

Article

Non-Destructive Analysis of a High-Power Capacitor Using High-Energy X-ray Compton Scattering

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Abstract: Changes in the internal state of a high-power capacitor during progressive charge–discharge cycling were measured non-destructively using high-energy synchrotron X-ray Compton scattering. The stacked structure of a laminated capacitor was clearly indicated by a Compton scattered X-ray intensity analysis and a line shape (S-parameter) analysis of a Compton scattered X-ray energy spectrum. Moreover, apparent differences in the progress of charge and discharge cycles were observed in the correlation between Compton scattered X-ray intensities and S-parameters obtained from the center and edge positions within the in-plane of the electrode. This difference in the correlation was obtained from the shifting of the stacked structure at the edge position, induced by the drift of the electrolyte material within the capacitor cells.

Keywords: high-power capacitor; non-destructive operando analysis; Compton scattering



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1. Introduction

Electrochemical capacitors (ECs), such as electric double-layer capacitors (EDLCs), super-capacitors, lithium-ion capacitors, and hybrid capacitors play a large role in achieving a sustainable society [1–3]. ECs have characteristics of high power per unit mass of the electrode, high stability for high-speed cycles, and high safety [4–6]. However, their energy density is at least one order of magnitude lower than that of lithium-ion batteries (LIBs) [7,8]. The differences in the characteristics between ECs and LIBs originate from their charge storage mechanisms. In ECs, charge storage is achieved by the formation of an electric double layer at an electrode–electrolyte interface, which consists of electrolyte ions adsorbed onto the electrode surface. In LIBs, charge storage is achieved via intercalation, alloying, and conversion [8]. ECs are used in many applications, including regenerative braking in hybrid and electric vehicles and energy storage systems in other electric devices, robots, and smart grid systems [2]. These applications require the high capacity of the ECs. While increasing the size of devices is the general strategy for increasing their capacity, it is thought that large unit sizes lead to an inhomogeneous ion distribution within the storage cells, which deteriorates the capacity more quickly and affects the structural stability of the cells. This is also a common problem for LIBs [9–11]. So far, observations of inhomogeneity in the ion distribution have only been performed using a disassembly analysis. However, it is difficult to fully understand the mechanism of the inhomogeneous distribution from a disassembly analysis because the distribution can be altered after cell relaxation or as a result of damage related to the disassembly.

Compton scattering technique using high-energy synchrotron X-rays of over 100 keV is a potential candidate method for the observation of large-sized closed electrochemical cells non-destructively. High-energy X-rays possess a high matter-penetrating power that can be used to probe inside an assembled (or commercialized) electrochemical cell without requiring any laboratory-based test cells or disassembly. This is the main advantage of the Compton scattering technique over other in-situ techniques such as nuclear magnetic resonance spectroscopy [3], infrared spectroscopy [3], and small-angle X-ray scattering techniques [12]. Furthermore, experimentally obtained Compton scattered X-ray analysis can reveal non-destructively the type of matter in the capacitor cell because the intensity of the Compton scattered X-rays is largely proportional to the electron density in the probing volume. Recently, we have developed an ion quantification method using the line shape of a Compton scattered X-ray energy spectrum, called S-parameter [13]. So far, the S-parameter analysis has quantitatively revealed the lithiation state of coin-type lithium rechargeable battery [14] and inhomogeneous spatiotemporal lithiation pattern on the cathode in a cylindrical LIB [15]. These advantages of Compton scattering have the potential to allow an operando observation of the ion distribution in large-size capacitors.

In this study, the high-energy synchrotron X-ray Compton scattering technique is used to investigate a large-size capacitor cell and non-destructively observe the changes in the internal state of the cell during charge–discharge cycling. Our technique, which is also applicable to commercialized capacitors, opens a novel pathway for operando analysis of capacitors that should accelerate the development of advanced high-performance capacitors.

2. Samples and Experimental Methods

We used a laminated capacitor cell composed of four alternatively stacked cathode layers and five anode layers, with each layer separated from the next by a polymer film, as shown in Figure 1a. Each cathode layer is composed of an Al current collector (20 μm thickness) that is coated on both sides with activated carbon (100 μm thickness), while each anode layer, also with an Al current collector, is coated with $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (26 μm thickness). The total thicknesses of the cathode and anode layers were 220 μm and 72 μm , respectively. The inside of the cell is filled electrolyte of a solution of 1 mol of LiBF_4 in propylene carbonate (PC). The top view (x/y) of the electrodes (of 100 mm lengths and 80 mm widths) is shown in Figure 1b. Compton scattering measurements were performed at the two positions marked by crosses in Figure 1b. One of these positions is located in the center of the electrode in the x-direction, which is called the center position, and the other is 45 mm away from the center position, which is called the edge position. The y-coordinate of both positions is located 30 mm from the end of the electrode. In the z-direction (i.e., the stacked view), the measurement positions are located between the 2nd and the 3rd cathode layers, as marked with the dashed red rectangle in Figure 1a.

Compton scattering measurements were performed at high-energy inelastic scattering beamline 08W of SPring-8, Japan. The experimental configuration is shown in Figure 2. Incident X-rays emitted from the multipole wiggler were used. The energy of the incident X-rays was monochromatized to 115.56 keV, and the scattering angle was fixed to 90° . The Compton scattered X-ray energy spectrum from the sample cell was measured by nine-elements pure Ge solid-state detectors. The sample cell was placed on a 5-axis movable stage, which allowed the movement of the sample in the x-, y-, and z-directions, as well as with respect to the tilt angle, θ , and azimuthal angles. This flexibility allowed the measurements to be made in specific internal regions within the sample cell. The observation region in the sample cell was determined by the incident and collimator slits, as shown in the enlarged view in Figure 2. The dimensions of the incident and collimator slits were 10 μm in height, 1 mm in width, and 1 mm in diameter. In this study, we ignored the degradation of the sample by X-rays because the Compton scattering cross-section and the size of incident X-rays were very small. The state of charge of the sample cell was adjusted using a potentiostat/galvanostat. The Compton scattered X-ray energy spectra at charged and discharged states were measured after 1, 2, 3, 4, 10, 15, 20, and 25 cycles. One

cycle was performed by first charging the cell using a constant current of 400 mA until the cell was fully charged, and then discharging it using a constant current of 400 mA until fully discharged. All measurements were performed at room temperature.

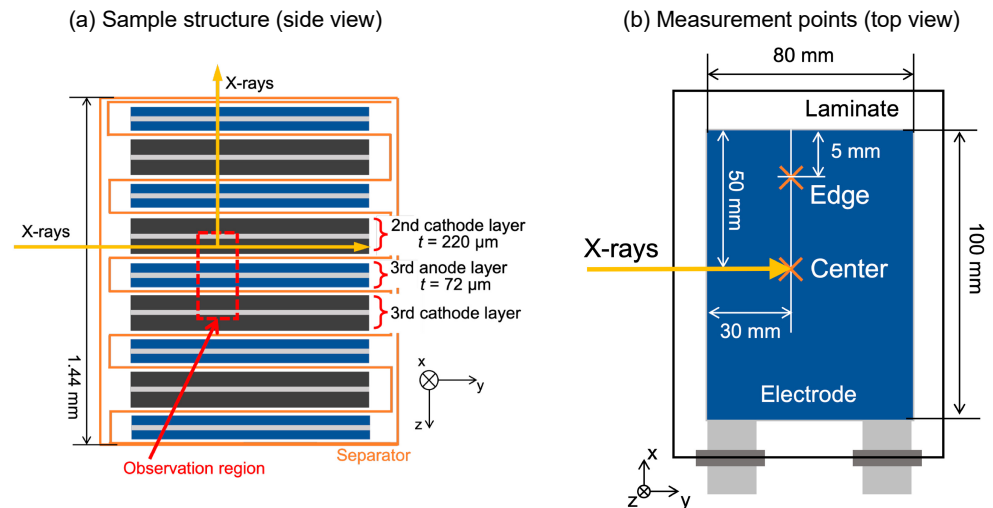


Figure 1. (a) Side view of stacked structure of laminated capacitor cell. Compton scattering X-ray energy spectra are measured within the dashed region using scanning incident X-rays; (b) top view of the electrode in the cell. Compton scattering measurements are performed on two positions marked by orange crosses.

Experimental setup

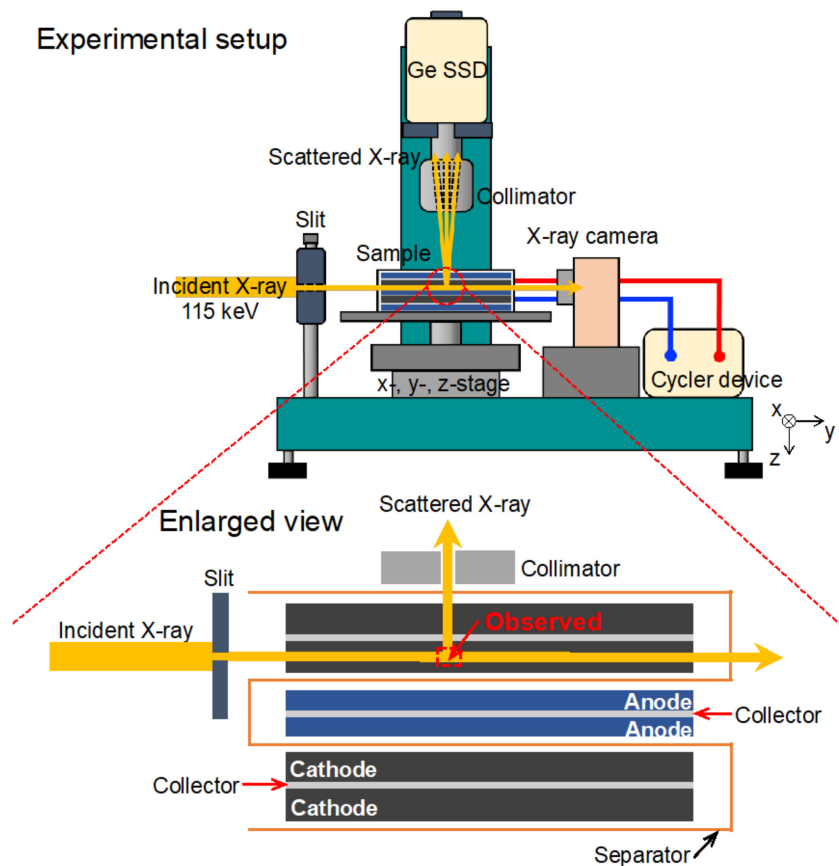


Figure 2. Schematic image of Compton scattering measurement system at BL08W of SPring-8. The enlarged view shows the detail of the observation area inside a capacitor cell.

Measured Compton scattered X-ray energy spectra were analyzed by Compton scattered X-ray intensity and the S-parameter. The Compton scattered X-ray intensity, dN , is described by Equation (1) [16], as follows:

$$dN = \Phi_0 t_1 t_2 \rho_e dV \frac{d\sigma_{KN}}{d\Omega}, \quad (1)$$

where Φ_0 is the photon flux of the incident X-rays; t_1 and t_2 are the path lengths of the incident and scattered X-rays in the sample, respectively; ρ_e is the average electron density in the probing volume; dV is the probing volume; and $d\sigma_{KN}/d\Omega$ is the Klein–Nishina differential cross-section. From Equation (1), when the X-ray path lengths can be treated as constants, the Compton scattered X-ray intensities are proportional to the electron density in the probing volume. Therefore, we can observe the change of the electron density with charge–discharge cycling in the electrodes. Compton scattered X-ray intensity analysis has been used for densitometry [16] and imaging [17]. Analytically, the dN corresponds to the area of the Compton scattered X-ray energy spectrum.

The S-parameter is used to characterize the modification of the line shape of a Compton profile [13]. The Compton profile, $J(p_z)$, is defined by the following equations [18–20]:

$$J(p_z) = \iint \rho(\mathbf{p}) dp_x dp_y, \quad (2)$$

$$\rho(\mathbf{p}) = (2\pi)^{-3/2} \sum_j n_j \left| \int \Psi_j(\mathbf{r}) \exp(-i\mathbf{p} \cdot \mathbf{r}) d\mathbf{r} \right|^2, \quad (3)$$

where $\mathbf{p} = (p_x, p_y, p_z)$ is the electron momentum, $\rho(\mathbf{p})$ is the momentum density distribution, $\Psi_j(\mathbf{r})$ is the wavefunction of an electron in the j -state, and n_j is the electron occupation. From Equation (3), $J(p_z)$ is linked directly to the wavefunction of the electrons. With this connection, the redox orbital studies regarding lithium insertion and extraction in LIB materials were performed by combining the Compton scattering measurement and first-principles calculations [21–26]. The line shape of the Compton profile depends on the distribution of $\rho(\mathbf{p})$, and its distribution is different for each atom. In our previous work, we found that the line shape of the Compton profile was sensitive to changes in right elements, such as Li [13]. Therefore, analyzing the line shapes of the Compton profiles allows us to quantify each element in the material. Analytically, the Compton profile was obtained by converting the x -axis of the Compton scattering X-ray energy spectrum from the X-ray energy to the electron momentum along z -direction, which can be achieved using the following conversion equation:

$$\frac{p_z}{mc} = \frac{E_0 - E_i + (E_i E_0 / mc^2)(1 - \cos \theta)}{\sqrt{E_i^2 + E_0^2 - 2E_i E_0 \cos \theta}}, \quad (4)$$

where mc is $1/\alpha$ and α is the fine structure constant; E_0 and E_1 are the incident and scattered X-ray energies, respectively; mc^2 is the rest mass energy of the electron; and θ is the scattering angle. Here, we introduce the S-parameter to analyze the line shape of the Compton profile. The S-parameter is defined as follows:

$$S = \frac{\int_{-d}^d J(p_z) dp_z}{\int_{-l}^{-d} J(p_z) dp_z + \int_l^d J(p_z) dp_z}, \quad (5)$$

where d and l are two parameters used to distinguish the contributions of lithium-rich and lithium-poor regions, respectively, on a Compton profile. The values of d and l were fixed to 1 atomic unit (a.u.) and 5 a.u., respectively, because the contribution of lithium was localized to $-2 < p_z < 2$ a.u. [27]. Our previous studies on lithium-ion rechargeable batteries have shown that the S-parameter can accurately indicate the lithiation state of

working batteries [14,15]. This study used the S-parameter for the analysis of high-power capacitor cells for the first time.

3. Results and Discussions

Figure 3a shows a Compton scattered X-ray energy spectrum for the corresponding regions inside the cell, including Al collector, activated carbon cathode, anode layers (including $\text{Li}_4\text{Ti}_5\text{O}_{12}$ anode active material and Al collector), and separator, which was obtained by scanning the incident X-rays along the stacking direction of the cell. As the Compton scattered X-ray energy spectrum of the cathode and separator regions shows a narrow distribution as compared with that of the anode and collector regions, the line shape of this energy spectrum is representative of the different materials in the cell. Generally, the inside of a capacitor cell is filled with electrolyte. A fitting analysis was performed on the Compton scattered X-ray energy spectrum obtained from the cathode to evaluate the contribution of the electrolyte, as shown in Figure 3b. Energy spectra of the active material and electrolyte, which were obtained from the pellets of activated carbon and a solution of 1 mol of LiBF_4 in PC, respectively, were used for the fitting analyses. From Figure 3b, the contributions of the active material and electrolyte to the Compton scattered X-ray energy spectrum of the cathode region are 86% and 14%, respectively. The non-destructively obtained Compton scattered X-ray intensity and S-parameter for the observation region marked in Figure 1a are shown in Figure 4a,b. The background shades in Figure 4 correspond to the different cell components, including the Al collector, cathode, separator, and anode. Notably, while the peaks corresponding to the Al collector, cathode, and separator are clearly visible, this study did not obtain enough spatial resolution to distinguish between the active material layer of the anode and Al collector, thereby, resulting in only a single peak for the anode region. Nonetheless, Figure 4 confirms that the Compton scattering technique can facilitate the internal-state analysis of a large-scale capacitor non-destructively.

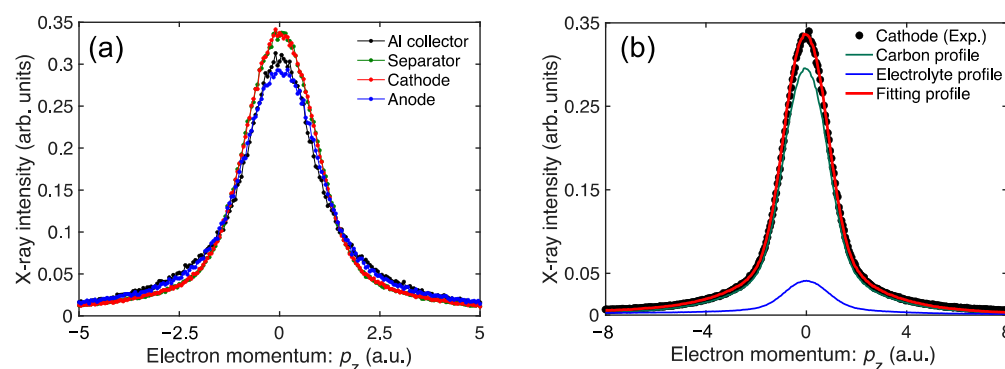


Figure 3. (a) Compton scattered X-ray energy spectrum obtained from Al collector, separator, activated carbon cathode and anode layer; (b) fitting analysis of the Compton scattered X-ray energy spectrum.

Compton scattered X-ray energy spectrum of the cathode was then measured at charged and discharged states after 1, 2, 3, 4, 10, 15, 20, and 25 cycles. Figure 5 shows the modification of the S-parameter at the center and edge positions at each charged and discharged state. The S-parameters are normalized by the S-parameter value of the initial (discharged) state, after averaging in S-parameters obtained from the cathode region which are highlighted by the dark gray background color in Figure 4. Evidently, the S-parameter values decrease as the capacitor cell is charged, and then increase to the value of the initial state as the capacitor cell is discharged. The S-parameter changes by about 1% with each charge and discharge, indicating that lithium moves between the cathode and the anode during each cycle, as expected. To investigate the structural changes inside the capacitor cell resulting from the cycling, we plotted and analyzed the correlation between the rate of change in the X-ray intensity and the rate of change in the S-parameter at the

center and edge positions, as shown in Figure 6. The plot of the initial state corresponds to the origin in Figure 6. The correlation at the center position appears to be relatively concentrated, with the distribution in the Compton scattered X-ray intensity ranging from -5% to 5% and that in the S-parameter from -1.5% to 0% . Interestingly, the correlation at the edge position shows a different distribution as compared with that of the center position. Furthermore, the Compton scattered X-ray intensities show an especially large modification with the progress of the charge–discharge cycles. The change in the correlation between the Compton scattered X-ray intensities and the S-parameters with progressive cycling has also been shown in our lithium rechargeable battery studies, where the correlation has shown a clear difference by degrading the battery [28]. Therefore, the changes observed in the correlation shown in Figure 6b indicate the presence of capacitor cell degradation. Figure 7 shows the amount of movement in the cathode Al collector position until four cycles. The amount of movement of the cathode Al collector position from 10 cycles to 25 cycles could not be measured because we did not have enough time. Although the Al collector position shifts with the cycles in both the center and edge positions, the amount of movement in the center position is relatively small as compared with the large movement detected in the edge position. Since the Compton scattered X-ray intensity reflects the electron density at the probing volume, as shown in Equation (1), and because the capacitor cell is filled with an electrolyte, the change in the Compton scattered X-ray intensities at the edge position implies that the drift observed is due to the movement of the electrolyte. Therefore, the drift of the electrolyte material caused by the repeated use of the capacitor cell is likely to influence the ion distribution in all subsequent charge–discharge cycles.

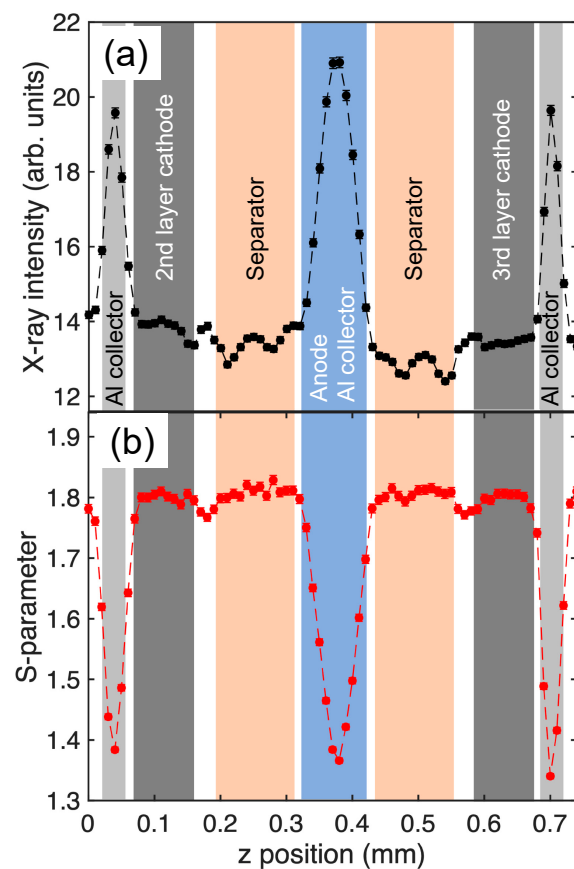


Figure 4. Schematic view of (a) Compton scattered X-ray spectrum; (b) S-parameter obtained non-destructively from inside the structure of the capacitor cell. Background shades correspond to the regions of the component of the capacitor cell.

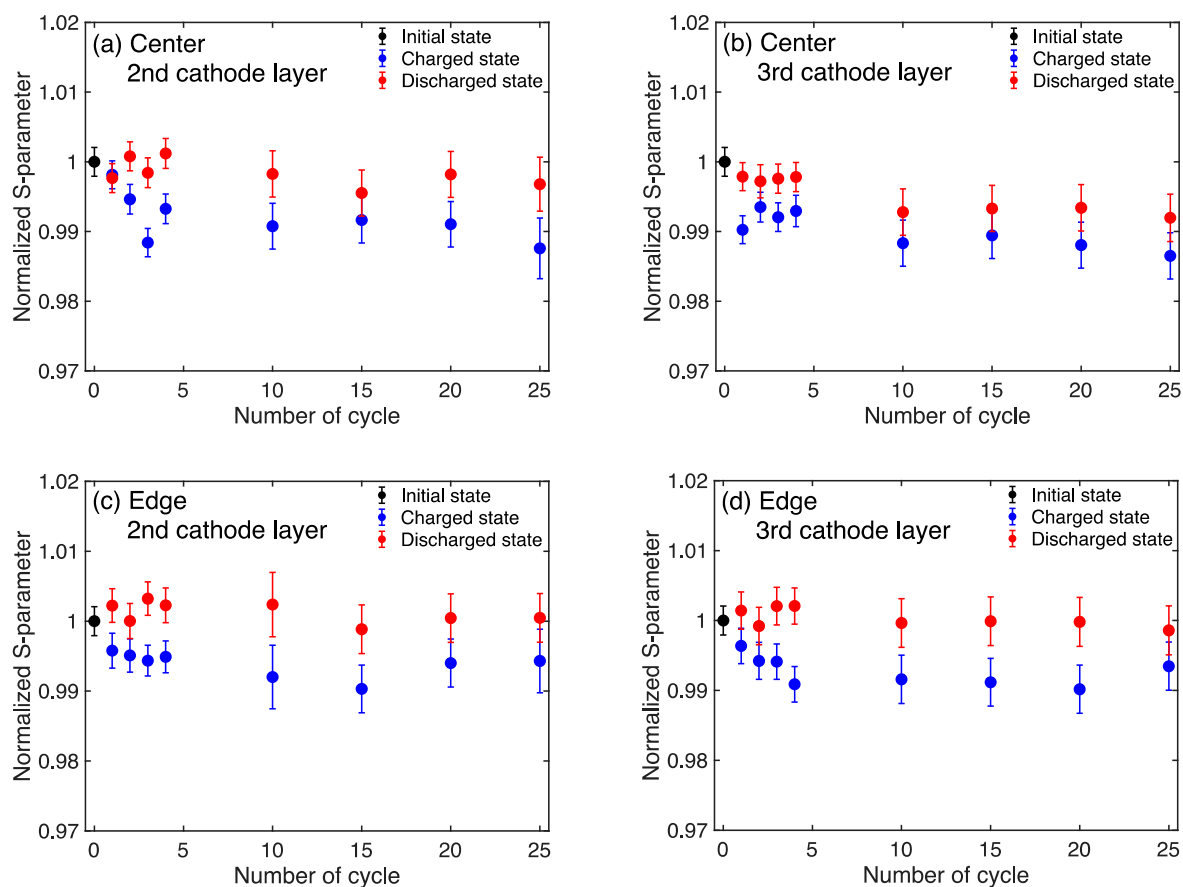


Figure 5. (a–d) Variation of the S-parameter with charge–discharge cycles obtained at the 2nd (left graphs) and 3rd (right graphs) layer cathodes for the center (top graphs) and edge (bottom graphs) positions. The initial state corresponds to a discharged state.

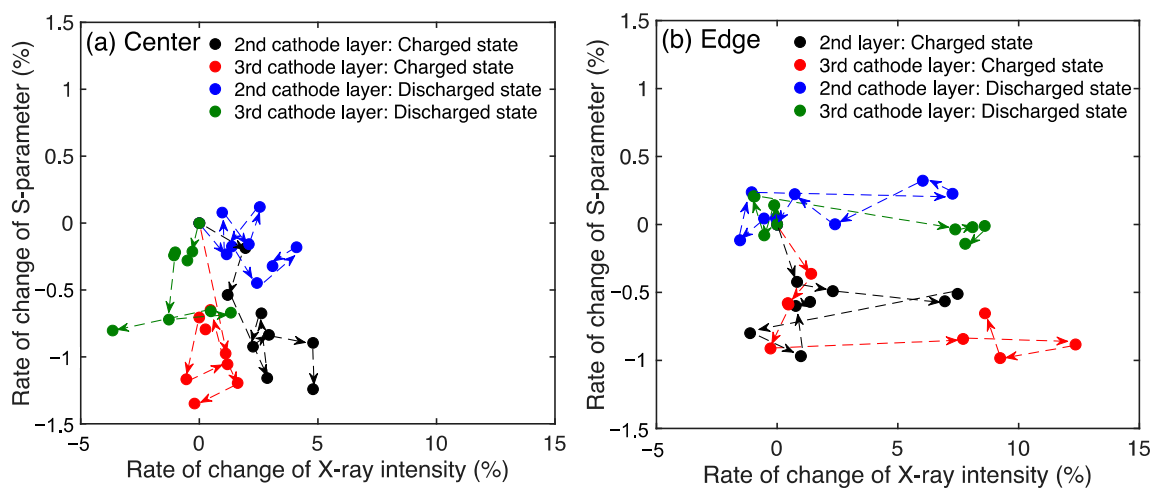


Figure 6. Correlation between the rate of change of the Compton scattered X-ray intensities and S-parameters at (a) the center position and (b) the edge position.

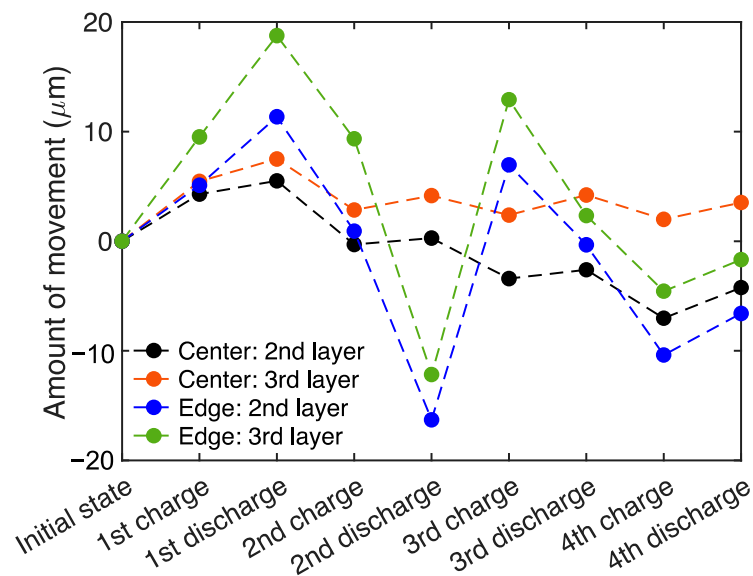


Figure 7. Amount of movement of cathode Al collector position corresponding to each cycle. The positive value of the y -axis corresponds to the z -direction shown in the coordinate in Figure 1a.

4. Conclusions

In this study, high-energy synchrotron X-ray Compton scattering techniques were used for a non-destructive investigation of a high-power capacitor cell during charge–discharge cycling. The Compton scattered X-ray intensity and the S-parameter clearly show the stacked internal structure of the capacitor cell. The S-parameter obtained at the cathode shows a variation corresponding to the charged and discharged states. Furthermore, the correlation between the rate of change of Compton scattered X-ray intensities and S-parameters with the progressive cycling differs between the center and the edge positions. It is thought that the shift in the stacked structure is induced by the drift of the electrolyte. Therefore, we have shown that Compton scattered X-ray intensity and S-parameter analysis allow monitoring of the lithiation of the capacitor cell.

Author Contributions: Conceptualization: Y.S., S.I. and H.S.; Methodology: N.T. and Y.S.; Software: K.S., K.T., S.S., T.A. and K.H.; Validation: all authors; Formal analysis: K.S., K.T., S.S., H.H., T.A., K.H., Y.M., S.I. and H.S.; Investigation: K.S. and H.H.; Resources: S.I., Y.S. and H.S.; Data curation: K.S., K.T., S.S., T.A., K.H. and H.S.; Writing—original draft preparation: K.S. and K.T.; Writing—review and editing: all authors; Visualization: K.S., K.T., S.S., T.A., K.H. and H.S.; Supervision: S.I., Y.S. and H.S.; Project administration: H.S.; Funding acquisition: K.S. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

References

1. Naoi, K. 'Nanohybrid Capacitor': The Next Generation Electrochemical Capacitors. *Fuel Cells* **2010**, *5*, 825–833. [\[CrossRef\]](#)
2. Wang, Y.; Wang, L.; Li, M.; Chen, Z. A review of key issues for control and management in battery and ultra-capacitor hybrid energy storage system. *eTransportation* **2020**, *4*, 100064. [\[CrossRef\]](#)
3. Pal, B.; Yasin, A.; Kaur, R.; Tebyetekerwa, M.; Zabihi, F.; Yang, S.; Yang, C.-C.; Sofer, Z.; Jose, R. Understanding electrochemical capacitors with in-situ techniques. *Renew. Sustain. Energy Rev.* **2021**, *149*, 111418. [\[CrossRef\]](#)
4. Pal, B.; Krishnan, S.G.; Vijayan, B.L.; Harilal, M.; Yang, C.-C.; Ezema, F.I.; Yusoff, M.M.; Jose, R. In situ encapsulation of tin oxide and cobalt oxide composite in porous carbon for high-performance energy storage application. *J. Electro. Chem.* **2018**, *817*, 217–225. [\[CrossRef\]](#)
5. Pal, B.; Yasin, A.; Kunwar, R.; Yang, S.; Yusoff, M.M.; Jose, R. Polymer versus Cation of Gel Polymer Electrolytes in the Charge Storage of Asymmetric Supercapacitors. *Ind. Eng. Chem. Res.* **2019**, *58*, 654–664. [\[CrossRef\]](#)
6. Seongki, A.; Haniu, Y.; Nara, H.; Momma, T.; Sugimoto, W.; Osaka, T. Synthesis of Stacked Graphene-Sn Composite as a High-Performance Anode for Lithium-Ion Capacitors. *J. Electrochem. Soc.* **2020**, *167*, 040519.
7. Bruce, P.G.; Freunberger, S.A.; Hardwick, L.J.; Tarascon, J.-M. Li-O₂ and Li-S batteries with high energy storage. *Nat. Mater.* **2012**, *11*, 19–29. [\[CrossRef\]](#)
8. Reddy, M.V.; Rao, G.V.S.; Chowdari, B.V.R. Metal Oxides and Oxyalts as Anode Materials for Li Ion Batteries. *Chem. Rev.* **2013**, *113*, 5364–5457. [\[CrossRef\]](#)
9. Suzuki, K.; Kanai, R.; Tsuji, N.; Yamashige, H.; Orikasa, Y.; Uchimoto, Y.; Sakurai, Y.; Sakurai, H. Dependency of the Charge-Discharge Rate on Lithium Reaction Distributions for Commercial Lithium Coin Cell Visualized by Compton Scattering Imaging. *Condens. Matter.* **2018**, *3*, 27. [\[CrossRef\]](#)
10. Kimura, Y.; Tomura, A.; Fakkao, M.; Nakamura, T.; Ishiguro, N.; Sekizawa, O.; Nitta, K.; Uruga, T.; Okumura, T.; Tada, M.; et al. 3D Operando Imaging and Quantification of Inhomogeneous Electrochemical Reactions in Composite Battery Electrodes. *J. Phys. Chem. Lett.* **2020**, *11*, 3629–3636. [\[CrossRef\]](#)
11. Orikasa, Y.; Yamamoto, K.; Shimizu, T.; Uchimoto, Y. Multiscale and hierarchical reaction mechanism in a lithium-ion battery. *Chem. Phys. Rev.* **2022**, *3*, 011305. [\[CrossRef\]](#)
12. Prehal, C.; Koczwar, C.; Jäckel, N.; Amenitsch, H.; Presser, V.; Paris, O. A carbon nanopore model to quantify structure and kinetics of ion electrosorption with in situ small-angle X-ray scattering, *Phys. Chem. Chem. Phys.* **2017**, *19*, 15549–15561. [\[CrossRef\]](#) [\[PubMed\]](#)
13. Suzuki, K.; Barbiellini, B.; Orikasa, Y.; Kaprzk, S.; Itou, M.; Yamamoto, K.; Wang, Y.J.; Hafiz, H.; Uchimoto, Y.; Bansil, A.; et al. Non-destructive measurement of in-operando lithium concentration in batteries via X-ray Compton scattering. *J. Appl. Phys.* **2016**, *119*, 025103. [\[CrossRef\]](#)
14. Suzuki, K.; Suzuki, A.; Ishikawa, T.; Itou, M.; Yamashige, H.; Orikasa, Y.; Uchimoto, Y.; Sakurai, Y.; Sakurai, H. In operando quantitation of Li concentration for a commercial Li-ion rechargeable battery using high-energy X-ray Compton scattering. *J. Synchrotron Rad.* **2017**, *24*, 1006–1011. [\[CrossRef\]](#) [\[PubMed\]](#)
15. Suzuki, K.; Suzuki, S.; Otauka, Y.; Tsuji, N.; Jalkanen, K.; Koskinen, J.; Hoshi, K.; Honkanen, A.-P.; Hafiz, H.; Sakurai, Y.; et al. Redox oscillations in 18650-type lithium-ion cell revealed by in operando Compton scattering imaging. *Appl. Phys. Lett.* **2021**, *118*, 161902. [\[CrossRef\]](#)
16. Sharaf, J.M. Practical aspects of Compton scatter densitometry. *Appl. Radiat. Isotop.* **2001**, *54*, 801–809. [\[CrossRef\]](#)
17. Harding, G.; Harding, E. Compton scatter imaging: A tool for historical exploration. *Appl. Radiat. Isotop.* **2010**, *68*, 993–1005. [\[CrossRef\]](#)
18. Cooper, M.J.; Mijnen, P.E.; Shiotani, N.; Sakai, N.; Bansil, A. The theory of Compton scattering. In *X-ray Compton Scattering*; Oxford Science Publications: London, UK, 2004; Chapter 2, pp. 22–69.
19. Barbiellini, B. A natural orbital method for the electron momentum distribution in matter. *J. Phys. Chem. Solids.* **2000**, *61*, 341–344. [\[CrossRef\]](#)
20. Barbiellini, B.; Bansil, A. Treatment of correlation effects in electron momentum density: Density functional theory and beyond. *J. Phys. Chem. Solids.* **2001**, *62*, 2181–2189. [\[CrossRef\]](#)
21. Suzuki, K.; Barbiellini, B.; Orikasa, Y.; Go, N.; Sakurai, H.; Kaprzyk, S.; Itou, M.; Yamamoto, K.; Uchimoto, Y.; Wang, Y.J.; et al. Extracting the Redox Orbitals in Li Battery Materials with High-resolution X-ray Compton Scattering Spectroscopy. *Phys. Rev. Lett.* **2015**, *114*, 087401. [\[CrossRef\]](#)
22. Barbiellini, B.; Suzuki, K.; Orikasa, Y.; Kaprzyk, S.; Itou, M.; Yamamoto, K.; Wang, Y.J.; Hafiz, H.; Yamada, R.; Uchimoto, Y.; et al. Identifying a descriptor for d-orbital delocalization in cathodes of Li batteries based on x-ray Compton scattering. *Appl. Phys. Lett.* **2016**, *109*, 073102. [\[CrossRef\]](#)
23. Hafiz, H.; Suzuki, K.; Barbiellini, B.; Orikasa, Y.; Callewaert, V.; Kaprzyk, S.; Itou, M.; Yamamoto, K.; Yamada, R.; Uchimoto, Y.; et al. Visualizing redox orbitals and their potentials in advanced lithium-ion battery materials using high-resolution X-ray Compton scattering. *Sci. Adv.* **2017**, *3*, e1700971. [\[CrossRef\]](#) [\[PubMed\]](#)
24. Hafiz, H.; Suzuki, K.; Barbiellini, B.; Orikasa, Y.; Kaprzyk, S.; Tsuji, N.; Yamamoto, K.; Terasaka, A.; Hoshi, K.; Uchimoto, Y.; et al. Identification of ferrimagnetic orbitals preventing spinel degradation by charge ordering in Li_xMn₂O₄. *Phys. Rev. B* **2019**, *100*, 205104. [\[CrossRef\]](#)

-
25. Hafiz, H.; Suzuki, K.; Barbiellini, B.; Tsuji, N.; Yabuuchi, N.; Yamamoto, K.; Orikasa, Y.; Uchimoto, Y.; Sakurai, Y.; Sakurai, H.; et al. Tomographic reconstruction of oxygen orbitals in lithium-rich battery materials. *Nature* **2021**, *594*, 213–216. [[CrossRef](#)] [[PubMed](#)]
 26. Suzuki, K.; Otsuka, Y.; Hoshi, K.; Sakurai, H.; Tsuji, N.; Yamamoto, K.; Yabuuchi, N.; Hafiz, H.; Orikasa, Y.; Uchimoto, Y.; et al. Magnetic Compton Scattering Study of Li-Rich Battery Materials. *Condens. Matter.* **2022**, *7*, 4. [[CrossRef](#)]
 27. Biggs, F.; Mendelson, L.B.; Mann, J.B. Hartree-Fock Compton profiles for the elements. *At. Data Nucl. Data Tables* **1975**, *16*, 201. [[CrossRef](#)]
 28. Suzuki, K.; Otsuka, Y.; Tsuji, N.; Hoshi, K.; Sakurai, Y.; Sakurai, H. Identifying the Degradation Mechanism in Commercial Lithium Rechargeable Batteries via High-Energy X-ray Compton Scattering Imaging. *Appl. Sci.* **2020**, *10*, 5855. [[CrossRef](#)]