in-situ X-Ray Imaging Analysis of Concentration Distribution in All-Solid-State Rechargeable Battery Using Ag-ion Conductor

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All-solid-state rechargeable batteries are highly expected as post lithium ion batteries, and development toward practical use is proceeding. In recent years, properties of the electrode active material and solid electrolyte have dramatically improved, but information on the behavior in the composite electrode has not been obtained in order to provide battery performance at the packing level. It has been shown that in a cell of a lithium ion battery using liquid electrolyte, decrease in effective ionic conductivity and salt concentration distribution in the composite electrode occur, which has a great influence on battery performance, forming a reaction distribution of electrodes during charge and discharge [1]. On the other hand, since the transport number of the solid electrolyte is almost one, it is considered that salt concentration distribution does not occur in principle. Therefore, all-solid-state batteries using thick electrodes have the potentiality to combine high energy density and high input / output characteristics. However, as far as the authors know, the dynamic behavior of carrier ions during operation of all-solid-state rechargeable batteries has not been reported. This is because it is extremely difficult to observe behavior of lithium ions which are general carrier ions but are light elements. Therefore, in this research, as a model case of all-solid-state rechargeable batteries, we used silver ion conductor with a large mass. Dynamic distribution of silver ion concentration in solid electrolyte and electrode during charge / discharge was investigated by X-ray imaging method with synchrotron X-ray with high transmittance, spatial resolution and time resolution.

$Ag_6I_4WO_4$ was used as the solid electrolyte, and in the case of the observation in the electrolyte, silver used as both electrodes. The pressed cell component was sliced with the thickness of 300 μm. In the case of the observation in the electrode, $TiTe_2$ was used as the working electrode and silver was used as the counter electrode. X-ray transmission imaging measurements were performed in SPring-8. The used X-ray energy was 33 keV or 65 keV.

Compared with the reference samples of $Ag_{6+x}I_4WO_4$ (x = 0, 0.06, 0.17, 0.31), it was shown that noticeable ion concentration distribution similar to that of the battery using the organic electrolytic solution as the electrolyte during charge / discharge does not occur. On the other hand, dendrite growth in the solid electrolyte due to silver deposition and reaction distribution in the electrode were observed.

References:
[1] H. Yamashige, N. Furuta, M. Mukouyama, K. Kawamura, H. Ota, T. Nonaka and H. Kawaura: ECS Meeting Abstracts, MA2014-02, 461 (2014).