7C/2007G687

XAFS study on the CuMn-spinel derived catalyst for the steam reforming of dimethyl ether

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Introduction

Fuel cells have attracted much attention as clean and high efficient power generators for stationary and mobile use. From the viewpoint of reformability, dimethyl ether (DME) is one of the most promising fuels¹.

CuMn-spinel combined with acidic oxides, *i.e.* H-ZSM-5, showed very high activity with good selectivity toward steam reforming of DME². To clarify the reason for the high activity, the nature of Cu in CuMn spinel was investigated by Cu K-edge XAFS measurements, and compared to Cu in non-spinel oxides.

Experimental

CuMn spinel oxide $(CuMn_2O_4)$ was prepared by calcination of CuMn mixed oxide powder at 1173 K. The non-spinel CuMn was obtained by calcination of the same powder at 573 K. Molar ratio of Cu:Mn is regulated to 1:2. The as prepared catalyst and the hydrogen reduced catalyst were denoted as (ox) and (red), respectively.

Cu K-edge XAFS spectra were collected at BL-7C with Si(111) double crystal monochromator in a transmission mode. The (ox) catalyst was pressed into a self-supporting disk. The (red) catalyst was transferred into the Al cell without contacting air, sealed with Kapton at the both ends of the X-ray path. The XAFS spectra were analyzed by the curve-fitting (CF) method with program REX2000 (Rigaku Co.).

Results and discussion

The catalytic activity for methanol steam reforming on the spinel derived catalyst is higher than that on the nonspinel derived catalyst. To clarify the differences in catalytic activity between two catalysts, the local structure of Cu in the spinel catalyst was characterized.

Figure 1 shows the Cu *K*-edge XANES spectra for CuMn catalysts and the Cu reference (foil). As prepared CuMn catalysts show the Cu²⁺ profile. On the other hand, the Cu⁰ profile was observed for hydrogen reduced catalysts both spinel and non-spinel derived one. From Cu K-edge XANES spectra, there seem no differences between the spinel derived and the non-spinel derived CuMn catalyst.

FTs of k^3 -weighted EXAFS oscillations are shown in Fig. 2. FT profiles of reduced CuMn catalyst show the resemblance in Cu foil. The peak intensity attributed to Cu-Cu coordination for spinel derived catalyst is smaller than that for non-spinel derived one. It seems that the higher catalytic activity is correlating with the smaller Cu metal particle size.



Figure 1. Cu *K*-edge XANES spectra for Cu foil and CuMn catalysts.



Figure 2. FT of Cu *K*-edge EXAFS spectra for Cu foil and CuMn catalysts.

References

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