Formation of droplets at very low Capillary Numbers

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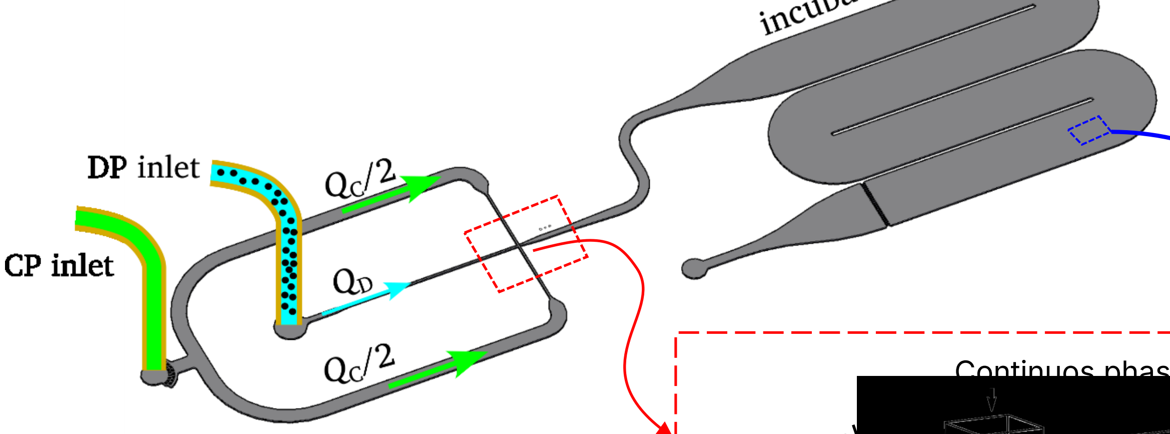
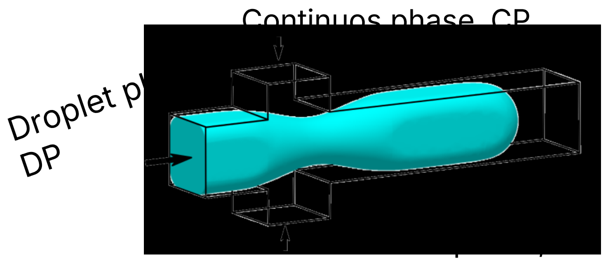
Droplet microfluidics

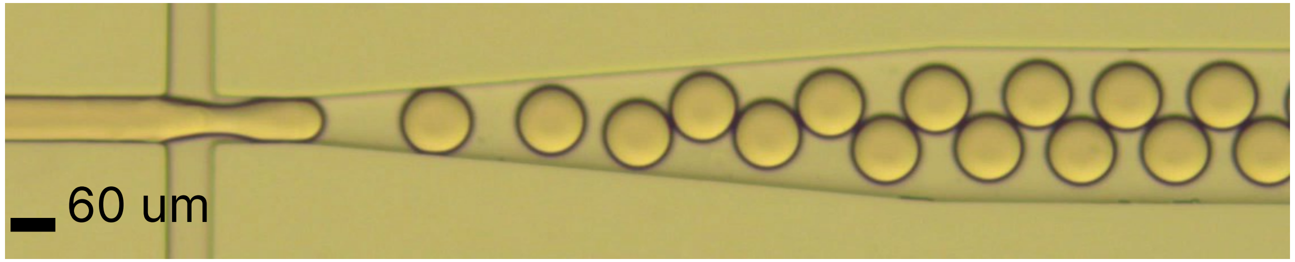
a technique that involves the generation, manipulation, and analysis of small droplets, usually ranging from picoliters to nanoliters, in a continuous fluid flow.

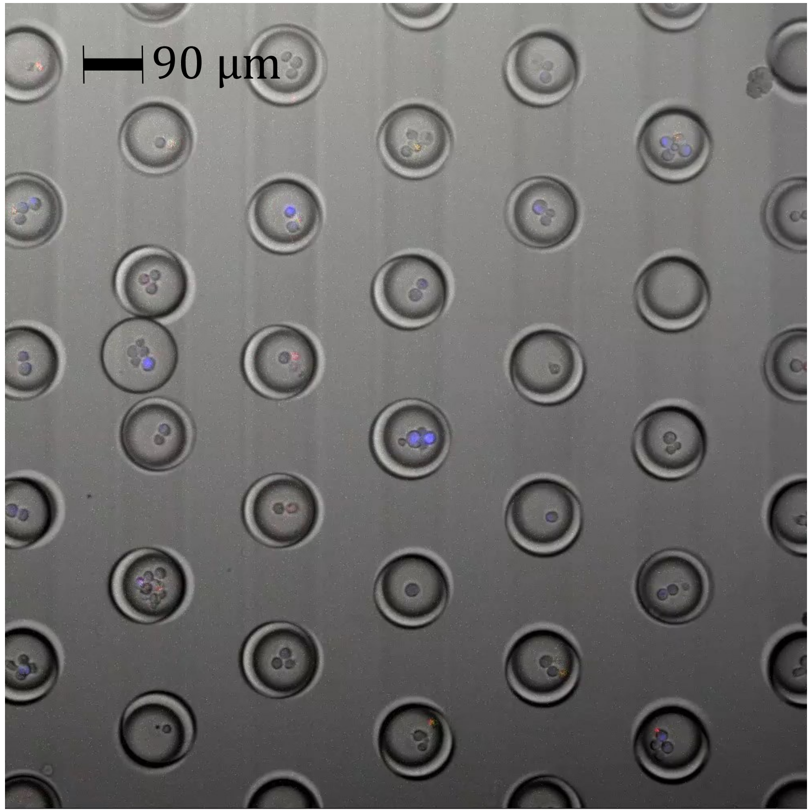
Example:
using droplets as cell incubator



A droplet generator “cross-junction”



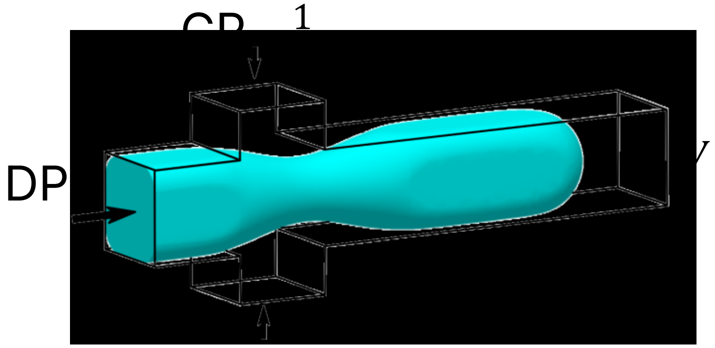




24 hr observation of Cancer cells + NK cells encapsulated in droplets.

The videos are my (remained) unpublished results.

Let's observe experiment result based on the variation of two parameters:



1. Flow rate ratio

$$q = \frac{Q_D}{Q_C}$$

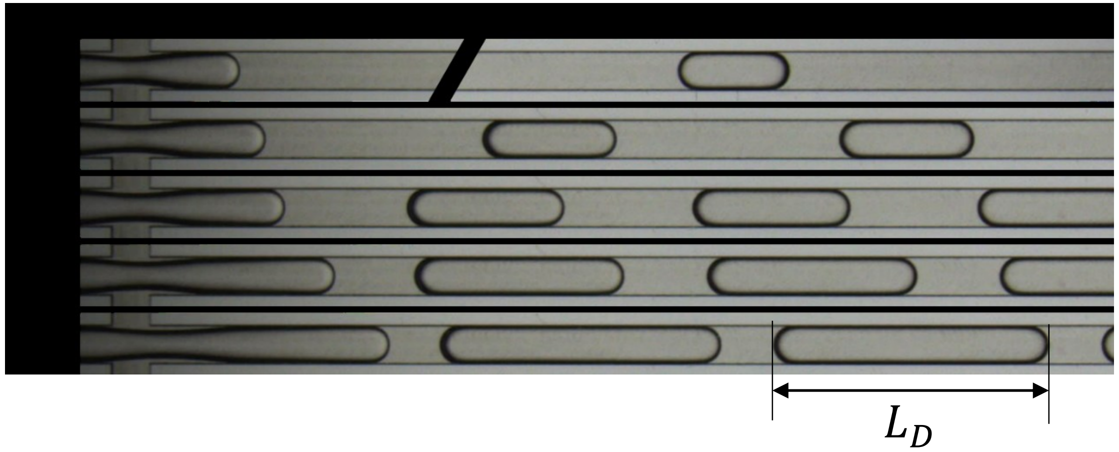
2. Capillary number

$$\mathrm{Ca} = \frac{Q_C \mu_C}{W H \gamma}$$

$$= \frac{\text{viscous forces}}{\text{surface tension forces}}$$

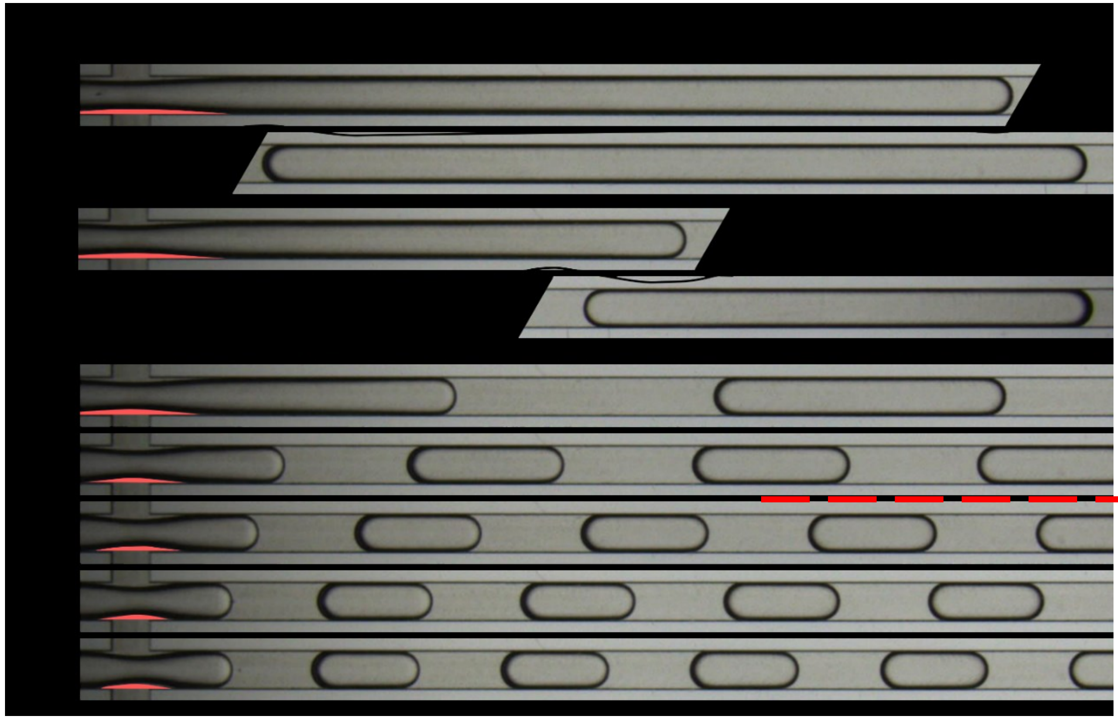
Dimensionless droplet volume: $l_D = \frac{L_D}{W} = \frac{V_D}{HW^2}$

1. Varying, q, while setting Ca constant:



l_D linearly dependent to q

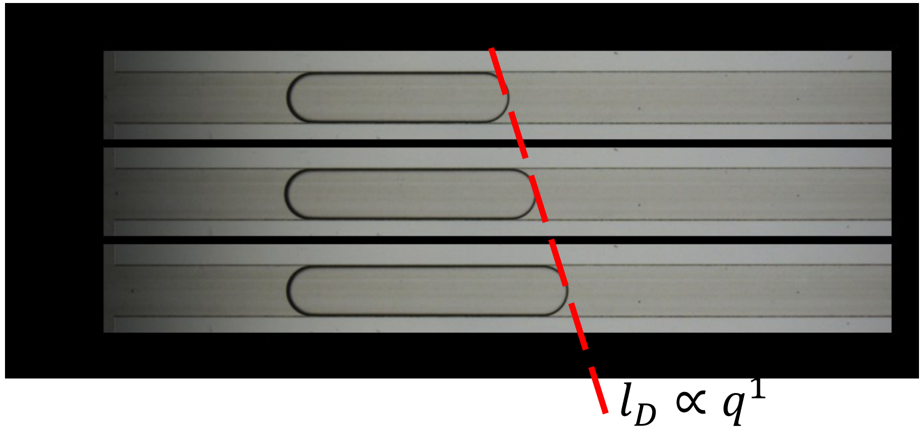
2. Varying Ca, while setting q constant:



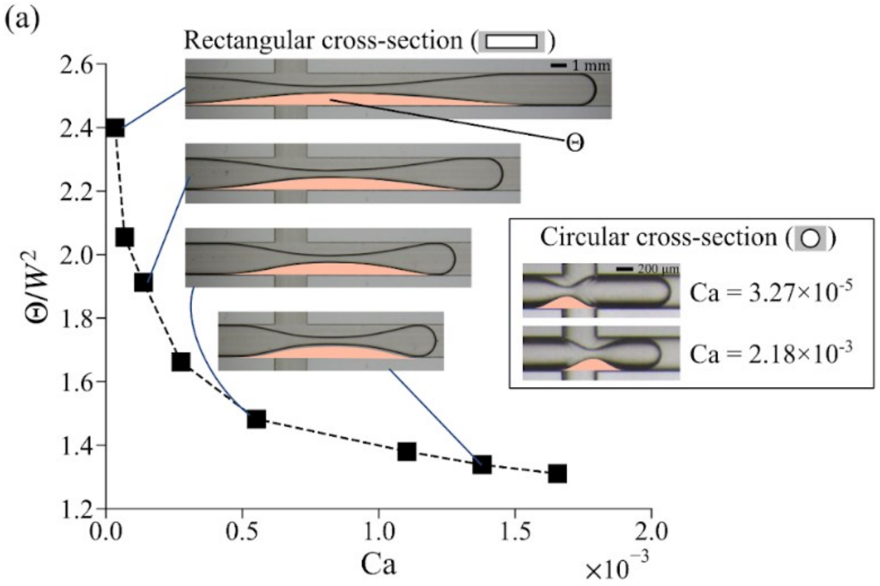
extreme increase of l_D at decreasing Ca

l_D almost independent to Ca (typical working regime)

Let's see another data sets...



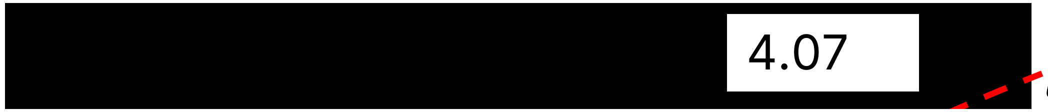
$l_D \propto q^1$



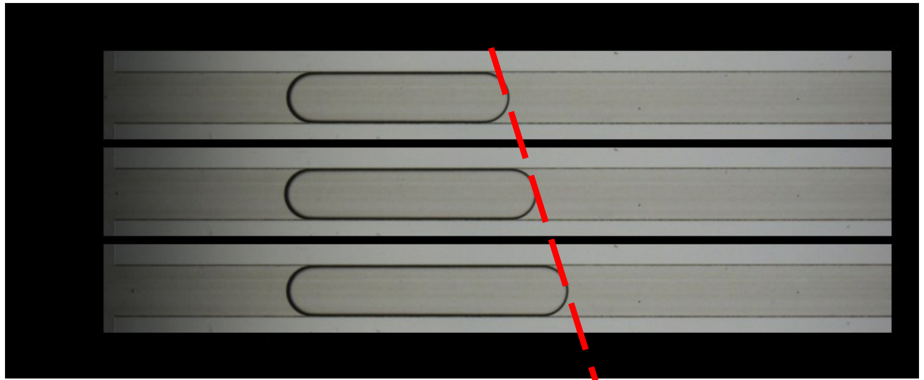
Additionally: the elongation of neck prior to pinch-off also observed at the very low Ca.

$l_D \propto \left(\frac{1}{Ca}\right)^?$

If we neglect the very low Ca droplets for a while..



$l_D \propto \left(\frac{1}{Ca}\right)^?$

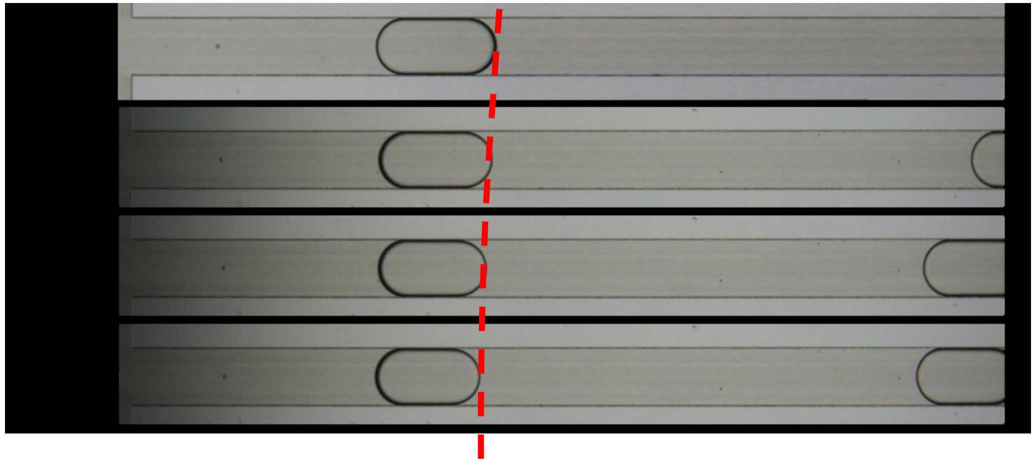


$l_D \propto q^1$

$$l_D = a + b\,q \quad *$$

- a and b are geometrical constants, not related to flow conditions.
- But if we include the very low Ca droplets, the scaling law breaks.
- The relation can be quite simple because of the squeezing rather than the shearing act of the CP against the DP that forms the droplet.
- Either one or both parameters a, b are not constant anymore
- It is known as the *squeezing* droplet formation.

Neglecting these for a while...



* First proposed in Garstecki (2006) with a= 1 and b~1 , verified experimentally using T-junction.

Let’s back to the original assumption:

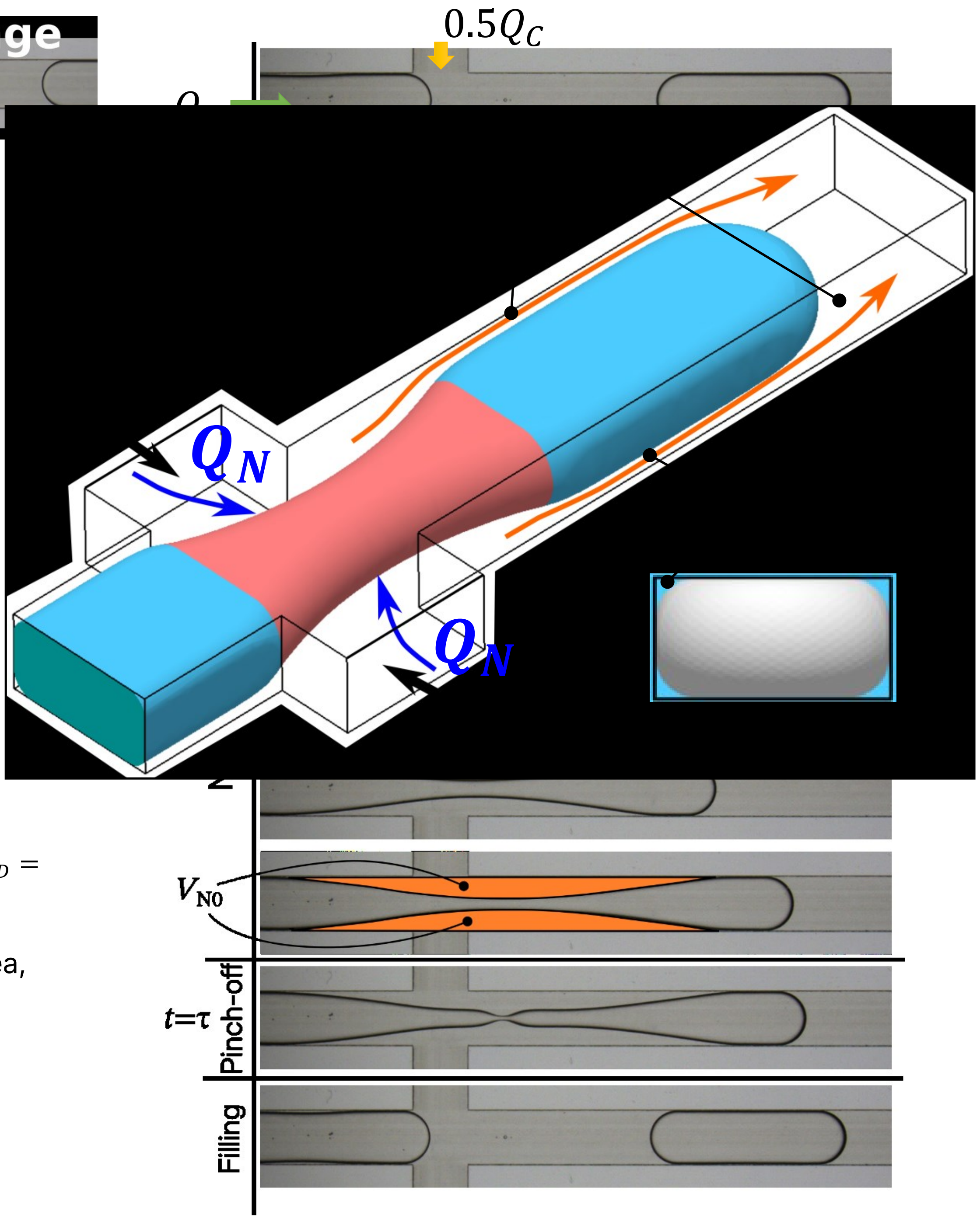
$$V_D = V_0 + \tau Q_D$$

Droplet volume — Initial volume — Necking duration — DP flow rate

In dimensionless form:

$$l_D = l_0 + \tau^* q$$

A general form of the scaling law which does not assume l_0 and τ^* as constants



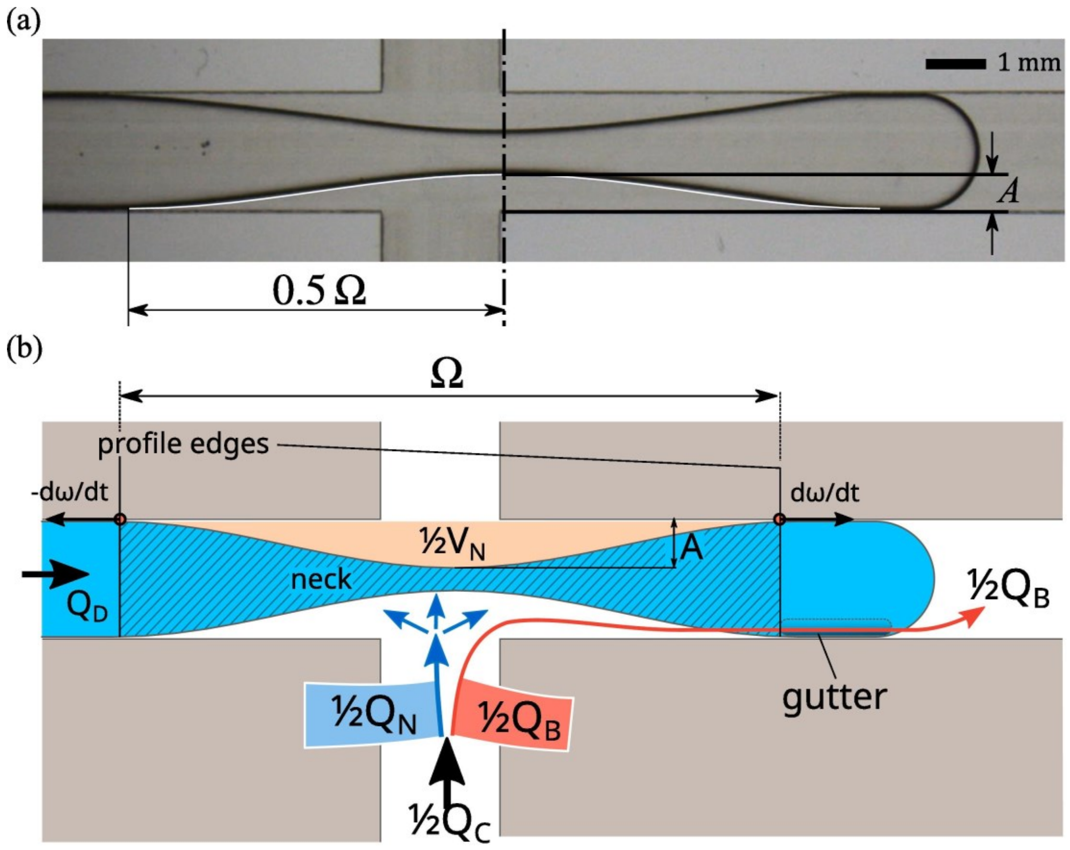
What is τ^*?

- If all CP flows to the necking area, $\tau = \frac{V_{N0}}{Q_C}$, where V_{N0} is the total DP volume displaced during the necking, which leads to the original squeezing equation, $V_D = V_0 + V_{N0} q$.
- However, in the very low Ca flow, only a fracti… o the necking area, Q_N such that $\tau = \frac{V_{N0}}{\langle Q_N \rangle}$.
- Defining $\langle q_N \rangle = Q_N/Q_C$ and $v_{N0} = V_{N0}/HW^2$, then

$$\tau^* = \frac{v_{N0}}{\langle q_N \rangle}$$

- Leaking flow has been described previously in T-junction device as $\langle q_N \rangle = \left(1 + \frac{\beta}{qCa}\right)^{-1}$, (Korczyk et. al., Nat. Commun., 2019).

Neck evolution



1. Assume the droplet edge as 2D neck profile and it has a shape following a sinusoidal shape.

   Neck profile: $h(x) = 0.5A\left[1 + \cos\left(\frac{2x}{\Omega}\pi\right)\right]$

   Volume under the neck: $V_N \approx 2H\int_{-\Omega/2}^{\Omega/2} h(x)dx = HA\Omega$

   Planar Curvature: $\kappa \approx d^2h(x)/dx^2 = 2\pi^2\frac{A}{\Omega^2}\cos\left(\frac{2x}{\Omega}\pi\right)$

2. Conservation of mass: $\frac{dV_N}{dt} = Q_N$

$$\frac{dA\Omega}{dt} = \frac{Q_N}{H} \quad (1)$$

3. Equation of motion:
   - We focus on the motion of two parameters of A and Ω.
   - In majority of necking process, the profile evolves in self-similar shape: we assume $d\Omega/dt \propto k\, dA/dt$, with k as a constant
   - With the viscous forces $\mu_C\, d\Omega/dt$ balanced by the surface tension effect, governs by the instantaneous profile curvature $\sim\gamma\frac{A}{\Omega^2}$.

   Put together: $$\frac{\mu_C}{\Xi}\frac{d\Omega}{dt} = \frac{\mu_C}{\Lambda}\frac{dA}{dt} + \gamma\frac{A}{\Omega^2} \quad (2)$$

μ_C: viscosity of CP,
Ξ, Λ are the constant parameters with dimension of $[L]$
γ: interfacial tension
W: Width of channel

1. Fill channel with DP, stationery,
2. Start necking (CP flow) and stop the flow (t= 0 s) before the breakup then observe.

Relaxation rate of the neck

- We cannot solve the previous set of equations: two equations vs three unknowns (A, Ω, and Q_{N})
- We can design a special experiment of which $Q_{\mathrm{N}} = Q_{\mathrm{C}} = 0$. And so we can solve the equations:

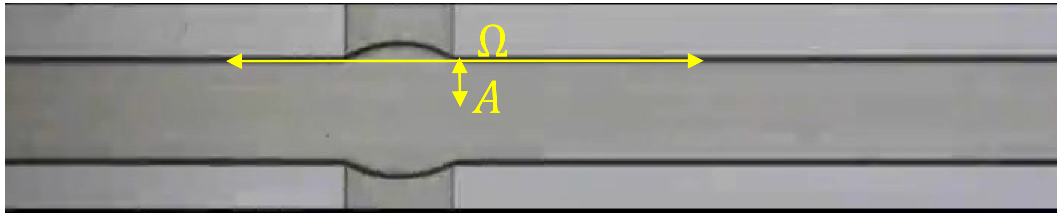
The solution* is:

$$\omega(\tilde{\tau}) = \omega_0 \sqrt{\sqrt{\left(\frac{a_0}{\omega_0}\frac{\xi}{\lambda} + 1\right)^2 + 4\xi\frac{a_0}{\omega_0^3}\tilde{\tau}} - \frac{a_0}{\omega_0}\frac{\xi}{\lambda}}$$

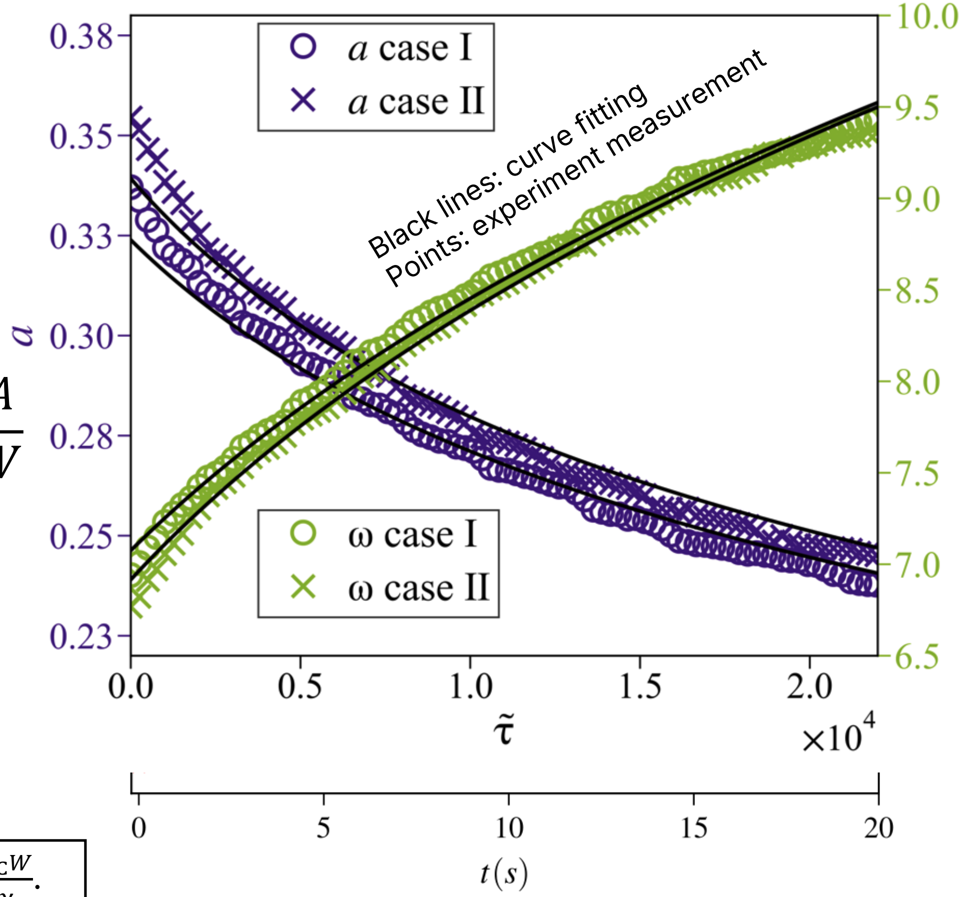
$$a = a_0\omega_0/\omega(\tilde{\tau})$$

- ξ and λ s the fitting parameters
- a_0 and ω_0 are the initial conditions.

The stop-flow experiment:



$a = \frac{A}{W}$



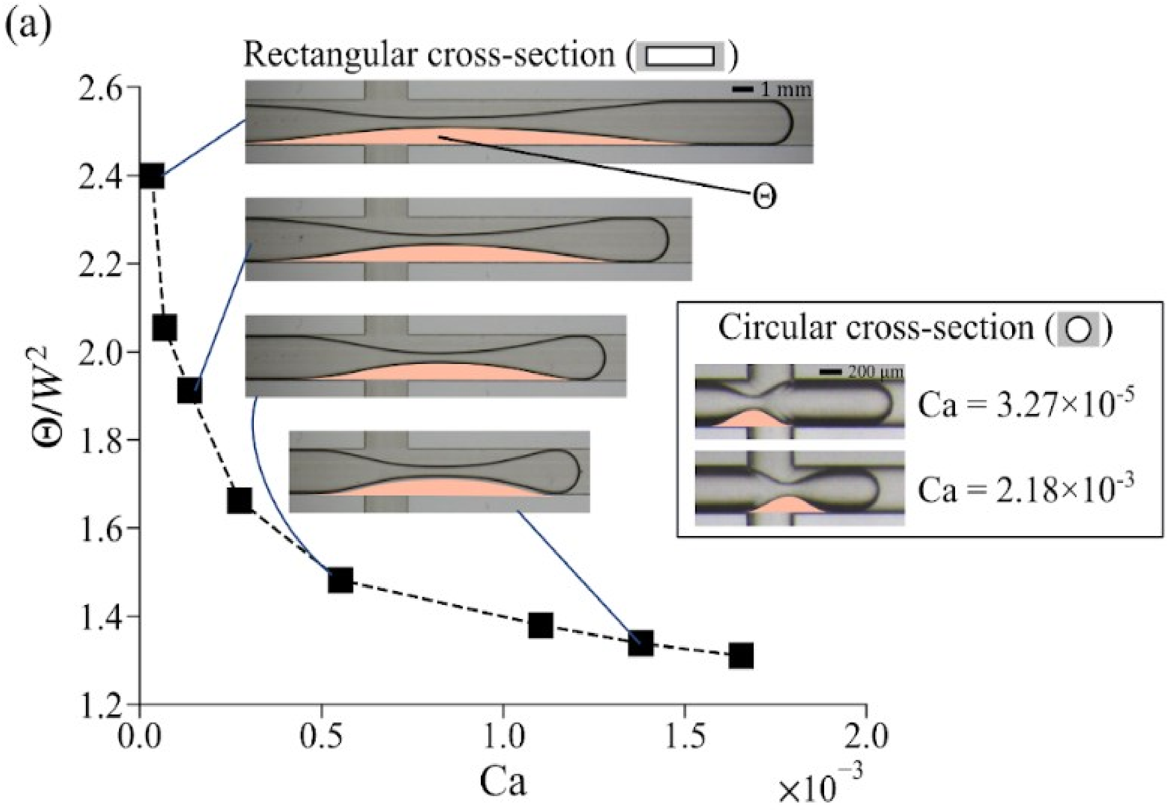
$\omega = \frac{\Omega}{W}$

*in non-dimensional form with $V_{\mathrm{N}}(t) = V_0$, $\lambda = \Lambda/W$, $\xi = \Xi/W$, $v_0 = \frac{V_0}{W^2 \mathrm{H}} = a_0\omega_0 = a\omega$, $t = \tilde{\tau}\frac{\mu_{\mathrm{C}} W}{\gamma}$.

Why the neck becomes wider as Ca decreases?

- The neck becomes wider due to:
  - Necking process $\propto \frac{dA}{dt}$
  - The relaxation of the surface, which we shown to be proportional to $\gamma \frac{A}{\Omega^2}$.

- So, the slower the necking process (due to low Ca + leaking effect), the more time for the neck to relax, causing the widening of neck and the increase of τ^*.



the elongation of neck prior to pinch-off also observed at the very low Ca.

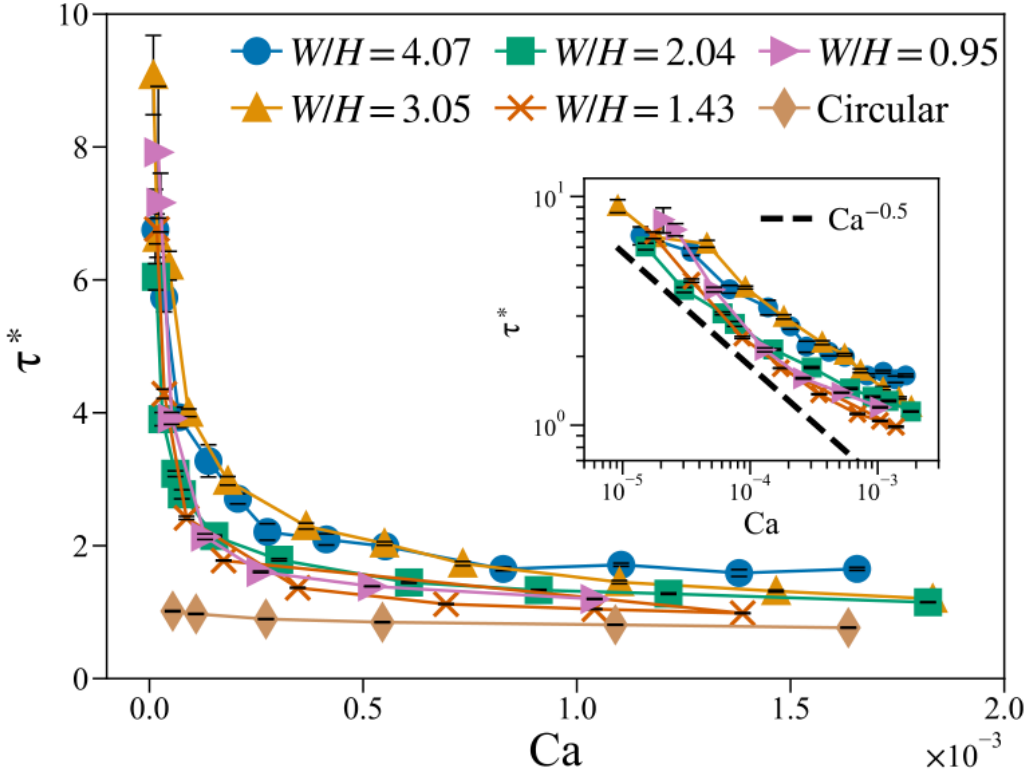
Conclussion

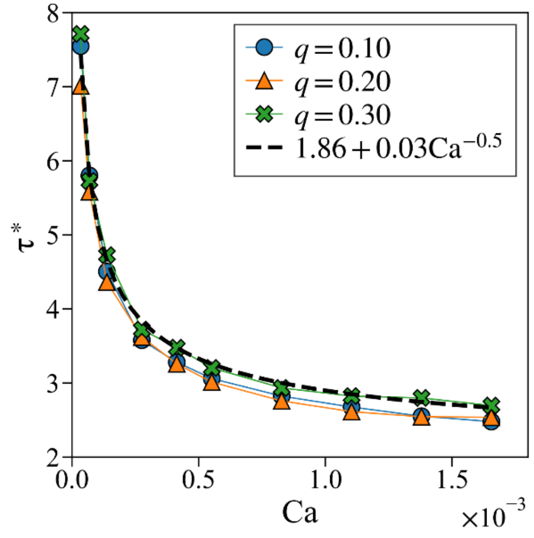
- Further analysis using the proposed equation of motions and other relations has shown the proportionality of $\tau^* \propto 1/\sqrt{\mathrm{Ca}}$.

$$\tau^* = \sqrt{\left(\frac{\xi}{\lambda}\right)^2 + \frac{8\xi}{\mathrm{Ca}}} + \frac{\xi}{\lambda}$$

- $\tau^* \propto 1/\sqrt{\mathrm{Ca}}$ correctly predicts the experimental measurement using variuos cross-junction device and different liquid pairs. The relation is an important milestone to generalize the equations of $l_D = l_0 + \tau^* q$.
- The complete story can be found in Kurniawan et. al. (J. Chem. Eng. ,2023).

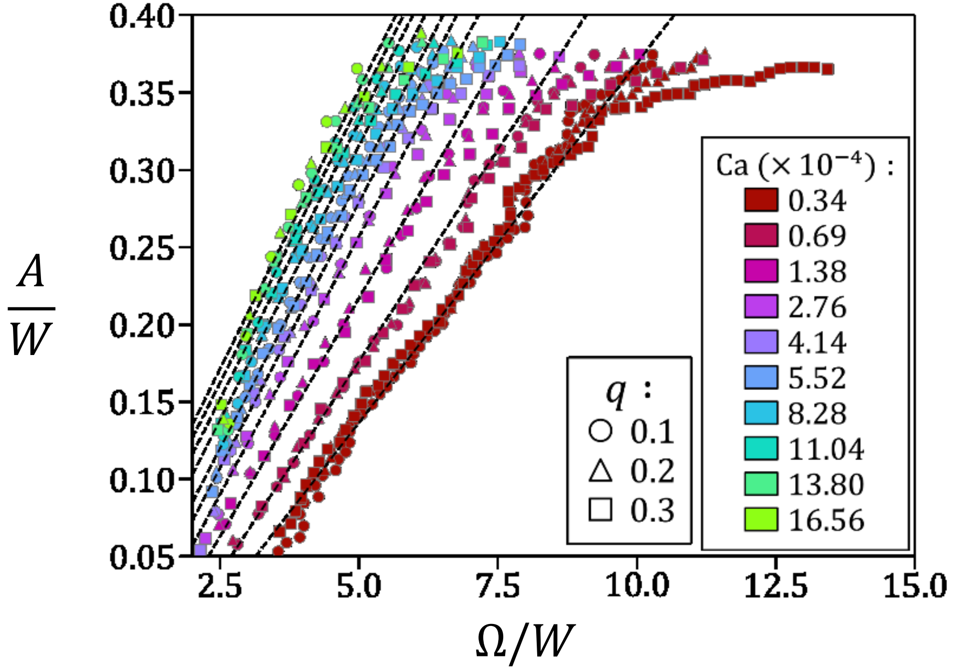
- *From simple model and special-tailored experiment, we can understand the elongation of neck during its evolution is due to the surface tension effect, which is proportional to the $\gamma \frac{A}{\Omega^2}$.*





Thank you for your attention!

Hopefully it is something that catch your interest!


0.40
0.35
0.30
0.25
0.20
0.15
0.10
0.05
A/W
2.5
5.0
7.5
10.0
12.5
15.0
Ω/W
q :
0.1
0.2
0.3
Ca (× 10⁻⁴) :
0.34
0.69
1.38
2.76
4.14
5.52
8.28
11.04
13.80
16.56