MONODISPERSE DROPLET GENERATION USING ELECTRICAL PULSES

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ABSTRACT

Microdroplet generation is of interest due to its use in applications such as drug delivery. Although flow focusing has emerged as the most popular droplet generation technique it does not offer rapid size variation or drop-on-demand capabilities. To overcome this limitation attempts have been made to use high voltage pulses to extract single droplets on demand. Although a step in the right direction, continuous droplet generation and its control have not been investigated. In this paper we study the effect of various electrical and flow parameters on the ability to generate monodisperse droplets (10 – 100 μm) continuously in a microfluidic setup through electro-generation.

KEYWORDS: Droplets, Flow Focusing, Electric, Drop-On-Demand,

INTRODUCTION

Microdroplets have a long list of uses, the most important of which are in drug delivery, combinatorial analysis and massively parallel assays in biology and medicine [1, 2]. Microgels are sought in drug delivery due to their monodispersity in size which effectively controls their drug release rates. Similarly, in biological studies, microdroplets are used to conduct large sets of screening experiments on small sample volumes consuming small quantities of expensive reagents. A variety of methods such as inkjet printing and emulsification are capable of microdroplet production. However, with these methods, control over droplet size is limited and droplets that are produced have large size distributions. Microfluidics overcomes many of these limitations offering the possibility of handling small volumes of fluid at low to medium throughput. Flow focusing, where a dispersed phase is co-flowed with a continuous phase through a narrow nozzle [3], has emerged as the most popular microfluidic method for producing monodisperse dispersions. However, this method lacks the ability to have dynamic control over droplet size nor can it produce drop-on-demand. One of the most promising technique for dynamic control of droplet size is the application of a high voltage electrical signal. Significant work has been done in electrospray technology where AC or DC signals are used to produce dispersions of aqueous/organic phase in air [4]. This technique is capable of large volume droplet production but is not well suited for many applications since the sizes are typically limited to under 10 μm, produces satellite droplets and control over size is difficult [5]. Investigations by Atten *et al.* [6] improves upon this method by introducing control over droplet sizes via pulsing and produced droplets 50-200 μm in size. However, their investigation focused on producing a single droplet without a satellite droplet and not on continuous production. Similarly, He *et al.* [7] demonstrated the use of high voltage pulses to form femtoliter sized single droplets in a microfluidic channel. Although these methods demonstrated droplet generation via electrical signals they do not offer the ability to generate large quantities of droplets whilst maintaining precise control over size or frequency of generation.

Here we present the design and analysis of a microfluidic device capable of continuous droplet production with the application of high voltage pulsed signals. Low frequency droplet generation is examined and control over droplet sizes is demonstrated in this device. This work successfully demonstrates monodisperse microdroplet generation at varying frequencies and introduces a method where microdroplets can be generated with dynamic control over size by varying the frequency of electrical waveforms.

WORKING PRINCIPLE

The droplet generation process in flow focusing microfluidic devices is triggered by the instability initiated by the flow of two phase liquid through a narrow orifice. But since the structure of the orifice is defined and constant for the same device, the only method to control the droplet formation process and the size and rate of droplet generation is through the control of flow rates. However, due to fluidic capacitances in the device, the transient time associated with the change in flow characteristics and the ensuing change in droplet generation process is quite long. Alternatively, flow instabilities can also be initiated by other methods. For instance, it is well known that electric potential change surface tension of a biphasic interface. Thus by application and removal of an electric potential temporal variations in surface tension can be initiated leading to flow instabilities. Here we use pulses to introduce interface instability on demand to initiate droplet formation. In this scheme, the application of an electric field reduces surface tension at the tip of capillary of the dispersed phase. In addition, it introduces a electrostatic force that pulls the interface towards the other electrode. This process will continue till the interface becomes unstable. Instability can be due to random disturbances in the surface or can be induced deliberately when the electric pulse is turned off. In the second case, the surface tension increases, and the present state of the interface cannot be sustained any longer leading to the breakup of the interface and droplet formation. Wright *et al.* [8] determined by simple analysis that in order to eject a droplet from a capillary one must overcome the capillary pressure which is given by:

$$P_{cp} = \frac{2\gamma}{r} \quad (1)$$

where P_{cp} is the capillary pressure, γ is the interfacial tension between the water/oil solutions, r is the capillary radius. In electrogeneration, the electrostatic pressure pulling the fluid out must overcome the capillary pressure holding the meniscus in place. The electrostatic pressure is given by:

$$P_e = {}^{1}\!/_{2}\ \varepsilon E_n^2 \qquad (2)$$

where P_e is the electrostatic pressure, ε is the permittivity of the insulating fluid, and E_n is the electric field normal to the meniscus surface. By equating the two pressures it is possible to estimate the minimum voltage required to eject a droplet from a capillary.

EXPERIMENTAL

The experimental setup consists of three modules for signal generation, imaging, and pumping. The signal generation module provides pulsed high voltage signals to the device produced by a combination of a function generator and an amplifier. A Tektronix AFG 3022B function generator was utilized to produce various pulsed waveforms of defined frequency and duty cycle. These signals were then amplified by a Trek 10/10b HS high voltage amplifier. The rise and fall time are under 1% of the lowest pulse duration and care was taken to ensure that the signal shape is not compromised. The working electrode is connected to the a syringe containing DI water with methylene blue added for visualization and conductivity. The ground is connected to a stainless steel needle placed 600 μm away from the capillary as shown in Figure 1.

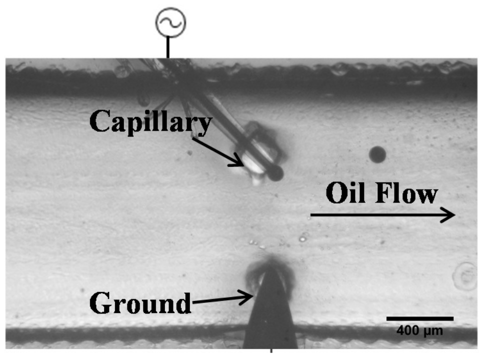

Figure 1: Microfluidic device for droplet generation, showing ground electrode, droplet injection capillary, and oil flow.

Two syringe pumps were connected to the microfluidic device to control the flow rates. A KDS Legato 270 (GeneQ Inc., Quebec, Canada) high precision syringe pump provided pulse free flow for the dispersed phase in the capillary. The dispersed phase used was DI water with 1% wt of methylene blue. . A Harvard Apparatus 22 infusion pump was used to control the flow of continuous phase. The continuous phase used was heavy paraffin oil with 1% by wt Span80 surfactant added after set up.

Imaging of the droplet generation process was done through a high speed camera attached to a microscope. A Photron Fastcam Troubleshooter high speed camera was used for imaging. Videos were recorded 3 minutes after the electrical signal was changed or applied at 1000 FPS to eliminate motion blurring.

Droplet diameter measurements were performed with ImageJ. With pictures having pixel resolutions lower than 1 μm the largest amount of error was due to the manual measurements. The 95% confidence interval of the measurements was determined to be +/-0.52% of the measured value. In all cases measurements were done on the primary droplet, which is in all cases the largest droplet ejected per pulse. This experimental setup was utilized in all experiments to determine the optimum design for a device capable of droplet generation via high voltage electrical signals.

RESULTS AND DISCUSSION

A fluidic device consisting of a channel 1.5 x 0.8 mm (Fig. 1) was designed which contained a 50 μm inner diameter fused silica capillary for droplet injection. The high voltage electrical signal is applied to the dispersed phase reservoir located outside of the device while the needle tip inside the channel is grounded. When a pulse (3 Hz, 1400 peak voltage, 0 bias, 670 μs pulse width) was applied, coulomb fission was observed (Fig. 2a) at the meniscus of the capillary. Coulomb fission is caused by excess charge accumulating in the meniscus and in the droplet formed. The repulsion of these like charges overcomes the surface tension and causes the droplet to split in order to increase surface area and accommodate the excess charge. To avoid this phenomenon and produce monodisperse drops, the peak voltage and pulse width were reduced to 1100 V and 560 μs respectively. As seen in figure 2b fission was eliminated and stable generation of a primary droplet with a single, small secondary droplet was observed. These experiments demonstrate that through the control of the voltage applied and the duration of the pulse the dynamics of droplet formation and ejection can be

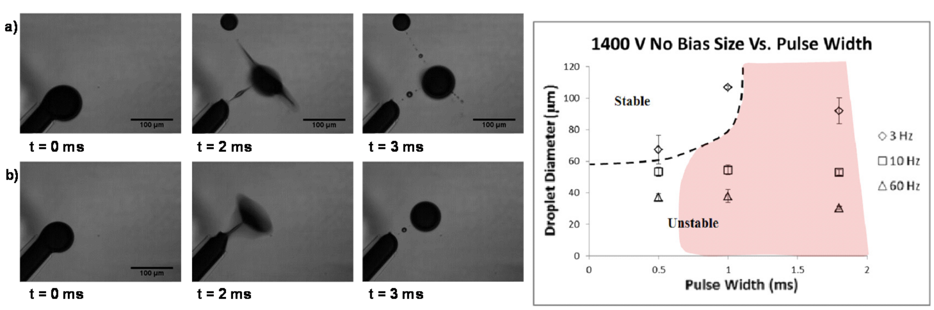

Figure 2: Modes of droplet generation when a single high voltage pulse is applied. a) When coulomb fission is present b) Ideal generation when no fission occurs.(Scale bar 100 μm).

controlled. In order to determine the parameter space suitable for monodisperse droplet generation, experiments were conducted at various frequencies, flow rates, bias voltages and pulse widths. Figure 2 shows that for a defined electric potential and flow rates, monodisperse droplet generation with minimal secondary droplets occur at low pulse widths. Additionally, it shows that modulating the pulse width over a limited range changes the diameter of the droplet generated.

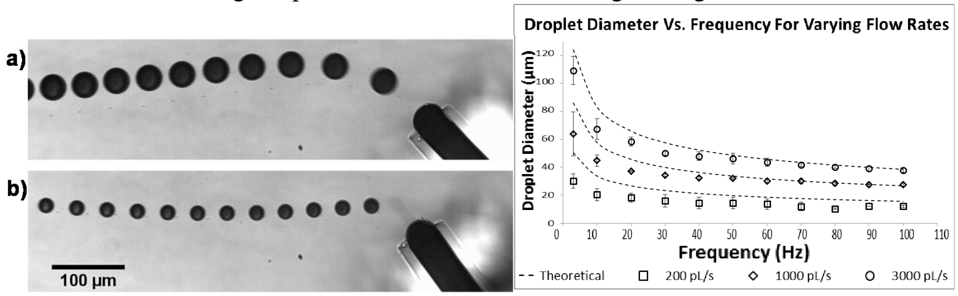

Figure 3: 90 Hz droplet generation at different capillary flow rates a) 3000 pL/s b) 1000 pL/s. Oil flow rate 8 mL/hr. Droplet generation is demonstrated to be stable and monodisperse.

When flow rates and frequencies were held constant the parameters of pulse width, peak voltage, and bias were varied to determine the optimum conditions where droplet generation is stable and monodisperse (figure 3). It is observed that for each frequency, a unique set of parameters exist where droplet generation is monodisperse. Series of experiments were performed to obtain results in figure 5 where monodisperse droplets (10-110 µm) are generated at different flow rates (200 – 3000 µL) and frequencies (3 - 100 Hz). Each point represents an unique combination of electrical (frequency, pulse width, peak and bias voltages) and flow (oil and water flow rates) that produce monodisperse droplets with minimal secondary droplet generation. Figure 5 also shows that droplet diameters can be changed by changing just the electrical parameters while holding the flow parameters constant. This offers a rapid way to instantaneously switch between droplet sizes without the need to wait for flow stabilization. Although droplet sizes do decrease proportionally with increasing frequency, some deviation existed between the experimental results and projected diameters (estimated utilizing the values for flow rates and frequencies). This was due to the changes in peak/bias voltages and pulse width at each frequency in order to maintain monodisperse stable droplet generation. These results show that control over droplet diameter is possible through changes in the capillary flow rate or frequency. Changes in the electrical signal enable the control of droplet diameter immediately, within seconds, whilst changes in flow rates require a period of up to 15 minutes for stability.

CONCLUSION

This method of droplet generation provides the user rapid control over droplet sizes via control of the electric signal with additional control possible through changes in the capillary flow rate. Using these results it is possible to electrogenerate monodisperse microdroplets in a microfluidic device at frequencies up to and exceeding 100 Hz.

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