Supplementary Material

Kosuke Suzuki [1,*], Ari-Pekka Honkanen [2], Naruki Tsuji [3], Kirsi Jalkanen [4], Jari Koskinen [4], Hideyuki Morimoto [1], Daisuke Hiramoto [1], Ayumu Terasaka [1], Hasnain Hafiz [5,6], Yoshiharu Sakurai [3], Mika Kanninen [4], Simo Huotari [2], Arun Bansil [6], Hiroshi Sakurai [1] and Bernardo Barbiellini [6,7]

[1] Faculty of Science and Technology, Gunma University, Kiryu, Gunma 376-8515, Japan
[2] Department of Physics, University of Helsinki, P.O. Box 64, FI-00014 Helsinki, Finland
[3] Japan Synchrotron Radiation Research Institute, SPring-8, Sayo, Hyogo 679-5198, Japan
[4] Akkurate Oy, Kaarikatu 8b, 20760 Kaarina, Finland
[5] Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA
[6] Department of Physics, Northeastern University, Boston, MA 02115, USA
[7] Department of Physics, School of Engineering Science, Lappeenranta University of Technology (LUT University), FI-53851 Lappeenranta, Finland
* Correspondence: kosuzuki@gunma-u.ac.jp; Tel.: +81-277-30-1714

Cycle Performance of 18650-Type Cell

Figure S1(a) shows the performance of the 18650-type cell as a function of the cycle number. The discharge capacity is seen to reduce by about 29% with cycling. Figures S1(b) and (c) show the impedance spectrum of the fresh and aged cells, respectively. A large semi-circular shape is observed in the low-frequency region in the aged cell.

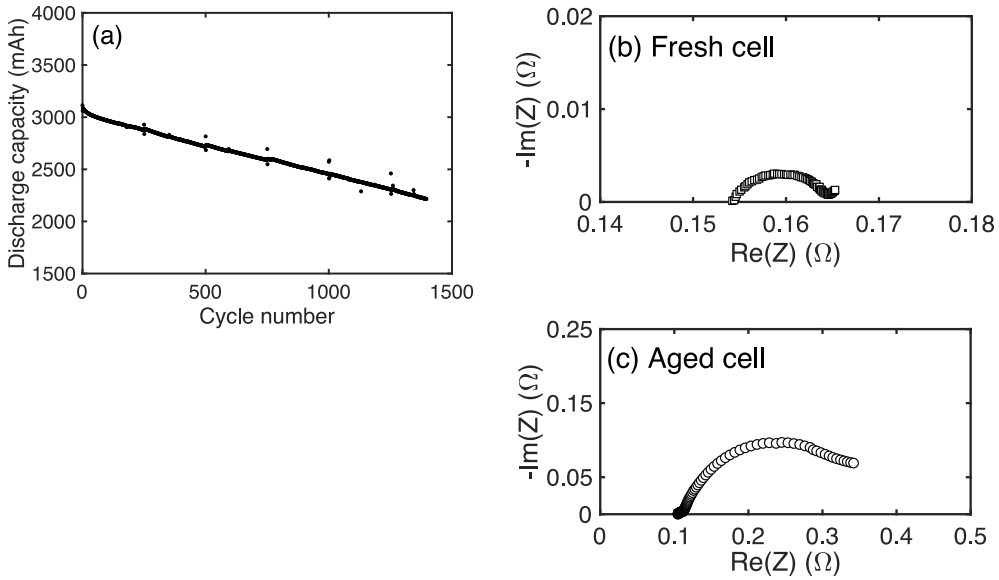

Figure S1. (a) Cycling performance of the 18650-type cell. (b) Impedance spectrum of the fresh cell. (c) Impedance spectrum of the aged cell.

Data Processing

Figure S2 shows raw Compton scattered X-ray energy spectra obtained by using nine Ge solid-state detectors. This energy spectrum was obtained from the graphite anode of the fresh cell in the discharged state. The conversion from the X-ray energy in figure S2(a) to the electron momentum (p_z) in Figure S2(b) was carried out by using the relationship [S1],

$$p_z = mc\frac{E_2 - E_1 + \left(E_1E_2/mc^2\right)\left(1-\cos\theta\right)}{\sqrt{E_1^2 + E_2^2 - 2E_1E_2\cos\theta}}, \quad \text{(S1)}$$

where m is electron mass, and c is the speed of light. E_1 and E_2 are energies of the incident and Compton scattered X-rays, respectively. θ is the scattering angle. The area under the low momentum region (H) and that under the high momentum region (W) was obtained for the various spectra, and the results are summarized in Table S1. (For this purpose, the low-energy cut-off d was set at 1 a.u., while the high-energy cut-off l was set at 5 a.u.) The overall S-parameter was computed from the values of H and W obtained by adding the values of H and W for the 9 detectors, which yields the S-parameter value of 1.807 ± 0.006.

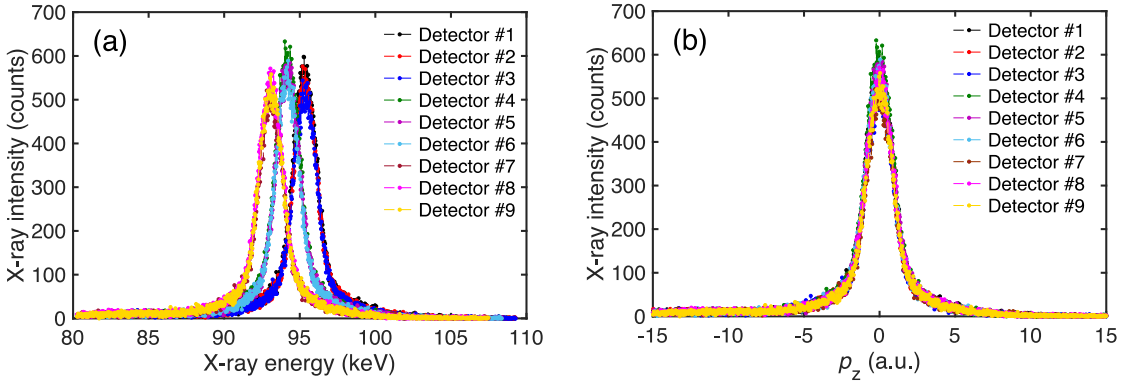

Figure S2. Raw Compton scattered X-ray energy spectra obtained from the graphite anode in the fresh cell. The horizontal scale in figure S2(b) is atomic momentum unit (a.u.). These energy spectra were obtained in 60 seconds.

Table S1. The areas under the low- (H) and high-momentum (W) regions of the 9 separate Compton spectra as discussed in the text.

	Det. #1	Det. #2	Det. #3	Det. #4	Det. #5	Det. #6	Det. #7	Det. #8	Det. #9	Sum
H	32377.53	30530.68	29100.24	34503.53	32724.05	31506.31	29260.84	31642.25	30586.71	282232.1
W	17909.53	16754.14	16284.96	18999.62	18063.54	17425.24	16545.89	17620.59	16558.5	156162.0

Figure S3 shows the Compton scattered X-ray energy spectrum obtained from the charged (I_C) and discharged (I_D) states of the cathode of the fresh cell. In order to compare the line-shapes of these spectra, the area under each energy spectrum was first normalized to the same value. The difference spectrum ($\Delta I = I_D - I_C$) then obtained is shown in Figure S3 as the blue line connecting the blue dots. This difference spectrum is seen to exhibit a line-shape, which is similar to that of the Compton profile of lithium atom (brown line) [S2]. The area under the difference Compton spectrum corresponds to the amount of lithium which moves to the cathode upon discharging the cell.

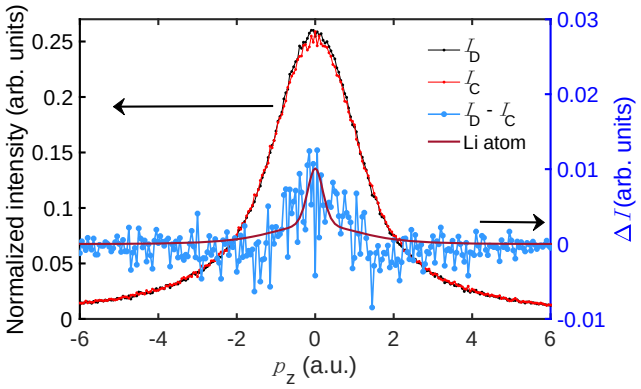

Figure S3. Normalized Compton scattered X-ray energy spectra obtained from the charged (red line) and discharged (black line) NMC cathode in the fresh cell. Blue line is the difference Compton spectrum. The brown line gives the Compton profile of lithium atom.

References

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[S1] Zukowski, E. The processing of experimental data. In *X-Ray Compton Scattering*, 1st ed.; Cooper, M.J., Mijnarends, P.E., Shiotani, N., Sakai, N., Bansil, A., Eds.; Oxford University Press: Oxford, UK, 2004; pp. 134–138.

[S2] Biggs, F.; Mendelson, L.B.; Mann, J.B. Hartree-Fock Compton profiles for the elements. *At. Data Nucl. Data Tables* **1975**, *16*, 201.