Redox oscillations in 18650-type lithium-ion cell revealed by in-operando Compton scattering imaging

K. Suzuki[1], S. Suzuki[1], Y. Otsuka[1], N. Tsuji[2], K. Jalkanen[3], J. Koskinen[3], K. Hoshi[1], A.-P. Honkanen[4], H. Hafiz[5], Y. Sakurai[2], M. Kanninen[3], S. Huotari[4], A. Bansil[6], H. Sakurai[1], B. Barbiellini[7,6]

[1] Faculty of Science and Technology, Gunma University, Kiryu, Gunma 376-8515, Japan, [2]Japan Synchrotron Radiation Research Institute, SPring-8, Sayo, Hyogo 679-5198, [3] Akkurate Oy, Kaarikatu 8b, 20760 kaarina, Finland, [4] Department of Physics, University of Helsinki, P. O. Box 64, FI-00014 Helsinki, Finland, [5] Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, USA, [6] Department of Physics, Northeastern University, Boston, Massachusetts 02115, USA, [7]Department of Physics, School of Engineering Science, LUT University, FI-53850, Lappeenranta, Finland

Email of communicating author kosuzuki@gunma-u.ac.jp

Compton scattering imaging is a unique technique to visualize lithiation state on electrodes of large-scale lithium-ion batteries in-situ and in-operando conditions. This technique characterized by high-energy synchrotron X-rays allows the non-destructive observation of the reaction in closed electrochemical cells and enables us to analyze quantitatively the concentration of light elements, like lithium since incoherent scattering effects are enhanced. In this study, Compton scattering imaging is applied to an 18650-type cylindrical lithium-ion cell to visualize a spatiotemporal lithiation state, called Turing pattern [1].

The Compton scattering imaging was performed at the high-energy inelastic scattering beamline BL08W of the SPring-8. The energy of the incident X-rays and the scattering angle is fixed at 115.56 keV and 90 degrees, respectively. The Compton scattered X-ray energy spectrum is measured by 9-segments Ge solid-state detector. An observation region of the cell is limited by incident and collimator slits. The size of these slits is 5 µm in height, 750 µm in width, and 500 µm in diameter, respectively. The state of charge of the sample cell was controlled using a potentiostat/galvanostat.

Figure 1 (a) shows the result of line shapes of the Compton scattering spectra, called *S*-parameter analysis [2], which obtained by changing the sample position along z-direction during the charging. By charging the cell, the position of each component of the cell is shifted, which is induced by intercalation/deintercalation of the lithium. Moreover, we observed *S*-parameter oscillations by a depth-resolved analysis of the anode and cathode. Fig. 1 (b) shows the space-time *S*-parameter modulation *S* obtained by subtracting from *S*-parameter its average value in the upper cathode region. A Fourier analysis of *S* shows that the dominating period of the *S*-parameter oscillation corresponds to the timescale of the charging curve and the dominating wavelength of the *S*-parameter oscillation is related to the size of the grains of the active material. The reason for the appearance of this *S*-parameter pattern is due to different mobilities of lithium ions and electrons and non-linear effects in the chemical reaction. Therefore, the existence of the *S*-parameter modulation implies that the cell can have an optimal cycle speed with a more homogeneous flow of ions.

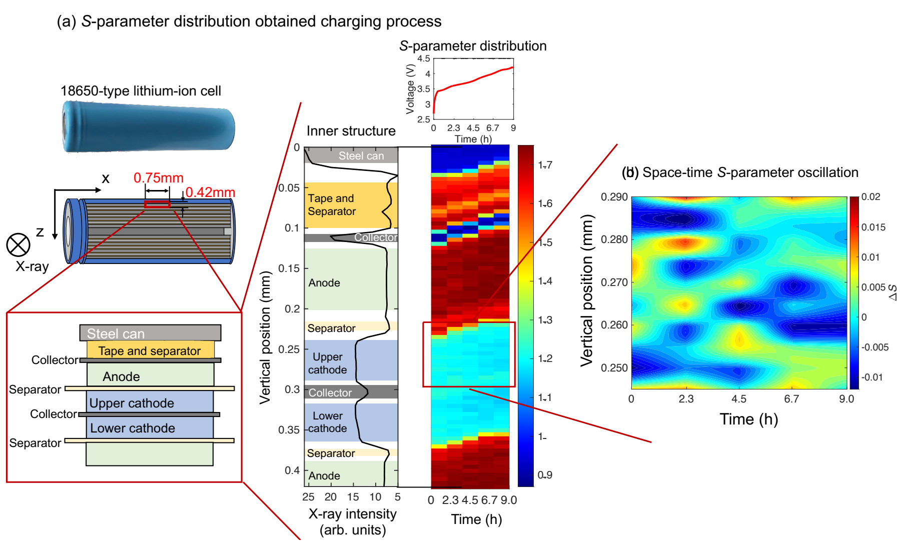

Figure 1. (a) S-parameter distribution obtained charging process. (b) the space-time S-parameter oscillation in the upper cathode.

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