

COMMUNICATION

Supporting Information

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One-pot synthesis of Au-Fe₂O₃@SiO₂ core-shell nanoreactors for CO oxidation

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Experimental Section

Materials. Tetraethyl silicate (TEOS), Polyoxyethylene (10) cetyl ether (Brij® C10), $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{HAuCl}_4 \cdot x\text{H}_2\text{O}$, were obtained from Aladdin Reagent Co., Ltd.; anhydrous ethanol, ammonium hydroxide ($\text{NH}_3 \cdot \text{H}_2\text{O}$, 15 M), and cyclohexane were purchased from Sinopharm Chemical Reagents Co., Ltd.; All reagents were used directly without further purification. Deionized water was used throughout. The gas mixture (carbon monoxide, 1%; O_2 , 20%; and the remainder, N_2) were purchased from Shanghai Weichuang Standard Reference Gas Analytical Technology Company.

Characterization. XRD patterns were collected on a Bruker AXS D8 Advance diffractometer using $\text{Cu K}\alpha$ radiation. Diffraction patterns were measured in the 2θ range from 10° to 90° . XRD samples were prepared by pressuring powders onto a glass plate. TEM images were obtained by a JEOL 2100 transmission electron microscope operated at 200 kV. The high angle annular dark field scanning transition electron microscopy (HAADF-STEM) image with EDS phase mapping of $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ nanoparticles were obtained using ThermoFisher Talos F200X. For TEM measurements, catalyst samples were dispersed in ethanol solution and were dropped onto a carbon-coated copper grid followed by solvent evaporation in air at room temperatures. BET surface area, pore size distribution and the adsorption/desorption isotherms of the nanocatalysts were measured by N_2 adsorption at 77 K, using a Micromeritics ASAP-2020 M automatic specific surface area and porous physical adsorption analyzer. UV-Vis absorption spectra were recorded on a Lambda 950 spectrophotometer at 300~900 nm. The nanoparticles are dispersed in a quartz cuvette.

Catalyst Preparation.

Preparation of $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ core-shell nanoparticles. In a typical synthetic process, 4.25 g Brij® C10 was dissolved in 40 mL of cyclohexane at 50°C . Then, 400 μL of an aqueous solution containing 0.03mmol HAuCl_4 and 0.03mmol FeCl_3 was added dropwise with stirring. After 20 min, 0.8 mL of $\text{NH}_3 \cdot \text{H}_2\text{O}$ (25%) and 1 mL of TEOS were added to the solution in sequence. The reaction mixture was kept stirring for 2.5h to obtain Precursor@ SiO_2 multinuclear/shell nanospheres. The product was centrifuged, washed with ethanol and dried at 100°C for 12h, and calcined at 350°C for 2h to obtain $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ nanocomposite catalyst.

In the process of synthesizing $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ nanocomposite catalyst, the addition amount of HAuCl_4 was 0.03 mmol, and the addition amount of FeCl_3 was 0 mmol, 0.01 mmol, 0.03 mmol, 0.06 mmol, 0.09 mmol, respectively, and the resulting products were labeled as Au_3Fe_0 , Au_3Fe_1 , Au_3Fe_3 , Au_3Fe_6 , Au_3Fe_9 . And the sample without HAuCl_4 addition was labeled as Au_0Fe_3 . The $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ (Au_3Fe_6)nanocomposite catalysts were subjected to thermal pretreatment at 450 and 550°C , respectively.

Catalyst Activity Measurements. The CO catalytic oxidation reaction was carried out in a fix-bed quartz reactor (U-type tube, 6 mm inner diameter). 50 mg of $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ nanocatalysts were charged into the U-type reactor, followed the system was pretreated at 100°C in a Ar flow for 0.5 h and cooled to the room temperature under Ar. And then the gas mixture (carbon monoxide, 1%; O_2 , 20%; and the remainder, N_2) were allowed to flow through the catalysts at a total flow rate of 30 mL/min at different temperatures for 3 h. The concentrations of CO and CO_2 were analyzed by FGA-4100 gas analyzer produced by Foshan Analyser Co., Ltd.

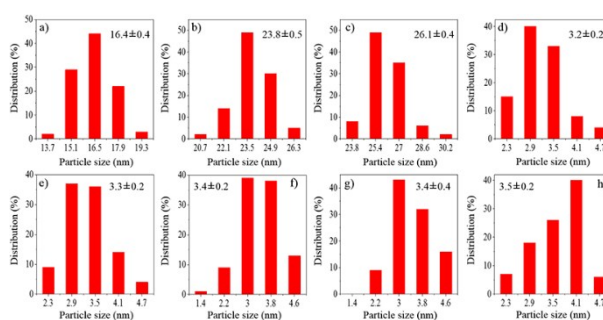
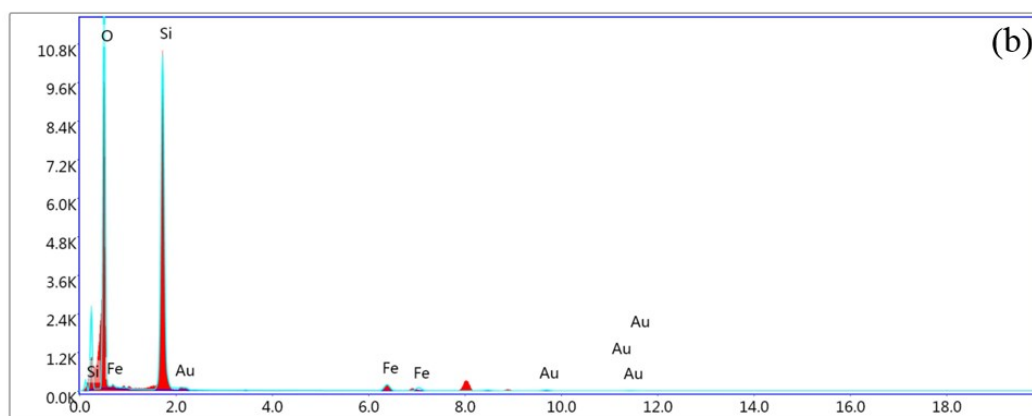
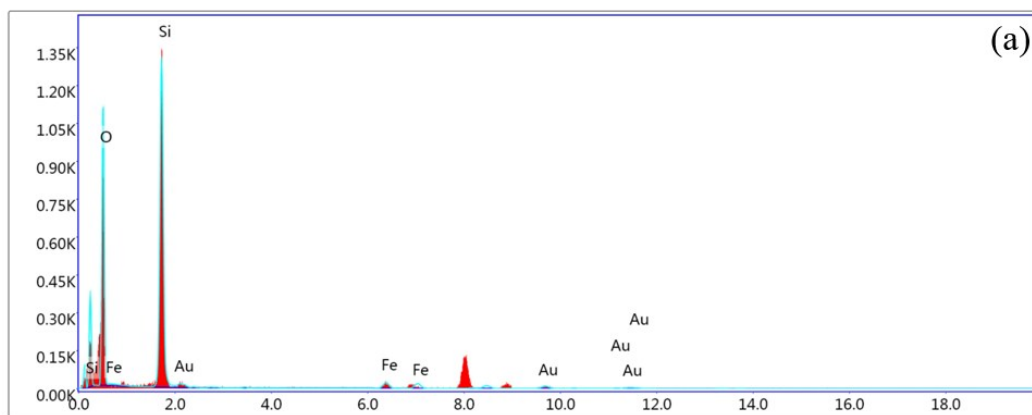
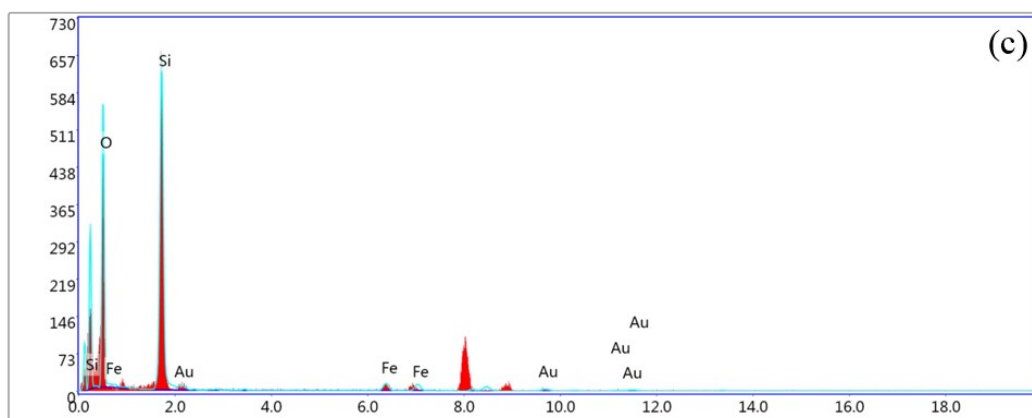


Figure S1. Particle size histograms: a)-c) $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ nanoparticles(Au_1Fe_3) synthesized at different TEOS concentrations of 0.1M, 0.2 M and 0.3 M, respectively; d)-h) $\text{Au-Fe}_2\text{O}_3 @ \text{SiO}_2$ nanoparticles with different Au/Fe ratios of Au_3Fe_0 , Au_3Fe_1 , Au_3Fe_3 , Au_3Fe_6 and Au_3Fe_9 , respectively.



Lsec: 30.0 15 Cnts 4.570 keV Det: Octane Elite TW 55



Lsec: 29.9 0 Cnts 0.000 keV Det: Octane Elite TW 55

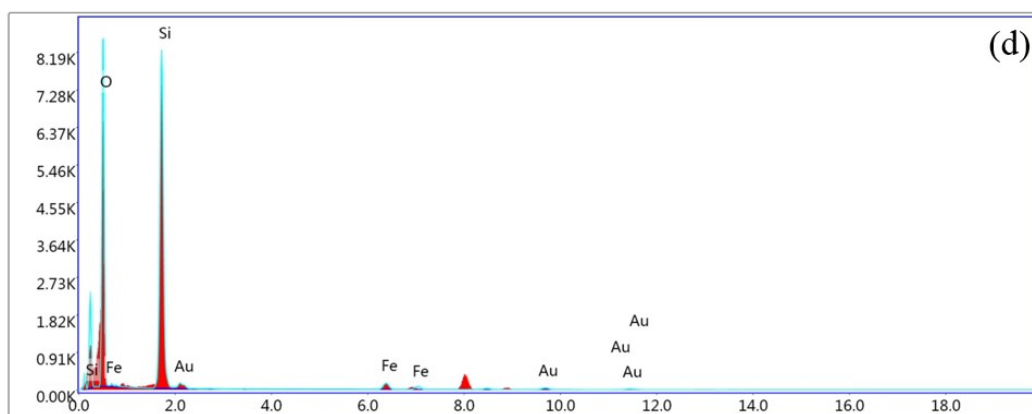
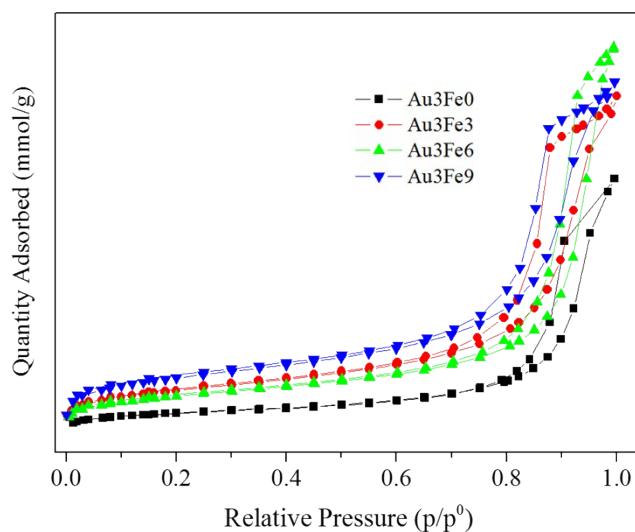
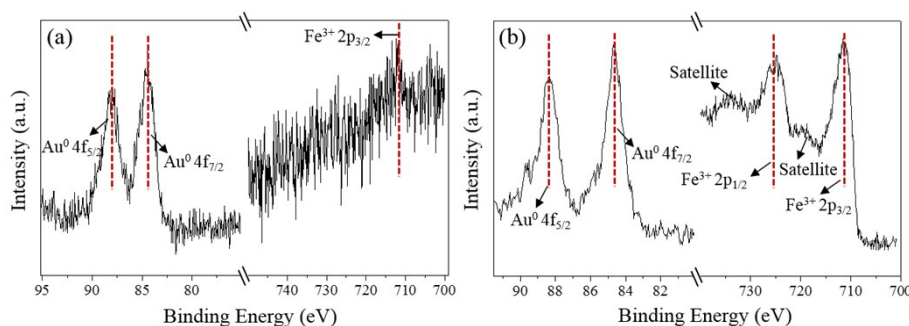


Figure S2. EDS spectra showing: a), Au₃Fe₁ NPs; b), Au₃Fe₃ NPs; c), Au₃Fe₆ NPs; d), Au₃Fe₉ NPs.Figure S3. N₂ sorption/isortherms and pore size distribution of Au-Fe₂O₃@SiO₂ NPs.Figure S4. XPS spectra showing (a) Au-Fe₂O₃@SiO₂(Au₃Fe₆) nanocatalyst calcined in air at 450 °C and (b) Au-Fe₂O₃/SiO₂ supported catalyst (The loadings of Au and Fe are 0.05 and 0.088, respectively.) calcined in air at 450 °C.

Preparation of Au-Fe₂O₃/SiO₂ catalysts. The Au-Fe₂O₃/SiO₂ catalysts were prepared by an incipient-wetness impregnation method. A certain amount of HAuCl₄·xH₂O and FeCl₃·6H₂O (The molar ratio of Au and Fe is 1/3) were dissolved in deionized water, and a calculated amount of silica was added into the above aqueous solutions. After 4 hours, the obtained samples were further dried at 120 °C overnight, and followed by calcination in air at 450 °C for 2 h to obtain Au-Fe₂O₃/SiO₂ catalysts.

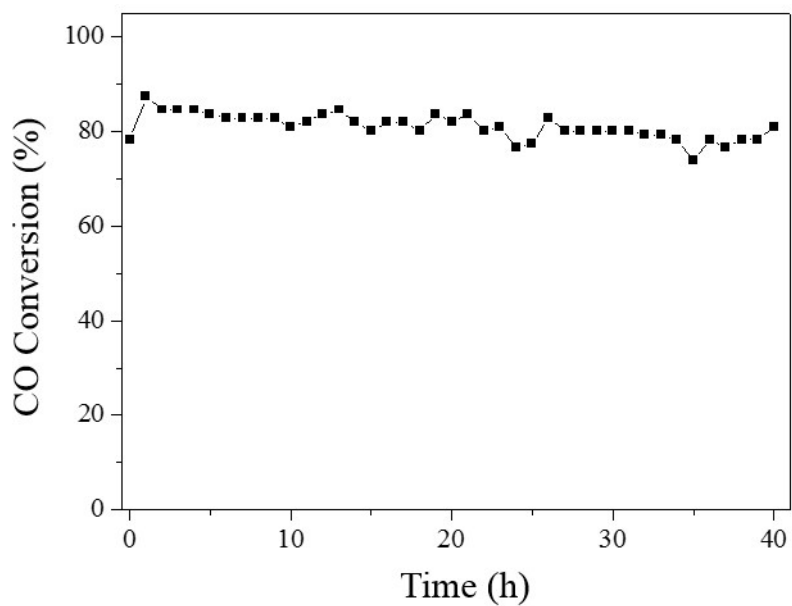


Figure S5. The effect of reaction time on CO oxidation over Au₃Fe₆ nanaocatalyst at 270 °C. Reaction conditions: Au-Fe₂O₃@SiO₂ catalysts-50 mg; Gas flow rate-30 mL/min; CO-1% (v/v), O₂-20% (v/v), and N₂-79% (v/v).