

Local structure of Ta in mesoporous tantalum oxide

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Introduction

The templating using primary amine as a structure directing agent offers a good synthetic method for silicon, titanium, niobium and tantalum oxides with an extremely high surface area. The BET area of mesoporous titania synthesized with dodecylamine exceeds 1200 m² g⁻¹ [1] while that of tantalum oxide synthesized recently by our group was 481 m² g⁻¹. It is reasonably assumed for these solid that most of the atoms are exposed at the surface. Although the mesostructure is characterized by a narrow distribution of mesopores (e.g. $r = 2.5$ nm for mesoporous tantalum), the chemical bond and coordination structure of these bulk mesoporous oxides are likely influenced by the nature of solid surface[2]. In this study we analysed the microstructure of mesoporous tantalum oxide by Ta K edge EXAFS spectroscopy.

Method

Mesoporous tantalum oxide was prepared using octadecylamine and tantalum pentaethoxide. The template amine was extracted with triflic acid.

X-ray absorption spectra of the Ta K edge were recorded on a BL-10B. The powder was diluted in boron nitride. A conventional transmission mode with detection using gas ion chambers was employed. The measurement of each spectrum was completed within 30 min. The spectra were acquired five times under the same measuring conditions and from the extracted spectra, the average $\chi(k)$ was calculated. The data were processed by a REX 2000 (Rigaku Co.) program assembly. The EXAFS oscillation was extracted by fitting a cubic spline function through the post edge region. After normalization using the McMaster tables, the k^3 -weighted EXAFS oscillation, $k^3\chi(k)$, in the 30 – 130 nm⁻¹ region was Fourier transformed into a radial distribution function. The amplitudes and phase-shift functions for Ta-O bond were calculated by FEFF 7.02 code. The amplitude reduction factor (S_0^2) was chosen to be 1 with the lifetime broadening (V_i) equal to 0.

Results and Discussion

Figure 1 shows the EXAFS function and the Fourier transform of Ta K edge spectra of mesoporous tantalum oxide. The characteristic oscillation for the heavy metal oxide at large k region ($k > 9 \text{ \AA}^{-1}$), which is found in the spectrum of conventional powder of Ta₂O₅, is lost in those of mesoporous tantalum oxide. A distinct peak appears at 3.8 Å in the radial distribution function of Ta₂O₅, while it

is hardly observed in those of mesoporous tantalum oxide. These findings demonstrate the loss of microscopic order of the oxide structure. No considerable difference is found between the spectra measured at 20 and 270 K, suggesting that, to the structural disorder, the contribution of thermal term is less important than that of static term.

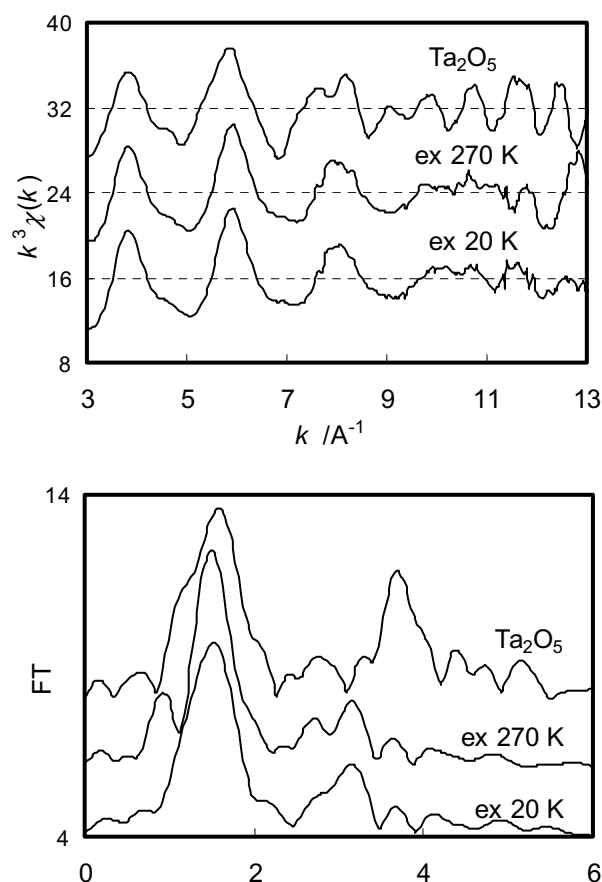


Figure 1 EXAFS oscillation and Fourier transform of Ta K edge spectra of mesoporous tantalum oxide. The measuring temperature is shown.

References

- [1] H. Yoshitake et al., Chem. Mater. 14, 1023 (2002); Stud. Surf. Sci. Catal. 141, 251 (2002).
- [2] H. Yoshitake et al., Phys. Chem. Chem. Phys. 5, 767 (2003).

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