Modulating patterns of two-phase flow with electric fields

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This paper describes the use of electro-hydrodynamic actuation to control the transition between three major flow patterns of an aqueous-oil Newtonian flow in a microchannel: droplets, beads-on-a-string (BOAS), and multi-stream laminar flow. We observed interesting transitional flow patterns between droplets and BOAS as the electric field was modulated. The ability to control flow patterns of a two-phase fluid in a microchannel adds to the microfluidic tool box and improves our understanding of this interesting fluid behavior. © 2014 AIP Publishing LLC.

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Microfluidic technologies have found use in a wide range of applications, from chemical synthesis to biological analysis to materials and energy technologies.^{1,2} In recent years, there has been increasing interest in two-phase flow and droplet microfluidics, owing to their potential for providing a high-throughput platform for carrying out chemical and biological analysis and manipulations.^{3–8} Although droplets may be generated in many different ways, such as with electric fields or extrusion through a small nozzle,^{9–12} the most common microfluidic methods are based on the use of either T-junctions or flow-focusing geometries with which uniform droplets can be formed at high frequency in a steady-state fashion.^{13,14} Various operations, such as cell encapsulation, droplet fusion, splitting, mixing, and sorting, have also been developed, and these systems have been demonstrated for a wide range of applications, including cell analysis, protein crystallization, and material synthesis.^{1–17}

In addition to forming discrete droplets, where a disperse phase is completely surrounded by a continuous phase, it is also possible in certain situations to have different phases flow side-by-side. In fact, multi-stream laminar flow, either of the same phase or different phases, has been exploited for both biochemical analysis and microfabrication.^{1,2,18–20} Beads-on-a-string (BOAS) is another potential flow pattern, which has been attracting attentions in microfluidics field. BOAS flow, owing to its special flow structures, may be particularly useful in some applications, such as optical-sensor fabrication.²¹ In BOAS flow, queues of droplets are connected by a series of liquid threads, which makes them look like a fluid necklace with regular periods.^{21–25} The BOAS pattern is easily found in nature, such as silk beads and cellular protoplasm, and is often encountered in industrial processes as well, such as in electrospinning and anti-misting.^{21,22} In general, it is thought that BOAS structure occurs mostly in viscoelastic fluids²² and is an unstable structure, which evolves continually and breaks eventually.^{21–29}

Flow patterns determine the inter-relations of fluids in a microdevice and are an important parameter to control. Common methods for adjusting microfluidic flow patterns include varying the fluid flow rates, fluid properties, and channel geometries. Additionally, the application of an electric field can be a useful supplement for adjusting microfluidic flow patterns, although most work in this area has been focused on droplets and in some cases also on multi-stream laminar flows.^{30–33} Here, in addition to forming droplets and two-phase laminar flow with electro-hydrodynamic actuation, we also observed a new stable flow pattern in a non-viscoelastic fluid, BOAS flow. Such flow patterns may find use in controlling the interactions between droplets, such as limited mixing by diffusion between neighboring droplets.

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FIG. 1. (a) Schematic of the setup. (b) Micrograph showing droplet generation in a flow-focusing junction. The scale bar represents $40\mu\text{m}$.

To generate droplets, we used the flow-focusing geometry (Figure 1(a)), in which aqueous phase (water) was flown down the middle channel and droplets were pinched off by the oil phase (1-octanol) from the two side channels at the junction; Figure 1(b) shows the droplets formed after the junction. To apply electric field along the main channel where the droplets were formed, we patterned a pair of electrodes upstream and downstream of the junction (Figure 1(a); for experimental details, please see Ref. 34 for supplementary material). The average electric field strength may be calculated from the voltages applied and the distance (1.7 mm) between the two electrodes. When a high voltage was applied along the channel between the two electrodes, the aqueous-oil interface at the flow-focusing junction became charged and behaved like a capacitor. As a result, more negative charges were drawn back upstream towards the positive electrode, and left behind more positive charges at the aqueous-oil interface, which then became encapsulated into the aqueous droplets dispersed in the oil phase.

The positively charged aqueous-oil interface was stretched under an applied electric field, and by adjusting the voltage and/or the two-phase flow-rate ratio, we found interestingly that various flow patterns emerged. We tested different combinations of applied voltages and flow-rate ratios and found that most of them resulted in similar flow patterns and transitions between flow patterns.

Figure 2 illustrates the effects of varying the applied voltages on droplets at a fixed liquid flow rate. With increasing electric-field strength and force, we found it was easier for the aqueous phase to overcome interfacial tension and form droplets. For example, as the voltage increased from 0.0 kV to 0.8 kV (average field strength increased from 0 to $0.47\text{ V}/\mu\text{m}$), droplet-generation frequencies became slightly higher, and the formed droplets were smaller in volume. Additionally, droplets gradually became more spherical in shape at higher voltages.

As the voltage increased further (e.g., up to 1.0 kV in Figure 3), the distance between neighboring droplets became smaller, and the aqueous-oil interface at the junction was stretched further toward the downstream channel. At a threshold voltage (1 kV here with corresponding average field strength of $0.59\text{ V}/\mu\text{m}$), the tip of the aqueous-oil interface would catch up with the droplet that just formed, and the tip of the interface of this newly captured droplet would in

FIG. 2. Images showing the effects of applied voltage on droplet shape and flow pattern. Oil-phase flow rate, $0.5 \mu\text{l}/\text{min}$; aqueous-phase flow rate, $0.2 \mu\text{l}/\text{min}$. The scale bar represents $40 \mu\text{m}$.

turn catch up with the interface of the droplet that formed before it. Consequently, a series of threads would connect all the droplets flowing between the two electrodes, thus resulting in a BOAS flow pattern.

At voltages near the threshold value, the flow pattern was not stable, but oscillated between droplets flow and BOAS flow. Figure 3 is a series of images captured by a high-speed camera that show the flow in this transition region. In Figures 3(a) and 3(b), the string of BOAS

FIG. 3. Series of images showing the reversibility and synchronicity of a transitional flow pattern between droplets and BOAS (bead-on-a-string). Voltage applied, 1.00 kV (corresponding field strength of $0.59 \text{ V}/\mu\text{m}$); oil-phase flow rate, $0.5 \mu\text{l}/\text{min}$; aqueous-phase flow rate, $0.2 \mu\text{l}/\text{min}$. The scale bar represents $40 \mu\text{m}$.

became thinner over time, and then the BOAS broke into droplets (Figures 3(c) and 3(d)). The newly formed droplets, however, were not stable either. Thin liquid threads would appear and then connect neighboring droplets, and a new switching period between discrete droplets and BOAS would repeat (Figures 3(e)–3(h)). In addition to this oscillation and reversibility, the flow pattern had a synchronous behavior: all the droplets appeared connected simultaneously by liquid threads or were separated at the same time.

When the voltage reached 1.3 kV, which corresponded to an average field strength of $0.76 \text{ V}/\mu\text{m}$, a stable BOAS flow was obtained (Figure 4(a)). BOAS structures are thought to be present mostly in viscoelastic fluids,²² because viscoelasticity is helpful in enhancing the growth of beads and in delaying breakup of the string; thus, the viscoelastic filament has much longer life time than its Newtonian counterpart. Here, with the help of electric field, regular BOAS structures are realized in a non-viscoelastic fluid (water) in microchannels.

Microenvironment and electric fields alter the common evolution of BOAS structure observed in macroscopic or unbound environments. The BOAS structure formed in our experiments is not a stationary pattern, but a steady-state flowing one. Electric-field force prevents liquid strings from breaking between beads, and thus plays a similar role as elastic force in viscoelastic fluids. Figure 4(b) shows the dynamic BOAS profile, obtained at a fixed plane (shown in Figure 4(a)) perpendicularly across the channel as the BOAS structure passed through it. Droplets and liquid-thread diameters were nearly constant during the sampling time. The longer term experiments (over 3 min) showed there were slight variations of the two diameters in time, but the essential BOAS structure still remained qualitatively the same as a whole.

When the voltage was further increased, the string diameter became larger and the droplet diameter became smaller. Because of the low flow-rate ratio (0.4) between the aqueous phase and oil phase used in the experiment depicted in Figure 4, the flow did not further develop into a multi-stream laminar flow, as would be expected at a higher voltage, and instead became

FIG. 4. (a) Micrograph showing BOAS flow in a channel. (b) Profile of the top-half of the BOAS flow recorded continuously at a cross-section (shown in Figure 4(a)) of a channel. Voltage applied, 1.30 kV (corresponding field strength of $0.76 \text{ V}/\mu\text{m}$); oil-phase flow rate, $0.5 \mu\text{l}/\text{min}$; aqueous-phase flow rate, $0.2 \mu\text{l}/\text{min}$. The scale bar represents $40 \mu\text{m}$.

FIG. 5. Micrograph showing multi-stream two-phase laminar flow in the channel. Voltage applied, 3.00 kV (corresponding field strength of 1.76 V/ μm); oil-phase flow rate, 0.5 $\mu\text{l}/\text{min}$; aqueous-phase flow rate, 0.5 $\mu\text{l}/\text{min}$. The scale bar represents 40 μm .

unstable and irregular. When the flow-rate ratio was increased to 1.0 and the voltage was adjusted to 3.0 kV (corresponding field strength of 1.76 V/ μm), we observed a stable multi-stream laminar flow (Figure 5). The aqueous stream flowed in the channel center surrounded by the oil phase on the sides. This experiment showed that higher electric-field strengths alone would not give rise to another stable flow pattern (i.e., multi-stream laminar flow), but a suitable flow-rate ratio of aqueous phase to oil phase is required for the formation of stable two-phase laminar flow.

The flow patterns we observed may be described by a phase diagram (Figure 6), which depends on two dimensionless numbers: capillary number, $\text{Ca} = \mu_a U_a / \sigma$, and electric Bond number, $\text{Bo}_e = E^2 (\epsilon D / \sigma)$. Ca and Bo_e describe the ratio of viscous force to interfacial tension force and the ratio of electric-field force to interfacial tension force, respectively. Here, μ_a (1 mPa s), σ (8.5 mN/m), ϵ (7.1×10^{-10} F/m), E , U_a , and D are, respectively, the aqueous-phase viscosity, aqueous-oil interfacial tension, aqueous-phase permittivity, electric field strength, aqueous-phase velocity, and the hydraulic diameter of the channel at the junction. Figure 6 shows clearly that at higher Ca , flow pattern changes gradually from droplet to BOAS and to multi-stream laminar flow with increasing Bo_e , which indicates the increasing importance of the electric-field force compared with the interfacial tension force. At lower Ca , flow pattern and transition show similar trend with increasing Bo_e as in the higher Ca case, except that multi-stream laminar flow is not observed. The relatively higher viscous force at higher Ca may be necessary for transitioning to the multi-stream laminar flow regime. In addition, Figure 6 shows that the BOAS window at the lower Ca is smaller than that at the higher Ca .

In summary, we showed the ability to use electric fields to generate and control different flow patterns in two-phase flow. With the aid of an applied field, we were able to generate BOAS flow patterns in a non-viscoelastic fluid, a pattern that typically requires a viscoelastic fluid. The BOAS structure was stable and remained as long as the applied electric field was on.

FIG. 6. Phase diagram showing different flow patterns in the Ca and Bo_e space. Hollow symbols: oil-phase flow rate, 0.5 $\mu\text{l}/\text{min}$; aqueous-phase flow rate, 0.5 $\mu\text{l}/\text{min}$. Solid symbols: oil-phase flow rate, 0.5 $\mu\text{l}/\text{min}$; aqueous-phase flow rate, 0.2 $\mu\text{l}/\text{min}$.

We also report transitional flow patterns, those between droplets and BOAS exhibited both good reversibility as well as synchronicity. And with a suitable flow-rate ratio between the two phases, BOAS flow could be transitioned into a stable two-phase laminar flow by applying a sufficiently high field strength. Finally, a phase diagram was presented to describe quantitatively the flow-pattern regimes using capillary number and electric Bond number. The phenomena we report here on the properties of two-phase flow under an applied electric field may find use in developing a different approach to exert control over droplet based or multi-phase laminar-flow based operations and assays, and also aid in understanding the physics of multi-phase flow.

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