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Quantification of deposit carbons on supported Pt particles by Pt L XANES

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Introduction

Carbon deposition is a major factor of catalyst deactivation. Thus, to know the amount of deposited carbon on catalyst surfaces spectroscopically is an important issue to understand the mechanism of the deposition and to develop robust catalysts against carbon deposition. X-ray absorption spectroscopy has a great advantage for in situ measurement, because gas ambient does not absorb hard X-rays. We measured Pt L edge XANES spectra of supported Pt catalysts at various amounts of deposited carbon. We related the change of whiteline intensity to the amount of deposited carbon. The effect of supports and carbon sources were also investigaged.

Experimental

Pt L edge XANES spectra were measured at BL-10B with a transmission mode at room temperature. CH_4, C_2H_4, and benzene were admitted to supported Pt catalysts at 573-673 K to deposit carbons. The amount of deposited carbon was estimated by the amount of CO_2 produced by the calcinations after XANES measurement.

Results

When carbon was deposit on Pt/SiO_2, the Pt L edge XANES was slightly changed. Substracted spectra between the spectra before and after the depositon were calculated to extract the change. Figure 1 shows the substracted Pt L_{III} edge XANES spectra. The intensity at 11563 eV decreased and that at 11569 eV increased by carbon deposition. The peak intensity at 11569 eV increased with carbon amount linearly. However, the increasing slope is affected by support and carbon source. When CH_4 was used as a source, the intensity changed heavily at a small amount of deposited carbon. On the other hand, when benzene was used, the intensity change was small. The result indicates that there are at least two types of deposited carbons on catalyst surfaces. It is well known that there are two types of deposted carbons, carbitic and graphitic carbons. It is probable that carbotic and graphitic carbons were formed by addmitting methane and benzene, respectivley. If so, the results of XANES study suggest that carbidic carbons affect the XANES spectra more than graphitic carbons.

Figure 2 shows difference spectra of Pt L_I edge XANES of Pt/SiO_2. Carbon deposition was done using methane, ethane, ane benzene. When the carbon was deposited from methane (carbidic carbon would be formed), a positive peak appeared at 13900 eV. On the other hand, a negative peak appeared when benzene was addmitted to the sample. This result indicates that the amount of carbidic and graphitic carbons can be obtained separately by combining L_I and L_{III} XANES. The detail is under investigation.

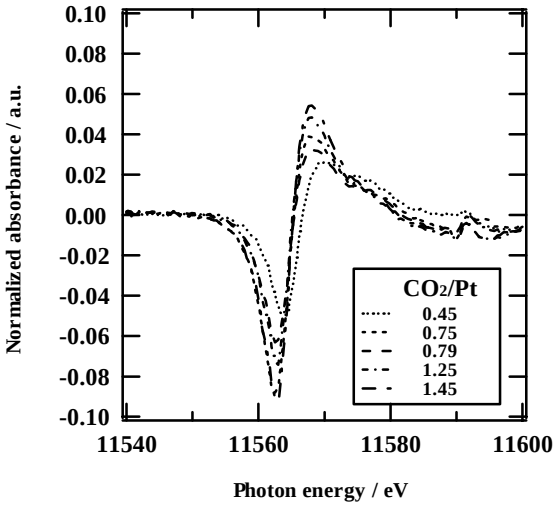

Figure 1. Changes in Pt L_{III} edge XANES by carbon deposition of Pt/SiO_2 at several carbon coverage.

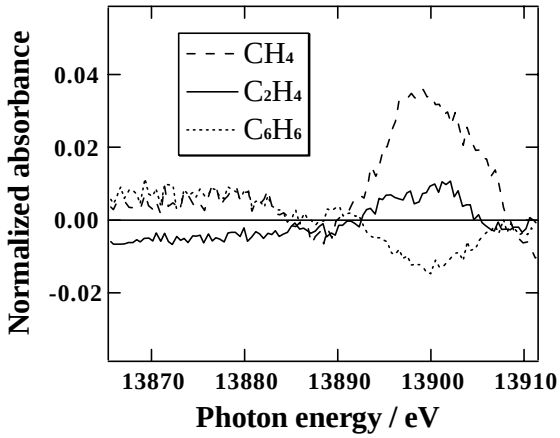

Figure 2. Changes in Pt L_I edge XANES by carbon deposition of Pt/SiO_2. Carbon was deposited from methane, ethylene, and benzene.