Supporting information for:

Mitigating X-ray-Induced Damage for Accurate Analysis of Battery Cathodes Using Transmission X-ray Microscopy

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Experimental methods.

Electrochemistry. Cathodes of $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ (NMC622) and $Li_{1.2}Ni_{0.15}Co_{0.1}Mn_{0.55}O_2$ were prepared with 80% active material, 15% carbon black, and 5% polyvinylidene difluoride (PVDF) binder. N-methyl pyrrolidone (NMP) was used as solvent to prepare the cathode slurry. For operando coin cell, cathode slurry with NMC622 in the above-mentioned ratio was casted on aluminum foil, dried on vacuum oven overnight at 120 ºC, and cut into 12 mm diameter size disks. Operando coin cells were then assembled with the cathode disk, Li metal as the counter electrode, Celgard 2320 separator, and 1.2 M $LiPF_6$ in a 3:7 ratio of ethylene carbonate and ethyl methyl carbonate as the electrolyte. The holes in the operando coin cells were sealed with Kapton tape and epoxy glue to prevent air leak. Operando pouch cells were made with $Li_{1.2}Ni_{0.15}Co_{0.1}Mn_{0.55}O_2$ cathode casted on an aluminum foil current collector, Li metal foil as anode with Cu strip placed on one side as current collector, Celgard 2320 as separator, and 1.2 M $LiPF_6$ in a 3:7 ratio of ethylene carbonate and ethyl methyl carbonate as the electrolyte. The whole assembly was housed in a polymer coated coffee bag and sealed with an impulse sealer inside the glovebox. A one-time pressure of 5psi was applied to the cell after assembly. During operando experimentation, a constant pressure was applied to the cell during the entirety of the experiment by two steel plates through tightening four screws in the corners all the way. All cells were cycled with a Biologic in the full field x-ray imaging beamline for operando image collection.

Operando transmission x-ray microscopy imaging. Transmission x-ray microscopy imaging was performed in full-field x-ray imaging beamline (18-ID) at National Synchrotron Lightsource II (NSLS-II) at Brookhaven National Laboratory. Images were collected at both a specific energy of 8.4 keV and scanning around the white line energy of Ni K-edge to create Ni K-edge x-ray

absorption near edge structure (XANES) maps. Image reconstruction and fitting around the white line of the Ni K-edge were performed in the TXM-Sandbox software package.

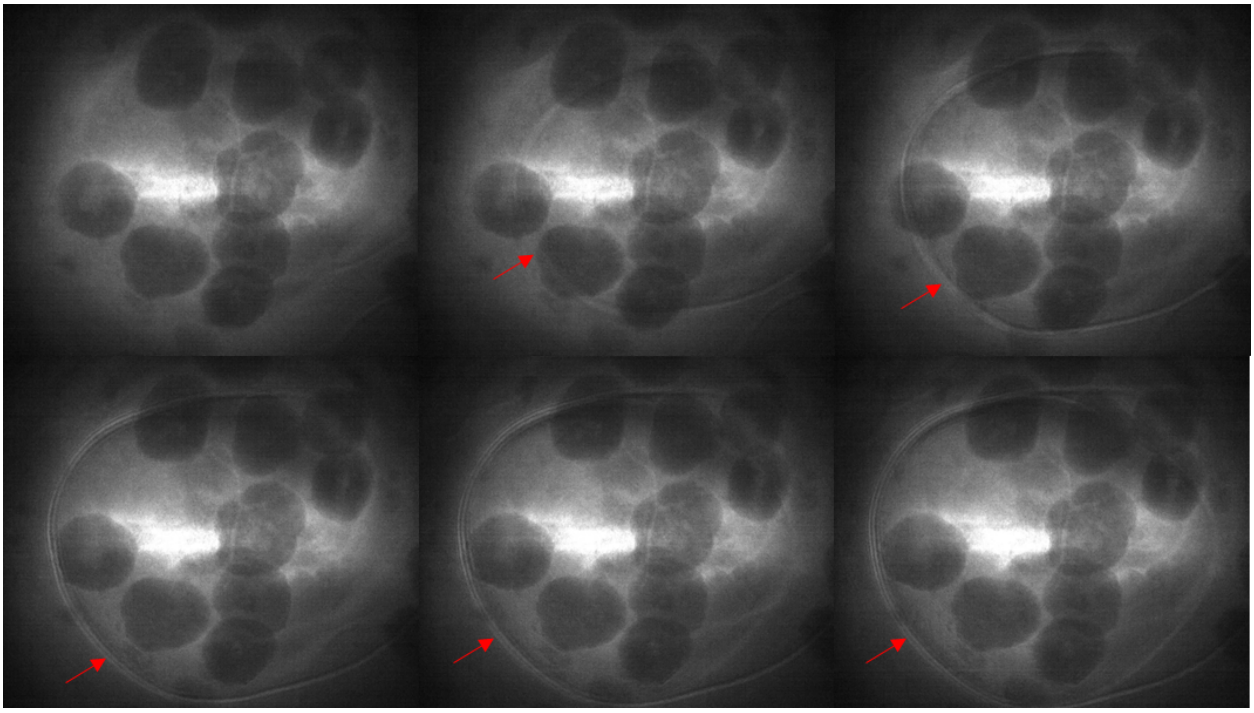

Figure S1. Bubble formation and propagation within a field of view exposed to x-ray at 8.4 KeV. The battery was at rest during this set of image collection.

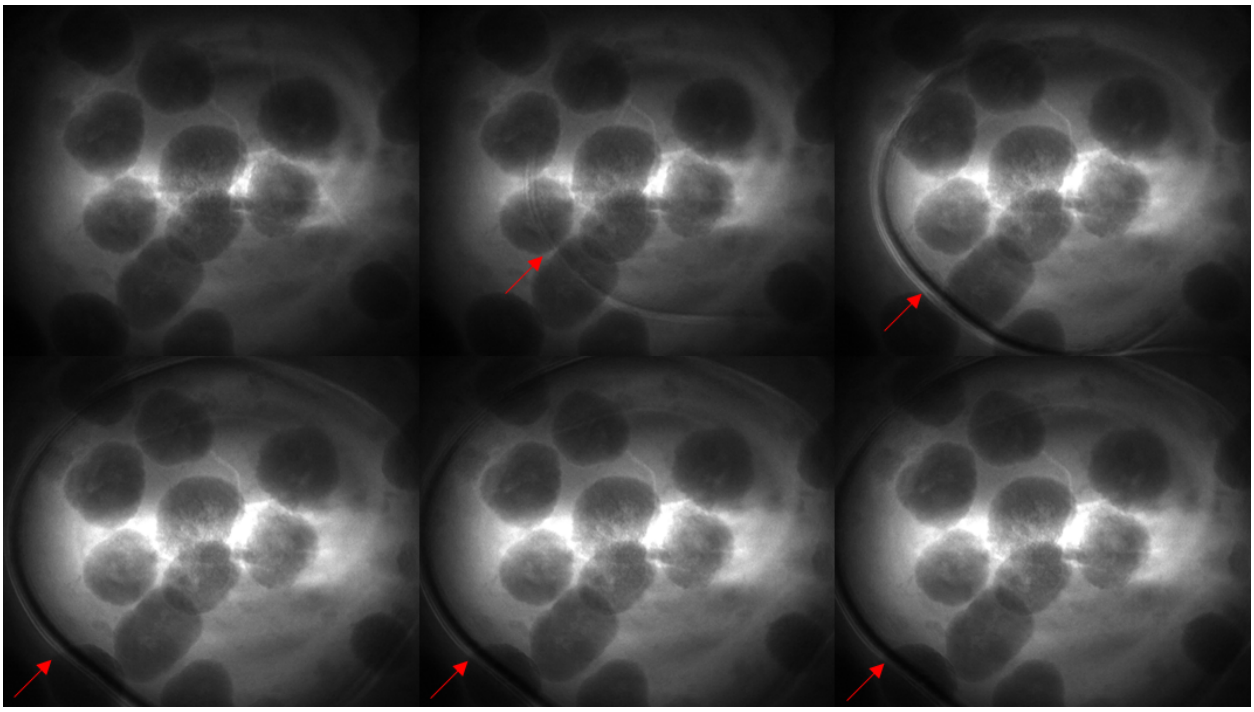

Figure S2. Bubble formation and propagation within a different field of view with prolonged exposure to x-ray at 8.4 KeV. The battery was at rest during this set of image collection.

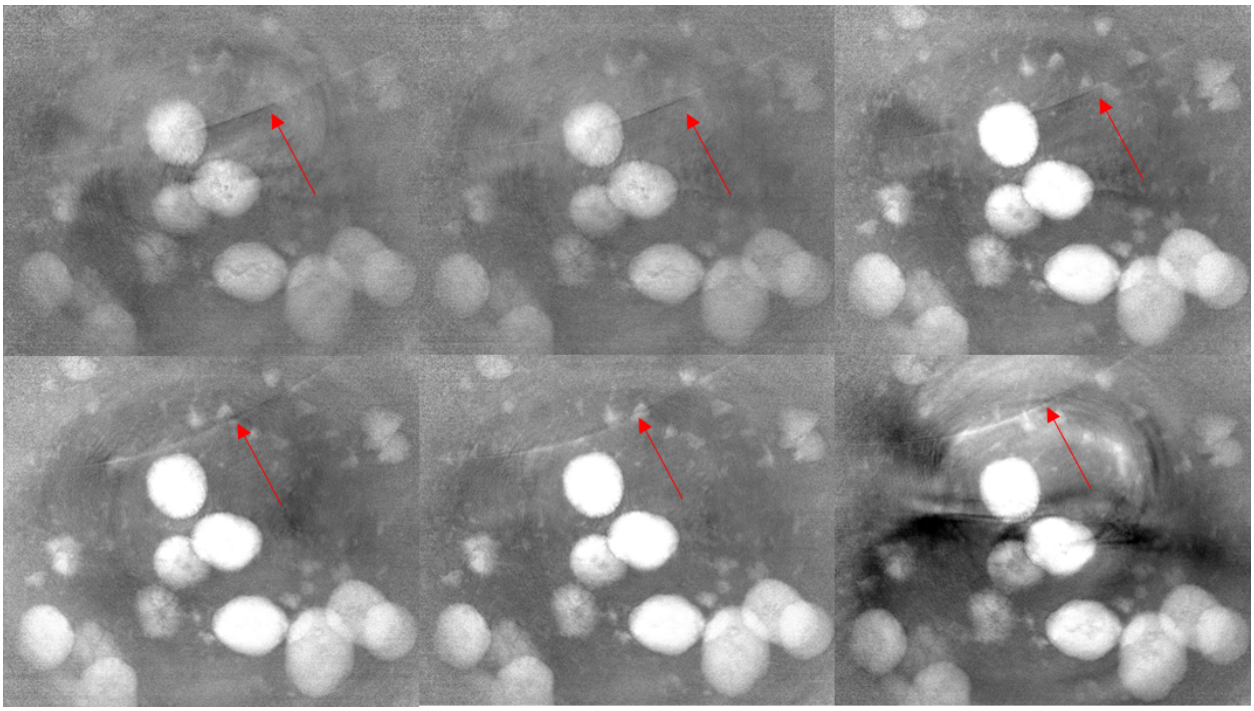

Figure S3. Bubble formation and propagation during the initial stage of battery operation. For this set of scans, the battery cycling was just commenced. All the images are from one set of scans with energy of image acquisition rising from left to right in top row to bottom row in the same direction. The particles appear increasingly brighter as the energy approaches the white line energy of Ni K-edge.

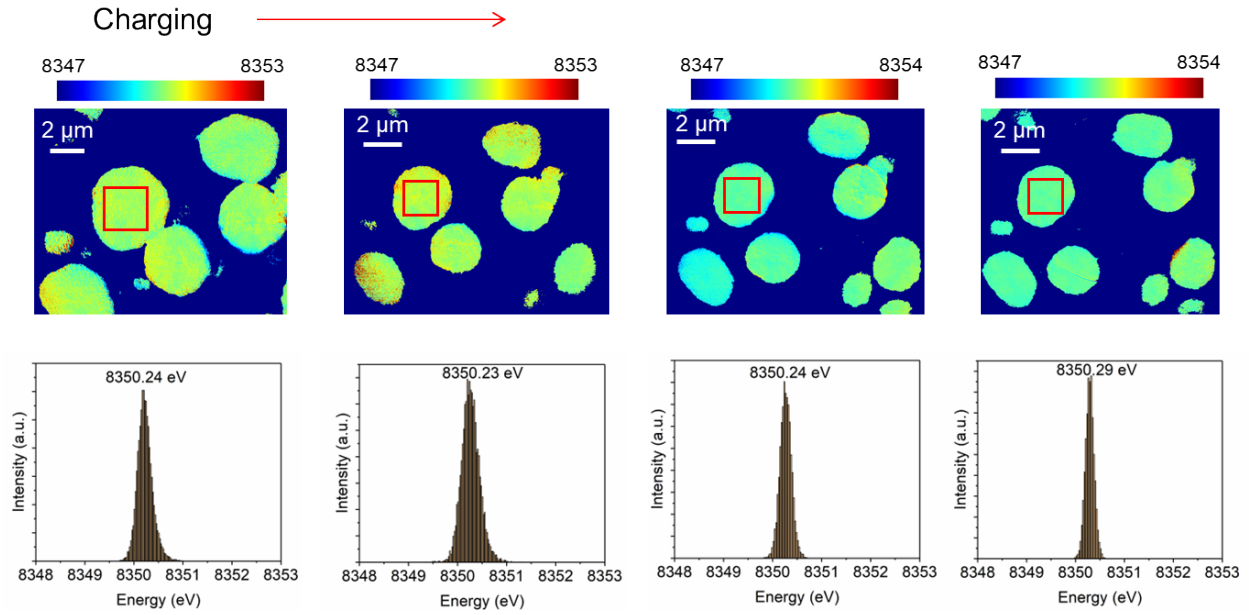

Figure S4. Histogram analysis of Ni white line energy distribution within the boxed region of the field of view.

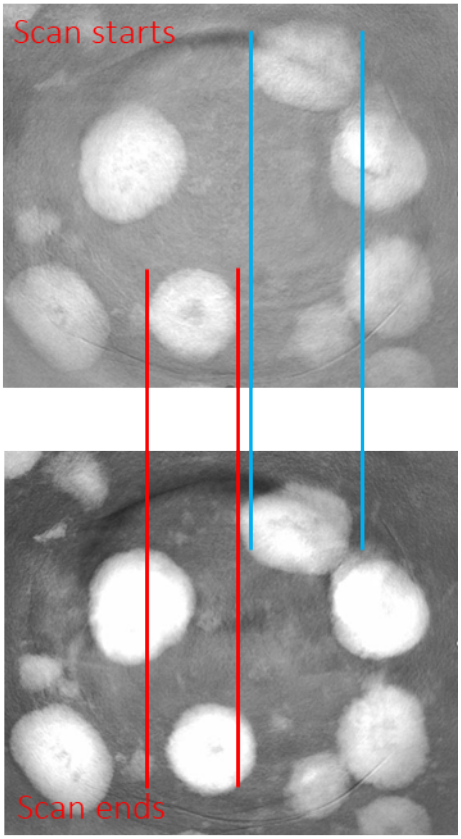

Figure S5. Particle movement within the field of view during scanning.