

Investigation on manganese oxide structures supported on USY zeolite

Hisahiro EINAGA*¹ and Masafumi HARADA²

¹ Department of Energy and Material Sciences, Faculty of Engineering Sciences,
Kyushu University, Kasuga, Fukuoka 816-8580, Japan

² Department of Clothing Environmental Science, Faculty of Human Life and Environment,
Nara Women's University, Nara, 630-8506 Japan

Introduction

Supported manganese oxides have been used for various types of catalytic reactions including catalytic combustion, selective reduction of NO_x, and oxidation of organic compounds. They are also effective for catalytic oxidation of volatile organic compounds with ozone.

It has been reported that the structures and the dispersions of manganese oxides on catalyst support strongly depend on the preparation methods and the kind of catalyst supports. For instance, manganese oxides are highly dispersed on γ -alumina by using manganese acetate as a precursor, while their aggregation easily takes place when Mn nitrate is used as the precursors.

Recently, we have investigated the structures of MnY catalyst, which was prepared by ion-exchange method and have reported that Mn is present in highly dispersed state [1]. In this study, we subsequently investigate the structures of manganese oxides supported on ultra stable Y (USY) zeolite and compared them with those on SiO₂ support, because the catalyst showed high activities for benzene oxidation with ozone at room temperature. Our studies provide us the information on the active sites for catalytic oxidation with ozone.

Experimental

XAFS measurements were carried out on the Photon Factory beam line BL-7C and BL-9A. Mn-loaded ultra stable Y (denoted by Mn/USY) was prepared by a conventional impregnation method from an aqueous solution containing Mn-acetates and USY (Zeolyst, SiO₂/Al₂O₃ = 180). Catalyst samples were dried at 383 K and calcined at 773 K. The catalyst samples were pressed into thin self-supporting wafers with 20 mm ϕ in diameter and set in an in situ cell with polyimide film (Du Pont-Toray Co., Ltd. Kapton 200H), which was connected to the flow-type reaction system. XAFS spectra were recorded at a temperature of 296 K in N₂ flow. Data reduction was carried out by using Rigaku REX-2000.

Results and Discussion

Figure 1 shows the Mn-K edge EXAFS Fourier transforms for the Mn/SiO₂ catalysts at different surface area and Mn loadings. The peak for Mn-O was observed at 1.5 Å for these samples. The intensity of the peaks due to Mn-Mn contribution was observed at 2.5-3.0 Å and their intensity decreased with decreasing the surface area

of the catalysts, indicating that the dispersion of Mn is improved with the increase in catalyst surface area.

Figure 2 shows the Mn-K edge EXAFS Fourier transforms for the Mn/SiO₂ catalysts at loading levels 5-15 wt%. Only the peak for Mn-O was observed for these samples and the peak due to Mn-Mn contribution was much lower in their intensities even at high loadings, implying that Mn was highly dispersed in the USY zeolite catalyst. Thus, USY zeolite is useful for the preparation of Mn loaded catalysts with highly-dispersions.

References

[1] H. Einaga and M. Harada, PF Activity Report 2005, B 27.

* einaga@mm.kyushu-u.ac.jp

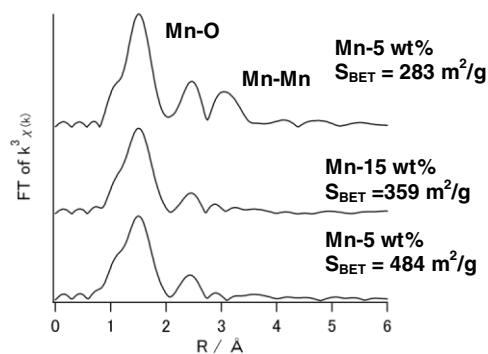


Fig. 1 EXAFS spectra of Mn/SiO₂ catalysts

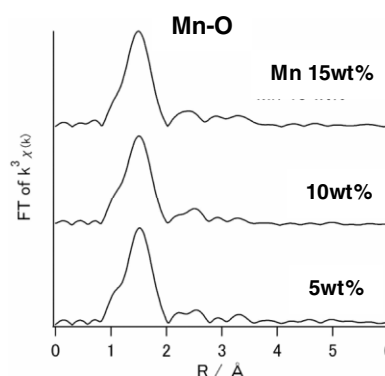


Fig. 2 EXAFS spectra of Mn-USY catalysts