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Supporting information

Encapsulation of Atomically Thin Gold Nanosheets within Porous Silica for Enhanced Structural Stability and Superior Catalytic Performance

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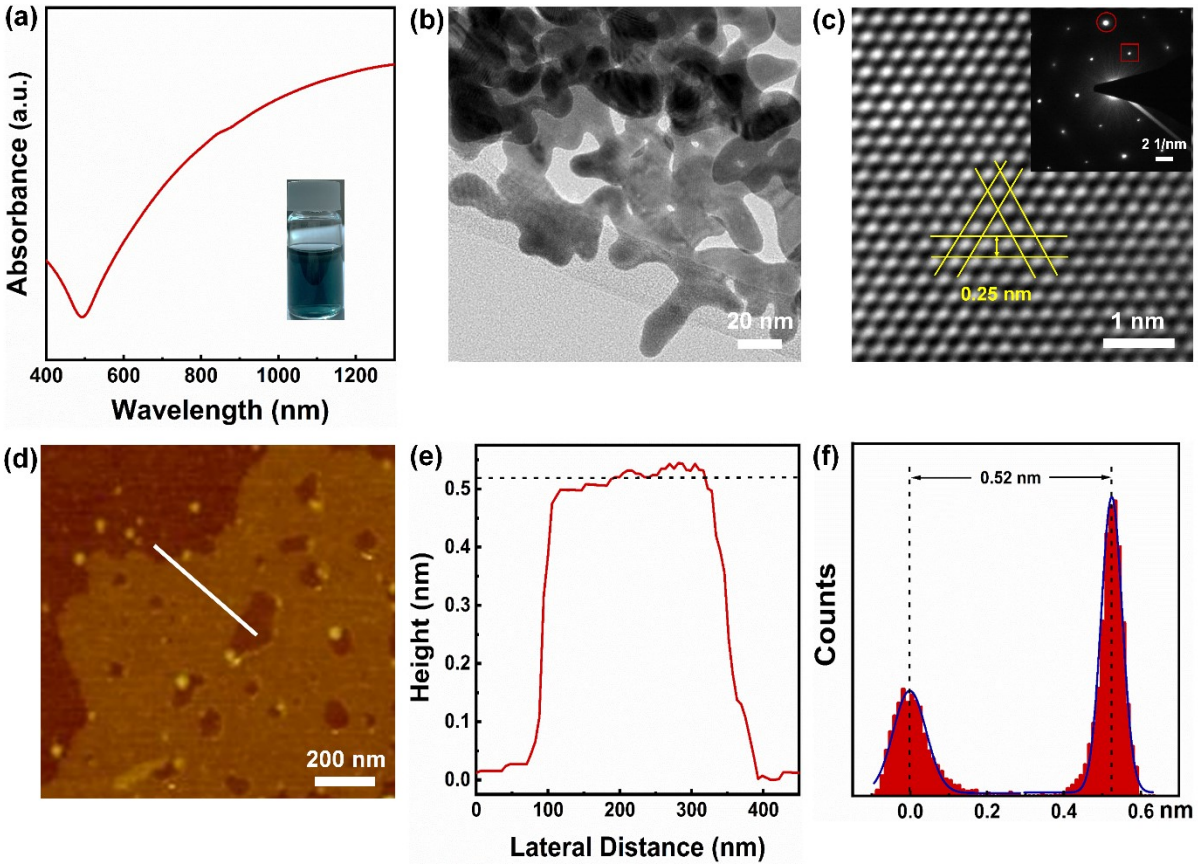

Figure S1. (a) UV-vis-NIR spectrum and photograph (inset) of as-prepared AuNSs dispersed in water (0.5 mg/ml). (b) TEM, (c) HR-TEM, (d) AFM images of isolated AuNSs for which samples were prepared in grids from the AuNSs dispersion in water at dilute concentration (0.05 mg/ml). The inset in (c) shows the SAED pattern. (e) Height profile (along the white line in (d)) and (f) Height histogram measured from the AFM image.

The as-prepared AuNSs samples dispersed in water (0.5 mg/ml) display green-blue color as shown in the inset of Figure S1a. The UV-vis-NIR spectrum of as-prepared AuNSs dispersed in water exhibit a broad absorption band in the region of 500-1300 nm (Figure S1a). For TEM and AFM measurements, as-prepared AuNSs dispersed in water at dilute concentration (0.05 mg/ml) was drop-casted on TEM grid and freshly cleaved muscovite mica, respectively, and dried at room temperature. The TEM image shows translucent and seaweed-like AuNSs (Figure S1b) and the HR-TEM image reveals a six-fold symmetric structure with

a lattice spacing of 0.25 nm (Figure S1c).[1,2,3] The SAED pattern in the [111] zone axis shows two sets of six-fold symmetric spots (inset of Figure S1c). One set of spots corresponds to the allowed {220} reflections with a lattice spacing of 0.144 nm and the other one corresponds to the forbidden 1/3 {422} reflections with a lattice spacing of 0.250 nm. The presence of the forbidden 1/3 {422} reflections suggests that the surface of ultrathin AuNS is atomically flat.[4,5,6] The HR-TEM and SAED results indicate the single crystalline nature of AuNSs.[1,6] The height profile and height histogram obtained from the AFM image show that the thickness of AuNSs is ca. 0.52 nm indicating that the as-prepared AuNSs are two atomic layers thick (Figure S1d, e, and f).

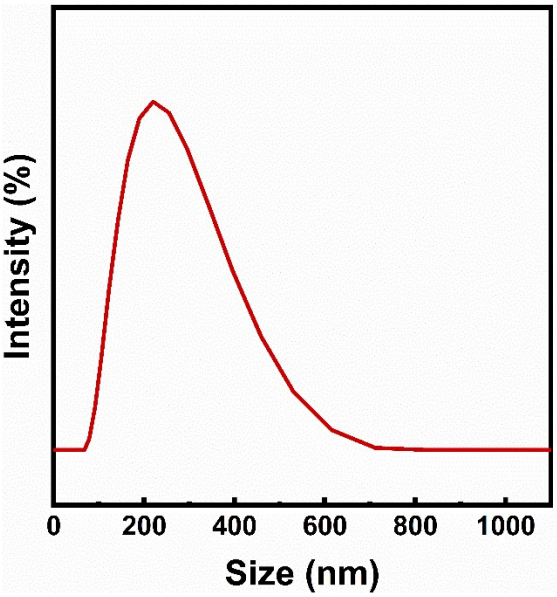

Figure S2. Distribution of hydrodynamic diameter of as-prepared AuNSs dispersed in water.

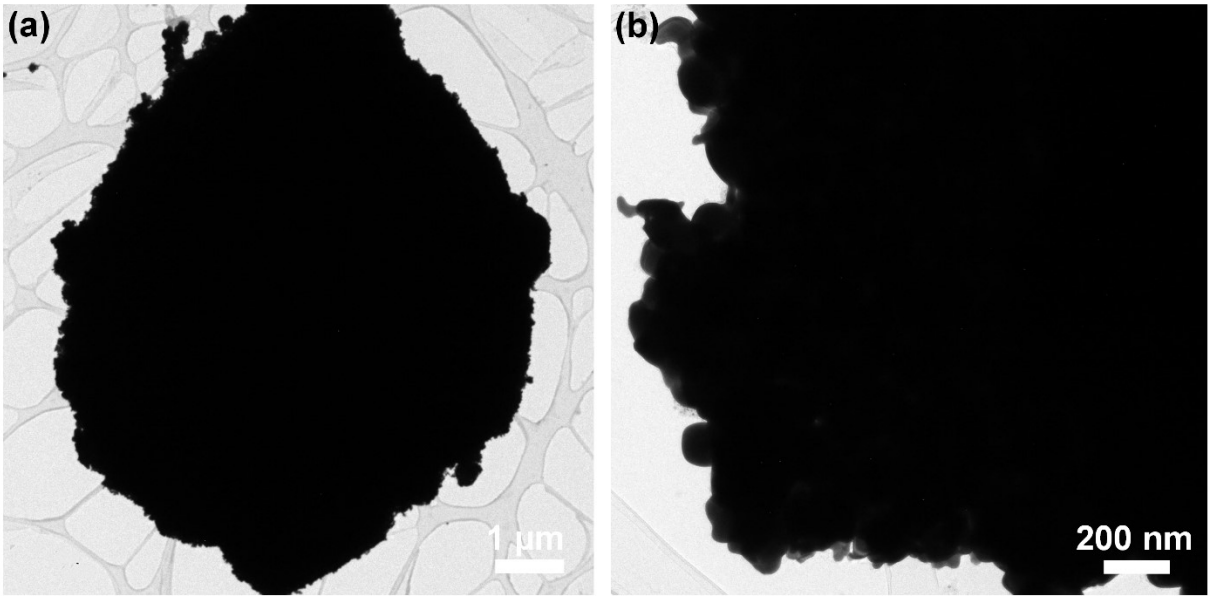

Figure S3: Low magnification TEM images of AuNSs in the dried state. The samples were prepared by placing the dispersions of AuNSs in water (0.5 mg/ml) on TEM grids and drying at room temperature.

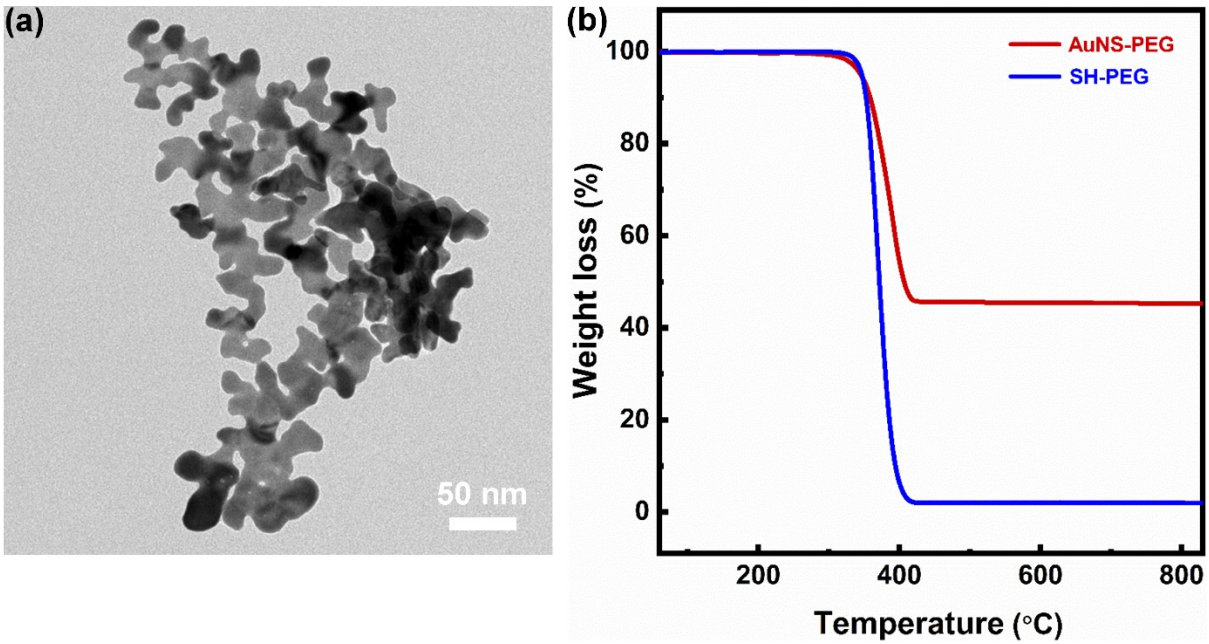

Figure S4. (a) TEM image of AuNSs functionalized with SH-PEG (AuNS-PEG). (b) TGA of AuNS-PEG and SHPEG. The TGA results of both AuNS-PEG and SHPEG show a drastic weight loss at around 400 °C, which is due to the degradation of the SHPEG molecules.

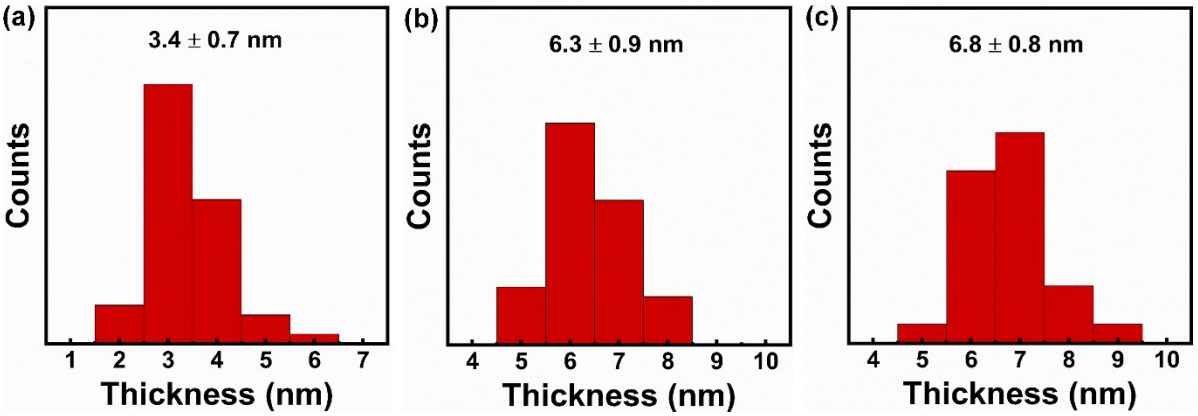

Figure S5. Distribution of silica layer thickness measured from the TEM images of silica-encapsulated AuNS-PEG prepared with different amounts of TEOS, (a) 30 µl, (b) 60 µl, and (c) 90 µl. The average thicknesses of silica layer are ca. 3.4, ca. 6.3, and ca. 6.8 nm, respectively.

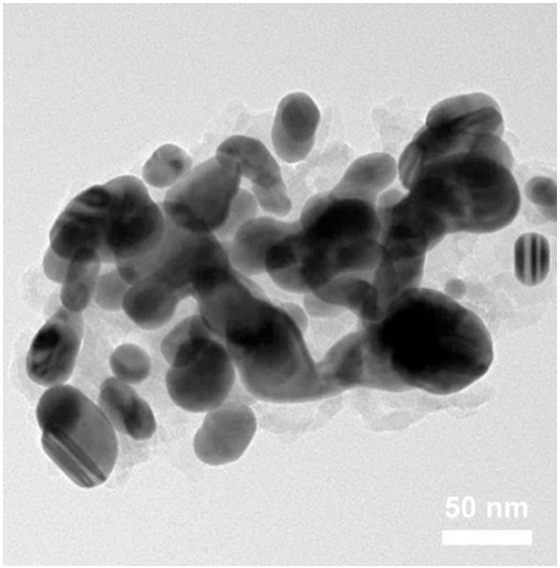

Figure S6. TEM image of AuNSs encapsulated with ca. 3.4 nm thick silica layer after UV-ozone treatment.

Table S1. Elemental analysis of AuNSs ecapsulated with ca. 6.3 nm thick silica layer before and after UV-ozone treatment.

Sample	C (wt%)	H (wt%)	S (wt%)
Before UV-ozone treatment	11.43	2.41	0.98
After UV-ozone treatment	0.35	0.25	0.02

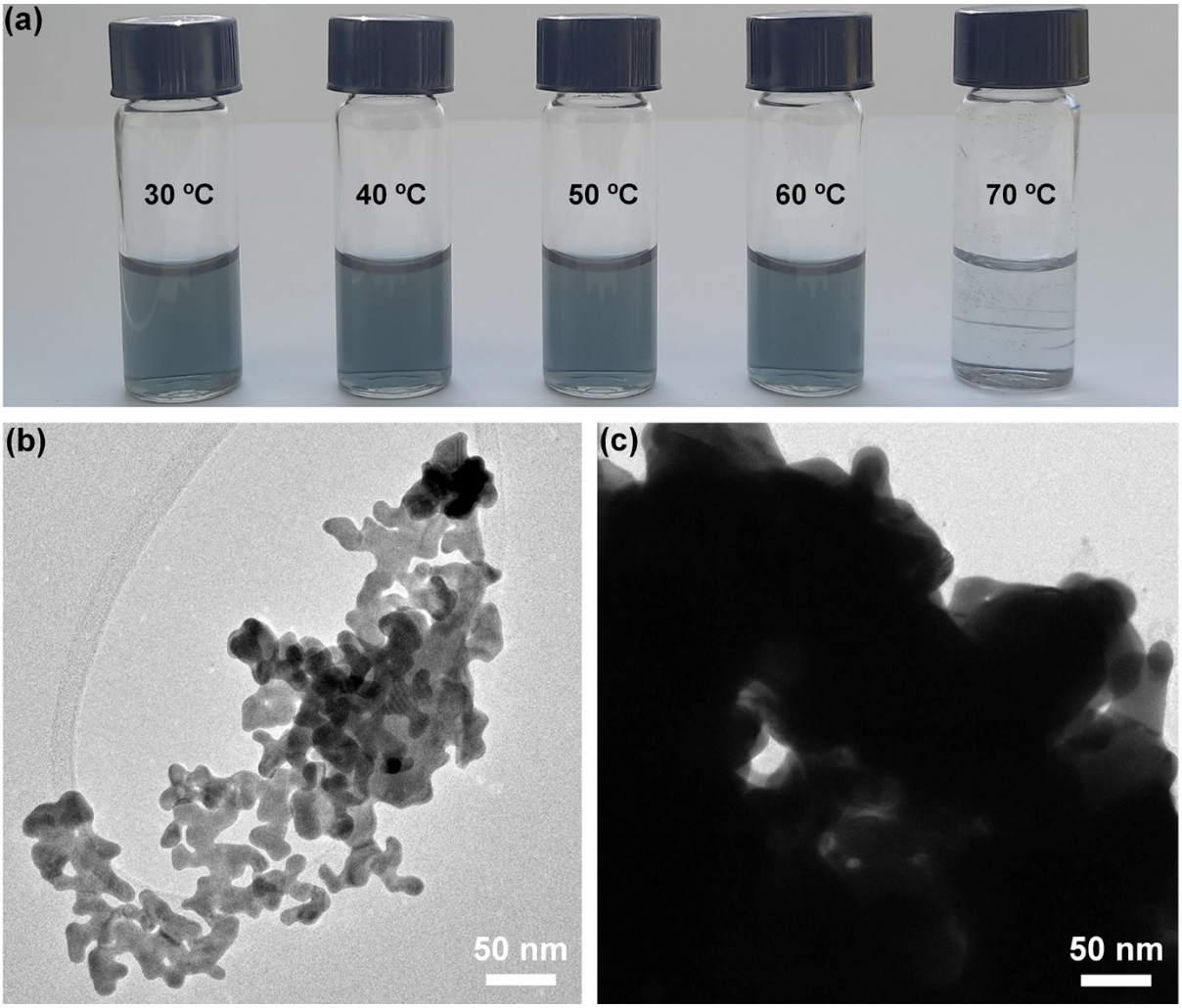

Figure S7. (a) Photographs of as-prepared AuNS dispersions in water (0.5 mg/ml) treated at different temperatures for 2 hours in a water bath. The TEM images were obtained for the samples treated at (b) 60 °C and (c) 70 °C.

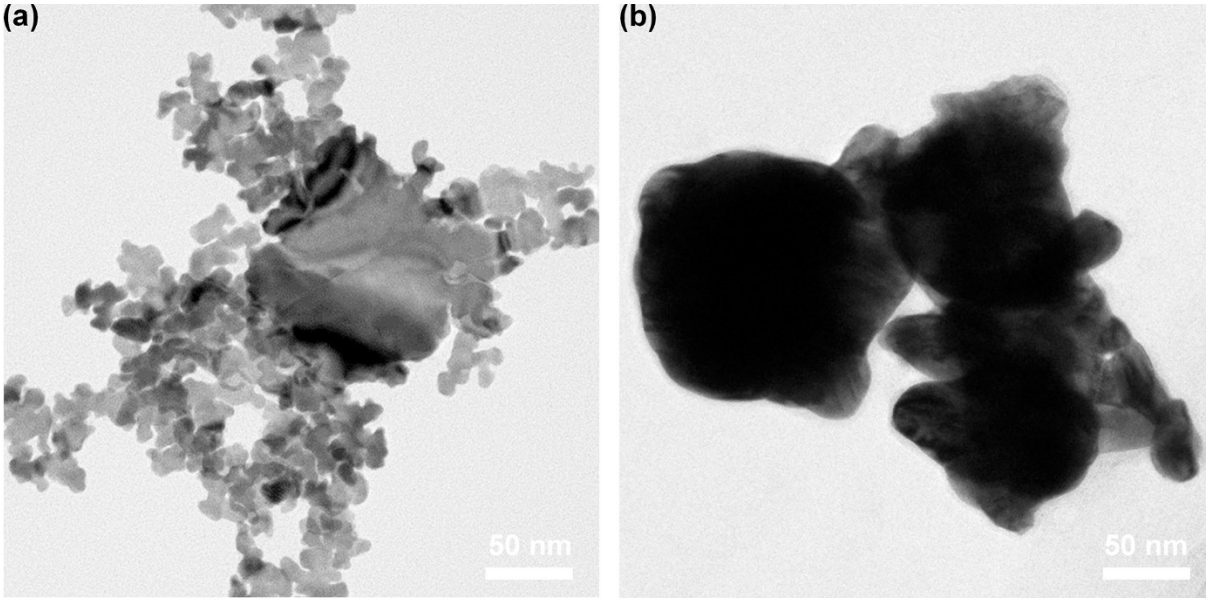

Figure S8. TEM images of isolated AuNSs deposited on TEM grids and treated at (a) 60 °C and (b) 70 °C for 2 hours in an oven.

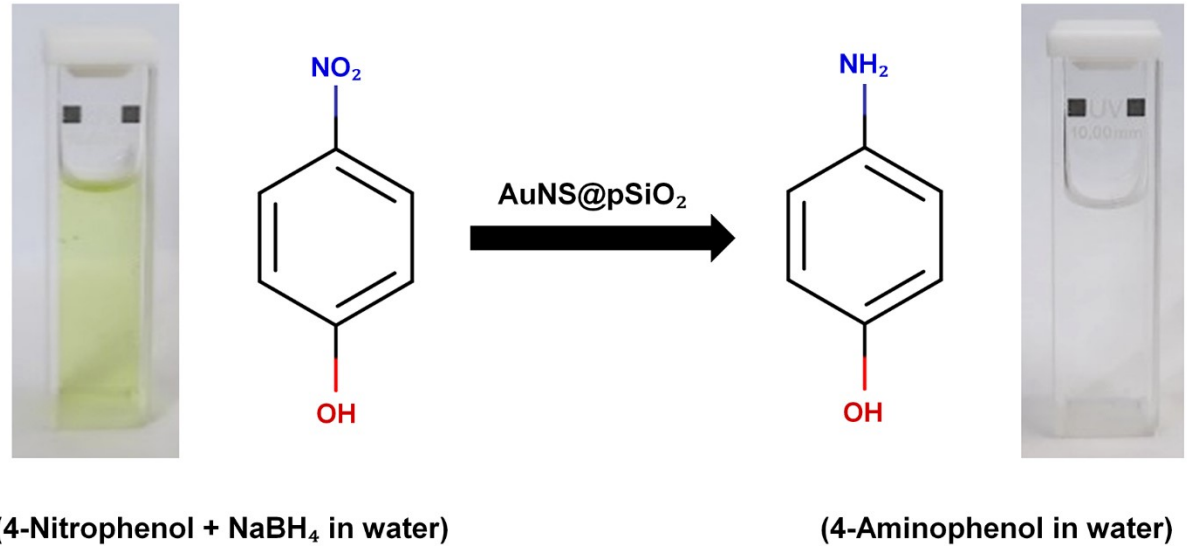

Figure S9. Photographs show the colour change of 4-NPh solution before and after catalytic reduction with AuNS@pSiO$_2$.

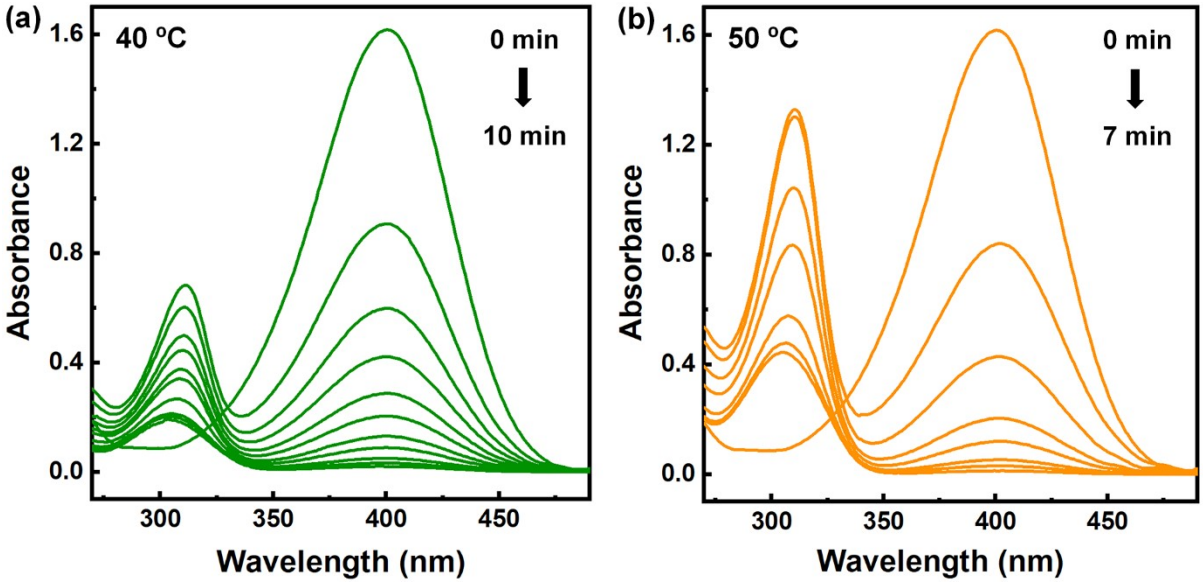

Figure S10. UV-vis absorption spectra monitoring the catalytic reduction of 4-NPh at (a) 40 °C and (b) 50 °C with time after the addition of AuNS@pSiO_2.

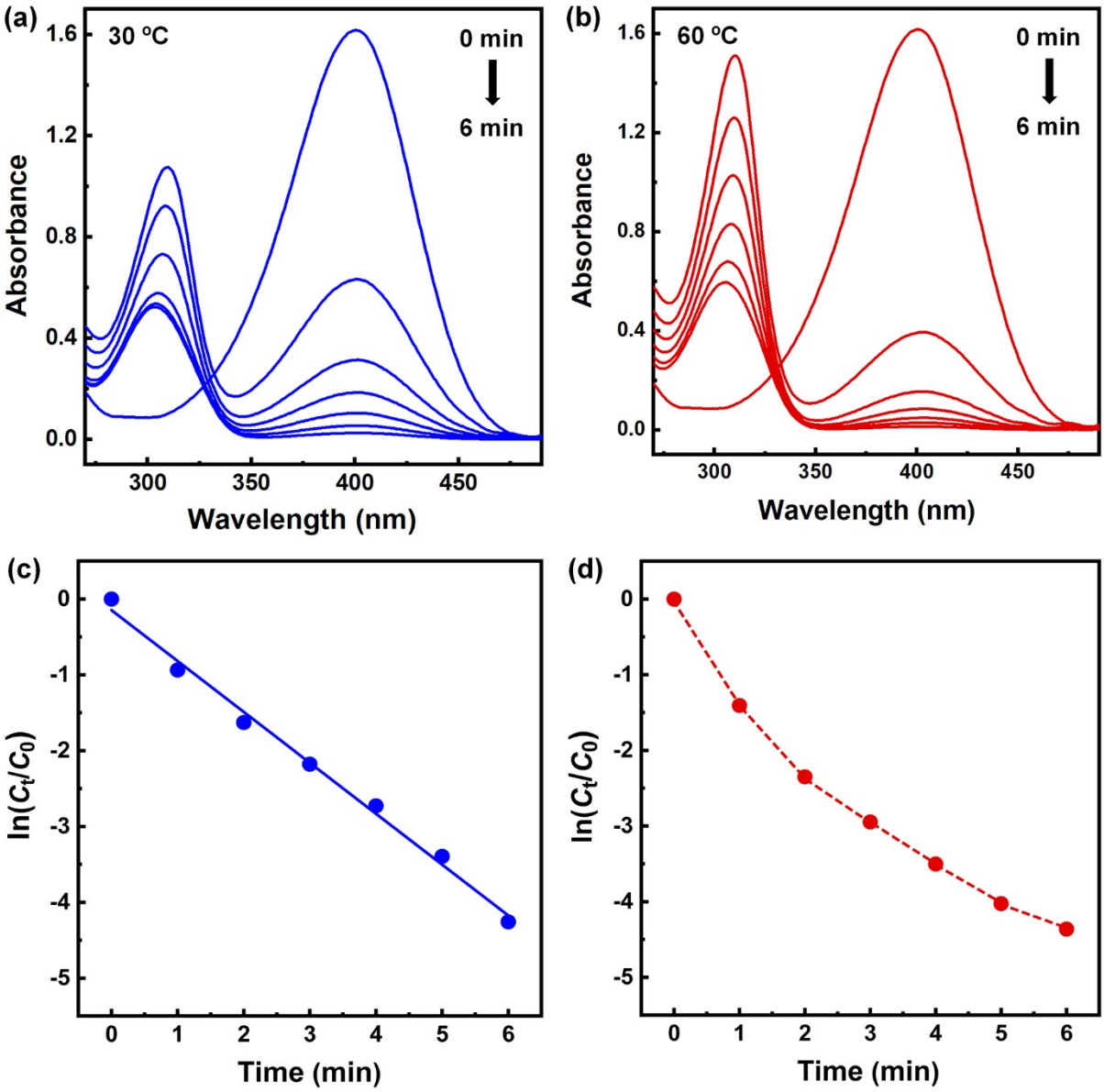

Figure S11. UV-vis absorption spectra monitoring the catalytic reduction of 4-NPh at (a) 30 °C and (b) 60 °C with time after the addition of as-prepared AuNSs. (c, d) The corresponding plots of $\ln(C_0/C_t)$ against reaction time, which is linear for the reaction at 30 °C (the estimated rate constant k is 0.67 min^{-1}) and non-linear for the reaction at 60 °C (indicating that as-prepared AuNSs might be aggregated during the catalytic reaction).

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