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Enhanced catalytic performance of rare earth-doped Cu/H-Sep for the selective catalytic reduction of NO with C_3H_6

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ABSTRACT

The undoped and rare earth oxide (REO_x, RE = La, Ce, Pr, Sm)-doped acid-treated sepiolite (H-Sep)-supported copper oxide catalysts (Cu/H-Sep and RE-Cu/H-Sep; Cu and RE loading = 15 and 4 wt%, respectively) were prepared using the incipient wetness impregnation method. The catalysts were characterized by means of the XRD, BET, XPS, H_2-TPR and NH_3-TPD techniques, and their catalytic activities were evaluated for the selective catalytic reduction (SCR) of NO with propylene. It is shown that the doping of REO_x led to an enhancement in catalytic performance of Cu/H-Sep in the SCR of NO with C_3H_6. Among the RE-Cu/H-Sep catalysts, Ce-Cu/H-Sep performed the best, with 62% NO conversion being achieved at 350 °C in the ($NO + C_3H_6 + O_2$) reaction. We believe that the high catalytic performance of Ce-Cu/H-Sep for the SCR of NO with C_3H_6 was associated with the good low-temperature reducibility, high copper oxide dispersion, and strong metal–support interaction.

1. Introduction

Nitrogen oxides (NO_x) from combustion of fossil fuels are one of the major pollutants to the atmosphere, which can cause photochemical smog, acid rain, and respiratory problems. It is known that ca. 50% NO_x are from the emission of automotives (e.g., gasoline cars and diesel engine trucks) and ca. 40% NO_x are from the stationary sources (e.g., power plants fueled by fossil fuel combustion). The selective catalytic reduction of NO_x using hydrocarbons (HC-SCR) has attracted much attention, because it is capable of eliminating NO_x emitted from the natural-gas fueled engines (e.g., lean-burn gas engines in cogeneration systems and lean-burn gasoline and diesel engines), where the noble-metal three-way catalysts are not effective in the presence of excess oxygen [1].

Iwamoto and co-workers [2] first reported that the Cu-exchanged ZSM-5 material was an effective catalyst for the SCR of NO. In the past two decades, much attention has been paid on the development of catalysts exhibiting better performance. Although a large number of studies have been focused on the ion-exchanged zeolites, Cu is believed to be one of the most active components in the HC-SCR processes. Various kinds of metal oxide or zeolite-supported Cu catalysts have also been thoroughly investigated for the HC-SCR reaction. These Cu-based catalysts, however, are susceptible to sintering at high temperatures (500–700 °C), resulting in a loss in catalytic activity. Moreover, a catalytic activity loss can be also observed due to the dealumination of such catalysts in the presence of water vapor at high temperatures [3]. Therefore, it is highly desired to select an appropriate support that possesses good hydrothermal stability. Clays represent a class of microporous materials, which have been found wide applications in catalysis and adsorption and separation. In the case of the HC-SCR process, clays are superior to ZSM-5 in hydrothermal stability [4]. Sepiolite is a natural clay mineral and its ideal formula is $[Si_{12}Mg_8O_{30}(OH)_4(H_2O)_4]\cdot 8\ H_2O$ [5]. Similar to other types of silicate minerals, sepiolite contains a continuous two-dimensional tetrahedral sheet of T_2O_5 (T = Si, Al, Be, etc.) but lacks continuous octahedral sheets [6]. This unique fibrous structure with interior channels (0.36 × 1.06 nm) renders sepiolite to be useful in adsorption and catalysis [7].

In order to improve the activity of copper-based catalysts for the HC-SCR, some additives, such as Fe, Co, Ni, V, Mn, W, Mo, and Cr, were doped for the promotion of the copper species dispersion. The addition of certain promoters could enhance the catalytic performance of supported Cu catalysts for the SCR of NO_x with hydrocarbons [8]. Cerium oxide is a commonly used promoter in automotive three-way catalysts [9]. Other rare earth oxides have also positive effects on the catalytic activity improvement. For example, rare earth oxide (La_2O_3, Sr/La_2O_3, Nd_2O_3, Sm_2O_3, Sr/Sm_2O_3, Tm_2O_3 and Lu_2O_3)-based catalysts were active for the

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