

Characterization of the Local Structures on Fe-ZSM-5 Zeolites by XAFS Spectroscopy

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Introduction

The Fe-ZSM-5 catalysts have been widely investigated for such significant reactions as the selective catalytic reduction (SCR) reaction of NO, one-step oxidation of benzene to phenol. Recently, it was found that Fe-ZSM-5 exhibits high photocatalytic reactivity for the decomposition of N_2O into N_2 and O_2 . The present study deals with the effect of the different Si/Al ratio on the local structures of the Fe-oxides within Fe-ZSM-5 by means of XAFS (EXAFS and XANES) spectroscopy.

Experimental

The Fe-ZSM-5 samples (1.0 Fe wt%) were prepared by the solid-state reaction of H-ZSM-5 having different Si/Al ratios (20, 34, 950) supplied by Tosoh Co with $FeCl_3 \cdot 6H_2O$. The samples were calcined at 823 K for 9 h in air. XAFS (XANES and EXAFS) spectra were obtained in the fluorescence mode at 295 K at the BL-9A facility of the High Energy Acceleration Research Organization (KEK) in Tsukuba. The EXAFS data were examined by the analysis program Rigaku EXAFS (REX).

Results and Discussion

Figure 1 shows the Fe K-edge XANES and EXAFS spectra of Fe-ZSM-5 (20, 34, 950). The characteristic feature of the XANES spectra of Fe-ZSM-5 is the appearance of a pre-edge peak assigned to a 1s-to-3d-like transition, which is forbidden in pure octahedral symmetry or in

tetrahedral symmetry. It was found that the shape of the XANES spectrum of Fe-ZSM-5 (Si/Al=20) is quite similar to that of the Fe_2O_3 compound, indicating that this sample consists of Fe-oxides with an octahedral coordination. The pre-edge peak intensity is the highest in Fe-ZSM-5 (Si/Al=950), however, it decreases slightly and the shape of the XANES spectra changes when the Si/Al ratio is decreased. From the EXAFS radial structure function of Fe-ZSM-5 (Si/Al=950) in the 3-10.5 \AA^{-1} region, only one peak due to the presence of neighboring oxygen atoms (Fe-O) was observed at ca. 1.0-2.0 \AA (without phase-shift correction), while an additional peak due to the Fe-O-Fe bond was observed at ca. 3.0 \AA with the averaged Fe-oxide clusters between two and three Fe ions in Fe-ZSM-5 (Si/Al=20).

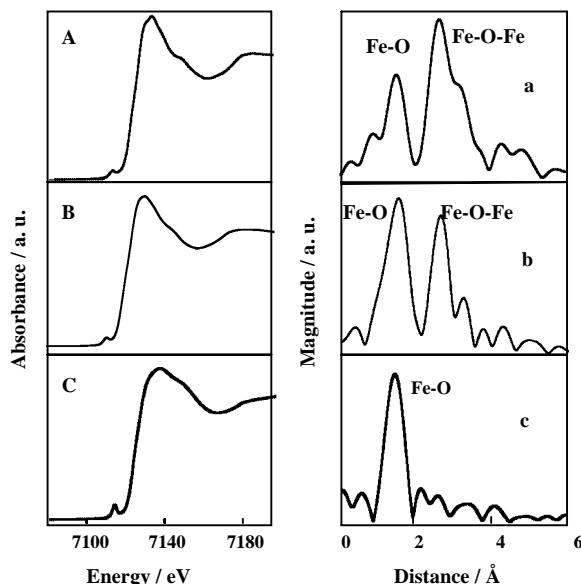


Fig. 1 XANES and EXAFS spectra of Fe-ZSM-5 (1.0 Fe wt%) with various Si/Al ratios of (A, a) 20, (B, b) 34, (C, c) 950 evacuated at 473K.