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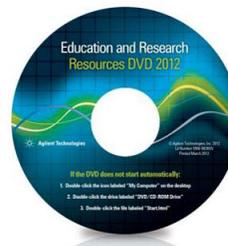


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Transport of a soft cargo on a nanoscale ratchet

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Surface ratchets can guide droplet transport for microfluidic systems. Here, we demonstrated the actuation of microgels encapsulated in droplets using a unidirectional nanotextured surface, which moves droplets with low vibration amplitudes by a ratcheting mechanism. The nanofilm carries droplets along the ratchets with minimal drop shape deformation to move the encapsulated soft cargo, i.e., microscale hydrogels. The tilted nanorods of the nanofilm produce unidirectional wetting, thereby enabling droplet motion in a single direction. Maximum droplet translation speed on the nanofilm was determined to be 3.5 mm/s, which offers a pathway towards high throughput microgel assembly applications to build complex constructs. © 2011 American Institute of Physics. [doi:10.1063/1.3625430]

Digital microfluidics¹ uses droplets to manipulate controlled volumes and chemical compositions. Various mechanisms for actuation of droplets have been proposed, such as capillary forces,² focused acoustics waves,³ surface tension,⁴ transistor based actuation,⁵ electrowetting,⁶ and chemical^{5,7} or temperature based Leidenfrost ratchet.⁸ Among these techniques, the concept of vibration on asymmetrically textured surfaces^{9,10} (i.e., surface ratchets) provides a unique opportunity to transport small solid or liquid materials without a directed force and eliminates the need for complex fabrication procedures and controls for droplet manipulation.

Microscale engineering requires methods to assemble small size objects to create larger and more complex constructs.¹¹ Therefore, there is a need for technologies to manipulate and assemble the building blocks, e.g., microgels for tissue engineering applications.¹¹ Recently, Demirel *et al.* have developed an engineered hydrophobic surface deriving its unidirectional wetting directly from its asymmetric nanoscale roughness.¹⁰ The nanofilm consists of a well-ordered array of polymeric nanorods fabricated via vapor-phase oblique angle polymerization.¹² In addition, Xu *et al.* presented assembly of microgels within microdroplets to build complex constructs using physical fields including acoustics and magnetic nanoparticles.¹¹ However, a directional mechanism to transport microgels to a target location has not been demonstrated. Technologies, such as surface ratchets, could be effective to manipulate and assemble microgels.

Here, we showed the manipulation of microgels on a unidirectional surface by a ratchet mechanism, which can move soft materials encapsulated in droplets. Specifically, we quantitatively studied transport of a microgel inside a droplet via vibrations on a polymeric textured nanofilm. By

inducing a vibrational field on the surface, droplets can be propelled on the unidirectional surface, which points in the direction of the asperities. Here, time-lapse frames of droplet motion with and without microgels were quantified as a function of vibration frequency.

Droplet transport on a surface requires a textured surface to move droplets with low vibration amplitudes by a ratcheting mechanism. Correspondingly, the nanofilm provides a hydrophobic directional surface due to its nanoscale texture.¹⁰ The smooth nanofilm surface, fabricated by oblique angle vapor deposition of poly(p-chloro-xylylene) (PPX), carries microliter drops along the half-pipes with minimal droplet shape deformations.¹⁰ The tilted fibers on our PPX nanofilm produce unidirectional wetting, thereby enabling droplet motion in a single direction. Though adhesive, our unidirectional hydrophobic nanofilm can support large drops in Cassie states, i.e., suspended above the nanorods.¹⁰

Figure 1(a) shows the frequency required to move the water droplets (Ultra pure, 18.2 MΩ cm) as a function of drop size. The water droplets were directionally transported along the half-pipes, which were attached horizontally to a mechanical vibrator (PASCO SF 9324). The oscillations of the mechanical vibrator were kept constant at low amplitude (0.585 mm) for all experiments. Experimental data were recorded with a digital camcorder (Sony DCRTRV50), and the velocities were estimated from the movies using ImageJ and Windows Movie Maker. These results showed that droplet volume has a specific frequency interval in which the droplet could be transported.¹³ Additionally, we demonstrated that the droplet volume is inversely proportional to the frequency. The scaling $\omega \sim 1/\sqrt{V}$ is due to Rayleigh¹⁴ for spherical volumes, V , which is given by $\omega = (3\pi\rho V/(8\gamma))^{1/2}$, where ρ is the density and γ is the surface tension of water. Figure 1(b) shows the velocities of these droplets as a function of normalized vibrations, ω/ω^* , where $\omega^* = (\gamma/m)^{1/2}$ for water and m is the mass of drop which varies between 2 to 12 μg . The data collected at

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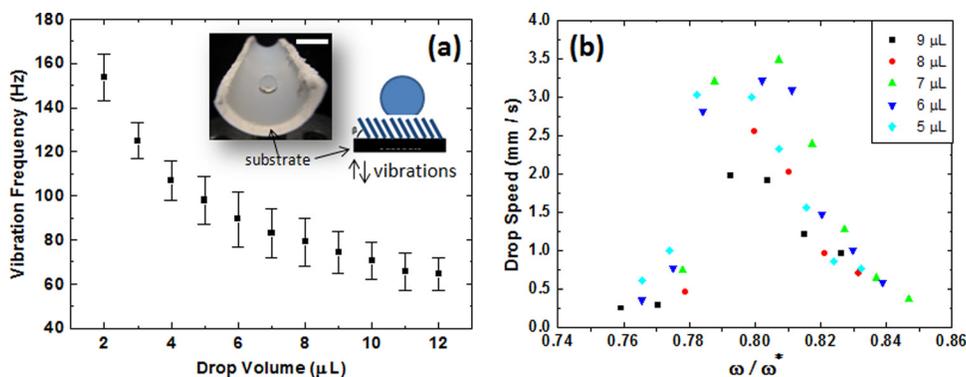


FIG. 1. (Color online) (a) Vibration frequency as a function of water droplet size. Inset shows the optical image of the directional nanofilm coated half-pipe and a schematic of vibration for droplet transport (b) Droplet transport speed at various normalized vibration frequencies shows clustering of the data around 0.8. Maximum droplet transport speed of 3.5 mm/s was observed (scale bar: 5 mm).

different frequencies and drop volume cluster around the same peak, which indicate that the velocity peaks are due to resonance effects arising from the matching of the vibrational and natural frequencies.¹⁵

The microgel preparation is shown schematically in Figure 2(a), and an optical image of the microgels on a glass substrate is shown as an inset.¹¹ Briefly, microgels were prepared using a pre-polymer solution by dissolving 20% poly(ethylene glycol) methacrylate (PEGMA, Sigma) in Dulbecco's phosphate buffer saline (DPBS). A photo-initiator, 2-hydroxy-4'-(2-hydroxy-ethoxy)-2-methyl-propiophenone (98%, Sigma), was added to the pre-polymer solution right before the UV exposure. The mixture was placed between two glass slides with a 600 μm spacer. UV light (UVP B-100A, 365 nm) was exposed from a 10 cm distance, through a photo-mask placed on the top glass slide, for 3 min to cure the pre-polymer solution into 500 μm^2 microgels. Microgels were washed off with distilled water on a glass slide and collected in a glass container. The Fourier transform infrared spectroscopy (FTIR) spectra in Figure 2(a) show the bands of 1724 cm^{-1} , 1090 cm^{-1} , 3446 cm^{-1} (wide area), and 2869 cm^{-1} , indicating the C=O and C-O stretching of PEGMA's carbonyl group, O-H stretching in water, CH₂ stretching and bending of PEGMA, respectively. Contact angle of droplet on the nanofilm without and with a microgel in the water droplet were measured as $117^\circ \pm 3^\circ$ and $88^\circ \pm 3^\circ$ (FTA 1000B, First Ten Angstroms). The change of surface wetting properties from hydrophobic to hydrophilic promotes water penetration into the interstitial regions between nanorods, leading to droplet impalement that renders the surfaces impractical for droplet transport. Therefore, a thin layer ($\sim 5 \mu\text{m}$) of planar poly(p-chloro-xylylene)

was deposited on the microgels using a vacuum deposition¹⁶ (SCS, PDS 2010) to prevent the hydrophilic transformation of the surface due to the presence of an encapsulated microgel in the droplet.

We analyzed the vibration frequency dependence as a function of droplet volume in Figure 2(b). An example of individual microgel transport in a water droplet on the directional nanofilms is shown in the inset of Figure 2(b). After placing a 5 μl water droplet on the half-pipe, a 600 μm^3 microgel was placed in the droplet by a pipette. Vibration of the surface at 94 Hz allowed the microgel to move within the 5 μl droplet along the ratchet. We also compared the frequency dependence of water droplets without an encapsulated microgel in Figure 2(b). The average frequency required to move the droplets with a microgel is similar in magnitude to the droplets without a microgel.

Transport of microgels by a sequential process (Figure 3) shows the ability to control microgel transport on the nano-coated surface via ratcheting. First, we demonstrated the sequential drop transport in Figure 3(a) by placing three 2 μl water droplets along the nanotextured half-pipe. A larger water drop (5 μl) was placed at one end of the half-pipe which moves when the vibration is set at 95 Hz. As noted earlier, the vibration frequency was sequentially decreased (i.e., 95-65 Hz) to move larger droplets (i.e., 5-11 μl) on the directional nanofilm. We repeated the same experiment by adding microgels into the droplets (Figure 3(b)).

In summary, we demonstrated that droplets with and without microgels move unidirectionally as a function of vibration frequency on a nanocoated surface. Directional transport of droplets on nanotextured surfaces is a promising

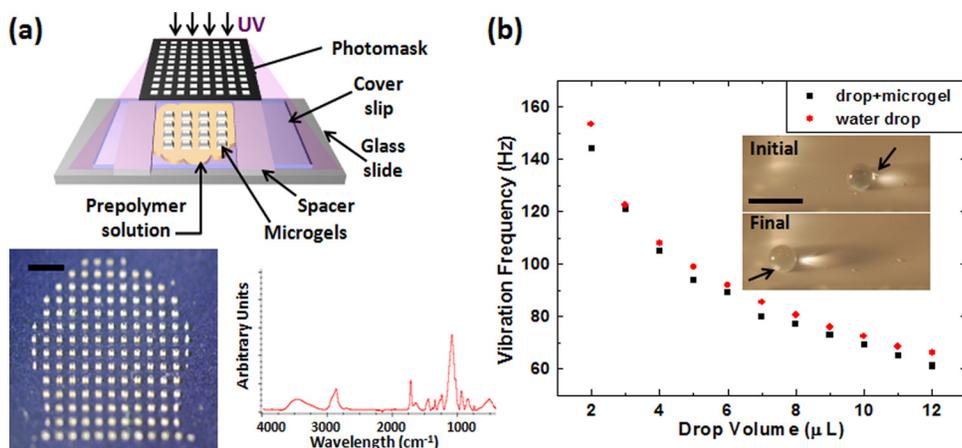


FIG. 2. (Color online) (a) Schematic of microgel synthesis, FTIR of the microgel, and optical image of fabricated microgels are shown (scale bar in the inset is 1 mm). (b) Vibration frequency of as a function of water droplet size with and without a microgel encapsulated within the transported droplet. The inset shows a microgel encapsulated by 5 μl water drop, vibrated at 94 Hz on directional nanofilm (scale bar: 5 mm).

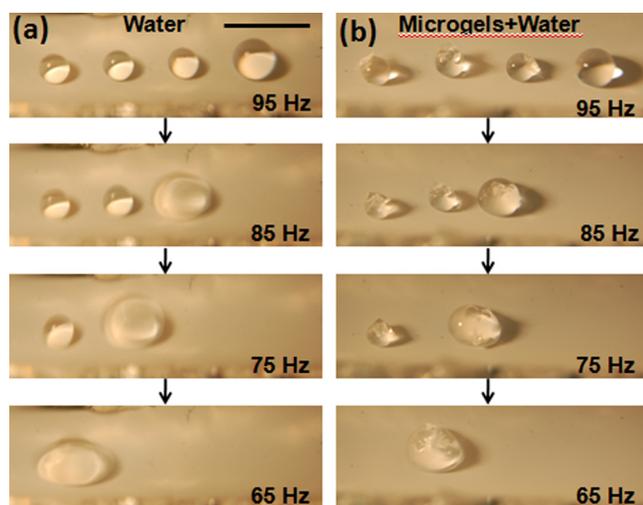


FIG. 3. (Color online) Optical images of the water droplets with and without a microgel on nanofilm coated half-pipes. A $5\ \mu\text{L}$ droplet moves from right to left collecting three $2\ \mu\text{L}$ droplets without (a) and with (b) microgels. As the drop volume gets larger the frequency of vibration is lowered to transport droplets. Three microgels were sequentially transported and merged into a single droplet on the nanofilm. (scale bar: 5 mm).

technology for moving soft cargo such as microgels. Future work will focus on two directions: (i) developing a surface chemistry that will be versatile for transport of droplets other than water and (ii) encapsulation of cells in microgels to achieve assembly of complex shapes. The research in the assembly of cell encapsulated microgels can potentially overcome the limitations to obtain control over cell-cell proximity with microscale resolution.

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and M.C.D. performed the experiments. All authors contributed to writing and revising the manuscript and agreed on its final content. K.S. and U.G. contributed equally to this work.

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