Influence of Al_2O_3 support on the activity of Ir/Al_2O_3 catalysts for NO reduction with C_3H_6 and CO in slight lean conditions

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The NOx abatement from lean-burn engines is still difficult problem because of the presence of excess O_2 in the exhaust. To overcome this problem, the selective reduction of NO with urea and NOx storage/reduction techniques have already been developed, and applied to diesel and gasoline lean-burn engines. The use of fuel-derived reductants such as hydrocarbons and CO for the reduction of NO is also regarded as practically effective way. Supported precious metal catalysts are known to show the NO reduction activity in the low and narrow temperature region. However, the removal of NOx in high speed driving conditions, which correspond to slight lean and high temperature exhaust gas, is also required to meet the strict emission regulations. We have recently found that supported Ir catalyst can effectively catalyze the NO reduction with C_3H_6 and CO in slight lean condition [1]. In this study, we have devoted to study the effect of Al_2O_3 support on the activity of Ir/Al_2O_3 for NO reduction.

A series of Ir (1wt%) supported on Al_2O_3 was prepared by impregnation with 5 different Al_2O_3 powders with $Ir(NO_3)_4$, followed by drying and calcination at 600 °C: commercial (NK-324 (Al-1), MSC (Al-2)), sol-gel (Al-3), hydrolysis (Al-4) and precipitation Al_2O_3 (Al-5). The catalytic activity test was carried out using a flow reactor system by passing a slight lean reaction gas mixture containing NO (500 ppm), CO (0.1%), C_3H_6 (300 ppm), O_2 (0.5%) and H_2O (5%) diluted in He at a rate of 50 cm^3 min^{-1} over 30 mg of catalyst, which had been pretreated *in situ* in the flow of reaction gas at 600 °C for 1 h.

The NO reduction activity of Ir/Al_2O_3 was found to be different depending on Al_2O_3 support. Among the catalysts tested here, Ir/Al-2 prepared by using MSC-Al_2O_3 purchased from Mizusawa Chemicals showed the highest NO reduction activity. Although Ir dispersion estimated by CO chemisorption was also different depending on Al_2O_3 support, no good correlation between NO reduction activity and Ir dispersion was observed.

According to the previous report [2], it can be expected that metallic Ir^0 act as catalytically active species. In fact, the NO reduction activity of Ir/Al-2 was significantly increased after the reduction treatment with H_2 at 400 °C. On the other hand, the reaction gas contains an excess amount of O_2, leading to the consideration that Ir species are oxidized during the reaction. We performed TPO measurements to examine the oxidation behavior of Ir species, and found that the temperature for the oxidation of Ir species is strongly dependent on Al_2O_3 support. This suggests that the oxidation state of Ir species during the reaction is different. Fig. 1 shows the relationship between the temperature for Ir oxidation and the reaction rate for NO reduction at 420 °C. It appears that the NO reduction activity depends on the oxidation state of Ir species. In order to clarify the role of Al_2O_3, the surface acid-base properties were evaluated by NH_3- and CO_2-TPD. As can be seen in Fig. 1, the stability of Ir species in the reducing state was found to be increased with increasing the surface acidity of Al_2O_3, where the surface acidity was estimated by NH_3/CO_2 ratio calculated from the amount of NH_3 and CO_2 desorption. We can conclude that the role of Al_2O_3 support is to stabilize the Ir species in catalytically active state.

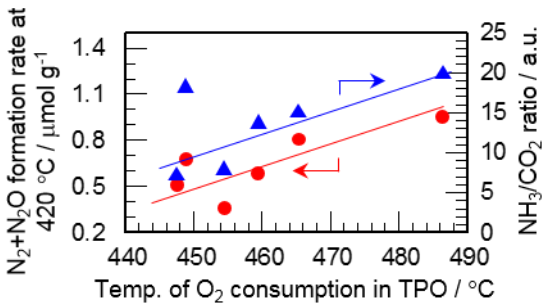

Fig. 1. Influence of oxidation behavior of Ir and surface acidity of Al_2O_3 on the NO reduction activity.

REFERENCES

[1] R. Burch, Catal. Rev., 46 (2004) 271.
[2] Y. Doi, M. Haneda, 118th Meeting of Catal. Soc. Jpn. A, (2016) 1H15.