



Short Communication

The oxidation of carbon monoxide over the palladium nanocube catalysts: Effect of the basic-property of the support



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ABSTRACT

The nanocatalysts of Pd nanocubes supported on SiO₂, TiO₂ and MgO were prepared and the CO oxidation activities over the three catalysts were evaluated. The acid–basic properties of the support materials were determined using the temperature-programmed desorptions of NH₃ (NH₃-TPD) and CO₂ (CO₂-TPD). The CO adsorptions over the three catalysts were investigated by the diffuse reflectance infrared Fourier transform (DRIFT). It was found that the Pd/MgO catalyst showed the strongest ability to adsorb CO molecules and performed best for CO oxidation.

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1. Introduction

The catalytic activity of supported metal catalysts can be influenced by the intrinsic properties (e.g., shape, size, and chemical valence) of metal nanoparticles (NPs) and the nature of support [1–9]. It is generally accepted that different types of supports can lead to different reaction mechanisms and different catalytic performances.

In 1970s, Tauster and co-workers [10] observed the “strong metal-support interaction (SMSI)” effect, which exerted an impact on the ability for adsorption of CO and H₂ molecules over catalysts with metal NPs supported on TiO₂, and consequently modified the catalytic properties. It is well known that the electron transfer phenomenon is involved in catalytic process, and the acid–basic property of the support is the crucial factor affecting the electron transfer, therefore, the effect of the acid–basic property of the support on the catalytic performance should be emphasized. As Vishwanathan and his co-workers reported [11], alkali promotion on phenol hydrogenation activity of Pd/Al₂O₃ rooted in the changed acid–base property and the altered net charge density at the reactive site. For Pd based three-way catalyst [12] and Pt/Al₂O₃ [13], the basic-property or the electron-donating ability of the basic additives or the support itself played an important role in the improved catalytic performances.

For CO oxidation, the adjustment in electronic structure of metal NPs induced by the support can directly alter the adsorption intensity of surface CO molecules, thus influencing the catalytic performances over the supported catalyst. In this study, Pd NPs with nanocube-like morphology were employed as the active phase, and materials with

different basic-property, i.e. SiO₂, TiO₂, and MgO, were used as the supports. The generated supported Pd catalysts were investigated to explore the effect of basic-property of support on the catalytic performance for oxidation of CO and the correlations of support nature, metal-support electron transfer with the catalytic performance were induced.

2. Experimental

2.1. Synthesis of Pd nanocubes

The synthesis procedure adopted was based on the work of Niu et al. [14] with minor modifications. In order to remove the excess CTAB, the as-prepared Pd nanocubes colloid was rinsed using the “deposition–redispersion strategy” [15]. The detailed synthesis and rinsing procedure were described in the Supplementary data.

2.2. Preparation of supported Pd catalysts

The supported Pd catalysts were prepared by a modified wetness impregnation method. Typically, a certain amount of SiO₂, TiO₂ or MgO was mixed with the desired amount of Pd nanocube ethanol solution under stirring to obtain black slurry. The excess ethanol was then removed by evaporation, and the resulted wet gray solid was dried at 80 °C overnight, thus giving the supported Pd nanocube catalysts, in which the theoretical Pd loading was 2.0 wt.%.

2.4. Evaluation of catalytic activity

The CO pulse oxidation over the supported Pd catalysts was carried out in a fixed-bed quartz microreactor. In the catalytic activity evaluation,

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the mixture gas of CO and O₂ was pulsed over the catalyst (50 mg, 40–60 mesh) through a six-way sampling valve (20 μL at a time) with He as the carrier gas. The concentrations of the reactants and products were monitored on-line by a quadrupole mass spectrometer (Hiden, HPR20).

2.5. Catalyst characterization

Transmission electron microscopic (TEM) images and selected-area electron diffraction (SAED) patterns of the catalysts were taken

on a JEOL transmission electron microscope (JEM-2100) operated at 200 kV. The actual Pd loading of the catalysts were determined by X-ray fluorescence spectrometer (Magix-PW2403). Besides, the catalysts were characterized using the techniques of X-ray diffraction (XRD) on an Advance X-ray diffractometer (Bruker-AXS D8) and N₂ adsorption–desorption on a physisorption apparatus (Micromeritics ASAP 2020). The temperature-programmed desorptions of NH₃ (NH₃-TPD) and CO₂ (CO₂-TPD) as well as O₂–H₂ titration experiments were carried out on a chemisorption analyzer (Micromeritics Autochem II 2920). The acid–basic properties of the supports were determined with NH₃-TPD

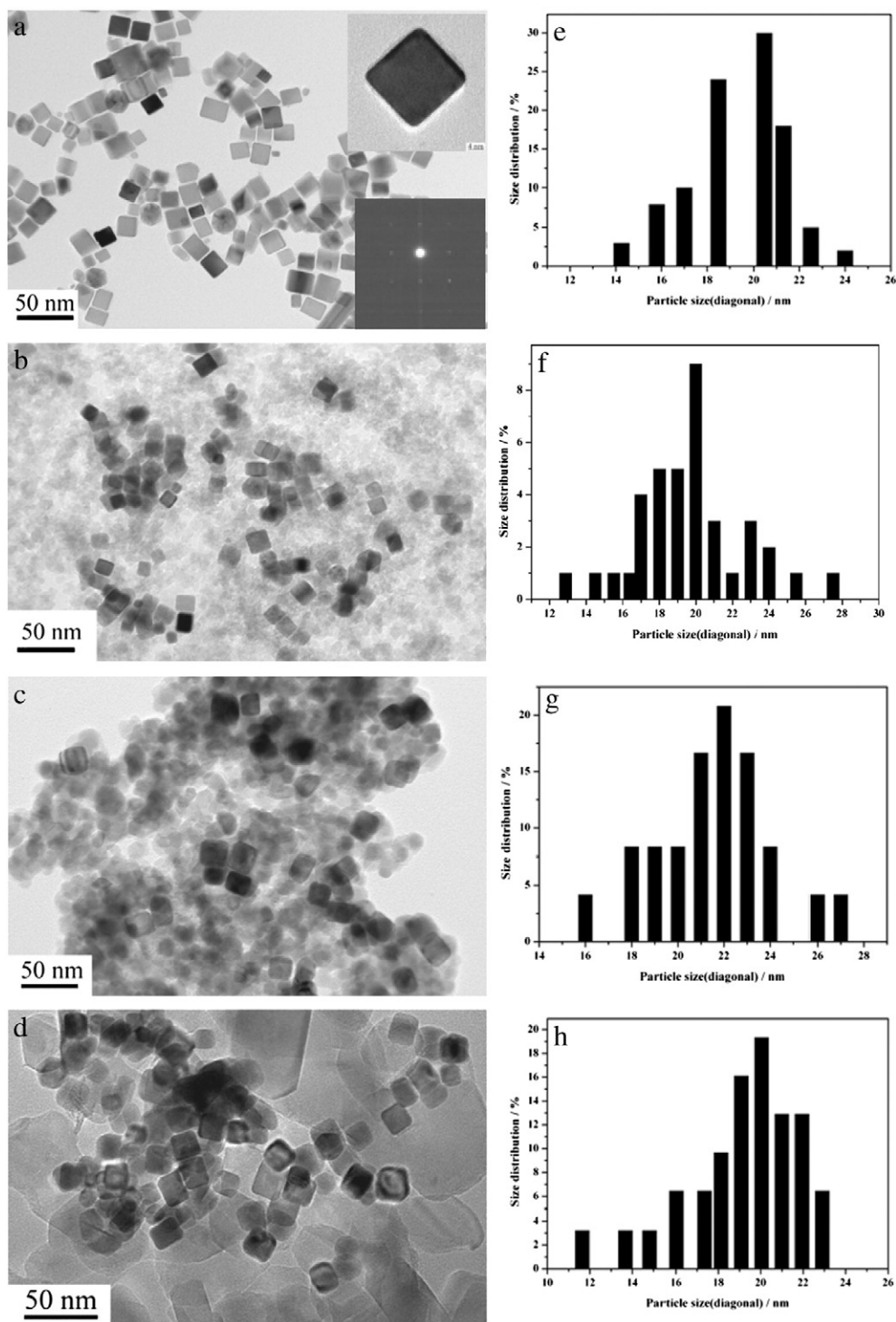


Fig. 1. TEM images of unsupported Pd nanocubes (a), the Pd/SiO₂ (b), Pd/TiO₂ (c) and Pd/MgO (d) catalysts, and the corresponding particle size distributions of Pd nanocubes (e–h), the inset in (a) is the SAED pattern and the HRTEM image of a single Pd nanocube.

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