DYNAMICS OF THE INTERFACE OF A GAS-LIQUID MENISCUS IN COMPLEX PDMS MICRO-GEOMETRIES

Jie Xu and Daniel Attinger

Laboratory for Microscale Transport Phenomena, Department of Mechanical Engineering, Columbia University, New York, NY 10027

ABSTRACT

This article describes the design and manufacturing of microfluidic chips, where an injected gas-liquid meniscus is used for actuation purpose. The dynamics of the meniscus during its controlled motion is studied. The critical pressures to move the meniscus are measured and compared to theoretical result. Three different micro-geometries are compared in term of their ability to stabilize the meniscus. Also, the meniscus response to ultrasonic excitation is studied. When the excitation is weak, traveling waves are found and their wavelength decreases with increasing excitation frequencies. When the ultrasonic excitation is stronger, subharmonic and superharmonic oscillations at the meniscus interface are observed.

INTRODUCTION

Actuators based on the controlled motion of a gas-liquid interface have applications in microelectromechanical systems (MEMS) [1-7]. The generation of bubbles is usually performed using either thermal [1] or electrolytic [2] methods. Both methods induce phase change, and the bubble growth occurs within milliseconds. During such fast transformation, the surrounding pressure, temperature and electrical field can experience drastic changes, with consequences for the liquid close to the bubble, for instance in biomedical applications. In this study, an alternative generation method is explored, where a gas-liquid meniscus is slowly injected from a microchannel into a microchamber, using a syringe pump. Since the bubbles in most MEMS applications are attached to at least one wall [2, 4, 5], the meniscus should perform similarly as a bubble. However, the control of its volume and of the pressure across its interface is much simpler. Also the system can be operated at room temperature. If we consider the actuation of the gas-liquid interface, rather than its generation, two types of actuation are useful for MEMS devices: 1) a large displacement (usually greater than 10 µm) can directly push mechanical parts [3] or pump a liquid [5]; 2) high-frequency (on the order of 100s of kHz [4]) oscillation of the interface with small oscillation amplitude can induce steady microstreaming that moves particles [7]. Using the first kind of motion, either thermally or electrolysis, Papavasiliou et al. [3] were able to displace a mechanical valve by about 100 µm, Deshmukh et al. [5] could drive a micropump at 0.5 µL/min (by periodically expanding and shrinking a bubble, also with the help of an one-direction microvalve) and Maxwell et al. [6] can manipulate particles by capturing and releasing them. Regarding the second kind of motion, Marmottant et al. [7] observed a strong microstreaming field, in which particles can be transported. Also Kao et al. [4] showed that a micro-rotor can be driven at a speed as high as 625 rpm by the microstreaming flow field induced by an oscillating bubble. In this paper, we describe the manufacturing and characterization of three types of microstructures made of glass and of cured Polydimethylsiloxane (PDMS). We first study the dynamics of large displacements of the meniscus (first kind of motion) by measuring the pressures needed to move the gas-liquid interface in a micro-geometry, and we classify the ability of different micro-features to trap the interface at a desired location. Then, we study the response of the meniscus interface to high-frequency oscillations.

CHIP DESIGN, FABRICATION AND ASSEMBLY

A typical microfluidic chip used in our study is shown in Figure 1: it involves a chamber fed by a fork-like network of four channels. Channels A, C are 400µm wide and Channel B is 1 mm wide. Channel D is 100 µm wide and is used to inject...
air into the water-filled chamber E. The control of the gas-liquid interface position is enhanced by microgeometrical features at the junction of Channel D and the chamber, as shown in Figure 3 to 5. The height of all micro-channels is 50 pm. The microfluidic chip is manufactured using soft lithography in the cleanroom of Columbia University, according to the following process. First, a 50µm thick SU-8 master (MicroChem) is cured on a silicon wafer with patterns transferred from a mask (CAD/Art Services Inc.). The chip is then manufactured from the master using PDMS Sylgard 184 Kit (Dow Corning). The PDMS chip is then covered with a PDMS cover plate and sandwiched between two glass slides held by lateral clamps. This type of assembly ensures that all channel walls are made of PDMS and have the same surface properties. For some experiments involving high-frequency oscillations of the interface, a ring-shaped piezoelectric actuator is embedded in the PDMS cover plate.

**EXPERIMENTAL SETUP**

![Experimental setup diagram](Image)

Figure 2 The experimental setup has three subsystems: a microfluidic platform, a piezoelectric actuation system and a visualization system. The microfluidic platform involves (A) a KDS 210 infusion/withdrawal syringe pump (flow rate range from 0.001µl/hr to 86 ml/min), (B) a Honeywell 143PC03D pressure sensor (pressure range ±17 kPa), (C) a right angle flow-switching valve (Upchurch). (During the experiment, we connect the valve C to syringe 2, so that water is infused into the system. Then, the valve is shifted to syringe 1 to add a controlled flow rate of air into the system and forms a meniscus.) (D) is an Optem long distance microscope with a Pixelink PL-776 high-speed camera. (E) is the microfluidic chip as shown in Figure 1. (F) is a ring shape piezoelectric transducer (2 mm thick). The electrical part involves a function generator (Agilent, 33120A), an amplifier (Krohn-Hite, 7600M), a frequency divider (BNC, 7010) and an oscilloscope (Agilent 54622A).

The experimental setup is described in Figure 2, and involves three subsystems: a microfluidic platform, a piezoelectric actuation system and a visualization system. The microfluidic system involves the microfluidic chip described above and allows for the controlled filling of the chamber and subsequent controlled injection of an air plug in channel D, using a KDS 210 syringe pump (flow rate range from 0.001µl/hr to 86 ml/min). Visualization is performed with an Optem long-distance microscope in the plane perpendicular to the microfluidic chip, with a space resolution of about 1 micrometer. A strobe microscopy technique is used for visualizing the shape taken by the meniscus while oscillating. A function generator (Agilent, 33120A) generates a square wave to both an amplifier (Krohn-Hite, 7600M) and a frequency divider. The amplifier controls the voltage, which is applied to the piezoelectric transducer, and the signal coming out from the frequency divider is connected to a delay generator (BNC, 7010), which controls the delay time between the illumination by a diode and the actuation of the piezoelectric transducer within one microsecond resolution. Therefore the diode frequency can be set to either the actuator frequency or half of its value, using the frequency divider.

The physical properties used in the experiments are described in Table 1. The surface tension, density and viscosity of water are from [8]. The contact angles are measured from the pictures using ScionImage.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Physical property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>σ</td>
<td>Surface tension of water</td>
<td>72.0 mN/m @ 25 ºC</td>
</tr>
<tr>
<td></td>
<td></td>
<td>69.6 mN/m @ 40 ºC</td>
</tr>
<tr>
<td>θ</td>
<td>Contact angle of water on PDMS</td>
<td>70º for receding</td>
</tr>
<tr>
<td></td>
<td></td>
<td>110º for advancing</td>
</tr>
<tr>
<td>ρ</td>
<td>Density of water</td>
<td>0.998 g/cm³</td>
</tr>
<tr>
<td>ν</td>
<td>Viscosity of water</td>
<td>900 µPas</td>
</tr>
</tbody>
</table>

Table 1 Physical properties

**CHARACTERIZATION OF MENISCUS DYNAMICS**

This section investigates the motion of the meniscus during the injection phase, as well as the response of the meniscus to ultrasonic excitation.

**Ability of three microgeometries to trap a contact line**

Repeatability is important in MEMS. Therefore we investigated the efficiency of microgeometrical features to trap the meniscus contact line during the injection process. Three types of features called pits, peaks and modified peaks are shown in the respective Figure 3 to 5, and are evaluated and compared. Sequence frames shown in Figure 3-5 show the time evolution of the interaction between the meniscus and the geometrical feature. From Figure 3, we observe that pits are not able to trap the meniscus contact line, while the peaks and modified peaks in Figure 4 and 5 can pin the meniscus contact line efficiently. A measure of the ‘pinning’ efficiency is the range of meniscus visible area for which the wetting line is pinned. Using image analysis of a movie taken during slow growth of the meniscus, we define S₂ as the visible area at the time when the wetting line starts to be pinned and S₁ as the area at the instant when the wetting lines moves again. The measured value ΔS = S₂ – S₁ is indicated in the Figure 3-5. The larger values of the peaks and modified peaks design show their better ability at trapping the contact line.

Theoretically, assuming the meniscus keeps a perfect circular shape during its growth and neglecting the influence of the top and bottom walls, the wetting line starts to be pinned when it reaches Location 1 as shown in Figure 6 with θ equals to the receding contact angle of water on PDMS, which is 70º. Figure 6(a) shows that for the pits design, when the wetting line is pinned along the interior side or the v-shaped pit, while
so also equals 70°, the receding contact angle of water on PDMS, theoretical value of even if the meniscus grows to infinity. Therefore, the wetting line is pinned between Location 1 and 2, where here. We will therefore rely on this theoretical approach for relative ability of various designs, such as the three tested geometry to trap a wetting line, it is useful at classifying the does not allow to quantitatively predict the ability of a interface curvature is in 2D. While the theoretical analysis progression of the meniscus, with a value

The fact that the meniscus cannot grow to infinity is explained by the finite dimensions of the chamber, the geometry of the microchip that exhibits tolerances of a few micron, and the interface curvature is in 2D. While the theoretical analysis does not allow to quantitatively predict the ability of a geometry to trap a wetting line, it is useful at classifying the relative ability of various designs, such as the three tested here. We will therefore rely on this theoretical approach for the rest of the interface is very close to the exterior side of the pit. As soon as the interface contacts with the exterior side of the pit, the wetting angle reaches a value near zero, which implies fast receding of the wetting line: the meniscus passes over the pit. This explains why $\Delta S$ for pits is nearly zero in Figure 3. Figure 6(b) shows that for the peaks design, the wetting line is pinned between Location 1 and 2, where $\theta_1$ also equals 70°, the receding contact angle of water on PDMS, so $\Delta S$ for peaks is calculated to be 0.042 mm². In Figure 6(c), we can see that for the modified peaks, $\theta_2$ will never reach 70° even if the meniscus grows to infinity. Therefore, the theoretical value of $\Delta S$ for modified peaks should be infinity. This explains why this design is best at stopping the progression of the meniscus, with a value $\Delta S$ of 0.021 mm². These critical pressures are plotted in Figure 7 as a function of the meniscus radius. Both lower and upper critical pressure decrease with the meniscus radius and their difference is constant, at a value of about 2000 Pa. The behavior in Figure 7 can be explained as follows. Across the interface of the meniscus between the air and the surrounding fluid, the pressure difference is expressed by the Laplace equation [9]:

$$\Delta P = \sigma \left( \frac{1}{R_h} + \frac{1}{R_w} \right)$$  

where $R_w$ and $R_h$ are radiiuses of the meniscus curvature in the respective parallel and perpendicular plane to the microfluidic chip. Experimentally we observe that when the pressure difference $\Delta P$ is modified, $R_h$ changes first while $R_w$ stays constant. The of $R_w$ modification occurs by a variation of the

Pressure measurement across the meniscus

Another way to study the meniscus stability is to measure the pressure amplitude that can be applied across the meniscus while keeping the contact line pinned on the microfeature of Figures 3-5. The pressure is measured using a differential pressure sensor (Honeywell 143PC03D), with one port of the sensor is connected to the output of Syringe 1 and the other port is open to the atmosphere, as shown in Figure 2. The pressure resolution is 0.4 Pa using an Agilent 54622A Oscilloscope to measure the voltage output. The measurement is performed on a chamber with the peaks microgeometry of Figure 4: starting from a meniscus at equilibrium, we either increase or decrease the pressure inside the meniscus by slowly operating the syringe pump. The pressure at the instant where the meniscus starts to grow is called upper critical pressure. The pressure at the instant where the meniscus starts to shrink is called lower critical pressure.

These critical pressures are plotted in Figure 7 as a function of the meniscus radius. Both lower and upper critical pressure decrease with the meniscus radius and their difference is constant, at a value of about 2000 Pa. The behavior in Figure 7 can be explained as follows. Across the interface of the meniscus between the air and the surrounding fluid, the pressure difference is expressed by the Laplace equation [9]:

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wetting angle between its receding value $70^\circ$ and advancing value $110^\circ$. $R_h$ can be expressed as

$$R_h = \frac{h}{2\cos\theta} \quad (2)$$

where $\theta$ is the advancing or receding wetting angle, and the thickness of the chamber $h$ is 50 µm. Once the pressure modification is too large to be compensated by the sole variation of $R_h$, $R_w$ will change and induce a visible motion of the meniscus interface in the visualization plane (see e.g. Figure 4). It is worth noting that there is a position where $R_w$ has a minimum value, which equals half of the width of the microchannel, namely, 50 µm. If the meniscus is further away than that position, once $R_w$ start changing, the motion of the meniscus becomes unstable, either rapidly expanding or rapidly shrinking, in virtue of Equation (1), similarly to the dynamic of an air bubble popping out of an underwater needle. The above theory predicts a range of $\Delta P$ where the visible shape of the meniscus does not change (i.e. $R_w$ is constant), while $R_h$ varies within the bounds of the wetting angle $\theta$ (see Equation (2) and Table 1). Using the surface tension of water at room temperature, this theoretical range of $\Delta P$ is plotted as two solid lines as a function of $R_h$ in Figure 7 and agrees relatively well with our measurements. The uncertainty in Figure 7 is determined by

$$u = (u_0^2 + u_1^2 + u_2^2)^{1/2} = 136 Pa \quad (3)$$

where $u_0$ is the resolution uncertainty (0.2 Pa), $u_1$ is the linearity error of the sensor (129 Pa) and $u_2$ is the repeatability and hysteresis error of the sensor (43 Pa).

**Response to ultrasonic excitation**

Another type of meniscus motion is its response to high-frequency pressure (or volume) oscillations. To induce these oscillations, we cure in the PDMS cover layer a commercially available piezoelectric transducer (PZT, ring-shaped with 2mm thickness). The transducer excites the water in the microchamber and induces various kind of surface waves on the water/air interface, as well as a streaming flow in the liquid close to the meniscus, which can be described as follows.

**Traveling wave**

For a discrete set of excitation frequencies, we observed waves traveling on the meniscus surface, with the same frequency as the excitation. Figure 8 shows a sequence taken at 150 kHz of excitation frequency, with a delay of 2 µs between each frame. Crests A and B are moving towards the center of symmetry of the meniscus. This phenomenon also occurs when the meniscus is retracted and assumes a flat shape in the chip plane, as shown in Figure 9: this configuration simplifies the measurement of the wavelength, as well as the analysis of the phenomenon. Right to the individual frames in Figure 9 are the values of the observed wavelengths at the given oscillation frequencies. We observe that the wavelength decreases as the excitation frequency increases, and this measurement is plotted in Figure 11. Since the wavelengths are small ($\lambda/\langle 2\pi \kappa^{-1}\rangle$, where $\kappa^{-1}$ is the water-air capillary length, with $2\pi \kappa^{-1}$ typically on the order of a centimeter), gravity can be neglected in the theoretical analysis: these waves are very likely capillary waves, caused by inertia and surface tension.

![Figure 8 Traveling waves on meniscus at 150 kHz. FIND EDGE function of Photoshop is used to make the edge clear.](image)

![Figure 9 Traveling waves at different frequencies. FIND EDGE function of Photoshop is used to make the edge clear.](image)

![Figure 10 Wave nomenclature. Where Z0 is the z distance between contact line and equilibrium.](image)
The wavelength-frequency relationship can be described in the classical framework of surface waves analysis [10], neglecting gravity. Figure 10 defines the nomenclature and reference frame with \( x-z \) plane parallel to the microfluidic chip. In a pure 2-D case in the \( x-z \) plane, a dispersion relation exists that relates the wavelength to the wave frequency [10]:

\[
\lambda = \left( \frac{2\pi \sigma}{f^2 \rho} \right)^{1/3}
\]  

(4)  

**Figure 11 Wavelength vs frequency**

This relation is plotted in Figure 11, for a water surface tension at the value of 40°C, corresponding to the measured water temperature increase due to the resistive dissipation in the piezoelectric. Both the theory and experiment show the trend that wavelength decreases as the frequency increases, with the predicted wavelength approximately 5% larger than the observed wavelength. However, in the derivation of Equation (4), the capillary restoring force is only due to the curvature in the plane of the chip (\( x-z \) plane). In our system, however, the capillary restoring force also depends on the curvature in the perpendicular plane (\( x-y \) plane), see Equation (1). In the experiment, the curvature in the perpendicular plane is caused by the wave amplitude and \( z_0 \), which is the distance between the average \( z \)-location of the meniscus and visible location of the wetting line on the top (and bottom) PDMS surface, (see Figure 10). These two effects imply that the visible interface is blurred as in Figure 8. The total contribution of \( z_0 \) (about 2 \( \mu \)m) and wave amplitude (about 2 \( \mu \)m) to the curvature in the perpendicular plane can be expressed as

\[
\frac{1}{R_b} = -\frac{2(\eta - z_0)}{(\eta - z_0)^2 - h^2/4}
\]

while the other curvature, in the plane of the chip is

\[
\frac{1}{R_w} = \frac{-\partial^2 \eta/\partial x^2}{1 + (\partial \eta/\partial x)^2} \approx -\frac{\partial^2 \eta}{\partial x^2}
\]

(5)  

(6)  

By assuming a sine-shape wave with wavelength values as reported in Figure 11, \( R_w \) is on the order of 10 \( \mu \)m, and \( R_b \) is much larger on the order of 80 \( \mu \)m. Therefore, we choose to neglect the 3-D effect due to \( 1/R_b \) in the analysis. Also, the good agreement in Figure 11 between the experimental data and the 2-D theory points to the fact that the flow field in our system is mostly 2-D, which will greatly simplify future developments of similar chips.

**Standing wave**

Increasing the actuation intensity – while keeping the same frequency as in Figure 8 – generates a standing wave oscillating at half the actuation frequency, and observed in Figure 12, using a diode and a frequency divider. The standing wave has larger amplitude (around 5 \( \mu \)m) than the traveling wave (around 2 \( \mu \)m). The experimental conditions at which these standing waves occur are in Table 2, which shows that the standing waves can be either subharmonic, harmonic or superharmonic, in opposition with the traveling waves, which were always observed to be harmonic. An analogous phenomenon known as the Faraday instability happens with gravity waves at the horizontal water surface of a container experiencing up and down oscillations: the fluid layer becomes unstable and exhibits standing waves at half the excitation frequency. The ratios of wave frequencies are given in Table 2, as well as the minimum excitation frequency needed to produce the standing wave. Since they all have integral times of half excitation (\( n/2 \), \( n = 1, 2, 3 \)), all could be observed using a frequency divider.

**Figure 12 Standing wave on meniscus interface at 150 kHz of excitation, the standing wave is oscillating at 75 kHz.**

<table>
<thead>
<tr>
<th>Excitation frequency (kHz)</th>
<th>150</th>
<th>75</th>
<th>50</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wave/excitation ratio</td>
<td>( \frac{1}{2} )</td>
<td>1</td>
<td>3/2</td>
</tr>
<tr>
<td>Smallest voltage needed to induce standing wave (V)</td>
<td>28</td>
<td>184</td>
<td>356</td>
</tr>
</tbody>
</table>

**Table 2 Conditions at the occurrence of standing waves**

**CONCLUSION**

We have described the manufacturing of microfluidic chips for bubble-based actuators. First, we studied the influence of three different geometries on the meniscus contact lines in microfluidic chips and quantitatively identified the efficiency of each geometry to trap the contact line of the gas-liquid meniscus. Theoretically, we found that each meniscus radius corresponds to a range of pressure that maintain the meniscus stable, because of the hysteresis of the wetting angle. This finding is confirmed by pressure measurements. Then, the meniscus response to ultrasonic excitation is studied. When the excitation is weak, traveling waves are found and the wavelength decreases with increasing
excitation frequency, which is well predicted by capillary wave theory. When the excitation becomes stronger, standing waves at the meniscus interface with different ratios (1/2, 1, 3/2) of frequency to excitation frequency are observed.

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REFERENCE: