Droplet Manipulation on a Hydrophobic Textured Surface With Roughened Patterns

Jing-Tang Yang, Julia C. Chen, Ker-Jer Huang, and J. Andrew Yeh

Abstract—A novel concept is proposed and verified, experimentally and theoretically, to manipulate droplets without external power sources. The proposed device is a hydrophobic surface containing specific roughness gradients, which is composed of several textured regions with gradually increased structural roughness. Hydrophobic materials of four types, photoresist AZ6112, Teflon, Parylene C, and plasma polymerization fluorocarbon film (PPFC)—are adopted to fabricate the textured surfaces, and are tested. Acting forces come from the different Laplace pressures exerted on a droplet across various hydrophobic surfaces, whereas resistance forces come from the contact-angle hysteresis. Two patterns of devices are shown in this article—chain-shaped and concentric circular. The former forms as a droplet transport route and the latter provides both transport and orientation functions. Theoretical estimation and experimental verification of the droplet motion, including actuation and resistance forces, on the devices are conducted. Optimal design is achieved based on accurate estimations of the acting forces. The proposed device provides a simplified fabrication process and shows superior biocompatibility for droplet manipulation in microfluidic systems.

Index Terms—Contact angle, droplet actuation and manipulation, microfluidics, textured surface.

I. INTRODUCTION

A technique for fluid transport is a key issue for the development of microfluidic systems, especially for chemical analytic and bioassay systems that must respond rapidly and be disposable and inexpensive. Surface tension overpowers the inertial force and becomes a dominant force in microscale systems; fluidic actuation driven by surface tension has, therefore, drawn much attention of researchers. Various mechanisms for droplet manipulation are based on thermal [1], electrical [2], [3], and photoresponsive [4], [5] principles, but inherent problems, such as chemical compatibility, temperature rise, and interference of electric potential, might induce side effects in bioapplications.

A surface wettability gradient has long been applied to control droplet transport. Such autonomous motion typically results from the nonequilibrium Laplace pressure generated on opposite sides of the droplet. Gallardo et al. [4], [5] created surface tension gradients using electrochemical principles; they created the magnitude and direction of spatial surface-tension gradients by generating surfactant species at one electrode and consuming them at another. Droplets of organic liquids have been guided through simple fluidic networks. Ichimura et al. [6] proposed a light-driven method; the surface free-energy gradient was generated photochemically by asymmetrical photoirradiation of a photoisomerizable monolayer covering a substrate surface. The motion of the liquid was manipulated reversibly by light. Daniel et al. [7], [8] combined Marangoni flow caused by a temperature gradient interior to a droplet with the directional flow generated by a substrate surface wettability gradient. Such a surface wettability gradient was prepared on depositing hydrophobic molecules on the central part of a hydrophilic surface and a temperature gradient was created on circulating cold fluid beneath it. A droplet was propelled toward the region of greater wettability much more rapidly than for typical Marangoni flows.

Previous work reveal that surface wettability can be tuned by microstructure geometry [9]–[12]. Shen et al. [13] theoretically analyzed and experimentally tested forces to actuate a mercury droplet on various structured surfaces; they reported that adhesion forces of droplets can be designed by controlling the surface roughness. Thus the design of surface patterns can play a significant role in microfluidic devices.

He and Lee [14] presented a roughness-switchable device, comprising flat and roughened polydimethylsiloxane (PDMS) surfaces. When a droplet was deposited across the boundary between flat and rough surfaces, it moved away from the boundary. This phenomenon indicated that a droplet tended to move from a more hydrophobic surface toward a less hydrophobic one. However, the motion described in their experiment might be regarded as a droplet collapsing process rather than transport motion, because the droplet moved no further than its base width.

The discoveries of previous work reveal that surface tension can be controlled by varying either physical or chemical properties of the surfaces. In this work, we attempt to create surface wettability gradients by patterning microstructure distributions and to develop a droplet manipulating device based on varied Laplace pressures of a droplet across distinct hydrophobic surfaces. The Laplace-pressure-driven mechanism provides a favorable concept for designing microfluidics systems because of its feature of no consumption of external power and lack of undesirable side effect from thermal, chemical, electrical problems.
II. BASIC CONCEPTS AND THEORY

A. Contact Angle

The intrinsic contact angle $\theta$ of a liquid droplet on a flat homogeneous solid surface is given by the classical Young's equation [15].

$$\cos \theta = \frac{\gamma_{SV} - \gamma_{SL}}{\gamma_{LV}}$$  \hspace{1cm} (1)

in which $\gamma_{SV}$, $\gamma_{SV}$, and $\gamma_{SL}$ are the interfacial free energies per unit area of the solid-vapor, liquid-vapor, and solid-liquid surface, respectively. As for the rough surface, Wenzel [16] proposed a theoretical model describing the contact angle $\theta'$ of a liquid droplet on the surface by modifying (1) as follows:

$$\cos \theta' = \frac{r(\gamma_{SV} - \gamma_{SL})}{\gamma_{LV}} = r \cos \theta$$  \hspace{1cm} (2)

in which $r$ is a roughness factor, which is defined as the ratio of actual surface area of a rough surface to the geometric projected area. Because a larger $r$ corresponds to a larger $\theta'$, hydrophobicity of a noncomposite surface is enhanced through increasing the solid-liquid contact area.

The composite interface beneath a suspended droplet contains both liquid-vapor and liquid-solid interfaces. Cassie and Baxter [17] proposed an equation to describe the apparent contact angle on a composite surface as follows:

$$\cos \theta' = f_1 \cos \theta_1 + f_2 \cos \theta_2$$  \hspace{1cm} (3)

in which $\theta_1$ and $\theta_2$ are the contact angles of the droplet on each surface, and $f_1$ and $f_2$ are the fraction of solid-liquid and gas-liquid interfaces below the droplet. As $f_2$ refers to the liquid-gas interface, $\cos \theta_2$ equals $-1$ in (3) for the suspended droplet (i.e., $\theta_2 = 180^\circ$). As $\theta'$ is greater for $f_1$ being smaller, the hydrophobicity of the composite surface is enhanced through decreasing the solid-liquid contact area.

B. Laplace Pressure

The pressure difference between the droplet and the surrounding medium $\Delta p$ can be estimated from the Laplace-Young equation [18]

$$\Delta p = \gamma_{LV} \left( \frac{1}{r_1} + \frac{1}{r_2} \right)$$  \hspace{1cm} (4)

in which $r_1$ and $r_2$ are the two radii of curvature of the surface. Whereas a droplet is deposited across the boundary between two areas with different roughness factors, there exists a pressure difference inside the droplet that results from the difference of radii of curvatures through the contact-angle difference. This pressure difference can move the droplet across the boundary between flat and roughened surfaces [14], [19]. Therefore, the droplet moves from the more hydrophobic region toward the less hydrophobic one.

C. Contact Angle Hysteresis

Contact angle hysteresis plays an important role with the surface dynamic wettability taken into consideration. The force $F$ needed to initiate motion of a drop moving over a solid surface [20], [21] is

$$F = \gamma_{LV} \cdot l \cdot (\cos \theta_R - \cos \theta_A)$$  \hspace{1cm} (5)

in which $l$ is the characteristic length of the droplet, and $\theta_A$ and $\theta_R$ are the advancing and receding contact angles, respectively. Dettre and Johnson [22] theoretically simulated the effect of surface roughness on contact angle hysteresis and suggested that hysteresis arises from surface roughness. However, hysteresis decreases greatly on a very rough surface, i.e., a droplet suspended atop the asperities and resolves into the composite configuration consisting of liquid-vapor and liquid-solid interfaces.

D. Droplet Transport Mechanism

When a droplet is placed on a sequence of textured surface regions with gradually increasing or decreasing densities of roughness patterns, it might move along these regions spontaneously by virtue of the unbalanced Laplace pressures, according to (4). Concurrently, the resistance force during the movement is induced due to the viscous effect between the droplet and the textured surface, and, is regarded as the contact angle hysteresis in this work. The mechanism of droplet transport is accordingly regarded as the reaction of the unbalanced Laplace pressures and contact angle hysteresis.

The power resource of the entire motion results from the induced Laplace pressure whenever the droplet is located on a boundary between two surfaces with different roughness. Hence a droplet moves continuously along a string of rough surfaces if the route is designed properly. Moreover, the moving process occurs spontaneously without relying on any external power source. The actuation force of the motion, $F_{act}$, can be expressed with an equation modified as follows:

$$F_{act} = \int \gamma_{LV} \left( \frac{1}{r_1} + \frac{1}{r_2} \right) ds_{left} - \int \gamma_{LV} \left( \frac{1}{r_1'} + \frac{1}{r_2'} \right) ds_{right}$$

$$+ \gamma_{LV} \cdot A_{eff} \cdot \left\{ \left( \frac{1}{r_1} + \frac{1}{r_2} \right) - \left( \frac{1}{r_1'} + \frac{1}{r_2'} \right) \right\}$$  \hspace{1cm} (6)

in which $A_{eff}$ is the droplet cross-sectional area corresponding to the maximum height, which is derived in the following section of theoretical analysis and discussion. The projection of the area $A_{eff}$ is orthogonal to the moving direction. $r_{1,2}$ and $r'_{1,2}$ represent the radii of curvature of the distinct regions on which the droplet lies simultaneously.

The droplet sticks on a surface when the resistance force is greater than the actuating force. Contact angle hysteresis is crucial in designing the route. As the droplet moves in only one direction, the base width of the droplet should be chosen the same as the characteristic length in (5) [23]. The resistance of the motion $F_{res}$ is

$$F_{res} = \gamma_{LV} \cdot f_1 \cdot w \cdot (\cos \theta_R - \cos \theta_A)$$  \hspace{1cm} (7)

in which $w$ is the droplet base width in a direction perpendicular to motion.
III. EXPERIMENTAL ANALYSIS

A. Microfabrication

Four types of hydrophobic materials, photoresist AZ6112, Teflon, Parylene C, and plasma polymerization fluorocarbon film (PPFC), were tested in this paper. The microstructures on the surfaces of these four materials were prepared through the standard lithography process.

Microstructures were fabricated on silicon wafer by the following procedures. Photoresist AZ6112 (PR) was first spun on silicon substrates, followed by the standard lithography process to create the tested pattern. Microstructure covered with Teflon, Parylene C, and PPFC was fabricated with the same procedures as AZ6112 with additional process described as follows. Inductively coupled plasma (ICP) was chosen to create surface microstructures using AZ6112 as etch mask. The hydrophobic material, Teflon, Parylene C, or PPFC, was coated after AZ6112 was removed. PPFC has the chemical compound of C₄F₈ and it was deposited using plasma enhanced chemical vapor deposition (PECVD). Teflon was spin coated on the substrates, followed by baking at 200 °C. Parylene C was deposited by vapor deposition, providing a conformal coating on the substrates. Microstructures of Parylene C were patterned using oxygen plasma.

B. Contact Angle Measurement

The sessile drop method was used for contact angle measurements with a commercial contact angle meter (OCA 20 Dataphysics Instruments, Germany). The volume of the deionized (DI) water used for these measurements was 3 μL. The equivalent diameter of the spherical droplet is presumed to be 1.8 mm in the air. The room temperature was controlled to be 25 °C and the corresponding surface tension was about 72.3 × 10⁻³ N/m. Sequential photographs during the droplet movement were taken every 20 ms with the contact angle meter equipped with a CCD video camera. Contact angles were measured at four points and the averaged values were then calculated for each sample. Dynamic advancing and receding contact angles were measured by the motor-driven and software control instrument (TBU 90E, Germany) with inclination up to an angle of 90°. Detailed microstructures were inspected by both scanning electron microscopy (SEM) and optical microscopy.

C. Static Contact Angle

Droplets were placed on different textured regions of these four hydrophobic surfaces (AZ6112, Teflon, Parylene C, and PPFC, respectively). All these textured regions were formed by the same groove structure, 5 μm in width and 1000 μm in length. The separations among the grooves, i.e., the f₁ values, were different in each region as shown in Fig. 1. Table I shows the measured contact angle in each region of these four hydrophobic surfaces, where data in the f₁ = 1 row are the intrinsic contact angles. θm represents the measured contact angle whereas θk represents the theoretical contact angle obtained from (3). Since θm and θk are quite coincident in the experiment, it is reasonable to anticipate the apparent contact angles by designing the f₁ values of the textured surfaces. Fig. 2 shows the droplet deposited on four AZ6112 surfaces with different f₁ values. Surfaces with lower f₁ values result in higher contact angles. These droplet images further indicate that droplets with higher contact angles correspond to lower radii of curvature. As a result, the design of the roughness patterns influences both the droplet apparent contact angle and the Laplace pressure.

D. Contact Angle Hysteresis

The strategy to measure the contact angle hysteresis on each textured surface is to place a sessile droplet on it, and then tilt the surface until the droplet begins to slide. The angle subtended at the front of the droplet is the advancing contact angle (θA), whereas that of the rear is the receding contact angle (θR). Fig. 3 shows a droplet on a 24° tilted PPFC surface with groove structures f₁ = 0.67. Table II lists the measured (θA) and (θR) on differently hydrophobic surfaces of AZ6112, Teflon, Parylene C, and PPFC. Not available (NA) represents the situation that droplet sticks on the surface even its tilting angle has reached 90°. This table shows that droplet adhesion occurs easier on less hydrophobic ones, such as AZ6112 and Parylene C surfaces with smaller f₁ values. Contact angle hysteresis (Δθ = θA - θR) decreases with f₁ value. However, this proportional relationship is not obvious on Teflon surfaces. The experimental inaccuracy might come from the defective coating of Teflon surfaces and need further verification. The uncertainty analysis of the contact angle measurement with respect to various f₁ values and to different patterns on Teflon surface (at 95% confidence level) is shown in Tables III and IV, respectively.

E. Device Test

The testing device is constituted with five textured regions with gradually increasing or decreasing f₁ values. Several previous theoretical and experimental studies have shown that sharp edges on the solid could be an important cause on contact angle hysteresis. The grooved surface thus shows a better water-shedding property in the parallel direction because of the low energy barrier for the movement of the three-phase line in this sliding direction [9], [24], [25]. For this reason, the grooved structure is chosen to form the textured regions of the device. Two configurations of the surface texture, chains and concentric circles, are designed for testing the proposed concept and the

Fig. 1. SEM Photos of the grooved surfaces with various f₁ values.
TABLE I
CONTACT ANGLE (°) FOR GROOVED SURFACES WITH VARIOUS \( f_i \) VALUES

<table>
<thead>
<tr>
<th>( f_i )</th>
<th>AZ 6112</th>
<th>Teflon</th>
<th>Parylene C</th>
<th>PPFC</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \theta_m )</td>
<td>( \theta_i )</td>
<td>( \theta_m )</td>
<td>( \theta_i )</td>
<td>( \theta_m )</td>
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<td>1</td>
<td>113</td>
<td>120</td>
<td>92</td>
<td>125</td>
</tr>
<tr>
<td>0.67</td>
<td>127</td>
<td>126</td>
<td>132</td>
<td>132</td>
</tr>
<tr>
<td>0.50</td>
<td>134</td>
<td>134</td>
<td>134</td>
<td>139</td>
</tr>
<tr>
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<td>148</td>
<td>151</td>
<td>151</td>
</tr>
<tr>
<td>0.14</td>
<td>157</td>
<td>156</td>
<td>158</td>
<td>158</td>
</tr>
<tr>
<td>0.10</td>
<td>159</td>
<td>160</td>
<td>160</td>
<td>162</td>
</tr>
<tr>
<td>0.08</td>
<td>160</td>
<td>162</td>
<td>162</td>
<td>164</td>
</tr>
</tbody>
</table>

As long as the droplet does not collapse, it shows no direct correlation with the apparent contact angle. The contact angle varies with the density of the textured structure, \( f_i \); the greater the value of \( f_i \), the apparent contact angle is much closer to the intrinsic angle.

The chain configuration on the surface functions as droplet transporting route, and the concentric circular configuration provides both transporting and orientation functions. Two sets of \( f_i \) value sequences used for chain configuration: (1) 0.1, 0.25, 0.5, 0.8, 1; and (2) 0.1, 0.2, 0.35, 0.65, 1. To achieve the function of orientation, the gradient of \( f_i \) value should decrease with increasing radial direction on the surface of concentric circular configuration, as shown in Fig. 4(b). The \( f_i \) value changes gradually in each concentric circular region as shown in Fig. 5. For example, the \( f_i \) distribution between \( f_{i,0} = 0.8 \) and 0.17 would be \( f_{i,x} = 0.8 - 0.63 \cdot x \) for \( x = 1 \text{ mm} \). Two sets of \( f_i \) value at most inward part of each concentric circular region are: (1) 0.01, 0.05, 0.17, 0.8, 1; and (2) 0.01, 0.04, 0.11, 0.67, 1. In other words, the hydrophobicity of concentric circular device is lower inward. Three joint patterns between each neighboring regions,
TABLE II
ADVANCING AND RECEDING CONTACT ANGLES (°) FOR GROOVED SURFACES WITH VARIOUS $f_1$ VALUES

<table>
<thead>
<tr>
<th>$f_1$</th>
<th>$\theta_A$</th>
<th>$\theta_R$</th>
<th>$\theta_A$</th>
<th>$\theta_R$</th>
<th>$\theta_A$</th>
<th>$\theta_R$</th>
<th>$\theta_A$</th>
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<td>NA</td>
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<td>123</td>
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<td>NA</td>
<td>138</td>
<td>122</td>
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<td>NA</td>
<td>140</td>
<td>135</td>
</tr>
<tr>
<td>0.25</td>
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<td>149</td>
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<td>142</td>
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<td>134</td>
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<td>152</td>
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<tr>
<td>0.08</td>
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<td>144</td>
<td>149</td>
<td>145</td>
<td>156</td>
<td>151</td>
</tr>
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</table>

TABLE III
THE UNCERTAINTY ANALYSIS OF THE CONTACT ANGLE MEASUREMENT WITH RESPECT TO VARIOUS $f_1$ VALUES ON TEFLOL SURFACE (AT 95% CONFIDENCE LEVEL)

<table>
<thead>
<tr>
<th>$f_1$</th>
<th>$\theta_{AVG}$</th>
<th>$\theta_L$</th>
<th>$\theta_U$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>121°</td>
<td>119°</td>
<td></td>
</tr>
<tr>
<td>0.08</td>
<td>159°</td>
<td>156°</td>
<td></td>
</tr>
<tr>
<td>0.10</td>
<td>159°</td>
<td>140°</td>
<td></td>
</tr>
<tr>
<td>0.25</td>
<td>156°</td>
<td>144°</td>
<td></td>
</tr>
<tr>
<td>0.50</td>
<td>156°</td>
<td>137°</td>
<td></td>
</tr>
<tr>
<td>0.67</td>
<td>156°</td>
<td>137°</td>
<td></td>
</tr>
</tbody>
</table>

TABLE IV
THE UNCERTAINTY ANALYSIS OF THE CONTACT ANGLE MEASUREMENT WITH RESPECT TO DIFFERENT PATTERNS ON TEFLOL SURFACE (AT 95% CONFIDENCE LEVEL)

<table>
<thead>
<tr>
<th>patterns of microstructure</th>
<th>$\theta_{AVG}$</th>
<th>$\theta_L$</th>
<th>$\theta_U$</th>
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</thead>
<tbody>
<tr>
<td>◊</td>
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<td>156°</td>
<td>140°</td>
</tr>
<tr>
<td>◯</td>
<td>159°</td>
<td>140°</td>
<td>157°</td>
</tr>
<tr>
<td>groove</td>
<td>159°</td>
<td>159°</td>
<td>159°</td>
</tr>
<tr>
<td>△</td>
<td>159°</td>
<td>159°</td>
<td>159°</td>
</tr>
<tr>
<td>□ 5 μm</td>
<td>156°</td>
<td>156°</td>
<td>155°</td>
</tr>
<tr>
<td>□ 15 μm</td>
<td>156°</td>
<td>155°</td>
<td>153°</td>
</tr>
<tr>
<td>□ 64 μm</td>
<td></td>
<td>153°</td>
<td>153°</td>
</tr>
</tbody>
</table>

overlapping, connecting, and separating, are also tested and the corresponding SEM photos are shown in Fig. 6(a), (b), and (c).

Once a droplet is extracted from the dosing needle onto the boundary of two regions, it moves continuously along the ascent of the $f_1$ values and stops on the smooth region ($f_1 = 1$) at last. For the device with concentric circular texture, the droplet moves inward continuously and pins at the central circle at last. The sequential pictures in Fig. 7 clearly depict the temporal and spatial variations of the droplet locations on the textured surface during the moving process. The $f_1$ distribution in Fig. 7 from left to right equals 1, 0.67, 0.11, and 0.04, respectively. The total moving length is about 2.5 mm and the average velocity of this droplet is about 62.5 mm/s.

In the experiments conducted, the droplet only moved on the surfaces of overlapping and connecting patterns. For the case of separating pattern, the droplet was unable to run across the gap between the bordered regions, and, thus, moved discontinuously along the route as shown in Fig. 8. Besides, the droplet collapses on the region of $f_1 = 0.01$ due to the wider gap of the grooved structures. Since the collapse phenomenon accompanies the large contact angle hysteresis as mentioned above, the droplet deposited on this region sticks tightly. Consequently, the moving behavior is sensitive to the surface energy barriers, such as the joint gap and contact angle hysteresis. It is necessary to avoid the gap and lower the hysteresis effects while designing such kind of device.

IV. THEORETICAL ANALYSIS AND DISCUSSION

Two dominant factors in designing the devices are: (a) the $f_1$ value distributions of each textured region, and (b) the contact angle hysteresis along the route. The former relates to the actuating force, whereas the latter to the dragging force of the droplet movement. The effects of these two factors are discussed as follows.

A. Actuating Force: Laplace Pressure

According to the Cassie’s equation, referring to (3), the apparent contact angle is governed by the $f_1$ value. The Laplace
equation reveals that the pressure difference is related to the droplet curvature radius, which is influenced by the \( f_1 \) value discussed in the previous experimental analysis. Therefore, deduction of the correlation between \( \theta \) and \( \Delta p \) provides the way to determine the droplet pressure difference between adjacent regions and to design the favorable distribution of \( f_1 \) value for actuating the droplet. When the force of surface tension is significantly greater than the gravity force, the contour of the droplet can be approximated by a section of a sphere and the following simple geometric relations are used:

\[
V = \frac{\pi}{3} h^2 (3r_1 - h) \quad (8)
\]

\[
h = r_1(1 - \cos \theta) \quad (9)
\]

where \( V \) is the volume of the droplet, \( h \) is the maximum height of the droplet, and \( r_1 \) is the curvature radius. Deriving from (8)

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**Fig. 4.** SEM photos of the textured surfaces. (a) Chain configuration. (b) Concentric circular configuration.

**Fig. 5.** The \( f_1 \) value distribution schematics of concentric circular configuration.

**Fig. 6.** SEM photos of three joint patterns between adjacent regions. (a) Overlapping. (b) Connecting. (c) Separating.
and (9), an equation that describes relationship between $r_1$ and $\theta$ is given as follows:

$$r_1 = \left\{ \frac{1}{4} \cdot (2 - 3 \cos \theta + \cos^3 \theta) \right\}^{\frac{-1}{3}} \cdot r_0. \quad (10)$$

Fig. 9 plots the correlation between the apparent contact angle and the curvature radius of a droplet settling on the textured surface. The line indicates the theoretically predicted curvature radii calculated from (10). The circle, triangle, square, and diamond marks represent the calculated curvature radii of Teflon, Parylene C, AZ6112, and PPFC surfaces, respectively, by substituting the experimental data of the droplet height and apparent contact angle into (9). Both theoretical and experimental results in Fig. 9 reveal that the curvature radii of a droplet decrease rapidly with increasing contact angle in regions of smaller contact angle ($\theta = 90^\circ - 130^\circ$). However, the curvature radii of experimental calculations are mostly greater than those of theoretical predictions. This disagreement could results from the spherical contour hypothesis of (10) since the contour of droplet on the grooved surface might deform on the heterogeneous surface. The Bond number (gravitational force/surface tension force) is estimated to be 0.436. The effect of gravitational force is about 40% of surface tension force. If the size of the droplet is further reduced to be 100 $\mu$m or the fluid with greater surface tension is adopted for experiment, the effect of surface tension should be more profound. More experimental work is needed in the future to deduce the more precise correlations between the curvature radius and contact angle for each kind of the textured surface.

The actuating force is estimated by substituting the radius on each side of the droplet into (6), although it is difficult to estimate precisely both $r_1$ and $r_2$ from experimental images. Basically, $r_1$ and $r_2$ are decided on the triple line and Fig. 10(a),
in Fig. 9, and the modified correlation between $\theta$ and $r_2$ for each material is obtained. The actuating forces within the device are calculated through the procedure described below. First, the design parameters ($f_1$ values) are substituted into (3) to estimate $\theta$ of each textured region. Then, the curvature radius is estimated by the correlation between $\theta$ and $r_1$ above. Finally the actuation force is calculated by substituting $w$, $\theta$, and $r_1$ into (11) and (12).

Tables V and VI list the calculated results of these two sets of designs on PPFC surfaces. The column of force represents the actuation force between two neighboring regions in the table. For example, the actuation force of chain configuration in case 1 between $f_1 = 0.8$ and $f_1 = 1.0$ equals to $4.7 \times 10^{-6}$ N, and so on. In case 2 of chain configuration, each contact angle difference between neighboring regions is about $10^\circ$, whereas the inducing force is higher in less hydrophobic (higher $f_1$ value) regions. This phenomenon is well correlated with the trends shown in Fig. 9. The gradients of the curves are greater and thus the actuation forces are all greater in the regions of smaller contact angle.

B. Resistance Force: Contact Angle Hysteresis

The force for starting the droplet movement is expressed as (7). After substituting into (7) the experimental data of the advancing and receding contact angles in Table II, the resistance forces are obtained. Fig. 11 shows the correlations between the apparent contact angle and the resistance force for surfaces of Teflon, Parylene C, AZ6112, and PPFC, respectively. The line represents the curve of PPFC data fitted with a power equation. The figure reveals that the hysteresis force decreases with increasing contact angle for each surface and the trend is consistent with the water-repellent phenomenon of the superhydrophobic surface. For the test that the droplet slides downward on an inclined plane, the actuation force is gravity and is used to estimate the resistance force (or the hysteresis effect). In our experiment, the hysteresis forces corresponding to contact angles below $130^\circ$ are too large that the droplet sticks on the surface tightly. Further experiments will be needed to develop the more accurate correlation between the hysteresis force and the contact angle for each kind of surface.

The PPFC textured surface is taken as an example to discuss forces exerted on a droplet during the moving period. The resistance force on PPFC surfaces with chain-configuration grooves at corresponding contact angles are listed in Tables V and VI by reading PPFC fitting curve from Fig. 11. Both forces of actuation and resistance on each boundary of cases 1 and 2 are also contrasted in the table. The actuation force is invariably greater than the resistance force in each boundary of case 2. Data of case 1, however, show a poorer force allotment (at boundary between $f_1 = 0.8$ and $f_1 = 1.0$) due to the improper arrangement of $f_1$ value. This reasonably interprets the better actuating performance of devices of case 2 in the experimental analysis. The forces of actuation and resistance on PPFC surfaces with concentric circular-configuration grooves listed in Table VI reveal similar trends.

According to the foregoing, the actuating and dragging forces are both important to droplet moving behavior. Thus these two forces should be estimated simultaneously while designing the device. For the actuating force, the slope of the curvature radius.
seems to be sharper at lower contact angle sections. In other words, we can obtain the same pressure drop with smaller contact angle difference at lower contact angle sections. From this, it seems to be more economical to design the $f_1$ distributions in the sections of lower contact angle. However, when considering the hysteresis force, the lower contact angle sections have larger dragging forces. After obtaining the accurate relationships between the droplet curvature radius versus contact angle and hysteresis force versus contact angle, the optimal design of the $f_1$ distributions can be achieved.

Besides the considerations aforementioned, there are still some details worth noticing while designing the device. For example, the width of each textured region should never be longer than the droplet baseline to ensure its contact with at least two different values textured regions during the moving process. Furthermore, the droplet size is also an important factor to the moving behavior. Smaller droplet sizes will be favorable for the droplet transport process for it can reduce the gravity effect and enhance the surface tension. Choice of materials influences the hysteresis force. Functions of the devices can be variable by

### Table V
**The Actuation and Resistance Forces on PPFC Surfaces with Chain-Configuration Grooves**

<table>
<thead>
<tr>
<th>Case 1 $f_1$</th>
<th>$\theta$(deg)</th>
<th>$r_1$(mm)</th>
<th>$A_{eff}$ (mm$^2$)</th>
<th>$F_{act}$ ($10^{-3}$ N)</th>
<th>$F_{res}$ ($10^{-3}$ N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>125</td>
<td>1.000</td>
<td>1.00</td>
<td>4.20</td>
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<tr>
<td>0.80</td>
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<td>0.980</td>
<td>3.09</td>
<td>0.47</td>
<td>1.49</td>
</tr>
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<td>0.50</td>
<td>142</td>
<td>0.952</td>
<td>3.14</td>
<td>0.68</td>
<td>0.25</td>
</tr>
<tr>
<td>0.25</td>
<td>153</td>
<td>0.937</td>
<td>3.31</td>
<td>0.41</td>
<td>0.05</td>
</tr>
<tr>
<td>0.1</td>
<td>163</td>
<td>0.934</td>
<td>4.14</td>
<td>0.09</td>
<td>0.012</td>
</tr>
</tbody>
</table>

### Table VI
**The Actuation and Resistance Forces on PPFC Surfaces with Concentric Circular-Configuration Grooves**

<table>
<thead>
<tr>
<th>Case 1 $f_1$</th>
<th>$\theta$(deg)</th>
<th>$r_1$(mm)</th>
<th>$A_{eff}$ (mm$^2$)</th>
<th>$F_{act}$ ($10^{-3}$ N)</th>
<th>$F_{res}$ ($10^{-3}$ N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>125</td>
<td>1.000</td>
<td>1.00</td>
<td>4.20</td>
<td></td>
</tr>
<tr>
<td>0.80</td>
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<td>0.9799</td>
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<td>0.474</td>
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</tr>
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<td>3.28</td>
<td>0.119</td>
<td>0.024</td>
</tr>
<tr>
<td>0.04</td>
<td>168</td>
<td>0.9330</td>
<td>5.13</td>
<td>0.006</td>
<td>0.005</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Case 2 $f_1$</th>
<th>$\theta$(deg)</th>
<th>$r_1$(mm)</th>
<th>$A_{eff}$ (mm$^2$)</th>
<th>$F_{act}$ ($10^{-3}$ N)</th>
<th>$F_{res}$ ($10^{-3}$ N)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>125</td>
<td>1.000</td>
<td>1.00</td>
<td>4.20</td>
<td></td>
</tr>
<tr>
<td>0.67</td>
<td>136</td>
<td>0.9656</td>
<td>3.09</td>
<td>0.813</td>
<td>0.651</td>
</tr>
<tr>
<td>0.11</td>
<td>162</td>
<td>0.9344</td>
<td>4.14</td>
<td>0.105</td>
<td>0.014</td>
</tr>
<tr>
<td>0.04</td>
<td>169</td>
<td>0.9330</td>
<td>5.13</td>
<td>0.006</td>
<td>0.005</td>
</tr>
</tbody>
</table>
properly arranging the distributions of these textured regions, such as transporting, mixing, or orientation.

V. CONCLUSION

In this paper, we propose and verify, experimentally and theoretically, the spontaneous transport mechanism of a single droplet by means of the hydrophobicity differences on various textured surfaces. A droplet on the patterned surface, which is constituted with a sequence of textured regions with gradually increased roughness, is capable of moving continuously along the ascent of the surface roughness. The transporting behavior of the droplet is irreversible and the velocity is about 62.5 mm/s. Two configurations of the micro-texture surfaces are fabricated, the shapes of chain and concentric circular. The former functions as a droplet transporting route and the latter provides both transporting and orientation functions. The performance of the device correlates to two factors: the distributions of roughness values on each textured region and the contact angle hysteresis along the route. The former relates to the actuation force, while the latter to the resistance force of the droplet movement. The actuation force and hysteresis force are estimated from correlations between the droplet curvature radius versus contact angle and hysteresis force versus contact angle. Thus, the optimal design of the roughness patterns is able to be achieved after establishing the accurate relationships among the control parameters.

Functions of the proposed droplet manipulation concept are variable and flexible, depending on appropriately arranging the roughness distributions of these textured regions. Moreover, the device needs no external power, and thus excludes the side effect problems in thermal, chemical or electrical applications. It provides a higher biocapability while applying for the biomicrofluidics products.

ACKNOWLEDGMENT

The authors would like to acknowledge Professor and Vice President W.-H. Chen of National Tsing Hua University, Taiwan, for providing instrument and measurement technique of surface tension. The authors express great appreciation to C.-S. Yu, J. Y. Hu, and Dr. C.-J. Chen, the Director of the Precision Instrument Development Center of NSC, for supporting the microfabrication work.

REFERENCES


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