[Regular Paper]

Selective Catalytic Reduction of NO with Propene in the Presence of Excess Oxygen over Supported Cerium Oxides

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Selective catalytic reduction of NO with propene in the presence of excess oxygen was investigated by using several CeO₂ supported on oxides. Unsupported CeO₂ was found not effective for the NO reduction; however it was highly active for oxidation of NO to NO₂ and C₃H₆ to CO and CO₂. The activity of CeO₂ for the reduction of NO to N₂ was greatly enhanced by supporting it on Al₂O₃, ZrO₂, Ga₂O₃, etc. Among them Al₂O₃ was the best support, but Al₂O₃ itself was almost inactive under the conditions presently used (300–400°C). CeO₂/Al₂O₃ was superior in activity to Pt/Al₂O₃ and to Pt–CeO₂/Al₂O₃. It was presumed that CeO₂ had the oxidizing capability, which allowed oxidative activation of NO without excessive oxidation of propene, and the oxidized NO was successively reduced on Al₂O₃.

1. Introduction

Selective catalytic reduction of NO with hydrocarbons in the presence of excess oxygen has actively been investigated because the reaction is a practical technique promising for removal of nitrogen oxides from exhaust gases of diesel and lean-burn gasoline engines^{1)–6)}. Since Cu–ZSM-5 was found to catalyze this reaction effectively⁷⁾, numerous catalysts have been reported. Furthermore, zeolites exchanged with metal ions such as Co⁸⁾, Ga⁹⁾, In⁹⁾, and Pd¹⁰⁾ have attracted attention because these catalysts were observed to be effective for NO reduction with methane in the presence of excess O₂. Among the ion-exchanged zeolites, Ce–ZSM-5 was found to exhibit high activity for selective reduction of NO with propene^{11),12)}. We have recently reported that Ce–mordenite was more active for the reduction of NO with propene than Cu–mordenite; moreover, it was the most active among the mordenites exchanged with various rare earth ions¹³⁾. These results indicated that Ce could be used as an effective catalyst component for selective NO reduction. However, the low thermal stability of the zeolites was disadvantageous.

Ce oxides have been used practically as promoters of the three-way catalysts, consisting of noble metals (Pt–Pd–Rh) and oxide supports such as Al₂O₃, for catalytic control of NO emission in automobile exhaust gases¹⁴⁾. In the three-way catalysts, Ce oxide functions as an oxygen storage component. Kakuta *et al.* investigated in detail the oxygen storage capacity of alumina-supported Ce oxides¹⁵⁾; however, the catalytic

performance of Ce oxides themselves or that supported on other oxides were not investigated in detail. Nevertheless, it should be helpful for clarifying a new aspect of Ce-based catalysts to investigate the activity of Ce oxides for selective NO reduction.

Conventional metal oxides also showed catalytic activities for NO selective reduction with hydrocarbons. Hamada *et al.* found that Al₂O₃ catalyzed the selective reduction of NO with propane¹⁶⁾. In addition, various oxides^{4),17)}, supported oxides such as Cu/Al₂O₃¹⁸⁾, and Co/Al₂O₃¹⁸⁾, and mixed metal oxides such as ZrO₂–TiO₂¹⁹⁾ also exhibited catalytic activities for the NO reduction. In addition, the supported metal catalysts^{20),21)} were reported to be also effective for selective catalytic reduction of NO. The oxide catalysts are expected to be thermally stable and durable although their activities may be somewhat lower than those of the zeolite catalysts⁴⁾.

Such background prompted us in this study to develop and prepare various supported Ce oxide catalysts for the selective reduction of NO with propene. We also investigated the effects of Pt addition to CeO₂/Al₂O₃, and we observed that CeO₂/Al₂O₃ was more active than cerium oxides supported on other oxides.

2. Experimental

Ce oxides supported on various metal oxides were prepared by an impregnation method. Al₂O₃, MgO, SiO₂, ZrO₂, Nb₂O₅, La₂O₃, and Ga₂O₃ were used as supports. We used three Al₂O₃ samples, JRC–ALO-4 and JRC–ALO-1 (supplied as reference catalysts from the Catalysis Society of Japan), and Aerosil C (Aerosil Nippon Co.) in which JRC–ALO-4 was mainly exam-

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Table 1 Catalytic Activities of Various Supported Ce Oxides for Selective Reduction of NO with C₃H₆ at 400°C^{a)}

Catalysts ^{b)}	Surface area [m ² · g ⁻¹]	Conversion [%]				Selectivity ^{c)} [%]
		NO to N ₂	NO to NO ₂	NO to N ₂ O	C ₃ H ₆ to CO and CO ₂	
CeO ₂ /Al ₂ O ₃ ^{d)}	158	33	3	0	61	13
CeO ₂ /Al ₂ O ₃ ^{e)}	145	12	4	0	46	6
CeO ₂ /Al ₂ O ₃ ^{f)}	92	30	3	0	25	24
CeO ₂ /MgO	78	11	5	2	52	5
CeO ₂ /SiO ₂	300	4	2	0	30	3
CeO ₂ /ZrO ₂	58	16	4	0	86	4
CeO ₂ /Nb ₂ O ₅	131	4	0	0	41	3
		(8	1	0	14	12) ^{g)}
CeO ₂ /La ₂ O ₃	14	3	3	1	6	10
		(17	5	3	74	6) ^{h)}
CeO ₂ /Ga ₂ O ₃	35	17	1	0	86	4
Al ₂ O ₃ ⁱ⁾	170	2	0	0	5	12
CeO ₂	76	7	14	0	87	2

a) Reaction conditions: NO 1000 ppm, C₃H₆ 500 ppm, O₂ 2% (He balance), total flow rate 90 cm³ · min⁻¹, catalyst 0.3 g (W/F=0.2 g · s · cm⁻³). Data of the catalyst, which gave the maximum activity for NO reduction at temperatures other than 400°C, are indicated in the parentheses.

b) The amount of CeO₂ loaded was 10 wt% (as CeO₂).

c) Selectivity [%] = (number of N₂ formed) × 2 / {(number of CO formed) + (number of CO₂ formed) × 2 + (number of H₂O formed)} × 100.¹²⁾

d) Prepared from JRC-ALO-4.

e) Prepared from JRC-ALO-1.

f) Prepared from Aerosil C.

g) Data obtained at 300°C.

h) Data obtained at 500°C.

i) JRC-ALO-4.

ined. MgO, La₂O₃, and Ga₂O₃ were prepared from an aqueous solution of the corresponding metal nitrate and aqueous ammonia, and calcined at 500°C for 3 h in air. ZrO₂ was obtained by calcination of Zr(OH)₄ (Daiichi Kigenso Kagaku Kogyo Co., Ltd.) at 500°C for 3 h in air. SiO₂ (Aerosil 300, Aerosil Nippon Co.) and Nb₂O₅ (NIOBIA HY-340, CBMM Co.) were used as received. Ce oxides were loaded onto the oxides from a 0.18 mol · dm⁻³ aqueous Ce(NO₃)₃ (Wako Pure Chemical Industries, Ltd.) solution using the incipient-wetness technique, and the solids obtained were heated at 500°C for 3 h in air. The amount of Ce oxide loaded was 10 wt% as CeO₂. The catalyst thus obtained will be denoted as CeO₂/oxide, for example CeO₂/Al₂O₃. Pt-Supported Al₂O₃ was prepared using the incipient-wetness method from an aqueous solution of [Pt(NH₃)₄](OH)₂ and the product was then calcined at 500°C for 3 h in air (the catalyst will be termed as Pt/Al₂O₃). The amount of Pt was 0.5 wt%. Pt was also loaded onto CeO₂/Al₂O₃ by the same manner, and the Pt-content was 0.5 wt% (the catalyst will be denoted as Pt/CeO₂/Al₂O₃). CeO₂ was prepared by adding aqueous ammonia to an aqueous Ce(CH₃COO)₃ solution, and calcined at 500°C for 3 h in air. Surface areas of the catalysts were determined by the BET analysis of N₂ adsorption at 77 K using an automatic gas adsorption apparatus (BEL Japan BELSORP28SA), and they are listed in Table 1. Powder X-ray diffraction (XRD) pattern was measured by a diffractometer

(Rigaku Geigerflex 2027) with monochromated CuKα radiation.

Selective catalytic reduction of NO with propene was conducted in a fixed-bed flow reactor (quartz tube, 8 mm of inside diameter). The catalyst was pretreated before use at 500°C for 2 h in a flow of He (90 cm³ · min⁻¹). A mixed gas of NO: 1000 ppm, O₂: 2%, and C₃H₆: 500 ppm (He balance) was fed to 0.3 g of catalyst at a flow-rate of 90 cm³ · min⁻¹ (W/F=0.2 g · s · cm⁻³). The reaction was at first carried out at 500°C, then it was lowered to 200°C with intervals of 50–100°C when the reaction became stationary at each temperature. Effluent gases were analyzed by gas chromatography (Nippon Aera M-200) using a Molecular Sieve 5A column (for N₂, O₂, and CO) and a Porapak Q column (for CO₂ and N₂O). Concentrations of NO and NO₂ at the outlet were determined by a chemiluminescence NO_x analyzer (Yanaco ECL-77A).

3. Results and Discussion

Figure 1 shows catalytic activities of CeO₂/Al₂O₃, Al₂O₃, and CeO₂ for the selective reduction of NO with C₃H₆ at various temperatures. Here JRC-ALO-4 was used as the Al₂O₃ source. CeO₂/Al₂O₃ catalyzed the reduction of NO to N₂ at the temperature region of 300–400°C much more efficiently than Al₂O₃ and CeO₂. The reductive activity decreased by elevating the temperature to 500°C. At this temperature, C₃H₆ was

The source of Al_2O_3 was JRC-ALO-4.

(A) Conversion of NO to N_2 , (B) Conversion of C_3H_6 to CO and CO_2 , (C) Conversion of NO to NO_2 , (D) Conversion of NO to N_2O .

Reaction conditions: NO 1000 ppm, C_3H_6 500 ppm, O_2 2% (He balance), total flow rate $90 \text{ cm}^3 \cdot \text{min}^{-1}$, catalyst weight 0.3 g ($W/F=0.2 \text{ g} \cdot \text{s} \cdot \text{cm}^{-3}$).

Fig. 1 Temperature Dependence of the Selective Reduction of NO with C_3H_6 in the Presence of Excess O_2 over CeO_2 , Al_2O_3 , and $\text{CeO}_2/\text{Al}_2\text{O}_3$

completely oxidized (**Fig. 1B**), which was responsible for the decrease in the activity. Under the present conditions, Al_2O_3 alone was almost inactive for the NO reduction and other related reactions at temperatures lower than 400°C . This observation was in harmony with previous results that Al_2O_3 showed the activity at temperatures higher than 500°C ^{16),22)}. On the other hand, the oxidation reactions, *i.e.*, C_3H_6 to CO and CO_2 , and NO to NO_2 , proceeded rapidly over CeO_2 , whereas it was less active for the reduction of NO to N_2 . These results indicated that the catalytic activity of CeO_2 for selective reduction of NO was greatly enhanced by the catalyst supported on Al_2O_3 . Other $\text{CeO}_2/\text{Al}_2\text{O}_3$ catalysts prepared from different Al_2O_3 samples showed similar behavior, although their activities were somewhat different (*vide post*).

Table 1 summarizes the activities of various CeO_2 /oxide catalysts for the selective NO reduction with C_3H_6 at 400°C . Temperature-dependencies of CeO_2/MgO , $\text{CeO}_2/\text{SiO}_2$, $\text{CeO}_2/\text{ZrO}_2$, and $\text{CeO}_2/\text{Ga}_2\text{O}_3$ for N_2 formation were similar to the dependency observed for $\text{CeO}_2/\text{Al}_2\text{O}_3$. The dependencies of $\text{CeO}_2/\text{Nb}_2\text{O}_5$ and $\text{CeO}_2/\text{La}_2\text{O}_3$ were different; these catalysts exhibited the maximum formation of N_2 at temperatures other than 400°C (the results obtained at these temperatures

are also listed in **Table 1**). However, the activities of all these CeO_2 /oxide catalysts were less than half of the activity of $\text{CeO}_2/\text{Al}_2\text{O}_3$ prepared from JRC-ALO-4. We note that the conversion of NO to N_2 over these CeO_2 /oxide catalysts except $\text{CeO}_2/\text{SiO}_2$ was higher than that over unsupported CeO_2 . These results show that the catalytic activity of CeO_2 for the selective reduction of NO with C_3H_6 is greatly promoted by supporting on metal oxides and that Al_2O_3 (JRC-ALO-4) is the best support.

The results in **Table 1** indicate that activities of the $\text{CeO}_2/\text{Al}_2\text{O}_3$ catalysts depended on the Al_2O_3 samples used. The catalyst prepared from JRC-ALO-4 was most active. We characterized the $\text{CeO}_2/\text{Al}_2\text{O}_3$ samples by using XRD to obtain information on structural differences. **Figure 2** shows the XRD patterns. Diffraction peaks due to CeO_2 and $\gamma\text{-Al}_2\text{O}_3$ were detected for all samples, indicating that Ce oxide was present as CeO_2 crystallites, and Ce-Al-O mixed oxides such as CeAlO_3 did not probably form because the diffraction peaks due to CeAlO_3 ($2\theta(\text{CuK}\alpha)=33.6, 23.6, 41.4$, etc.)²³⁾ were not detected. However, the sharpness of the XRD peaks due to CeO_2 clarified that crystallinity of the cerium oxide differed with the Al_2O_3 supports; CeO_2 on JRC-ALO-4 was most highly crystalline

CeO₂ was supported on (a) JRC-ALO-4, (b) JRC-ALO-1, and (c) Aerosil C. All samples were calcined at 500°C in air and analyzed before catalytic tests.

Fig. 2 XRD Patterns of CeO₂/Al₂O₃

whereas the sample prepared from JRC-ALO-1 exhibited the most broad diffraction peaks. This difference suggests that the crystallinity of CeO₂ on Al₂O₃ is more or less related to the activity of CeO₂/Al₂O₃ catalysts; thus crystalline CeO₂ is apparently suitable. On the other hand, Okazaki *et al.* reported that the catalytic activity of Al₂O₃ itself for selective NO reduction with C₂H₄ was largely dependent on the type of Al₂O₃; JRC-ALO-4 exhibited the highest activity²²⁾. They explained the difference by the purity of the Al₂O₃ samples. Such a difference in catalytic performance of Al₂O₃ itself can also be a reason for the difference in the activities of various CeO₂/Al₂O₃ catalysts. These results indicate that catalytic activities of the CeO₂/oxide catalysts for selective reduction of NO can be influenced by such properties as size, crystallinity, surface state, purity, etc. of CeO₂ and those of the oxide supports.

Figure 1 shows that CeO₂ was more active than Al₂O₃ for the reactions of NO to NO₂ and C₃H₆ to CO and CO₂. Thus, C₃H₆ was not efficiently utilized for the NO reduction on CeO₂ because it was combustibly consumed. In oxidation of various hydrocarbons, CeO₂ showed activity, which was rather high²⁴⁾. Unsupported Al₂O₃, in contrast, scarcely catalyzed the NO reduction to N₂, C₃H₆ oxidation, and NO oxidation in the temperature region (300–400°C, **Fig. 1**) where CeO₂/Al₂O₃ was active. Hence, CeO₂ must have played an important role in the selective catalytic reduction of NO over CeO₂/Al₂O₃.

It has often been claimed that selective catalytic reduction of NO with hydrocarbons proceeds via oxidation of NO to NO₂^{17),18),21),25)–29)}. The mechanism of NO selective reduction with C₃H₆ over Ce-ZSM-5 has been explained by oxidation of NO to NO₂ on Ce ions and by the following reaction between NO₂ and C₃H₆

that produced N₂, CO, and CO₂ via the formation of an organic intermediate containing N and O²⁵⁾. For the case of selective NO reduction over Pt/SiO₂^{21),26)} and Pt/Al₂O₃^{27),28)}, Pt has been regarded as an oxidizing site to form NO₂ from NO, and the oxidized species has been assumed to transfer onto the surface of SiO₂ or Al₂O₃ where NO₂ is reduced to N₂ with hydrocarbons^{20),21)}. Moreover, the selective reduction over Co/Al₂O₃ has also been presumed to involve oxidation of NO on the metals²⁹⁾. Since unsupported CeO₂ is shown to be highly active for oxidation of NO to NO₂, it is likely that CeO₂ on Al₂O₃ contributes to the oxidation of NO to NO₂ like Pt on Al₂O₃. The selective NO reduction over CeO₂/Al₂O₃ will proceed by oxidation of NO on CeO₂ and by the following reduction of NO₂ to N₂ on Al₂O₃, since Al₂O₃ has been found to be effective for NO₂ + C₃H₆ reaction⁴⁾.

Figure 3 compares catalytic activities of CeO₂/Al₂O₃, Pt/Al₂O₃, and Pt/CeO₂/Al₂O₃. They were prepared from JRC-ALO-4 used as the Al₂O₃ source. Pt/Al₂O₃ showed the maximum reduction of NO to N₂ at 300°C, the temperature which was lower than the temperature at which CeO₂/Al₂O₃ showed the maximum. The maximum conversion of NO to N₂ over Pt/Al₂O₃ was lower than that over CeO₂/Al₂O₃. The decrease in the activity of Pt/Al₂O₃ for the NO reduction at higher temperatures is due to the consumption of C₃H₆, which was completely oxidized to CO and CO₂ at temperatures higher than 300°C (**Fig. 3B**). Similar temperature dependence was observed for Pt/CeO₂/Al₂O₃ whose activity was somewhat lower than that of Pt/Al₂O₃.

The difference in the catalytic behavior between CeO₂/Al₂O₃ and Pt/Al₂O₃ would be related to the oxidizing capability of CeO₂ and Pt. We had reported elsewhere the relationship between catalytic activity for

The source of Al_2O_3 was JRC-ALO-4.

(A) Conversion of NO to N_2 , (B) Conversion of C_3H_6 to CO and CO_2 , (C) Conversion of NO to NO_2 , (D) Conversion of NO to N_2O .

Reaction conditions: NO 1000 ppm, C_3H_6 500 ppm, O_2 2% (He balance), total flow rate $90 \text{ cm}^3 \cdot \text{min}^{-1}$, catalyst weight 0.3 g ($W/F = 0.2 \text{ g} \cdot \text{s} \cdot \text{cm}^{-3}$).

Fig. 3 Temperature Dependence of the Selective Reduction of NO with C_3H_6 in the Presence of Excess O_2 over $\text{CeO}_2/\text{Al}_2\text{O}_3$, $\text{Pt}/\text{Al}_2\text{O}_3$, and $\text{Pt}/\text{CeO}_2/\text{Al}_2\text{O}_3$

$\text{NO} + \text{C}_3\text{H}_6 + \text{O}_2$ reaction and that for $\text{C}_3\text{H}_6 + \text{O}_2$ reaction, by using metal ion-exchanged mordenite catalysts¹³⁾. This relationship showed that Ce-mordenite, which had a moderate oxidizing capability in the $\text{C}_3\text{H}_6 + \text{O}_2$ reaction, exhibited the highest catalytic activity for NO reduction in the $\text{NO} + \text{C}_3\text{H}_6 + \text{O}_2$ reaction. The catalytic activity of the ion-exchanged mordenite for the selective reduction decreased as the oxidizing capability of the catalyst decreased, because NO is not efficiently oxidized to NO_2 on such materials. On the other hand, Pt-mordenite oxidized C_3H_6 very rapidly, and thus the activity of the Pt-contained catalyst for the selective reduction of NO was much lower than that of Ce-mordenite. Since Pt has a high oxidizing capability, C_3H_6 cannot be utilized efficiently for the NO reduction, but it was combustibly consumed over Pt-mordenite. This should be the case for the present system, in which the oxidizing capability of CeO_2 should be more moderate than that of Pt. Thus, $\text{CeO}_2/\text{Al}_2\text{O}_3$ would have high catalytic activity for the selective reduction of NO at relatively high temperatures at which $\text{Pt}/\text{Al}_2\text{O}_3$ oxidizes C_3H_6 excessively. The oxidizing capability of CeO_2 would be balanced with the ability of Al_2O_3 to reduce NO_2 to N_2 by utilizing C_3H_6 . From such a point of view, addition of Pt

onto $\text{CeO}_2/\text{Al}_2\text{O}_3$ would break the balance. In fact, the catalytic activity of $\text{Pt}/\text{CeO}_2/\text{Al}_2\text{O}_3$ for the selective reduction was somewhat lower than that of $\text{CeO}_2/\text{Al}_2\text{O}_3$.

In conclusion, $\text{CeO}_2/\text{Al}_2\text{O}_3$ is an efficient catalyst for selective reduction of NO with propene in the presence of excess oxygen in the temperature region of 300–400°C. CeO_2 on Al_2O_3 is probably the oxidation site of NO to NO_2 , and its moderate oxidizing capability suppresses propene waste; thus catalytic activity becomes higher than that of Al_2O_3 and $\text{Pt}/\text{Al}_2\text{O}_3$ in this temperature region.

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要 旨

担持酸化セリウムを触媒とする酸素共存下での NO のプロペンによる選択還元

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種々の酸化物に担持した酸化セリウムを触媒とする酸素共存下での NO のプロペンによる選択還元反応を検討した。CeO₂ を担持せずに触媒として用いた場合、NO の還元はあまり進行せず、むしろ酸化反応 (NO からの NO₂ の生成やプロペンからの CO, CO₂ の生成) が促進されたが、Al₂O₃, ZrO₂, Ga₂O₃ などの酸化物に担持することによって 300~400℃ の温度範囲で良好な NO 還元活性が得られた。中でも Al₂O₃ 担持 CeO₂ が最

も高い活性を示した (Al₂O₃ そのものはこの温度範囲ではほとんど活性を示さない)。また、この触媒の活性は同温度範囲で Pt/Al₂O₃ の活性よりも高かった。このような Al₂O₃ 担持 CeO₂ の触媒作用は、CeO₂ の適度な酸化力のため、CeO₂ 上でプロペンが過度に酸化消費されることなく NO が酸化活性化され、これが Al₂O₃ 上で還元されることによって進行する、と推定した。

Keywords

Nitrogen oxide, Oxide catalyst, Selective reduction, Cerium oxide