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Elucidation of Charge-discharge Elementary Process of Si-based Negative Electrode Material for Li-ion Secondary Battery

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As a technology for increasing the capacity of lithium-ion batteries (LIB), the development and the study of negative electrodes using Si-based materials is in progress [1,2]. In order to elucidate the mechanism of capacity decrease due to charging and discharging of these Si-based negative electrodes, the XANES spectra of Li K-edge, O K-edge, and Si K-edge are shown for changes in substances in the negative electrode with different charge and discharge depths. It is clarified that the formation behavior of LiF generated by the chemical change occurring at the interface with the electrolytic solution and the formation of lithium silicate and lithium silicide are occurred.

Lithium-ion batteries were constructed using negative electrodes containing SiO, and discharge samples were created after 50% charge, 100% charge, discharge, and 300 cycles. In the soft X-ray XAFS measurement, the XANES spectra of the Li K-edge and the O K-edge were acquired at BL-11 of the SR center, Ritsumeikan University, and the XANES spectra of the Si K-edge were acquired at BL-10.

According to the Li K-edge XANES spectra shown in Figure 1(a), the peak of 61.8 eV, which is thought to be derived from lithium fluoride, is observed to increase, and decrease during initial charge and discharge. In addition, by repeating charging and discharging 300 times of LIB, a large peak of 61.8 eV appears even during discharging. The XANES spectra were also observed on low energy side and high energy side of 61.8 eV during charging, suggesting that lithium oxide, lithium silicate, and lithium silicide were generated.

Figures 1(b) show the Si K-edge XANES spectra related to charging and discharging of LIB using a negative electrode containing SiO. According to these spectra shown, the first edge of 1840.7 eV shifts to the low energy side near 1839.0 eV, which lithium silicate is formed. As charging more, further shifts to the lower energy side are observed. In addition, the initial peak of 1846.7 eV shifted to the 1845.6 eV side due to charging and discharging, and the formation of lithium silicate was confirmed. The formation of lithium silicate was also confirmed in the spectra at the O K-edge, and it is thought that the irreversibility of LIB in this study is increased by repeating the charge/discharge cycle.

These results indicate one of the causes of deterioration associated with the charge/discharge cycle of LIB using a negative electrode containing SiO.

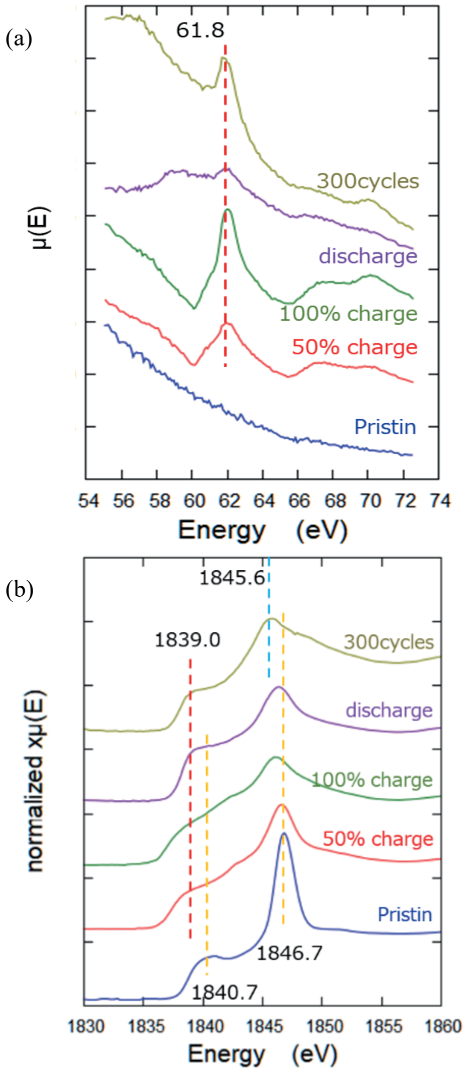

Fig. 1 XANES spectra of SiO anode at Li K-edge (a) and Si K-edge (b).

References

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