

# CONTROLLED DROP GENERATION FOR DIGITAL MICROFLUIDIC SYSTEMS BY MEANS OF ELECTROWETTING

Hao Gu\*, Michel H. G. Duits and Frieder Mugele

Physics of Complex Fluids, MESA+ Institute, University of Twente,  
PO Box 217, 7500 AE, Enschede, The Netherlands

## ABSTRACT

Digital microfluidic two-phase flow systems have achieved an impressive degree of flexibility and versatility. Yet, the primary generation of drops is generally still controlled solely by the imposed flow rates of both continuous and dispersed phase and the resulting hydrodynamic forces. Here we present a novel method to overcome this lack of precise on-demand control of the drop generation using hybrid microfluidic channel systems with incorporated electrowetting functionality. Using a fixed flow rate ratio in combination with freely programmable electrical waveforms on the electrodes, arbitrary sequences of drops with variable volume ( $\approx 0.5\text{fL} \sim 1.6\text{pL}$ ) can be generated.

**KEYWORDS:** Microfluidics, Electrowetting, Drop Generation

## INTRODUCTION

There is a growing interest in two-phase flow microfluidic systems for highly controlled generation of drops in the past few years. Present technology for drop generation is mainly based on the imposed flow rates of both continuous and dispersed phases in combination with an appropriately designed geometry. Alternatively, electrostatic actuation mechanisms provide active and precise control on the drop generation. Recently, Kim *et al.*[1] integrated electrodes into a microchannel to control micrometer-sized drops in a electrosprayinglike process requiring voltages in excess of 1000V. At same time, we introduced the concept of integrating electrowetting (EW) into a flow-focusing device (FFD) and demonstrated drop size control at a much lower voltage. [2-4]

In current paper, we explain the general concept of hybrid EW-enhanced digital microfluidic systems and present in particular a simple, rapid and inexpensive method to fabricate FFD and insulator-covered EW electrodes. Use of a thiolene precursor allows defining the channel geometry via soft imprint lithography, as well as bonding of the chip via exposure to UV light. Compared to earlier polydimethylsiloxane (PDMS)-based designs, this method allows us to make microchannels with smaller dimensions, lower aspect ratios, and symmetric electrodes both on the top and the bottom of the channel. We demonstrate their performance on drop generation controlled by tuning ac voltage  $U$  [0, ..., 100V<sub>rms</sub> (root mean square)] with various ac frequencies  $f$  [100Hz, 1kHz, 10kHz].

## THEORY

EW is an increasingly popular method for modulating the surface tension because of its simple device configuration, ease of electronic control and low energy consumption. EW refers to an electrically induced reduction in contact angle of a conductive liquid on a dielectric coated electrode. The standard EW configuration comprises a solid electrode surface covered with a thin dielectric material. In case the latter is not hydrophobic, it is coated with an additional thin hydrophobic layer. Application of an electric potential between the solid electrode and the aqueous liquid then allows a quantitative control over the deformation of the liquid surface. The apparent contact angle  $\theta(U)$  depends on both Young's angle  $\theta_Y$  and the applied voltage  $U$ :

$$\cos\theta(U) = \cos\theta_Y + \frac{c}{2\sigma}U^2 \quad (1)$$

Where  $c$  is the capacitance per unit area between the drop and the substrate, and  $\sigma$  is the interfacial tension between the drop and the ambient medium. Note that equation (1) remains valid for ac voltage if one replaces  $U$  with its root mean square (rms) value  $U_{\text{rms}}$ .

## EXPERIMENTAL

Fig. 1(a) shows the schematic of EW-based FFD. To implement EW into microchannels, we use commercial indium tin oxide (ITO)-coated glass as a substrate, covered with dip-coated Teflon AF as an insulator. Microchannels are fabricated by soft imprint lithography using NOA 81 (Norland optical adhesive). NOA 81 is imprinted by a structured PDMS mold and then cured by UV irradiation. After peeling the PDMS mold from the crosslinked NOA 81, microchannels are replicated on the bottom substrate. Meanwhile a very thin uncured NOA 81 layer remains because porous PDMS facilitates the delivery of oxygen to the surface of NOA 81. Top substrate is the same as the bottom one. The holes are drilled on it for inlet and outlet connections. The bonding between top substrate and the bottom part is facilitated by previously mentioned thin uncured NOA 81 layer which causes adhesion between the top and bottom surfaces. Using UV irradiation for 180s permanently seals microchannels.

The last step for chip fabrication is a surface modification. NOA surfaces are prone to show poorly controllable wetting properties: both hydrophilic and oleophilic behaviors have been observed. Such variations pose a serious obstacle for controlling two-phase flows, which can be sensitive to small changes in wettability. For the requirement of EW, we apply a silanization treatment to make the NOA 81 surface hydrophobic. 1% (v/v) 1H, 1H, 2H, 2H perfluorodecyltrichlorosilane (FDTs) in isooctane solution is injected into the microchannel. After 15 min of incubation, the channel is

flushed by isooctane and isopropanol respectively, each for 20 min. The device is ready for use after drying overnight at room temperature. The detailed fabrication process and characterization of modified NOA surface are previously reported in ref. [5].

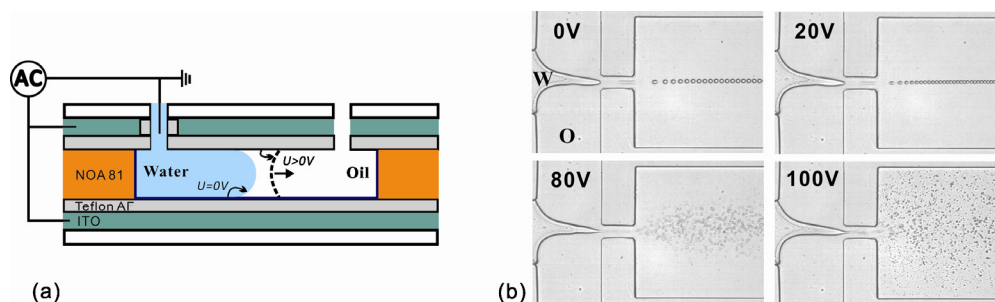


Figure 1: (a) Side-view schematic of EW-based FFD. Voltage is zero, contact angle is larger than  $90^\circ$ ; voltage is on, contact angle is less than  $90^\circ$ . (b) Top views in the drop generation modes at different ac rms voltages with a frequency of 10kHz. (flow rate ratio  $Q_w/Q_o=1/600$ )

For two-phase flow system, the aqueous dispersed phase is de-ionized water with dissolved NaCl (conductivity is 5-7 mS/cm). Mineral oil (viscosity:  $\mu_o = 0.03$  Pa·s) with Span 80 (5 wt%) added as a surfactant is utilized as oil phase. The interfacial tension ( $\sigma$ ) of water-oil with Span 80 was measured to be 5 mN/m. The electrodes underneath the channel connect to an ac voltage source operating at variable rms voltages from 0 to 100V and three different frequencies (100Hz, 1kHz and 10kHz), meanwhile the aqueous phase was connected to ground. In the experiment of water-in-oil drop generation, the water and oil flow was driven using syringe pump (Harvard PHD 2000, Harvard Apparatus).

## RESULTS AND DISCUSSION

For drop generation, a flow-focusing geometry is used [see Fig. 1(b)]. The height of channel is  $10\mu\text{m}$  and the width of orifice is  $20\mu\text{m}$ . In the experiments reported here, the flow rate of continuous phase (oil phase),  $Q_o$ , was kept fixed at  $60\mu\text{L/h}$ , corresponding to a capillary number  $Ca = \mu_o v / \sigma$  of 0.5, where  $v$  is the average velocity of oil phase. Fig. 1(b) demonstrates two drop generation modes: dripping (0V and 20V) and conical spray mode (80V and 100V). To explore the influence of voltage  $U$  at a constant ac frequency (10kHz) on drop generation control, three different flow rate ratios of dispersed phase (aqueous phase) and continuous phase were chosen, ( $Q_w/Q_o$ : 1/6, 1/60 and 1/600). The different regimes of drop generation are found for various combinations of  $U$  and  $Q_w/Q_o$ .

In Fig. 2 we show the drop size and the drop generation rate as a function of voltage  $U$ , as obtained by quantitatively analyzing video data such as in Fig. 1(b). Fig. 2(a) indicates that the variation of  $Q_w/Q_o$  and  $U$  gives various ways for generating drops in the size range of  $\approx 0.5\text{fL} \sim 1.6\text{pL}$ . At any  $Q_w/Q_o$ , the drop size decreases with increasing  $U$  (up to 60V). In the case of  $Q_w/Q_o = 1/600$ , fairly monodisperse size distributions are found (polydispersity  $\sigma_D \approx 1\% - 4\%$ ) for  $U$  up to 40V. For  $U > 60\text{V}$ , we observe a conical spray, in which the drops spread out over a much wider range in the expanded downstream. Interestingly, the tiny drops ( $\approx 0.5\text{fL} \sim 26\text{fL}$ ) in this regime are rather monodisperse, which could be attractive for application. When knowing the volumes of drops and  $Q_w$ , also the drop generation rate can be calculated. [see Fig. 2(b)] We note that in all cases of  $Q_w/Q_o$  drop generation rate follows an increasing trend with increasing  $U$  (up to 60V). For  $Q_w/Q_o = 1/600$ , the generation rate of the drops still increases when  $U$  goes from 60V to 100V. In particular to the case of  $U = 100\text{V}$ , the generation rate is quite high ( $6 \times 10^4 \text{ s}^{-1}$ )

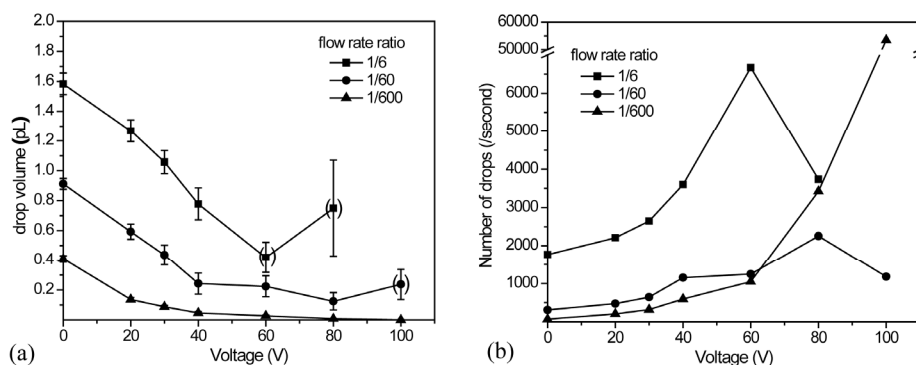


Figure 2: (a) Characterization of drop size evolution with an increasing ac voltage  $U$  at constant ac frequency 10kHz and different  $Q_w/Q_o$ . Bracket data points indicate cases where significant polydispersity was encountered; here only the primary drops were incorporated in the analysis. Error bars indicate the standard deviation of the size distribution. (b) Drop generation rate as a function of applied voltages at constant ac frequency 10kHz and different  $Q_w/Q_o$ .

The ac frequency also can be used to influence the drop generation. Fig. 3 shows representative snapshots of drop generation for variable  $U$  and ac frequency  $f$  at constant  $Q_w/Q_o = 1/600$ . At low  $f$  (100Hz), tri-disperse drops (three different drop sizes) are generated at 40V [Fig. 3(a)]. With the increase of  $U$ , the drops spread out over a much wider range in the downstream channel. The drop sizes become polydispersed [(b)-(d)]. At moderate  $f$  (1kHz), rather monodisperse primary drops with tiny satellites drops are generated at 40V. Also the drop sizes become polydispersed with increasing  $U$  [(e)-(h)]. At high  $f$  (10kHz), we are able to generate very fine monodisperse drops in all cases of  $U$  [(i)-(l)]. It is clearly shown that a higher ac frequency offers smaller and more monodisperse drops.

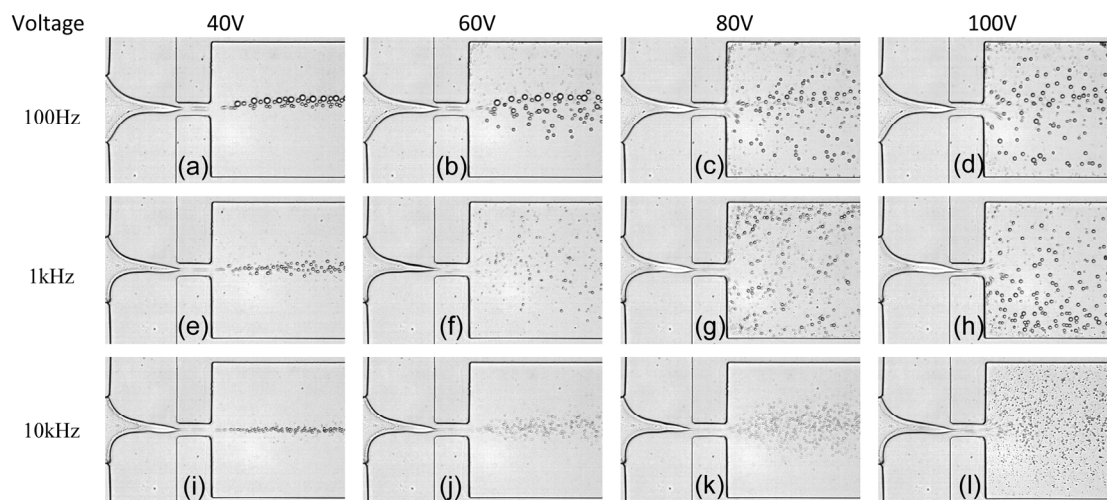


Figure 3: Representative images of drop generation at different  $U$  and ac frequencies  $f$ . ( $Q_w/Q_o$ : 1/600)

Here we interpret the role of EW in the drop generation process. The operation of water-oil interface is mainly controlled by hydrodynamic forces (pressure gradient, viscosity, interfacial tensions) at low to moderate voltage (up to 40-60V). In this range, EW primarily changes the global shape of the water-oil interface by reducing the water CA on the channel walls. This leads to a rather efficient fine control over the drop sizes. At voltages of order 60V and higher, the conical spray regime indicates that EW become more dominant and the drops in this regime carry electrical charge. This may be rationalized assuming that the drops are formed by a mechanism involving the electrical Maxwell stress as a primary driving force, similar to the satellite drops generation during the instability of contact lines at high voltage in EW. The influence of ac frequency on drop generation is not fully understood. It is assumed that EW with different ac frequencies can change the charge distribution inside generated drops, and then affect the drop size distribution because unexpected coalescence events occur at certain conditions.

## CONCLUSION

In conclusion, we demonstrated a simple and low-cost method to construct a digital microfluidic system in combination with EW. The powerful control on drop generation can be achieved by varying ac voltages and ac frequencies. The EW-based approach offers an active and precise control on a wide and continuous range of drop size (0.5fL~1.6pL). Compared to other electrically assisted routines, our method offers these capabilities at a much lower voltage.

## ACKNOWLEDGEMENTS

The authors acknowledge support from the MicroNed programme, part of the Decree on subsidies for investments in the knowledge infrastructure (Bsik) from Dutch government for financial support, as well as the research institutes IM-PACT and MESA+ at Twente University.

## REFERENCES

- [1] H. Kim, D. Luo, D. Link, D. A. Weitz, M. Marquez, and Z. Cheng, "Controlled production of emulsion drops using an electric field in a flow-focusing microfluidic device", *Appl. Phys. Lett.*, vol. 91, pp. 133106, 2007
- [2] F. Malloggi, S. Vanapalli, H. Gu, D. van den Ende, F. Mugele, "Electrowetting-controlled droplet generation in a microfluidic flow-focusing device", *J. Phys.: Condens. Matter*, vol. 19, pp. 462101, 2007
- [3] F. Malloggi, H. Gu, A.G. Banpurkar, S.A. Vanapalli, F. Mugele, "Electrowetting – A versatile tool for controlling microdrop generation", *Eur. Phys. J. E*, vol. 26(1-2), pp 91-96, 2008
- [4] H. Gu, F. Malloggi, S. A. Vanapalli, and F. Mugele, "Electrowetting-enhanced microfluidic device for drop generation", *Appl. Phys. Lett.*, vol. 93, pp. 183507, 2008
- [5] H. Gu, M. H. G. Duits and F. Mugele, "A hybrid microfluidic chip with electrowetting functionality using ultraviolet (UV)-curable polymer", *Lab Chip*, vol. 10, pp. 1550-1556, 2010

## CONTACT

\*Hao Gu, tel: +31-53-4894236; h.gu@tnw.utwente.nl