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Promotion effect of H_2 pretreatment on CeO_2 catalyst for NH_3-SCR reaction

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Abstract: In this study, the promotion effect of H_2pretreatment on the SCR performance of CeO_2 catalyst was investigated based on the characterization results of XRD, H_2-TPR, Raman and *in situ* DRIFT techniques. Lower crystallinity, higher reducibility and surface acidity can be found on CeO_2-H catalyst. The results of DRIFT study reveal that the pretreatment of CeO_2 catalyst with H_2 can facilitate the adsorption of NH_3 and NO_x species, while the adsorbed NO_x is basically inactive in the NH_3-SCR reaction. Moreover, the reaction mechanism of the NH_3-SCR reaction over CeO_2 catalyst is not changed by H_2 pretreatment, which is mainly under the control of Eley-Rideal (E-R) mechanism. The enhanced SCR performance of CeO_2-H catalyst is mainly due to the promoted NH_3 adsorption and the subsequent facilitation of SCR reaction through E-R pathway.

Keywords: CeO_2 catalyst; SCR; H_2 pretreatment; *in situ* DRIFT; mechanism; Rare earths

1. Introduction

Due to its high reliability and NO_x removal efficiency, selective catalytic reduction (SCR) of NO_x with NH_3in the presence of excess oxygen has been applied widely for controlling NO_x emission from stationary and mobile sources[1,2]. For this purpose, V-based catalyst (including V_2O_5-WO_3/TiO_2 and V_2O_5-MoO_3/TiO_2) is the most commercially available catalyst used in SCR reactor[3]. However, the utilization of V-based SCR catalyst has brought about some inevitable problems including narrow operation temperature, low selectivity at high temperature and the toxicity of VO_x species to human health and environment[4–6]. Therefore, developing alternative SCR catalyst without V species is of great importance for future NO_x emission control.

As a well-known alternative SCR catalyst, ceria has attracted considerable attention due to its strong oxygen transportcapacity of Ce^{4+}/Ce^{3+} pair and the high oxygen storage ability[7–10]. Recently, ceria-based catalysts supported on TiO_2, Al_2O_3 and TiO_2-SiO_2 have been successfully used in NH_3-SCR reaction[11–13]. To further enhance its SCR performance, the modification of Ce-based SCR catalyst by some transition metals such as W, Fe, Cu, Sb, Nb and Mo has been validated to be an effective approach by several groups[14–19]. Moreover, the precursortype and surface acid-base property had a significant impact on the SCR performance of Ce-based catalysts[20,21]. Besides that, the pretreatment of Ce-based SCR catalyst by some acidic or reductive gases including SO_2, HF, CO and H_2 has been proven to be another pathway for improving its SCR activity[22–25]. Yu et al.[23] found that the pretreatment of Ce/TiO_2 catalyst by H_2could enhance its SCR activity. To further understand the promotion mechanism, the effect of H_2 pretreatment on the NH_3-SCR reaction over CeO_2 catalyst was investigated based on different characterization techniques. And the effect of H_2 pretreatment on the NH_3-SCR reaction mechanism over CeO_2 catalyst is also be discussed.

2. Experimental

2.1 Catalyst preparation

In this study, pure CeO_2 catalyst sample was prepared by the thermal decomposition of cerium nitrate in a muffle furnace under air atmosphere at 550 ºC for 4 h. The H_2-pretreated catalyst sample was obtained by treating the