

Supplementary Information

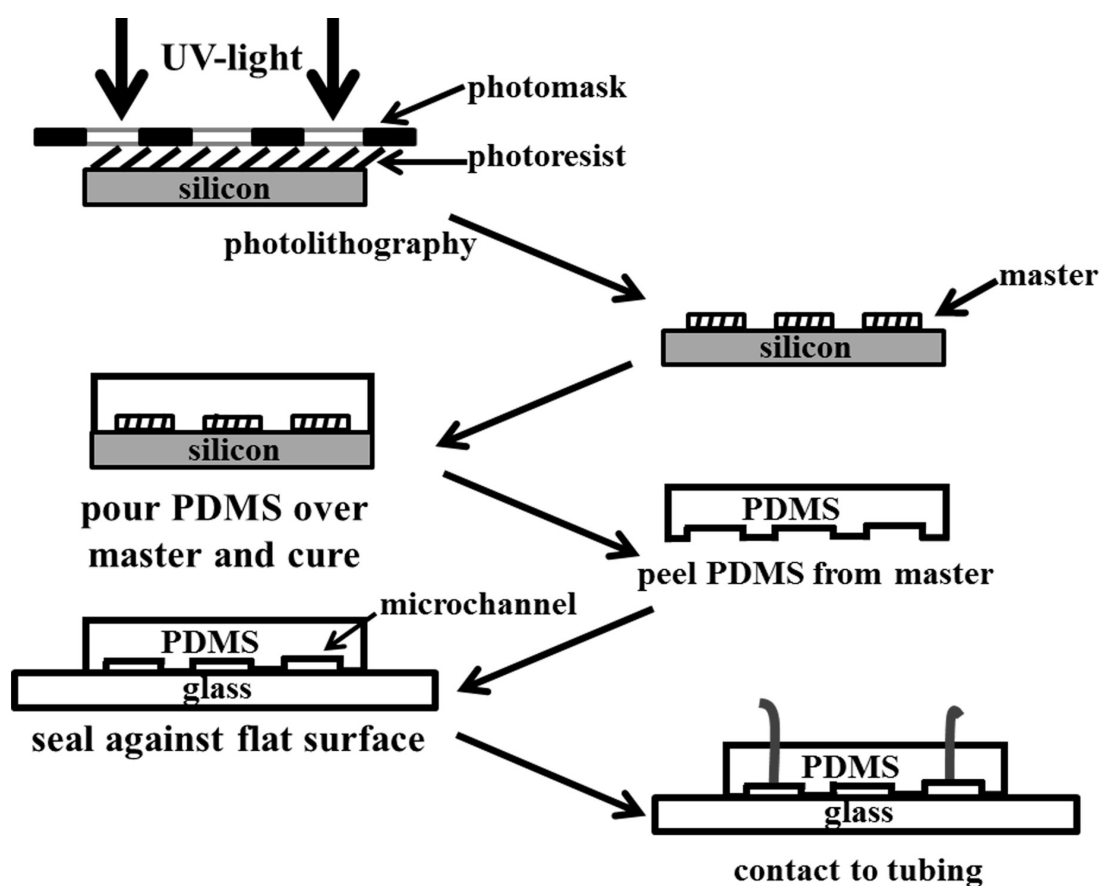


Figure S1. Schematic illustration of the poly(dimethylsiloxane) (PDMS)-based microfluidic device fabrication process.

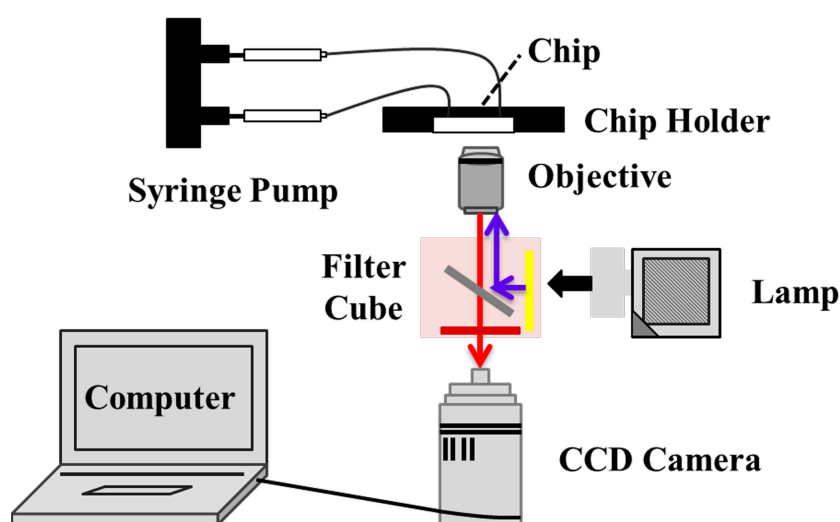


Figure S2. Schematic illustration of the experiment setup.

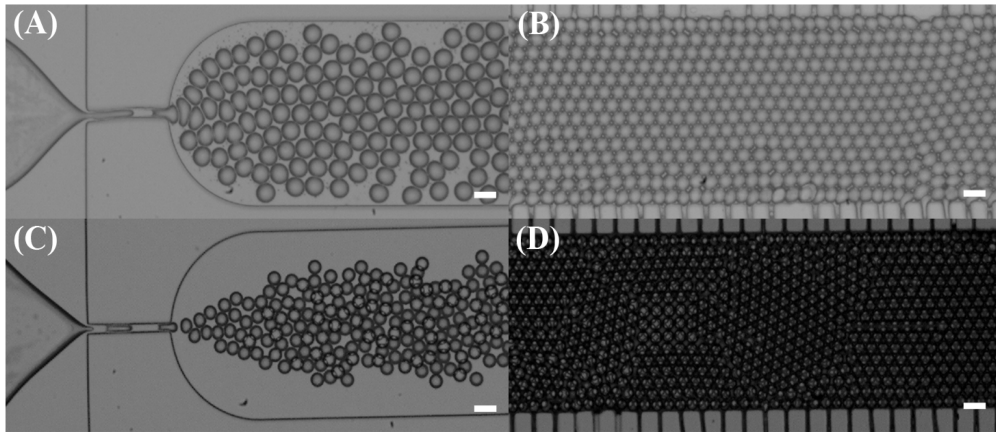


Figure S3. Optical microscopic images of the microdroplets fabrication and assembled 2D and 3D configurations in the PDMS-based flow-focusing microfluidic device with the characteristic size of $w_f = 25 \mu\text{m}$, $W_a = 400 \mu\text{m}$ and $d_c = 40 \mu\text{m}$. (A) and (C) are the images of droplets generation at the flow-focusing area; (B) and (D) are the microdroplets assembled arrangements in single-layer and double-layer, respectively. Q_o and Q_w were 50 and $10 \mu\text{L}\cdot\text{h}^{-1}$ for (A) and (B), 60 and $20 \mu\text{L}\cdot\text{h}^{-1}$ for (C) and (D). In (A) and (B), the oil phase was mineral oil with 21.2 wt % ABIL EM 90, and the aqueous phase was 9 wt % NIPAM, 2.3 wt % MBA and 2.0 wt % I 2959 dissolved in DI water. In (C) and (D) the oil phase was mineral oil with 11.2 wt % ABIL EM 90, and the aqueous phase consisted of 6 wt % NIPAM, 2.0 wt % MBA and 0.5 wt % I 2959. Scale bars are $50 \mu\text{m}$.

Movie S1: Microfluidic induced generation and assembly of microdroplets into controllable close-packed configurations. Q_o and Q_w were 600 and $100 \mu\text{L}\cdot\text{h}^{-1}$. Oil phase and Water phase were hexadecane with 2.7% (w/v) Span80 and DI water, respectively. The microfluidic device with the characteristic size of $w_f = 25 \mu\text{m}$, $W_a = 400 \mu\text{m}$ and $d_c = 40 \mu\text{m}$ was used. (See the separate Movie S1.mpg).

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