Electronic Supplementary Information (ESI)

Double-step emulsification device for direct generation of double emulsion†

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1 Estimate real droplet diameter from compressed pancake shape

To determined diameter of droplet from its apparent diameter obtained from image processing (Fig. S1 Top). The shape of droplet is assumed axisymmetric meaning it's laterally unconfined. The channel top and bottom walls in vertical direction confine a droplet equal to a height of the channel height h. In the case of a non-wetting droplet, curvature intersects the plates tangentially with a contact angle 180°. The thickness of the lubrication film between droplet and channel walls is neglected, so the shape only given by capillary force and the effect of shear force is negligible. To meet the criteria, the shape of droplet has been described as axisymmetric surface having a constant mean curvature which is a portion of nodoid. However, the equation can only be numerically solved. As a simple approximation, here we use filled torus model, which suggests low relative error (less than 0.8%) in comparison to solution of portion of nodoid[1].

Hence we describe relationship of apparent diameter D'_i obtained from image (Fig. S1 Bottom) into spherical diameter D_i as following expression:

For $D'_i \leq h$,

$$D_i = D'_i \tag{1}$$

For $D'_i > h$,

$$D_i = D'_i\left(\frac{3h}{2D'_i}\left(1-2\frac{h}{D'_i}+\frac{5}{3}\left(\frac{h}{D'_i}\right)^2+\frac{\pi h}{2D'_i}\left(1-\frac{h}{D'_i}\right)\right)\right)^{\frac{1}{3}} \tag{2}$$

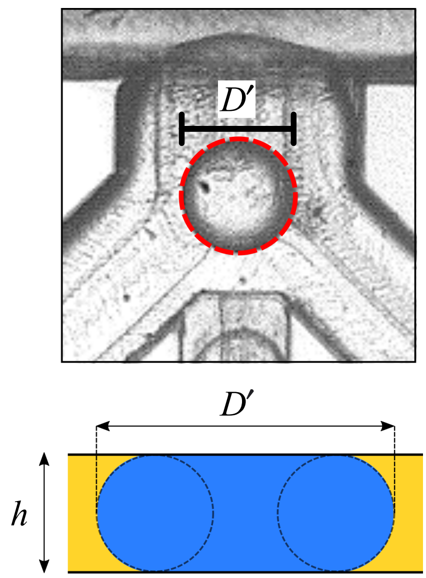

Figure S1 Outline of droplet captured from image processing (red dash line) determines apparent diameter of droplet in between parallel plates (D'_i). Top: image acquired from camera. Bottom: Cross section of compressed droplet while $D'_i > h$.

2 Surface measurement

In the Fig. S2 we show images of surface profile. The data is acquired by optical profilometer (Contour GT-K,Bruker,USA) followed by image post processing on software Gwyddion[2]. The extracted cross section of each step (Fig. S2 Right) allowed to determine step dimension.

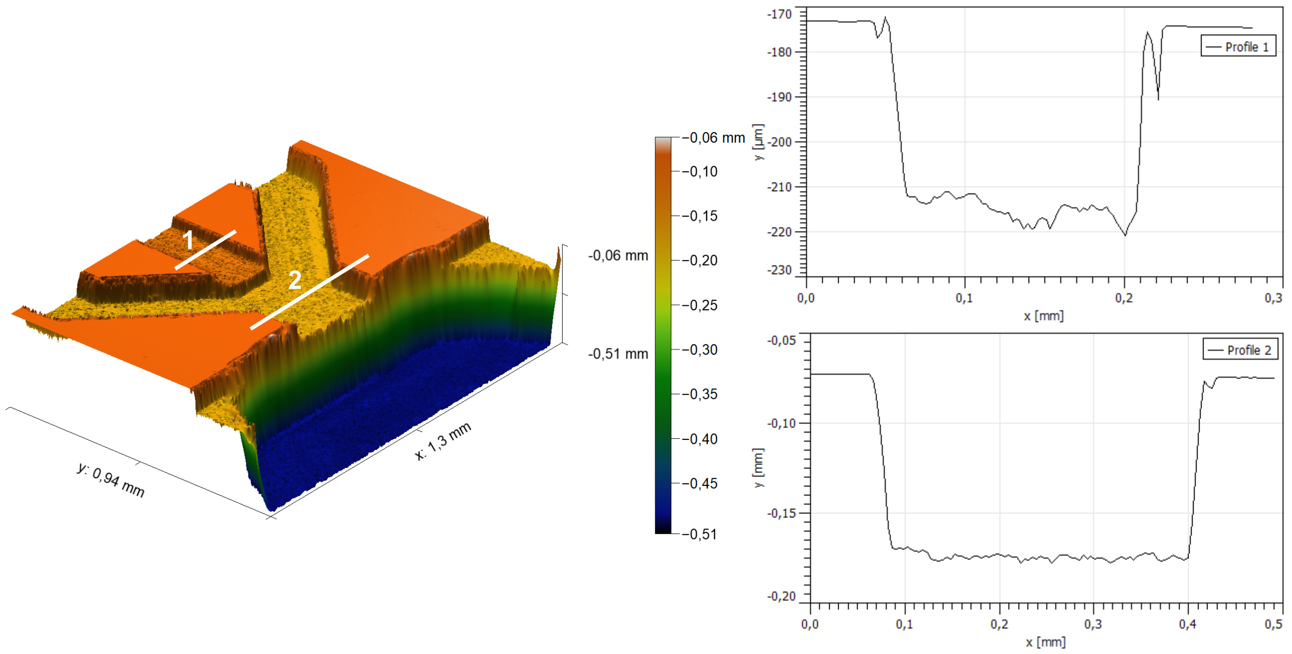

Figure S2 Surface measurement from profilometer showing the substantial geometry, which is two-stair nozzle after the device is milled. Left: 3D topography image. The white lines approximately depict the route of profile line. Right: profile of the scanned image.

3 Surface modification for W/O/W double emulsion

Such modification would require very precise hydrophilicity patterning. In particular, the water inlet channel and the vertical wall behind the first step would need to be modified hydrophobic, while the rest of the device - hydrophilic (Fig. S3). Such selective modification is challenging (we have not attempted it in this work) which may be a limitation of our device.

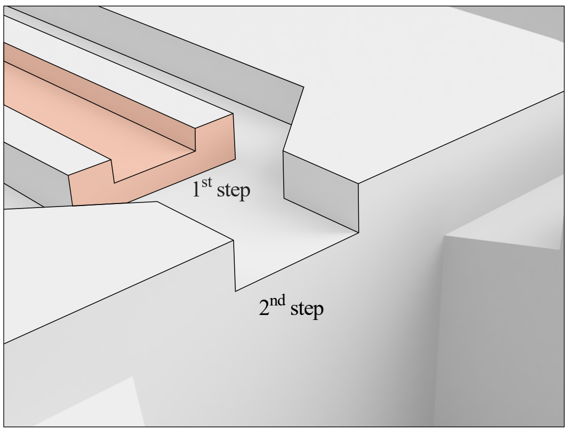

Figure S3 The schematic illustration of the device with selective surface modification for W/O/W double emulsion. The area labelled with orange (channel of the core phase and the vertical wall on which the core droplet generating nozzle is) must be rendered hydrophobic and the rest of the device should be hydrophilic.

References

[1] J. Lim, O. Caen, J. Vrignon, M. Konrad, V. Taly and J.-C. Baret, *Biomicrofluidics*, 2015, **9**, 034101.

[2] D. Nečas and P. Klapetek, *Open Physics*, 2012, **10**, 181–188.