

## Supporting information

### **A region-selective modified capillary microfluidic device for fabricating water-oil Janus droplets and hydrophilic-hydrophobic anisotropic microparticles**

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The following are some of our analysis about the prediction of different Janus structure in the manuscript and supporting information.

Torza and Mason theory<sup>1, 2</sup> used spreading coefficient ( $S_i$ ) to predict the morphology of three fluids, as follows,

$S_i = \sigma_{jk} - (\sigma_{ij} + \sigma_{ik})$  ( $i \neq j \neq k = 1, 2, 3$ ),  $\sigma_{jk}$  represent the interfacial tension between j and k fluids.

$S_1 < 0$   $S_2 < 0$   $S_3 > 0$ , engulfing structure

$S_1 < 0$   $S_2 < 0$   $S_3 < 0$ , Janus structure

$S_1 < 0$   $S_2 > 0$   $S_3 < 0$ , separating

Here in our work, the interfacial tensions of three fluids are diverse because we use continuous phase with different mass fraction of surfactant. The continuous phase is liquid paraffin in which the mass fraction of the surfactant Span80 is adjustable. The two inner phases are ETPTA and Water. The relationship between interfacial tensions of fluids and the mass fraction of Span80 in continuous phase is shown in Fig. 6d in the manuscript.

The interfacial tension between ETPTA and Water is not shown in the picture because it doesn't change through the Span 80. ( $\gamma_{EW} = 6.13 \text{ mN/m}$ )

In our manuscript,  $\gamma_w$  represents the interfacial tension between water and liquid paraffin,  $\gamma_E$  represents the interfacial tension between ETPTA and liquid paraffin, and  $\gamma_{EW}$  represents the interfacial tension between water and ETPTA.

To make the interfacial tensions more clearly, we change picture above into table, when the mass fraction of Span 80 changed from 0 wt.% to 2.0 wt.%, the interfacial tensions are shown in Table 1 in the manuscript

Then we use the Torza and Mason theory to predict the morphologies, for  $S_i = \sigma_{jk} - (\sigma_{ij} + \sigma_{ik})$  ( $i \neq j \neq k = 1, 2, 3$ ), continuous phase represents 2, water represents 1 and ETPTA represents 3. Thus,  $\gamma_{EW} = \sigma_{13}$ ,  $\gamma_E = \sigma_{23}$ ,  $\gamma_W = \sigma_{12}$ . Then let's take several data above to predict their morphology and compare with the Torza and Mason theory.

For examples,

### #1

The Span80 fraction is 0w%,  $\gamma_W = 17.19$  (mN/m),  $\gamma_E = 3.63$ (mN/m),

$$\gamma_{EW} = 6.13 \text{ (mN/m)}$$

The spreading coefficient are:

$$S_1 = \sigma_{23} - (\sigma_{12} + \sigma_{13}) = 3.63 - (17.19 + 6.13) = -19.69 < 0$$

$$S_2 = \sigma_{13} - (\sigma_{12} + \sigma_{23}) = 6.13 - (17.19 + 3.63) = -14.69 < 0$$

$$S_3 = \sigma_{12} - (\sigma_{13} + \sigma_{23}) = 17.19 - (6.13 + 3.63) = 7.43 > 0$$

According to the Torza and Mason theory, it should form engulfing structure. In our work, it formed engulfing structure. The microscopic photo is shown in down (as shown in Fig. 6a). It clearly shows that it forms engulfing structure.

### #2

The Span80 fraction is 0.5w%,  $\gamma_W = 4.12$ (mN/m),  $\gamma_E = 3.53$ (mN/m),

$$\gamma_{EW} = 6.13 \text{ (mN/m)}$$

The spreading coefficient are:

$$S_1 = \sigma_{23} - (\sigma_{12} + \sigma_{13}) = 3.53 - (4.12 + 6.13) = -6.72 < 0$$

$$S_2 = \sigma_{13} - (\sigma_{12} + \sigma_{23}) = 6.13 - (4.12 + 3.53) = -5.01 < 0$$

$$S_3 = \sigma_{12} - (\sigma_{13} + \sigma_{23}) = 4.12 - (6.13 + 3.53) = -5.54 < 0$$

According to the Torza and Mason theory, it should form Janus structure . In our work, it formed Janus structure. In our manuscript, the microscopic photo show the structure in 0.4 wt.%. (as shown in Fig. 6b). It clearly shows that it forms Janus structure.

### #3

The Span80 fraction is 1w%,  $\gamma_w = 3.77(mN/m)$ ,  $\gamma_E = 3.48(mN/m)$ ,

$$\gamma_{EW} = 6.13(mN/m)$$

$$S_1 = \sigma_{23} - (\sigma_{12} + \sigma_{13}) = 3.48 - (3.77 + 6.13) = -6.42 < 0$$

$$S_2 = \sigma_{13} - (\sigma_{12} + \sigma_{23}) = 6.13 - (3.77 + 3.48) = -1.12 < 0$$

$$S_3 = \sigma_{12} - (\sigma_{13} + \sigma_{23}) = 3.77 - (6.13 + 3.48) = -5.84 < 0$$

According to the Torza and Mason theory, it should form Janus structure. In our manuscript, the microscopic photo show the structure in 0.8 wt.%. ( as shown in Fig. 6c). It clearly shows that it forms Janus structure. If we use all data to calculate the Spreading coefficient, we could obtain as following:

Mass fraction of Span80(w%)	$S_1$	$S_2$	$S_3$
0	-19.69	-14.69	7.43
0.05	-7.55	-2.23	-4.71
0.1	-6.98	-1.7	-5.28
0.15	-6.9	-1.64	-5.36
0.5	-6.72	-1.52	-5.54
1	-6.42	-1.12	-5.84
2	-5.37	-0.05	-6.89

According to the Torza and Mason theory, expect for 0 wt. %, other fluids used in our experiment would form Janus structure.

### References

1. S. Torza and S. G. Mason, *Science*, 1969, **163**, 813.
2. S. Torza and S. G. Mason, *Journal of colloid and interface science*, 1970, **33**, 67-83.