Electronic Supplementary Material (ESI) for Nanoscale

Supporting Information

A Novel Method of Preparing Metallic Janus Silica Particles Using Supercritical Carbon Dioxide

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Calculations for experimental section

The calculation of the excess amount of SiO_2-NH_2 particles that were added into the suspension is shown in the following:

The expressions for the number of SiO_2 particles (N_s) (Eq. 1) and PS spheres (N_p) (Eq. 2) are:

$$N_S = \frac{3}{4\pi}\left(\frac{W_S}{\rho_S}\right)\left(\frac{1}{r}\right)^3 = \frac{3}{4\pi}\left(\frac{c_S V_S}{\rho_S}\right)\left(\frac{1}{r}\right)^3 \text{................................}Eq.1$$

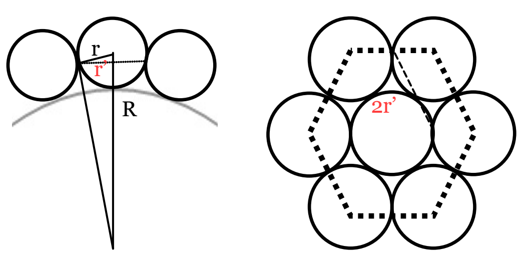
$$N_P = \frac{3}{4\pi}\left(\frac{W_P}{\rho_P}\right)\left(\frac{1}{R}\right)^3 = \frac{3}{4\pi}\left(\frac{c_P V_P}{\rho_P}\right)\left(\frac{1}{R}\right)^3 \text{................................}Eq.2$$

where N_S and N_P are the numbers of SiO_2 particles and PS spheres; w_S and w_P are their total weights; r and R are the radii of SiO_2 particles and PS speres; c_S and c_P are the concentration of SiO_2 particles and PS spheres in the suspension; V_S and V_p are the volumes of SiO_2 particles and PS spheres in suspension.

From eq 1 and 2, the real number ratio (n) between SiO_2 particles and PS spheres can be calculated by Eq.3:

$$n = \frac{N_S}{N_P} = \frac{\frac{3}{4\pi}\left(\frac{c_S V_S}{\rho_S}\right)\left(\frac{1}{r}\right)^3}{\frac{3}{4\pi}\left(\frac{c_P V_P}{\rho_P}\right)\left(\frac{1}{R}\right)^3} = \left(\frac{\rho_P}{\rho_S}\right)\left(\frac{C_S V_S}{C_P V_P}\right)\left(\frac{R}{r}\right)^3 \text{..................}Eq.3$$

In the following we assume for simplicity that: (a) PS spheres are fully covered with SiO_2 particles; (b) there are no free SiO_2 particles in the solution; (c) both SiO_2 particles and PS spheres are monodisperse; (d) SiO_2 particles are hexagonally packed on the surface of PS spheres.



Thus the theoretical maximal number (n_{max}) of SiO_2 particles on a PS sphere can be calculated by Eq.4:

$$n_{\max} = \frac{A_P}{A_S} = \frac{4\pi R^2}{2\sqrt{3}r'^2} = \frac{2\sqrt{3}}{3}\frac{R}{R+2r}\left(\frac{R+r}{r}\right)^2 \approx \frac{2\sqrt{3}}{3}\left(\frac{R}{r}\right)^2 \text{........}Eq.4$$

Where *A*p is the surface area that the PS sphere can provide; *A*s is the surface area that a SiO_2 particle occupied on a PS sphere.

Thus the ratio *f* between the experimentally used ratio (*n)* and the theoretical maximal number (n_{max}) of SiO_2 particles on PS spheres denominates the excess ($f = n / n_{max} > 1$)

According to the value of each parameters provided in the experimental section, the value of *f* for both 10 µm/500 nm and 2 µm/100 nm systems can be calculated as $f_{10\ \mu m/500\ nm} \approx 3.0$ and $f_{2\ \mu m/130\ nm} \approx 1.7$, respectively.

All silica particles that are embedded on the surface of PS templates can be converted to Janus particles, leading to a high yield of Janus particles. The highly permeable small molecular CO_2 - in supercritical state - provides a homogeneous environment for plastifing treatment of PS templates and thus renders evenly embedment of silica particles, leading to homogeneous Janus balance control. In addition, only embedded silica particles were kept on the surface of the PS templates as excess silica particles can be removed easily before and after the embedding treatment by filtration. Therefore, the amount of Janus particles prepared in one batch can be calculated by deducting the weight of the dried filtrate from the total amount of silica particle added. Actually, the productivity deeply depends on how much excess silica particles were added into the suspension. Since the excess of silica particles is $f \approx 3.0$ (three times the amount of silica particles that can adsorb on the PS template surface) for 10 µm/500 nm in our experiment, the yield of Janus particles is around 30%. The yield is increased to 83% by appropriately reducing the excess of silica particles degree to $f = 1.2$.

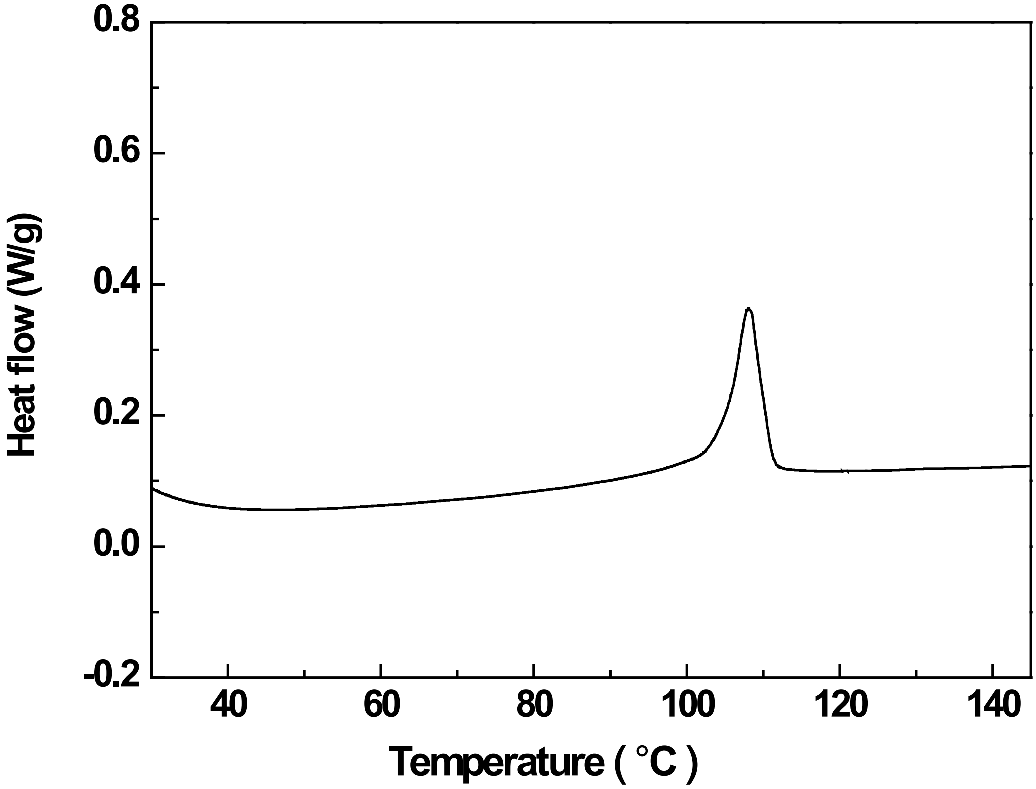


Fig. S1. DSC curve (second heating) of 10 μm PS-COOH particles.

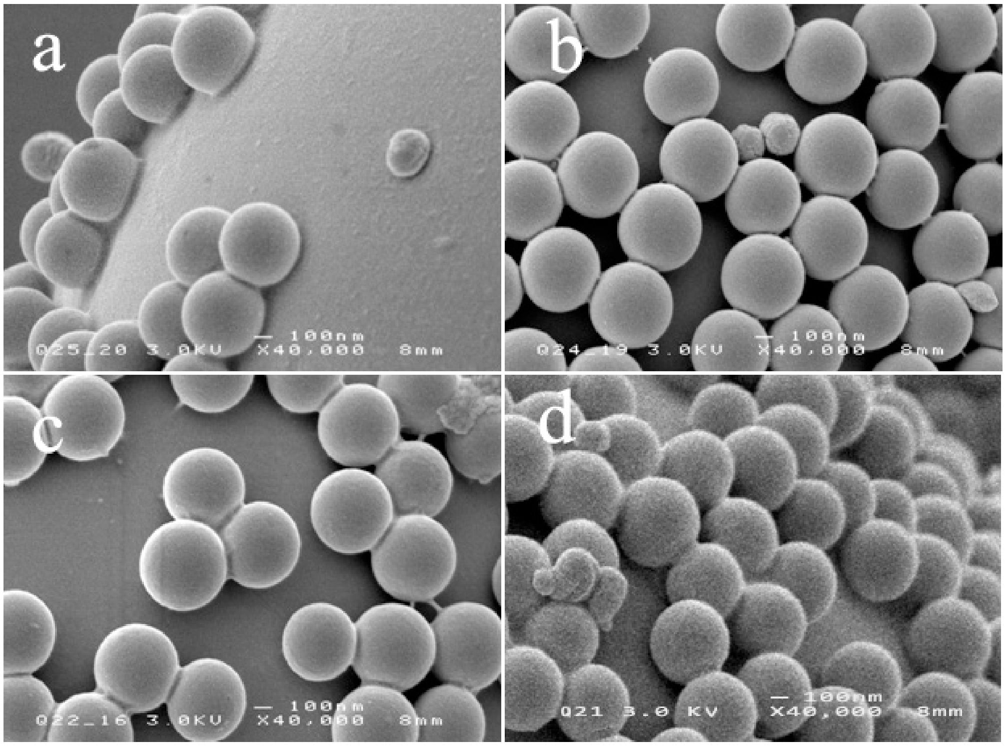


Fig. S2. SEM images of embedded silica particles into PS sphere templates after treatment by sc CO_2 under 126 bar for 4 h at different temperatures: (a) 55 ˚C, (b) 45 ˚C, (c) 33 ˚C, (d) 25 ˚C.

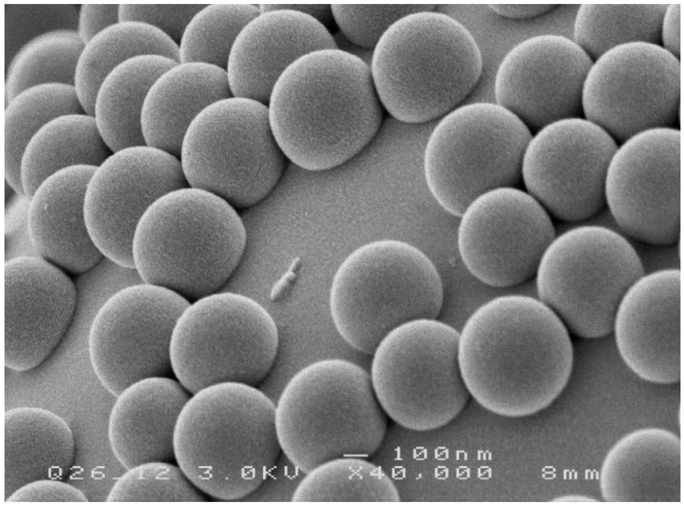


Fig. S3. SEM images of embedded silica particles into PS sphere templates after treatment by sc CO_2 under 60 ˚C, 126 bar for 1 h.

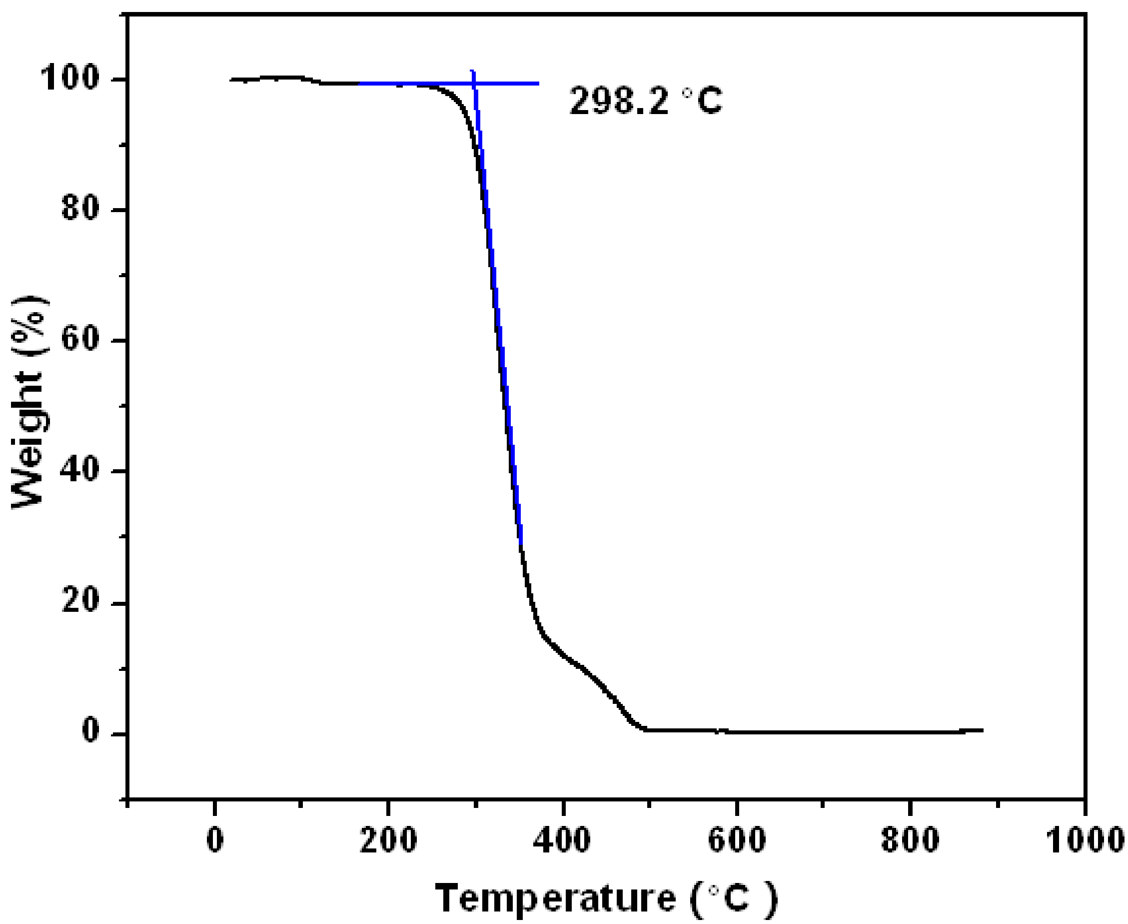


Fig. S4. TGA curves of10 μm PS-COOH particles.

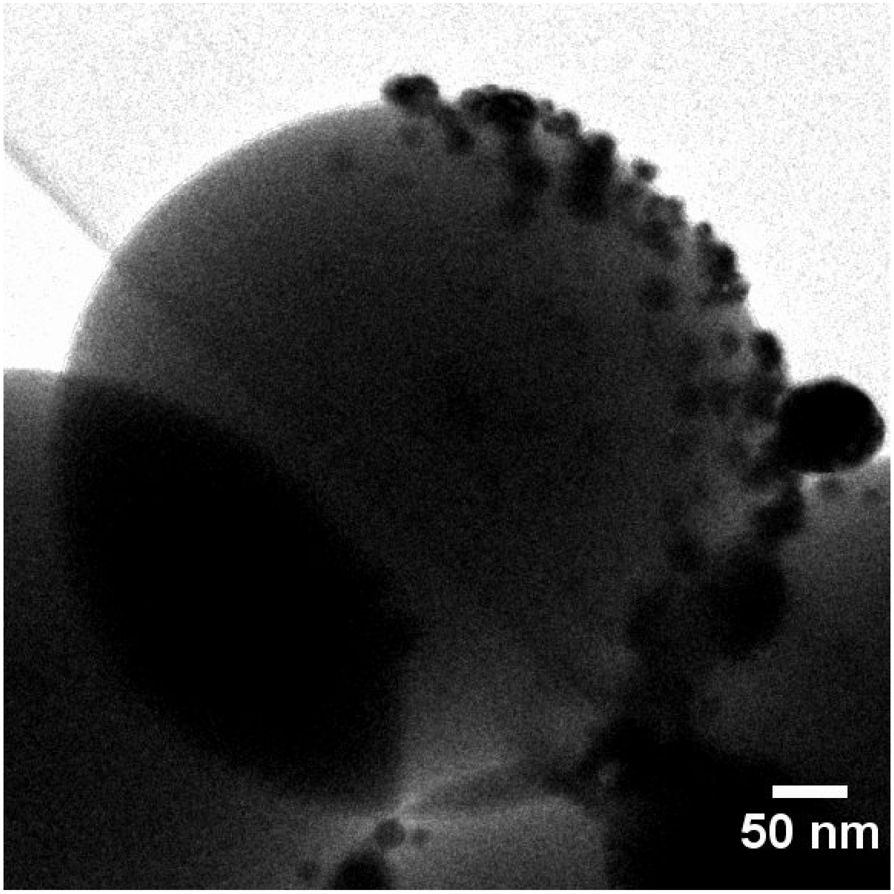


Fig. S5. TEM image of gold nanoparticles labeled Janus silica particles obtained by burning PS sphere templates in a furnace overnight at 400 ˚C. The embedment was conducted in sc CO_2 under 60 ˚C, 170 bar for 4 h.