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Numerical Study of the Controlled Droplet Breakup by Static Electric Fields inside a Microfluidic Flow-focusing Device

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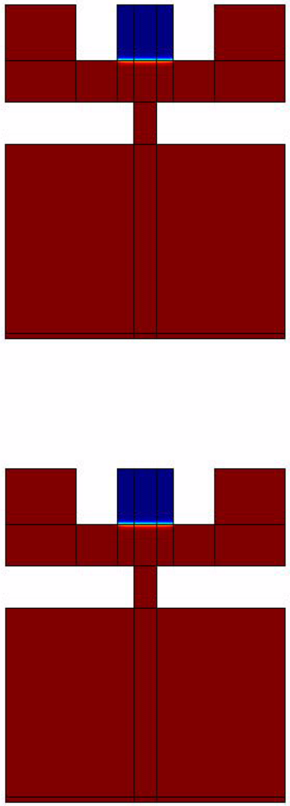
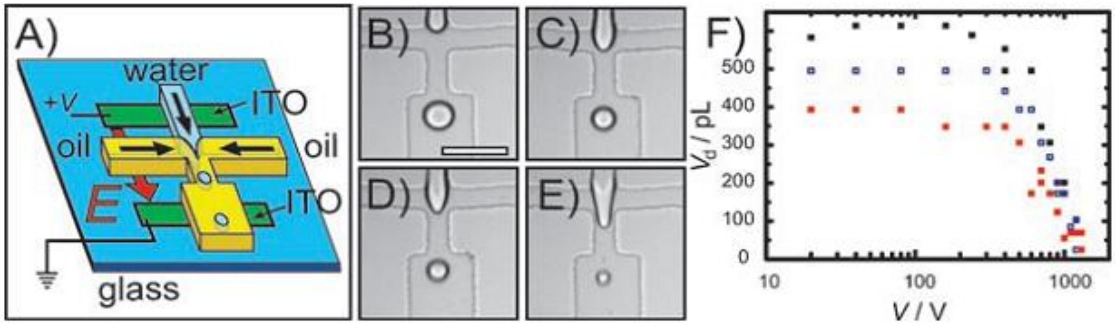
Outline

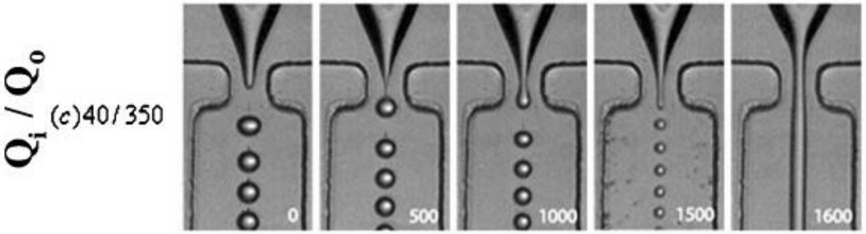
- Background: using electric fields in droplet-based microfluidics
- Numerical methods to model droplet breakup in electric field
- Droplet breakup controlled by applying electric field with low / medium / high strength
- Using electric field to control poly-dispersed droplet breakup
- Conclusion



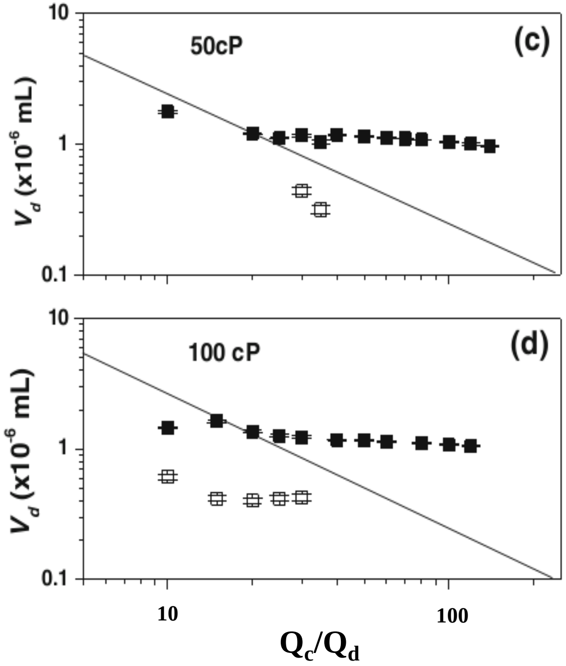
Droplet-based microfluidics coupled with electric field



Platform of using electric field to control droplet breakup (Link, et al. 2006)



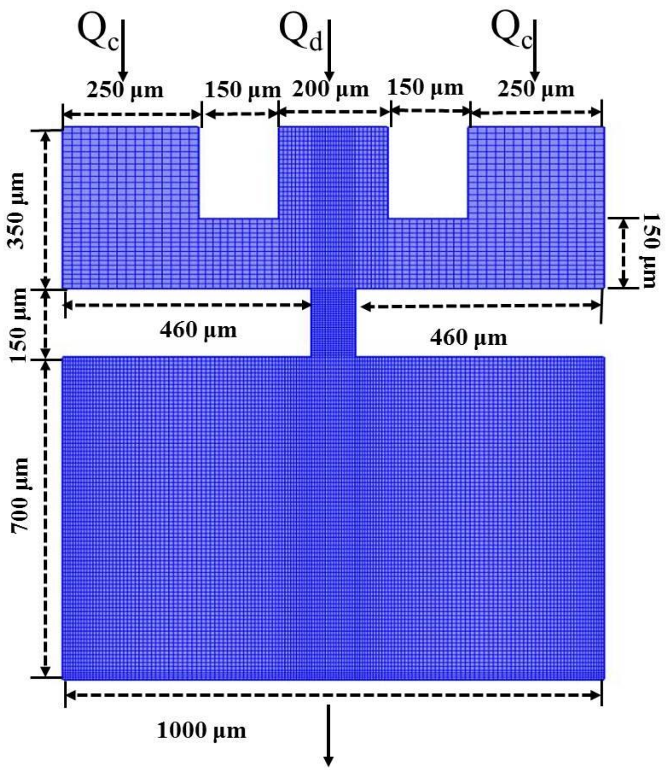
Droplet size as a function of electric field strength (Kim, et al., 2007)



Breakup of silicone oil droplet in water (Nie, et al. 2008)

- Droplet-based microfluidics utilize an immiscible fluid to segment the reagents into individual droplets. This technique has been widely used in DNA analysis, protein crystallization, nanomaterial synthesis and etc.
- Link proposed to use electric field as an additional approach to control droplet microfluidics.
- The electric field can effectively control the droplet sizes, and it can help to control the breakup of high viscous droplets.

Simulation setup



Geometry of the microfluidic flow-focusing device (MFFD).
Orifice size: 86 µm.
Typical grid resolution: 13,160 elements.

Operating conditions:

- Q_d =0.04 mL/h and Q_c/Q_d=10, 20, 50 and 100;
- Applied voltage: 12 V to 660 V.
- Dispersed phase: silicone oil (10, 20, 50cp and 100 cp)
- Continuous phase: water

Liquids	Water (continuous phase)	Silicone oil (Dispersed phase)
Density (kg/m^3)	1000	960
Viscosity (mPa*s)	1	10/20/50/100
Relative permittivity	78.5	2.8

Numerical methods

Electrostatic

$$\nabla \cdot (-\varepsilon \nabla V) = \rho_f \qquad (1)$$

$$\vec{E} = -\nabla V \qquad (2)$$

Perfect dielectric model:

Free charge density $\rho_f = 0$

- Due to the low conductivities of the both fluids, the effect of conductivity on droplet is neglected in this stage.
- The governing equations are solved by Comsol Multiphysics 4.3a.
- The simulations are performed on high performance computers (HPC) in Louisiana State University.
- The typical simulation time is 24 hours if one node (8 processors) is used.

Fluid Flow: Conservative Level-set Method

$$\frac{\partial \phi}{\partial t} + \vec{u} \cdot \nabla \phi = \gamma \nabla \cdot \left(\epsilon \nabla \phi - \phi(1-\phi) \frac{\nabla \phi}{|\nabla \phi|} \right) \qquad (3)$$

$$\hat{n} = \frac{\nabla \phi}{|\nabla \phi|} \qquad (4)$$

$$\kappa = -\nabla \cdot \hat{n}|_{\phi=0.5} \qquad (5)$$

$$\vec{F}_{sf} = \sigma \kappa \delta \hat{n} \qquad (6)$$

$$\delta = 6|\nabla \phi||\phi(1-\phi)| \qquad (7)$$

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{u}) = 0 \qquad (8)$$

$$\frac{\partial (\rho \vec{u})}{\partial t} + \nabla \cdot (\rho \vec{u}\vec{u}) = -\nabla P + \nabla \cdot \left[\mu \left(\nabla \vec{u} + \nabla \vec{u}^T \right) \right] + \vec{F}_{sf} + \vec{F}_{ef} \qquad (9)$$

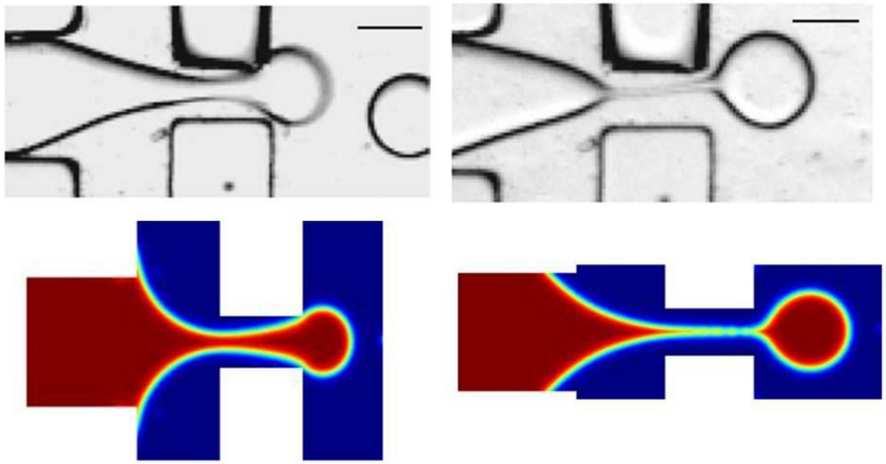
$$\rho = \rho_1 + (\rho_2 - \rho_1)\phi$$
$$\mu = \mu_1 + (\mu_2 - \mu_1)\phi$$
$$\varepsilon = \varepsilon_1 + (\varepsilon_2 - \varepsilon_1)\phi$$

$$\vec{F}_{ef} = \nabla \cdot T_{MW} = -\frac{1}{2}(\vec{E} \cdot \vec{E})\nabla \varepsilon$$

E. Olsson and G. Kreiss, *J. Comput. Phys.*, 2005, **210**, 225-246.

Validation of the numerical model

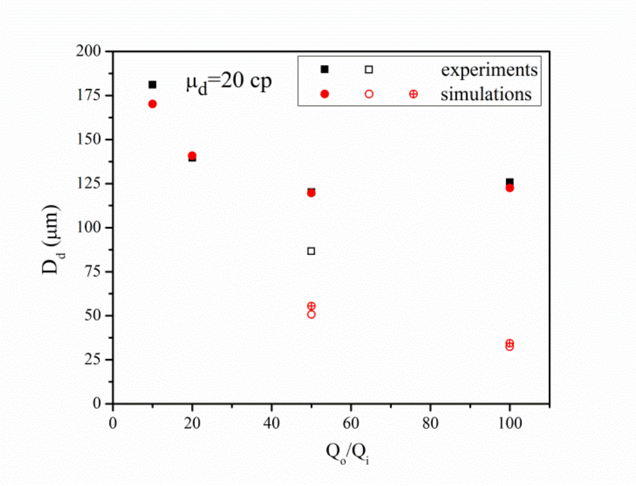
Snapshots from experiments and simulations

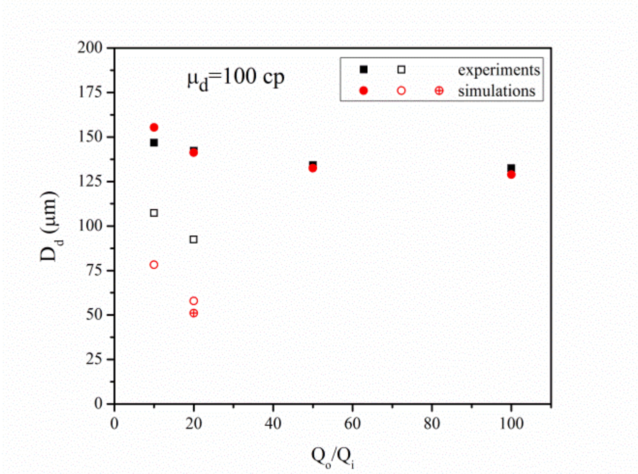


(a) μ_d = 20 cp

(b) μ_d = 100 cp

Comparison of Observed droplet sizes

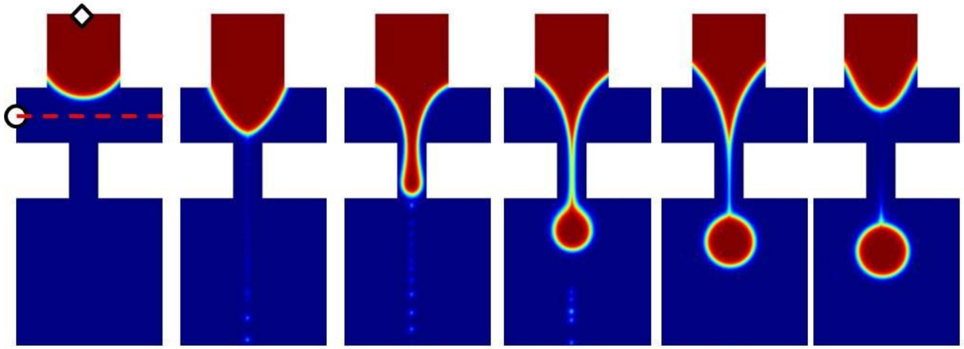




- Good agreement of the level-set method with the experimental observations by Nie et al., 2008.
- In this study, the exploration focuses on typical “dripping” regime.

Z. H. Nie, M. S. Seo, S. Q. Xu, P. C. Lewis, M. Mok, E. Kumacheva, G. M. Whitesides, P. Garstecki and H. A. Stone, *Microfluid. Nanofluid.*, 2008, **5**, 585-594.

Droplet formation process without electric field



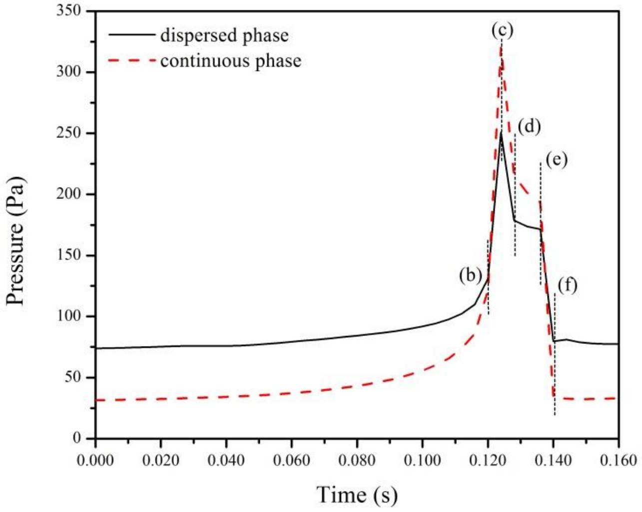
(a) t=0.000 s (b) t=0.120 s (c) t=0.124 s (d) t=0.128 s (e) t=0.136 s (f) t=0.140 s

Droplet formation process at μ_d=50 cp, Q_i =0.04 mL/h and Q_o/Q_i=50

(1) In dripping regime, the formation process undergoes:

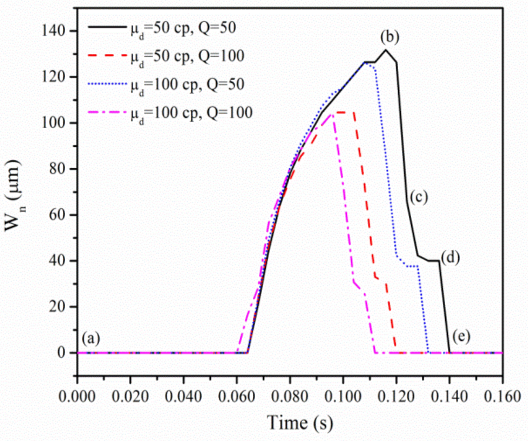
- Expanding ((a)~(b))
- Squeezing ((c)~(e))
- Pinching-off (f)

(2) Droplet size depends on squeezing time and flow rate of the dispersed phase.

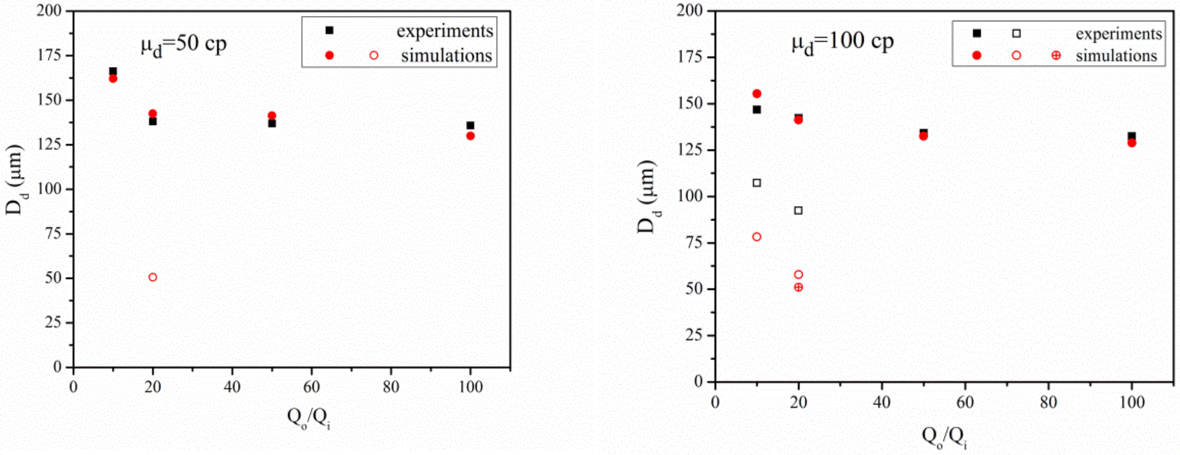


Upstream pressure evolution of the continuous phase and dispersed phase for the droplet breakup process at μ_d=50 cp, Q_d =0.04 mL/h and Q_o/Q_i=50

Droplet formation process without electric field



Evolutions of neck width at different viscosity and flow ratio



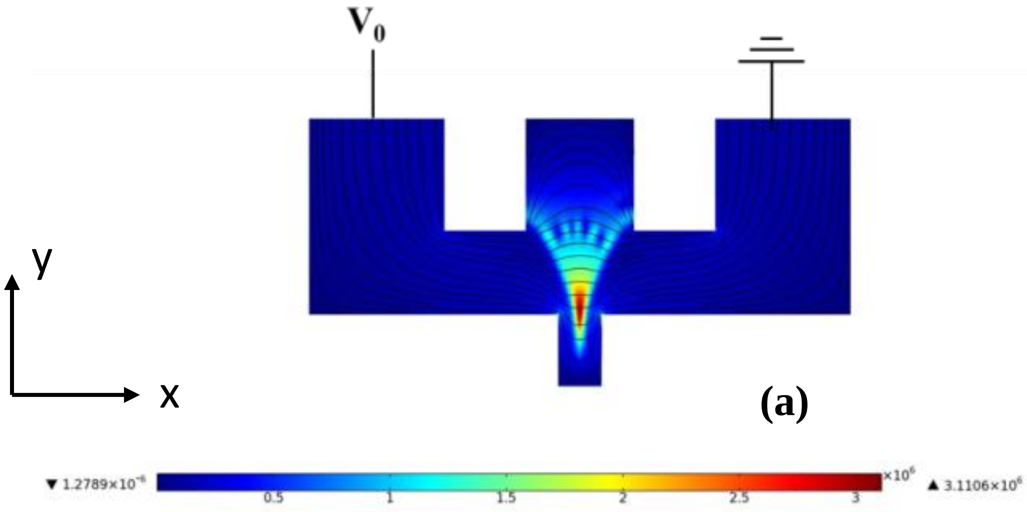
(a)

(b)

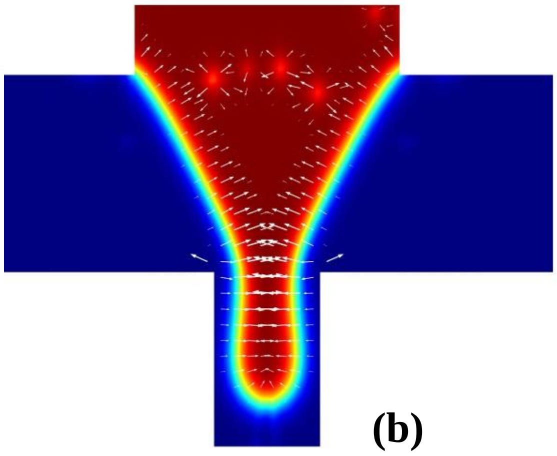
Droplet size as a function of flow ratio. (a) μ_d=50 cp (b) μ_d=100 cp

- The common approach to reduce the droplet size relies on increasing the flow ratio of continuous phase to dispersed phase (Q_o/Q_i). The higher flow ratio, the larger pressure difference exists between the two phases.
- The pressure difference cannot effective squeeze the neck when $\mu_d >> \mu_c$. The squeezing rate (indicated as the slope of the neck evolution) is marginally affected by increasing the flow ratio.
- Large droplets are observed for a wide range of flow ratio if $\mu_d >> \mu_c$.
- Need an effective external force to control droplet size.

Effect of electric field on droplet breakup



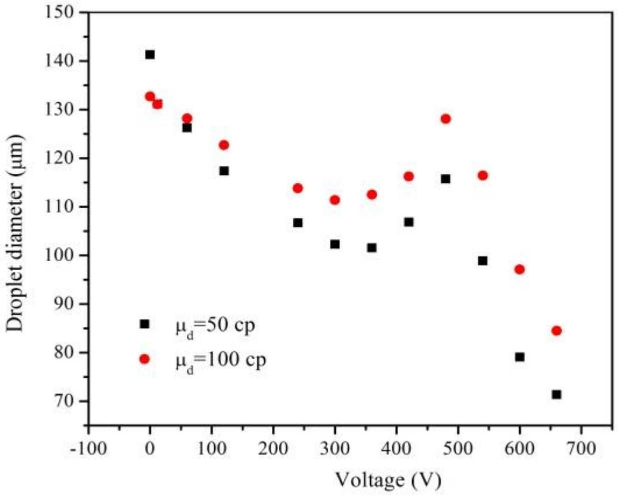
(a) Contour plot of the electric field strength during the droplet formation at μ_d=50 cp, Q_i =0.04 mL/h and Q_o/Q_i=50.

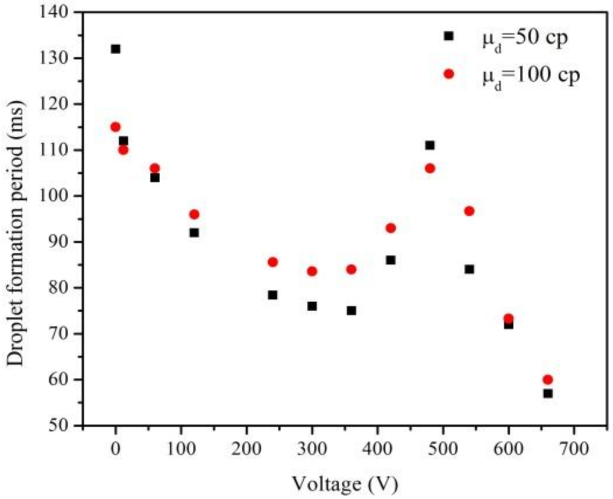


b) Phase surface plot of the focusing region at the same time as (a). The white arrows indicate the electric force

- High potential V_0 is added on the left inlet of the continuous phase. The right inlet is connected to ground ($V_0 = 0$ V).
- Polarization charges are developed along the interface due to the difference of electric properties. As $\varepsilon_2 < \varepsilon_1$, high electric field is formed inside the dispersed phase.
- The electric force has two effects: compression on x direction; retardation on y direction.
- The compression effect helps to squeeze the neck while the retardation effect repels the propagation of the dispersed phase.

Electric field strength on droplet size

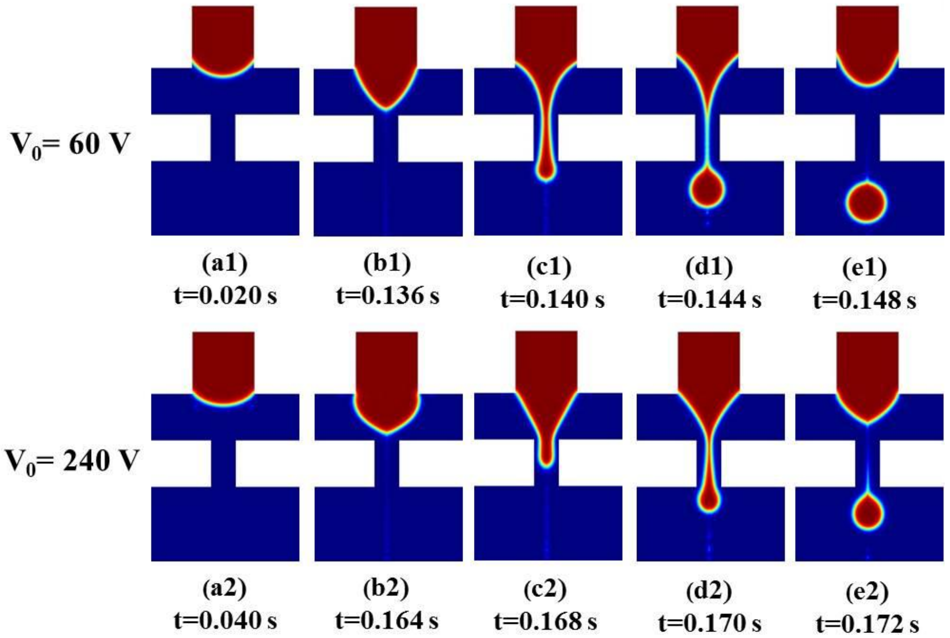


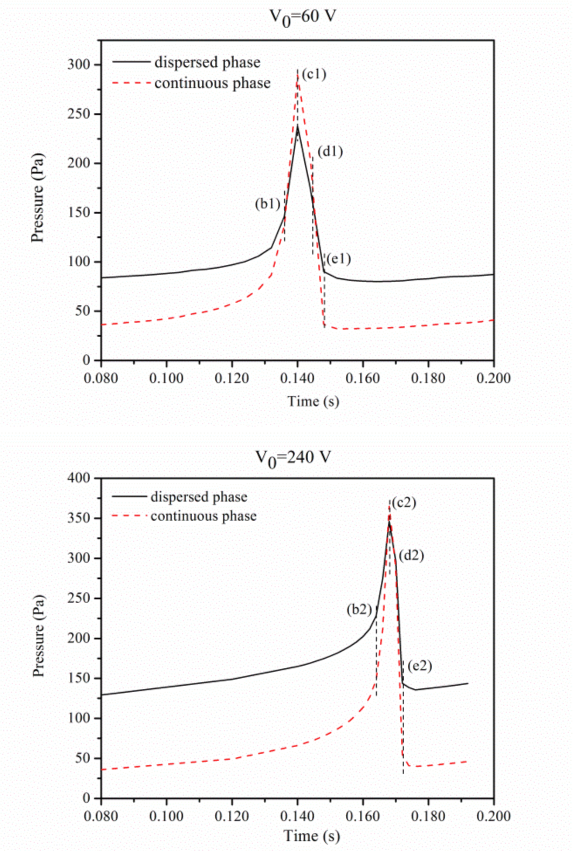


Three zones are observed in the observed voltages:
(1) V_0 = 12 V ~ 300 V: the droplet size and formation period decrease with the applied voltages.
(2) V_0 = 300 V ~ 500 V: the droplet size and period slightly increase with the applied voltages.
(3) V_0 = 500 V ~ 660 V: The droplet size and period decreases with the applied voltages.

This presentation focus on the low voltage zone.

Droplet formation in low electric field

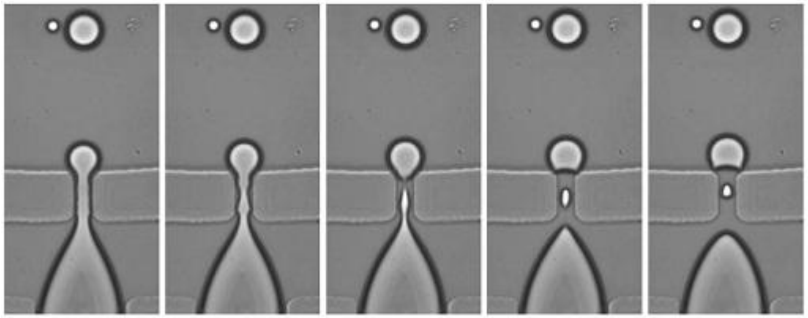




Upstream pressure evolution

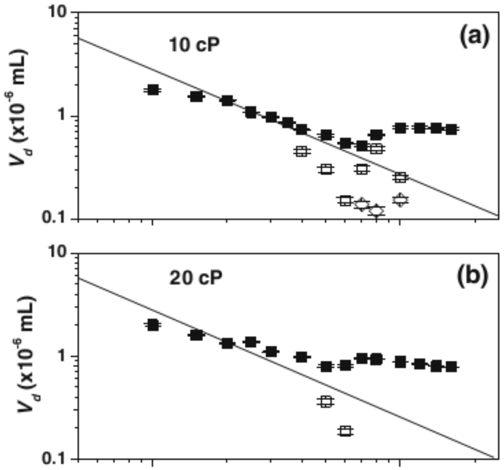
- The compression effect from the electric force can reduce the squeezing time ((c) to (e)) thus reduce the droplet size.
- The initial expanding time ((a) to (b)) increases due to the retardation effect of the electric force.
- To overcome the retardation force, the dispersed phase builds up a high pressure. The pressure difference does not play role in squeezing the neck when $V_0 > 240$ V.

Poly-dispersed droplet breakup in MFFD

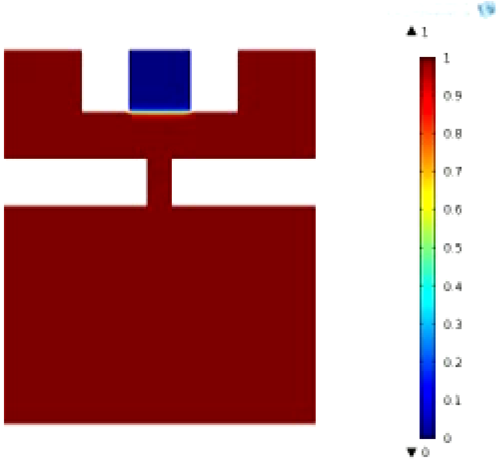


Experimental observation of poly-dispersed droplet breakup

(S. L. Anna, et al., *Applied Physics Letters*, 2003, **82**, 364-366)

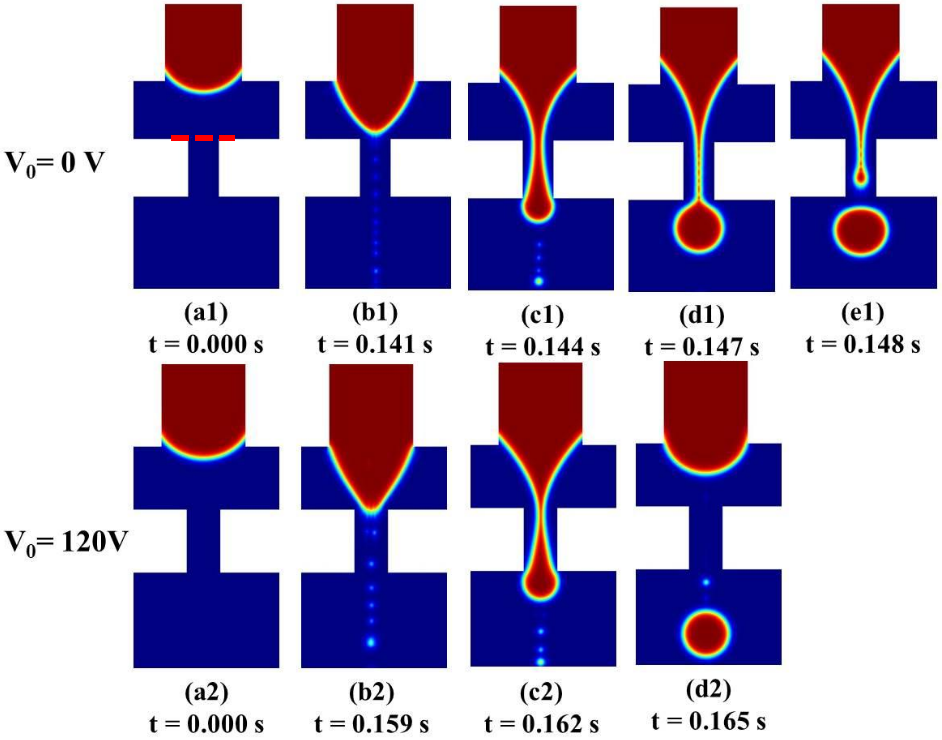


Secondary droplet sizes (indicated by empty squares)
(Z. H. Nie, et al., *Microfluid. Nanofluid.*, 2008, **5**, 585-594.)



- Poly-dispersed droplet breakup are commonly observed in dripping regime, resulting in droplets of broad size distributions.
- After the primary droplet is pinched off, the neck continues to grow in the orifice and breaks into secondary (satellite) droplets.
- Depending on operating conditions, one or multiple secondary droplets can be generated in the orifice.
- The poly-dispersed droplet breakup in dripping regime is governed by capillary instability. The capillary instability needs time to develop. (H. A. Stone and L. G. Leal, *J. Fluid Mech.*, 1989, **198**, 399-427.)

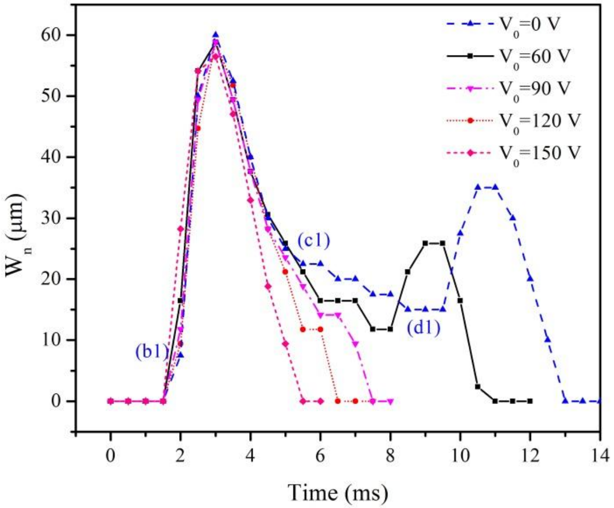
Controlling poly-dispersed breakup by electric field



(a1) t = 0.000 s
(b1) t = 0.141 s
(c1) t = 0.144 s
(d1) t = 0.147 s
(e1) t = 0.148 s

(a2) t = 0.000 s
(b2) t = 0.159 s
(c2) t = 0.162 s
(d2) t = 0.165 s

Droplet formation process at μ_d=20 cp, Q_i =0.04 mL/h and Q_o/Q_i=50



Neck evolution as a function of applied voltage

- The electric force reduces the squeezing time thus suppresses the growth of capillary instability.
- If the applied voltage exceeds 90 V, the poly-dispersed breakup is avoided.

Conclusion

- We have studied viscous droplet breakup inside a MFFD controlled by electric fields. We use conservative level-set method coupled with electrostatic model in Comsol 4.3a to study the droplet breakup process.

- If the viscosity of the dispersed phase is much larger than the continuous phase, the traditional way of controlling droplet size by varying flow ratio is not effective.

- By applying external electric field, a electric force is exerted on the fluid interface that helps to squeeze the dispersed phase. The droplet size can be significantly reduced especially for those situations that the dispersed phase has large viscosity.

- By reducing the droplet formation time, the electric force can suppress the capillary instability; therefore, it can avoid the poly-dispersed droplet breakup.

Acknowledgement

- Guidance from Prof. Nandakumar and Dr. Mranal Jain.
- Help from our group members.
- HPC facility of LSU is well acknowledged.

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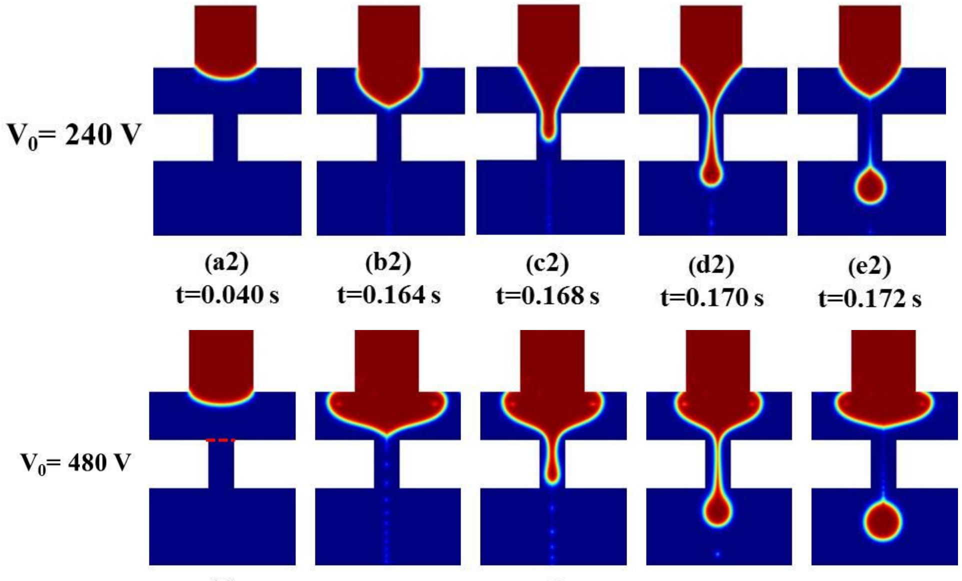
LSU HPC

LONI: Louisiana Optical Network Initiative

Thanks for your attention!

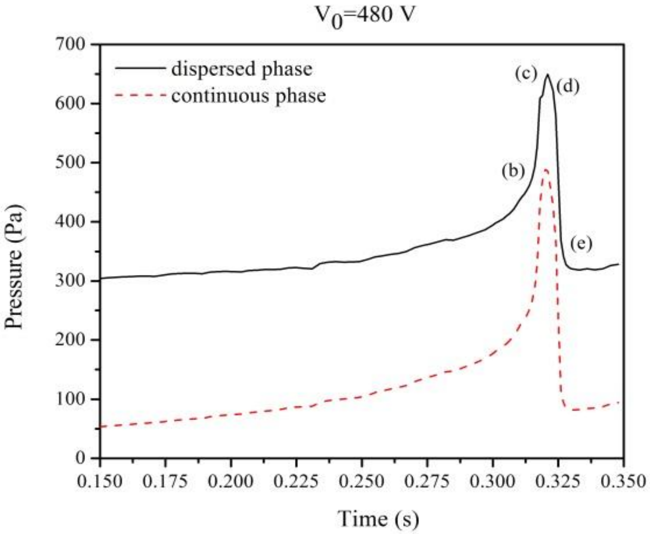
Questions ?

Droplet formation in medium electric field



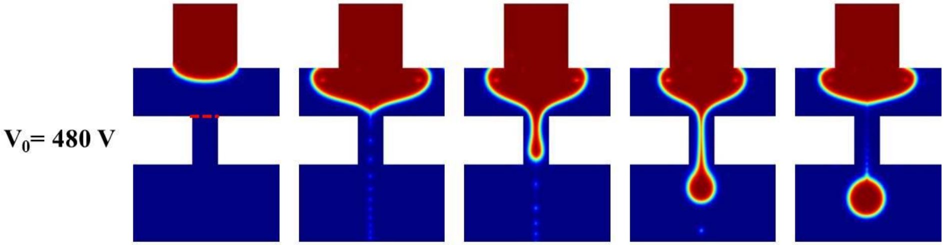
(a2) t=0.040 s (b2) t=0.164 s (c2) t=0.168 s (d2) t=0.170 s (e2) t=0.172 s

(a) t=0.050 s (b) t=0.316 s (c) t=0.320 s (d) t=0.323 s (e) t=0.326 s

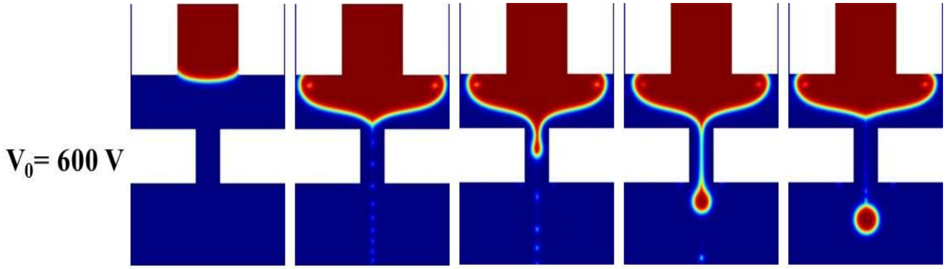


- Further increase the applied voltage causes significant increase of pressure in dispersed phase.
- The dispersed phase has a broad deformation during the formation process.
- The high pressure in dispersed phase stabilizes the interface which increases the squeezing time.
- The droplet size increases with the applied voltage in this region.

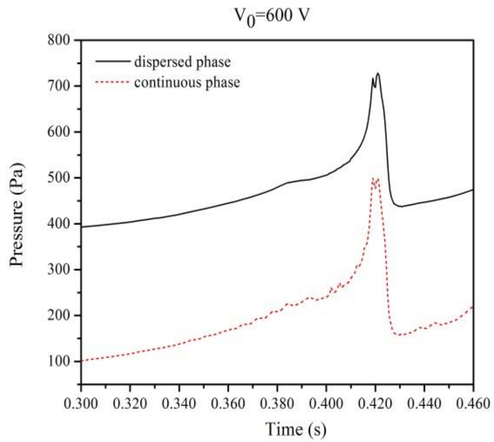
Droplet formation in high electric field

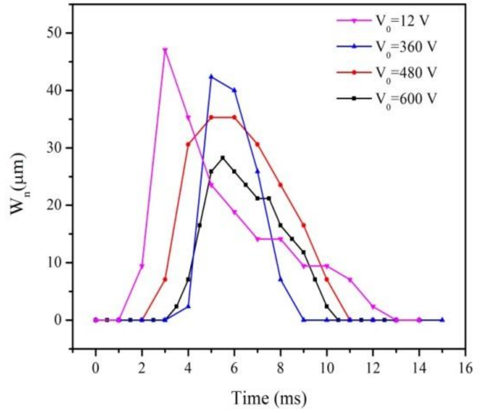


(a) t=0.050 s (b) t=0.316 s (c) t=0.320 s (d) t=0.323 s (e) t=0.326 s



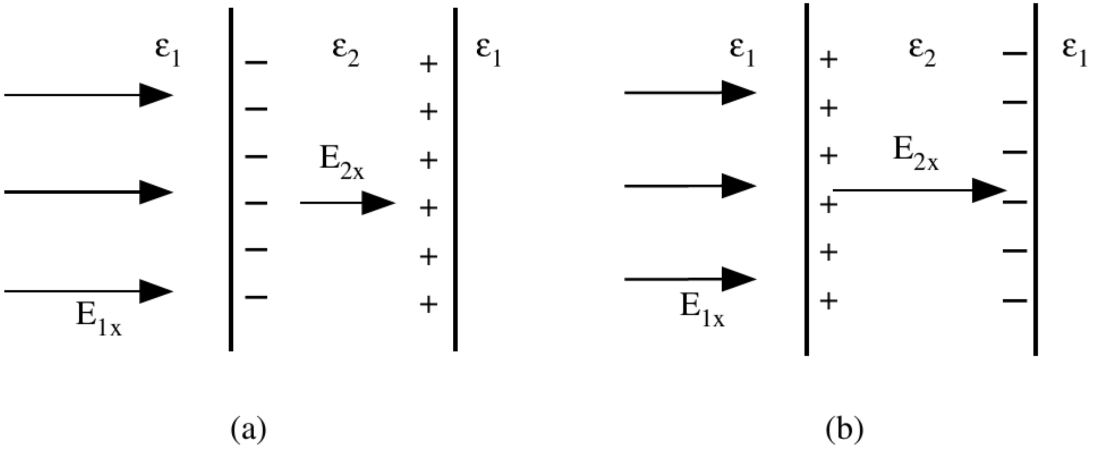
(a) t=0.020 s (b) t=0.414 s (c) t=0.420 s (d) t=0.423 s (e) t=0.426 s





- In high electric field regime, the droplet size decreases with the applied electric field.
- The maximum neck width in the orifice entrance is remarkably reduced due to the high pressure of the continuous phase.
- The neck width quickly reaches the critical value that pinch-off occurs.

Overview of the effect of electric field



- The polarization charge density and electric field strength depend on the sign of $(\varepsilon_2 - \varepsilon_1)$.
- If $\varepsilon_2 - \varepsilon_1 > 0$, negative charge on left hand side surface, $E_{2x} < E_{1x}$
- If $\varepsilon_2 - \varepsilon_1 < 0$, negative charge on right hand side surface, $E_{2x} > E_{1x}$
- If $\varepsilon_2 - \varepsilon_1 = 0$, $E_{2x} = E_{1x}$

In this work:

- Continuous phase: water

$\varepsilon_{r1} = 78.5$, K= 5.5 μS/m

- Dispersed phase: silicone oil

$\varepsilon_{r2} = 2.8$, K= 10^{-7} μS/m

J. H. Masliyah and S. Bhattacharjee, *Electrokinetic and colloid transport phenomena*, Wiley-Interscience, Hoboken, N.J., 2006.