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Effect of Sb additive on Structure of Pt catalyst supported on silica

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Introduction

The selective oxidation of CO in hydrogen is a challenging issue in the domain of catalytic science. Recently, the development of highly selective and durable catalysts has attracted much interest for forming pure hydrogen from methanol reforming gas mixture. We have found that Sb added Pt catalyst showed significant enhancement of CO removal accompanied with a suppression of hydrogen consumption. We report here the structural transformation of Pt particle studied by XAFS spectroscopies.

Experimental

Pt supported on silica catalysts were prepared as follows. SiO_2 (Davisil^{TM} 635, surface area 480 $m^2 \cdot g^{-1}$) was impregnated with aqueous solution of $H_2PtCl_6 \cdot 6H_2O$ followed by drying at 393 K and calcination at 773 K. The loading of Pt was 1 wt %. Transmission electron microscopic observation showed that the particle size was distributed with centring at 8 nm. The above Pt/SiO_2 was impregnated in an aqueous solution of $SbCl_5$, dried and finally calcined at 673 K. The five kinds of catalysts (*i.e.* Sb/Pt = 0, 0.01, 0.05, 0.1 and 0.5) were stored in a desiccator. In addition of Sb, metal particles redispersed into the distributions around 5 nm.

0.05 g catalyst was mounted in a Pyrex closed circulating system and calcined at 673 K followed by 673 K reduction. The powder was transferred into an EXAFS cell in vacuum or in 133 kPa of CO. All spectra were measured in a transmission mode at BL-10B. Data were processed by EXAFS analysis software, REX2000 (Rigaku).

Results and Discussion

Fourier Transforms of Pt L_3 EXAFS spectra were shown in Fig. 1. Only Pt shell was observed. The curve fitting results were summarized in Table 1. The coordination number increased according to Sb addition in spite of the decrease in particle size in TEM photographs. In the presence of CO, it decreased considerably, which is probably caused by the split of Pt-Pt bond by adsorption of CO. However, the reduction of *N* is abnormally large for the Pt particle more than 5 nm. On the other hand, Pt-Pt bond length did not change at all.

The intensity of white lines of Pt L_2 and L_3 edges decreased with the amount of Sb, suggesting the increase of 5d electron density. This can be simply explained by the difference in the ionic potential (IP(Pt) < IP(Sb)) and the electron affinity (EA(Pt) > EA(Pt)).

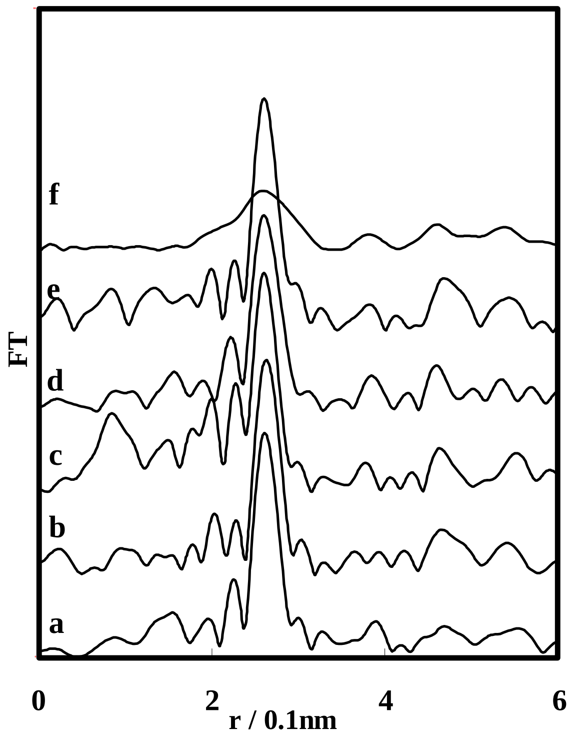

Figure. 1 Fourier Transform of Pt L_3 edge EXAFS of the catalysts. a : Pt/SiO_2, b: Pt-Sb/SiO_2 (Sb/Pt = 0.01), c: Pt-Sb/SiO_2 (Sb/Pt = 0.05), d: Pt-Sb/SiO_2 (Sb/Pt = 0.1), e: Pt-Sb/SiO_2 (Sb/Pt = 0.5) and f: CO adsorbed on Pt-Sb/SiO_2 (Sb/Pt = 0.05).

Table 1. Structural parameters of Pt

Sb/Pt	*N*	*r* /nm	*E$_0$* /eV	*Dw*
0	9.4	2.77	1.35	0.062
0.01	9.5	2.78	2.18	0.062
0.05	9.8	2.77	2.20	0.050
0.1	10.8	2.77	-0.30	0.065
0.5	10.1	2.77	0.30	0.061
1.0	11.6	2.76	-0.91	0.07
0.05 in CO	7.3	2.77	-0.31	0.057

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