

## Electronic Supplementary Information

### 3D Printing of Monolithic Gravity-Assisted Step-Emulsification Device for Scalable Production of High Viscosity Emulsion Droplets

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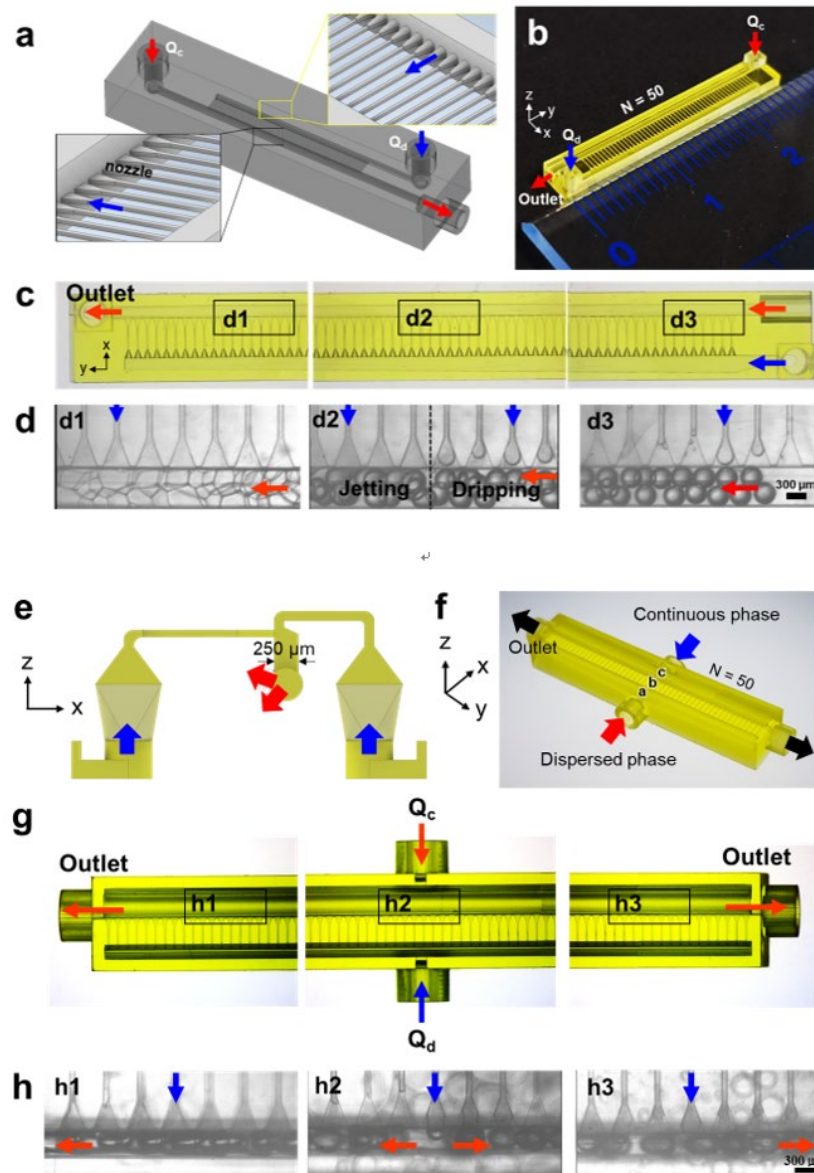
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#### Keywords

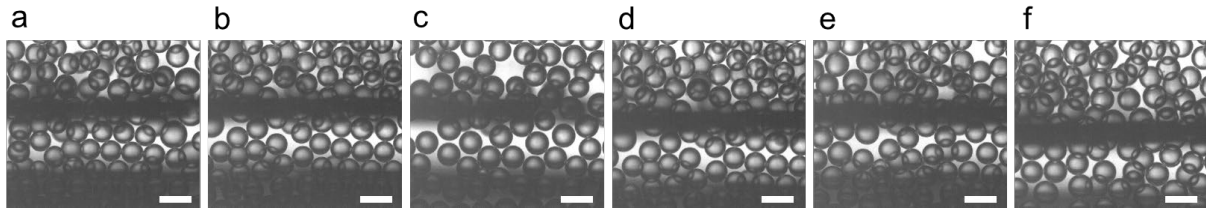
Step-emulsification; 3D-printing; Emulsions; Microfluidics; Microparticle; Parallelization

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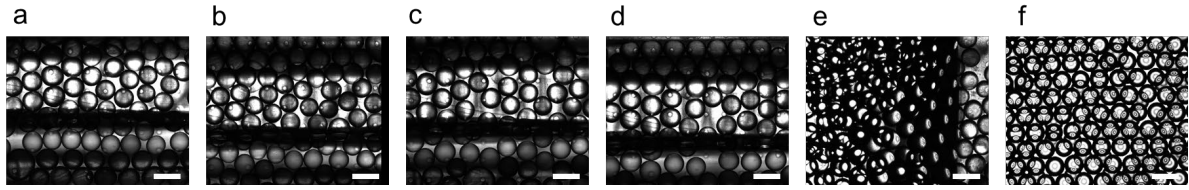
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**Fig. S1.** (a) Schematics illustrating the 3D-printed step-emulsification device with one horizontal outlet. (b) Photograph and the (c) magnified photograph of the device. (d) Optical micrograph showing the droplet formation behavior at different drop generator nozzles (d1-d3). (e) Schematics illustrating the 3D-printed step-emulsification device with two horizontal outlets. (f) Photograph and the (g) magnified photograph of the device. (h) Optical micrograph showing the droplet formation behavior at different drop generator nozzles (h1-h3). The continuous phase is 2 wt% Span80 in mineral oil and the dispersed phase is DI water.



**Fig. S2.** Series of micrographs showing the monodisperse W/O emulsion droplets produced from the 3D-PSD at different flow rates. The average diameter of the droplets ( $d$ ) are (a) 193.5  $\mu\text{m}$  (CV = 3.2%), (b) 195.4  $\mu\text{m}$  (CV = 2.1%), (c) 198.4  $\mu\text{m}$  (CV = 2.6%), (d) 194.1  $\mu\text{m}$  (CV = 3.2%), (e) 195.8  $\mu\text{m}$  (CV = 2.5%), and (f) 193.4  $\mu\text{m}$  (CV = 2.9%), which are operated at ( $Q_d = 5 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 10 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 20 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 30 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 40 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), and ( $Q_d = 50 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), respectively. Scale bar presents 300  $\mu\text{m}$ .



**Fig. S3.** Series of micrographs showing the monodisperse O/W emulsion droplets produced from the hydrophilically modified 3D-PSD at different flow rates. The average diameter of the droplets ( $d$ ) are (a) 193.5  $\mu\text{m}$  (CV = 3.2%), (b) 195.4  $\mu\text{m}$  (CV = 2.1%), (c) 198.4  $\mu\text{m}$  (CV = 2.6%), (d) 194.1  $\mu\text{m}$  (CV = 3.2%), (e) 195.8  $\mu\text{m}$  (CV = 2.5%), and (f) 193.4  $\mu\text{m}$  (CV = 2.9%), which are operated at ( $Q_d = 5 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 10 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 20 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 30 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), ( $Q_d = 40 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), and ( $Q_d = 50 \text{ mL h}^{-1}$ ,  $Q_c = 50 \text{ mL h}^{-1}$ ), respectively. The continuous phase is 2 wt% PVA aqueous solution and the dispersed phase is DCM. Scale bar presents 300  $\mu\text{m}$ .

**Table S1.** Physical properties and parameters used for the CFD simulation.

Notation	Description	Value
$\rho_c$	Density of the continuous phase	770 kg/m <sup>3</sup>
$\rho_d$	Density of the dispersed phase	998 kg/m <sup>3</sup>
$\mu_c$	Dynamic viscosity of the continuous phase	3.0 x 10 <sup>-3</sup> Pa·s
$\mu_d$	Dynamic viscosity of the dispersed phase	8.9 x 10 <sup>-4</sup> Pa·s
$^a\sigma$	Interfacial tension between the continuous and dispersed phases	0.0050 N/m
$g$	Gravitational acceleration	9.8 m/s <sup>2</sup>
$\theta_{dc}$	Contact angle between the continuous and dispersed phases on the channel wall	150°