Supporting Information

Battery Cycling

Experiments were carried out on a single LG MJ1 18650 cell, which contained a mixed Si/graphite anode and nickel manganese cobalt oxide (NMC) cathode (INR18650, 3400 mAh, LG Chem Ltd., Seoul, South Korea). The cell had a maximum rated discharge of 10 A with a maximum charging current of 1C. Battery cycling was performed using a Maccor 4300 (Maccor Inc., Oklahoma, USA) with all testing conducted on a fresh cell within the manufacturer's standard operating limits. Charging of the cell was conducted at 1700 mA until 4.2 V, with the voltage then held constant until the current tapered to 50 mA. Discharge was at 680 mA to 2.5 V. Resistance measurements were also performed throughout the cycling using an electrical pulse technique. The resistance of the cell also increased during cycling. The resistance at the top of charge increased from 57 mΩ initially to 104 mΩ, whereas a smaller increase was calculated at the bottom of discharge (from 114 mΩ to 121 mΩ). The capacity of the cell was seen to fade with the increasing cycle number as shown in Figure 2(a).

Tomography

The battery was imaged fully charged before any cycling and then after 1061 cycles using a Nikon XT H 225 instrument (Nikon Metrology, Tring, UK), operating with a source voltage of 205 or 210 kV at 330 µA. A total of 3176 projections of 1s exposure were obtained for each scan, recorded through 360 degrees. The X-ray transmission images were reconstructed using a proprietary reconstruction algorithm (CT Pro 3D, Nikon Metrology). The reconstructed volumes had a voxel size of 36.0 µm and 36.6 µm for the pre-cycle and post 1061 cycles respectively.

The high speed scans were acquired on the same instrument with varying conditions. The 26 min scan used a total of 3176 projections with an exposure time of 534 ms. The 160 second scan used 600 projections with a 267 ms exposure time and the 80 second scan was done with 300 projections at 267 ms exposure.

Region of interest (ROI) scans were performed using the Zeiss Xradia Versa 520 micro-CT instrument (Carl Zeiss XRM, California, USA) operating at 159 kV and 63 µA. A total of 1601 projections of 40 s were recorded through 360 degrees and were reconstructed using a filtered back-projection reconstruction algorithm (XM Reconstructor, Zeiss). The larger field of view tomogram used a 0.4× objective lens resulting in a 10 µm voxel size, while the smaller, more detailed, ROI used a 4× objective lens yielding a 1.97 µm voxel size.