

## Supporting Information

Vesicle-Coacervate-Driven efficient deposition of Sprayed droplet on Hydrophobic Soybean Leaf Surfaces

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## S1. Experimental Section

### 1. Materials

1,6-Dibromohexane ( $C_6H_{12}Br_2$ , 97%), 1,4-Dibromobutane ( $C_4H_8Br_2$ , 98%), 1,8-Dibromooctane ( $C_8H_{16}Br_2$ , 98%), N,N-Dimethyldodecylamine ( $C_{14}H_{31}N$ , >99%), octadecyltrichlorosilane ( $C_{18}H_{37}Cl_3Si$ , >95%, OTS), Sodium dodecyl sulfonate ( $C_{12}H_{25}NaO_3S$ ), Filter paper (medium speed qualitative, 11 cm) was purchased from Xinxing (Hangzhou, China), Milli-Q water (18.2 MU cm) was used in all the experiments.

### 2. Synthesis of 12-n-12 Gemini surfactants

In a 50 mL three-necked flask, N,N-dimethyldodecylamine (1.35 mL, 5 mmol L<sup>-1</sup>) and 1,4-dibromobutane or 1,6-dibromohexane or 1,8-dibromooctane (0.27 mmol L<sup>-1</sup>) were dissolved in 10 mL acetonitrile, and the reaction mixture was heated at 80 °C for 24 h. Then, the reaction mixture was concentrated under vacuum to remove the solvent and the crude product was recrystaled by acetone for three times, and dried at 40 °C under vacuum<sup>1</sup>.

### 3. Sample preparation

The 12-4-12, 12-6-12, 12-8-12, and SDS powders were completely dissolved in water

by stirring or ultrasound treatment to the desired concentration as stock solutions. Subsequently, 12-n-12/SDS mixed solutions with different total concentrations ( $C_T$ ) and molar fractions ( $X_{12-n-12} = C_{12-n-12}/(C_{12-n-12} + C_{SDS})$ ,  $n = 4, 6, 8$ ) were obtained by blending the 12-n-12 and SDS stock solutions in specific volumes, and fully mixed by stirring at 25.0 °C<sup>2</sup>.

#### **4. z-Potential and size measurements**

The z-potential and size measurements of the mixed solutions containing 12-n-12/SDS ( $n = 4, 6, 8$ ) at a constant concentration ( $C_T = 5 \text{ mmol L}^{-1}$ ) and various  $X_{12-n-12}$  values were conducted at  $25.0 \pm 0.1^\circ\text{C}$  using a Malvern Zeta Sizer Nano-ZS instrument. For z-potential assessments, a folded capillary cell (DTS1070) was employed, while a disposable polystyrene cuvette (DTS0012) was used for size distribution measurements. The experiments were repeated at least three times for each sample to ensure the accuracy and reproducibility of the results.

#### **5. Preparation and characterization of hydrophobic surfaces**

In order to achieve a consistent contact angle of  $130^\circ \pm 1^\circ$  on filter paper surfaces, a structured coating protocol was implemented. First, a mixture of 10 mL of n-hexane and 100  $\mu\text{L}$  of pure OTS was prepared in a tube and vigorously shaken for homogeneity prior to application. For the surface modification of solid substrates, the substrates were immersed in the coating solutions overnight. Subsequently, the treated surfaces were carefully extracted from the solution, underwent a thorough washing

process with n-hexane repeated three times to remove excess coating, and were then left to air dry. Contact angle measurements were conducted at multiple locations on the coated surfaces using a contact angle device. This approach ensured a comprehensive evaluation of surface properties, providing reliable and uniform results across the substrate<sup>3</sup>.

## **6. Droplet impact test**

The free-falling velocity of the 12-n-12/SDS (n = 4, 6, 8) water droplets was 1.98 m s<sup>-1</sup>, which was achieved by setting the release height of the droplets from a glass syringe needle as 20 cm. The size of the droplets was about 2 mm, which was fixed by the inner diameter of the needle. Side views of the droplets impacting the hydrophobic surface were recorded using a high-speed camera (FHX-7S) at a frame rate of 1000 frames per second (fps) with a shutter speed of 1/10000 • s. The experiment was repeated at least three times for each type of droplet.

## **7. Dynamic surface tension (DST)**

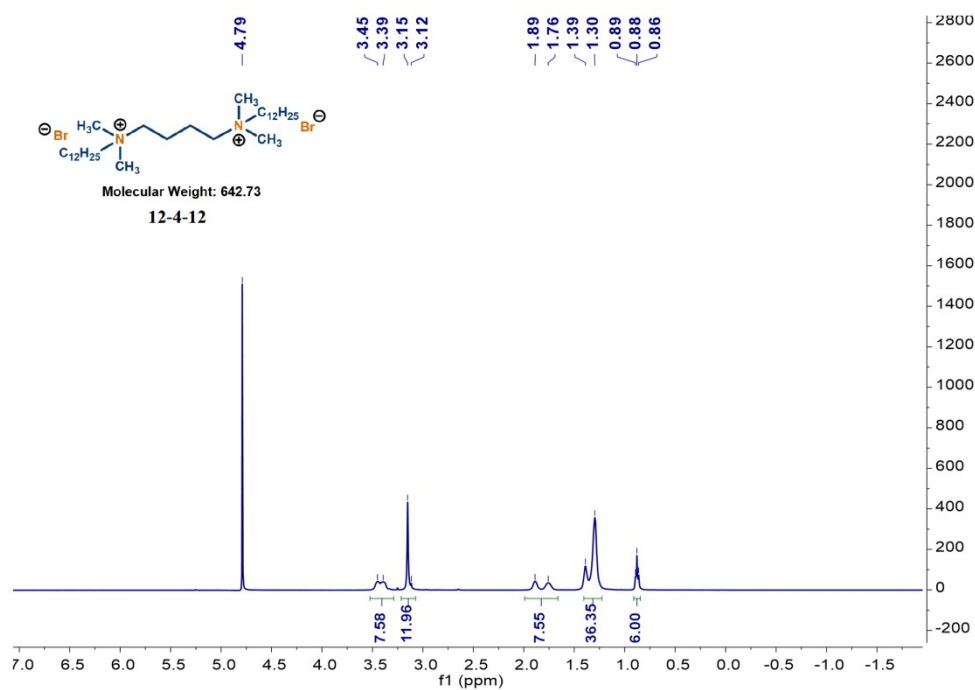
The experiment was meticulously replicated a minimum of three times for each droplet type. Dynamic surface tension measurements for the 12-n-12/SDS (n = 4, 6, 8) mixtures were conducted employing an automatic maximum bubble pressure tensiometer (Krüss-BP-100). The measurement time span ranged from 8 ms to 10 s, utilizing a capillary with a diameter of 0.275 mm. To ensure the robustness and consistency of the findings, the experiment was repeated at least three times for each

individual sample.

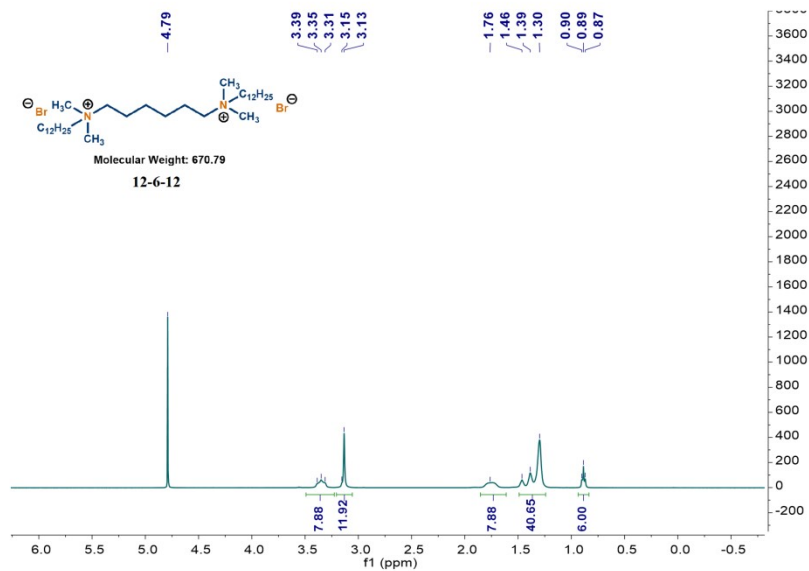
## **8. Mass spectrometry**

Mass spectra were acquired using a Bruker Compact ESI-TOF and collected in positive ion mode or negative ion mode. The relative intensity of the fragment ion peaks ( $m/z$ ) of 12-n-12/SDS indicates the interaction between 12-n-12 and SDS.

## S2. Supplementary Figure

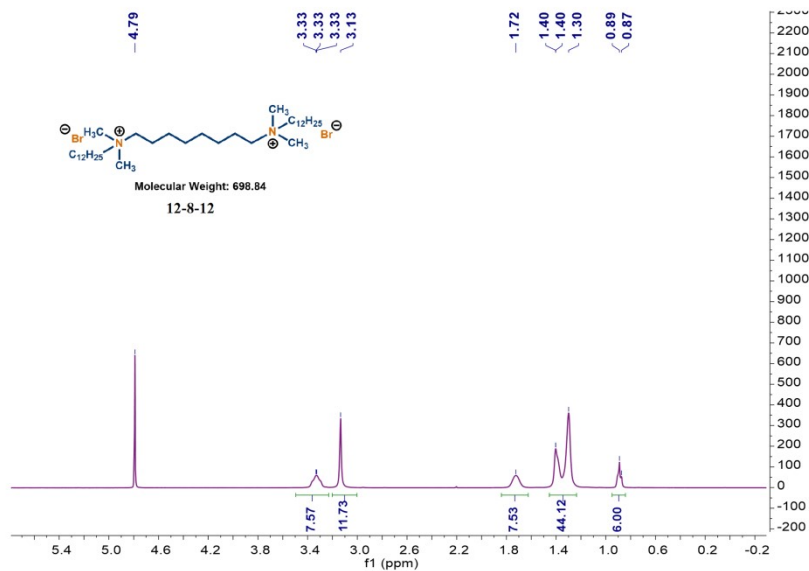


**Fig. S1** <sup>1</sup>H NMR spectra of compound 12-4-12. <sup>1</sup>H NMR (400 MHz, Deuterium Oxide) δ 3.45 (s, 3H), 3.39 (s, 3H), 3.15 (s, 11H), 1.89 (s, 3H), 1.76 (s, 3H), 1.34 (d, J = 37.2 Hz, 37H), 0.88 (t, J = 6.3 Hz, 6H).



**Fig. S2**  $^1\text{H}$  NMR spectra of compound 12-6-12.  $^1\text{H}$  NMR (400 MHz, Deuterium Oxide)  $\delta$  3.35 (t,  $J$  = 14.9 Hz, 8H), 3.13 (s, 12H), 1.76 (s, 8H), 1.46 (s, 4H), 1.39 (s, 8H), 1.30 (s, 30H), 0.89 (t,  $J$  = 6.5 Hz, 6H).

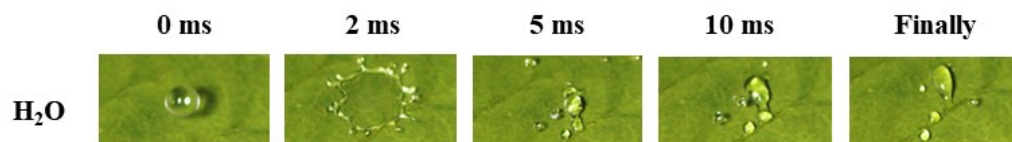




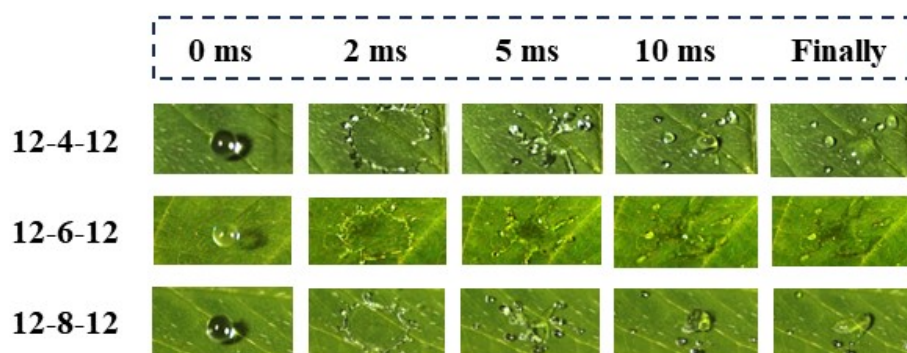
**Fig. S3**  $^1\text{H}$  NMR spectra of compound 12-8-12.  $^1\text{H}$  NMR (400 MHz, Deuterium Oxide)  $\delta$  3.33 (s, 8H), 3.13 (s, 12H), 1.72 (s, 8H), 1.35 (d,  $J = 41.7$  Hz, 46H), 0.88 (d,  $J = 6.6$  Hz, 6H)

Compound	Molecular formula	Calculated (%)			Found (%)		
		C	H	N	C	H	N
12-4-12	$\text{C}_{32}\text{H}_{70}\text{Br}_2\text{N}_2$	59.80	10.98	4.36	59.86	11.04	4.39
12-6-12	$\text{C}_{34}\text{H}_{74}\text{Br}_2\text{N}_2$	60.88	11.12	4.18	60.89	11.15	4.19
12-8-12	$\text{C}_{36}\text{H}_{78}\text{Br}_2\text{N}_2$	61.87	11.25	4.01	61.92	11.31	4.04

**Fig. S4** Organic element analysis of 12-4-12, 12-6-12, and 12-8-12.



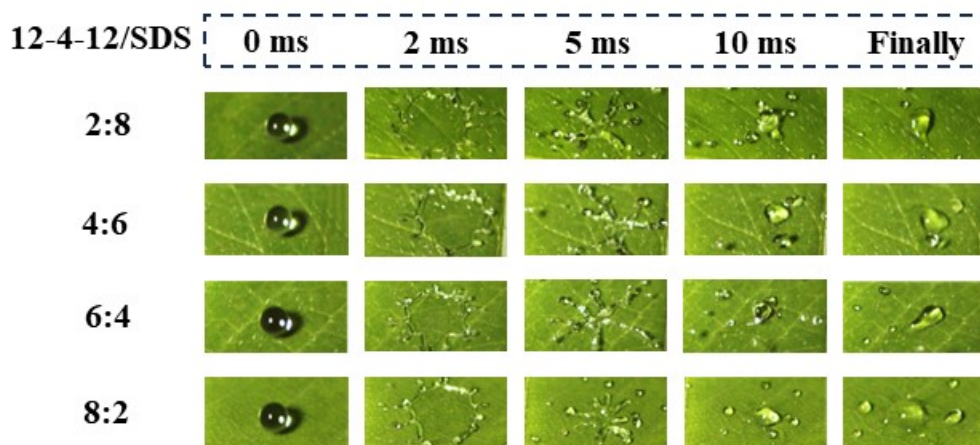
**Fig. S5** The deposition of high-velocity impacting drops on the soybean hydrophobic leaf surface.



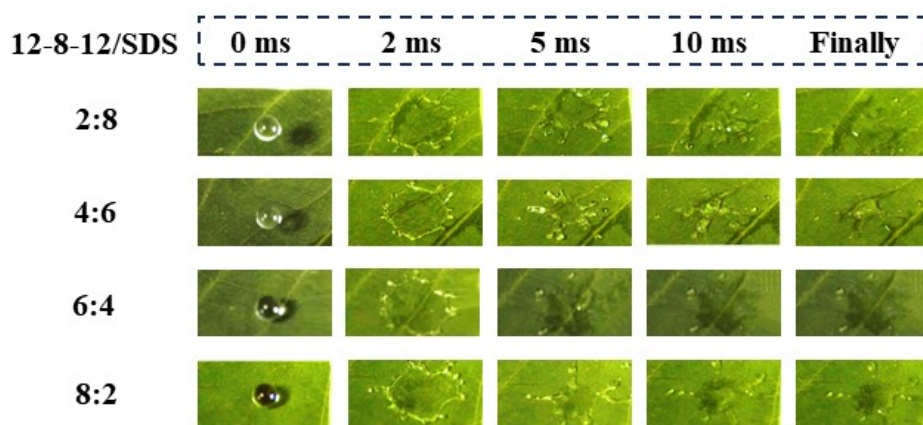
**Fig. S6** The deposition of a high-speed ( $\sim 1.98 \text{ m s}^{-1}$ ) 12-n-12 (n=4, 6, 8) droplets on soybean hydrophobic leaf surface.



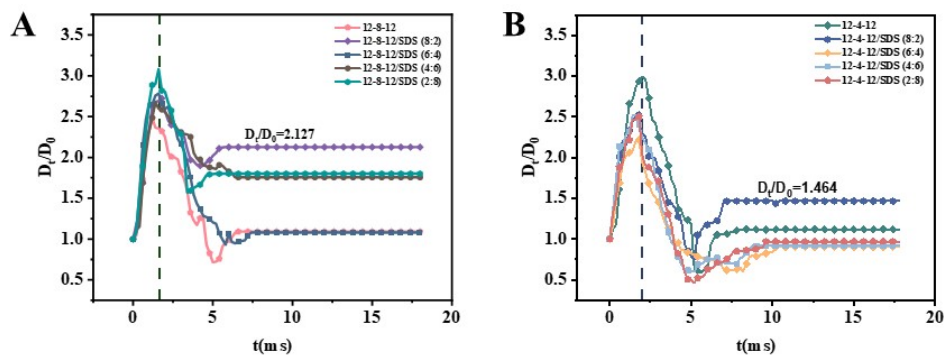
**Fig. S7** The deposition of a high-speed ( $\sim 1.98 \text{ m s}^{-1}$ ) SDS droplets on soybean hydrophobic leaf surface.



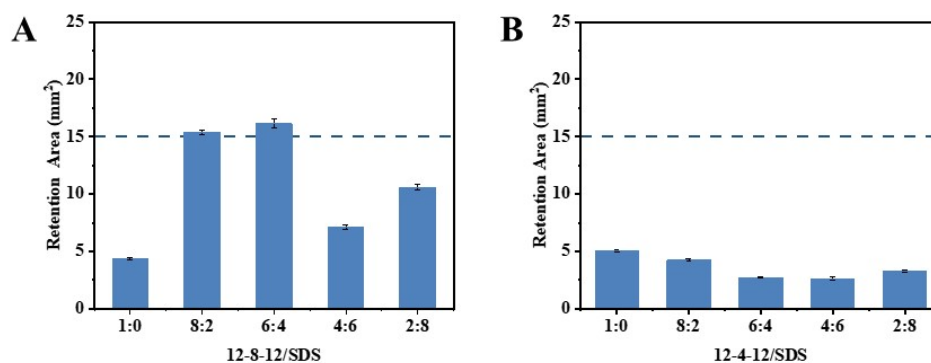
**Fig. S8** The deposition of a high-speed ( $\sim 1.98 \text{ m s}^{-1}$ ) 12-4-12/SDS droplets at various molar fractions ( $X_{12-4-12}$ ): 0.20, 0.40, 0.60, 0.80, and 1.00 on soybean hydrophobic leaf surface



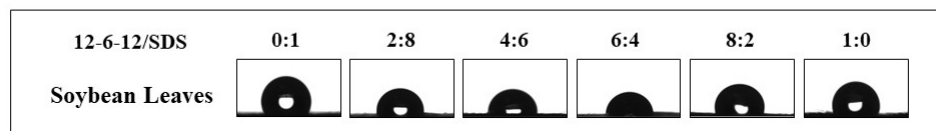
**Fig. S9** The deposition of a high-speed ( $\sim 1.98 \text{ m s}^{-1}$ ) 12-8-12/SDS droplets at various molar fractions ( $X_{12-8-12}$ ): 0.20, 0.40, 0.60, 0.80, and 1.00 on soybean hydrophobic leaf surface.



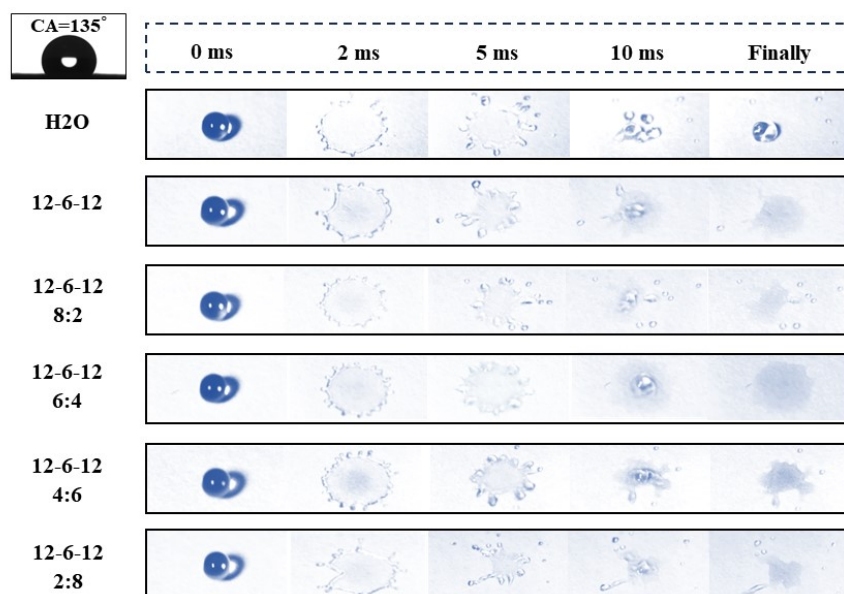
**Fig. S10** (A)-(B) Time evolution of the spreading factor  $D_t/D_0$  in the drop impact process on a superhydrophobic surface for 12-8-12/SDS and 12-4-12/SDS water droplets.  $D_0$ : initial droplet diameter,  $D_t$ : spread area diameter at time  $t$ .



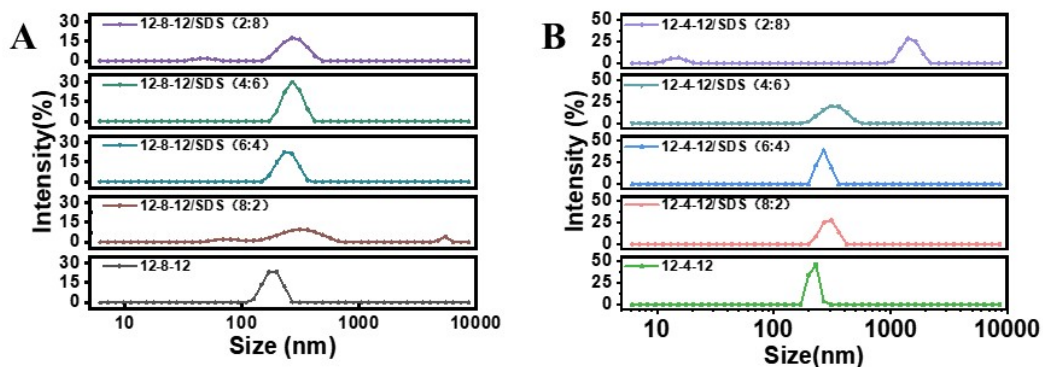
**Fig. S11** (A)-(B) The final deposition area in the drop impact process on a superhydrophobic surface for 12-4-12/SDS and 12-8-12/SDS water droplets.



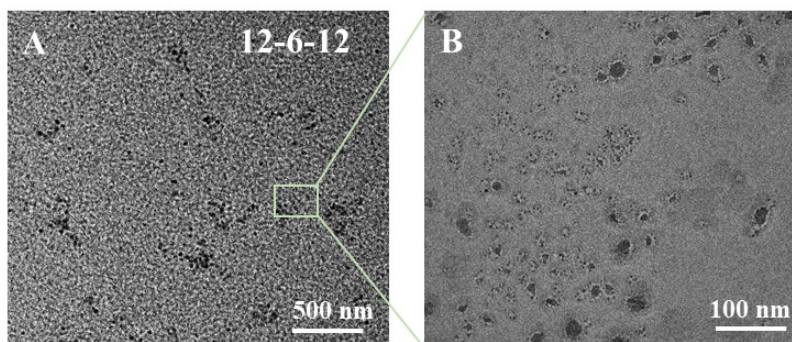
**Fig. S12** For 12-6-12/SDS droplets, the final contact angle at  $X_{12-6-12}=0.6$  ratio is  $114.3 \pm 1.1^\circ$ , indicating better wetting performance compared to other proportions.



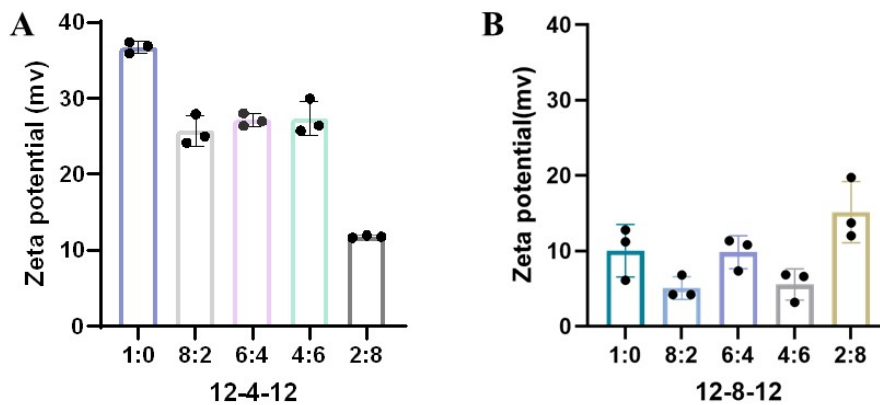
**Fig. S13** The deposition of a high-speed ( $\sim 1.98 \text{ ms}^{-1}$ ) 12-6-12/SDS droplets at various molar fractions ( $X_{12-6-12}$ ): 0.20, 0.40, 0.60, 0.80, and 1.00 on a hydrophobic paper.



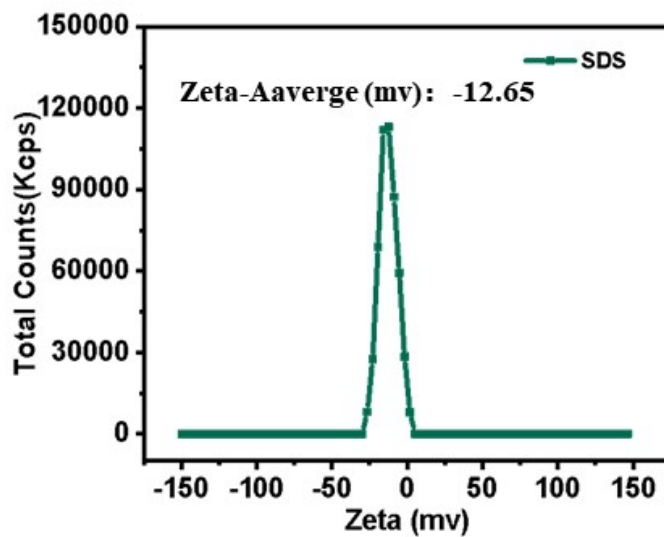
**Fig. S14** (A) and (B) Size distributions of aggregates from dynamic light scattering (DLS) for 12-8-12/SDS and 12-4-12/SDS water droplets at various molar fractions (0.20, 0.40, 0.60, 0.80, and 1.00).



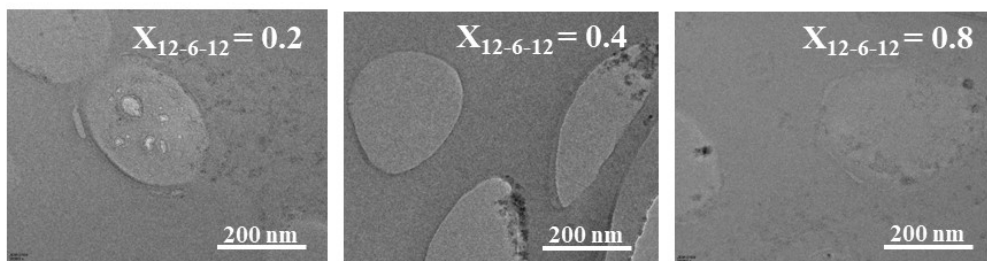
**Fig. S15** (A) 12-6-12 forms small spherical micelles, highlighted with dotted rectangles. (B) is a larger magnification image of A.



**Fig. S16** (A) Zeta potential (mV) for 12-4-12/SDS water droplets at various molar fractions ( $X_{12-4-12}$ ): 0.20, 0.40, 0.60, 0.80, and 1.00. (B) Zeta potential (mV) for 12-6-12/SDS water droplets at various molar fractions ( $X_{12-6-12}$ ): 0.20, 0.40, 0.60, 0.80, and 1.00.



**Fig. S17** Zeta potential (mV) for SDS



**Fig. S18** TEM figures of 12-6-12/SDS at various molar fractions ( $X_{12-6-12}$ : 0.20, 0.40, and 0.80)



## References

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