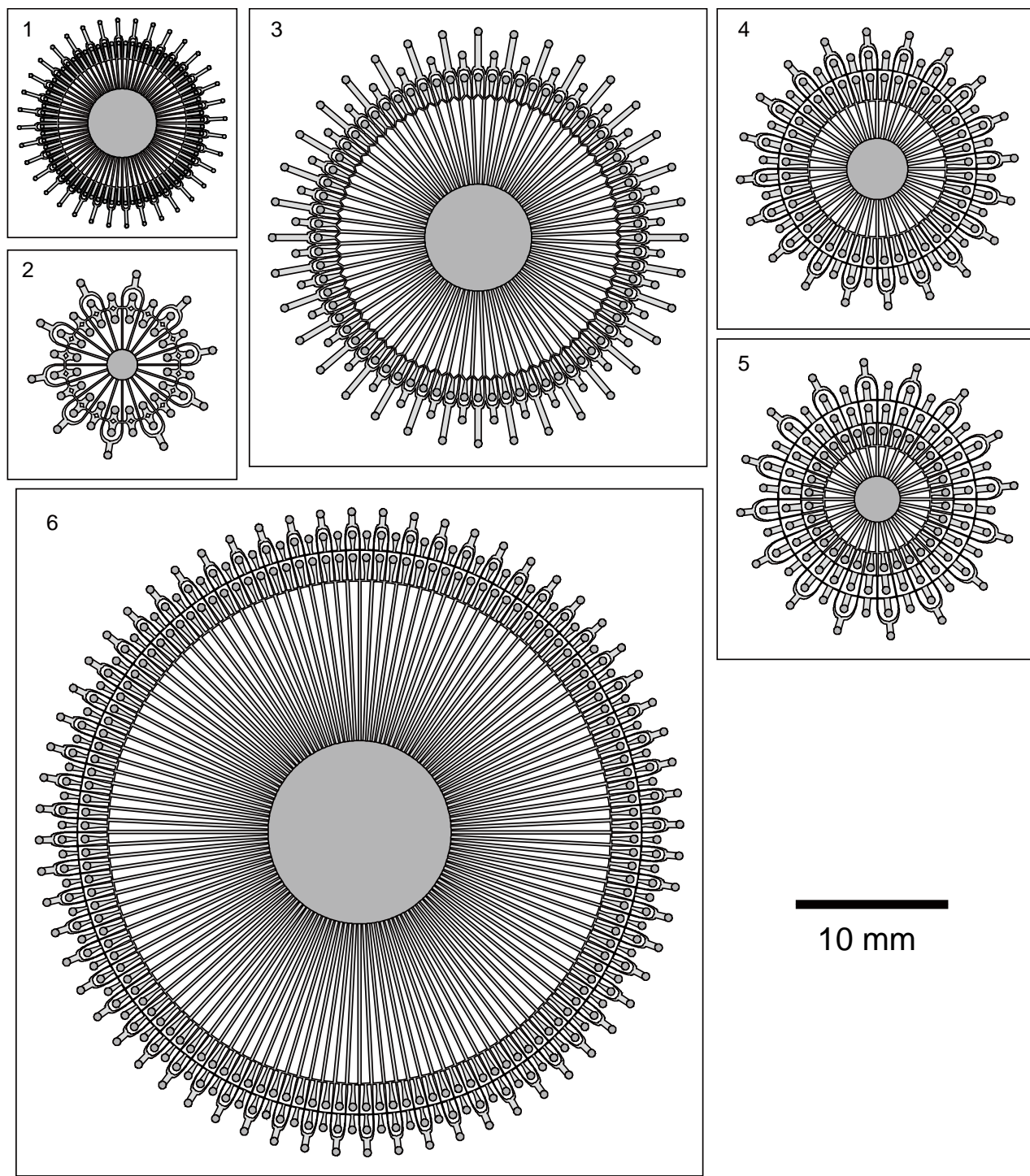


## Electronic Supplementary Information

### 1. Microfluidic chips

The six microfluidic chips used in the experiments are shown schematically in Fig. S1. The height of the microchannels is uniformly 100  $\mu\text{m}$ . Details of each chip are listed in Table S1 and S2.



**Figure S1.** Schematic illustrations of the six microfluidic chips used.

**Table S1.** Detailed information on the microfluidic chips.

	Chip size (mm <sup>2</sup> )	Size of input holes (μm)	Number of input holes	Number of infused fluids	Type of emulsions	Number of final droplet generators	Related figures in the text
1	15 × 15	250	108	2	single	144	Fig. 1, 5
2	15 × 15	500	50	3	Janus	40	Fig. 7
3	30 × 30	500	144	3	Janus	72	Figs. 1, 6
4	21 × 21	500	100	3	double	40	Fig. 8
5	21 × 21	500	112	4	triple	32	Figs. 1, 9
6	45 × 45	500	320	3	double	128	Fig. 8

**Table S2.** Widths of microchannels around junctions.

#	Innermost junctions		Middle junctions		Outermost junctions	
	Output (μm)	Input (μm)	Output (μm)	Input (μm)	Output (μm)	Input (μm)
1	100	100	-	-	-	-
2	100	100	-	-	-	-
3	200	100	-	-	-	-
4	200	100	100	50	-	-
5	200	100	100	50	50	50
6	200	100	100	50	-	-

## 2. Supporting modules with annular channels

Details of supporting modules with annular channels are listed in Table S3.

**Table S3.** Supporting modules with annular channels

#	Chip #	Module size (mm <sup>3</sup> )	Number of annulus	1 <sup>st</sup> annulus (outer)		2 <sup>nd</sup> annulus		3 <sup>rd</sup> annulus		4 <sup>th</sup> annulus (inner)	
				i.d. (mm)	o.d. (mm)	i.d. (mm)	o.d. (mm)	i.d. (mm)	o.d. (mm)	i.d. (mm)	o.d. (mm)
1	1	36 × 36 × 24	2	13	14	10	11	-	-	-	-
2	2	36 × 36 × 32	3	11.5	12.5	8.5	9.5	5.5	6.5	-	-
3	3	48 × 48 × 32	3	26.5	27.5	23.5	24.5	20.5	21.5	-	-
4	4	40 × 40 × 32	3	17.5	18.5	14.5	15.5	11.5	12.5	-	-
5	5	40 × 40 × 40	4							8.5	9.5
6	6	64 × 64 × 32	3	41.5	42.5	38.5	39.5	35.5	36.5	-	-

### 3. Production rates

Table S4 lists the production rates demonstrated in this study. These flow rates were not the maximized flow rates in the microfluidic modules we used.

**Table S4.** Production rates demonstrated in this study.

Chip#	Type of emulsions	Number of final droplet generators	Production rate	Related figures in the text
1	single	144	180 mL h <sup>-1</sup>	Fig. 1, 5
2	Janus (immiscible)	40	20 mL h <sup>-1</sup> (10+10)	Fig. 7
3	Janus (miscible)	72	72 mL h <sup>-1</sup> (36+36)	Fig. 6
4	double	40	25 mL h <sup>-1</sup> (5+20, two-step) 20 mL h <sup>-1</sup> (10+10, one-step)	Figs. 8b-g Fig. 8h
5	triple	32	31 mL h <sup>-1</sup> (5+6+20, three-step)	Fig. 9
6	double	128	75 mLh <sup>-1</sup> (15+60, two-step)	Fig. 8i

### Supplemental Movie Captions:

1\_cross144.mpg: Movie clip of the formation of O/W emulsion droplets in the module having 144 cross-flowing MFDGs, recorded at 6000 fps. Disperse phase flow rate ( $Q_d$ ) was  $180.0 \text{ mL h}^{-1}$  and continuous phase flow rate ( $Q_c$ ) was  $270.0 \text{ mL h}^{-1}$ .

2\_sheath72-Janus.mpg: Movie clip of the formation of Janus droplets of two miscible segments in the module having 72  $\psi$ -shaped MFDGs, recorded at 4000 fps. Flow rates were  $Q_d = 72.0 \text{ mL h}^{-1}$  ( $36.0 \text{ mL h}^{-1} \times 2$ ) and  $Q_c = 576 \text{ mL h}^{-1}$ .

3\_cross40-Janus.mpg: Movie clip of the formation of Janus droplets in the module having 40 cross-flowing MFDGs, recorded at 3000 fps. Flow rates were  $Q_d = 20.0 \text{ mL h}^{-1}$  ( $10.0 \text{ mL h}^{-1} \times 2$ ) and  $Q_c = 40.0 \text{ mL h}^{-1}$ .

4\_40-double-twostep-single.mpg: Movie clip of the two-step formation of single core O/O/W double emulsion droplets in the module having 40 sets of two-consecutive cross-shaped MFDGs, recorded at 3000 fps. Flow rates were  $Q_d = 25.0 \text{ mL h}^{-1}$  (core  $5.0 \text{ mL h}^{-1}$  + shell  $20.0 \text{ mL h}^{-1}$ ) and  $Q_c = 500.0 \text{ mL h}^{-1}$ .

5\_40-double-twostep-dual.mpg: Movie clip of the two-step formation of dual core O/O/W double emulsion droplets in the module having 40 sets of two-consecutive cross-shaped MFDGs, recorded at 3000 fps. Flow rates were  $Q_d = 25.0 \text{ mL h}^{-1}$  (core  $5.0 \text{ mL h}^{-1}$  + shell  $20.0 \text{ mL h}^{-1}$ ) and  $Q_c = 250.0 \text{ mL h}^{-1}$ .

6\_40-double-onestep.mpg: Movie clip of the one-step formation of single core O/O/W double emulsion droplets in the module having 40 sets of two-consecutive cross-shaped MFDGs, recorded at 1000 fps. Flow rates were  $Q_d = 20.0 \text{ mL h}^{-1}$  (core  $10.0 \text{ mL h}^{-1}$  + shell  $10.0 \text{ mL h}^{-1}$ ) and  $Q_c = 150.0 \text{ mL h}^{-1}$ .

7\_128-double-twostep.mpg: Movie clip of the two-step formation of O/O/W double emulsion droplets in the module having 128 sets of two-consecutive cross-shaped MFDGs, recorded at 2000 fps. Flow rates were  $Q_d = 75.0 \text{ mL h}^{-1}$  (core  $15.0 \text{ mL h}^{-1}$  + shell  $60.0 \text{ mL h}^{-1}$ ) and  $Q_c = 600.0 \text{ mL h}^{-1}$ .

All movies are prepared in MPEG-1 format.