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Hierarchical nanotexturing enables acoustofluidics on slippery yet sticky, flexible surfaces

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transport

Abstract. The ability to actuate liquids remains a fundamental challenge in smart microsystems, such as those for soft robotics, where devices often need to conform to either natural or three-dimensional solid shapes, in various orientations. Here we propose a hierarchical nanotexturing of piezoelectric films as active microfluidic actuators, exploiting a unique combination of both topographical and chemical properties on flexible surfaces, whilst also introducing design concepts of shear hydrophobicity and tensile hydrophilicity. In doing so, we create nanostructured surfaces that are, at the same time, both slippery (low in-plane pinning) and sticky (high normal-to-plane liquid adhesion). By enabling fluid transportation on such arbitrarily shaped surfaces, we demonstrate efficient fluid motions on inclined, vertical, inverted, or even flexible geometries in three dimensions. Such surfaces can also be deformed and then reformed into their original shapes, thereby paving the way for advanced microfluidic applications.

Precision control of microfluids on a small scale, particularly in a microdroplet format, underpins a range of smart microsystems, such as those for soft robotics and lab-on-chip.¹⁻³ However a major challenge remains in the complexity of assembling all the necessary functionalities using conventional microfluidic devices which are generally based upon rigid substrates, and are thus unable to conform to the flexing of natural shapes.⁴⁻⁷ Future applications for the directional transport of fluids will require liquid actuation on flexible and deformable surfaces, on three dimensional shapes, such as those in robotics.

Thin film piezoelectric surface acoustic wave (SAW) devices have previously been shown to enable all necessary microfluidic functions in diagnosis and sensing applications⁸. Material

stoichiometry, texture and crystallinity can be easily controlled, whilst remaining biocompatible.^{9–11} They have previously been developed on conformable substrates, and have been used for sensing and microfluidics with similar performances to their rigid counterparts.^{12–14} However, to date, their deformability has not been exploited due to challenges in realizing both long-range wave propagation across the surfaces as well as in implementing liquid handling on curved surfaces (where gravity causes the droplet to detach).

To address these issues, we now show that we can deposit and control the surface structures of ZnO on the nanoscale, on aluminium foils (with thickness from 50 to 600 μm) as flexible and acoustically actuated microfluidic devices, which can be deformed (and maintain their temporal shape) before being returned to their original shapes. Importantly, we show that the acoustic waves not only propagate over long distances of several centimetres without significant dissipation, but also overcome the major challenge of enabling efficient liquid handling on curved surfaces – providing the designated slipperiness necessary for motion along the surface, whilst ensuring sufficient stickiness for the retention of the liquid when the surface is inclined, vertical or even inverted.

Hydrophobic coatings such as CYTOP¹⁵, or topographic structures (including roughness) have previously been combined with hydrophobic materials^{16–18} to create surfaces with superhydrophobicity, minimizing droplet pinning forces and creating a slippery surface. However, this also minimizes the surface contact area, which itself is directly related to the quanta of energy that can be transferred from the acoustic waves into the liquid.

The reduced contact area for superhydrophobic surfaces also necessarily results in a reduced droplet adhesion normal to the plane (“stickiness”). Additionally, as the surface oscillates with a nanometer-scale amplitude generated by the acoustic excitation, this increases the probability that the liquid penetrates the surface texture, resulting in an irreversible pinning of the droplet. Such issues are counter-intuitive to the key design criteria for a three-dimensional and bendable smart system for droplet transport, which critically requires retaining droplets on the surface as the physical orientation is varied (e.g. either as the angle of inclination or curvature).

We demonstrate that by engineering the material stack and surface texture of a ZnO/Al piezoelectric flexible sheet for our SAW-based devices, we can achieve droplet manipulation on large scales and conformable shapes not previously possible, opening a new way for flexible devices to perform complex smart microfluidic systems. We combine the microscale roughness of the metal foil substrate with the nanoscale structures of ZnO film to create a hierarchically textured surface, showing not only a low contact angle hysteresis (i.e. shear hydrophobicity¹⁹ allowing easy translation of droplets across the surface), but also a receding contact angle allowing droplets to be retained in a wide range of surface orientations (i.e. with sufficient tensile hydrophilicity¹⁹). Importantly, we not only demonstrate that such ZnO/Al sheets support low power wave propagation and efficiently drive liquids on horizontal, inclined, vertical and arbitrarily shaped surfaces, but also show that both the contact angle hysteresis and receding contact angle are key design parameters for requiring droplet transportation and adhesion.

Surface design considerations

Figure 1a depicts a droplet in sequential positions along a curved surface driven by SAW propagating along the surface. For microliter-sized droplets (for instance 1 μL with a radius of ~ 1 mm), the radius of curvature of surfaces conforming to geometric features can be considered infinitely large (>10 cm) as compared to the droplet, so that we can estimate the forces applied to the droplet by considering a flat sheet tangential to the curved surface. For a droplet placed on a horizontal flat surface (see Fig. 1b), the idealized static equilibrium state is described using Young's equation,

$$\gamma_{sl} + \gamma_{lv} \cos \theta_Y = \gamma_{sv} \quad (1)$$

where, γ_{sl} is the surface tension of the liquid-solid interface, γ_{lv} is the liquid surface tension, γ_{sv} is the solid surface tension, and θ_Y is the contact angle in the idealized equilibrium state.

When the SAW propagates, and thus moves a droplet along the horizontal surface, the advancing and receding contact angles, θ_{adv} , and θ_{rec} , define the contact hysteresis and the pinning of the contact line²⁰. To agitate and move the droplet, the externally applied driving force must overcome the pinning force of the droplet due to this contact angle hysteresis. Assuming there is a perfect circular contact line for the droplet, the lateral (radial) pinning force can be formulated as^{21,22}:

$$F_{cl} = \frac{24}{\pi^3} \gamma_{lv} D_0 (\cos \theta_{rec} - \cos \theta_{adv}) \quad (2)$$

where D_0 is the equivalent base diameter in a static state.

The force can be reduced by decreasing the difference between the advancing and receding contact angles, or by tuning the surface topography and hydrophobicity, thus enabling the liquid to ball up and changing the length of the contact line (represented in Eq. (2) by the equivalent base diameter).

Considering the case of driving a hanging droplet, e.g., perpendicular to a surface and upwards, the actuation force must overcome both the work of adhesion (given by $\gamma_{lv}(1+\cos\theta_{rec})$ ^{19,23}) and gravity. Use of a superhydrophobic surface is difficult to implement, because although droplets can be pushed on inclined or inverted superhydrophobic surfaces (at an angle α , $0^\circ < \alpha \leq 180^\circ$), in practice, once the actuation force stops, gravity will often cause the moving liquid droplet to detach from the surface and fall off.

An alternative approach that reduces the contact line force is to reduce the contact angle hysteresis (i.e. increase shear hydrophobicity¹⁹), whilst retaining a receding contact angle at a value appropriate to ensure that the droplet adheres to the surface when subject to normal forces (i.e. tensile hydrophilicity¹⁹). In this approach, the reduction in contact angle hysteresis does not require the droplet to be balled-up or the contact line length to be reduced to achieve an easy droplet transport along the surface. Moreover, since the contact line length does not have to be reduced, the droplet-surface area is large enough for the acoustic wave to interact with the liquid droplet, and hence enable an efficient wave energy dissipation, thus low power actuation is maintained.

Once the droplet starts moving under agitation of a SAW, the viscous resistance force becomes significant due to a strain formed along the contact line. This force, $F_r(v)$, can be written as²⁴:

$$F_r(v) = \int_0^{\frac{D_0}{2} - \varepsilon_c} \frac{3\mu v}{h(r)} (2\pi r) dr \quad (3)$$

where ε_c is the cut-off length to prevent the singularity at the contact line, μ is the liquid viscosity, v is the droplet velocity, and $h(r)$ is the height of the droplet at certain position dependent on the radial coordinate r . $F_r(v)$ is equal to zero when the droplet is immobile.

Figures 1b to 1e compare the total SAW streaming forces, F_s , applied to the droplet in both static and dynamic states of different substrate orientations, induced by an acoustic pressure, which is defined as²⁵⁻²⁷:

$$F_s = -\left(1 + \theta_1^2\right)^{3/2} A^2 \omega^2 k_i \exp\left(2(k_i x + \theta_1 k_i z)\right) \quad (4)$$

where A is the wave amplitude, ω is the angular velocity and k_i is the imaginary part of k_i which is the wave number of the leaky surface acoustic wave. $\theta_1 = j\theta$ (with $j^2 = -1$) is the attenuation constant where $\theta^2 = 1 - (v_{Le}/v_l)^2$, v_{Le} is the leaky wave velocity, while v_l is the sound velocity in liquid.

The direction of the pressure/force follows the Rayleigh angle Θ_R which is defined by:

$$\Theta_R = \sin^{-1}(v_l/v_s) \quad (5)$$

where v_s is the sound velocity in the flexible substrate. The minimum force required to move the droplet can be written as:

$$F_s \sin \Theta_R - F_{cl} = 0 \quad (6)$$

The relationship among the different forces applied to the droplet which are balanced during droplet movement, can be described using the following equation:

$$F_s \sin \Theta_R - F_{cl} - F_r(v) = m \frac{dv}{dt} \quad (7)$$

where m is the mass of the droplet. This relationship predicts that the projection of acoustic force will be increased with the Rayleigh angle for a given input power. If the surface is tilted at a certain inclination angle $0^\circ < \alpha \leq 180^\circ$, the gravitational force is involved at both static and dynamic states, thus the force balance in both the z' and x' directions should be considered (Figs. 1c to 1e). In the static state, the forces are balanced to keep the droplet stable in its position, then we have:

$$\begin{cases} mg \sin \alpha - F_{cl} = 0 & 0^\circ < \alpha < 180^\circ \\ mg - \pi D \gamma_{lv} \sin \theta_{rec} = 0 & \alpha = 180^\circ \end{cases} \quad (8)$$

where g is the acceleration of gravity. Accordingly, in a dynamic state, the acoustic force needed to initiate the droplet movement is:

$$F_s \sin \Theta_R - F_{cl} - mg \sin \alpha = 0 \quad (9)$$

Meanwhile Eq. (7), which describes the relationship between the droplet velocity and the forces, can be further modified as:

$$F_s \sin \Theta_R - F_{cl} - F_r(v) - mg \sin \alpha = m \frac{dv}{dt} \quad (10)$$

Based upon the above discussion and analysis, in order to realize three-dimensional manipulations of droplets on inclined or even curved surfaces driven by SAW, the surface needs to be prepared not only with a low contact angle hysteresis for easy transportation of droplets, but also with a suitable receding contact angle to retain the droplet on it.

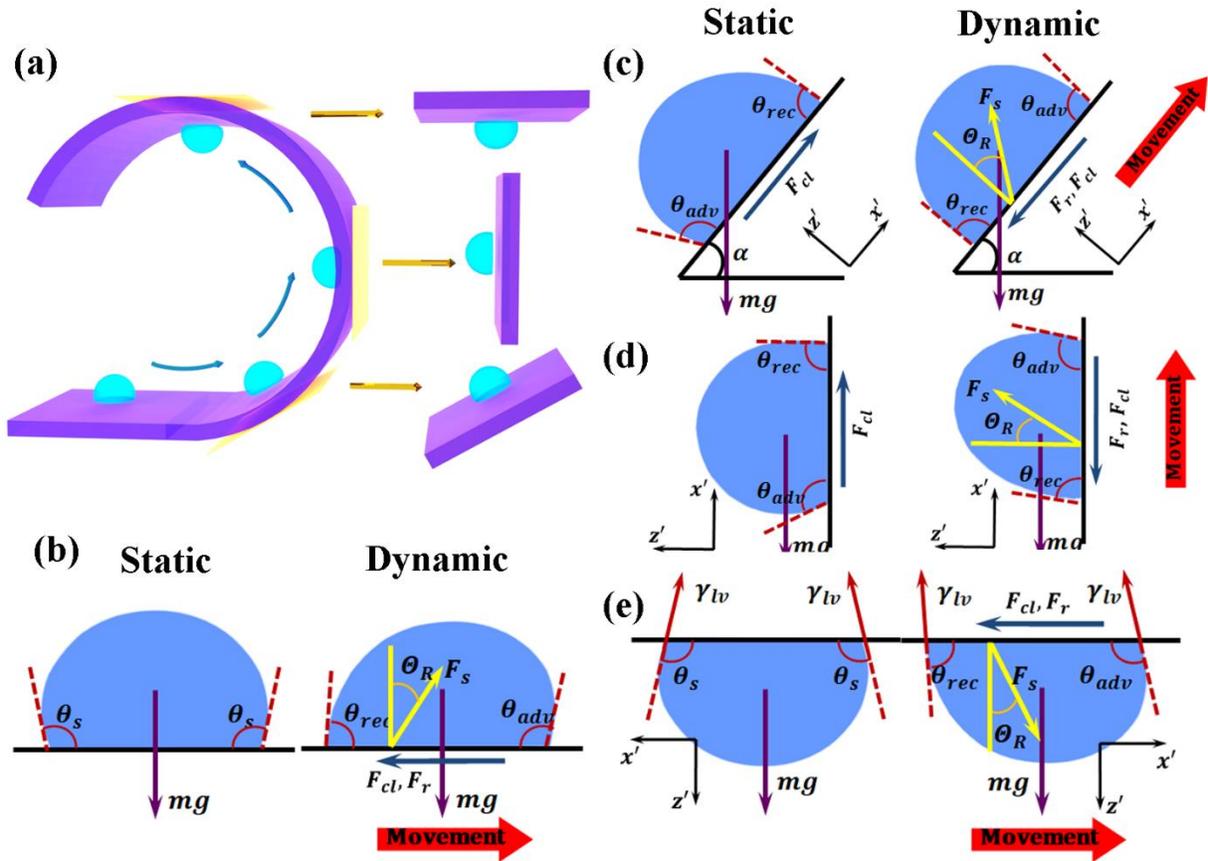


Figure 1. Principles of droplets moving on curved surface. (a) Schematic diagram of droplet movement on a curved surface, which can be treated as a sum of movements on tangential planes. (b-e) Force analysis of the water droplet retained and moving on hydrophobic surfaces: (b) placed horizontally, (c) tilted at angle α , (d) placed vertically ($\alpha=90^\circ$) and (e) placed up-side down ($\alpha=180^\circ$) in static and dynamic (surface acoustic wave power is on) states.

The contact angle hysteresis and receding contact angle can be regarded as two key parameters describing a design space, with the first parameter controlling whether a droplet is pinned (thus preventing motion along it), and the second parameter controlling how strongly a droplet adheres in the direction perpendicular to the surface. Figure 2 summarizes these four fundamental combinations of slipperiness and stickiness (e.g., showing low/high pinning and low/high

adhesion) and provides exemplars of potential applications²⁸⁻³⁰. For our application in this study requiring droplet motion along a surface that can have multiple orientations including vertical and inverted, the surface wettability design requires ease of transport and sufficient adhesion and this was achieved by creating a hierarchical texture of nano- and microscale features, as shown in Figs. 3a and 3b.

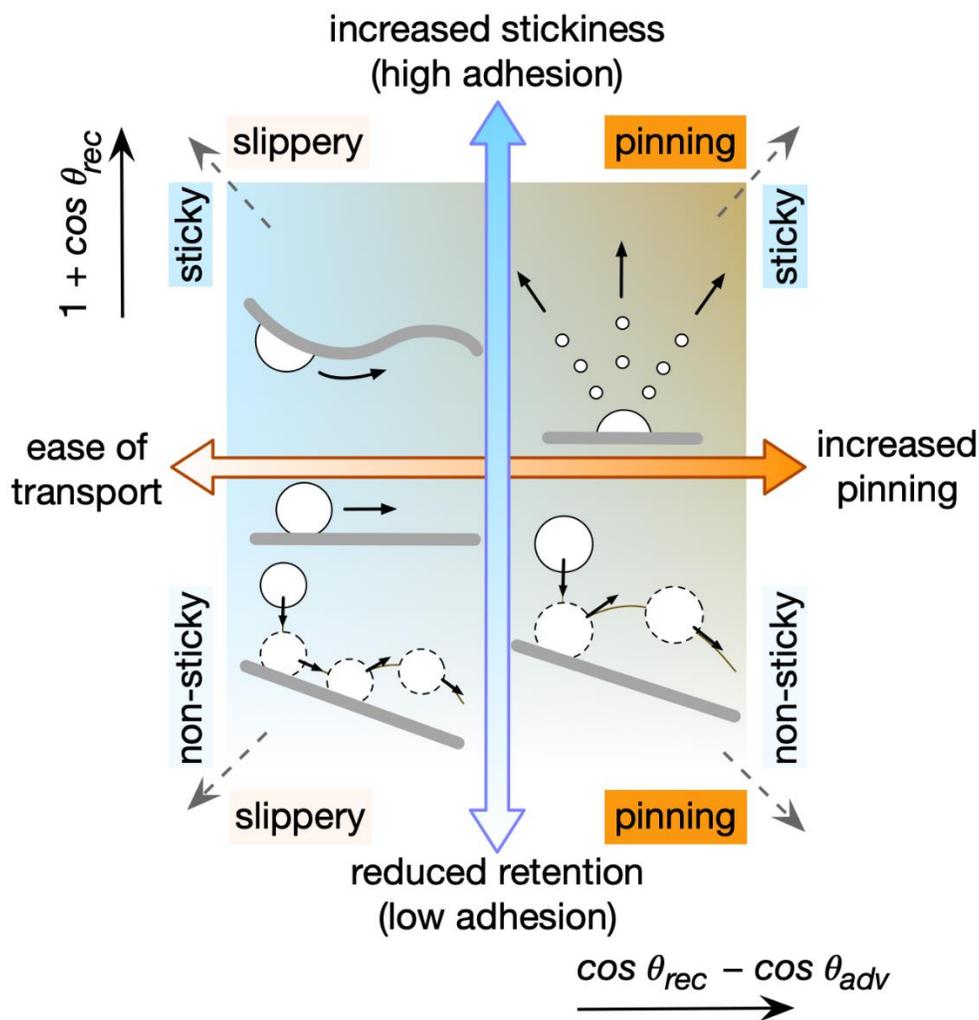


Figure 2. Combinations of four different surface types arising from contact angle hysteresis and receding contact angle. Scheme defining the design space of slippery/non-sticky²⁸,

pinning/non-sticky²⁹, pinning/sticky³⁰, and slippery/sticky surface types with exemplars of possible applications.

Hierarchically textured surface with shear hydrophobicity and tensile hydrophilicity

Here we used Al sheets with a thickness of 600 μm as the underlying substrates. There are surface grooves with microscale roughness on the surface, generated by cold rolling manufacturing processes (Fig. S1), and the root mean square roughness of grooved surfaces is in the range of $0.72 \pm 0.05 \mu\text{m}$, measured using a profilometer. ZnO thin film was deposited on top of the microstructure, providing a hierarchical and fractal surface with an average roughness value of 73.6 nm (shown in Fig. 3c - AFM image, $5 \mu\text{m} \times 8 \mu\text{m}$).

Sputtered ZnO thin films have a relatively low surface energy, and contact angles of a water droplet on such a surface vary between 70° to 100° ³¹. When the droplet is located on the ZnO/Al sheets structure, a Wenzel state is formed on the surface as shown in Fig. 3a. This Wenzel state could be transformed into a Wenzel/Cassie-Baxter mixed state by coating the ZnO surface with a thin hydrophobic layer (300 nm - 500 nm thick) of CYTOP, an amorphous fluoropolymer with high transparency and water and oil repellency. Figures 3c and 3d show that the nanoscale roughness is reduced after the surface is coated with CYTOP. For a comparison, we also measured the surface roughness of a ZnO film deposited onto a standard polished (100) Si wafer using the AFM and obtained a homogeneous and smooth surface with an average roughness of $15.05 \pm 4.58 \text{ nm}$ over an area of $100 \mu\text{m} \times 100 \mu\text{m}$, which is much smaller than those on the foil substrates.

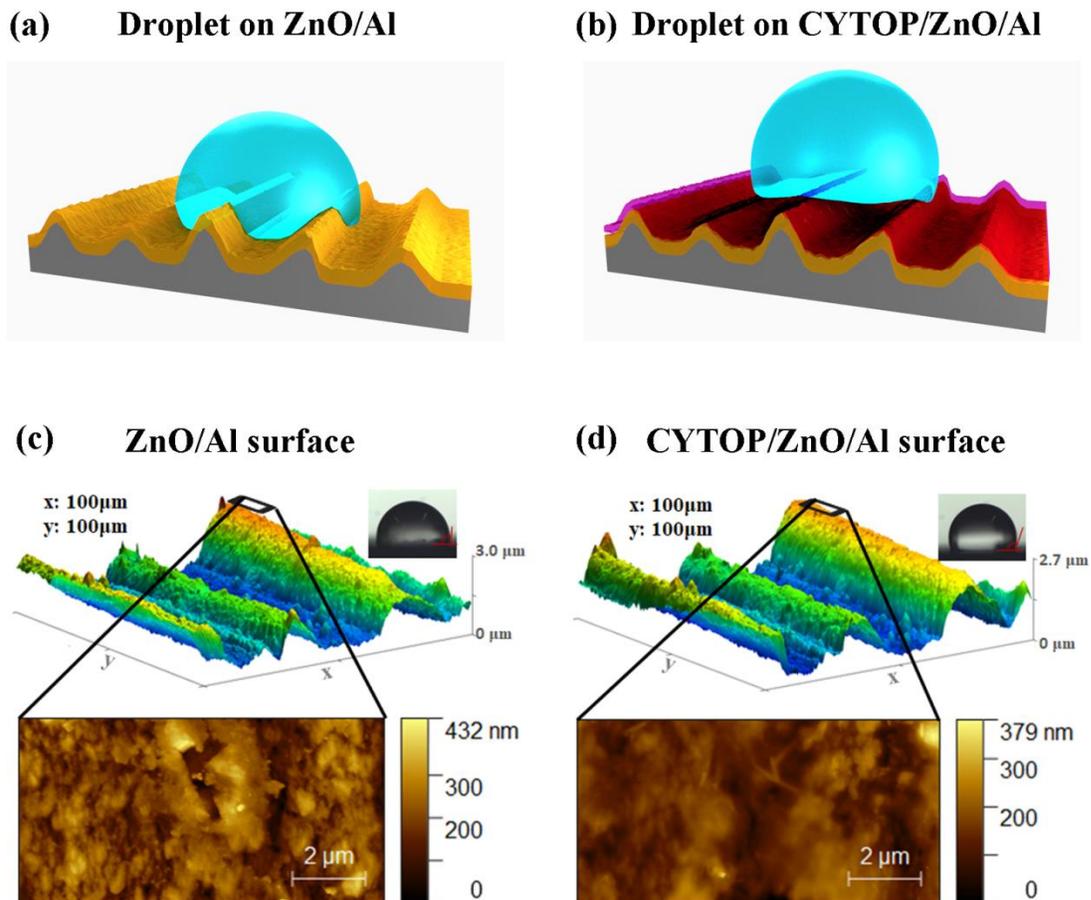


Figure 3. Surface morphology of hierarchical textures. (a) 3D schematic diagram of the bi-layer structure composed of Al substrate (grey) and ZnO thin film (orange). (b) 3D scheme of the tri-layer structure composed of the Al substrate, ZnO thin film and CYTOP coating (red). AFM images showing the root mean squared (RMS) roughness of the hierarchical structure (c) before and (d) after CYTOP coating.

To further understand the wettability of the ZnO/Al sheet surface, we measured the static contact angle θ_s , and the advancing and receding contact angles, giving us a contact angle hysteresis, $\Delta\theta = \theta_{adv} - \theta_{rec}$, for 1 μL water droplets with and without CYTOP treatment as shown in Fig. 4a (values are tabulated in Table S1 in Supplementary Information). The untreated ZnO/Al sheet

delivered good tensile hydrophilicity with a receding contact angle of $49^\circ \pm 14^\circ$ retaining the droplet against any normal force. However, the droplet on such a surface is also pinned in the direction of movement due to the large contact angle hysteresis of $63^\circ \pm 15^\circ$ and, hence, poor shear hydrophobicity. Similarly, the untreated ZnO/Si surface provides a smaller receding contact angle ($27^\circ \pm 4^\circ$), indicating a better tensile hydrophilicity, whilst having poor shear hydrophobicity due to the contact angle hysteresis staying the same as the untreated ZnO/Al surface. After the surface was coated with CYTOP, the surface wetting properties were modified, and the shear hydrophobicity was enhanced with a decreased contact angle hysteresis of $28^\circ \pm 6^\circ$. The receding contact angle of CYTOP/ZnO/Si surface increased to $95^\circ \pm 4^\circ$ but was still able prevent the droplet from being detached perpendicularly providing acceptable tensile hydrophilicity (adhesion) compared to superhydrophobic surfaces. By using the ZnO/Al surface, we could further optimize the shear hydrophobicity and reduce the contact angle hysteresis to $9^\circ \pm 2^\circ$ whilst achieving a receding contact angle of $105^\circ \pm 1^\circ$, thus creating a surface that simultaneously has excellent shear hydrophobicity (slipperiness) and sufficient tensile hydrophilicity (adhesiveness). In such cases, the surface acoustic wave actuation force required to transport a droplet along a surface, which is proportional to $\gamma_{lv}(\cos\theta_{rec} - \cos\theta_{adv})$, is reduced, whilst the force normal to the surface required to remove a droplet, which is proportional to $\gamma_{lv}(1 + \cos\theta_{rec})$, remains sufficiently large to prevent droplet detachment.

Droplet transport enhanced on horizontal and tilted flat surfaces

The input power determines the displacement amplitude of the Rayleigh wave, thus the streaming force F_s . According to Eq. (9), when $F_s \sin \Theta_R$ is larger than the lateral pinning force F_{cl} ($mg \sin \alpha = 0$ applied for a horizontal surface), the droplet starts to move forwards. During the

movement, Eq. (10) can be used to explain the behavior of the droplet shown in Fig. 3b, where the instant velocity presents multiple peak values. The streaming force F_s decreases exponentially as the droplet moves away from the wave source due to its attenuation in the substrate. Meanwhile, the viscous resistance force $F_r(v)$ is proportional to the instant velocity. At the starting point, $F_s \sin \Theta_R$ is much larger than the sum of F_{cl} , F_r and $mg \sin \alpha$, creating a positive acceleration dv/dt . The instant velocity increases accordingly while $F_s \sin \Theta_R$ decreases and F_r increases until the forces reach their first balance point, where $dv/dt = 0$. Afterwards, the instant velocity decreases rapidly due to the negative acceleration, causing the fast decrease of F_r until once again $F_s \sin \Theta_R$ is dominant. This process would repeat several times before the droplet moves to the position where the damped acoustic wave cannot initiate the movement.

We also investigated the droplet transport characteristics of the hydrophobically treated ZnO/Al SAW devices by measuring average moving velocities and accelerations at different power levels (Fig. 4c), and compared them with those on a ZnO/Si surface. The average accelerations on both the CYTOP/ZnO/Al and CYTOP/ZnO/Si surfaces are increased with the input power, reaching $42.1 \pm 7.1 \text{ mm/s}^2$ and $0.05 \pm 0.003 \text{ mm/s}^2$, respectively for an applied power of 0.72 W, corresponding to average velocities (averaged over time) of $8.0 \pm 0.8 \text{ mm/s}$ and $0.2 \pm 0.01 \text{ mm/s}$. Importantly, the threshold power to initiate droplet movement on the CYTOP/ZnO/Al surface is much smaller than that on the CYTOP/ZnO/Si device. In addition, the droplet average velocities are higher on the CYTOP/ZnO/Al surface than that on the CYTOP/ZnO/Si device at the same power, indicating that the acoustic devices on the ZnO/Al with a hierarchically textured surface are more efficient for droplet transport.

This enhanced performance can, in the first instance, be attributed to the hierarchical CYTOP/ZnO/Al surface, which reduces the contact angle hysteresis to 9° , significantly smaller than on the CYTOP/ZnO/Si surface at 28° , decreasing the threshold power required to initiate the droplet movement along the surface, and also reducing the power needed to overcome the lateral pinning force which could restrict the droplet movement. The surfaces also retained a receding contact angle of 105° , ensuring that droplets do not detach from the surface when curved/bent.

For completeness, the Rayleigh angle also plays an important part in the enhanced performance. We calculated it as 30.8° on ZnO/Al, as opposed to 19.7° on ZnO/Si (using phase velocities of 2868 m/s on the ZnO/Al sheet, 4351 m/s on ZnO/Si, and 1468 m/s in water) and verified experimentally (e.g., 33.6° and 15.8° as shown in Fig. 4d and 4e). As predicted by Eq. (10), the driving force is the projection of the streaming force on the moving surface, which can be calculated by $F_s \sin \theta_R$. For ZnO/Al, $\sin \theta_R$ is around 0.55, whilst it is around 0.27 for ZnO/Si. Therefore, given the same input power (same F_s), the driving force of ZnO/Al devices is twice that of ZnO/Si devices along the moving direction, thus is more efficient for droplet transport.

Furthermore, the enhanced actuation of the liquid can be attributed to the different ways in which the droplets interact with the substrate surface. For the droplet on the CYTOP/ZnO/Al substrate, assuming it is in a mixed Wenzel/Cassie-Baxter state, the entrapped air prevents the droplet from flowing into the surface's topology and reduces the contact area between the liquid and the solid surface, thus lowering the friction and causing efficient transport. In contrast, there is little air entrapped under the droplet on the silicon substrates with smooth surfaces. The larger Rayleigh angle and smaller contact angle hysteresis for the CYTOP/ZnO/Al device during movement imply

that liquid droplets move using a combination of rolling and sliding (Fig. S2a). On the contrary, the droplet movement is dominated by sliding and jumping modes on the CYTOP/ZnO/Si surface (Fig. S2b).

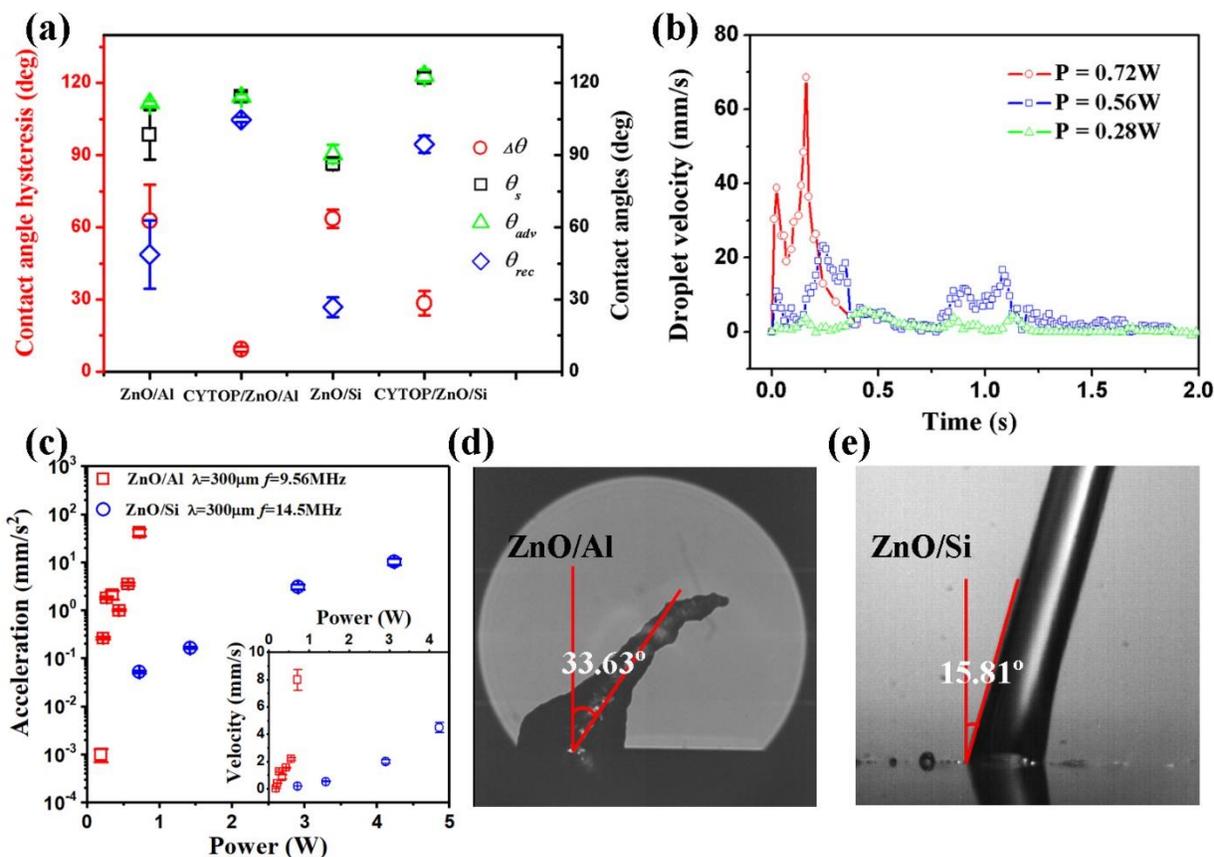


Figure 4. Contact angle measurements and pumping experiments on horizontal surfaces. (a) Comparison of the static contact angle θ_s , the receding contact angle θ_{rec} , and the advancing contact angle θ_{adv} , and hence the contact angle hysteresis, $\Delta\theta = \theta_{adv} - \theta_{rec}$ of water droplets (1 μL) on ZnO/Si and ZnO/Al surfaces before and after CYTOP coating; (b) Pumping velocity as a function of time with different input powers when the water droplet is driven by an acoustic wave on a 600 μm thick horizontal Al plate; (c) Average acceleration calculated from videos of horizontal pumping on ZnO coated 600 μm thick Al plates and ZnO coated Si wafer increasing with the input power. The wavelength of the acoustic wave was 300 μm for both devices. The inset figure shows

the average velocity (over time). Rayleigh angles of surface acoustic wave can be observed from the jetting trajectory of 2 μL water droplets on **(d)** ZnO/Al device (jetted at 8 W) and **(e)** ZnO/Si device (jetted at 14 W).

This mechanism enables us to demonstrate movement on inclined surfaces (Fig. S3) by tilting the device plate into different orientations, e.g., inclined angled slopes, vertical and up-side down positions. Here, an important consideration is the droplet size, as it has a significant influence on pumping at different inclination angles due to the effect of gravity. This is especially important at inclination angles around 90° , and we experimentally determined a maximum volume for our device to be 3 μL . Larger droplets cannot be pumped upwards but move downwards due to gravity.

Demonstration of programmable, flexible and bendable acoustofluidics

To demonstrate the concept of programmable, flexible and bendable three-dimensional acoustofluidics using hierarchical CYTOP/ZnO/Al nanostructured surfaces, we deformed the CYTOP/ZnO/Al (600 μm thick) device into a circle (see schematic illustration in Fig. 1a). Figures 5a to 5c show that a water droplet could be efficiently pumped forward on different segments of its surface with a curvature varying from 54.5 m^{-1} to 85.7 m^{-1} .

To further extend the flexibility of our platform, we reduced the thickness of Al sheets with the corresponding acoustic wave modes changing from those associated with Rayleigh waves to Lamb waves. Acoustic wave devices were designed and fabricated on 200 μm -thick ZnO/Al sheets, and deformed in shapes of a “roller coaster” (Figs. 5d and 5e, and Movies S1 and S2). Finally, we also

show that the droplet can be easily pumped along arbitrary topographies using a randomly twisted CYTOP/ZnO/Al (50 μm -thick) sample (Fig. 5f and Movie S3).

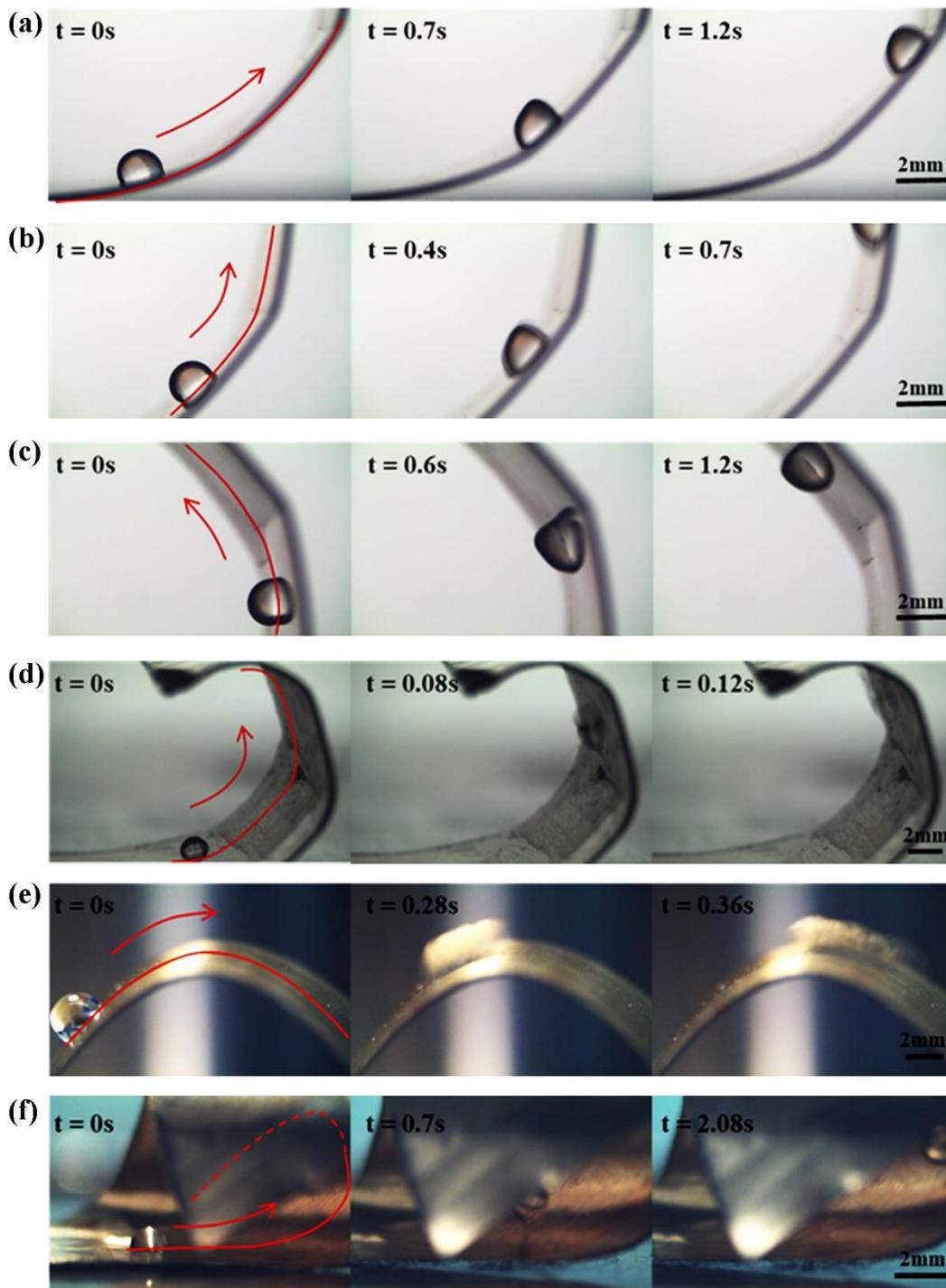


Figure 5. Experimental demonstration of 3D pumping driven by ZnO based acoustic wave on bendable substrates. (a)-(c) 1 μL droplets moving at different stages on curved surfaces ($P = 15\text{W}$). (d) 0.5 μL droplet moving on a ‘roller coaster’ formed by curled acoustic wave devices ($P = 20\text{W}$). (e) 2 μL droplet moving past a hilly surface ($P = 6\text{W}$). (f) 0.5 μL droplet moving on a twisted surface ($P = 3\text{W}$).

Conclusions

In summary, we designed nanostructured surfaces using hierarchical textures to optimize shear hydrophobicity and tensile hydrophilicity to tune droplet transport and adhesion on arbitrarily oriented devices. We demonstrated that the CYTOP/ZnO/Al sheets enabled a significantly better actuation performance when compared to conventional ZnO/Si acoustic wave devices. These CYTOP/ZnO/Al sheets were uniquely capable of efficiently driving droplets across a wide range of inclination angles ($0^\circ < \alpha \leq 180^\circ$). Our designed devices can be flexibly deformed and then maintain their shape, while enabling efficient liquid pumping. We envisage that the surface design principles and demonstrated devices/applications will be pivotal for liquid sampling for smart diagnostic systems within mobile and disposable settings.

Methods

Sample preparation. ZnO/Al and ZnO/Si substrates were prepared using magnetron sputtering (NS3750, Nordiko). ZnO films with thicknesses of 5 μm were deposited on 50 μm thick Al foils, 200 μm and 600 μm thick Al thin plates, and 500 μm thick 4-inch (100) Si wafers in vacuum with a pressure of 0.65 Pa. A 99.99 % pure Zn target was used while Ar/O₂ gases with flow rates of 10/13 sccm were introduced to form a plasma and to provide oxygen ions (power, 400W).

The acoustic wave devices included interdigital transducers (IDTs) on top of the ZnO thin films, fabricated through photolithography and lift-off processes. The Cr/Au electrodes with thickness of 20 nm/100 nm were deposited using a thermal evaporator. For hydrophobic surface treatment, 1% CYTOP solution (L-809A) was dip-coated on ZnO surface and heated to 180 °C for 1 min.

Experimental methods. The surface roughness of all samples was characterized using an interferometer (Bruker Contour GT-K) and an AFM (Veeco Dimension 3100). The static, advancing and receding contact angles were measured using sessile droplets and volume addition and withdrawal on a droplet shape analyzer (Kruss DSA30S). In the acoustofluidic experiments, the acoustic wave devices were connected to an RF signal generator (9 k-2.4 GHz 2024, Marconi Instruments) through a power amplifier (Model 75A250, Amplifier Research). The movements of water droplets were imaged by both a high-speed camera (Phantom V2511, 3000 FPS) and a standard CMOS camera.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at <http://pubs.acs.org>.

Figure S1. Surface morphology of 600 μm thick Al substrate.

Figure S2. Deformation of 1 μL droplet in the movement driven by acoustic waves.

Figure S3. Droplets moving on various tilted flat surfaces.

Table S1. Contact angle hysteresis measurements.

Movie S1. Droplet moving on a ‘roller coaster’.

Movie S2. Droplet moving past a hilly surface.

Movie S3. Droplet moving on a twisted surface.

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Author Contributions

Y.Q. F., G. M., J. R. and J. M. C. proposed the idea. R. T., Y.Q. F., J.T. L. and P. C.-P. performed the experimental work. R. T. and Y.Q. F. wrote the manuscript and performed analysis. G. M., J. R., H. T., Q. W., J. L., X. Y., J. Z. and J. M. C. were involved in writing, discussions and modifications of the article.

Notes

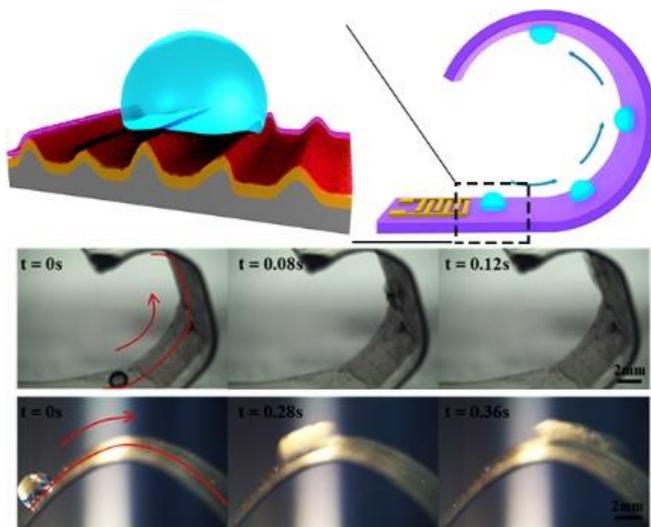
The authors declare no competing financial interest.

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TOC GRAPHIC



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