

(Supporting Information)

**Controlled production of size-tunable Janus droplets for submicron  
particle synthesis using an electrospray microfluidic chip**

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### (1) Electrospray microfluidic chip fabrication

The microfluidic chip comprises a PDMS layer and a conductive glass layer. The PDMS channel layer was fabricated using soft lithography techniques.<sup>1</sup> SU-8 2050 (Micro-Chem, Newton, MA, USA) was used to make a master mold of channel patterns. The PDMS replica was prepared using the mold. The electrodes on the conductive glass layer were fabricated on an indium tin oxide (ITO) glass substrate by using photolithography and chemical wet etching. AZ P4620 photoresist (AZ Electronic Materials K. K., Japan) was used to form a sacrifice layer with the patterns of electrodes. The glass substrate was treated with 37.5% hydrochloric acid to etch off the undesired ITO, and the ITO electrodes were formed after removal of the photoresist. The PDMS layer was bonded to the conductive glass via air plasma treatment (Model PDC-32G, Harrick Scientific Co., USA). The electrodes were connected to the high voltage source using copper wires fixed onto the ITO electrode surface by conductive glue.

### (2) Data of diameters and coefficients of variation of various droplets

**Table S1** Droplet diameters and coefficient of variation values.

Voltage (V)	DI water droplets		Ink labelled Janus droplets		Alginate droplets		PLGA droplets	
	Diameter (μm)	CV (%)	Diameter (μm)	CV (%)	Diameter (μm)	CV (%)	Diameter (μm)	CV (%)
0	89.8	1.84	135	0.733	119	1.97	77.7	4.70
400	86.4	2.49	133	1.35	114	1.31	68.0	2.42
800	62.7	1.91	120	1.06	106	1.68	50.1	4.38
1200	54.4	2.72	115	1.71	96.1	2.20	37.4	3.24
1600	4.03	3.33	3.14	8.54	2.28	8.93	35.2	4.73
1600*			85.3	4.40	69.3	3.07		

\* The last row presents the diameters and CVs of the mother droplets in the electrospray mode.

### (3) Post-processing and characterization parameters of Janus particles

After collected into a modified centrifuge tube, the particles were pipetted into another container along with the dispersing liquid, and stirred for 4 h to remove the organic solvent. Then the particles were washed with DI water for three times by means of centrifugation at 5000 rpm for 5 min each time, and at last dispersed in DI water for further characterization. The particles on a silicon substrate were coated with a thin gold film by vacuum evaporation coating, and characterized by scanning electron microscopy (the EHT was set at 20 kV). For the observation of particles under a fluorescent microscope, 10 μL particle dispersion was dripped on a clean glass slide, and dried in a vacuum oven, leaving particles on the slide surface. Laser excitation modules with the wavelengths of 495 nm and 550 nm were used for producing green and red fluorescent respectively.

**Movie S1:** Electrospray of pure water to form tiny droplets under an electric field with the voltage set at 1600 V.

**Movie S2:** Formation of ink-doped Janus droplets without an electric field.

**Movie S3:** Electrospray of an ink-doped Janus mother droplet to form tiny droplets at 1600 V.

### References

1 Y. N. Xia and G. M. Whitesides, *Annu. Rev. Mater. Sci.*, 1998, **28**, 153.