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Dynamic Manipulation by Light and Electric Fields: Micrometer Particles to Microliter Droplets

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We demonstrate a new hybrid optoelectric technique that can manipulate objects across several length scales. The technique leverages a variety of different physical mechanisms to achieve the dynamic manipulation of droplets and also the in situ concentration of colloidal particles suspended in the droplets. Various physical mechanisms such as optoelectrowetting, electrothermal flows, and ac electroosmosis are leveraged through different modes of operation of the device. Each operational mode, which is activated through the proper combination of an applied ac bias and the illumination used, is characterized by the ability to manipulate objects on a certain length scale. We also demonstrate that the device lends itself to the active control of microstructure patterns that emerge from a droplet evaporation process.

Introduction

Digital microfluidics, with its “digitization” of liquid streams into discrete droplets, facilitates a hierarchical architecture for lab-on-a-chip devices, thus allowing diverse biomedical applications to be parsed into easily realizable elemental fluidic building blocks.1 Such a hierarchical approach allows for highly efficient, compact lab-on-a-chip devices with a low response time and point-of-care diagnostics, which promise to be the face of future medical diagnostics.2–4 In digital microfluidics, droplets containing various reagents in particulate or liquid phases can be dispensed from reservoirs, and these droplets can be independently and even simultaneously addressed using electric fields. The in situ concentration, sorting, or manipulation of particles, cells, viruses, or molecules inside droplets is an operation that is often necessitated by sample reaction and detection processes in lab-on-a-chip devices.5 In addition, the redistribution of organic solutes or particulate phases inside droplets during a droplet evaporation process6,7 is a microscale and/or nanoscale assembly process (ex situ process) with relevance to various technologies ranging from biosensors8,9 to the synthesis of functionalized biomaterial surfaces.10 Control over the microstructures and/or nanostructures that emerge during a droplet evaporation process is necessary and is usually pursued through passive techniques such as control of the evaporation rate by altering the humidity or temperature11–13 and also through the addition of surfactants,14,15

Digital microfluidic platforms employ the principles of electrowetting on a dielectric (EWOD),1.6–9 where reaction vessels (droplets) are manipulated by the use of electric fields. Typically, volumes for droplets range from 10−9 to 10−6 L, and thus electrodes, which can produce localized forces on length scales (Lc) of ~10−3 m, are required for digital microfluidic systems. In stark contrast, the manipulation of particulate phases inside droplets typically requires localized forces at Lc ≈ 10−9−10−6 m. The cross-scale manipulation required for the simultaneous control of the droplet movement and manipulation of particulate phases inside the droplet can be especially challenging, but the idea has recently ignited efforts to create cross-scale manipulation


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platforms and has resulted in limited progress. Although there exists a crucial need for further progress toward the goal of an integrated platform, in existing platforms in situ particulate manipulation can be spatially constrained by permanent electrodes or by sandwiched structures, where the extensibility is seriously limited. Cross-scale techniques are yet to demonstrate control of an ex situ process, such as control of particulate patterns that emerge during droplet evaporation.

We demonstrate a hybrid optoelectric technique that can achieve the multiscale manipulation of droplets and particulate phases inside the droplets. The technique can not only optically address droplets in a nonsandwiched configuration but also dynamically concentrate particles inside the droplet (in situ concentration). Droplet manipulation and particulate manipulation take advantage of separate physical phenomena on different length scales—optoelectrowetting (OEW) for droplet manipulation and electrokinetic flow for particulate manipulation. Furthermore, as an application of this multiscale platform, we demonstrate the novel “active” control of a particle-patterning process with microsphere solutions (ex situ control).

Materials and Methods

Figure 1 illustrates the structure of the chip. Constructed over a transparent glass substrate, the first layer consists of transparent indium titanium (ITO) electrodes, followed by a photoconductive layer (amorphous silicon, a-Si) of 450 nm. On the top of the photoconductive layer, there is an insulating layer (silicon dioxide, SiO2) of approximately 115 nm and finally the topmost layer consists of a hydrophobic coating (Teflon AF400S2-100-1, DuPont) that is less than 50 nm. The interdigitated ITO electrodes were fabricated through a wet etching process and have a height of 100 nm and an average width of 750 μm with a 50 μm separation between electrodes. The subsequent layers consisting of a layer of a-Si and a layer of SiO2 were deposited through a plasma-enhanced chemical vapor deposition process (PECVD). A thin layer of a hydrophobic Teflon coating is achieved through a spin-coating process. The choice of materials for the electrode layer and the photoconductive layer are important for the operation of the hybrid device. ITO is transparent, allowing for optical access from beneath. Moreover, ITO absorbs strongly in the infrared (IR) spectrum, thus an ITO film can be locally heated by shining a focused IR laser beam. This property will be constructively utilized in creating electrothermal flows for the particulate concentration. However, amorphous silicon, a-Si, is a strong photoconductor in the visible spectrum. This property will facilitate the creation of virtual electrodes with most common illumination sources.

The interdigitated electrodes are biased with an ac signal. Typically, the biasing voltage, V, is between 25 and 50 Vrms (root-mean-square volts) for particle concentration and approximately 50 Vrms for droplet manipulation, and the ac frequency, f, is determined by the mode of operation of the device. When an ac signal is biased across the coplanar interdigitated electrodes, subcircuit loops are formed as droplets roll over the electrodes. This eliminates the need to sandwich the droplet, and such an open architecture was adopted because of its ready adaptability to complex operations such as human interfacing for pipetting operations. Active control of the droplet-patterning process was also facilitated by the open architecture.

The hybrid device utilizes a combination of different illumination schemes to achieve its cross-scale functionality. The light illumination for droplet actuation can come from a variety of sources. A He–Ne laser (632.8 nm) beam, with a total power of 20 mW, was used to direct the droplet movement. Alternatively, movement can also be directed by loosely focusing broadband illumination from a 120 W lamp (X-cite 120, Exfo, Quebec, Canada). The light spot size in either case is of the same order of magnitude as the droplet. A separate, tightly focused infrared (IR) laser beam with a wavelength of 1064 nm from a Nd:YAG laser served as the source of optical illumination for particle aggregation. A 40× (0.8 numerical aperture, 2 mm working distance) Nikon microscope objective lens was used to focus either broadband illumination or the IR laser. A power of 30 mW for the infrared laser was used for particle aggregation. This stated laser power value refers to the total laser power impinging the back focal plane of the focusing lens.

To illustrate the operation of the hybrid device, microliter-sized aqueous droplets (~20 μL) carrying a dilute suspension of the 1.0 μm carboxylate-coated polystyrene microspheres were prepared. The 1.0 μm carboxylate-modified polystyrene microspheres (Invitrogen, MD) used in the present investigation were fluorescent red. These microspheres of nearly unity specific gravity can be considered to be neutrally buoyant in the immersion medium.

Results and Discussions

The hybrid open chip (Figure 2A) has distinct modes of operation, and whereas in one mode it can manipulate droplets, in another it can concentrate particulate phases inside droplets. Each mode is characterized by the ability to localize forces on a particular length scale and is activated by the proper combination of the applied ac bias and illumination used. To demonstrate manipulation at lE ~10−3 m, droplets were dispensed on the substrate surface while being suspended in an oil medium. Under the application of a low-frequency ac bias (~10 Hz), these aqueous droplets could be manipulated dynamically on the coplanar electrodes. The movement of the droplets was determined by the light-induced virtual electrodes. The droplets could
be actuated by focusing broadband illumination from a 120 W lamp onto a millimeter-sized spot, as in Figure 2B–D; alternately, a millimeter-sized He–Ne laser beam with a total power of 20 mW could also be used for droplet control. To localize forces at \( l_e \approx 10^{-6} \) m, a separate mode of operation is required. This mode is activated through the use of a separate micrometer-sized IR laser illumination spot and a higher-frequency ac bias (10^3–10^5 Hz). The localization of forces on such length scales allowed the in situ manipulation of particulate phases inside the droplet. For droplets placed directly on the electrode surface, particulate phases inside the droplets were concentrated on the water–electrode interface by simultaneously focusing IR illumination on the electrode surface and the application of the appropriate ac bias across the electrodes (Figure 2E,F and Supporting Information Movie 1). Particle transport in the device is rapid and can be faster than achieved with many other optical techniques.\(^{29}\) A translation of the laser focal point results in a consequent translation of the vortex center, and hence particle aggregation in this hybrid device is dynamic in nature (Supporting Information Movie 1). When the aqueous droplets were suspended in a silicone oil medium, the particulate phases could be concentrated on the water–oil interface (data not shown).

Cross-scale manipulation in this hybrid device constructively utilizes many different phenomena. The guiding principle of droplet manipulation is OEW. Virtual electrodes are created by utilizing the photoconductive nature of a-Si. The typical illumination used lies in the visible spectrum, and the spot size is approximately the same size as the droplet. Virtual electrodes are determined by the illuminated site through a shift in the principal voltage drop.\(^{24}\) The terms \( C_i, C_w, \) and \( C_{ph} \) are the capacitances of the insulator, the droplet, and the photoconductor, respectively; \( R_w \) and \( R_{ph} \) are the resistances of the droplet and the photoconductor, respectively.

Figure 3. (A) For droplet motion, virtual electrodes are created by utilizing the photoconductive nature of a-Si. The typical illumination used lies in the visible spectrum, and the spot size is approximately the same size as the droplet. Virtual electrodes are determined by the illuminated site through a shift in the principal voltage drop.\(^{24}\) The terms \( C_i, C_w, \) and \( C_{ph} \) are the capacitances of the insulator, the droplet, and the photoconductor, respectively; \( R_w \) and \( R_{ph} \) are the resistances of the droplet and the photoconductor, respectively. (B) Illustration of the mechanism of the particle concentration inside the droplet. An IR laser is focused onto a spot through a microscope objective lens, and suspended particles are transported to the IR-illuminated region. The blue arrows indicate the direction of electrothermal flow. AC electroosmosis also contributes to fluid flow (indicated by orange arrows). The transported particles are trapped on the electrode surface by the concerted action of different electrokinetic mechanisms.

aggregation, activated by IR illumination, was also noticed at the water–oil interface when the dispersed droplet was immersed in an oil medium. Such aggregation at the water–oil interface is not fully understood and needs further investigation. A partially similar mode of in situ particle concentration for a continuous microfluidic system was demonstrated recently by Williams et al.33 Despite some progress,26,27 a comprehensive modeling of the forces present in their system is yet to be achieved. Although those forces are definitely also present in our system, a complete understanding of the particle-concentration process is exacerbated by the numerous additional effects that are present.

Thus, the hybrid optoelectric chip utilizes multiple phenomena, activated by the proper combination of the electric field and the illumination landscape, to achieve manipulation across scales. The hybrid technique can lend itself to the active control of particle patterns resulting from dispensing and evaporating colloidal droplets on the electrode surface. To illustrate this application, a droplet containing 1 \( \mu \)m polystyrene particles was dispensed directly onto the solid surface. An ac electric field (ac frequency \( \sim 10^4 \) Hz) was applied, and an IR laser spot was placed approximately in the middle of one of the electrodes covered by the droplet. The application of a suitable ac electric field and IR illumination initiates rapid particle aggregation at the illumination spot, similar to that depicted in Figure 2E. These conditions are maintained until the complete evaporation of the droplet (active process). Upon the evaporation of the first droplet, a second droplet was dispensed nearby and allowed to evaporate under normal laboratory conditions. No electric field was applied across the electrodes, and only normal laboratory lighting was present (passive process). The second droplet serves mainly as a control. Although evaporation in both cases leaves an assembled structure of polystyrene beads reflecting a result of acting forces, imaging of the assemblies demonstrates striking differences between the two cases (Figure 4). Figure 4 is a composite image, created by juxtaposing parts of epifluorescent images of the particulate deposits. The bright areas indicate the particle deposits. In Figure 4, the right quarter circle demonstrates the

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microstructure formed by the passive process, where a ringlike structure is formed by the aggregation of the particulates at the droplet periphery. Such a ringlike structure is an assembly formed commonly by a passive evaporation process.14 The left quarter circle in Figure 4 shows that particle deposition for the active process is almost exclusively concentrated at the location of the IR spot. Electrokinetic flow, as depicted in Figure 2E,F and Supporting Information Movie 1, thus alters the assembly process resulting from the droplet drying process. To confirm the role of electrokinetic flow, experiments were also conducted in the absence of electric fields and with the sole application of an IR beam. Although the sole application of IR illumination could also result in some localized deposits, such deposits were found to be far less pronounced (Supporting Information Figure S1).

Figure 4 demonstrates the ability of the hybrid platform to control particle deposition actively. Prior to this control over particle assembly, structures have been attempted through passive processes such as control of the ambient environment or the introduction of surfactants. Scanning electron microscope (SEM) imaging established the presence of at least five particle layers in both deposition patterns. However, particle packing in the deposits achieved through the active and passive processes show very different structures. The deposit formed by the active process (left half of Figure 4) exhibits a loose packing of particles (Figure 5A–C); in contrast, the ringlike deposit formed by the passive process (right half of Figure 4) exhibits a hexagonal close-packing of particles that had been fused together during the assembly process (Figure 5D–F). This difference can be understood in terms of the dipolar forces, which would cause higher interparticle repulsive forces in the active process as compared to those in the passive process. Thus, the active process results in a reconfigurable assembly.

Conclusions

We demonstrate a hybrid optoelectric device that has the ability to manipulate objects ranging from microliter droplets to micrometer particles. The ability to control the transport of droplets dynamically and the in situ localization of particles will enable the development of novel, integrated sensing devices including preconcentrators that can collect and transport analytes to nanosensors, including electrodes nanopatterned with biological molecules, among many others. Although applications such as those mentioned above definitely stand to benefit, we also demonstrate the novel ex situ control of microstructures created through a droplet evaporation process. Such an application, at present, is conceivable only through a hybrid optoelectric approach. Our multiscale, multiphysics approach will enable a new class of assays that can be translated to clinical or point-of-care use.

Supporting Information Available: Assemblies formed by the aggregation of fluorescent 1.0 μm polystyrene beads after the evaporation of two droplets placed on the hybrid device. Movie depicting the dynamic aggregation of 1.0 μm polystyrene beads in a droplet when ITO electrodes are illuminated with 1064 nm light. This material is available free of charge via the Internet at http://pubs.acs.org.