



Full Length Article

Acid-treated TiO₂ nanobelt supported platinum nanoparticles for the catalytic oxidation of formaldehyde at ambient conditions



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ABSTRACT

In this work, two kinds of novel Pt/TiO₂ catalysts were prepared by impregnation method using hydrothermally synthesized TiO₂ nanobelt (TiNB) and sulfuric acid-treated TiO₂ nanobelt (TiNB-ac) as supports, and their catalytic properties were investigated in the oxidation of formaldehyde. It was found that Pt/TiNB-ac exhibits much higher catalytic activity, which can efficiently convert formaldehyde to CO₂ and H₂O at ambient temperature. The addition of water vapor into the feed stream can further promote the catalytic activity of Pt/TiNB-ac catalyst. A variety of characterization results showed that TiNB-ac possesses much rougher surface, and more defect sites (including abundant chemisorbed oxygen and surface hydroxyl species) due to the treatment by sulfuric acid. These features should be beneficial to achieve high dispersion of Pt nanoparticles on the rough-surface of TiNB-ac, to produce more interface active sites like Pt-O(OH)_x-Ti species through the interaction between the Pt nanoparticles and the surface hydroxyl species of TiNB-ac support, thus resulting in the formation of highly efficient Pt/TiNB-ac catalyst for the oxidation of formaldehyde under mild conditions.

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1. Introduction

As a major indoor air pollutant, formaldehyde (HCHO) has been classified as a human carcinogen Group 1 of hazardous substances by International Agency for Research on Cancer (IARC) [1–4]. For removing trace HCHO in air, different methods have been developed, including adsorption [5,6], plasma [7], degradation [8], photo-catalysis [9,10] and catalytic oxidation [11–36]. Among them, catalytic oxidation has been regarded as one of the most efficient and environmentally benign technologies. So far, a variety of supported noble metal catalysts (e.g., Au [11,12], Pt [13–31], Pd [32,33], Ag [34,35]) have typically been employed for the removal of HCHO through the catalytic oxidation process. By selecting suitable supports and optimizing preparation conditions, some highly active supported Pt nanoparticles catalysts such as Pt/TiO₂, Pt/Fe₂O₃, Pt/AlOOH, Pt/CeO₂-AlOOH, and PtNi(OH)_x/Al₂O₃ have been developed, and part of them could even achieve excellent catalytic property at ambient temperatures [13–31].

A few literature works have shown that the morphology, the microstructure and the surface properties of TiO₂ supports can sig-

nificantly influence the catalytic performance of TiO₂ supported Pt catalysts for HCHO oxidation [18,20–23,30]. For instance, by using TiO₂ nanosheet with exposed (001) facet or using mat-like hierarchically porous TiO₂ nanofiber as support, Nie et al. [18,22] obtained two kinds of Pt/TiO₂ catalysts with high catalytic activity for room-temperature oxidation of HCHO. Chen et al. [20,21] reported that highly efficient Pt/TiO₂ catalyst could be prepared using ordered pore-through TiO₂ nanotube arrays as supports. Qi et al. [30] found that mesoporous TiO₂ hollow chain supported Pt catalyst exhibits very high catalytic activity toward HCHO oxidation. These results suggested that changing the microstructure and the surface properties of TiO₂ supports can influence the dispersion of Pt nanoparticles, and adjust the interaction between Pt and TiO₂, which may finally enhance the catalytic performance for HCHO oxidation. At present, it is still an interesting subject to explore how the surface chemistry and the structure factors of TiO₂ support could influence the nature of active sites of Pt/TiO₂ catalysts, and what are the main features of a highly efficient Pt/TiO₂ catalysts for the low-temperature oxidation of HCHO.

In this work, we tried to use TiO₂ nanobelt (TiNB) and sulfuric acid-treated TiO₂ nanobelt (TiNB-ac) as supports to prepare two kinds of Pt/TiO₂ catalysts (Pt/TiNB and Pt/TiNB-ac) for the application in low-temperature oxidation of HCHO. Previous literatures have shown that TiO₂ nanobelt with high surface-to-volume

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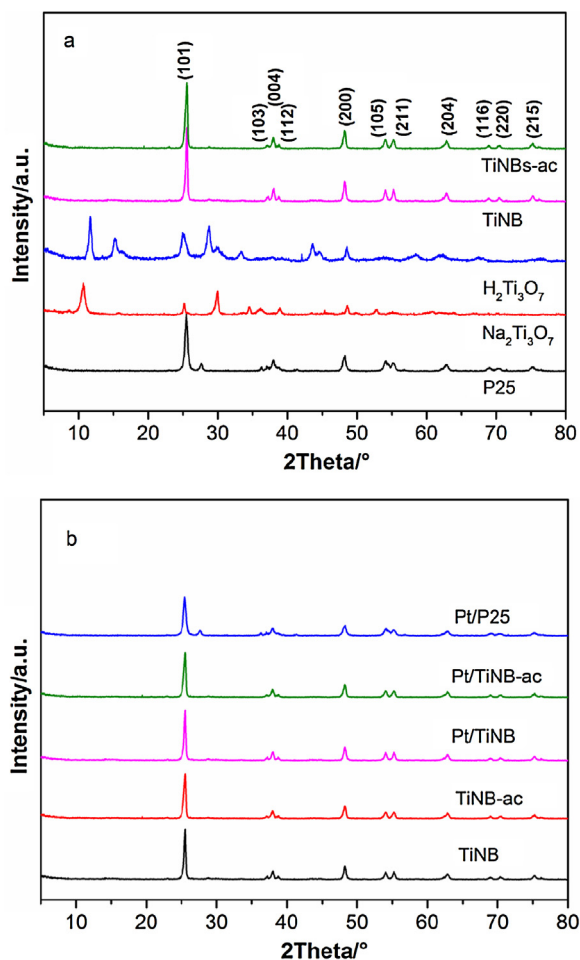


Fig. 1. X-ray diffraction patterns for all samples.

ratio and defined crystalline orientation can be used as suitable support to prepare efficient noble metal supported catalysts for some significant catalytic processes [37–39]. More interesting, the surface chemistry of TiO_2 nanobelt could be further adjusted by acid-treatment, which may produce more rough and activated surface species for special application [37,40]. Here, we found that the Pt/TiNB-ac catalyst derived from the acid-treated TiO_2 nanobelt exhibits excellent catalytic activity toward HCHO oxidation at room temperature. The existence of abundant hydroxyl species on the surface of TiNB-ac can provide more anchor sites for achieving high dispersion of Pt nanoparticles, and can also produce more interface active sites like $\text{Pt-O(OH)}_x\text{-Pt}$ species through the metal-support interaction, which can finally resulting in the formation of highly efficient Pt/ TiO_2 catalyst for the oxidation of HCHO at ambient conditions.

2. Experimental section

2.1. Catalyst preparation

TiO_2 nanobelt materials were synthesized through a simple hydrothermal procedure reported in literatures [37,39]. Typically, 0.6 g TiO_2 (Degussa P-25, composed of 80% anatase and 20% rutile crystallites of TiO_2) was mixed with 100 ml of 10 M NaOH aqueous solution, followed by hydrothermal treatment at 200°C in a 150 ml Teflon-lined autoclave for 72 h. The product of $\text{Na}_2\text{Ti}_3\text{O}_7$ nanobelt was collected and washed thoroughly with deionized water until a pH value of about 7 is reached. The washed samples were immersed

in 0.1 M HCl aqueous solution for 24 h and then washed thoroughly with deionized water to get $\text{H}_2\text{Ti}_3\text{O}_7$ nanobelt. After dried at 70°C for 10 h, $\text{H}_2\text{Ti}_3\text{O}_7$ nanobelt was thermally annealed at 600°C for 3 h to produce TiO_2 nanobelts (TiNB). Sulfuric acid-treated TiNB (denoted as TiNB-ac) were prepared through an acid-assisted hydrothermal approach [37,40]. Typically, 0.6 g of TiNB samples were added in 100 ml of 0.02 M H_2SO_4 aqueous solution under vigorous stirring, and the mixture was transferred to Teflon-lined autoclave and kept at 100°C for 12 h. After filtrating, washing and drying, the resultant TiNB-ac materials were obtained via calcining the solid powder at 600°C for 3 h.

Pt/TiNB and Pt/TiNB-ac catalysts were prepared by impregnation method. Typically, 0.6 g of TiNB or TiNB-ac sample was added into H_2PtCl_6 aqueous solution under vigorous stirring. After impregnation for 1 h, 4.0 ml of a mixed aqueous solution containing NaOH (0.5 M) and NaBH_4 (0.1 M) were quickly added into the slurry and vigorously stirred for another 1 h. After that, the slurry was evaporated at 80°C under stirring. Finally, the solid sample was dried at 80°C for 12 h. For comparison, a Pt-supported P25 catalyst (Pt/P25) was also prepared under similar conditions by using the commercial Degussa P25 as support. The loading of Pt is approximately 1.0 wt% for each catalyst, which is calculated from the amount of H_2PtCl_6 used for catalyst preparation.

2.2. Catalyst characterization

The X-ray diffraction (XRD) analyses of the catalysts were carried out using a D/Max-rA X-ray diffractometer operated at 30 kV and 40 mA employing nickel-filtered Cu K α radiation. Transmission electron microscopy (TEM) images and the high-angle annular darkfield STEM (HAADF-STEM) images were obtained using a FEI Tecnai F20 EM operated at 200 kV and equipped with an energy-dispersive spectroscopy analyzer. $\text{H}_2\text{-O}_2$ titration experiments were carried out on a Builder PCA-1200 Pulse Chemisorption System to determine the Pt dispersion of the catalysts. The X-ray photoelectron spectra (XPS) measurements were carried out on an ESCALAB250 X-ray photoelectron spectrometer with Al K α radiation as excitation source. The XPS spectra were corrected by adjusting the C 1s peak to a position of 284.6 eV. Temperature programmed reduction by H_2 measurements ($\text{H}_2\text{-TPR}$) were carried out using an adsorption instrument equipped with a TCD. The samples were loaded and pretreated with Ar at 100°C for 30 min. The $\text{H}_2\text{-TPR}$ experiment was performed under the mixture of 5% H_2 in N_2 flow (30 ml/min) over 0.1 g of catalyst at a heating rate of $10^\circ\text{C}/\text{min}$.

2.3. Catalytic test

For the HCHO oxidation tests, 0.1 g of solid catalyst (40–60 mesh) was loaded in a quartz tube reactor. The gas mixture consisted of 400 ppm HCHO, 20 vol% O_2 , and a certain amount of water vapor (with relative humidity RH of 0–30%), and balanced by N_2 . Gaseous HCHO was generated by flowing N_2 (coming from a mass flow controller) through aqueous formaldehyde in an incubator. Products and reactants were analyzed by Techcomp GC-7900 gas chromatograph equipped with TCD detector, and the catalytic activity of the catalysts was evaluated by the conversion of formaldehyde to CO_2 .

3. Results and discussion

3.1. Characterization of the catalysts

XRD patterns of TiO_2 nanobelts and their precursors are presented in Fig. 1. The samples of $\text{Na}_2\text{Ti}_3\text{O}_7$ nanobelt and $\text{H}_2\text{Ti}_3\text{O}_7$ nanobelt show some intense and sharp diffraction peaks, which

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