

Direct decomposition of nitrous oxide in the presence of oxygen over iridium catalyst supported on alumina

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Abstract

Direct decomposition of nitrous oxide (N_2O) on noble metal catalysts supported on alumina was examined in the presence of oxygen. The iridium catalysts supported on alumina showed higher activities than the other noble metal catalysts. Although the catalyst activity was affected by oxygen formed by N_2O decomposition at lower temperatures, desorption of oxygen proceeded promptly at the temperature $> 350^\circ C$, and the catalytic activity was recovered by increasing the reaction temperature from 350 to $400^\circ C$. Therefore, the Ir/ Al_2O_3 catalyst can be used for N_2O decomposition in the presence of oxygen at relatively higher temperatures.

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1. Introduction

Nitrous oxide (N_2O) is one of harmful gases in our environment because of contribution to the destruction of the stratospheric ozone layer. Nitrous oxide also has the greenhouse effect and its global warming potential is 310 times larger than that of CO_2 (Houghton et al., 2001; Centi et al., 1999). To protect our environment, N_2O must be decomposed or reduced to harmless N_2 . Nitrous oxide is produced by both natural and anthropogenic sources, and the latter includes nitric acid and adipic acid plants and fluidized bed combustors for sewage-sludge or industrial wastes (Centi et al., 1997) besides the medical exhaust and agricultural emission.

Various types of catalysts such as noble metals (Centi et al., 1997; Haber et al., 2004; Kapteijn et al., 1996; Imamura et al., 1996; Yuzaki et al., 1998), metal oxides (Satsuma et al., 2000; Drago et al., 1997; Kannan and Swamy, 1999; Armor et al., 1996; Chellam et al., 2000), and ion-exchanged zeolites (Pieterse et al., 2004; da Cruz et al., 1998; Kameoka et al., 2000; Pérez-Ramírez et al., 2001) are

reported to be active for N_2O decomposition. However, few of them are active and stable enough for practical applications, because their activities are severely inhibited by the presence of other gases such as O_2 . Noble metal catalysts are known to have quite high activities for decomposition of N_2O , and Rh catalyst was reported to catalyze the reaction even at room temperature (Centi et al., 1997). However, oxygen atoms formed by the decomposition of N_2O were hardly desorbed from the catalyst surface and accumulated on the Rh surface; the activity of the catalyst decreased suddenly after keeping high N_2O conversion for a while (Centi et al., 1997; Tzitzios and Georgakilas, 2005). Therefore, further development of the catalysts is required for the practical use of the catalyst to remove N_2O from the exhaust gases. In the present study, N_2O decomposition in the presence of oxygen over iridium catalysts was examined.

2. Experimental

2.1. Preparation of supported catalysts

Chlorides of iridium ($IrCl_4$; Wako), ruthenium ($RuCl_3 \cdot nH_2O$ ($n \approx 1.5$); Wako), rhodium ($RhCl_3 \cdot 3H_2O$; Wako) and palladium ($PdCl_2$; Kanto Chemical) were used without further

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purification. For alumina supports, γ -alumina (JRC-ALO-8; standard catalyst from Catalysis Society of Japan) and spherical alumina (γ - and δ -alumina phases, Nanophase Tech. Corp.) were used as received and another γ -alumina sample was prepared by calcination of pseudoboehmite (Vista Chemical) at 800 °C for 30 min in air. Preparation of the other catalyst supports was described elsewhere (Ohnishi et al., 2006, 2007; Inoue et al., 1991, 1993). The catalysts were prepared by impregnation of the supports with aqueous solutions of noble metal chlorides at 80 °C. The dried catalysts were calcined in air at 700 °C for 30 min.

2.2. Catalyst test

Catalyst tests were carried out in a fixed-bed flow reactor. The catalyst was tableted and pulverized into 10–22 mesh, and 0.5 g of the catalyst was set in the reactor. The catalyst bed was heated to 500 °C in a helium gas flow and held at that temperature for 30 min. Then, the reaction gas composed of 5000 ppm N_2O , 2% O_2 , and He balance was introduced to the catalyst bed at $W/F = 0.3 \text{ g s ml}^{-1}$. The effluent gases from the reactor were analyzed every 5 min with an on-line micro-gas-chromatograph (CP 2002, Chrompack, Netherlands) and data after keeping the reaction temperature for 30 min are given in this paper. Reaction temperature was decreased from 500 °C to the temperature where the catalyst showed negligible N_2O conversion (1d stage). Then, the catalyst activity was measured by increasing the reaction temperature (2i stage).

Note that for the catalysts examined in this paper, formation of NO was not observed, and the yield of N_2 was essentially identical with the conversion of N_2O , indicating that selectivity for N_2 was 100%.

2.3. Characterization

Powder X-ray diffraction (XRD) patterns were recorded on a Shimadzu XD-D1 diffractometer using Cu $K\alpha$ radiation and a carbon monochromator. The specific surface area was calculated by the BET single-point method on the basis of nitrogen uptake measured at 77 K. Temperature-programmed desorption of O_2 was measured using the fixed-bed flow reactor. After an adequate pretreatment of the catalyst, it was heated to 500 °C at a constant heating rate in a He flow and desorbed O_2 was analyzed with a Pfeiffer Vacuum Oministar GSD 301 O 1 quadrupole mass spectrometer.

3. Results

3.1. N_2O decomposition over various noble metal catalysts supported on alumina

Activities of various noble metal catalysts supported on γ -alumina (JRC-ALO-8) for N_2O decomposition in the presence of 2% oxygen were examined first. The noble metal catalysts showed high activities, while the catalyst support, γ -alumina, exhibited negligible activity. The catalytic activity

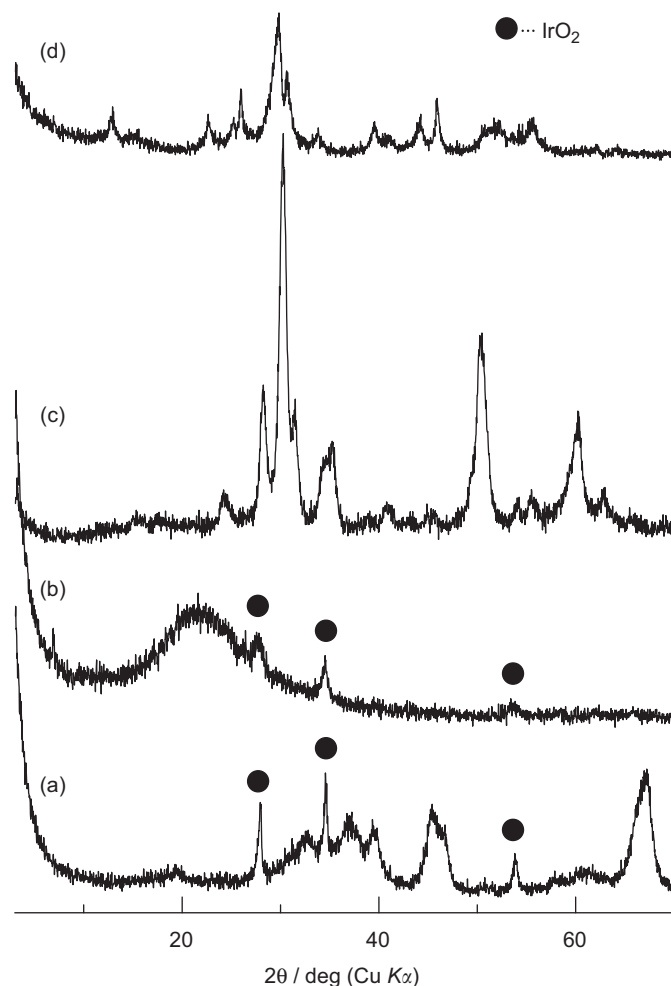


Fig. 1. XRD patterns of the iridium catalysts supported on: (a) Al_2O_3 (JRC-ALO-8); (b) SiO_2 ; (c) ZrO_2 , (d) La_2O_3 . The loading of iridium metal was adjusted to 1 wt.%.

decreased according to the following order: $\text{Ir}/\text{Al}_2\text{O}_3 > \text{Rh}/\text{Al}_2\text{O}_3 > \text{Ru}/\text{Al}_2\text{O}_3 > \text{Pd}/\text{Al}_2\text{O}_3 \gg \text{Al}_2\text{O}_3$.

3.2. N_2O decomposition over the iridium catalysts supported on various metal oxides

Fig. 1 shows the XRD patterns of the iridium catalysts supported on various metal oxides. The sample supported on alumina showed the relatively sharp, intense peaks due to iridium oxide. The catalyst that exhibited the intense diffraction peaks due to iridium oxide showed higher activity for N_2O decomposition in the presence of 2% oxygen than the samples that showed only the peaks due to the support.

The catalytic activity decreased according to the following order of the support: $\text{Al}_2\text{O}_3 > \text{Co}_3\text{O}_4 > \text{SiO}_2 \approx \text{TiO}_2 \approx \text{ZrO}_2 > \text{ZnO} \approx \text{NiO} > \text{La}_2\text{O}_3 > \text{CuO} \approx \text{MgO}$. Note that some supports such as Co_3O_4 and NiO had activities for decomposition of N_2O (Ohnishi et al., 2006, 2007), and loading of the iridium species rather destroyed the activity of the support; thus, the $\text{Ir}/\text{Co}_3\text{O}_4$ catalyst showed lower activity than the Co_3O_4 catalyst (Ohnishi et al., 2006).

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