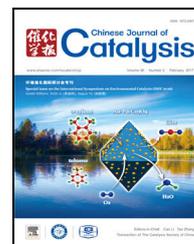


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Effects of hierarchical structure on the performance of tin oxide-supported platinum catalyst for room-temperature formaldehyde oxidation

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ABSTRACT

Flower-like tin oxide-supported platinum (Pt/SnO_x) with a hierarchical structure was synthesized by a hydrothermal method and characterized by XRD, SEM, TEM, high resolution TEM, XPS and nitrogen adsorption. The flower-like Pt/SnO_x microspheres of 1 μm in diameter were composed of staggered petal-like nanosheets with a thickness of 20 nm. Pt nanoparticles (NPs) of 2–3 nm were well dispersed on the SnO_x nanosheets. The catalyst was tested in the catalytic oxidation of gaseous formaldehyde (HCHO) at room temperature, and exhibited enhanced activity compared to Pt NPs supported on commercial SnO and ground SnO_x. HCHO removal of 87% was achieved over the hierarchical Pt/SnO_x after 1 h of reaction, which was 1.5 times that over the ground SnO_x-supported Pt (Pt/g-SnO_x), and the high activity was maintained after six recycles, showing the high stability of this catalyst. HCHO decomposition kinetics was modeled as a second order reaction. The reaction rate constant for Pt/SnO_x was 5.6 times higher than Pt/g-SnO_x. The hierarchical pore structure was beneficial for the diffusion and adsorption of HCHO molecules, and the highly dispersed Pt NPs on the SnO_x nanosheets were the active sites for the oxidative decomposition of HCHO into CO₂ and H₂O. This study provided a promising approach for designing efficient catalysts for indoor HCHO removal at ambient temperature.

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1. Introduction

Formaldehyde (HCHO) is a hazardous pollutant common in indoor air, which is emitted from building and decoration materials [1–3]. In the last decade, many efforts have been made for developing methods for gaseous HCHO removal, including

adsorption [4–6], photocatalytic oxidation [7–11] and catalytic oxidative decomposition [12–29]. Among these methods, catalytic oxidation is appealing because HCHO is continuously decomposed into harmless carbon dioxide (CO₂) and water (H₂O) over well designed catalysts at low temperature and with low energy use [30,31]. Most of the catalysts capable of

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room-temperature HCHO decomposition are supported noble metals, such as platinum (Pt) [12–18], palladium (Pd) [19,20], gold (Au) [21–23] and silver (Ag) [24], while recently non-noble metal materials such as manganese oxide have been reported to have high HCHO decomposition activity at room temperature [25–27].

Besides the selection of the active component with an intrinsic high activity [21,32], various strategies have been adopted to improve the performance of catalytic HCHO oxidation, such as increasing the dispersion of the noble metal, removing residual halogen ions, enhancing metal-support interaction, and increasing HCHO adsorption affinity (e.g. by introducing a higher amount of surface hydroxyl) [16,19,33]. In addition to the above factors, which focus on the chemical properties of the catalyst, the diffusion of reactants and products to and from the catalyst interior also plays a key role in determining the rate of catalytic reactions [34,35]. Thus, materials with a hierarchical structure of different length scales have come to the attention of researchers, due to the efficient mass transfer within the hierarchical architecture [36,37]. Hierarchical micro- and nanostructures have recently attracted considerable interest in many fields, such as energy conversion and storage [38,39], gas sensor [40], adsorptive removal of water pollutants [41,42], as well as environmental catalysis and photocatalysis [8,36,43–50].

Catalysts with hierarchical structures can take various forms, e.g. a nanofiber mat [36], assembly of lab-synthesized or commercial nanoparticles [8,37], and open-ended macroporous channels [43]. These different morphologies would influence the catalytic performance [44]. Here, we report the fabrication of flower-like tin oxide-supported platinum (Pt/SnO_x) with a hierarchical structure for the catalytic oxidation of HCHO into CO₂ and H₂O at room temperature. The hierarchical SnO_x with the flower-like morphology was beneficial for the transport and adsorption of HCHO molecules, and the highly dispersed Pt nanoparticles (NPs) were active components for HCHO decomposition. This research can help the design of a catalyst for efficient air purification at ambient temperature.

2. Experimental

2.1. Preparation of catalysts

All chemicals were analytical grade and used without further purification. In a typical procedure to synthesize the flower-like SnO_x, 3.2 mmol of SnF₂ and 12.8 mmol of urea were added into 160 mL of deionized water and stirred for 0.5 h. The mixed solution was then transferred into a dried 200 mL Teflon-lined stainless steel autoclave and kept at 180 °C for 18 h. After the hydrothermal reaction, the grey precipitate was collected by centrifugation, washed with distilled water and ethanol three times, and dried in an oven at 80 °C for 6 h.

To investigate the role of the hierarchical structure on HCHO oxidation performance, two additional types of SnO_x were used as the catalyst support for comparison, namely, a commercial tin(II) oxide (SnO) (denoted as c-SnO) and ground

flower-like SnO_x (g-SnO_x). Specifically, to prepare the g-SnO_x sample, a portion of the flower-like SnO_x was suspended in ethanol and ground in a ball mill (QM-3SP04) for 6 h.

To synthesize the SnO_x-supported Pt catalyst, 1 g of the SnO_x support (flower-like SnO_x, c-SnO or g-SnO_x) was dispersed into 10 mL of deionized water by ultrasonic treatment, and 2.65 mL of H₂PtCl₆·6H₂O solution (1 wt%) was added into the suspension under magnetic stirring. After impregnation for 1 h, 2.5 mL of a mixed solution of NaBH₄ (0.1 mol/L) and NaOH (0.5 mol/L) was quickly added into the suspension under vigorous stirring for 0.5 h. The precipitate obtained was washed with deionized water twice, and then dried at 80 °C for 12 h. The samples obtained with the different SnO_x supports were denoted as Pt/SnO_x, Pt/c-SnO and Pt/g-SnO_x, respectively.

2.2. Characterization

The morphology and structure of the samples were characterized by field emission scanning electron microscope (FESEM) (S-4800), transmission electron microscopy (TEM) (JEOL-JEM-2100F) and high resolution TEM (HRTEM) (JEM-2100). The elemental composition of the Pt/SnO_x sample was determined by an energy dispersive X-ray spectroscopy (EDX) module attached to the TEM. The phase structure was analyzed by X-ray diffraction (XRD) (Rigaku, D/Max-RB, Japan) with Cu K_α radiation at a scan rate (2θ) of 0.05°/s. The surface chemical state of the samples was analyzed by X-ray photoelectron spectroscopy (XPS) (VG ESCALAB 210). The Brunauer-Emmett-Teller (BET) specific surface area and pore volume were measured by N₂ adsorption at -196 °C (Micromeritics ASAP 2020). The pore size distribution was calculated from the adsorption branch of the N₂ adsorption isotherm by the Barret-Joyner-Halender (BJH) method.

2.3. Catalytic test

The catalytic HCHO oxidation tests were performed in the dark at room temperature (25 °C). In each experiment, 0.1 g of catalyst was dispersed on the bottom of a glass Petri dish (8 cm diameter), which was placed in a 6 L organic glass box and covered with a glass slide. Then, the box reactor was sealed, and 12 μL of condensed HCHO solution (38%) was injected into the reactor. A fan was turned on to accelerate the volatilization of HCHO, and the HCHO concentration in the reactor was stabilized at 172–177 ppm after 1 h. Then, the glass slide on the Petri dish was removed to start the catalytic oxidation reaction. The concentrations of HCHO, CO₂, CO and water vapor were monitored online with a photoacoustic field gas monitor (INNOVA AirTech Instruments, Model 1412). The HCHO removal ratio (η) was calculated by equation:

$$\eta(\%) = \frac{C_0 - C_{\text{final}}}{C_0} \times 100 \quad (1)$$

where C₀ is the initial HCHO concentration, and C_{final} is the final HCHO concentration.

3. Results and discussion

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