Supplementary Information

Dynamics of liquid droplets in an evaporating drop: Liquid droplet "Coffee Stain" effect

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1 Schematic of the Droplet Generation Chip

In the Figure below, we show the schematic of the droplet generation chip, as well as the mechanism of the droplet generation.

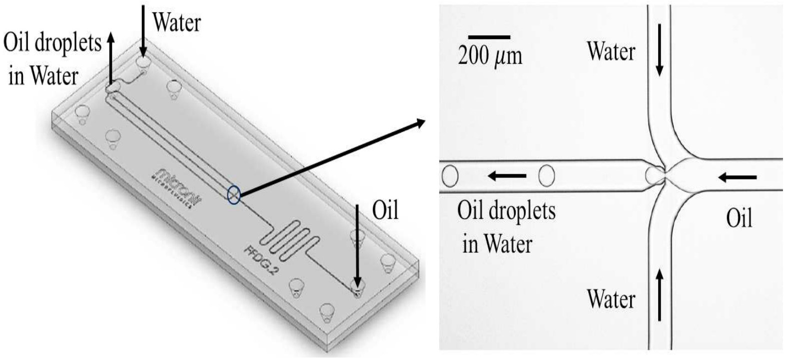

Figure 1 (left) Schematic of the Focussed Flow Droplet Generation Chip. (Right) Schematic of the droplet generation in the Droplet Generator (arrows are added to indicate the direction of the flow).

2 Derivation of the geometric relationship for the "enclosure" phenomenon

In this section, we derive the geometric relationship [eq.(3) in the main text] that define the "enclosure" distance as a function of the system parameters.

First, we provide the derivation for the case where the oil droplets make acute contact angle with the solid substrate (oil droplets on polycarbonate in water medium). The corresponding picture is shown in figure (2a) in the main text. We attempt to quantify the "enclosure" effect by obtaining x [see figure (2a) in the main text], which is the distance of closest approach of the

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droplet to the TPCL, as a function of the system parameters . From ΔAMC and ΔBMC in figure (2a), we can obtain:

$$\begin{aligned} y = AM = CM\tan\theta = R\cos\theta_0\tan\theta, \\ z = R - CA = R - \frac{CM}{\cos\theta} = R\frac{(\cos\theta - \cos\theta_0)}{\cos\theta}, \\ r_c = R\sin\theta_0, \end{aligned} \tag{1}$$

where θ is the sessile droplet contact angle, θ_0 is the oil-droplet contact angle, R is the radius of curvature of the oil droplet and r_c is the contact radius of the oil droplet. In ΔABC,

$$\sin\theta = \frac{BC}{CA} = \frac{z}{x + r_c - y}. \tag{2}$$

Using eqs.(1,2), we get after certain trivial simplifications:

$$\frac{x}{r_c} = \mathrm{cosec}\theta\,\mathrm{cosec}\theta_o - \cot\theta\cot\theta_o - 1. \tag{3}$$

We next consider the case where the oil droplets make obtuse contact angle with the solid substrate (oil droplets on glass in water medium). The corresponding picture is shown in Figure (2b) in the main text. In this figure, we have ΔABC and ΔADC congruent (since $BC = CD = R$, angleABC=angle$ADC$$= 90^\circ$, and AC is the common side, i.e., by RHS congruency). Therefore angleBAC=angle$DAC$$= 90^\circ - (\theta_0 - \theta)/2$ (since in ΔAOB, exterior angleDAB=2angleBAC=2angleDAC=angleAOB+angle$ABO$$=180^\circ - \theta_0 + \theta$). Therefore, in ΔABC,

$$\frac{BC}{AB} = \tan\left(90^\circ - \frac{\theta_0 - \theta}{2}\right) \Rightarrow AB = \frac{R}{\cot\left(\frac{\theta_0 - \theta}{2}\right)}. \tag{4}$$

Similarly, in ΔAMB, using eq.(4), we shall get:

$$\begin{aligned} \frac{BM}{AB} = \cos(180^\circ - \theta_0) \Rightarrow BM = -\frac{R\cos\theta_0}{\cot\left(\frac{\theta_0-\theta}{2}\right)}, \\ \frac{AM}{AB} = \sin(180^\circ - \theta_0) \Rightarrow AM = \frac{R\sin\theta_0}{\cot\left(\frac{\theta_0-\theta}{2}\right)}. \end{aligned} \tag{5}$$

Finally in ΔAOM, using eq.(5), we get:

$$\frac{AM}{OM} = \tan\theta \Rightarrow OM = \frac{R\sin\theta_0}{\tan\theta\cot\left(\frac{\theta_0-\theta}{2}\right)}. \tag{6}$$

Therefore, we can write the enclosure distance $x = OB = OM + BM$, with $R = r_c/\cos(\theta_0 - 90^\circ) = r_c/\sin\theta_0$, as [using eqs.(5,6) and the identities $\cot\alpha/2 = \sin\alpha/(1-\cos\alpha)$]:

$$\begin{aligned} \frac{x}{r_c} &= \frac{\cot\theta}{\cot\left(\frac{\theta_0-\theta}{2}\right)} - \frac{\cot\theta_0}{\cot\left(\frac{\theta_0-\theta}{2}\right)} \\ &= \frac{(\cot\theta - \cot\theta_0)\left[1 - \cos(\theta_0 - \theta)\right]}{\sin(\theta_0 - \theta)} \\ &= \frac{(\sin\theta_0\cos\theta - \sin\theta\cos\theta_0)\left[1 - \cos(\theta_0 - \theta)\right]}{\sin\theta_0\sin\theta\sin(\theta_0 - \theta)} \\ &= \mathrm{cosec}\theta\,\mathrm{cosec}\theta_o - \cot\theta\cot\theta_o - 1. \end{aligned} \tag{7}$$

Eqs.(3,7) prove that the geometric relationship defining the “enclosure” effect is given by eq.(2) in the main text, and this relationship remains the same irrespective of whether the oil droplets form the acute or the obtuse angles.