

Supplementary Material

for

Quantification of Single Crystallinity in Single Crystal Cathodes for Lithium-Ion Batteries

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Supplementary Note 1. Rationale for considering agglomerates as single particles in the calculation of single crystallinity.

The agglomeration of single crystal particles can be attributed to three primary factors: (i) simple overlapping of separate particles, (ii) insufficient pulverization during the synthesis process, and (iii) imperfect grain growth and single crystallization. Simple overlapping of separate particles is characterized by irregular interfaces and surface shapes, making it easily distinguishable. As depicted in the figures of the manuscript (Fig. 2a, Fig. 5k, and Fig. 6a), the agglomerates exhibit well-connected interfaces, indicating that they should not be regarded as merely overlapped particles.

The second and third factors are interrelated. All cathode powders underwent pulverization using Jet-mill after heat treatment to separate individual single crystal particles, but some powders remained inadequately pulverized, leaving behind agglomerates. However, the sample sintered at 770 °C exhibited more pronounced agglomeration compared to others, despite undergoing the same pulverization procedure. We attribute this to imperfect grain growth and single crystallization at relatively lower sintering temperature, resulting in incomplete single crystallization yet tightly bound grain boundaries, making them difficult to separate during the pulverization process. Consequently, we determined that these agglomerates should be considered as the final particles with low single crystallinity. In essence, the presence of agglomerates lowers the calculated single crystallinity value, but we believe they are intrinsic and must be accounted for.

Table A1. Complete data for single crystallinity calculation results for the Particles in Figure 5.

According Particle Orientation	Particle 1			Particle 2			Particle 3		
	X	Y	Z	X	Y	Z	X	Y	Z
7.5 °	99.3 %	99.3 %	100 %	96.5 %	71.7 %	94.2 %	54.9 %	55 %	54.9 %
3.75 °	99.1 %	98.4 %	97.0 %	71.8 %	64.5 %	78.1 %	46.3 %	37.3 %	53.3 %
1.88 °	90.2 %	-	-	59.5 %	47.4 %	-	26.6 %	-	37.8 %

Fig. A1. Histograms of the statistically analyzed single crystallinity of individual SCC particles for each sample shown in Table 1.

