

**Supplementary Information**

**Reaction Uniformity Visualized by Raman Imaging in  
Composite Electrode Layers of All-Solid-State Lithium Batteries**

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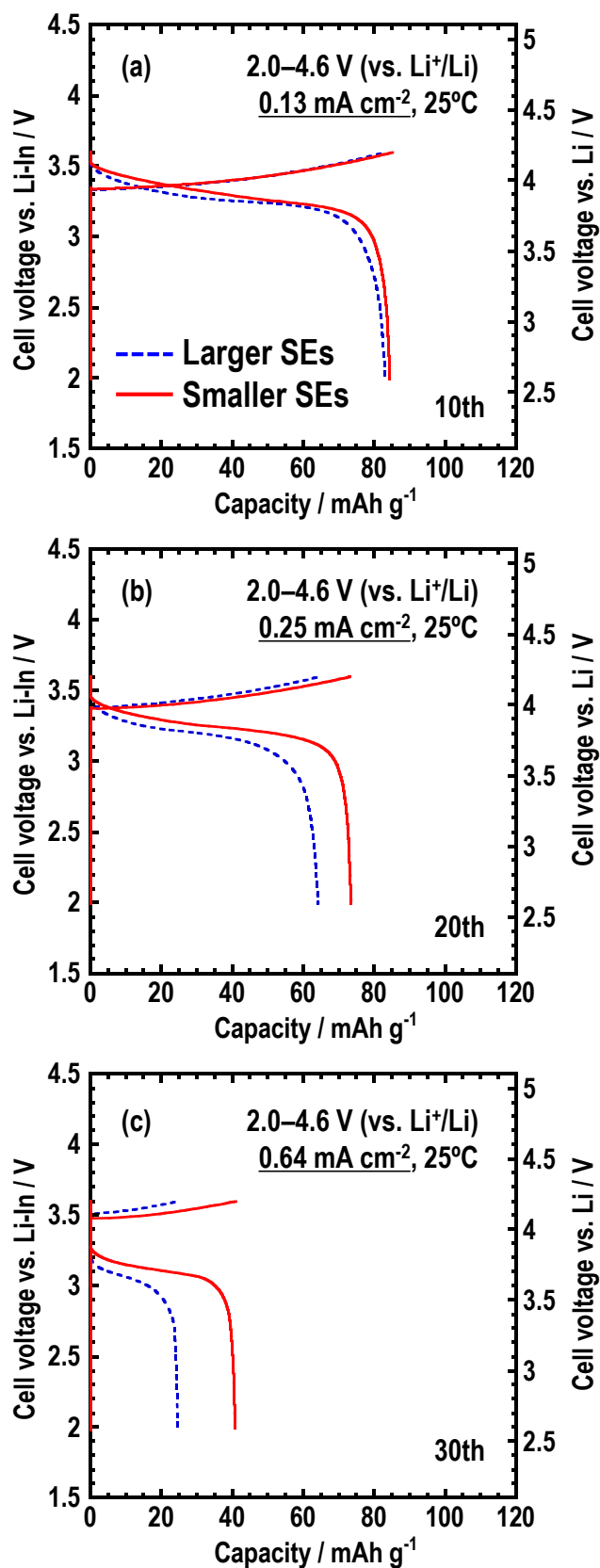


Fig. S1 (a) 10th, (b) 20th and (c) 30th charge-discharge curves in the cycle performance test shown in Fig. 2. The cells were operated under a current density of (a) 0.13, (b) 0.25 and (c) 0.64 mA cm<sup>-2</sup> at 25°C with a cut-off voltage of 2.6–4.2 V (vs. Li<sup>+</sup>/Li).

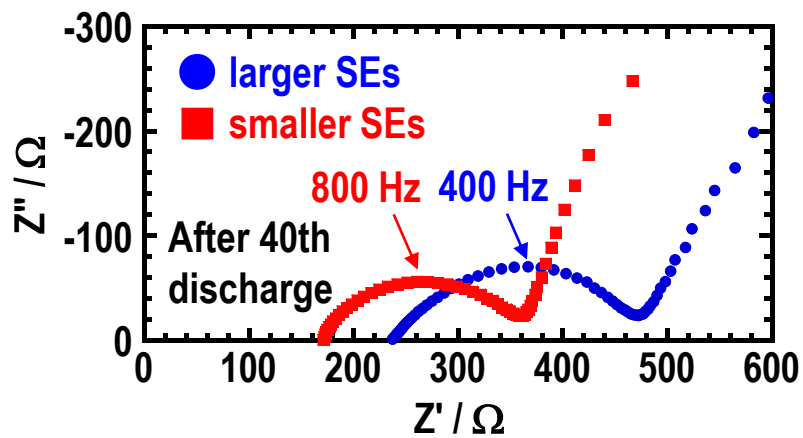


Fig. S2 Nyquist plots of the cells with larger SE and smaller SE particles after the 40th discharge test in Fig. 2.

Table S1 Abundance ratio of high SOC, low SOC and  $\text{Co}_3\text{O}_4$  areas in the Raman mapping images of the cells with larger and smaller SEs after the charge and discharge tests.

|             |            | low SOC | high SOC | $\text{Co}_3\text{O}_4$ |
|-------------|------------|---------|----------|-------------------------|
| Larger SEs  | Charged    | 14.8%   | 84.7%    | 0.5%                    |
|             | Discharged | 47.8%   | 49.6%    | 2.6%                    |
| Smaller SEs | Charged    | 2.0%    | 98.0%    | 0%                      |
|             | discharged | 97.6%   | 2.3%     | 0.1%                    |

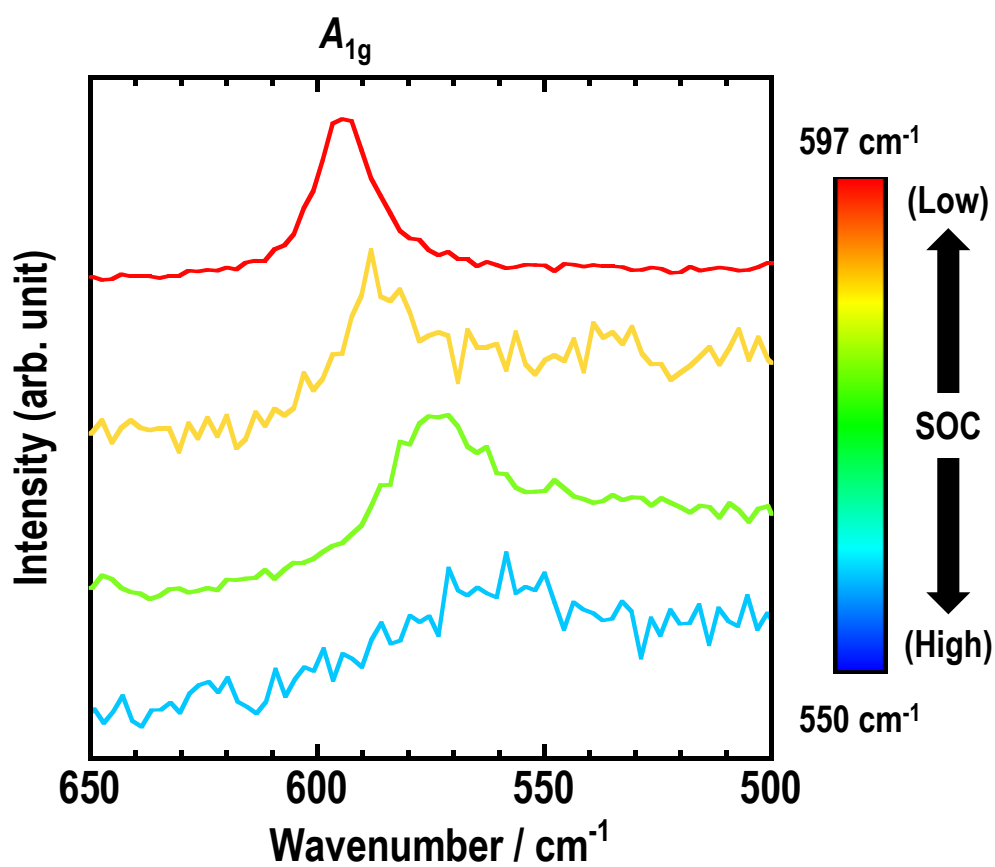


Fig. S3 Raman spectra focused on  $\text{LiCoO}_2$  Raman band attributable to the  $A_{1g}$  mode with various SOC corresponding to the color bar.

## After charge (Larger SEs)

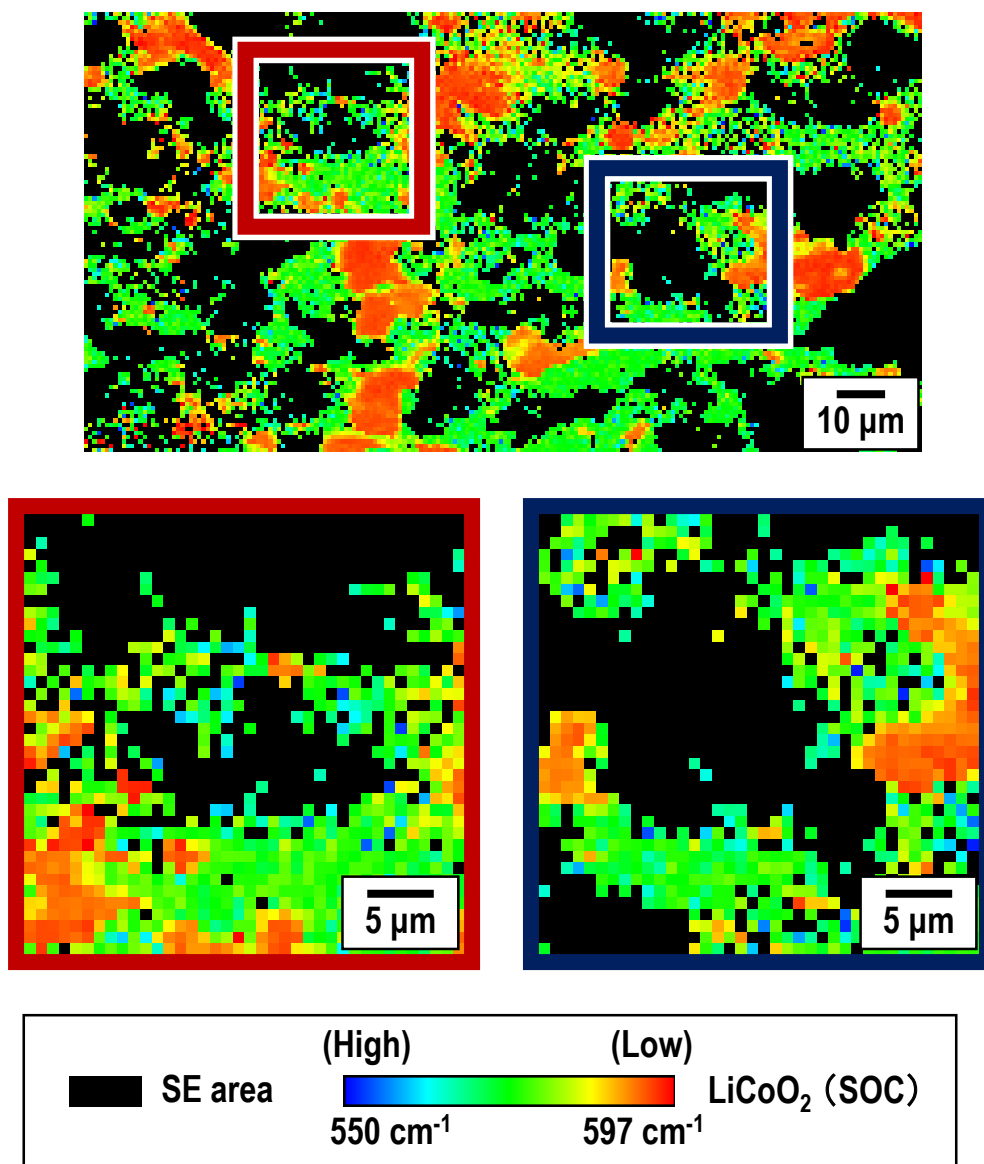


Fig. S4 Enlarged Raman mapping images of the charged  $\text{LiCoO}_2$  electrodes using larger SEs shown in Fig. 4.

## After discharge (Larger SEs)

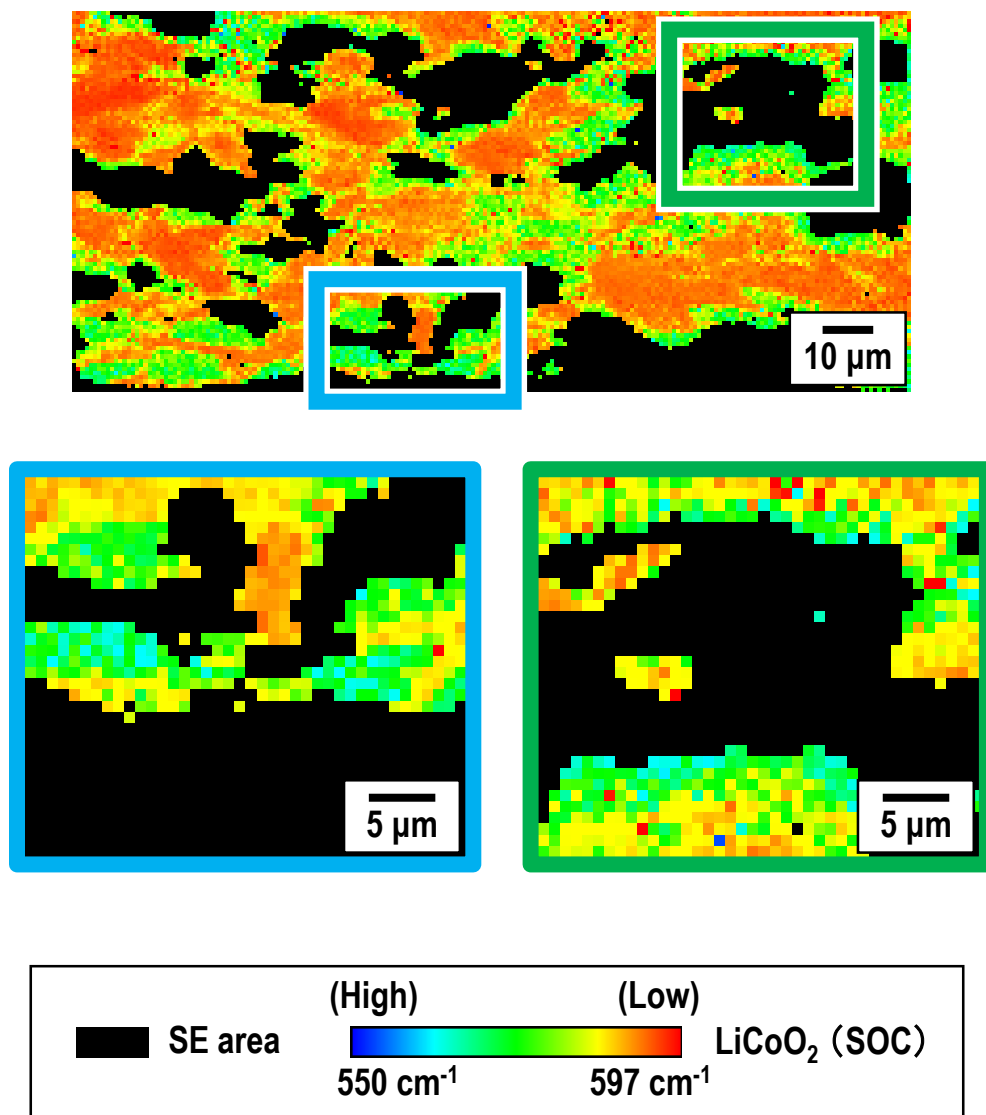


Fig. S5 Enlarged Raman mapping images of the discharged  $\text{LiCoO}_2$  electrodes using larger SEs shown in Fig. 5.