

## Effect of Support on Activity of Selective Reduction of NO by Hydrogen over Pt-based Catalysts

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### Introduction

The catalytic property of supported noble metal catalysts can be modified with acid-base property of support materials. We have studied the support effect on the activity of Pt catalyst in propane combustion, and concluded that more metallic platinum shows higher catalytic activity, which was obtained on the acidic support. The electrophilic property of the support improves the oxidation-resistance of platinum, and as a result, the catalyst exhibits high catalytic activity [1].

Selective catalytic reduction (SCR) of NO from fuel exhausts is of major environmental importance, and low temperature SCR system is desired. SCR by H<sub>2</sub> over Pt-based catalysts is one of the promising candidates [2]. In the present report, in order to obtain a controlling factor for H<sub>2</sub>-SCR over Pt-based catalysts, support effect on the activity of Pt-based catalysts was investigated by using various types of Pt-based catalysts.

### Experimental

Pt/zeolite catalysts were prepared by a conventional ion-exchange in an aqueous Pt(NH<sub>3</sub>)<sub>4</sub>Cl<sub>2</sub> solution at room temperature. Pt/metal oxide catalysts were prepared by impregnating an aqueous Pt(NH<sub>3</sub>)<sub>4</sub>Cl<sub>2</sub> solution. These precursors were calcined at 773 K for 3 h in a flow of dried air. Pt content was 1 wt% for all the catalysts. The catalytic activity was measured with a fixed-bed flow reactor at atmospheric pressure. The reaction gas contained a mixture of 1000 ppm NO, 5000 ppm H<sub>2</sub>, 6.7% O<sub>2</sub> in He was fed to a 0.05 g of catalyst at a total flow-rate of 100 cm<sup>3</sup>min<sup>-1</sup>, corresponding to GHSV = 78,000 h<sup>-1</sup>. After reaching a steady-state, the effluent gas was analysed by a gas chromatograph and a chemiluminescence NOx analyzer.

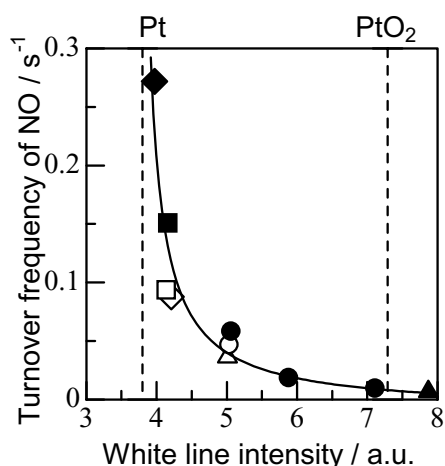
The Pt L<sub>III</sub>-edge XAFS measurement of platinum catalyst used in the reaction was performed at room temperature under atmosphere in transmission mode at the BL-10B station at KEK-PF [3] with Si (311) channel cut monochromator.

### Results and discussion

Fig. 1 shows a correlation between turnover frequency (TOF) of NO reaction at 348 K and white line intensity of Pt L<sub>III</sub>-edge XANES, which is an index of oxidation state of Pt particles [1]. In the separate study, the linear correlation between the oxidation state and the white line area intensity has been confirmed [4]. As shown in the

figure, turnover frequency of NO is strongly dependent on the oxidation state of Pt, i.e., metallic platinum on SiO<sub>2</sub> shows high activity, and while highly oxidized Pt on MgO only shows poor activity. This is also in good agreement with our previous report on propane combustion over Pt-based catalysts [1]. It should be noted this correlation is common to both metal oxide supported catalysts and zeolite supported catalysts. Therefore, this figure clearly indicates that oxidation state of Pt is an essential factor for H<sub>2</sub>-SCR activity.

In this result, a good correlation was observed between the acid-base property of support and the oxidation state of the platinum, except for silica-supported Pt catalyst; the acidic support provided metallic platinum (low white line intensity).



**Fig. 1** Turnover frequency of NO over Pt-based catalysts as a function of white line area intensity estimated from Pt L<sub>III</sub>-edge XANES. (◇) Pt/MOR, (□) Pt/MFI, (△) Pt/BEA, (○) Pt/Y, (■) Pt/SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>, (◆) Pt/SiO<sub>2</sub>, (●) Pt/Al<sub>2</sub>O<sub>3</sub> and (▲) Pt/MgO.

### References

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