Strong Enhancement of CO-H₂-SCR of NO over Ir Catalyst Supported on Na-zeolite

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Introduction
A selective catalytic reduction of NO in the presence of excess oxygen is a potential method to remove NOx from lean-burn and diesel exhausts. Among a number of catalysts, Ir catalysts are effective in the reduction of NO by not only unsaturated hydrocarbon, but also CO and hydrogen. [1-4]. However, Ir catalysts show lower NO reduction activity at lower temperatures below ca. 500 K. Hence, the extension of active window of Ir catalysts to lower temperatures is strongly desired. In this study, we discover that Ir catalysts show high activity below 500 K by combination of co-presence of CO and H₂ and use of Na-zeolite supports. By means of Ir LIII-edge XANES, the reason for high activity is clarified to be low oxidation state and high dispersion of iridium species [5].

Experimental
Ir catalysts were prepared by impregnating supports with an aqueous solution of IrCl₄ followed by evaporation to dryness at 303 K and by calcination in air at 773 K for 4 h. Loaded Ir amount was 0.5 wt%. The catalytic test was performed with a fixed-bed flow reactor by passing a mixture of 1000 ppm NO, 3000 ppm CO, 5000 ppm H₂ and 2% O₂ in He at a rate of 100 cm³ min⁻¹ over 0.05 g catalyst. Ir LIII-edge XANES spectra of the samples treated with exposing reaction gas mixtures were recorded in fluorescence mode by using Lytle detector at the BL-10B station [6] at KEK-PF with a Si(311) channel cut monochromator.

Results and discussion
The SCR under the co-presence of CO and H₂ was carried out with various supports at 448-673 K. Among Ir/zeolites, Ir/Na-MOR exhibited higher NO reduction activity than Ir/H-MFI below 500 K. Particularly, NO conversion on Ir/Na-MOR was 38% at 473 K. This low temperature catalytic activity is much higher than that of 5wt% Ir/SiO₂ for the CO-SCR in the presence of SO₂ [5]. On the other hand, Ir/H-MOR showed NO reduction around 573 K, and little activity below 500 K. NO reduction activity of Ir/MgO was very low over the whole reaction temperatures. The order of NO reduction activity at 473 K was Ir/Na-MOR > Ir/Na-MFI > Ir/H-MFI = Ir/SiO₂ > Ir/SiO₂-Al₂O₃ > Ir/H-MOR > Ir/Al₂O₃ > Ir/MgO.

Figure 1A shows Ir LIII-edge XANES spectra of IrO₂, Ir/H-MFI, Ir/Al₂O₃, Ir/H-MOR, Ir/Na-MOR, Ir/SiO₂ and Ir metal. Figure 1B shows Ir LIII-edge XANES spectra of various Ir catalysts after the SCR under the co-presence of CO and H₂. The white line of Ir LIII-edge XANES spectrum is assigned to the electron transition to 5d³/² and 5d⁵/² orbitals, it is supposed that the white line intensity is an informative indication for the oxidation state of iridium. Comparing with the catalytic tests, very high SCR activity of Ir catalysts Na-zeolite supports can be attributed to the remarkable formation of highly active metallic iridium species.

References

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