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Article

The role of graphene coating on cordierite-supported Pd monolithic catalysts for low-temperature combustion of toluene

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ABSTRACT

In the present work, a Pd/graphene/cordierite (Pd/Gr/Cor) composite was prepared as a monolithic catalyst for low-temperature combustion of toluene. We mainly focused on understanding the role of graphene coating through investigation of catalytic performance and adsorption behavior of the composite. Compared with the traditional Pd/Cor catalyst without graphene coating, Pd/Gr/Cor catalyst delivered much higher activity and stability for toluene catalytic combustion in both dry and moist conditions. Transmission electron microscopy (TEM) and hydrophobic characterizations indicated that graphene coating can considerably improve the dispersity of Pd nanoparticles and enhance the hydrophobicity of the cordierite support. The adsorption behavior of the above two catalysts, including adsorption isothermal, adsorption kinetics, and adsorption thermodynamics were carefully investigated. The simulation results indicated that a large amount of toluene was adsorbed on graphene surface through relatively weak interaction, whereas only a relatively small amount of toluene was adsorbed on Pd surface with strong affinity. The adsorption thermal calculation indicated that the adsorption of toluene on graphene was a process with reduced entropy, indicating highly-ordered assembly of toluene molecular on graphene. It is the significant concentration and affinity gap between graphene and Pd that ensures a simultaneously and rapid transfer of toluene during the reaction process.

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1. Introduction

Catalytic combustion is one of the most important technologies for eliminating volatile organic compounds (VOCs) pollutants [1,2]. The development of high-performance monolithic catalyst has been viewed as a promising research direction to achieve effective removal of VOCs by catalytic combustion, owing to the catalyst's low pressure drop, mass transfer enhancement, ease to scale-up and separation regeneration [3,4]. However, the surface area of most traditional monolithic catalyst supports, e.g., cordierite is relatively low, thus making them always required modification with activated alumina or other high-specific-surface coatings [5,6]. As for active catalyst components, noble metals such as Pd and Pt have been known as highly effective catalysts [7]. The dispersity and particle size of the noble metals are two key factors. The particles with high dispersity and uniform size distribution could not only enhance the catalytic combustion activity but also reduce the used amount of noble metals to minimize the cost. Besides, since the catalyst particles are closely interrelated to the coating of catalyst supports, the catalytic activity is normally affected also by the surface properties of coating layers as well as interactions

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between the coating and noble metals [8,9]. The other challenge is the water vapor resulting from combustion reaction and the VOC feed, which can be easily adsorbed on the hydrophilic oxide surface and thus significantly reduce the catalytic activity of the noble metal catalysts [10,11]. Therefore, choosing an appropriate coating layer is vital for developing highly active noble-metal monolithic catalysts for VOC elimination.

To improve the moisture resistance of the catalyst, hydrophobic active carbon, carbon nano-fiber and other carbon materials have been explored as catalyst support for low-temperature catalytic combustion [12,13]. Zhang and coworkers [14] compared toluene catalytic combustion activity of the Pt catalyst supported on activated carbon and alumina. When the reaction was carried out in the wet gas stream at 130 ˚C, the activity of alumina-supported catalyst fell by 67%, while the activated carbon dropped by only 14% compared with dry conditions. Graphene is an extensively explored carbon material as a catalyst support because of its large specific surface area and high chemical stability [15,16]. Furthermore, our previous studies revealed that graphene nanosheets can be firmly immobilized on the surface of cordierite by hydrogen bonding or electrostatic adsorption. In addition, Pd nanoparticles loaded on the graphene exhibited high dispersity and a uniform size of less than 5 nm. The prepared catalyst exhibited excellent catalytic performance and stability in hydrogenation reactions [17, 18]. However, the role or the mechanism of graphene coating layers in the monolithic catalysts was unknown in our previous work [17,18]. In the present work, we mainly focused on understanding the role of graphene coating through investigation of catalytic performance and adsorption behavior of Pd/Cor and Pd/Gr/Cor catalysts. The catalytic performance for the two catalysts were studied in both dry and moist conditions. Additionally, as graphene with a perfect sp^2 hybridization of carbon atoms may absorb benzene series with benzene ring structure due to the π-π bonding effect [19], it is very beneficial to remove low concentrations of VOCs by amplifying the adsorption-catalytic synergistic effect. The graphene coating layer is demonstrated to play important roles in enhancing the dispersion of noble-metal particles, thus improving the hydrophobicity of catalyst support and endowing the catalyst with excellent adsorption for benzene series. As a result, the Pd/Gr/Cor catalyst exhibited high performance for catalytic combustion of toluene.

2. Experimental

2.1. Raw material

Cordierite, from Jiangxi Xingfeng Chemical Packing Company, and $PdCl_2$ with a Pd content of 60%, from Shaanxi Kaida Chemical Ltd, were purchased. The H_2PdCl_4 aqueous solution (0.01 mol/L) was used in the present work and prepared by completely dissolving 445 mg $PdCl_2$ in 250 mL HCl (6 mol/L) aqueous solution.

2.2. Catalyst preparation

Graphene oxide was prepared using the modified Hummers method, and Pd/Gr/Cor was prepared by ultrasonic impregnation techniques, as demonstrated in our previous work [18]. By contrast, the cordierite was immersed in the H_2PdCl_4 aqueous solution by ultrasound for 20 min, dried at 60 ˚C for 6 h, calcined in air at 350 ˚C for 2 h and then followed by reduction in a potassium borohydride (KBH_4) solution, the resulting catalyst is denoted as Pd/cordierite (Pd/Cor). The loading of Pd in Pd/Gr/Cor was 0.08% by ICP measurement and the loading of Pd in the Pd/Cor catalyst was controlled at a similar level.

2.3. Catalyst characterization

The morphology and size of palladium particles on monolithic catalysts were recorded by transmission electron microscopy (TEM) on a JEOL JEM-2100 (Japan) operated at 200 kV. X-ray diffraction (XRD) measurement of monolithic catalyst was carried out on a 3014-Z2 diffractometer (Japan) with a Cu K_α radiation at a scanning rate of 5°/min. The Brunauer-Emmett-Teller (BET) surface area of monolithic catalysts was calculated by the BET model at pressure ranging from 0.005 to 0.02 MPa on an Autosorb-iQ (Quantachrome) analyzer at –196 ˚C. The contact angle measurements were recorded on contact angle tester JC2000D1.

2.4. Toluene catalytic combustion reaction experiments

Catalytic performance of the monolithic catalysts was tested through catalytic combustion of toluene. The catalytic reaction was carried out in a tubular reactor. Toluene was carried by nitrogen spurge, mixed with air in a mixer, and then transferred into the reactor for reaction. The toluene concentration and airspeed can be changed by adjusting the flow rate of nitrogen and air. The reaction was monitored by gas chromatographic analysis (SHIMADZU 2010 plus, column RTX-5 with an inner diameter of 0.25 mm, a film depth of 0.25 μm and a length of 30 m) of the toluene outlet concentration withdrawn from the reactor.

Stability tests for the catalyst in moisture were as follows: water vapor was carried by bubbling air, mixed with toluene carried by nitrogen gas, and then carried out of the reaction. The conditions were as follows: toluene concentration = 15 g/m^3, relative humidity = 81%, space velocity = 4000 h^{-1}, the reaction temperature = 220 ˚C.

2.5. Toluene adsorption experiments

Toluene adsorption experiments were carried out through a similar process to that of the toluene catalytic combustion reaction. At the same temperature, the breakthrough curve would be drawn by detecting the outlet concentration of toluene. The simulations were further conducted based on the tested data.

3. Results and discussion

3.1. Characterization of catalysts

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