Supplementary Information

Fluoropolymer surface coatings to control droplets in microfluidic devices

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Video S1. Droplet formation in a coated T-junction device with main and inlet channel widths of 400 and 100 μ m, respectively. The dispersed phase flow rate was 20 mL/h and the continuous phase flow rate was 200 mL/h. Images were captured at 159250 frames per second with a 1.76 μ s exposure and they are played back at 100 frames per second.

Video S2. Droplet formation in a coated T-junction device with main and inlet channel widths of 400 and 100 μ m, respectively. The dispersed phase flow rate was 100 mL/h and the continuous phase flow rate was 200 mL/h. Images were captured at 159250 frames per second with a 1.76 μ s exposure and they are played back at 100 frames per second.

Description of Image Analysis:

Each captured image was processed as a 2-D matrix with its row and column indices representing pixel numbers and each entry representing the intensity at the corresponding pixel. To simplify the calculation, we averaged the intensity of each column to convert the 2-D matrix into a 1-D vector. At droplet edges, the intensity changed drastically (at left edges, intensity decreased rapidly and at right edges, intensity increased rapidly). We calculated the derivatives of the intensity plots and then searched for local optima, which occurred at the droplet edges.

We processed the intensity data to eliminate imaging artifacts, mainly originating from the reflection at the center of the droplets. To achieve this, we used a Gaussian convolution filter. The convolution of functions f(t) and g(t) is defined as:

$$(f * g)(t) = \int_{-\infty}^{\infty} f(\tau)g(t-\tau)d\tau$$
(1)

In this specific case, $f(\tau)$ is the column-averaged intensity in a single image and the weighting factor $g(\tau)$ is the Gaussian kernel. The 1-D Gaussian kernel is

$$g(x) = \frac{1}{\sqrt{2\pi\sigma}}e^{-\frac{\left(x - x_{center}\right)^2}{2\sigma^2}}$$
(2)

where the x_{center} is the center of the function and σ defines the width of the kernel. We used a σ value of 32 or 64 pixels to locate the approximate droplet edges and avoid detecting artifacts. Then we used these initial positions and halved the values of σ down to one to precisely calculate the exact edge positions. A representative image of droplets with the detected edges overlaid is shown in Fig. S1.



Figure S1. Micrograph of coated channel with main channel width of 400 μ m and the detected droplet edges are drawn in red.



Figure S2. Polymer thicknesses measured by profilometry at two different locations in the serpentine channels for samples coated for various times. Thicknesses were measured on a silicon wafer masked by the PDMS channel.

Description of PDMS Organic Phase Absorption:

We used slabs of PDMS that were 2 mm thick. A conservative estimate treating the PDMS as a membrane reveals that >20 μ L/h of hexanes should diffuse through the slabs.¹ This is more than the estimated 15 μ L/h of hexanes that each PDMS device was exposed to during a typical experiment, indicating the PDMS was not saturated. There was no observable trend in terms of droplet merger time with respect to the amount of time the device had been used. Reducing the thickness of the PDMS slab would linearly increase the rate of organic transport through the PDMS during an experimental run.

1. Stafie, N.; Stamatialis, D. F.; Wessling, M., Insight into the transport of hexane–solute systems through tailor-made composite membranes. *Journal of Membrane Science* **2004**, 228, (1), 103-116.