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The mutual promotional effect of Au–Pd/CeO_2 bimetallic catalysts on destruction of toluene

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ABSTRACT

Bimetallic Au–Pd nanoparticles were deposited on CeO_2 and used in the destruction of toluene, which exhibited the mutual promotional effect compared with Au/CeO_2 and Pd/CeO_2 catalysts. This is due to the synergistic effect between Au and Pd. The addition of suitable amount of gold to Pd/CeO_2 increased the proportion of metallic palladium and the catalytic activity.

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1. Introduction

It is well-known that adding a second metal enhances the activity, selectivity, and stability of pure metal catalysts [1]. One of the most popular systems in catalysis research is Au–Pd bimetallic catalyst, which is miscible at any ratio as can be seen in their phase diagram [2]. Chen *et al.* [3] reported the promotional effect of gold on palladium. Shao *et al.* [4] demonstrated the mutual promotional effect of bimetallic Au–Pd catalyst in destruction of p-aniline.

Catalytic oxidation is an effective way for reducing the emissions of volatile organic compounds (VOCs) [5,6]. Supported noble metal catalysts are highly active catalysts for oxidation reactions and have been preferentially used in commercial practices [7]. Many researchers have reported that the gold nanoparticle deposited on transition metal oxide is highly active in some oxidation reaction [8,9]. These oxides alone are generally also active catalysts. However, a combination with Au nanoparticles produced spectacularly high activity [10]. According to one of the explanations, this is due to the increased number of oxygen vacancies responsible for O_2 activation in the oxides in vicinity of the gold/oxide interface [11]. Palladium on different supports is a well-known oxidation catalyst [10–15]. Therefore, combination of gold and palladium is an interesting candidate to achieve higher activity and to better understand the nature of the outstanding activity of palladium-containing catalysts [16–20]. In this paper, by modifying the Pd (0.5 wt.%)/CeO_2 with a small amount of highly dispersed Au, we successfully synthesized highly active AuPd/CeO_2 catalysts by the routine deposition–precipitation (DP) method with $HAuCl_4$ as the precursor. The resulting AuPd/CeO_2 catalyst demonstrated excellent catalytic activity in complete oxidation of toluene. Complete oxidation of toluene on various catalysts has been studied by many researchers [21–24]. Aromatic compounds are more difficult to oxidize than non-aromatics. This reaction was chosen as the test reaction in this study.

2. Experimental

2.1. Catalyst preparation

The conventional incipient-wetness impregnation method was used to prepare 0.5 wt.% Pd/CeO_2 catalyst with aqueous solution of palladium nitrate (Alfa Aesar) and powder CeO_2 (Nikki), 0.8 ml D.I. water and 0.0126 g palladium nitrate were needed per g CeO_2. The Pd/CeO_2 samples were dried at 80 °C in an oven for 4 h, and then calcined at 350 °C for 8 h. The resulting Pd/CeO_2 was used directly to prepare the Au–Pd/CeO_2 catalyst by the deposition–precipitation (DP) method using $HAuCl_4 \cdot 4H_2O$ as the precursor. The $HAuCl_4 \cdot 4H_2O$ aqueous solution and the ammonia water were slowly co-added into a three-necked bottle containing Pd/CeO_2, with pH controlled at 8. The system was stirred at 65 °C for 24 h, after which the solid was filtered and washed several times. The resulting powder was dried at 80 °C for 24 h, followed by

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calcination at 180 °C for 4 h. The Au/CeO_2 and Pd/CeO_2 catalysts were also prepared similarly for comparison purposes.

2.2. Characterization

The exact palladium and gold content was analyzed with inductively coupled plasma (ICP-MS, PE-SCIEX ELAN 6100 DRC). A microwave (CEM MDS-2000, Matthews, NC, USA) equipped with Teflon vessel was used to digest the powder sample. The XRD experiments were performed using a Siemens D500 powder diffractometer. The XRD patterns were collected using Cu $K\alpha_1$ radiation (1.5405 Å) at a voltage and current of 40 kV and 40 mA, respectively. The sample was scanned over the range 2θ from 20 to 70° at a rate of 0.05°/min to identify the crystalline structure. The morphologies and particle sizes of the samples were determined by transmission electron microscopy (TEM) on a JEM-1200 EX II microscope operated at 160 kV and HRTEM on a JEOL JEM-2010 operated at 160 kV. A small amount of sample was put into the sample tube filled with a 95% ethanol solution. After agitating under ultrasonic environment for 60 min, one drop of the dispersed slurry was dipped onto a carbon coated copper mesh (300#) (Ted Pella Inc., CA, USA), and dried at room temperature in vacuum overnight. The XPS spectra were recorded with a Thermo VG Scientific Sigma Prob spectrometer. The XPS spectra were collected using Al Kα radiation at a voltage and current of 20 kV and 30 mA, respectively. The base pressure in the analyzing chamber was maintained in the order of 10^{-9} torr. The spectrometer was operated at 23.5 eV pass energy and the binding energy was corrected by contaminant carbon (C 1s = 284.5 eV) in order to facilitate the comparisons of the values among the catalysts and the standard compounds.

2.3. Catalytic activity

The oxidation of toluene was performed in a fixed-bed continuous-flow, glass-tubular reactor loaded with 0.2 g catalyst. The reaction was carried out from 50 to 310 °C in intervals of 20 °C. The reactant gas containing air/toluene (8.564 g/m^3 of toluene) was fed into the reactor with a total flow rate of 40 ml/min (space velocity = 10,000/h). The reactor was heated in a temperature-regulated furnace (heating rate = 4 °C/min) and the temperature was measured using a thermocouple placed inside the catalyst bed. After reaching the temperature for 10 min, the product was taken by a 6-port sampling valve and was analyzed by a gas chromatograph equipped with a flame ionization detector using Supel-Q PLOT column (30 m × 0.53 mm ID) with helium as the carrier gas.

3. Results and discussion

3.1. ICP-MS

The ICP-MS results shown in Table 1 show the exact amounts of gold and palladium loadings in the catalysts. In this study, the original amount of palladium was fixed at 0.5 wt.%, and the content of gold was between 0.1 and 1 wt.%. The data show that the amount of palladium did not change a lot after depositing gold, which was within experimental error. It is difficult to gain complete deposition of gold on the support by DP method, as reported in the literature. Only about 70% of gold was deposited by DP method.

Table 1
Actual gold and palladium loadings on support of the catalysts.

Catalyst	Nominal (wt.%)		Actual (wt.%)	
	Au	Pd	Au	Pd
Pd/CeO_2	0	0.5	0	0.508
0.1 wt.% $Au–Pd/CeO_2$	0.1	0.5	0.077	0.476
0.5 wt.% $Au–Pd/CeO_2$	0.5	0.5	0.362	0.442
1.0 wt.% $Au–Pd/CeO_2$	1.0	0.5	0.794	0.466

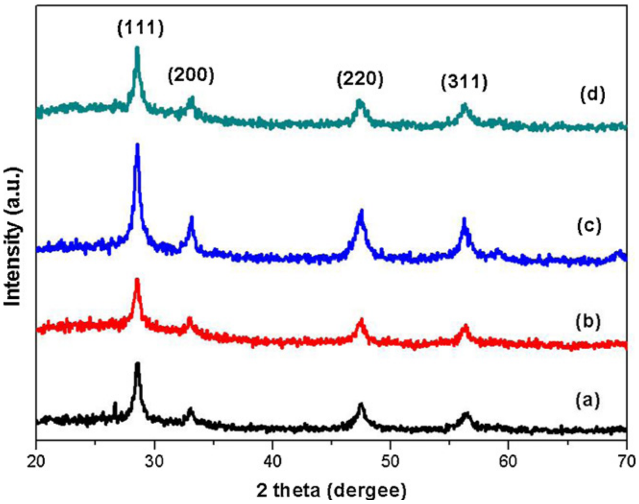

Fig. 1. XRD pattern of (a) Pd/CeO_2, (b) 0.1 wt.% $Au–Pd/CeO_2$, (c) 0.5 wt.% $Au–Pd/CeO_2$, and (d) 1.0 wt.% $Au–Pd/CeO_2$.

3.2. XRD

Fig. 1 shows the XRD patterns of Pd/CeO_2 with different amounts of Au. All catalysts containing CeO_2 support showed intense XRD peaks at $2\theta = 28.6°$ (1 1 1), 33.1° (2 0 0), 47.6° (2 2 0), 56.3° (3 1 1) and 59.1° (2 2 2). No distinct palladium peaks at $2\theta = 40.1°$ (1 1 1) and 46.7° (2 0 0) were observed, because the palladium loading was too small to detect. The peaks at $2\theta = 38.2°$ and 44.5° corresponding to Au metal were not observed in the XRD patterns. This can be attributed to the low Au metal loading in this study.

3.3. TEM and HRTEM

The TEM images of $Au–Pd/CeO_2$ show that the CeO_2 particles are rod-like shape. Fig. 2 shows that the lengh of the CeO_2 particle

Fig. 2. The TEM image of CeO_2 (Nikki).

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