**ARTICLE TYPE** 

## Chemically induced droplet coalescence with microfluidics $^{\dagger}$

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## **Supporting Information**

Computational fluid dynamics simulations of steady-state, undisturbed fluid flow through the section of the microfluidic channel where droplet merging occurs. Simulations are performed in COMSOL 4.3a as a two dimensional laminar flow model. At low PFB flow, streamlines from the main channel flow around both sides of the oval obstruction. Droplets entrained in this flow follow the nearest streamlines: hence we steer them to the lower wall of the main channel so that they interact with the incoming PFB and flow under the obstruction. As PFB flow is increased, streamlines from the main channel are forced to flow primarily above the obstruction, causing droplets in the flow to be driven away from the PFB and making it impossible for them to merge at this point. These results agree with experimental observations, in which higher PFB flows do not result in droplet merging at the oval obstruction and instead push droplets to flow in the upper pathway around the obstruction

Fluorescence assay experiments (see Fig.S4) are performed by encapsulating dextran labeled with Rhodamine B (Mw $\sim$ 70kDa, Sigma-Aldrich) and dextran labeled with Fluorescein (Mw $\sim$ 70kDa, Sigma-Aldrich) in droplets and characterized by confocal microscopy (Leica SP5). The time stability of fluorescence assay droplets are done by storing the collected droplets in closed vials for three days and imaging the droplets on confocal microscopy at 24, 48, and 72 hours.

In the high speed movie, droplet pairs enter the constriction region and coalesce after exposure to the PFB stream. The movie is recorded at 2000 frames per second with a 40X objective.



Fig. S1 Fluid streamlines for operational flow rates in the merging microfluidic geometry generated by computation fluid dynamics in COMSOL. Flow rates for PFB addition are (A)  $5\mu$ L/hr and (B)  $100\mu$ L/hr.



Fig. S2 Complete scale device drawing.



**Fig. S3** Comparison between microfluidic chemical and electro-coalescence. (A) a collected merged droplet population using PFB to destabilize, (B) collected merged droplet poluation using an electro-coalescence device operating at 0.8kV.



**Fig. S4** Two-color overlay fluorescence images of droplets collected from the outlet of the microfluidic device using: (A) HFE-7500, (B) PFO, and (C) PFB; scale bar is 150mm. 2-D histogram of all individual droplet fluorescence intensities obtained from coalesced droplet pairs in (D) HFE-7500, (E) PFO, (F) PFB.

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