Controlled Formation of All-Aqueous Janus Droplets by Liquid-Liquid Phase Separation of an Aqueous Three-Phase System

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The supporting information contains the following components:

1) A typical movie showing the evolution of an initial single-phase droplet into a Janus droplet after phase separation. The scale bar is 300 μm. **(Movie S1)**

2) The table containing the osmolality of aqueous drop phases with different solute concentrations. (**Table S1**)

3) The figure illustrating the diameter of the jets under different outer fluid flow rates in glass-capillary microfluidic devices. (**Figure S1**)

4) A typical movie showing the droplet generation with perturbation in glass-capillary microfluidics and the obtained Janus droplets. The scale bar is 200 μm. **(Movie S2)**

5) The table showing the density of each phase of phase-separated DEX, PEtOx and PEG with three different compositions.

6) The figure illustrating the partitioning behavior of 50 wt% DOPC-50 wt% DOPS liposomes in the Janus droplets.

1) See the uploaded video content.

2) The table illustrating the osmolality of the drop phases with different compositions

Table S1. Osmolality of the aqueous solutions*

NO.	DEX T10 (wt%)	PEtOx (wt%)	Osmolality (mOsm/kg)
1	8	2	54±1
2	3	6	57±2
3	5	5	73±1

NO.	DEX T500 (wt%)	PEtOx (wt%)	Osmolality (mOsm/kg)
1	3	5	32±1
2	4	2	20±1

* Average values and standard errors are obtained from 3 independent measurements.

3) The figure shows the diameter of the jets under different outer fluid flow rates in a glass-capillary microfluidic device at constant inner fluid flow rate Q_{in} without perturbation.
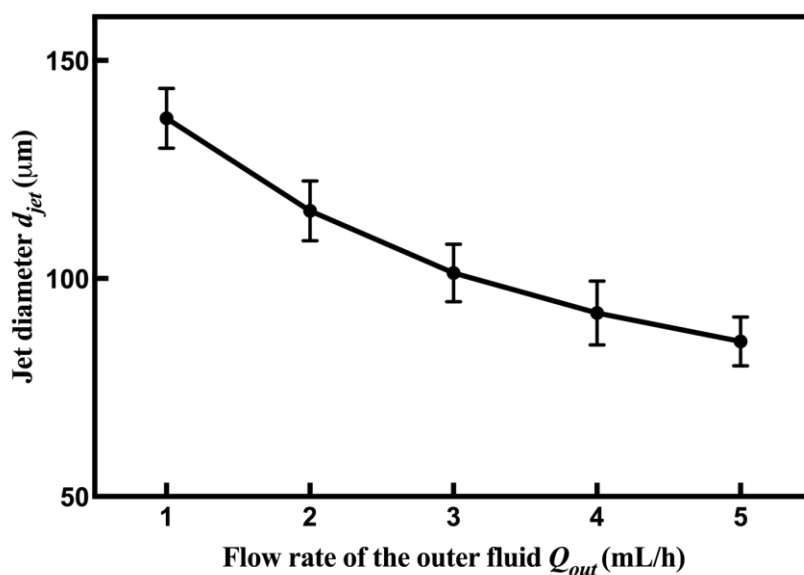

Figure S1. Plot of the diameter of the inner jets d_{jet} as a function of the outer fluid flow rates Q_{out}, ranging from 1.0 to 5.0 mL/h. $Q_{in} = 20$ μL/h.

4) See the uploaded video content.

5) The table showing the density of each phase of the phase-separated DEX, PEtOx and PEG with three different compositions.

Table S2. Density of three phases from the phase-separated A3PSs with different compositions*

Composition of A3PSs	Phase	Density (g/cm³)
10 wt% DEX-10 wt% PEtOx-2 wt% PEG	PEG-rich phase	1.039
	PEtOx-rich phase	1.040
	DEX-rich phase	1.090
10 wt% DEX-15 wt% PEtOx-2 wt% PEG	PEG-rich phase	1.041
	PEtOx-rich phase	1.047
	DEX-rich phase	1.110
8 wt% DEX-12 wt% PEtOx-3 wt% PEG	PEG-rich phase	1.037
	PEtOx-rich phase	1.042
	DEX-rich phase	1.105

* Average values are obtained from 3 independent measurements.

6) The figure illustrating the partitioning behavior of 50 wt% DOPC-50 wt% DOPS liposomes in the Janus droplets
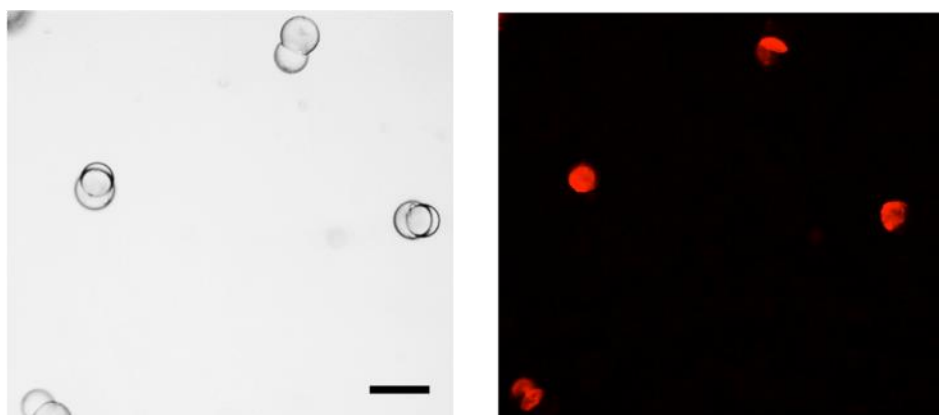

Figure S2. The microscope and fluorescence microscope images showing the partitioning of 50 wt% DOPC-50 wt% DOPS Rhodamine-labelled (red) liposomes in the Janus droplets. The 50 wt% DOPC-50 wt% DOPS liposomes mainly partition into the DEX-rich phase of the Janus droplets. The scale bar is 200 μm.