

Local Structure Analysis of Rhodium Supported Alumina for Dry-reforming Reaction using In-situ XAFS

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The dry-reforming reaction is a useful reaction by which hydrogen and carbon monoxide are produced from methane and carbon dioxide which are greenhouse gases as industrial raw materials. Recently, the importance of this reaction is increased more and more. However, various discussions are performed for this reaction mechanism but a definite conclusion is not obtained.

In this research, the approach by XAFS was attempted to elucidate the reaction mechanism about the rhodium supported alumina catalyst which is carbon-free catalyst. Rh K-edge XAFS was measured by the transmission mode with BL38B1 of SPring-8 (edge energy=23.2keV).

Samples were set in the In-situ cell, and pretreated for one hour at 600°C under the hydrogen gas flow. And so Samples were treated with carbon dioxide and CO₂-CH₄ mixture gas for one hour at 600°C. After there treatments, Rh K-edge XAFS was measured at the same temperature and atmosphere.

The results were shown in Fig.1 and Fig.2. In XANES spectra (Fig-1), a structural change had not taken place around rhodium after carbon dioxide treatment and CO₂-CH₄ reforming. In EXAFS spectra (Fig:2), the intensity of Rh-Rh bond has not changed

after carbon dioxide treatment and dry-reforming. As for the activation of carbon dioxide, it is understood not to be caused by rhodium. Perhaps, the activation of carbon dioxide is presumed to cause on supports. The dry-reforming reaction does not take place on a single alumina, and so it is thought that the activation of methane is caused by rhodium metals.

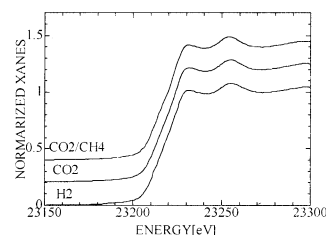


Fig.1 Rh K-edge XANES spectra of 0.5%Rh/Al₂O₃ catalysts under the reaction condition (600°C)

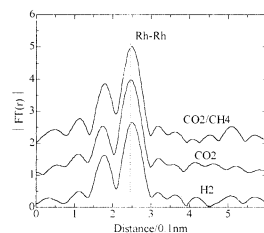


Fig.2 FT magnitude of k -weighted EXAFS for 0.5%Rh/Al₂O₃ catalysts under the reaction condition (600°C)

XAFS studies of Local Structure about Mn atoms in Diluted Magnetic Semiconductors (Ga,Mn)N

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The GaN-based diluted magnetic semiconductors (DMS) have been attracting much attention because of a theoretical prediction that this system has high Curie temperature (T_c) exceeding a room temperature.[1] We succeeded to grow a film of the GaN-based DMS doped with Mn of about 3% ((Ga,Mn)N) by use of the molecular beam epitaxy (MBE) system. This sample showed ferromagnetism with T_c much higher than 400 K.[2]

In order to investigate the local structure of Mn atoms in this film, we carried out the extended x-ray absorption fine structure (EXAFS) measurement of the K-edge of Mn at BL38B1. The experiments were performed by a fluorescence-yield mode using an array of 19 elements of Ga solid-state detectors because this sample was a thin film with a thickness of 360 nm. As a reference, we measured the Ga K-edge. The figure shows the Fourier transform spectra of the EXAFS oscillations. These spectra indicate the radial distribution of atoms around Mn and Ga, respectively. This figure shows clearly that the distribution of atoms around Mn is in good agreement with that around Ga.

This result suggests that Mn is substituted for Ga site of GaN in this film. The RHEED observation and the x-ray diffraction revealed that the single crystal films of the wurtzite type (Ga,Mn)N are grown. Therefore, it can be regarded that the peaks of the atomic distributions indicated by arrows in the figure are corresponding to the first nearest N atoms and the second nearest Ga atoms in the wurtzite GaN lattice, respectively.

[1] T. Dietl et al., Science **287** (2000) 1019.

[2] S. Sonoda et al., to be published in J. Crst. Growth.

