

# Supporting Information

## Preparation of asymmetric Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> Janus nanoparticles in aqueous phase and its interfacial property

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### Table of contents da rivedere

Text S1. ....	2
Equation S1.....	2
Equation S2.....	2
Text S2. ....	2
Figure S1. ....	3
Figure S2.....	3
Figure S3. ....	4
Figure S4. ....	5
Figure S5. ....	5
Figure S6. ....	6
Figure S7. ....	6

**Text S1.** Titration of carboxyl groups on the surface of Al<sub>2</sub>O<sub>3</sub> nanoparticles.

Since carboxyl groups are acidic in water, sodium hydroxide solution was used to directly titrate the carboxyl groups on the surface of Al<sub>2</sub>O<sub>3</sub> nanoparticles. Approximately 0.2 g of dried carboxylated xerogels was weighed using an analytical balance and placed into a 200 mL beaker, along with 50 mL of deionized water. The dried carboxylated xerogels are sufficiently dispersed in water by ultrasonic dispersion. The three drops of phenolphthalein solution were added to the mixture. The prepared solution was then titrated with a 0.004 M sodium hydroxide solution. The endpoint of the titration was reached when the solution turned red and remained unchanged for 1 minute.

**Equation S1.** Equation for determining the conversion rate (CR) of SA is shown as follows:

$$CR = \frac{n_{SA} \times M_{SA}}{m_{total\ SA}} \times 100\% \quad (S1)$$

**Equation S2.** Equation for determining  $n_{SA}$  in 1g of sample is shown as follows:

$$n_{SA} = \frac{C_{OH} \times V_{OH}}{m_{sample}} \times 1000 \times 1g \quad (S2)$$

where  $n_{SA}$  is the molar mass of SA,  $C_{OH}$  is the concentration of NaOH standard solution,  $V_{OH}$  is the volume of NaOH standard solution needed to reach the equivalence point for the sample,  $m_{sample}$  is the mass of the sample, and  $m_{total\ SA}$  is the total mass of SA.  $M_{SA}$  is the molecular weight of SA.

**Text S2.** Preparation of Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> film by sol-gel dip-coating process

Since the Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> hydrosols used in the experiment are highly polar water-based solutions, it is not suitable for coating onto the silicon wafer. Therefore, methanol with the second polarity was chosen to dilute the Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> hydrosols before and after modification, as well as the Al<sub>2</sub>O<sub>3</sub>+SiO<sub>2</sub> hydrosol after coupling. The freshly prepared alcohol hydrosols were

then deposited on the silicon wafer to form coatings via dip-coating process with a dip coater (SYDC-100, Shanghai Sanyan Technology, China). The ambient temperature and relative humidity (RH) were  $30\pm 2^\circ\text{C}$  and  $35\pm 5\%$ . The resulting films were subsequently dried at  $100^\circ\text{C}$  for 1h to allow complete solvent evaporation and obtain dry gels. The substrates were washed successively in dilute hydrofluoric acid (0.1 wt.%), deionized water and ethanol and wiped carefully before dip-coating process. The substrates were immersed in the alcohol hydrosols for five seconds and then lifted vertically at a constant withdraw rate. Finally, each coating was used to test the water contact angle (WCA) of the samples.

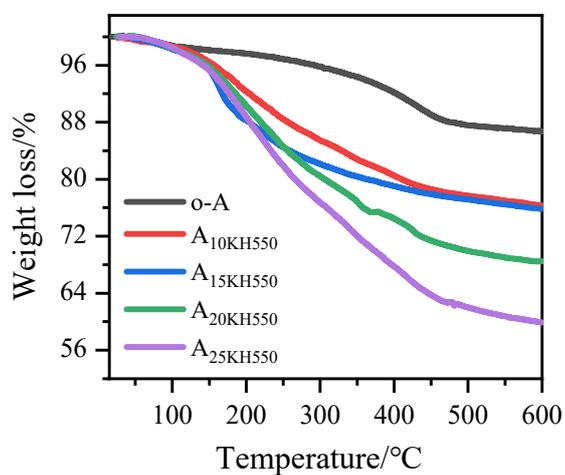


Figure S1. TGA curves of KH550 modified  $\text{Al}_2\text{O}_3$  nanoparticles

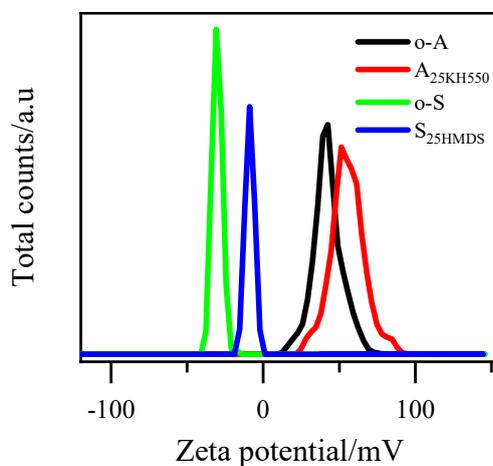
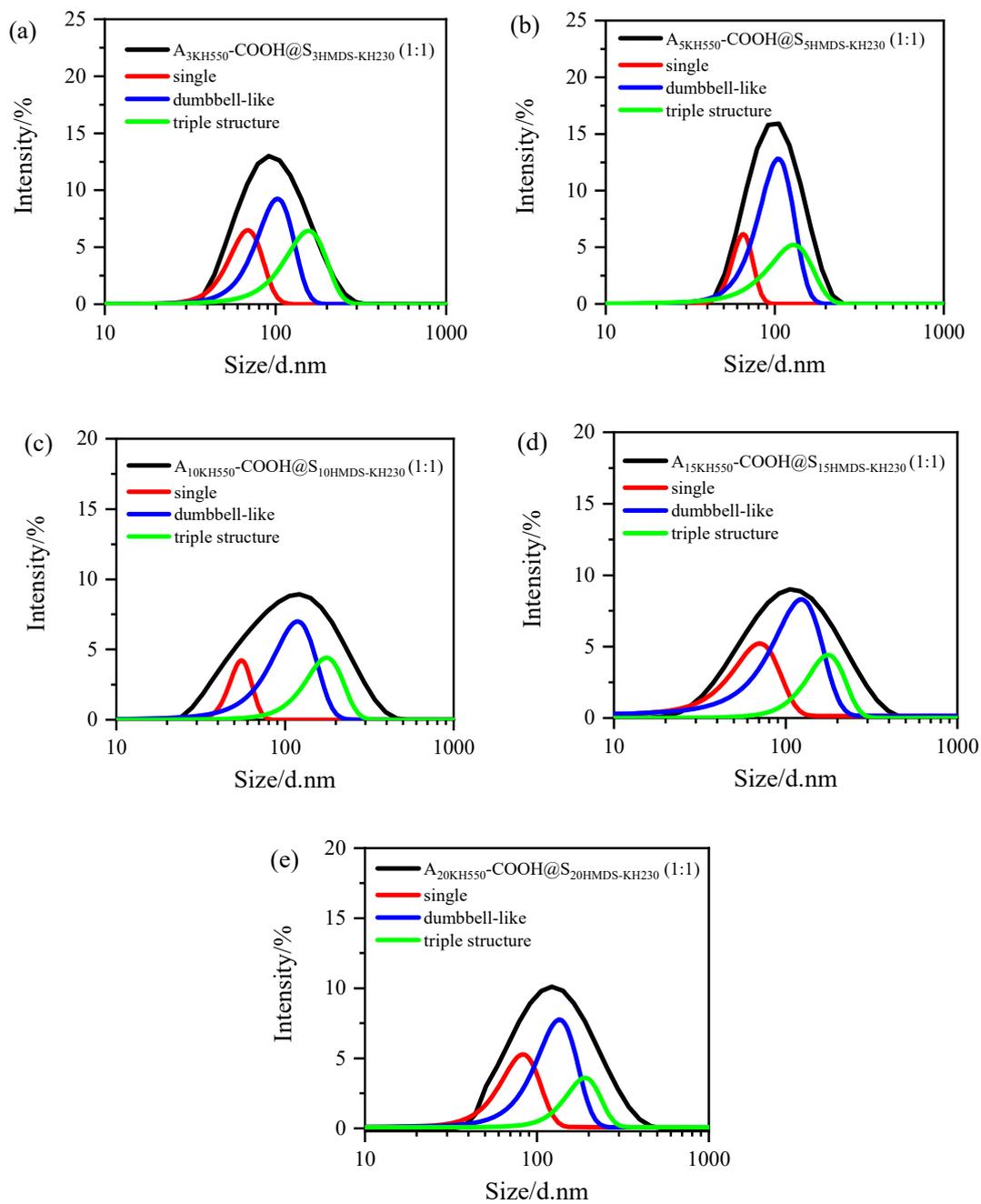
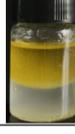


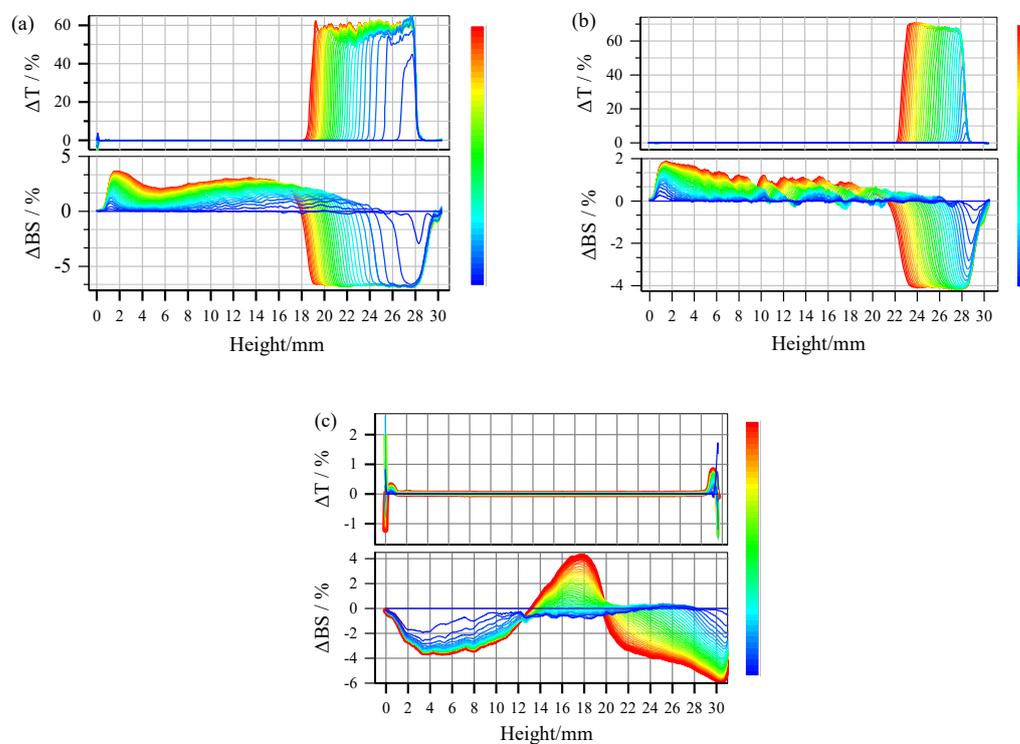
Figure S2. Zeta potential of o-A, A<sub>25</sub>KH550, o-S and S<sub>25</sub>HMDS.



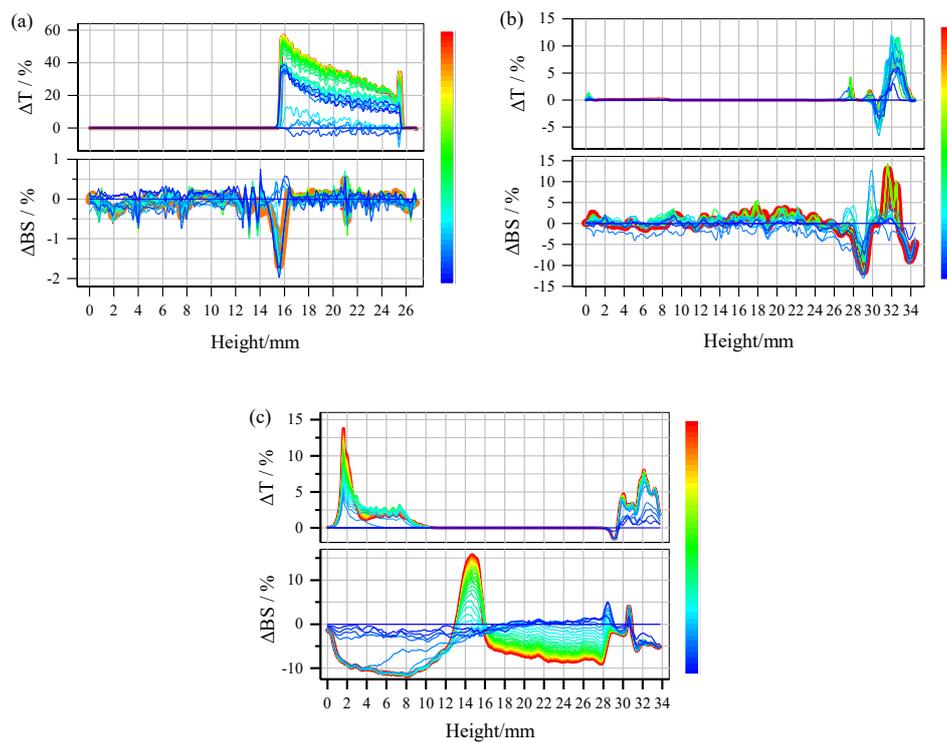
**Figure S3.** Peak fitting analysis of the particle size distribution of various dumbbell-like  $\text{Al}_2\text{O}_3$ - $\text{SiO}_2$  nanoparticles: (a)  $\text{A}_{3\text{KH550}}\text{-COOH@S}_{3\text{HMDS-KH230}}$  (1:1); (b)  $\text{A}_{5\text{KH550}}\text{-COOH@S}_{5\text{HMDS-KH230}}$ ; (c)  $\text{A}_{10\text{KH550}}\text{-COOH@S}_{10\text{HMDS-KH230}}$ ; (d)  $\text{A}_{15\text{KH550}}\text{-COOH@S}_{15\text{HMDS-KH230}}$ ; (e)  $\text{A}_{20\text{KH550}}\text{-COOH@S}_{20\text{HMDS-KH230}}$

Sample Oil-water system	o-A	A <sub>25KH550</sub> -COOH	o-S	S <sub>25HMDS</sub>	A <sub>25KH550</sub> -COOH@ S <sub>25HMDS</sub> -KH230	A <sub>25KH550</sub> -COOH@ S <sub>25HMDS</sub>
cyclohexane-water						
toluene-water						
silicone oil -water						
vegetable oil-water						

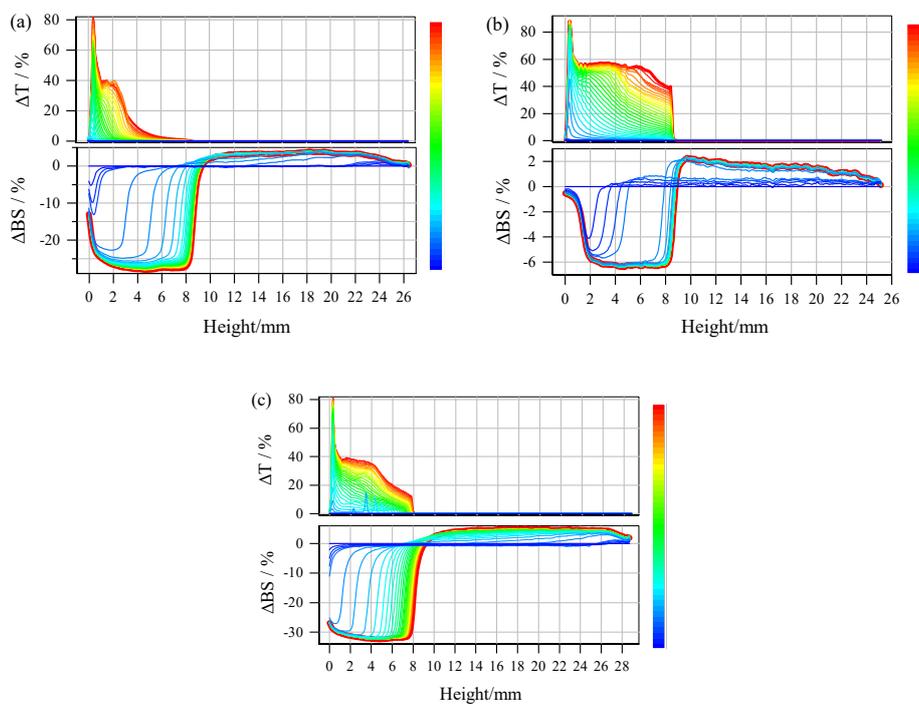
**Figure S4.** Photographs of various nanoparticles stabilization of four oil-water model systems after stirring for 10min at room temperature and standing for a moment (oil is the phase on top, water is the phase at the bottom, emulsifier dosage is 0.05g)



**Figure S5.** Delta transmission ( $\Delta T$ , top) and delta backscattering profiles ( $\Delta BS$ , bottom) of oil-water systems after emulsification by A<sub>25KH550</sub>-COOH@S<sub>25HMDS</sub>-KH230: (a) toluene-water emulsions; (b) cyclohexane-water emulsions; (c) vegetable oil-water emulsions (Time axis: blue to red represents from 0 to 3h)



**Figure S6.** Delta transmission ( $\Delta T$ , top) and delta backscattering profiles ( $\Delta BS$ , bottom) of oil-water systems after emulsification by  $A_{25KH550}\text{-COOH}@S_{25HMDS}$ : (a) toluene-water emulsions; (b) cyclohexane-water emulsions; (c) vegetable oil-water emulsions (Time axis: blue to red represents from 0 to 3h)



**Figure S7.** Delta transmission ( $\Delta T$ , top) and delta backscattering profiles ( $\Delta BS$ , bottom) of oil-

water systems after emulsification by sodium dodecyl sulfate: (a) toluene-water emulsions; (b) cyclohexane-water emulsions; (c) vegetable oil-water emulsions (Time axis: blue to red represents from 0 to 3h)