

Supplementary Information

Universality in microdroplet nucleation during solvent exchange in Hele-Shaw-like channels

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1. Contact angle of decane

Since it is impossible to measure the contact angle of decane directly in the channel, we measure the contact angle separately with a silicon wafer and a glass wafer. The silicon wafer is first etched with the same procedure so as to have the same roughness as the silicon walls in the channel. Then both the silicon wafer and the glass wafer are hydrophobized with the same procedure. Later they are immersed in decane, and a drop of water is released on top of each sample to measure the contact angle, see figure 1. The contact angle of decane on the hydrophobic silicon (immersed in water) is $\theta = 15 \pm 3^\circ$, and the contact angle of decane on the hydrophobic glass (immersed in water) is $\theta = 11 \pm 2^\circ$.

2. Finding the corresponding point S for point M

Again let $x = w_e$ and $y = w_o$. The binodal $w_{o,s} = w_{o,s}(w_e)$ can be rewritten in a new format:

$$y = y_s(x) \quad (2.1)$$

Then the Y -coordinate of point A is $y_A = y_s(x_A)$. Since the real Y -coordinate of point B: $y_B = 9 \times 10^{-9}$ is almost zero, we consider B to locate approximately at the origin, i.e., $y_B \approx 0$. Then the equation for the diffusion path AB is:

$$y = \frac{y_s(x_A)}{x_A} x, \quad 0 \leq x \leq x_A \quad (2.2)$$

The equation for the dilution curve is:

$$y = 1 + kx \quad (2.3)$$

Let (m, n) be the coordinate of point M, and (a, b) the coordinate of point S. Since M

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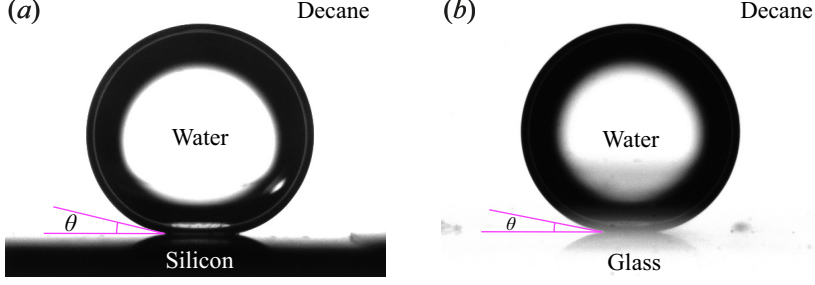


FIGURE 1. Contact angle of a water drop immersed in decane on (a) the silicon wafer and (b) glass. The silicon wafer is etched by the same method to achieve the same roughness as those in the channel. The silicon and the glass wafer are both hydrophobized in the same way as the channels. The contact angle of decane on such treated hydrophobic silicon wafer is $\theta = 15 \pm 3^\circ$, and the contact angle of decane on such treated hydrophobic glass wafer is $\theta = 11 \pm 2^\circ$.

is the intersection of the dilution curve and the diffusion path, and S is the intersection of the dilution curve and the binodal, we have:

$$\left. \begin{aligned} n &= \frac{y_s(x_A)}{x_A} m \\ n &= 1 + km \\ b &= y_s(a) \\ b &= 1 + ka \end{aligned} \right\} \quad (2.4)$$

First cancel out n and b in Eq.(2.4), then cancel out k , we obtain:

$$\frac{y_s(x_A)}{x_A} - \frac{1}{m} = \frac{y_s(a) - 1}{a} \quad (2.5)$$

x_A is the initial condition, y_s is a known function, then a as a function of m , $a = g(m)$, can be solved implicitly, because a appears in the function of the binodal $y_s(a)$. Or in other words, from the following equation:

$$\frac{w_{o,s}(w_{e,A})}{w_{e,A}} - \frac{1}{w_e(M)} = \frac{w_{o,s}(w_e(S)) - 1}{w_e(S)} \quad (2.6)$$

we obtain $w_e(S) = g[w_e(M)]$. Then we have:

$$c_e(S) = \rho(S) \cdot g \left[\frac{c_e(M)}{\rho(M)} \right] \quad (2.7)$$

where $\rho(S)$ and $\rho(M)$ are the density of mixture S and M, respectively.

Or Eq.(2.7) can also be simply noted as:

$$c_e(S) = f[c_e(M)] \quad (2.8)$$

For cases when the diffusivities are not equal, Eq.2.3 is changed to the diffusion path as calculated by Eqs.(C3) and (C4) in Appendix C.

3. Density of the mixture

We couldn't find any data on the density of the decane-ethanol-water mixture. However, since the oil weight fraction in all the solutions are very low, we approximate the density of the ternary mixture $\rho(w_e, w_o)$ by the density of the binary ethanol-water

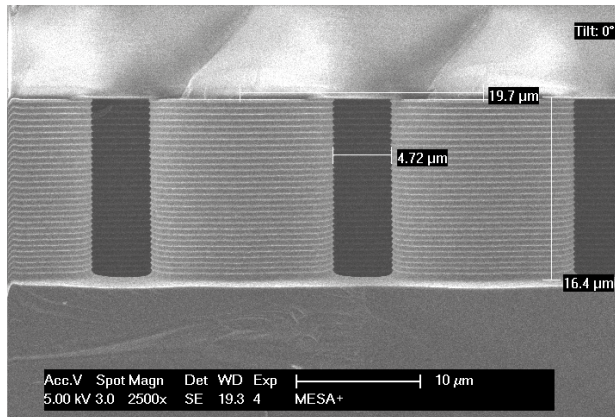


FIGURE 2. SEM image of the cross-section of a chip. The height of the channel is $h = 19.7 \mu\text{m}$.

mixture $\rho_{\text{binary}}(w_e)$. That is to say, $\rho(\text{S}) \approx \rho_{\text{binary}}(w_e(\text{S}))$ and $\rho(\text{M}) \approx \rho_{\text{binary}}(w_e(\text{M}))$. The density ρ_{binary} and the viscosity μ_{binary} of ethanol-water binary mixtures as functions of w_e is taken from [Khattab *et al.* \(2012\)](#).

4. SEM image of the cross-section of the chip

Figure 2 shows the cross-sectional Scanning Electron Microscope (SEM) image of one of the chips from the same batch. The height of the channel is measured to be $h = 19.7 \mu\text{m}$.

5. Confocal snapshots of the entire channel

The mid-plane snapshots of the entire channel are shown in figure 3. Figure 3(a) & (b) correspond to figure 2(a) & (b), respectively. As is shown, for channels with the porous region, no droplets are observed after the porous region (figure 3(a)). For channels without the porous region, droplets are observed across the entire channel. Note that for the channels without the porous region, L is defined as the length of the entire channel.

REFERENCES

- KHATTAB, I. S., BANDARKAR, F., FAKHREE, M. A. A. & JOUYBAN, A. 2012 Density, viscosity, and surface tension of water+ ethanol mixtures from 293 to 323k. *Korean J. Chem. Eng.* **29** (6), 812–817.

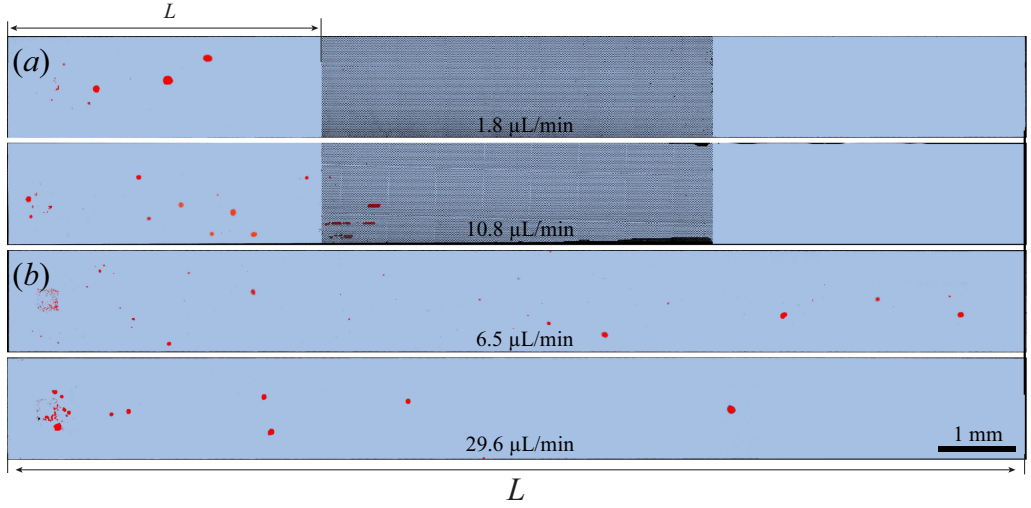


FIGURE 3. (a) Mid-plane snapshots of the entire channel of Chip No. 1, corresponding to figure 2(a). L is the length to the porous region. (b). Mid-plane snapshots of the entire channel of Chip No. 6, corresponding to figure 2(b). Red signals oil, light blue signals water, and black the pillars. Scale bar indicates 1 mm. L is the length of the entire channel, since no porous region is presented.