Sequential droplet manipulation via vibrating ratcheted microchannels

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\textbf{A B S T R A C T}

This work reports on a simple scheme for transport and sequential manipulation of droplets using ratcheted-shaped microchannels. Once subjected to lateral vibration through a sinusoidal shaker with a controllable frequency and amplitude, droplets move along the direction of least resistance with velocities depending on vibration parameters (amplitude and frequency), channel geometry and material properties, and the angle between the channel and vibration direction (lateral offset angle). Using channels having different lateral offset angles, we achieved a controllable time delay between the transports of various droplets. Based on the same principal, we also designed a multi-functional droplet manipulation platform with the capability to transport, collect, mix, and discharge several droplets.

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1. Introduction

The ability to control the dynamics of liquid droplets on solid surfaces is critical in many microfluidic applications that rely on manipulating such droplets for microanalytical processes (i.e., digital microfluidics) \cite{1–3}. The imbalance of surface tension forces across liquid–solid boundaries resulting in the difference in advancing and receding contact angles at the front and rear ends of the droplet is known to play the key role in such manipulations. This surface tension gradient has been generated by chemical treatments \cite{4,5}, thermal methods \cite{6,7}, photochemical methods \cite{4,5}, electrochemical modulation \cite{10}, and electric field (electrowetting on dielectric \cite{11} and dielectrophoresis \cite{12}). These techniques have been explored in detail with many interesting experimental demonstrations. However, there are also limitations associated with each method. These include complexity of the required surface chemistry, large voltages, and the application of heat which can easily evaporate the aqueous droplets. In this paper, we report on a simple method for manipulating droplets overcoming the aforementioned shortcomings. Our technique is based on moving droplets along ratcheted microchannels using mechanical vibration. Through simple geometrical arrangements, we could also accomplish time-multiplexed manipulation (collection, transport, and mixing) of μL-scale droplets using vibrating ratcheted microchannels.

2. Design and fabrication

Before describing our approach and design procedure, it is useful to review a few basic facts regarding the phenomena of capillarity and wetting on textured surfaces \cite{13,14}. For planar non-textured surfaces a macroscopic finite equilibrium contact angle $\theta$ between the droplet and different surfaces can be defined (Fig. 1a) as

$$\cos \theta = \frac{\gamma_{SG} - \gamma_{SL}}{\gamma_{LC}}$$  \hfill (1)

where $\gamma_{SG}$, $\gamma_{SL}$, and $\gamma_{LC}$ are the solid–gas, solid–liquid, and liquid–gas interfacial energies. Wetting phenomena on textured structures is however more complex. Fig. 1b shows the side views of several droplet morphologies in a groove with rectangular cross section assuming that the contact angle of liquid on a planner surface made of the same material is greater than 45° (after Ref. \cite{14}). If only a small amount of liquid is deposited, it prefers to adhere to the groove corners since this allows contact with both the bottom and the sidewall of the groove minimizing the total energy (case (i) in Fig. 1b). Increasing the liquid volume further moves the upper contact line segments towards the edge of the groove with subsequent pinning at the edge Fig. 1b(ii). Further increase in the droplet volume results in the growth and merger of the two cornered droplets and their subsequent overspilling (Fig. 1b(iii)) and extended filament formation (Fig. 1b(iv)). Since the channels described in this paper are in the millimeter scale range width and the droplets are
several $\mu$L in volume, we only consider the cases of overspilling droplets in static and dynamic situations.

2.1. Single channel and functional platform design

Fig. 2 shows the design parameters for a ratchet-shaped fluidic channel as defined by the channel width at its narrowest part $\alpha$, the ratchet depth $\beta$, and the ratchet periodic length $\gamma$. With such an asymmetric design, a droplet located in such a channel and subjected to horizontal vibration will move in the direction of least resistance (to the right in this case). The movement velocity depends on several factors including: (1) ratchet angle ($\theta = \tan^{-1}(\gamma/\beta)$), (2) ratchet density (periodic length), (3) materials used to fabricate the channel, (4) channel depth ($d$), (5) vibration frequency and amplitude, and (6) the angle between the channel and vibration direction (lateral offset angle, LOA). Among these some such as ratchet angle has a much lesser effect on the velocity of the droplets while others including vibration frequency, amplitude, and LOA have a more pronounced influence. Channel material and depth also affect the droplet movement with deeper channels providing more sidewall contact area and hence larger driving force on the droplets. Channel material influences the contact and sliding angles (defined as the critical angle where a water droplet with a certain weight begins to slide down an inclined surface), so it is desirable to choose a material with higher contact angle and lower sliding angle for easier droplet manipulation.

We designed two sets of ratcheted fluidic channels, one for sequential droplet manipulation based on different LOAs and the other for multi-functional performance (i.e., transport, mixing, and disposal). Fig. 3a shows the design of the time-multiplexed droplet manipulation platform with five branches each having a different LOA (ranging from $0^\circ$ to $40^\circ$). Each channel has a total length of 32 mm and ratchet parameters of $\alpha = 1$ mm, $\beta = 0.5$ mm, and $\gamma = 2$ mm. The second design is shown in Fig. 3b with four branches having a fixed LOA of $15^\circ$ and different ratchet geometrical parameters ($\alpha$, $\beta$, and $\gamma$) on each branch intended to create a time delay between the motions of droplets. Droplets will move towards the center in three branches, mixed in the central reservoir, and then guided to the fourth branch for disposal.

2.2. Fabrication process

As mentioned above, the channel surface has to be hydrophobic (nonwettable) in order to reduce the resistive force against which the droplet has to move. One can either use a naturally hydrophobic surface or a hydrophobized naturally hydrophilic surface. In this research, we fabricated channels made of the following materials: (1) SU8 on glass slide (SU8-glass channel), (2) silicon, (3) polydimethylsiloxane (PDMS), and (4) UV-curable epoxy. SU8-glass channels were fabricated using lithography on glass slides while silicon channels were fabricated by deep reactive ion etching (DRIE). In order to improve hydrophobicity, both of these channels were silanized with a 5 vol% solution of dichlorodimethylsilane (DCMS) (Sigma–Aldrich) in toluene [15]. PDMS channels were casted from a silicon mold while the UV-curable epoxy channels were casted from a negative PDMS mold. The UV-curable epoxy used in these experiments was a monomer mixture of isobornyl acrylate (IBA)
(Sigma–Aldrich), 9.7 wt% of tetraethylene glycol dimethacrylate (TeEGDMA) (Sigma–Aldrich), and 3.0 wt% of Irgacure 651 (Ciba Specialty Chemicals Corp.) as photoinitiator [16]. The UV curing source was a mercury lamp with 300 mJ/cm² energy density and 1 min exposure time. After exposure, the liquid prepolymer mixture turns into a light yellow rigid polymer with hydrophobic surfaces.

3. Experimental methods and results

3.1. Experiment setup

In this study, droplet movement was actuated by a sinusoidal vibrating shaker table. Fig. 4 shows the schematic view of the experimental setup which consisted of a function generator, a vibrating shaker table with an aluminum platform for supporting the fabricated substrate/platform with fluidic channels, and an accelerometer. The accelerometer was used to extract the vibration parameters (frequency and amplitude). A high speed digital camera was used to capture the droplet motion.

![Schematic of experiment setup showing the vibration table, various monitoring sensors, and droplet manipulation platform.](image)

Fig. 4. Schematic of experiment setup showing the vibration table, various monitoring sensors, and droplet manipulation platform.

As mentioned previously, the ratchet design introduces an asymmetrical resistance (equivalent to a pressure gradient), hence, creating a net force to drive the droplets. The droplet velocity is directly proportional to the vibration amplitude and frequency. Due to the experimental limitations associated with our shaker table, we could not test the system under frequencies higher than 200 Hz and vibration amplitudes of larger than 6.3 mm. Fig. 5 shows

![Test results showing the effect of droplet size on its movement for a fixed channel geometry (α = 1 mm, β = 0.5 mm and γ = 2 mm, channel depth = 200 μm). Tests were performed using 50 Hz and 3.5 mm actuation parameters. (Scale bar: 10 mm.)](image)

Fig. 5. Snapshots of a droplet (8 μL) moving along a SU8-glass channel (a) 0.5 s; (b) 1.2 s; (c) 3.5 s. (Scale bar 5 mm.)
Table 1

<table>
<thead>
<tr>
<th>Droplet speed Properties of residues Remarks</th>
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<tbody>
<tr>
<td>SU8-glass channel 60 mm/s Residue droplets left almost on each ratchet corner Hard to fabricate, SU8 can peel off the substrate</td>
</tr>
<tr>
<td>Epoxy channel 43 mm/s Some ratchets have residues some others do not Cheap, easy to fabricate, slow speed</td>
</tr>
<tr>
<td>Silicon channel 100 mm/s Residues left on each ratchet corner Last long, the highest speed, but has more residues</td>
</tr>
<tr>
<td>PDMS channel 38 mm/s Only few of ratchets have residues at the corners Cheap, easy to fabricate, slow speed</td>
</tr>
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All of the above tests were performed under the condition of 50Hz frequency and 3.5 mm vibration amplitude.

Table 2

<table>
<thead>
<tr>
<th>Contact angle (CA)</th>
<th>Sliding angle</th>
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<tbody>
<tr>
<td>Contact angle (CA)</td>
<td>Sliding angle</td>
</tr>
<tr>
<td>25 µL droplet</td>
<td>50 µL droplet</td>
</tr>
<tr>
<td>Silanized silicon  99</td>
<td>24.4°</td>
</tr>
<tr>
<td>SU8                79</td>
<td>50.7°</td>
</tr>
<tr>
<td>Silanized glass    71</td>
<td>60.6°</td>
</tr>
<tr>
<td>UV-curable epoxy   76</td>
<td>73.7°</td>
</tr>
<tr>
<td>PDMS               113</td>
<td>No sliding (&gt;90°)</td>
</tr>
</tbody>
</table>

* No sliding happens even when the tilt angle is larger than 90°.

We also investigated the effect of droplet size in a SU8-glass channel with a fixed geometry (\(\alpha = 1\) mm, \(\beta = 0.5\) mm and \(\gamma = 2\) mm, channel height = 200 µm). Fig. 7 shows the results for droplet volumes of 2–48 µL under similar actuation conditions of 50Hz and 3.5 mm. The smallest droplet size that can be successfully transported across the length of the channel is limited by the ratchet periodic length (\(\gamma\)). As can be seen, a 2 µL droplet covers one periodic length and when actuated it breaks up into smaller satellite droplets. A 4 µL droplet meanwhile covers approximately two ratchet lengths and is successfully transported across the channel.

The maximum droplet size is limited by channel lateral dimensions and beyond a certain limit the droplet will spread over the channel.

As mentioned above, channel depth is an important factor in moving velocity since it is related to the effective contact area which determines the net driving force on the droplet. In order to study the effect of channel depth, we focused on SU8-glass channels due to their easier fabrication. Fig. 6 shows the results of channel depth vs. droplet velocity for SU8-glass channels (\(\alpha = 1\) mm, \(\beta = 0.5\) mm, \(\gamma = 2\) mm). In the experiment, the vibration amplitude and frequency were fixed at 3.5 mm and 50Hz, respectively. As shown in the figure, the droplet velocity increased with increasing channel depth approximately in a linear fashion. However, due to the lack of enough driving force, the droplets did not move when the depth was below 100 µm.

![Fig. 8. Droplet velocity vs. lateral offset angle (LOA).](image-url)

![Fig. 9. Test results showing the effect of increasing LOA on droplet (8 µL) movement (\(\alpha = 1\) mm, \(\beta = 0.5\) mm, and \(\gamma = 2\) mm, channel depth = 200 µm). Numbers on the right of each figure indicate the vibration amplitude when the droplet started moving and the amplitude at which the droplet split or the shaker table amplitude limit was approached (6.3 mm).](image-url)
edges and overfill the reservoirs (see for example the 48 µL case). It is also worth noticing that the droplet velocity depends on its size. Although the larger droplets have more contact area with the ratchet side wall, the inertia of the droplet takes over the driving force, making the larger droplets to move slower than the smaller ones.

The effect of materials used to fabricate the fluidic channels on the droplet movement was also studied. Keeping the droplet volume and channel geometrical parameters constant, we evaluated different materials described in the fabrication section. These materials have different contact and sliding angles which correlates to the mobility of the droplets. In addition, they influence the droplet residues if sharp corners are used. Table 1 summarizes the experimental results. As can be seen, under the same testing conditions (50 Hz and 3.5 mm in amplitude) silicon channels had the highest droplet velocity (100 mm/s) but left the most unwanted residues. Channels fabricated from polymers (PDMS, UV-curable epoxy) had lower droplet velocities and left fewer residues. These results can be understood in light of different sliding angles as summarized in Table 2 along with contact angles (sliding angle measurements were performed by slowly increasing the surface incline angle and measure the angle at which the droplet begins to move. For comparison, we chose 25 and 50 µL droplets since the sliding angle is dependant on the size of the droplets). As to be seen, there is an inverse relationship between the droplet velocity and sliding angle, i.e., PDMS having the highest sliding angle has the smallest velocity whereas silanized silicon with the smallest sliding angle has the highest velocity. As can also be seen from Tables 1 and 2, the corner residues is also related to the velocity and sliding angle (with silicon having the most residues and PDMS having the least). These correlations are understandable since a small sliding angle allows a lower resistance to the movement of droplets hence a greater velocity and larger corner residues.

3.3. Functional platforms

As mentioned previously, the LOA has a pronounced effect of the droplet velocity. Fig. 8 shows droplet velocity vs. lateral offset angle. When the lateral offset angle is introduced, the vibration amplitude on the channel can be decomposed to a parallel component (the part in the channel direction) and a perpendicular component (the part perpendicular to the channel direction). The perpendicular vibration component has no effect on the movement while the parallel component is the effective vibration amplitude which
It was noticed that for LOAs more than 50°, the perpendicular component can become exceedingly large forcing the droplets out of the channel. Fig. 9 shows the test results for LOAs of 0–60° using a fixed droplet volume of 8 μL (actuation frequency of 50 Hz and amplitude of 2–6.3 mm). For LOAs of 0–40°, the droplet did not escape the channel even when vibrated at maximum amplitude of 6.3 mm. When the LOA was 50°, even though one could move the droplet across the channel at 5 mm actuation amplitude, the droplet was split during the movement and the satellite droplets were forced out of the channel (circled by white dot line). However, when LOA was increased to 60°, the droplet did not move even using the maximum vibration amplitude. Instead, it was split and splashed when the vibration amplitude was too high.

Fig. 10 shows snapshots of a time-multiplexed droplet manipulation experiment with five droplets (volume 8 μL) moving along SU8-glass channels having different LOAs (ranging from 0° to 40°). Each channel was 32 mm in length and had similar ratchet parameters of α = 1 mm, β = 0.5 mm, γ = 2 mm. Five colored droplets started from the outer edges of each channel and gathered/mixed in the central reservoir one after another. As expected, the velocity in each channel reduces linearly with increasing LOA. The total droplet manipulation process was ∼6 s with 1 s separation between different droplets arriving at the center. Although this time is adequate for applications such as investigating the kinetics of chemical reactions between various droplets; the time separation can be increased or decreased by changing the design of the channel (e.g., channel length).

Another example demonstrating the multi-functionality of such platforms is shown in Fig. 11. In this case, four channels are designed such that they can transport, collect, mix, and dispose three different droplets (all channels have the same LOA). Arrows are added in order to make the ratchet direction more clear. Since the ratchet geometries did not affect the droplet velocity appreciably, the time lag effect could not be observed in these snapshots. The channels were tested by fixing the frequency (50 Hz) and gradually increasing the shaking amplitude. As can be seen, each branch transports its droplet to the center, mix it there, and then deliver the mixture to the fourth outlet channel for disposal.

A demonstration indicating a more practical aspect of the platform was performed using luminol (3-aminophthalhydrazide, Sigma–Aldrich) chemiluminescence reaction, Fig. 12. The solution in branch 1 was the luminol stock solution made from 160 mg luminol and 1.2 g potassium hydroxide (KOH) in 20 ml DI water. Branch 2 contained 3% hydrogen peroxide while a droplet of blood was driven towards the center. The reaction was observed to take place at the center, producing a bright luminescence that was recorded by a camera.
placed in branch 3 as a catalyst. Droplets in channels were intentionally dyed for higher contrast. The middle subfigure shows the case when droplets in branches 1 and 2 were mixed at the central reservoir, while the blood droplet in branch 3 was still on its way. In this case, no luminescence was observed. After the blood droplet in branch 3 reached the central reservoir, chemiluminescence was initiated due to the catalytic action of iron in the hemoglobin. The right picture in Fig. 12 shows the emission of luminescence which could be seen when the room light was turned off.

3.4. Residue elimination

A closer look at Fig. 5 reveals small residues left behind at ratchet concave corners once the droplet passes through the channel. These residues can be a drawback since they cause cross-contamination if several different samples have to be manipulated sequentially over a single channel. Another drawback of the residues could be the droplet volume change leading to inaccuracy in sensitive chemical analysis. Fig. 13a (left panel) shows a close-up view of the residues on the channel ($\alpha = 1$ mm, $\beta = 0.5$ mm, $\gamma = 2$ mm). Although we performed all of our aforementioned experiments with channels having sharp corners, one can eliminate the residues by designing a channel with streamline rounded corners. Fig. 13b shows such a channel and a corresponding close-up view (right panel) showing practically no residues at the streamlined corners.

4. Conclusions

In this paper, we demonstrated and characterized a simple method for the movement of droplets confined in hydrophobic asymmetrically ratched microchannels. Upon the application of lateral vibration, the droplets moved at velocities that were dependent on vibration parameters, channel geometry and its construction material, and the channel lateral offset angle. A maximum droplet velocity of 100 mm/s was achieved with a vibration frequency and amplitude of 50 Hz and 3.5 mm, respectively. Two separate platforms were designed to demonstrate various microfluidic operations such as sample transport, mixing, and disposal.

References


Biographies

Zhenwen Ding received the B.S. degree in Physics from University of Science and Technology of China (USTC), Hefei, China, in 2003, and the M.S. degree in Physics from Purdue University, West Lafayette, IN, in 2005. He is currently working in Birck Nanotechnology Center toward the Ph.D. degree in Physics at Purdue University, West Lafayette, IN. He has authored and co-authored more than fifteen peer-reviewed journal and conference publications. His research interests include biomedical applications of MEMS and Microsystems, specifically, active flow control and droplet manipulation, biomimetic sensors and actuators, and the applications of smart polymers (environmental-sensitive hydrogel) for ultra-sensitive biosensing.

Woo-Bin Song received his B.S. and M.S. degrees in Physics from Konkuk University, Seoul, Korea where he received the distinguished Dean Award of the Graduate School. He received his Ph.D. degree in Electrical Engineering at the University of Minnesota, Twin Cities, MN, in 2004 where he received a Student Travel Scholarship to the Hilton Head Workshop. After completion of his doctoral study, he joined the School of Electrical and Computer Engineering at Purdue University in West Lafayette, IN, and the School of Dentistry at the University of Minnesota, where he served as a Post-Doctoral Research Associate to extend his research in BioMEMS and Bio-Nanotechnology. He is currently working as senior engineer at Semiconductor R&D Center, Samsung Electronics Co., Ltd., where he is developing the process of selective epistral growth to boost the performance of transistors for DRAM and Logic devices. He has authored and co-authored more than ten peer-reviewed journal and conference publications. Further, he is a reviewer for the IEEE Sensors Journal, the IEEE Transactions on Electron Devices, and Sensors and Actuators A, and a member of IEEE since 2002. His current research interests lie in the development of new active epistral growth technology and new device structures for thermal imaging, simple platforms for manipulation of micro-droplets, micro-/nano-devices using smart materials, and implantable devices using biodegradable materials.

Babak Ziaie received his doctoral degree in Electrical Engineering from the University of Michigan in 1994. His dissertation was related to the design and development of an implantable single channel microstimulator for functional neuromuscular stimulation. From 1995 to 1999 he was a post-doctoral fellow and an assistant research scientist at the Center for Integrated Microsystems (CIMS) of the University of Michigan. He subsequently joined the Electrical and Computer Engineering Department of the University of Minnesota as an assistant professor (1999–2004). Since January 2005, he has been with the School of Electrical and Computer Engineering at Purdue University where he is currently a professor. His research interests are related to the biomedical applications of MEMS and Microsystems (BioMEMS). These include implantable wireless microsystems, smart polymers for physiological sensing and active flow control, micromachined interfaces with the central nervous system, biomimetic sensors and actuators, and ultra-sensitive sensors for biological (molecular and cellular) applications. Dr. Ziaie is the recipient of the NSF Career award in Biomedical Engineering (2001) and McKnight Endowment Fund Award for Technological Innovations in Neuroscience (2002). Dr. Ziaie is a member of the IEEE and the American Association for the Advancement of Science.