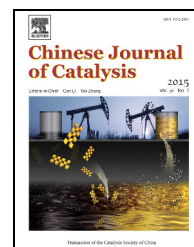


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Article

Titania-supported Pt catalyst reduced with HCHO for HCHO oxidation under mild conditions

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ABSTRACT

A series of Pt/TiO₂(P25) catalysts were prepared by both impregnation (IM) and deposition-precipitation (DP) methods followed by reduction using either a HCHO solution or a H₂ stream. The effects of the preparation and reduction conditions and the Pt loading level on structural properties and performance during HCHO oxidation were then studied based on the assessment of adsorbed species on the catalyst surfaces using *in-situ* diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS). The results indicated that Pt/P25 prepared by DP and reduced with HCHO exhibited homogeneously dispersed Pt nanoparticles with appropriate particle size in addition to a high concentration of chemisorbed oxygen, and also showed high activity for HCHO oxidation. A HCHO conversion of 98% with stable performance over more than 100 h was achieved over Pt/P25 produced using DP and HCHO reduction at 30 °C, even with a 0.4% Pt loading. Pt/P25 synthesized using DP with H₂ reduction was less effective at promoting formate decomposition and thus showed less HCHO oxidation activity, likely because of lower levels of chemisorbed oxygen. The Pt/P25 made using IM and H₂ reduction had the highest amount of chemisorbed oxygen but also the largest Pt particles, resulting in the lowest activity for both the formation of formate species and formate decomposition into CO species, and thus showed low HCHO oxidation activity.

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1. Introduction

Formaldehyde (HCHO) is one of the most common toxic volatile organic compounds; it is known to be carcinogenic and may cause leukemia [1]. Consequently, many methods have been proposed to remove trace levels of HCHO in air, including adsorption, photocatalysis, and catalytic oxidation [2]. Among these, catalytic oxidation is one of the most effective and economically feasible technologies because HCHO can be oxidized to CO₂ over catalysts at much lower temperatures than those are required for thermal oxidation [3]. The catalytic decompo-

sition of HCHO at room temperature is particularly desirable because of the attendant environmentally friendly reaction conditions. Recently, significant progress has been achieved with regard to decreasing the reaction temperature necessary for HCHO oxidation by employing Pt catalysts with various supports [4–11], especially when using titania-supported Pt catalysts.

Despite this success, Pt/TiO₂ materials are frequently reported to exhibit inconsistent activity for HCHO oxidation even when identical Pt loadings and TiO₂ support structures are applied. As an example, Peng and Wang [7] reported that the

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optimal Pt loading in a series of Pt/TiO₂ catalysts reduced in H₂ at 450 °C was 0.6%. The room temperature HCHO conversions obtained in their study were 0.8%, 1.0%, and 41.5% over 0.01%, 0.1%, and 0.6% Pt/TiO₂, respectively, while the conversion obtained with 1% Pt/TiO₂ dropped to 14.3%, although this value increased to 97% at 120 °C. In contrast, Zhang and He [6] reported that HCHO could be completely oxidized into CO₂ and H₂O at room temperature over Pt/TiO₂ with a relatively high 1% Pt loading level. Recently, Huang et al. [12] found that nearly 100% conversion of HCHO was achieved over sodium borohydride-reduced Pt/TiO₂ catalysts even with a low Pt loading of 0.1%.

Differences in experimental parameters, such as the Pt loading method, the TiO₂ support properties, and the reduction procedure, have been suggested to account for such discrepancies. Huang et al. [12] prepared a series of Pt/TiO₂ catalysts by the impregnation and deposition-precipitation method while applying different reduction processes and evaluated their activity for the catalytic decomposition of HCHO at room temperature. This study showed that the reduction treatment had a significant effect on the structural properties and HCHO oxidation activity of the Pt/TiO₂ catalysts. The authors concluded that well-dispersed and negatively charged metallic Pt nanoparticles together with high levels of chemisorbed oxygen were likely responsible for the high catalytic activity of the NaBH₄-reduced Pt/TiO₂ catalysts. However, further study is still required to relate the catalyst preparation and reduction conditions to the structural properties of the catalyst, and to elucidate the detailed mechanism leading to the high catalytic activity and stability of these catalysts. Moreover, because some of the above catalysts were reduced in a NaBH₄ solution, the finished materials may have contained residual Na and B [12], which could have had various effects on the performance of the catalysts. Zhang et al. [3] reported that the presence of Na⁺ had a positive effect on the catalytic activity of Pt/TiO₂, to the extent that it changed the HCHO catalytic oxidation mechanism. Thus, in those catalysts contaminated with residual ions, it is difficult to ascertain which factors are actually affecting the performance of the catalyst. Recently, Qu et al. [13] reported that Ag/SBA-15 catalyst treated with a HCHO solution also showed high activity for HCHO oxidation. In comparison with NaBH₄ solution reduction, reduction with HCHO solution leaves no additional elements on the treated catalyst, which provides an opportunity to study the effects of the reduction conditions on the structure, properties, and catalytic performance of the catalyst. To the best of our knowledge, the open literature contains very few reports focusing on the HCHO reduction process and its effects on the structure, properties, and catalytic performance of Pt-based catalysts.

The present paper reports the synthesis of a highly efficient Pt/TiO₂ catalyst intended for HCHO oxidation by a deposition-precipitation (DP) method followed by reduction via treatment with HCHO. A detailed study of the effects of the Pt/TiO₂ preparation and reduction conditions and the Pt loading amount on the structural properties and performance during trace HCHO oxidation is presented. The mechanism responsible for the high catalytic activity and stability of these materi-

als is examined using *in-situ* diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS), which represents a powerful technique for the investigation of surface species in real-world conditions [6,14].

2. Experimental

2.1. Catalyst preparation

Two different Pt loading methods were used to prepare Pt/TiO₂ catalysts, the impregnation method and DP method, producing materials denoted as Pt/P25(IM) and Pt/P25(DP), respectively. The Pt/P25(IM) catalyst was prepared by uniformly dispersing P25 powder (Degussa) into a H₂PtCl₆ solution (H₂PtCl₆·6H₂O, Alfa Aesar) with the appropriate H₂PtCl₆ concentration. After vigorous stirring, the sample was washed with distilled water and centrifuged three times to remove any impurities on the surface of the catalyst. The catalyst was then dried overnight at 120 °C to evaporate the solvent. The Pt/P25(DP) catalysts were prepared by first dispersing the P25 support in deionized water, following which a H₂PtCl₆ solution was added dropwise into the P25 slurry with vigorous stirring at 70 °C. After stirring for 1 h, the pH of the solution was adjusted to 8 by adding an aqueous NaOH solution, and deposition by precipitation was allowed to continue for 2 h under vigorous stirring at 70 °C. The catalyst was subsequently washed and centrifuged three times to remove any surface impurity, and the resulting powder was dried overnight at 120 °C. Two different reduction methods were used when preparing catalysts, H₂ reduction and HCHO solution reduction, producing specimens referred to as Pt/P25(IM-H₂), Pt/P25(DP-H₂), and Pt/P25(DP-HCHO). During a typical H₂ reduction process, the as-prepared Pt/P25(IM) or Pt/P25(DP) was calcined in a H₂ stream (40 ml/min) at 300 °C for 3 h. Pt/P25(DP-HCHO) was prepared using a deposition-precipitation method similar to that described above, the only difference being that a HCHO solution (37%–40%, Guangzhou Reagent Company) was added to the DP solution after the pH of the solution was adjusted. Note that the Pt loading levels in this work are expressed as the mass percent of Pt in the catalyst, as confirmed by inductively coupled plasma (ICP, IRIS (HR)) elemental analysis.

2.2. Catalyst characterization

In-situ DRIFTS analyses were carried out using an EQUINOX-55 FFT instrument (Bruker) equipped with a diffuse reflectance accessory and an MCT detector. During each analysis, a finely ground sample (ca. 10 mg) was placed in a ceramic crucible within the *in-situ* chamber. A 100 ml/min flow of nitrogen gas containing HCHO was then passed through the chamber. Spectra under reaction conditions were recorded by averaging 64 scans with a resolution of 4 cm⁻¹. Metal nanoparticle size distributions were observed using a JEOL 2100F transmission electron microscope (TEM), and the compositions of the samples were characterized by an energy-dispersive spectrometer fitted to a JEM-2010HR TEM. BET surface areas of the samples were determined by N₂ adsorption at -196 °C

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