Local Structure Analysis of Mo, Co in Co-Mo Catalysts Supported on Mesoporous Materials

Takeshi KUBOTA, Yasuaki OKAMOTO*
Department of Material Science, Shimane-Univ.,
Matsue, Nishikawatsu, Shimane 690-8504, Japan

Introduction

Recently, regulation of sulfur content in gasoline has become severer to reduce for particulate materials and SO_x in exhaust. For these reasons, development of new catalysts for ultra-deep-Hydrodesufurization (HDS) treatment is strongly needed now. Currently, Co-Mo sulfide catalysts are used for HDS treatment. To improve HDS activity, it is effective to disperse the active species highly on the catalyst supports. Mesoporous materials such as MCM-41 have very large surface area and uniform pore size. So, Mesoporous materials can be expected as a support for highly dispersed metal sulfides. In this study, we investigated the structure of Co sulfides and oxides supported on MCM-41.

Experiments

CoO/MCM-41 catalysts were prepared by an impregnation method using a methanol solution. The sample was calcined at 673 K. The sulfided CoS/MCM-41 was prepared in H_2S/H_2 flow at 673 K. Co K-edge XAFS spectra for Co/MCM-41 were measured at BL-10B in a transmission mode. The synchrotron radiation was monochromatized by a Si(311) monochromator.

Results and Discussion

Fig.1 shows Fourier transform for Co K-edge EXAFS oscillation of Co/MCM-41 and reference compounds. It is obvious that the Co oxides are highly dispersed in CoO/MCM-41 and CoO/Al-MCM-41, because the 2nd shell is not observed differently from Co_3O_4. It is found that an acidity of MCM-41 does not affect the structure of Co oxides. By sulfiding CoO/MCM-41 and CoO/Al-MCM-41, the peaks assigned to Co-S bonding appeared in Fourier transform. The shifts to a shorter distance compared to Co9S8 indicate very little contribution of Co-Co binding. This result indicates the presence of highly dispersed Co sulfides in CoS/MCM-41 and CoS/Al-MCM-41. Fig.2 shows Co K-edge XANES spectra for Co/MCM-41 catalysts and reference compounds. The spectra for Co oxides supported on MCM-41 shows strong white line on absorption edge, but these spectra are different from that for Co_3O_4. Probably, this difference is due to dispersion of Co oxides or oxidation states of Co. On the other hand, the XANES spectra for sulfided Co/MCM-41 were influenced by the presence of acidity (small white line appeared in CoS/Al-MCM-41). It is considered that Co sulfides are oxidized by the acidic sites formed during a loading of Co on Al-MCM-41.

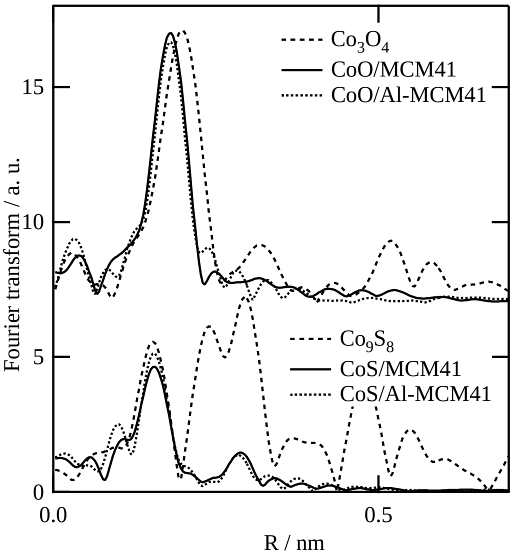

Fig.1 Fourier transform for Co K-edge EXAFS oscillation of Co/MCM-41 and reference compounds.

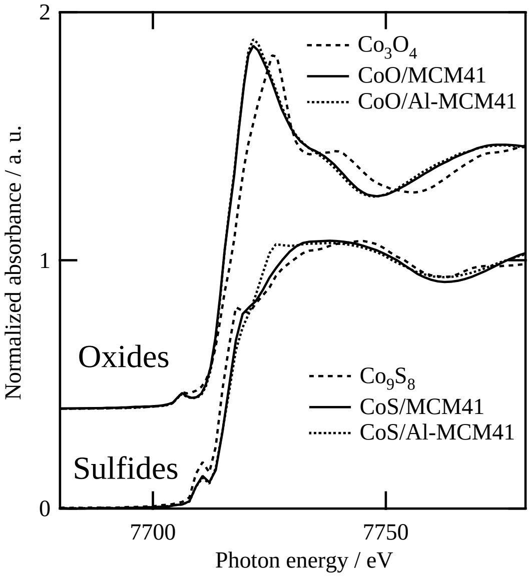

Fig.2 Co K-edge XANES spectra for Co/MCM-41 and reference compounds

*yokamoto@riko.shimane-u.ac.jp