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Supporting Information

Understanding the multi-scale battery degradation with a macro-to-nano zoom through its hierarchy

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Electrochemical measurements

We carried out a systematic investigation on an 18650-type commercial LIBs with polycrystalline $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ cathode that failed the factory line's quality control inspection (QCI) due to a self-discharging effect. A standard battery cycler (Shenzhen Neware, CT-4008T-5V6A-S1) was used to perform the galvanostatic charging/discharging tests. The battery was cycled at 2.5-4.2 V with a current density of 2 A/4 A (charging/discharging) for 1 time (see Fig. S1). For the self-discharging test, the battery was firstly charged to 4.2 V with a current density of 1 A, then rested for 25 days at room temperature. Fig. S2 shows the self-discharge curves of 18650-type lithium-ion battery.

Supplementary Figures

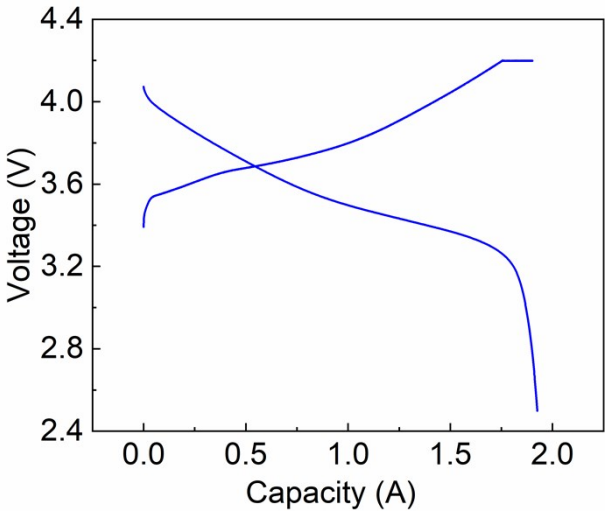

Fig. S1 The charging/discharging curves of 18650-type lithium-ion battery at 2.5-4.2 V with a rate of 1 C/2 C (1 C=2 A).

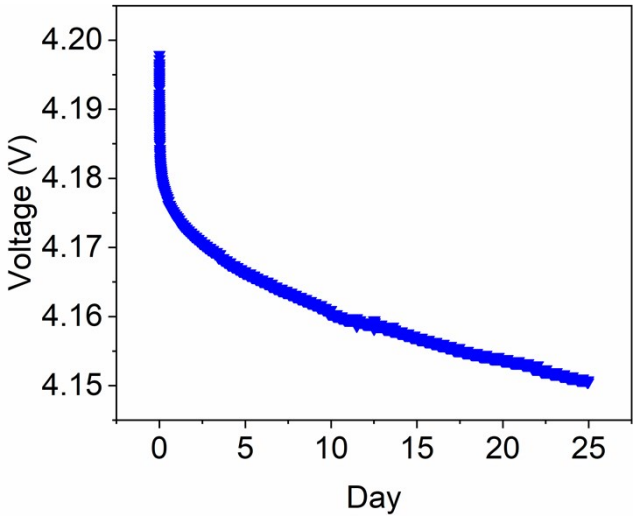

Fig. S2 The self-discharge curves of 18650-type lithium-ion battery.

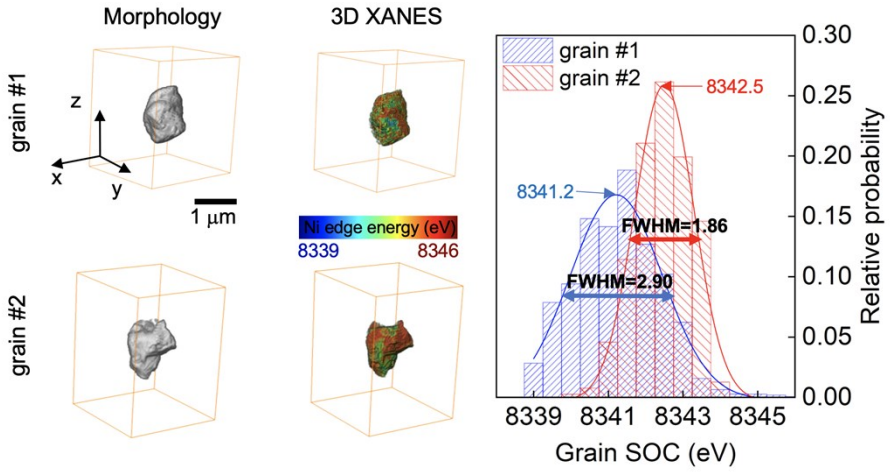

Fig. S3 The inter-granular and intra-granular heterogeneities between two randomly selected primary grains. The 3D morphology and Ni K-edge energy heterogeneity of two different grains are shown on the left. The relative probability distributions of the Ni K-edge energy value for these two grains are plotted on the right. Grain #2 shows a higher averaged Ni K-edge energy value at 8342.5 eV, indicating that grain #2 is more oxidized than grain 1#. The FWHM of the curves on the right effectively measures the degree of energy spread. Grain 1# appreas to be more non-uniform than grain 2#.