

## XAFS Study of Stability of Pt on Pt/Nb-AlMCM-41 in a Redox Atmosphere

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### 1 Introduction

The selective catalytic reduction of NO with hydrogen (H<sub>2</sub>-SCR) in the presence of excess oxygen has attracted attentions due to the activity at temperatures as low as 373 K. Several studies on Pt- or Pd-supported catalysts report two active temperature ranges in H<sub>2</sub>-SCR [1-3]. Two explanations were suggested for this: different states of active metals [2,3] and change in reaction mechanism [1]. Recently, we found that Pt-loaded AlMCM-41 (aluminum-containing mesoporous silica MCM-41, Pt/AlM41) also shows two active temperature ranges (approximately 373 and 500 K) in H<sub>2</sub>-SCR and that addition of niobium to Pt/AlM41 (Pt/Nb-AlM41) greatly enhances catalytic activity at 373-650 K. In this study, the possibility that differences between reduced and oxidized metals might result in two active temperature ranges on H<sub>2</sub>-SCR over Pt/Nb-AlM41 was investigated using the XAFS spectroscopy.

### 2 Experiment

M41 was synthesized using colloidal silica Snowtex-20 (Nissan Chemical Industries, Ltd.) and C<sub>12</sub>H<sub>25</sub>N(CH<sub>3</sub>)<sub>3</sub>Br (Tokyo Kasei Kogyo Co., Ltd.). Niobium ion was loaded by the template ion-exchange (TIE) method [4]. The resulting powder was calcined at 873 K for 6 h in air to remove the templates. Platinum was then impregnated onto calcined and Nb-loaded M41 using aqueous H<sub>2</sub>PtCl<sub>6</sub>. Platinum loading was 2 wt% metal. The sample was again calcined at 873 K for 6 h in air. The catalyst was abbreviated as Pt/NbX-M41, where X is the Si/Nb atomic ratio. The XAFS spectrum of Pt L<sub>3</sub>-edge was recorded on BL-9C of PF-KEK.

### 3 Results and Discussion

Fig. 1a shows the Pt L<sub>3</sub>-edge XANES spectra of Pt/AlM41 and Pt/Nb16-AlM41 samples. Both spectra were similar to platinum foil. The coordination numbers of platinum for Pt/AlM41 and Pt/Nb16-AlM41, which were calculated from the EXAFS data using the least-square method, were 9.8 and 10.0, respectively. These results indicate that platinum on M41 was not influenced by the co-loaded niobium.

The oxidation state of platinum under reducing or oxidizing atmospheres was measured using the Pt L<sub>3</sub>-edge XANES spectrum (a white line) at 11.564 keV, which is attributed to the electron transition from the 2p to 5d orbitals, and is a good indicator of the oxidation state of platinum [5,6]. Fig. 1b summarizes the change in white line intensity during redox cycles for Pt/AlM41 and Pt/Nb16-AlM41. Two findings are apparent. Peak heights were low upon reduction and high upon oxidation

of both samples. Pt species supported on oxides have been reported to give peak heights of 1.8-2.0 and 1.2-1.4, respectively, upon oxidation and reduction [6-8]. The values in Fig. 1b, 1.42-1.50, indicate that Pt supported on AlM41 was usually in a reduced state. In addition, the range for Pt/Nb16-AlM41 was smaller than for Pt/AlM41, revealing that Pt loaded on the former remained metallic. This finding is in good agreement with reports that platinum remained metallic on acidic supports [5,9]. Both findings indicate that the two active temperature ranges observed in H<sub>2</sub>-SCR did not result from differences between Pt and PtO (or PtO<sub>2</sub>).

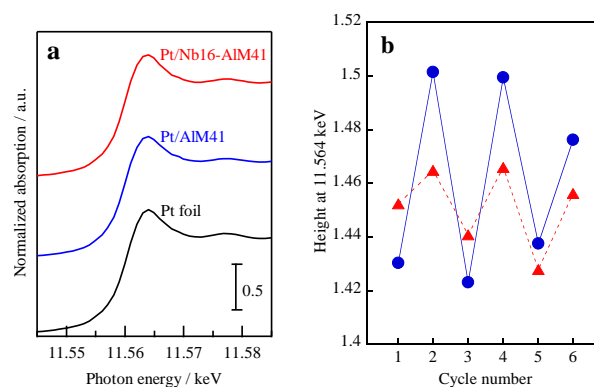


Fig. 1: (a) Pt L<sub>3</sub>-edge XANES spectra of Pt/AlM41 and Pt/Nb16-AlM41 evacuated at 773 K. (b) Change in height of white line at 11.564 keV in Pt L<sub>3</sub>-edge XANES spectra of (circle) Pt/AlM41 and (triangle) Pt/Nb16-AlM41 with redox treatments. The sample was reduced by H<sub>2</sub> at cycle numbers 1, 3, and 5, and oxidized by O<sub>2</sub> at 2, 4, and 6. Each reduction or oxidation was performed at 773 K (P<sub>H2</sub> 13.3 kPa or P<sub>O2</sub> 13.3 kPa) for 10 min.

### References

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