

Analysis of lithium ion diffusion in silicon single crystal using X-ray absorption spectroscopy

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Although graphite has been used as a negative electrode material for long time, it is not expected to drastically improve its energy density. On the other hand, silicon is focused as alternative material of graphite. The theoretical energy density of silicon is about 10 times that of graphite. However, degradation during charge / discharge reaction is remarkable, which is estimated to be due to expansion and shrinkage of silicon [1]. As a method of controlling the electrode materials, attempts of nanolization of silicon [2], wire formation [3], plating [4], have been made to relax distortion. For the practical battery using, the further development of performance is required. In this research, we aimed to understand lithium ion diffusion phenomenon in silicon negative electrode, which is extremely important for designing silicon negative electrode. Diffusion behavior of lithium ions into silicon was analyzed by X-ray absorption spectroscopy using silicon single crystal which is the simplest model.

The silicon single crystal pretreated with HF aqueous solution was contacted with the lithium foil in a glove box and held for 1 to 5 days. These samples were put in a transfer vessel in a glove box and brought into the beam line. X-ray absorption measurement was done at BL-10 of Ritsumeikan University SR Center. For the measurement, Si K-edge XAS was measured by the fluorescence mode.

As the reaction time increased, a change in X-ray absorption spectrum was observed. That is, it shows that the spectrum changes due to lithiation of silicon. When the reaction time is 4 days and 5 days, there is hardly any change in the spectrum, indicating that the state of lithiation is sufficiently progressed. On the other hand, it was revealed that at reaction time of 2 days, the reaction of silicon (111) did not progress more, compared with that of silicon (100). In order to quantify these, the amount of reaction was calculated by fitting with two spectra in which the reaction was sufficiently reacted with the initial state. From this result, it is possible to calculate the lithium ion diffusion coefficient in silicon.

References:

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