## Detection of Unbalanced Voltage Cells in Series-connected Lithium-ion Batteries Using Single-frequency Electrochemical Impedance Spectroscopy

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Fig. S1. Nyquist plots from the graphite/NCA lithium-ion cells with SOCs set to be SOC10\%, SOC20\%, SOC40\%, SOC60\%, and SOC80\%, respectively. (a) Nyquist plots in the frequency region from 10 kHz to 0.01 Hz and (b) a magnification.


Fig. S2. (a) Nyquist plots obtained from four individual cells (cell A, cell B, cell C, and cell D) with SOCs adjusted to be SOC20\%. (b) Nyquist plots of the sums of the impedances obtained from the results of (a) (green) and shifted $+60 \mathrm{~m} \Omega$ toward the real axis (yellow). The resistance of $60 \mathrm{~m} \Omega$ is an additional wiring resistance to connect cell A , cell B , cell C , and cell $D$ to form four-series connected cells for impedance measurement. Nyquist plots of the total impedance measured using ModuLab (blue) are also shown in (b). (c) Residual errors of $Z, Z^{\prime}$, and $Z^{\prime \prime}$ between the yellow and blue Nyquist plots. The Nyquist plots of the sums of the impedances obtained from the results of (a) (green) have a similar shape to those of the total impedance measured using ModuLab (blue) but they do not match. This may be because of the different wiring resistances of the measuring equipment for the individual cells and the series-connected cells. When the green Nyquist plots shift to $+60 \mathrm{~m} \Omega$ toward the real axis (yellow), they correspond to the total impedance measured using ModuLab (blue). This indicates that $60 \mathrm{~m} \Omega$ corresponds to the outer wiring resistance to connect four-series batteries and the total impedance of series-connected cells corresponds to the sum of the impedances from individual cells. The residual errors of $Z, Z^{\prime}$, and $Z^{\prime \prime}$ in the frequency range of 100 Hz to 0.1 Hz are less than $1 \%$.


Fig. S3. (a) Calculated value of the total impedance $Z^{\prime \prime}$ of series-connected cells from individual cells as a function of cell numbers. Error bar in the figure is the dispersion of impedance $Z^{\prime \prime}( \pm 3 \sigma)$. (b) Expand Fig. S3 (a) inset to show small number of series-connected cells. (c) Dispersion of impedance $Z^{\prime \prime}( \pm 3 \sigma)$ obtained from the result measured 10 individual battery cells in the same product lot.

Table S1. Conditions of total cell numbers in series and their constituents of individual cell SOC for the calculation of impedance $Z$ " for Fig. 3S. Total cell number is varied from 4 to 100 with the same ratio of the constituents of individual cells shown in Table 1 (Group C); No. 1 is composed of SOC $10 \%$ : SOC $20 \%$ : SOC $30 \%=0: 4: 0$, No. 2 is composed of is SOC $10 \%: \operatorname{SOC} 20 \%: \operatorname{SOC} 30 \%=1: 2: 1$, and No. 3 is composed of $\operatorname{SOC} 10 \%: \operatorname{SOC} 20 \%: \operatorname{SOC} 30 \%=2: 1: 1$ regardless of the total cell number in series.

| No. | Total cell number in series | Constituents (cell numbers) |  |  | Total voltage [V] |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | SOC10\% | SOC20\% | SOC30\% |  |
| 1 | 4 | 0 | 4 | 0 | 13.9 |
| 2 | 4 | 1 | 2 | 1 | 13.8 |
| 3 | 4 | 2 | 1 | 1 | 13.8 |
| 1 | 8 | 0 | 8 | 0 | 27.8 |
| 2 | 8 | 2 | 4 | 2 | 27.6 |
| 3 | 8 | 4 | 2 | 2 | 27.6 |
| 1 | 16 | 0 | 16 | 0 | 55.6 |
| 2 | 16 | 4 | 8 | 4 | 55.2 |
| 3 | 16 | 8 | 4 | 4 | 55.2 |
| 1 | 32 | 0 | 32 | 0 | 111.2 |
| 2 | 32 | 8 | 16 | 8 | 110.4 |
| 3 | 32 | 16 | 8 | 8 | 110.4 |
| 1 | 64 | 0 | 64 | 0 | 222.4 |
| 2 | 64 | 16 | 32 | 16 | 220.8 |
| 3 | 64 | 32 | 16 | 16 | 220.8 |
| 1 | 100 | 0 | 100 | 0 | 347.5 |
| 2 | 100 | 25 | 50 | 25 | 345.0 |
| 3 | 100 | 50 | 25 | 25 | 345.0 |

