Control the poly-dispersed droplet breakup mode inside a microfluidic flow-focusing device by external electric field

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Outline

Objective of this study:

- Capture the droplet breakup modes by level-Set method;
- Test the capability of using electric field to control the droplet breakup mode.

Introduction:

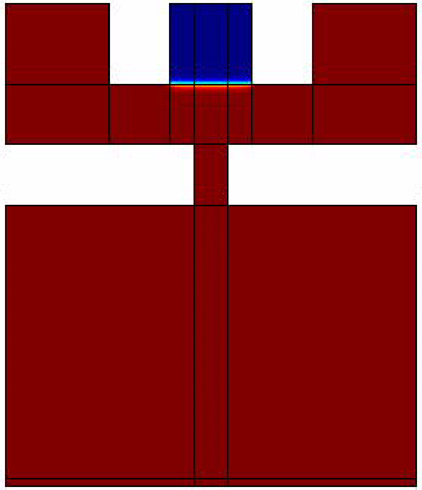
(1) droplet-based microfluidics;
(2) droplet generator; breakup regimes and breakup modes;
(3) control the droplet breakup by electric fields

Numerical methods

(1) Conservative level-set & Electrostatics;
(2) Simulation setup

Results from simulations

Questions and Discussions

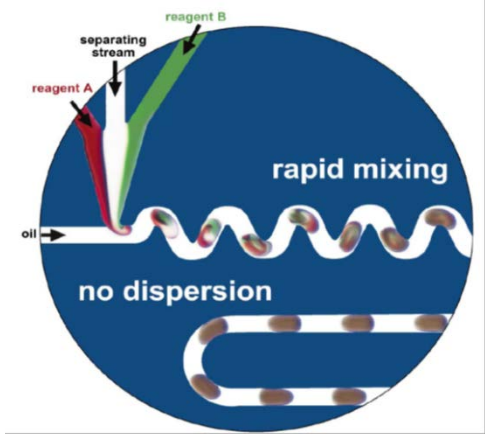


Introduction to droplet-based microfluidics

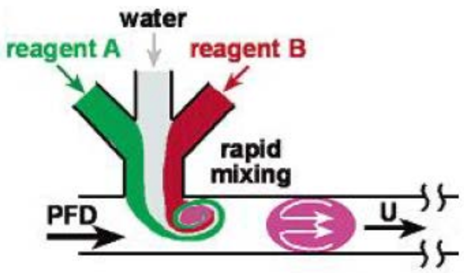
- The droplet-based microfluidics overcomes the drawbacks of the conventional single-phase microfluidics.
- **Approach**: introduce an immiscible carrier fluid (continuous phase) to encapsulate the reagents (secondary phase) inside discrete droplets / slugs.
- **Advantages**: Rapid mixing; no dispersion; minimized surface fouling.
- **Applications**:
  - (1) Nano-particle (NP) synthesis;
  - (2) In-situ kinetic measurement;
  - (3) Various other applications in chemistry and biology.
- **Challenges:**

(1) Control the droplet breakup to obtain droplets of desired sizes and distributions.

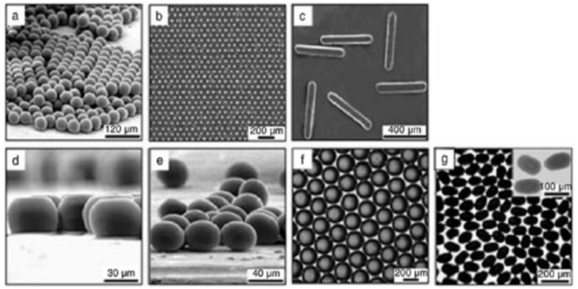
(2) Obtain “mono-dispersed” droplet sizes.



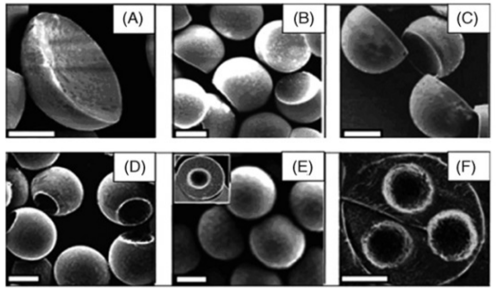
(Song, 2006)



(Tice, 2003)



NPs of various shapes (Nie, 2005)



core – shell structures (Xu, 2005)

References:

1. H. Song, D. L. Chen, and R. F. Ismagilov, Angew. Chem.-Int. Edit. **45** (44), 7336 (2006).
2. J. D. Tice, H. Song, A. D. Lyon, and R. F. Ismagilov, Langmuir **19** (22), 9127 (2003).
3. Z. H. Nie, S. Q. Xu, M. Seo, P. C. Lewis, and E. Kumacheva, J. Am. Chem. Soc. **127** (22), 8058 (2005).
4. S. Q. Xu, Z. H. Nie, M. Seo, P. Lewis, E. Kumacheva, H. A. Stone, P. Garstecki, D. B. Weibel, I. Gitlin, and G. M. Whitesides, Angew. Chem.-Int. Edit. **44** (5), 724 (2005).

Droplet generations in microfluidics

- **Passive droplet / slug generation:**

(1) Utilize device geometry and fluid flow;
(2) Three types of generators:
I. Co-flow device;
II. Cross-flow device (T-junction);
III. Hydrodynamic flow-focusing device.

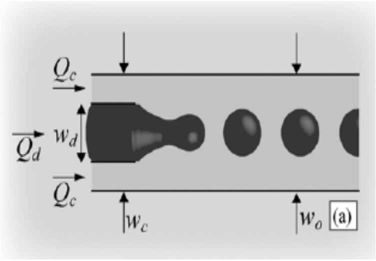
- **Droplet breakup dynamics:**

(1) Three forces:
Pressure force, **viscous shear** and **surface tension force**;
(2) Breakup regimes: **Squeezing, Dripping, Jetting**;
(3) Critical parameters:
Capillary number ($Ca = \mu_c U_c / \sigma$)
Flow ratio ($Q = Q_c / Q_d$)
Viscosity ratio ($\lambda = \mu_d / \mu_c$)

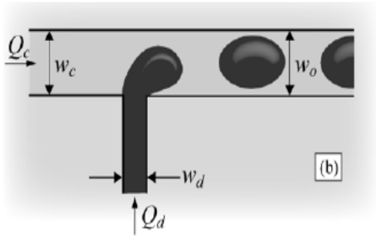
References:
1. G. F. Christopher and S. L. Anna, J. Phys. D-Appl. Phys. **40** (19), R319 (2007).
2. M. De Menech, P. Garstecki, F. Jousse, and H. A. Stone, J. Fluid Mech. **595**, 141 (2008).

Passive droplet generators (Christopher, 2007)

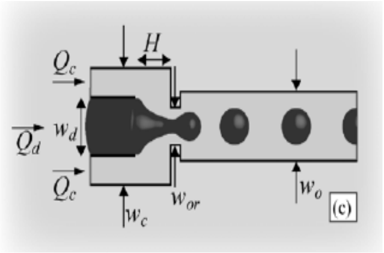
Co–flow device



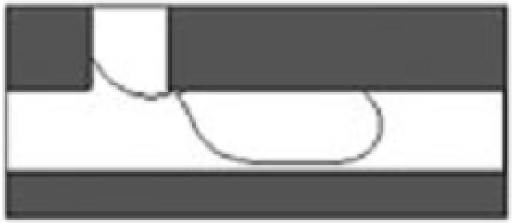
Cross – flow device (T-junction)



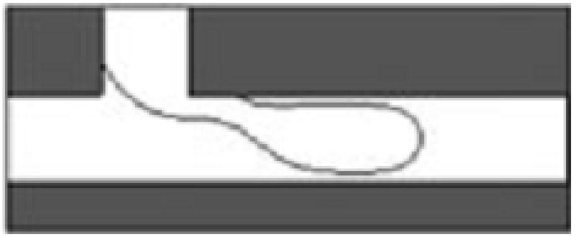
Hydrodynamic flow–focusing device



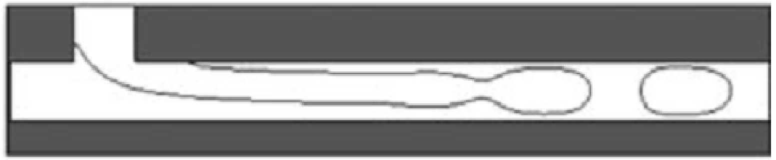
Droplet breakup regimes (De Menech, 2008)



Squeezing (Ca <0.01)



Dripping (0.01<Ca <0.04)



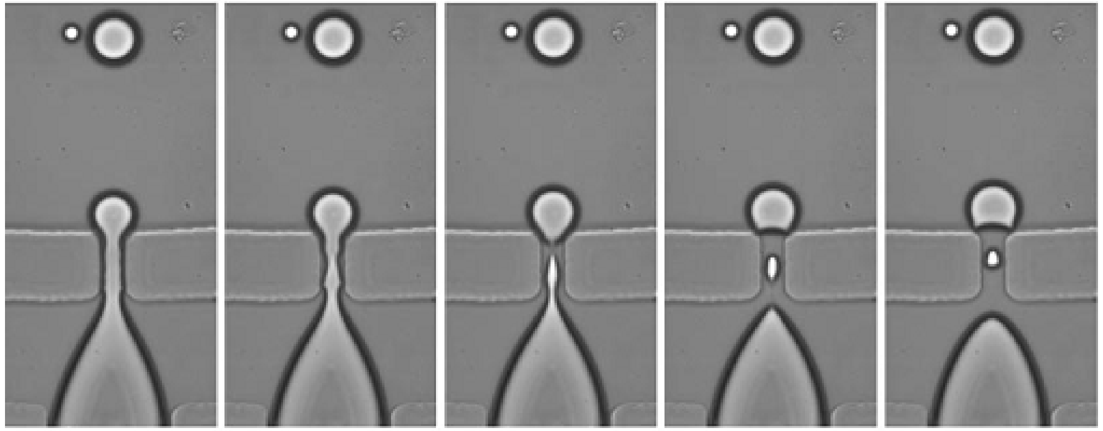
jetting (Ca > 0.04)

Droplet breakup modes

- Mono-dispersed breakup: uniform droplets, size variation < 2%;
- Poly-dispersed breakup: droplets of broad size distributions
- Typical poly-dispersed breakup modes:

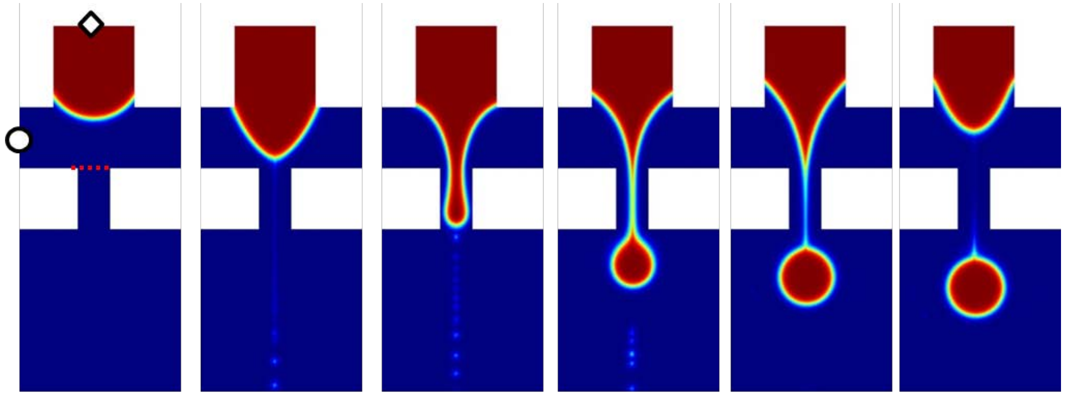
I. Single secondary (satellite) droplet after the primary droplet;
II. Multiple secondary droplet after the primary droplet.

Poly-dispersed breakup mode seen in experiments (Anna, 2003)



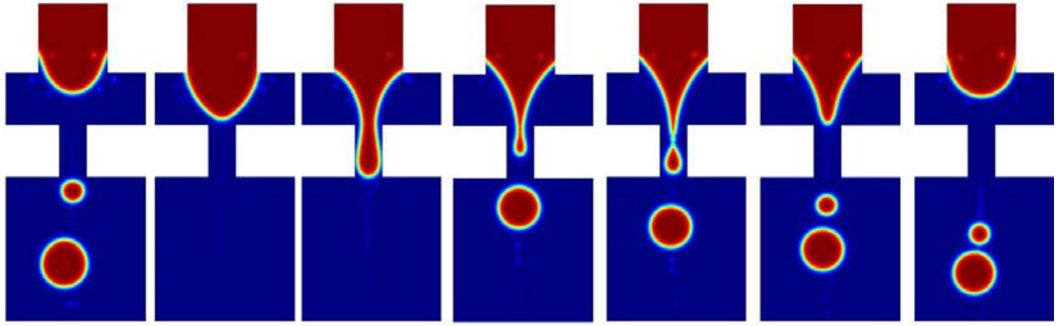
Reference: S. L. Anna, N. Bontoux, and H. A. Stone, Applied Physics Letters **82** (3), 364 (2003).

Mono-dispersed breakup mode



(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

A typical poly-dispersed breakup mode



(a) t = 0.000 s (b) t = 0.113 s (c) t = 0.119 s (d) t = 0.127 s (e) t = 0.130 s (f) t = 0.134 s (g) t = 0.139 s

Governing mechanisms of poly-dispersed breakup mode

- **Conclusions from literatures and previous simulations:**

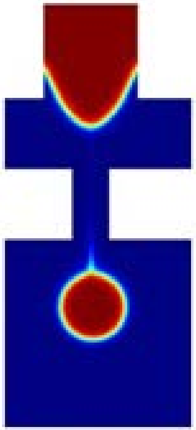
I. Poly-dispersed breakup mode is governed by the **non-linear dynamics.**

II. **Initiation**: imbalance of the three forces;

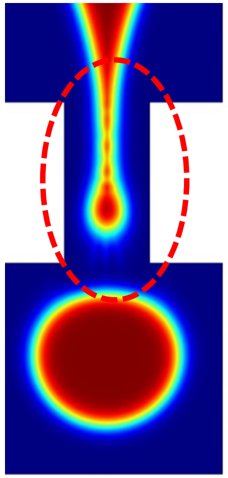
III. Two mechanisms: **end-pinching** & **capillary instability**;

IV. Comsol can capture these two modes and the wave shape.

V. Capillary instability needs time to develop.



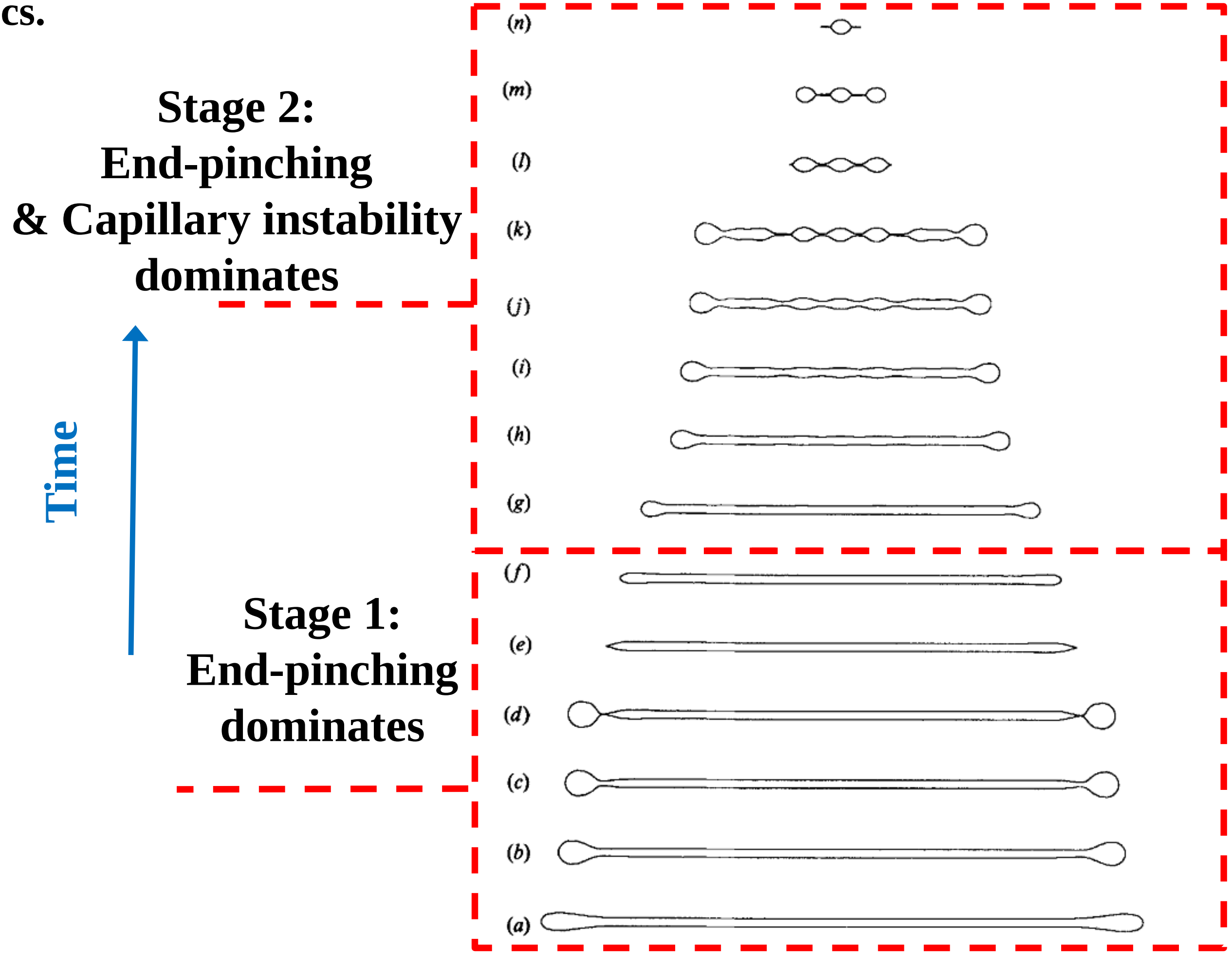
Mono-dispersed:
neck retract



Wave shape in the neck

Poly-dispersed:
neck does not retract

End-pinching & Capillary instabilities (Stone, 1989)



Stage 2:
End-pinching
& Capillary instability
dominates

Time

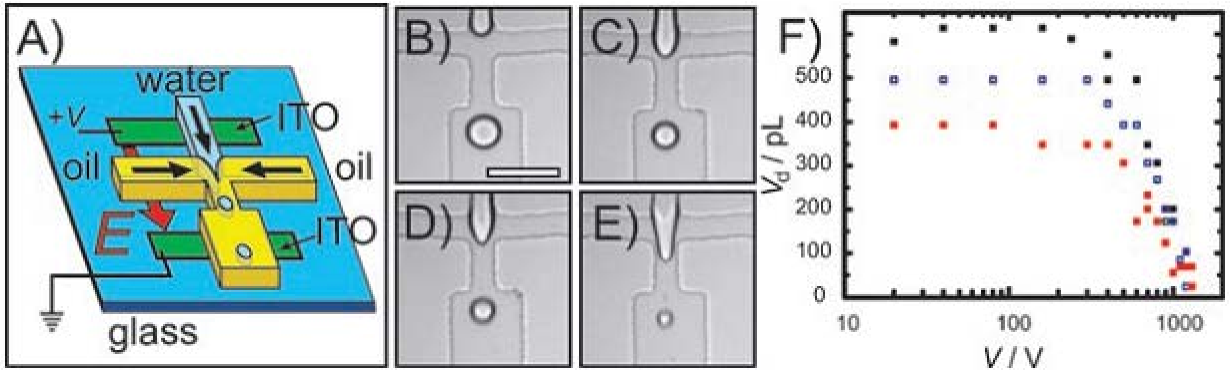
Stage 1:
End-pinching
dominates

Reference:

1. Y. C. Tan, V. Cristini and A. P. Lee, *Sens. Actuator B-Chem.*, 2006, **114**, 350-356.
2. H. A. Stone and L. G. Leal, *J. Fluid Mech.*, 1989, **198**, 399-427.

Apply external electric field to control droplet breakup

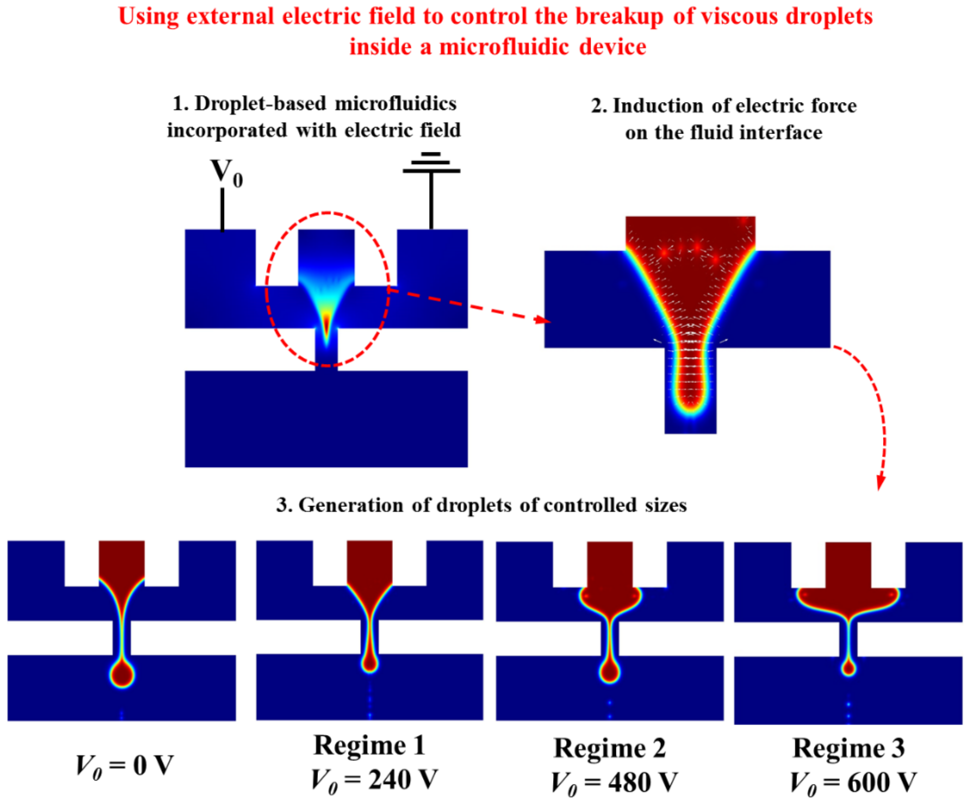
Apply electrical field to control droplet sizes (Link, 2006)



- Electric field has been coupled with conventional droplet-based microfluidics to enhance the droplet manipulations (breakup, coalescence, sorting and etc) .
- The different electric properties (permittivity, conductivity) induce electric charges on the fluid interface.
- The interactions between electric field and the induced charges generate electric forces (Maxwell stress) on the fluid interface.
- The electric force has shown the ability to control the droplet sizes.

Hypothesis: The electric field can control the droplet breakup mode in droplet-based microfluidics.

Use electrical field to control the breakup of viscous droplets (Li, 2015)



Reference:

1. D. R. Link, E. Grasland-Mongrain, A. Duri, F. Sarrazin, Z. D. Cheng, G. Cristobal, M. Marquez and D. A. Weitz, *Angew. Chem.-Int. Edit.*, 2006, **45**, 2556-2560.
2. Y. Li, M. Jain, Y. Ma and K. Nandakumar, *Soft Matter*, 2015, DOI: 10.1039/C5SM00252D.

Numerical methods

Electrostatics: Poisson equation

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

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

Fluid flow: Conservative Level-Set Method (LSM)

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

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

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

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

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

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

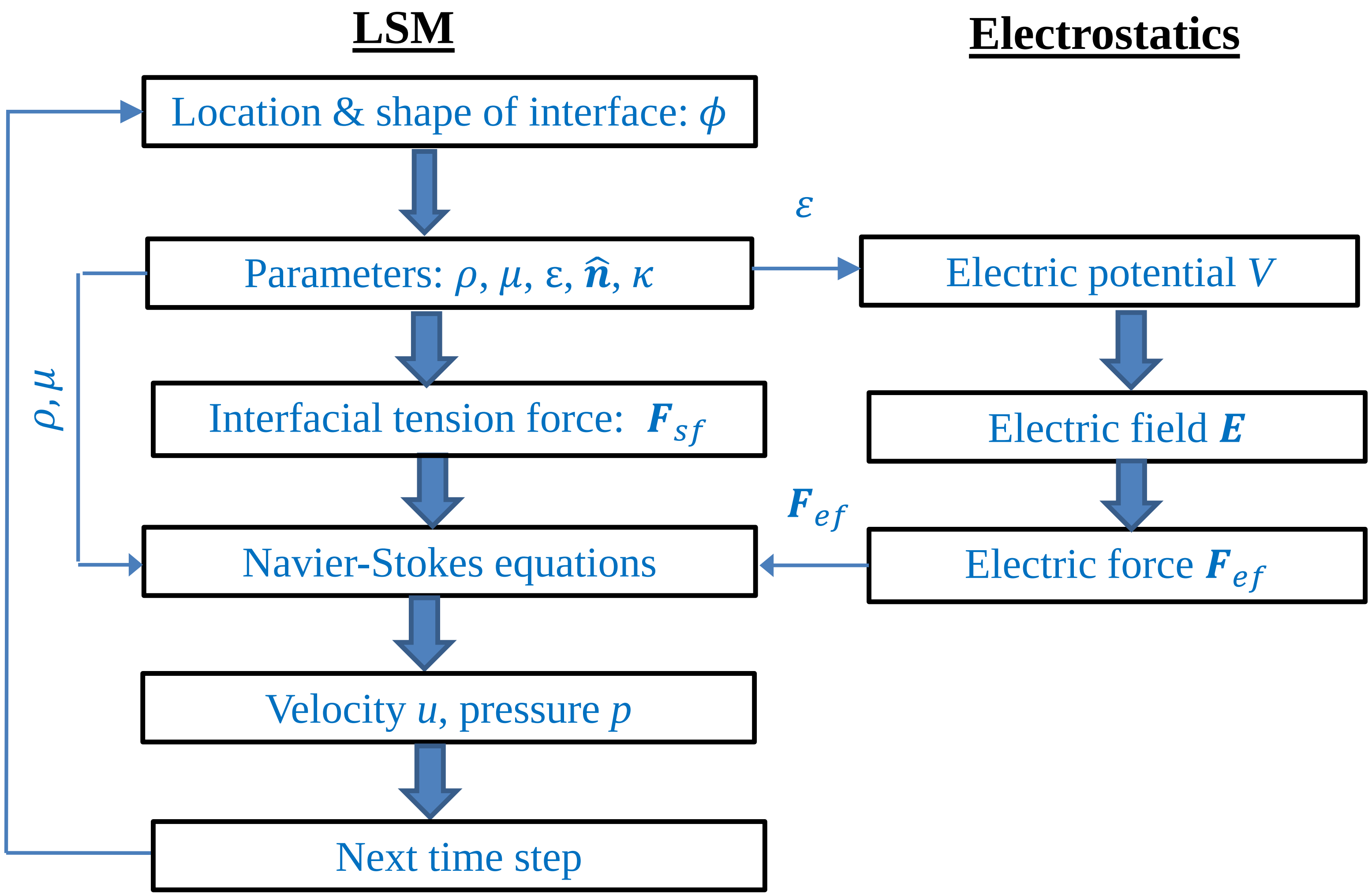
$$\frac{\partial(\rho\boldsymbol{u})}{\partial t}+\nabla\cdot(\rho\boldsymbol{u}\boldsymbol{u})=-\nabla p+\nabla\cdot[\mu(\nabla\boldsymbol{u}+\nabla\boldsymbol{u}^T)]+\boldsymbol{F}_{sf}+\boldsymbol{F}_{ef} \qquad (9)$$

$$\rho=\rho_1+(\rho_2-\rho_1)\phi$$

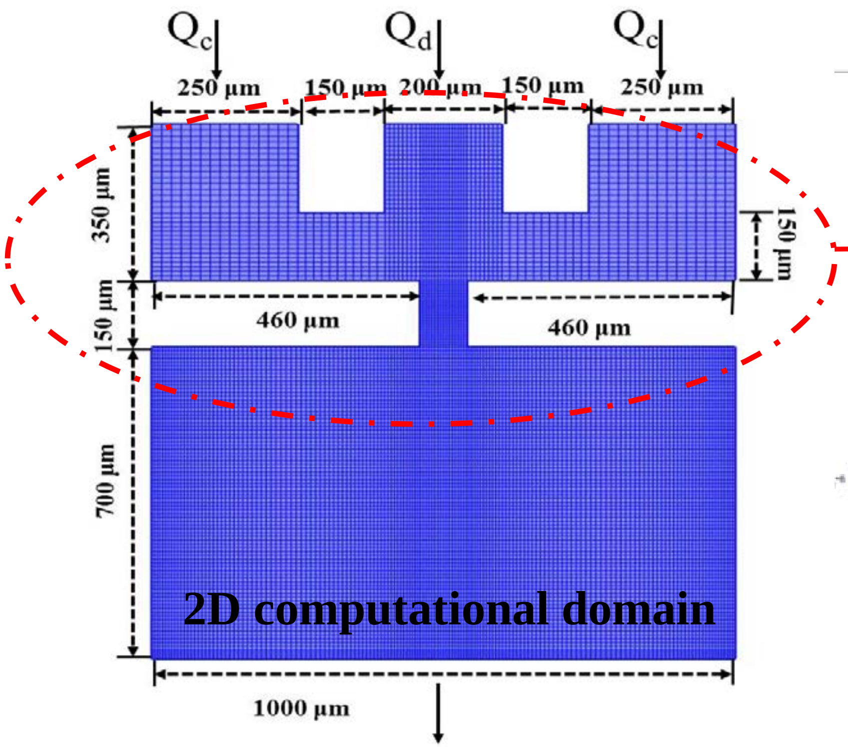
$$\mu=\mu_1+(\mu_2-\mu_1)\phi$$

$$\varepsilon=\varepsilon_1+(\varepsilon_2-\varepsilon_1)\phi \qquad (10)$$

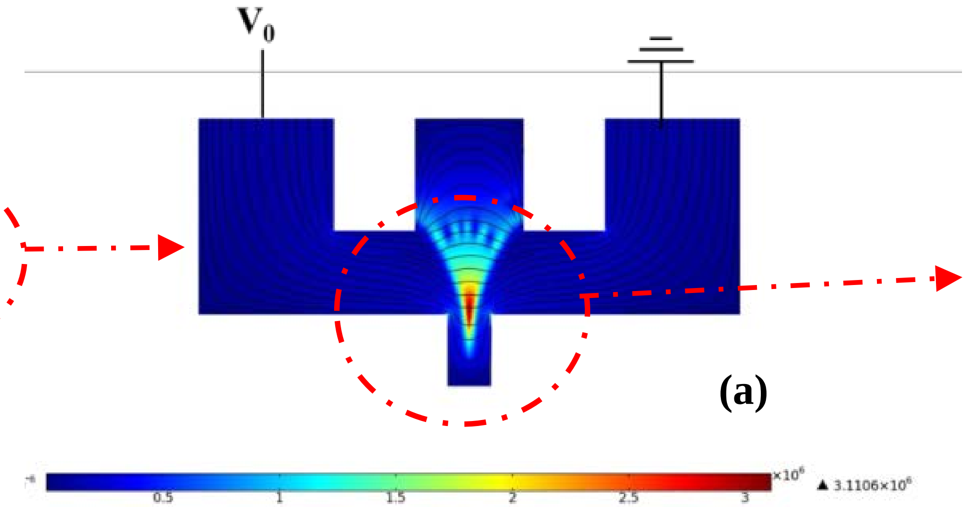
$$\boldsymbol{F}_{ef}=\nabla\cdot T_{MW}=-\frac{1}{2}(\boldsymbol{E}\cdot\boldsymbol{E})\nabla\varepsilon \qquad (11)$$

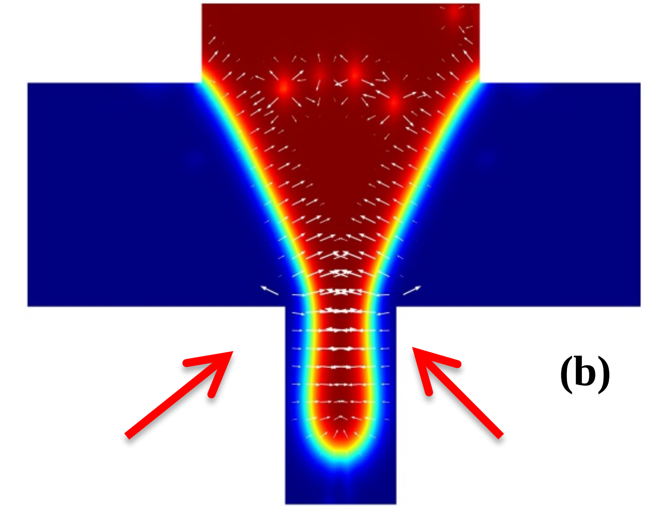


Simulation setup



2D computational domain

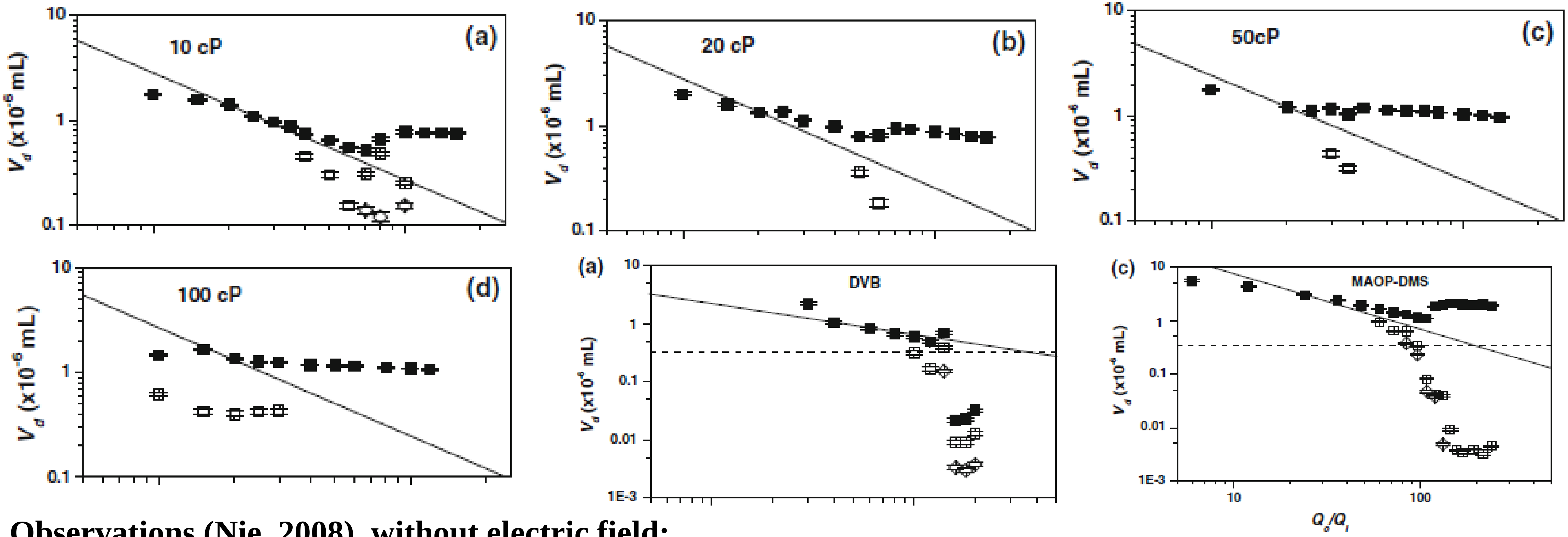




	Continuous phase	Dispersed phase
Density (kg/m^3)	1000	960
Viscosity (mPa*s)	1	10/20/50/100
Relative permittivity	78.5	2.8
Qc/Qd	10~100 (Qd = 0.04 mL/h)	
V0	0 ~ 150 V	

- Field configuration: high potential V_0 left, ground right.
- Strong field in the dispersed phase ($\varepsilon_2 < \varepsilon_1$).
- Electric force is induced on the fluid interface.
- Electric force “squeezes” the fluid neck.

Effect of flow ratio on poly-dispersed breakup mode (“poly-dispersed breakup window”)

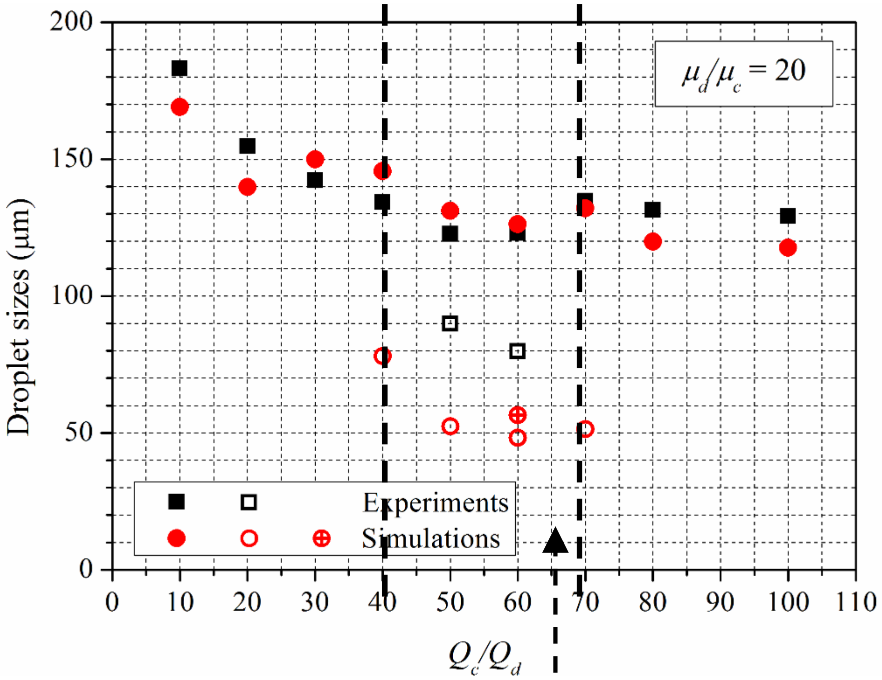


Observations (Nie, 2008), without electric field:

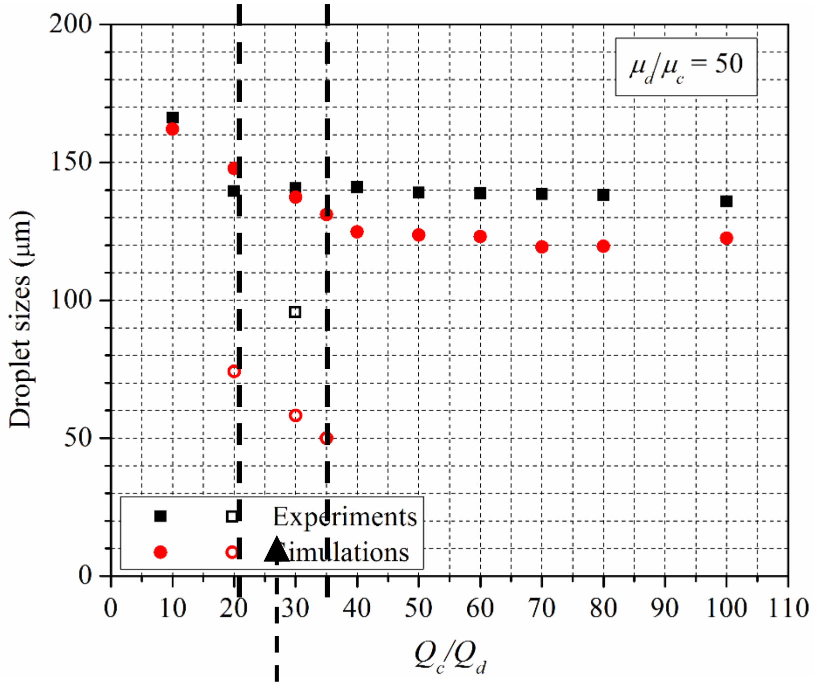
- Poly-dispersed breakup mode occurs in certain ranges of flow ratios (“poly-dispersed breakup window”).
- When the flow ratio increases beyond critical values, the poly-dispersed mode shifts to mono-dispersed mode.
- The locations and size of “windows” are functions of viscosity ratio ($\lambda = \mu_d/\mu_c$).
- The span of “window” is large when the viscosity ratio is small.

Reference: Z. Nie; *Microfluid. Nanofluid.*, 2008, **5**, 585-594.

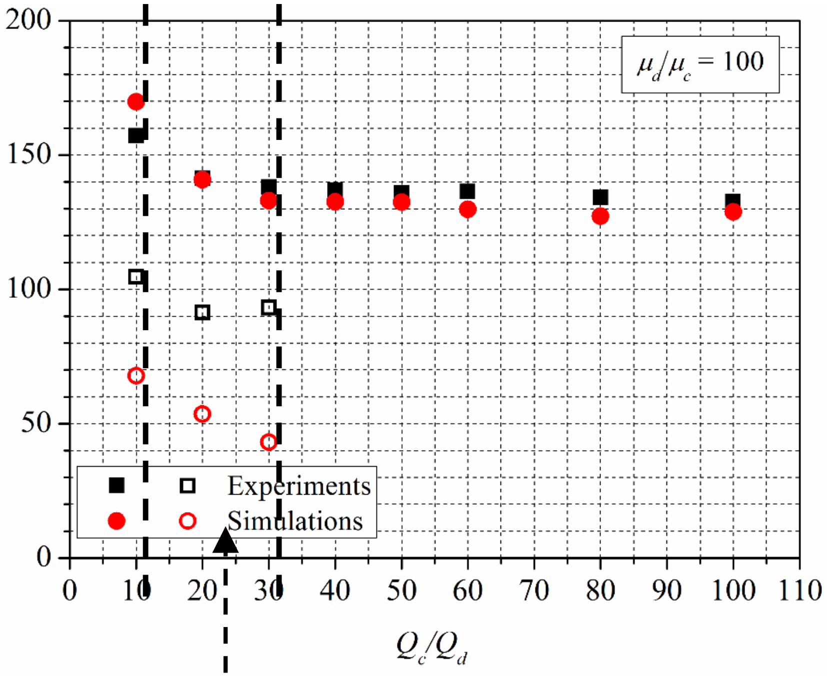
Simulation results: droplet breakup without electric field



Poly-dispersed breakup "window"



Poly-dispersed breakup "window"



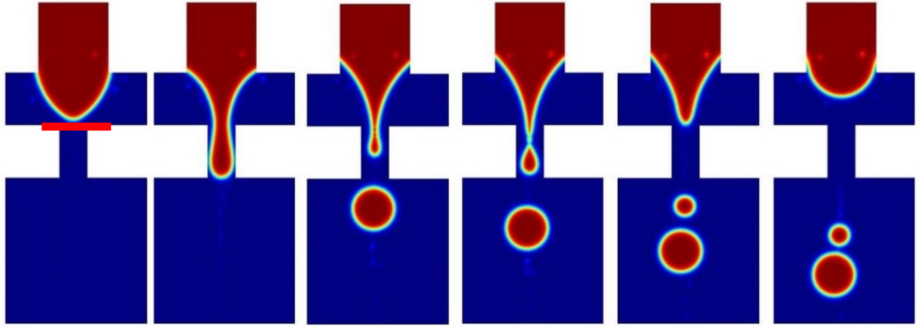
Poly-dispersed breakup "window"

Observations from simulations:

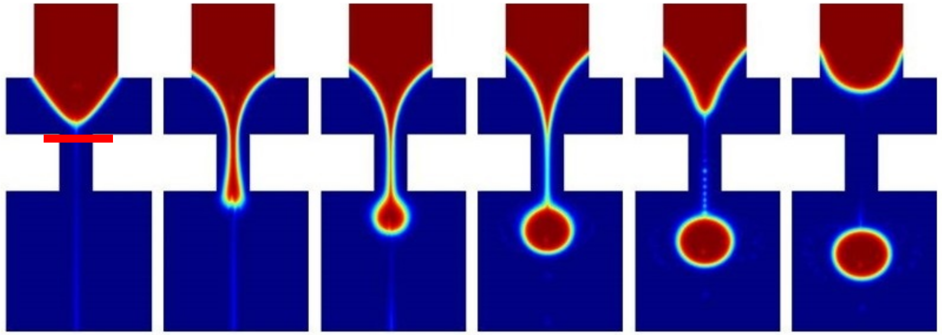
- The numerical model (LSM) can capture the "poly-dispersed breakup window" qualitatively.
- Good agreement of primary droplet sizes with experiments (Nie, 2008).

Effect of flow ratio on breakup modes

Poly-dispersed breakup mode



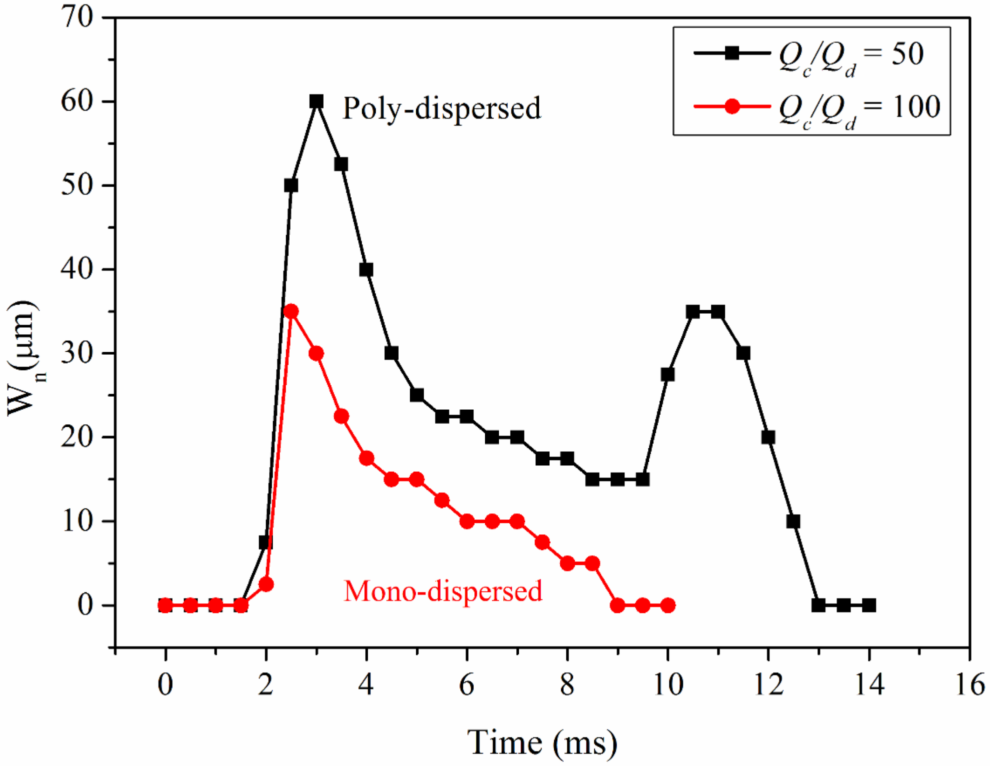
Mono-dispersed breakup mode



Increase the flow ratio

Measure the neck width in the orifice entrance

Neck width (W_n) as a function of time

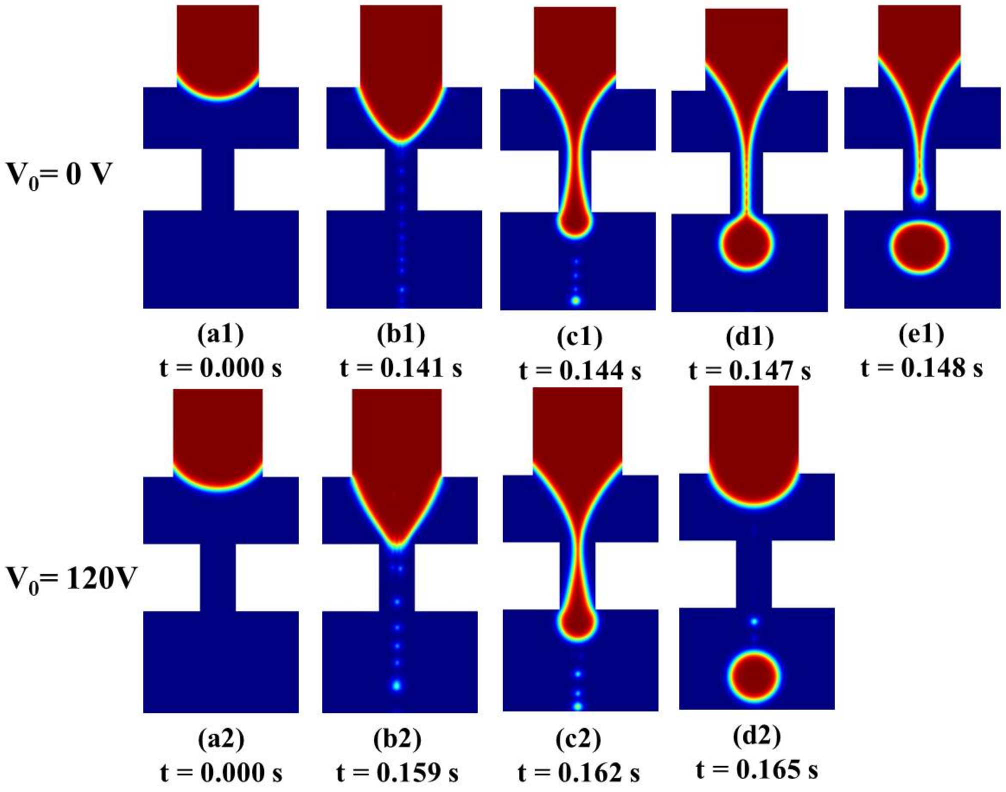


Observations:

- Increase the flow ratio from 50 to 100 → total droplet breakup time is reduced from 13 ms to ~ 8.5 ms.
- Reduce droplet breakup time → suppress the development of capillary instability → mono-dispersed breakup mode
- Hypothesis: apply electric field to speed up the breakup process thus to suppress the capillary instability.

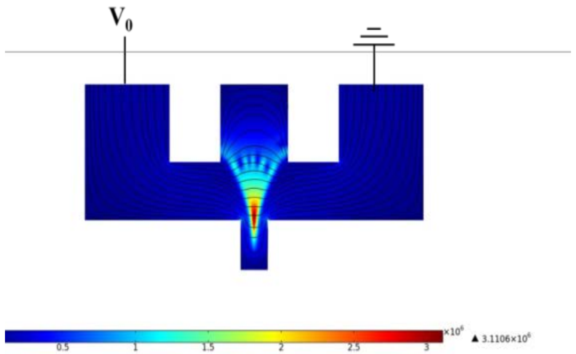
Effect of electric field on breakup mode

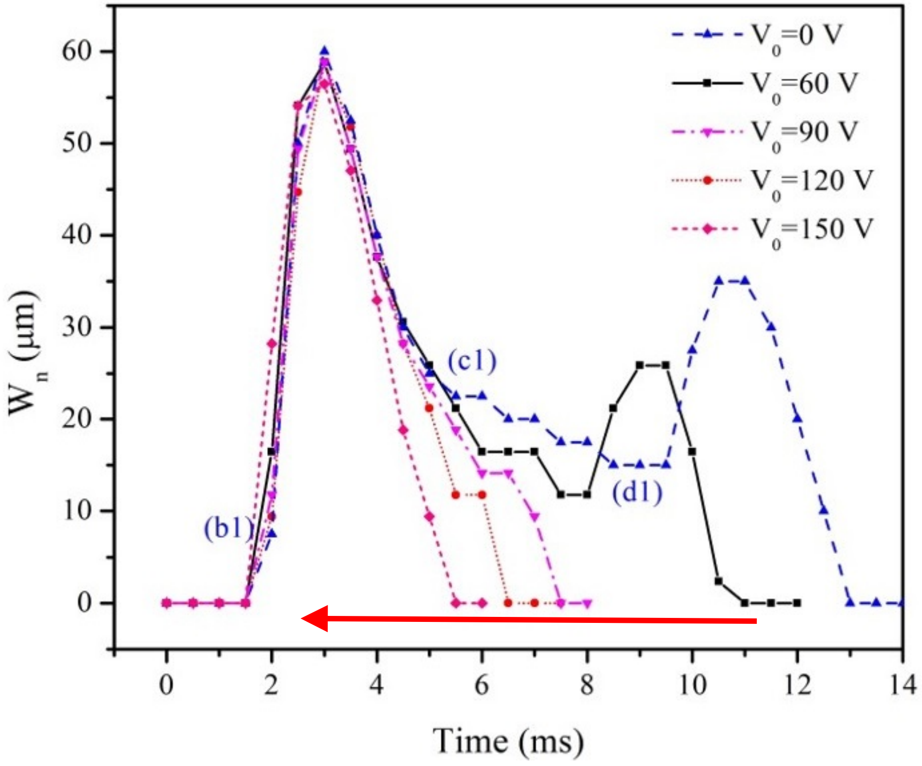
$\mu_d / \mu_c = 20$, $Q_c/Q_d = 50$, $Q_d = 0.04$ mL/h



(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





- The electric force squeezes the fluid neck thus reduces the droplet breakup time.
- When V_0 = 120 V is applied, the total droplet breakup time is reduced from 13 ms to ~ 7 ms.
- As the capillary instability does not have sufficient time to develop, the poly-dispersed breakup mode is eliminated.

Conclusions

- The simulations using Comsol have captured the droplet breakup modes successfully.
- The poly-dispersed breakup mode occurs due to the effect of capillary instability.
- The capillary instability requires certain time to develop before it can take effect.
- By shortening the droplet breakup time, the capillary instability can be suppressed, which can avoid the poly-dispersed breakup mode.
- By applying the external electric field, the electric force is induced on the fluid interface. The electric force helps to reduces the droplet breakup time thus to avoid the poly-dispersed breakup mode.
- As the applied voltage exceeds certain threshold value, the droplet breakup mode shifts from the poly-dispersed to the mono-dispersed one.

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LSU Campus

Chemical Engineering Dept, LSU

Questions?