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RESEARCH ARTICLE

Characterization and performance of V_2O_5/CeO_2 for NH_3-SCR of NO at low temperatures

Caiting LI (✉), Qun LI, Pei LU, Huafei CUI, Guangming ZENG

Key Laboratory of Environmental Biology and Pollution Control (Ministry of Education), College of Environmental Science and Engineering, Hunan University, Changsha 410082, China

Abstract A series of CeO_2 supported V_2O_5 catalysts with various loadings were prepared with different calcination temperatures by the incipient impregnation. The catalysts were evaluated for low temperature selective catalytic reduction (SCR) of NO with ammonia (NH_3). The effects of O_2 and SO_2 on catalytic activity were also studied. The catalysts were characterized by specific surface areas (S_{BET}) and X–ray diffraction (XRD) methods. The experimental results showed that NO conversion changed significantly with the different V_2O_5 loading and calcination temperature. With the V_2O_5 loading increasing from 0 to 10 wt%, NO conversion increased significantly, but decreased at higher loading. The optimum calcination temperature was 400°C. The best catalyst yielded above 80% NO conversion in the reaction temperature range of 160°C–300°C. The formation of $CeVO_4$ on the surface of catalysts caused the decrease of redox ability.

Keywords V_2O_5/CeO_2 catalysts, NH_3-SCR (selective catalytic reduction), the incipient impregnation, low temperatures

1 Introduction

The emission of NO_x contributes to acid rain, photochemical smog, ozone depletion, and greenhouse effects [1]. The pollution sources of NO_x are divided into stationary sources (i.e. power plants) and mobile sources (i.e. transportation). Power plant boilers produce about 40% of the NO_x emissions from stationary sources. In recent decades, the catalytic technologies of NO_x have become attractive. The selective catalytic reduction(SCR) of NO_x has been well reported for some time for the treatment of flue gas, by using reducing agents such as CO, H_2, NH_3, and aliphatic carboxylic acid [2,3]. The main reducing agent for reducing NO emissions from power plant sources is ammonia(NH_3). The reaction is as follow equation:

$$4\,NO + 4\,NH_3 + O_2 \rightarrow 4\,N_2 + 6\,H_2O \qquad (1)$$

The reported commercial catalysts for SCR of NO_x with NH_3 usually include V_2O_5/TiO_2 [1,4], or a V_2O_5/TiO_2 catalyst promoted by WO_3 or MoO_3 [5–9], CuO/TiO_2, MnO_x/TiO_2 [10]and so on. The required operating temperature for the above industrial catalysts is typically 300°C–400°C. This makes it necessary to locate the SCR unit upstream of the desulphurizer and/or particulate control device in order to avoid reheating the flue gas and causing deposition of dust on the catalyst, where the life of the catalyst is shortened because of high concentrations of SO_2 and ash in the flue gas [1]. Therefore, it is important to investigate a catalyst with high activity at low temperatures, which would be placed downstream of the desulphurizer and electrostatic precipitator, operating at a temperature of 80°C–300°C. Some transition-metal-containing catalysts have been investigated for the low temperature SCR reaction, such as MnO_x/Al_2O_3 [11], V_2O_5/AC [12], MnO_x/NaY [13], iron-silica aerogels [14], MnO_x/CeO_2 [15–18], MnO_x/TiO_2(prepared from two different precursors) [19], amorphous MnO_x [20] and other oxides [21].

Rare earth oxides have been widely investigated for catalysis. Ceria (CeO_2) is one of the most significant. And it is a crucial component in the so-called “three-way catalysts” for the elimination of toxic exhaust gases in automobiles [22]. The most important property of CeO_2 is as an oxygen reservoir, which stores and releases oxygen via the redox shift between Ce^{4+} and Ce^{3+} under oxidizing and reducing conditions, respectively. Ceria also improves the dispersion of the metal and the activity [22,23]. The vanadium-based catalysts are highly active and resistant to

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E-mail: ctli@hnu.cn

SO_2 [16]. Xiaodong Gu et al. [24] have found that various amounts of V_2O_5 on Ceria and different calcination temperatures brought about the changes of surface structure of dispersed vanadium species, which have an effect on the activity. Recently, Chen et al. [25] have found that the Ce doped V_2O_5-WO_3/TiO_2 catalyst could enhance the NO_x adsorption and then accelerate the SCR reaction due to the synergetic interaction among the Ce and V, W species. They [26] also have studied the W-doped CeO_2/TiO_2 catalysts by co-impregnation method, and found the co-impregnation method could make CeO_2 and WO_3 crystallites disperse better over the catalyst surface, and much stronger interaction happened between Ce and W with the increased appearance of Ce^{3+}.

In this work, we studied the CeO_2 with different loadings of V_2O_5 for SCR of NO with NH_3 at low temperatures (120°C–300°C). The activities of catalysts that were prepared at different calcination temperatures were compared. The effects of O_2 and SO_2 also were studied. And the CeO_2 and the V_2O_5/CeO_2 catalysts were characterized by means of specific surface areas (S_{BET}), X–ray diffraction (XRD).

2 Experiment

2.1 Catalyst characterization

The specific surface area (S_{BET}) of each prepared sample was determined by nitrogen physisorption at −196°C (Micromeritics ASAP 2010). The specific surface area was determined from the linear portion of the BET plot. Prior to the surface area measurements, the samples were degassed in vacuo at 120°C for 1 h.

The X–ray diffraction (XRD) measurements were carried out with a Rigaku Rotaflex D/Max–C system with Cu Kα (λ = 0.1543 nm) radiation. The samples were loaded on a sample holder with a depth of 1 mm.

2.2 Catalyst preparation

CeO_2 was prepared by thermal decomposition of $Ce(NO_3)_2 \cdot 6H_2O$ (4 moL·L^{-1}, Sinopharm Chemical Reagent Co., Ltd,) in air at 500°C for 5 h. The V_2O_5/CeO_2 catalysts with various loadings (1–20 wt% V_2O_5) were prepared by using the incipient impregnation method. Ammonium metavanadate (NH_4VO_3) and oxalate were mixed in the desired proportions (about 1:2, excess oxalate). A certain amount of CeO_2 powder was added into the aqueous solution and was stirred. After being kept at room temperature overnight, it was dried at 100°C for 12 h and then calcined at 400°C, 500°C and 600°C for 4 h. The catalysts were marked *x*% V_2O_5/CeO_2(y), *x*% is V_2O_5 loading and *y* is the calcination temperature. V_2O_5 was obtained by calcining NH_4VO_3 at 400°C for 5 h.

2.3 Catalytic activity tests

The SCR activity measurement of the V_2O_5/CeO_2 catalysts was carried out in a fixed-bed stainless steel continuous flow reactor at atmospheric pressure. The range of experimental temperature was 120°C–300°C. The typical reactant gas composition was as follows: 1×10^{-9} NO, 1.08×10^{-9} NH_3, 5 vol% O_2, and balance N_2. A 500 mg sample was used in each run. The total flow rate was 100 mL/min. To prevent formation and deposition of ammonium nitrate, the tubing of the reactor system was heat–traced. NO concentrations in the inlet and outlet gases were measured by Hand–held Combustion Analyzer (KM900, Kane International Limited, UK.). The accuracy is ±5% of the reading, and its resolution is 10^{-6}.

3 Results and discussion

3.1 Characterizations of catalysts

3.1.1 BET

The specific surface areas (S_{BET}) of the V_2O_5/CeO_2 catalysts with various V_2O_5 loadings at different calcination temperaturess are summarized in Table 1. It is clear that the S_{BET} of pure CeO_2 is the largest (83 m^2·g^{-1}). The S_{BET} of V_2O_5/CeO_2 catalysts decrease compared to pure CeO_2. It is clear that the S_{BET} decreases as the V_2O_5 loading increases from 0 to 20 wt%. The S_{BET} of V_2O_5 is only 15 m^2·g^{-1}. The reasons for this result may be: 1) the blockage of some pores by the supported species, 2) the destruction of the texture of ceria during impregnation, 3) the sintering of the catalysts, 4) the formation of new low surface area compounds due to solid reactions occurring between the support and supported species [24]. However, temperature higher than 400°C is needed for the solid reaction between CeO_2 and V_2O_5.

The S_{BET} decreased with the increase of V_2O_5 loading, which is due to either the blockage of some pores by the

Table 1 Specific surface area of the catalysts

catalysts	S_{BET} /(m^2·g^{-1})
CeO_2	83
V_2O_5	15
1%V_2O_5/CeO_2(400°C)	80
5%V_2O_5/CeO_2(400°C)	72
10%V_2O_5/CeO_2(400°C)	56
15%V_2O_5/CeO_2(400°C)	53
20%V_2O_5/CeO_2(400°C)	48
5%V_2O_5/CeO_2(500°C)	60
5%V_2O_5/CeO_2(600°C)	43

supported species or the destruction of the texture of ceria during impregnation. With the increase of calcination temperature, the increased order of the surface areas of three catalysts with different calcination temperatures is: 5% V_2O_5/CeO_2 (400°C) > $5\%V_2O_5/CeO_2$ (500°C) > $5\%V_2O_5/CeO_2$ (600°C). This is evidently related to the formation of new compounds and the sintering of the catalysts. Daniell et al. [27] found that VO_x and CeO_2 began to react to form $CeVO_4$ on the surface over 400°C. The sintering of the catalysts would be analyzed on XRD as demonstrated below.

3.1.2 XRD

XRD patterns of V_2O_5/CeO_2 catalysts with various V_2O_5 loadings and pure CeO_2 (commercial) are shown in Fig. 1. The cubic fluorite structure of CeO_2 (JCPDS #43-1002) was clearly detected in all catalysts. No XRD pattern for crystalline V_2O_5 was observed for the catalysts with the loading of V_2O_5 lower than 15%. This demonstrates that the V_2O_5 loading is too low to be detected or that vanadium species are in a highly dispersed state in the catalysts. With the V_2O_5 loading increased, a weak peak of V_2O_5 crystal was observed at 20% V_2O_5 loading. This indicates the coverage of V_2O_5 exceeds the monolayer on CeO_2 and the particle size of V_2O_5 is much larger than other catalysts calcined at 400°C.

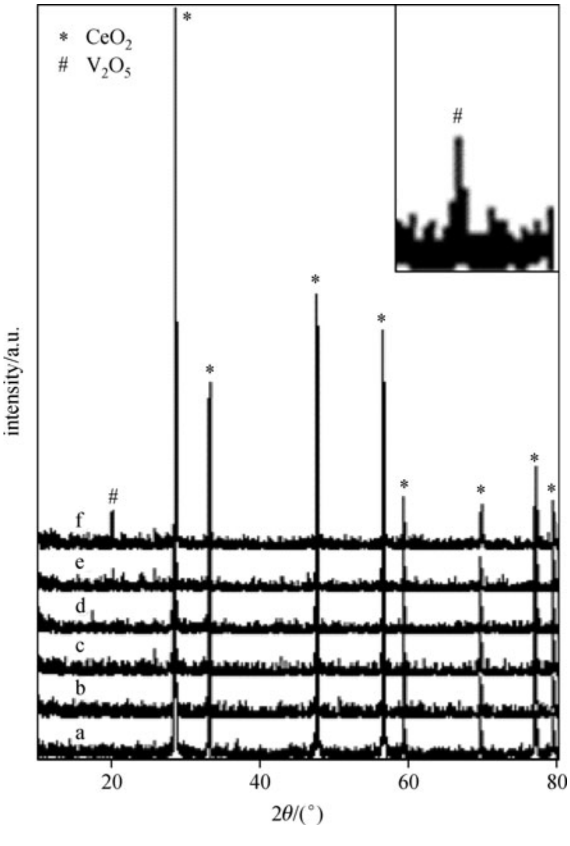

Fig. 1 XRD patterns of the V_2O_5/CeO_2 (400°C) catalyst with various V_2O_5 contents. (a) Pure CeO_2; (b) 1% V_2O_5/CeO_2; (c) 5% V_2O_5/CeO_2; (d) 10% V_2O_5/CeO_2; (e) 15% V_2O_5/CeO_2; (f) 20% V_2O_5/CeO_2

Figure 2 shows the XRD patterns of the $5\%V_2O_5/CeO_2$ (400°C), $5\%V_2O_5/CeO_2$ (500°C), and $5\%V_2O_5/CeO_2$ (600°C). With the increase of calcinations temperature, the characteristic peaks for $CeVO_4$ become intensive. This illustrates that more $CeVO_4$ forms and the particle size increases [24]. We can also observe that the peaks of CeO_2 become sharper with the increase of calcinations temperature, indicating the sintering of CeO_2, which is consistent with the results of the measurements of the S_{BET}. The oxygen anions in $CeVO_4$ are less active than in V_2O_5 and CeO_2, because they are more strongly bonded into the lattice of $CeVO_4$. With the formation of $CeVO_4$, the Ce^{4+} in CeO_2 is reduced to Ce^{3+} in $CeVO_4$, more stably, which in turn weakens the redox ability of the catalysts.

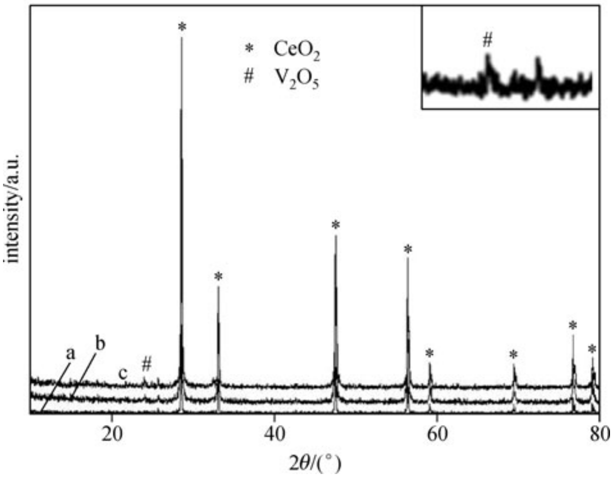

Fig. 2 XRD patterns of the $5\%V_2O_5/CeO_2$ catalyst at different calcination temperatures: (a) $5\%V_2O_5/CeO_2$ (400°C); (b) $5\%V_2O_5/CeO_2$ (500°C); (c) $5\%V_2O_5/CeO_2$ (600°C)

3.2 Activity evaluation

3.2.1 Effect of V_2O_5 loading on NO conversion

Figure 3 illustrates the results of the NO conversion of the catalysts with various V_2O_5 loadings for SCR of NO with NH_3 at low temperatures (120°C–300°C). Pure CeO_2 showed slight activity (not shown in Fig. 3). With an increase of the V_2O_5 loading, the activity of the catalysts was significantly enhanced, compared with pure CeO_2. However, the catalytic activity didn't always increase with the enhancement of the V_2O_5 loading. When the V_2O_5 loading exceeded 15%, the catalytic activity clearly decreased. For 5% and 10% V_2O_5/CeO_2, the trend of NO conversion was consistent, and higher than 90% in a temperature range (200°C–300°C). However, the NO maximum conversion of 15% V_2O_5/CeO_2 was 80% at

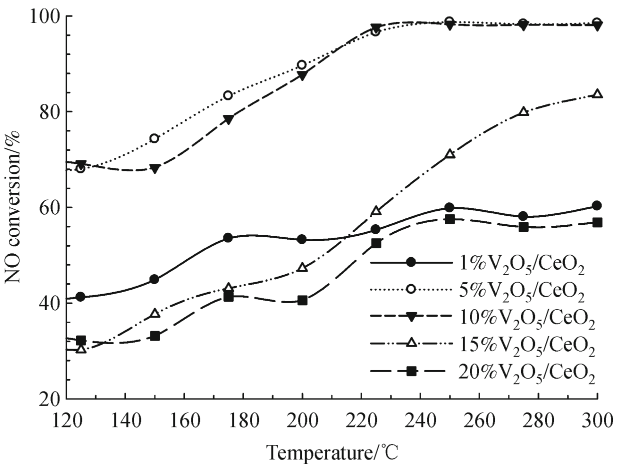

Fig. 3 NO conversion of the V_2O_5/CeO_2 catalyst with various V_2O_5 loadings calcined at 400°C.Reaction conditions: 500 mg catalyst, total flow 100 mL·min^{-1}, NO 1×10^{-9}, NH_3 1.08×10^{-9}, O_2 5 vol%, N_2 balance

300°C and the NO conversion of 20% V_2O_5/CeO_2 was only about 50% at 300°C. For all catalysts, the NO conversion increases gradually with the raising of the reaction temperature, and tended to be stable.

3.2.2 Effect of calcination temperature on NO conversion

The catalytic activities of the 5% V_2O_5/CeO_2 catalysts calcined at 500°C and 600°C also were studied with the same reaction conditions as the catalysts calcined at 400°C. Figure 4 shows the NO conversion of catalysts for 5% V_2O_5 loadings at different calcination temperatures (500°C, 600°C). With the enhancement of calcinations' temperature from 400°C to 600°C, the NO conversion of catalysts decreased. The 5%V_2O_5/CeO_2 (400°C) catalyst showed the widest reaction window and the highest NO conversion (nearly 98% at 250°C). The NO conversion of other catalysts 5%V_2O_5/CeO_2(500°C) and 5%V_2O_5/CeO_2(600°C) was only about 50%–70%. That indicated the calcinations' temperature of catalysts was one important factor for the catalytic activity. On the one hand, the calcinations' temperature increasing resulted in the formation of $CeVO_4$ which reduced the redox ability of the catalysts. In fact, we can observe the faint XRD pattern of the new compound $CeVO_4$, which is related to the V_2O_5 loading. On the other hand, the increase of calcination temperature led to the sintering of the catalyst to reduce activity.

3.2.3 Effect of O_2 on NO conversion

Xu et al. [28] reported that O_2 plays an important role in the reaction between NO with NH_3. Figure 5 showed the NO conversion of the 5%V_2O_5/CeO_2 catalysts in the presence of O_2 and in the absence of O_2. As shown in Fig. 5, the NO conversion of the 5%V_2O_5/CeO_2 catalyst increased gradually and reached 98% at 250°C in the presence of O_2.The NO conversion maintained a steady-state at the initial stage of the reaction in the absence of O_2, but then decreased at 200°C. Accordingly, we inferred that lattice oxygen participated in the reaction in the absence of O_2. The steady-state was mainly due to the CeO_2. With the whole reaction completed, the lattice oxygen of CeO_2 was consumed and the NO conversion decreased significantly.

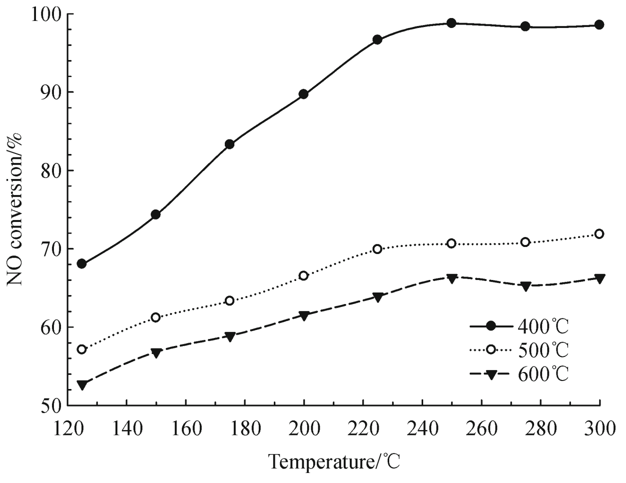

Fig. 4 NO conversion of the 5%V_2O_5/CeO_2 catalyst calcined at 400°C, 500°C, 600°C. Reaction conditions: 500 mg catalyst, NO 1×10^{-9}, NH_3 1.08×10^{-9}, O_2 5 vol%, N_2 balance, total flow 100 mL·min^{-1}

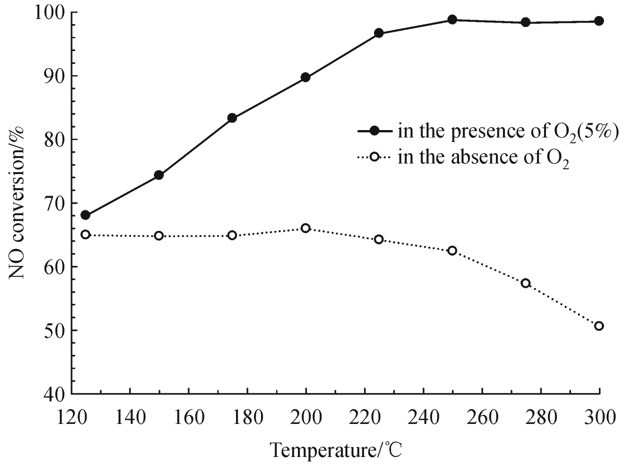

Fig. 5 NO conversion of the 5%V_2O_5/CeO_2 catalyst under different reaction conditions: in the presence of O_2 and in the absence of O_2. Reaction conditions: 500 mg catalyst, NO 1×10^{-9}, NH_3 1.08×10^{-9}, O_2 0–5 vol%, N_2 balance, total flow 100 mL·min^{-1}

3.2.4 Effect of SO_2 on NO conversion

We further studied the effect of SO_2 on the NO conversion of the 5%V_2O_5/CeO_2 catalyst. The results are shown in Fig. 6. After adding SO_2, the NO conversion increased in the 120°C–300°C range. This was due to the reaction between SO_2 and NH_3, which formed SO_4^{2-}. As is well known, V_2O_5 is a very active catalyst for SO_2 oxidation and much easier to transform into SO_4^{2-}. The SO_4^{2-} stimulated NH_3 to be adsorbed onto the surface of the 5%V_2O_5/CeO_2 catalyst to form NH_4^+. Then the reaction between NH_4^+ and NO occurred and promoted the NO conversion. Zhu et al. [29,30] also found that SO_2 had positive influences on the NO conversion when the V_2O_5 loading was 1%–5% and the reaction temperature was 250°C. On the other hand, the ammonium salts deposited on the face of the catalyst and acted as poisons to offset the promoting catalytic role of SO_2 with time process at higher temperature(> 200°C) [12].

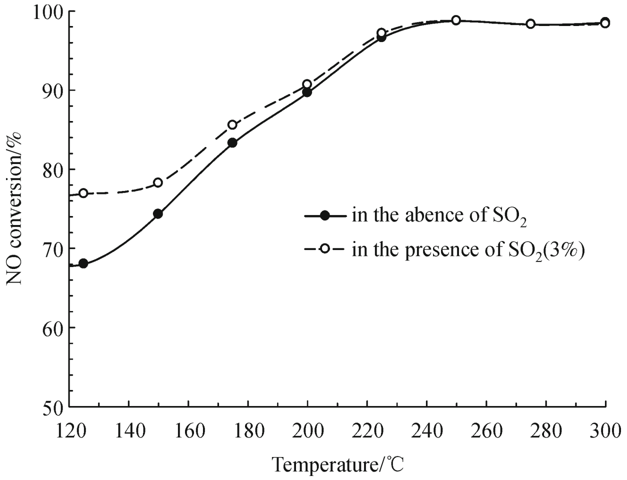

Fig. 6 NO conversion on 5%V_2O_5/CeO_2 catalyst under different reaction conditions: in the presence of SO_2 and in the absence of SO_2. Reaction conditions: 500 mg catalyst, NO 1×10^{-9}, NH_3 1.08×10^{-9}, SO_2 0–3vol%, N_2 balance, total flow 100 mL·min^{-1}

4 Conclusions

The V_2O_5/CeO_2 catalyst with various V_2O_5 loadings is highly active in the presence of oxygen and is resistant to SO_2. The V_2O_5 loading and the calcinations temperature of catalysts have significant effects on the activity and surface structure of the catalysts. O_2 has an important role in the reaction between NO and NH_3. By XRD and activity test, it is concluded that the formation of $CeVO_4$ has influenced catalytic efficiency by changing the structure of the catalysts, facilitating the reduction of Ce^{4+} to Ce^{3+}, which weakens the catalytic activity.

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