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Structural Analysis of Pd loaded MCM-41 Catalysts for Hydrogenation of Benzene by Means of XAFS

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Introduction

Metal–support interaction is important in understanding the structure and catalytic performance of supported metal catalysts. Indeed, the acidity of supports had crucial influence on the dispersion and catalysts of PdO [1]. In the previous studies, we found that the Pd loaded Al-MCM-41 was active in the hydrogenation of benzene. It could be supposed that the Lewis acid sites generated on Al influenced the generation of the active Pd species. In this study, in order to elucidate the structural and electronic effects of Lewis acidity on Pd in Al-MCM-41, Pd K-edge EXAFS of Pd/Al-MCM-41 was measured and the data was correlated with the catalytic performance of Pd.

Experimental

Pd was loaded on Al-MCM-41 with different Al concentration and through the impregnation method. Pd loadings were fixed at 1 wt% in every sample. The samples were calcined at 773 K in an O_2 flow, followed by reduction in 6% H_2 at 423 K. The acid amount of catalysts was measured using NH_3 TPD method. EXAFS spectra were measured at BL10B station of PF. A Si(311) single crystal was used to obtain monochromatic X-ray beam. Two ion chambers filled with Ar and Ar50%/Kr50% were used as detectors of I_0 and I, respectively. The sample was transferred to an aluminum cell with two Kapton windows connected to a flow system without contacting air. The analysis of the data was carried out according to the usual curve fitting method. In the analysis, the phase shift and backscattering amplitude were extracted from the spectrum of Pd foil measured at the same conditions.

Results and Discussion

Figure 1 shows the Fourier transforms of the Pd loaded on Al-MCM-41 with different concentrations as well as Pd on SiO_2-Al_2O_3 and Al_2O_3 supports. As can be seen in the figure, the intensity of the nearest-neighbor Pd-Pd boned changed depending on the kind of supports.

Figure 2 shows the acid amount of the catalysts and coordination numbers of the nearest-neighbor Pd-Pd bond determined from the EXAFS spectra plotted as a function of Al concentration of Al-MCM-41 support. From the figure, a good correlation was obtained between changes in the Pd-Pd coordinating numbers and the acid amount of Al-MCM-41. Namely, the highest acid amount was observed when Al concentration reached to 2.5 mol kg^{-1}. At this loading, the Pd-Pd coordination number was lowest among the tested samples. At the same time, Pd/Al-MCM-41 exhibited the highest hydrogenation activity at 2.5 mol kg^{-1}. Taking the above data into account, it can be supposed that the presence of Lewis acid sites in Al-MCM-41 affected the generation of the well-dispersed Pd particles, which was active in the hydrogenation of benzene.

References

[1] K. Okumura et al., Catal. Surv. Japan 5, 121 (2002).

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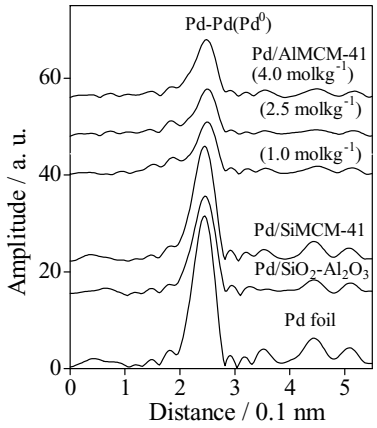

Figure 1. Pd K-edge EXAFS Fourier transforms of Pd loaded on Al, Si-MCM-41, SiO_2 –AlO_3 and Pd foil.

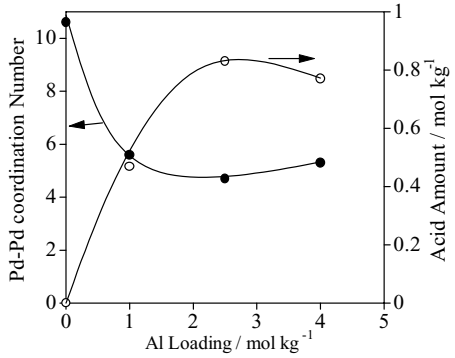

Figure 2. Relationship between nearest-neighbour Pd-Pd coordination number, acid amount of Pd/Al-MCM-41 and Al loading of Al-MCM-41.