Electronic Supplementary Material (ESI) for RSC Advances

Supporting Information

Microfluidic Preparation and Structure Evolution of Double Emulsions with Two-phase Cores

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a) Formation rules of double emulsion with single phase cores

To research to our microfluidic device clearly, we first drive only one phase into a pinhead while the other one was sealed to produce W/O/W double emulsion. We obtained monodispersed double emulsions with accurate control in the number and the size of the inner droplet.

We first explored the influence of inner flow rates to the size of inner droplets. We changed inner flow rates from 3 μL/min to 11 μL/min while kept middle and outer flow rates in 60 μL/min and 100 μL/min The relationship is shown in Figure S1.

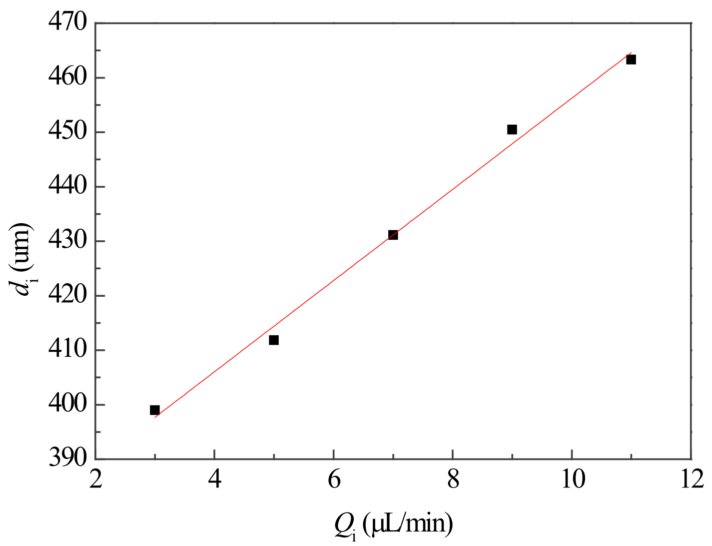

Figure S1. The relationship of inner droplets size and inner flow rates. Q_i means inner flow rates. d_i means the size of inner droplets

Then we explored the influence of middle flow rates to the size of inner droplets. We changed middle flow rates from 30 μL/min to 160 μL/min while keep inner and outer flow rates in 3μL/min and 100 μL/min. The relationship is shown in Figure S2.

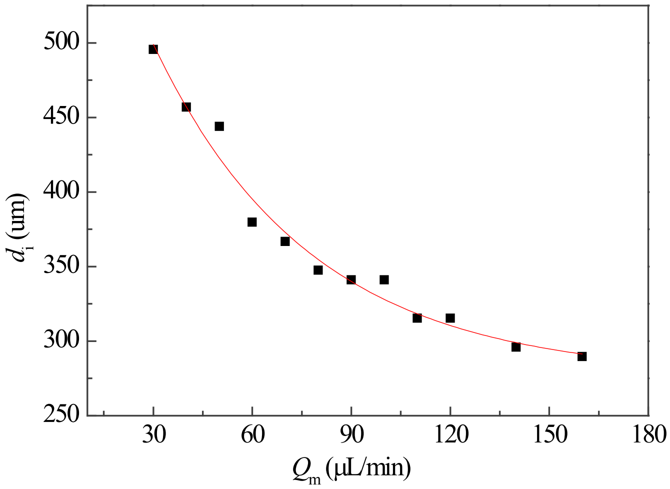

Figure S2. The relationship of inner droplets size and middle flow rates. Q_m means inner flow rates.

Furthermore, we realized good control in the number of inner droplets by changing inner flows. Figure S3 and Figure S4 show different number of inner droples encapsulated in the same level while changing inner droplets from 3 μL/min to11 μL/min.

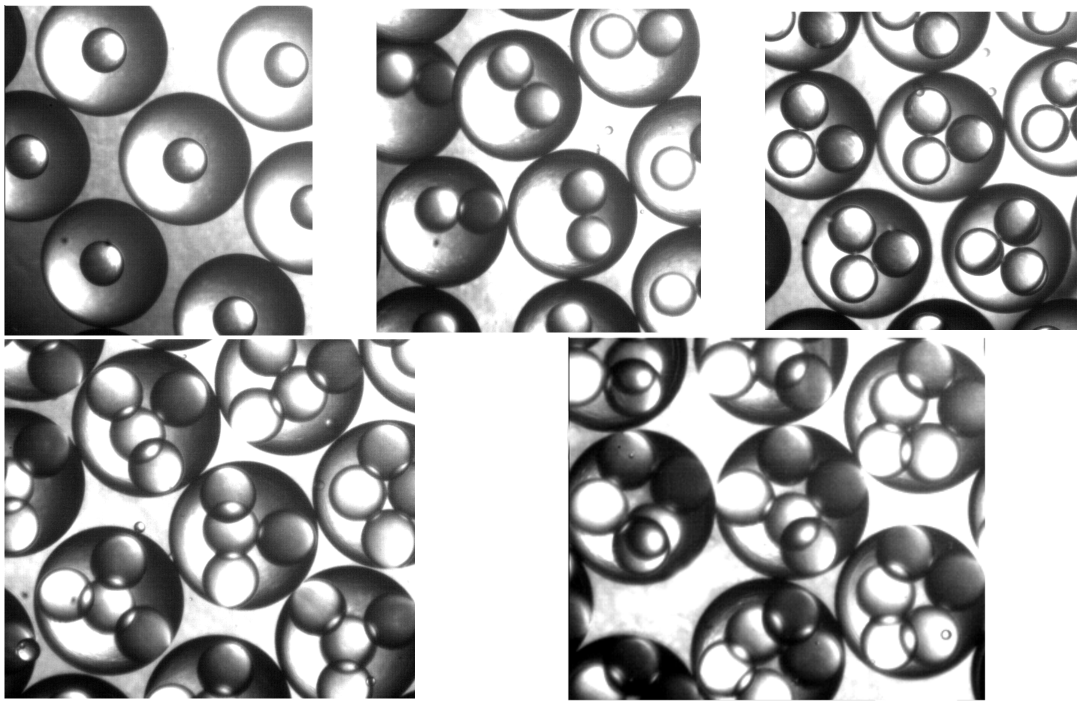

Figure S3. Different number of inner droplets encapsulated in the same level. The middle and outer flow rates are 60 μL/min and 100 μL/min. Inner flow rates are 3, 5, 7, 9, 11μL/min, respectively.

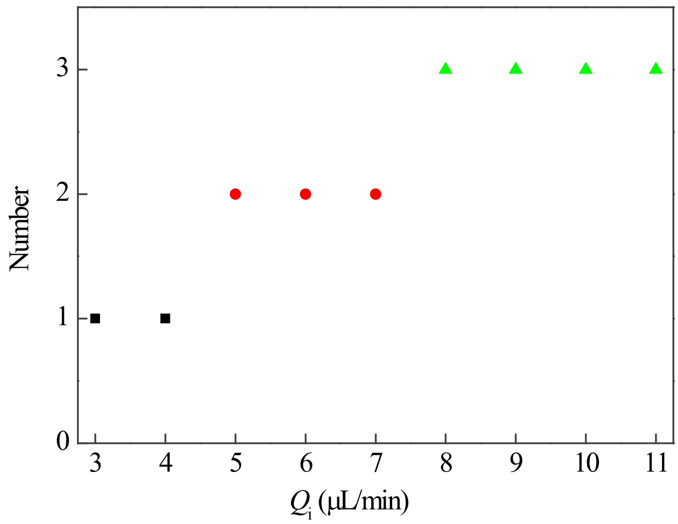

Figure S4. The relationship of the number of inner droplets and inner flow rates.

b) **The interfacial tensions measured using the pendent drop technique (OCAH200, DataPhysics Instruments GmbH, Germany**)

Table S1 The interfacial tensions and spreading coefficients of different systems

	Phase 1	**Phase 2**	**Phase 3**	δ_{12}	δ_{13}	δ_{23}
1	air	TPGDA	tetradecane	25.15	26.98	0.78
2	Water[a]	Silicon oil[a]	air	13.90	35.05	0.22
3	Silicon oil[b]	TPGDA	mineral oil	10.64	1.64	0.37
4	air	Water[a]	TPGDA	35.05	25.15	5.40
5	Silicon oil[a]	Water[a]	TPGDA	13.9	1.18	5.4
6	air	TPGDA	Silicon oil[b]	25.15	13.8	10.64
7	tetradecane	Water[a]	TPGDA	5.88	0.78	5.40
8	Silicon oil[a]	Water[a]	HDDA	13.90	2.98	11.49
9	Water[b]	octanol	air	3.70	27.20	29.10

Water[a] here means water+1%PVA+2%SDS. Water[b] means Water+0.5% Tween80. Silicon oil[a] refers to silicon oil (50cp) with Dowcorning 749 (10 wt.%). Silicon oil[b] refers to silicon oil (100 cp) without surfactant.