## Formation of dual-size droplets in the microfluidic cross-junction device

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We investigated droplet formation in a cross-junction microchannel which is a special type of hydrodynamics flow focusing device as shown in Fig. 1(a). At relatively low capillary number ( $Ca = \mu_C Q_C / \gamma HW$ , where  $\mu_C$  is the viscosity of the continuous phase,  $\gamma$  is the interfacial tension, W and H are the channel width and height, respectively), also known as the squeezing regime [1], droplets are generated by the squeezing action of the continuous phase (CP) to both sides of droplet liquid (DP). Constant flow rate source for CP ( $Q_C$ ) and DP ( $Q_D$ ) were supplied by syringe pumps. The formation process can be divided into three stages: I) Filling, ii) Necking and iii) Breakup. These three stages are clearly visualized by tracking the droplet tip position  $\ell_T$  and extreme width of the filament  $\varepsilon$  (both parameters were normalized by W), as shown in Fig. 1(b).

Depending on the flow parameters, there can be various flow patterns induced in the cross-junction [2]. At the squeezing regime, the three basic flow patterns are i) droplet formation at the junction "at Junction", ii) droplet formation at downstream "Downstream" and ii) the co-flow of both liquids without droplet-formation "Parallel Flow" (see Fig. 2). However, by using cross-junction device with various aspect ratio W/H, we have found that different flow patterns, "at Junction" and "Downstream" may co-exist at fixed flow rate parameters. We call this phenomenon as the bi-modal, shown as the yellow cross marker in Fig. 2. Contrary to the formation mechanism "at Junction", Downstream droplet formation does not include the Filling stage. After breakup, the filament recedes but stays inside the main-channel. Necking stage starts immediately without the free-filling stage. As the bi-modal alternates these two modes simultaneously, the it produces dual-sized droplets. We observed that the higher W/H the more likely the bi-modal appears. On the other hand, the use of higher W/H limits the range of q ( $=Q_D/Q_C$ ) that allows droplet formation. For devices with  $W/H \gg 1$ , the value of q in the transition from droplet formation to parallel flow is found to be linear function to device aspect-ratio,  $q_{trans}^{-1}=0.46(W/H)-0.45$ , confirming previous work of Humphry et. al. [3].

The relaxation of the filament following a droplet breakup is a crucial factor determining the occurrence of either droplet-formation modes. Approximation of the evolution of the maximum filament width (D) during relaxation process as a function of the droplet tip position  $(L_T)$  (see Fig. 3(a)) is written as

$$D = 0.5 E \left( 1 + \sqrt{1 + 4 (L_0 - L_T) / E + 4 Q_D t / (E^2 H)} \right)$$
(1)

where E is the fitting constant that represent the critical width of the filament. The fitting results of Eqn. (1) agrees very well to the experimental measurements as shown as the solid lines in Fig. 3(b).

## References

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Fig. 1 (a) simple cross-junction device (b) Three stages of droplet-formation : Filling (Blue), Necking (orange) and Breakup (at final time).



Fig 2 Regime map that shows different flow pattern for device with W/H=3.05. The bi-modal produces droplet by alternating two different mechanism simultaneously so that it generates dual-size droplets.



Fig. 3 (a) Overlay of filament outline (white lines) for every 0.1s depicting the relaxation process following droplet breakup. (b) The measurement result of D as a function of  $L_T$ . The solid lines represent fitting result of Eqn. (1). Fitting results for the circle (blue) and triangle (orange) data series are  $E=0.50 \& L_0=4.32$  and  $E=0.55 \& L_0=3.14$ , respectively (all units are in mm).