, the acceleration of the droplet would rapidly decrease when the P_L starts to decrease. The final acceleration would be less than zero, resulting in a rapid decrease of the droplet velocity. E) Estimated from the final velocity of droplets, the theoretical predicted data (dashed line) of maximum water self-ascending height were exhibited in the figure as well as the experimental results (point).

Considering the detailed process of the droplet self-ascension, the mechanism was further investigated in this work. A quasi-static model was proposed on the basis of the ideal droplet assumption, which can provide a certain Laplace pressure for calculation (Figure S6, Supporting Information). The observation of a high speed video camera showed that the contact line between the water droplet and superhydrophobic mesh remained pinned during the penetration of water droplets (Figure 3A and Movie S6, Supporting Information). The pinning of the contact line reduced the horizontal energy loss arising from the water-spreading on the lower surface, and then maximally preserved the Laplace pressure for the self-ascending of water droplets. Combining the above discussion, the optimized quasi-static model for antigravity delivery of water can be explained by the following considerations (Figure 3B). Under a constant static water pressure (P_{SWP}), the droplet ascending process for the superhydrophobic “pump” can be divided into four stages. At the first stage, the water droplet will self-ascend when P_{L1} is larger than P_{SWP} ($P_{L1} > P_{SWP}$). P_{L1} is defined as the initial Laplace pressure of the water droplet first contacting with the lower surface of the superhydrophobic mesh. At the second stage, the droplet will be accelerated to ascend with an

increased acceleration (Figure 3C), arising from an increase of the Laplace pressure induced by the spontaneous decrease of the curvature. The Laplace pressure reaches a maximum (P_{L2}) when the instantaneous CA is equal to 90° . Subsequently, the decrease of the difference between P_L and P_{SWP} resulted in a reduced upward driven force. At the third stage, the acceleration reduced to zero at the point of P_{L3} ($P_{L3} = P_{SWP}$). The velocity of the droplet is decreasing continuously and rapidly when the Laplace pressure is lower than P_{SWP} ($P_{L4} < P_{SWP}$). Finally, when the velocity reaches zero, the ascending of the droplet tends to stop. In order to calculate the instantaneous acceleration (Equation (4)) and velocity (Equation (4)) of the lower droplet, the water droplet was simplified and considered as an ideal mass-losing mass-point with a varied velocity, without the consideration of the complex droplet behaviors such as liquid deformation^[12]

$$a_i = \frac{F_i}{m_i} = \frac{\left(\frac{4\gamma \cdot \sin \theta_{ca(i)}}{D} - P_{SWP} \right) \cdot \pi \left(\frac{D}{2} \right)^2}{\frac{2}{3} \pi \left(\frac{D}{2 \sin \theta_{ca(i)}} \right)^3 \left[1 - \frac{3}{2} \cos \theta_{ca(i)} + \frac{\cos^3 \theta_{ca(i)}}{2} \right] \cdot \rho} \quad (4)$$

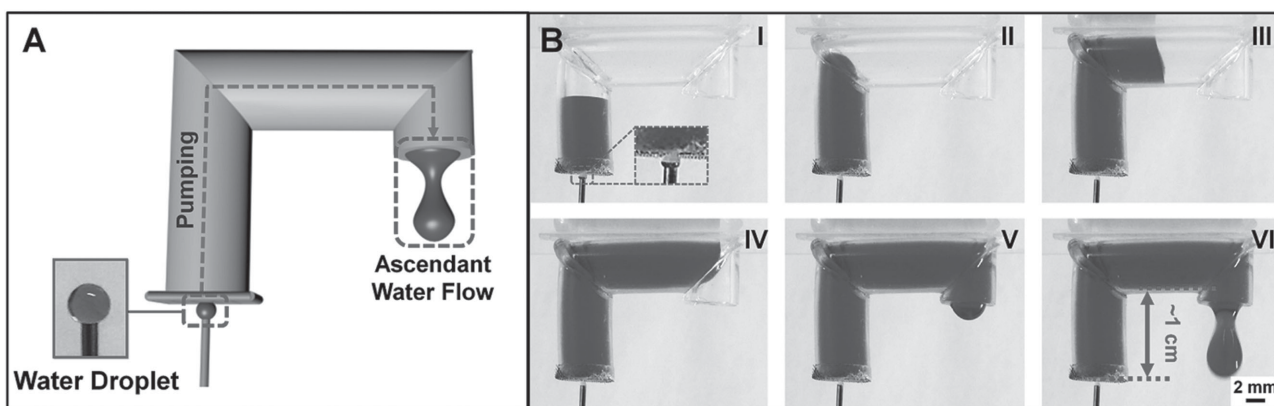


Figure 4. A continuous and spontaneous antigravity water delivery system. A) Schematic presentation of the integrated plumbing to simultaneously capture droplets and upward transport water without additional driving forces. B) Demonstration of the process for antigravity water transportation. The syringe needle did not directly contact the lower surface of pump. Accordingly, water droplets are continuously applied on the low end and water flow is successively discharged from the high end with a height difference in a centimeter scale.

$$V_i = \sum_{n=1}^{i-1} a_n \cdot t_n \quad (5)$$

where $\theta_{ca(i)}$ and D are the instantaneous CA and the diameter of the contact circle, respectively. P_{SWP} is a given constant static water pressure.

Using Equations (4) and (5), we can calculate the instantaneous Laplace pressure, mass, and velocity derived from the instantaneous CA of the applied water droplet on the lower surface of the superhydrophobic mesh. By comparison, Figure 3C displays the behavior of the instantaneous acceleration (a_i) during the droplet ascent as a function of the instantaneous $\theta_{ca(i)}$ under different static water pressure (P_{SWP}). The continuous changes of both Laplace pressure and droplet mass resulted in a variable acceleration, with an initial increased acceleration followed by a reduced acceleration. The acceleration of droplets was slowly increasing in accordance with the enhancement of Laplace pressure. Afterward, the acceleration of the droplet would rapidly decrease on the condition that the Laplace pressure cannot provide enough uplifted force. The final acceleration would be less than zero, indicating the droplet suffered a rapid deceleration. Introducing Equation (4) in Equation (5) yields the instantaneous velocity (V_i) (Figure 3D). The plot of velocity as a function of the variation θ_{ca} under different static water pressure P_{SWP} exhibited a similar trend shown in Figure 3C. The velocity increases gradually and then decreases dramatically. Under a critical P_{SWP} , the final velocity could be just reduced to zero, which indicated the self-ascent of the droplet was stopped. The maximum water ascending height can be deduced from the critical P_{SWP} and derived from Equation (5) accordingly (Figure 2E and Equations (S11)–(S17), Supporting Information). It was found that the theoretical value of the water ascending height is relatively in agreement with the experimental data, especially for the meshes with contact angles of 150° and 137.5° . Arising from the undesired extension of the contact line between the applied droplet and the lower surface of the mesh during the water-ascent, the proposed model is incapable to predict the self-ascending height of the liquid level for the mesh with a contact angle less than 130° (Figure S8,

Supporting Information). Due to the complex of the droplet during its penetration and self-ascent processes, the proposed model should be further optimized to clarify the mechanism and to accurately predict the self-ascending height in future work.

2.4. Antigravity Water Delivery via the Superhydrophobic “Pump”

Inspired by the observed new phenomenon, we design a novel antigravity water delivery system by integrating the superhydrophobic “pump” with a bend liquid pipe (Figure 4A). In order to efficiently achieve the successive antigravity water delivery, spherical water droplets were continuously applied on the lower surface of the “pump” using a hydrophobic syringe needle (Movie S7, Supporting Information). The applied droplet was spontaneously and rapidly captured and pumped, resulting in the release of fresh lower surface for the following applied droplets. The mass flow of liquid water was efficiently transported from the low end to the other high end without additional driving forces (Figure 4B). Utilizing this superhydrophobic “pump,” the self-ascending height of water level can reach a centimeter scale solely driven by the surface energy release of tiny water droplets. This work demonstrated a spontaneous and continuous antigravity delivery of water droplets, which may serve as a source of inspiration to further improve the existing antigravity water transportation strategies.

3. Conclusion

In conclusion, a new self-ascending phenomenon driven only by the surface energy release of water droplets was reported in this work. Inspired by the novel observation, we demonstrated the spontaneous water ascent via a superhydrophobic pump. Water can be uplifted to a centimeter scale height. We also designed an integrated antigravity water delivery system. Without external forces, water droplets were continuously and

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spontaneously transported upward from the lower end and were discharged at the higher end as water flow, indicating a continuous and self-propelled antigravity water delivery. Although the mechanism of the present antigravity water transportation is complex and its detailed investigations should be further studied, this work should serve as a source of inspiration to construct advanced devices and to develop new technology in the field of actuators, microdetectors, microfluidics, water collection and transportation, etc.

4. Experimental Section

Fabrication of the Superhydrophobic “Pump”: The superhydrophobic mesh was fabricated by combining 5 min alkali erosion (2.5 M of sodium hydroxide and 0.13 M of ammonium persulfate in aqueous solution) and further modification by a $\approx 1 \times 10^{-3}$ M dodecanethiol/ethanol solution for 24 h. The superhydrophobic “pump” was constructed by the copper mesh and polymer tube with an inner diameter of 8 mm. In order to diminish the probable influence of water self-ascending height induced by the capillarity of the upper tube, the hydrophobic polymer tube with a CA about 90° was selected to fabricate the pump. An initial water column about 3 mm height was prior incorporated into the tube for the droplets capture.

Instruments and Characterization: All the contact angles of the meshes were measured by a SCA-20 contact angle goniometer (Dataphysics, Germany), and the high speed video was captured by Phantom V9.1 video camera (America). The water droplet was continuously supplied by a syringe needle with a speed of $\approx 6 \text{ mL h}^{-1}$.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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