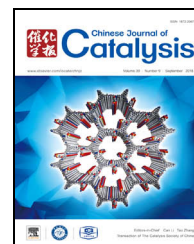


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Article

Pt supported on octahedral Fe₃O₄ microcrystals as a catalyst for removal of formaldehyde under ambient conditions

Weiyi Cui ^{a,b}, Dan Xue ^c, Naidi Tan ^a, Bin Zheng ^b, Mingjun Jia ^{b,*}, Wenxiang Zhang ^{b,#}^a Key Laboratory of Chemical Cleaner Production Technology of Jilin Province, Jilin Institute of Chemical Technology, Jilin 132022, Jilin, China^b Key Laboratory of Surface and Interface Chemistry of Jilin Province, College of Chemistry, Jilin University, Changchun 130021, Jilin, China^c School of Chemistry & Chemical Engineering, Xi'an Shiyou University, Xi'an 710065, Shaanxi, China

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ABSTRACT

Several catalysts comprising Pt supported on octahedral Fe₃O₄ (Pt/Fe₃O₄) were prepared by a facile method involving co-precipitation followed by thermal treatment at different temperatures. A variety of characterization results revealed that this preparation process afforded highly crystalline octahedral Fe₃O₄ with a uniform distribution of Pt nanoparticles on its surface. The thermal-treatment temperature significantly influenced the redox properties of the Pt/Fe₃O₄ catalysts. All the Pt/Fe₃O₄ catalysts were found to be catalytically active and stable for the oxidation of low-concentration formaldehyde (HCHO) with oxygen. The catalyst prepared by thermal treatment at 80 °C (labelled Pt/Fe₃O₄-80) exhibited the highest catalytic activity, efficiently converting HCHO to CO₂ and H₂O under ambient temperature and moisture conditions. The excellent performance of Pt/Fe₃O₄-80 was mainly attributed to beneficial interactions between the Pt and Fe species that result in the formation a higher density of active interface sites (e.g., Pt-O-FeO_x and Pt-OH-FeO_x). The introduction of water vapor improves the catalytic activity of the Pt/Fe₃O₄ catalysts as it participates in a water-assisted dissociation process.

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1. Introduction

Eliminating formaldehyde (HCHO) from indoor air is extremely important to meet stringent environmental regulations and preserve human health [1–4]. The catalytic oxidation of HCHO with oxygen at low temperature is an innovative and promising technique for this purpose owing to its high removal efficiency, low energy consumption, and lack of secondary pollution [5–8]. Among the numerous catalysts investigated for this process [9–19], those comprising Pt supported on iron-based oxides, such as Fe₂O₃, Fe₃O₄, and ferrihydrite, are extremely attractive owing to their remarkable activities in

HCHO oxidation at low temperatures [17–19].

It has been previously reported that Pt/Fe₂O₃ catalysts prepared by the colloid deposition method exhibit much higher catalytic activities for room-temperature oxidation of HCHO than those prepared by co-precipitation or impregnation method [17,18]. The effects of various factors on the catalytic performance of Pt/Fe₂O₃ catalysts have been studied, and the existence of beneficial interactions between the Pt and Fe species therein has been proposed to lead to the formation of highly efficient active interface sites. For example, Yan et al. [19] reported that mesoporous ferrihydrite and Fe₃O₄ nanoparticles obtained by a microemulsion-assisted method could

* Corresponding author. Tel: +86-431-85155390; Fax: +86-431-85168420; E-mail: jiamj@jlu.edu.cn# Corresponding author. Tel: +86-431-85155390; Fax: +86-431-85168420; E-mail: zhwenx@jlu.edu.cn

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be used as support materials in the preparation of highly active supported-Pt catalysts for HCHO oxidation. They suggested that the main features responsible for the superior catalytic activities of these supported-Pt catalysts were their surface hydroxyl groups, high Pt dispersions, and excellent adsorption performance.

Recently, significant progress has been made on the controlled synthesis of Fe_3O_4 materials with different crystallographic forms and morphologies, such as spheres, cubes, wires, nanorods, and octahedra [20–24]. Of these, well-defined octahedral Fe_3O_4 sub-microcrystal materials have drawn particular attention for their potential applications in environmental remediation and heterogeneous catalysis [25–27]. The thermodynamically stable octahedral Fe_3O_4 can provide an interface on which to assemble nanoparticles with high packing efficiency and structural stability. Furthermore, the bulky Fe_3O_4 crystallites are redox active, and their surface compositions can be adjusted by slight changes in oxygen partial pressure and substrate temperature. These features present numerous opportunities to develop highly efficient and thermally stable Fe_3O_4 -based catalysts for application in many important processes [28–31].

In the current study, Pt catalysts supported on octahedral Fe_3O_4 crystals ($\text{Pt}/\text{Fe}_3\text{O}_4$) were prepared by a simple co-precipitation/thermal treatment method, and their catalytic properties were investigated for the low-temperature oxidation of HCHO. The effect of thermal-treatment temperature on the physicochemical properties and catalytic performance of the $\text{Pt}/\text{Fe}_3\text{O}_4$ catalysts was investigated using a variety of characterization techniques. As a result, it was found that the $\text{Pt}/\text{Fe}_3\text{O}_4$ catalyst thermally treated at a relatively low temperature (80 °C) exhibited very high catalytic activity and stability in HCHO oxidation under ambient temperature and moisture conditions.

2. Experimental

2.1. Catalyst preparation

$\text{Pt}/\text{Fe}_3\text{O}_4$ catalysts were synthesized through a simple co-precipitation method previously reported in the literature with some modifications [25,32]. Typically, 5.189 g $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ was dissolved in 10 mL water and then mixed with 1.33 mL of a solution of $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$ (15 g/L) in a 250-mL three-neck round-bottom flask. The solution was then heated at 90 °C for 10 min to form a homogeneous solution. This mixture was then added to 100 mL KOH solution and vigorously stirred for 12 h. Finally, the resultant precipitate was washed with deionized water, separated via centrifugation, and dried at 80 °C for 6 h without any further thermal treatment. This as-prepared catalyst was labelled $\text{Pt}/\text{Fe}_3\text{O}_4$ -80. Two more catalysts were obtained by thermal treatment of the above material at 200 and 300 °C in static air, and denoted as $\text{Pt}/\text{Fe}_3\text{O}_4$ -200 and $\text{Pt}/\text{Fe}_3\text{O}_4$ -300, respectively. The Pt loading was approximately 1.0 wt% for each catalyst. A reference sample of the Fe_3O_4 support alone was also prepared by the same method but without the addition of $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$.

2.2. Catalyst characterization

Powder X-ray diffraction (XRD) patterns were obtained using a D/Max-rA X-ray diffractometer equipped with nickel-filtered $\text{Cu } K_\alpha$ radiation. ^{57}Fe Mössbauer spectra were obtained using an OIMS-500 Mössbauer spectrum instrument. ^{57}Co (Pd) was used as the γ -ray radioactive source and the velocity was calibrated against a standard α -iron foil. Magnetic measurements were performed on a SQUID-VSM magnetometer at room temperature. Transmission electron microscopy (TEM) images and high-angle annular dark field STEM (HAADF-STEM) images were acquired using a FEI Tecnai F20 EM operated at 200 kV and equipped with an energy-dispersive spectroscopy analyzer. N_2 -adsorption-desorption isotherms were obtained using a Micromeritics analyzer (ASAP 2010N). The Brunauer-Emmett-Teller (BET) model was used to calculate the surface areas. X-ray photoelectron spectroscopy (XPS) spectra were collected using an ESCALAB250 spectrometer with $\text{Al } K_\alpha$ radiation as the excitation source. All the elemental binding energies were corrected by adjusting the C 1s peak to 284.6 eV. Temperature-programmed reduction by H_2 (H_2 -TPR) analysis was performed by an adsorption instrument equipped with a thermal conductivity detector (TCD). For each analysis, the samples were purged in a flow of Ar at 100 °C for 30 min. The H_2 -TPR experiments were performed using a 5% H_2/Ar mixture (30 mL/min) over 0.1 g of catalyst at a heating rate of 10 °C/min. The Fourier-transform infrared (FTIR) spectra of the samples were recorded using KBr disks on a Nicolet 6700 spectrometer.

2.3. Catalytic tests

The HCHO oxidation tests were carried out in a quartz tube reactor containing 0.1 g of solid catalyst (40–60 mesh). The gas mixture consisted of 400 ppm HCHO, 20 vol% O_2 , a certain amount of water vapor chosen to provide a relative humidity (RH) of 0–80%, and balanced with N_2 . Gaseous HCHO was generated by flowing N_2 (coming from a mass flow controller) through aqueous formaldehyde kept in an incubator. Products and reactants were analyzed using a Techcomp GC-7900 gas chromatographer equipped with a TCD, and the catalytic activity of the catalyst under inspection was evaluated by the conversion of formaldehyde to CO_2 .

3. Results and discussion

3.1. Characterization of the catalysts

The XRD patterns of the three $\text{Pt}/\text{Fe}_3\text{O}_4$ catalysts ($\text{Pt}/\text{Fe}_3\text{O}_4$ -80, $\text{Pt}/\text{Fe}_3\text{O}_4$ -200, and $\text{Pt}/\text{Fe}_3\text{O}_4$ -300) as well as the Fe_3O_4 support alone are presented in Fig. 1. All the samples show diffraction peaks characteristic of inverse spinel magnetite iron oxide (Fe_3O_4 , JCPDS 65-3107) located at $2\theta = 30.2^\circ$, 35.6° , 43.3° , 53.8° , 57.3° , 63.0° , and 78.3° , which can be attributed to the (220), (311), (400), (422), (511), (531), and (533) planes of Fe_3O_4 , respectively [25,29]. For the three $\text{Pt}/\text{Fe}_3\text{O}_4$ catalysts, the intensity of the diffraction peaks as-

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