

Electronic Supplementary Information (Figs. S1-S3 and Tables S1-S2).

Fluid displacement during droplet formation at microfluidic flow-focusing junction

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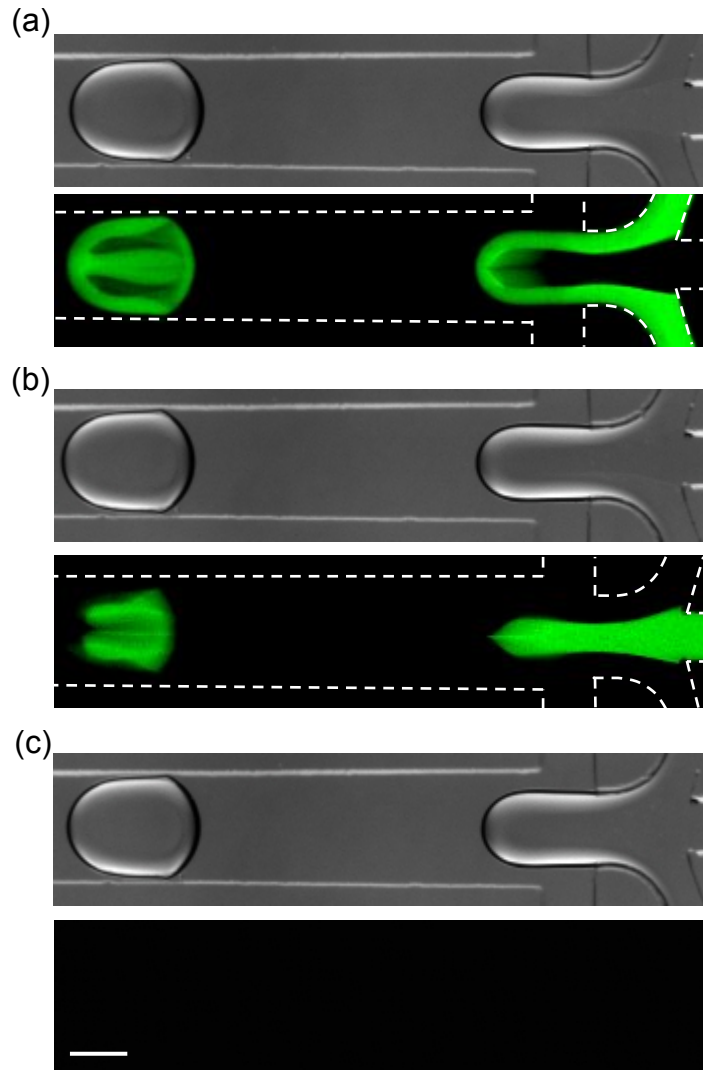


Fig. S1. Bright field and fluorescence images of pinched-off and pendent microdroplets showing the addition of fluorescein isothiocyanate-dextran (500 kDa) in the shell or core fluids does not affect droplet formation at the microfluidic flow-focusing junction. (a) The images of microdroplets formed with the fluorophore in shell fluid; (b) The images of microdroplets formed with the fluorophore in core fluid. (c) The images of microdroplets formed with no fluorophore in any fluid. Scale bar: 200 μm . Experimental condition: $\mu_{core} = \mu_{shell} = 23.4 \text{ mPa s}$, $\mu_c = 30.9 \text{ mPa s}$, $q_{core} = q_{shell} = 0.1 \text{ ml hr}^{-1}$, $q_c = 1 \text{ ml hr}^{-1}$, and $\sigma = 5.55 \text{ mN m}^{-1}$.

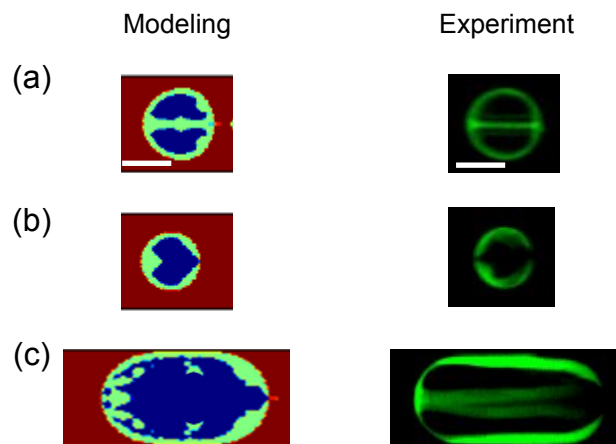


Fig. S2. Comparisons of results from modeling and experiments on the morphology of droplets generated by microfluidic flow-focusing junction. (a) Θ -shaped distribution of the shell solution, $\mu_{core} = \mu_{shell} = 23.4 \text{ mPa s}$, $q_c = 2 \text{ ml h}^{-1}$. (b) O-shaped distribution of the shell solution, $\mu_{core} = \mu_{shell} = 599 \text{ mPa s}$, $q_c = 2 \text{ ml h}^{-1}$. (c) Reduced fluid displacement in large microdroplets, $\mu_{core} = \mu_{shell} = 23.4 \text{ mPa s}$, $q_c = 0.2 \text{ ml h}^{-1}$. For all the studies, $q_{shell} = 33.3 \text{ } \mu\text{l h}^{-1}$, $q_{core} = 166.7 \text{ } \mu\text{l h}^{-1}$. Scale bars, $200 \text{ } \mu\text{m}$

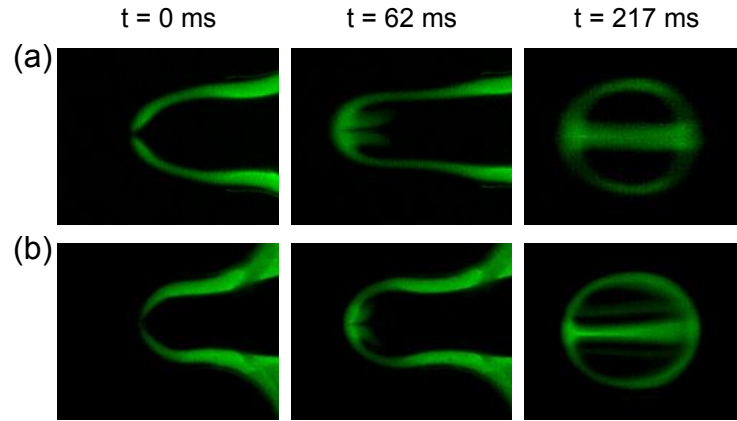


Fig. S3. The effect of interfacial tension on fluid displacement during droplet formation at microfluidic flow-focusing junction. (a) Strong interfacial tension causes fast but short-lasting capillary recoil, $Ca = 339$. (b) Small interfacial tension induces slow but long-lasting capillary recoil, $Ca = 144$. For both (a) and (b), $\zeta = 3.5$, $\lambda = 1.3$, $I_{in}/I_{out} = 1.2$ in the final droplets. Scale bar: $200 \mu\text{m}$.

Table S1. Dynamic viscosities of various fluids determined experimentally using rheometer

Fluid	Mean viscosity (mPa s)	Standard deviation (mPa s)
Water	1.0	0.1
0.5% (w/v) M cellulose* in water	23.4	0.2
1% (w/v) M cellulose in water	93.2	3.7
2% (w/v) M cellulose in water	598.8	9.3
1% (w/v) H cellulose# in water	797.1	41.5
Mineral oil	30.0	0.0
1% (v/v) span 80 in mineral oil	30.9	0.0
2% (v/v) span 80 in mineral oil	32.4	0.0
5% (v/v) span 80 in mineral oil	33.2	0.3
10% (v/v) span 80 in mineral oil	36.8	0.5
20% (v/v) span 80 in mineral oil	42.6	0.5
30% (v/v) span 80 n mineral oil	49.2	0.7
Mineral oil-CaCl ₂ emulsion	107.7	1.5
2% (w/v) alginate in water	23.6	0.4

*Medium viscosity carboxymethyl cellulose from Sigma

#High viscosity carboxymethyl cellulose from Sigma

Table S2. Interfacial tension of various fluid pairs determined experimentally using contact angle goniometer

Fluid pair	Mean interfacial tension (mN m ⁻¹)	Standard deviation (mN m ⁻¹)
Water - mineral oil	60.0	0.6
Water - mineral oil with 1% (v/v) span 80	6.3	0.1
Water - mineral oil with 2% (v/v) span 80	5.3	0.1
Water - mineral oil with 5% (v/v) span 80	5.2	0.3
Water - mineral oil with 10% (v/v) span 80	5.0	0.2
Water - mineral oil with 20% (v/v) span 80	4.7	0.2
Water - mineral oil with 30% (v/v) span 80	4.5	0.1
0.5% (w/v) M* cellulose in water - mineral oil with 1% (v/v) span 80	5.6	0.2
0.5% (w/v) M cellulose in water - mineral oil with 2% (v/v) span 80	4.6	0.2
0.5% (w/v) M cellulose in water - mineral oil with 5% (v/v) span 80	4.2	0.1
0.5% (w/v) M cellulose in water - mineral oil with 10% (v/v) span 80	4.1	0.2
0.5% (w/v) M cellulose in water - mineral oil with 20% (v/v) span 80	4.0	0.3
0.5% (w/v) M cellulose in water - mineral oil with 30% (v/v) span 80	3.8	0.3
1% (w/v) M cellulose in water - mineral oil with 1% (v/v) span 80	4.8	0.1
1% (w/v) M cellulose in water - mineral oil with 2% (v/v) span 80	3.9	0.0
1% (w/v) M cellulose in water - mineral oil with 5% (v/v) span 80	3.7	0.2
1% (w/v) M cellulose in water - mineral oil with 10% (v/v) span 80	3.5	0.3
1% (w/v) M cellulose in water - mineral oil with 20% (v/v) span 80	3.3	0.4
1% (w/v) M cellulose in water - mineral oil with 30% (v/v) span 80	3.0	0.5

*Medium viscosity carboxymethyl cellulose from Sigma