Multiphase, Multiscale Chemomechanics at Extreme Low Temperatures: Battery Electrodes for Operation in a Wide Temperature Range

Lithium-ion battery (LIB), a tremendously successful technology for energy storage, has become the dominant power source in applications ranging from consumer electronics to electric vehicles. There is an increasing desire for stable storage and utilization of electrical energy under extreme conditions such as cold-climate, aerospace exploration, and subsea operations. These applications feature highly diverse and varying working environments with overcharge/discharge, high pressure, external forces and, often most critical, high/low temperature. For example, LIBs lose most of their capacity, power, and cycle life when operated below ambient temperature. Therefore, it is crucial to gain more insights into the mechanisms causing the LIB performance degradation upon exposure to low-temperature conditions, which could aid engineering of active materials, electrode structure, and charge/discharge protocols.

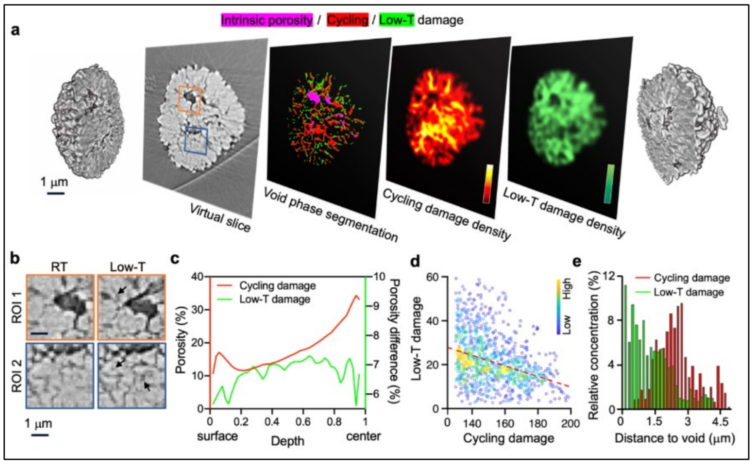

Figure 1. TXM characterization of single NMC particle damages. (a) A 3D rendering of single NMC particle with its central slice shown in the middle. The particle's structural damages of different origins are segmented and then used for calculating of the local damage density. (b) Two selected regions of interest, as indicated in (a), are compared before and after the low-temperature exposure. (c) Depth-dependent profiles of the damages caused by electrochemical cycling and by exposure to -40 °C. (d) The spatial association of the cycling-induced cracks and the low-temperature-induced damages, showing a mild negative correlation. (e) The relative frequency histogram of cycling-induced damage and low-temperature-induced damage as a function of the distance from the nearest pore phase, which suggests a closer association of the low-temperature-induced damage with the intrinsic porosity.

Immense efforts have been devoted to improving lithium-ion transport in the electrolyte and through the solid electrolyte interphases (SEI). A significant irreversible LIB performance degradation upon exposing to low-temperature conditions, however, is not understood and rarely tackled. A research team led by Drs. Yijin Liu (SSRL), Peter Cloetens (ESRF) and Profs. Feng Lin (Virginia Tech) and Kejie Zhao (Purdue) systematically elucidated multiphase, multiscale chemomechanical behaviors in composite $LiNi_xMn_yCo_zO_2$ (NMC, x+y+z=1) cathodes at extreme low temperatures. By utilizing a suite of advanced characterization techniques including synchrotron-based x-ray powder diffraction (XRD, beamline 14-1 at SSRL), X-ray absorption near edge structure (XANES, beamline 7-3 at SSRL), extended x-ray absorption fine structure (EXAFS, beamline 7-3 at SSRL), full-field transmission X-ray microscopy (TXM, beamline 6-2c at SSRL), and x-ray phase contrast nano-holo-tomography (beamline ID16A-NI at ESRF), the authors observe that, upon exposure to low temperature and then recover to room temperature, the reversible and anisotropic lattice deformation could lead to irreversible cracking of active cathode particles (**Figure 1**). At the electrode scale, the morphological deformations of different cathode components mismatch at low temperatures, causing structural disintegration that could irreversibly provoke the development of local impedance and particle deactivation (**Figure 2**).

This work presents a fundamental understanding of the mechanisms behind the irreversible performance degradation of LIBs upon exposure to extreme low temperature. Their results suggest that, in order to design batteries for use in a wide temperature range, it is critical to develop electrode components that are structurally and morphologically robust when the cell is switched between different temperatures.

Primary Citation

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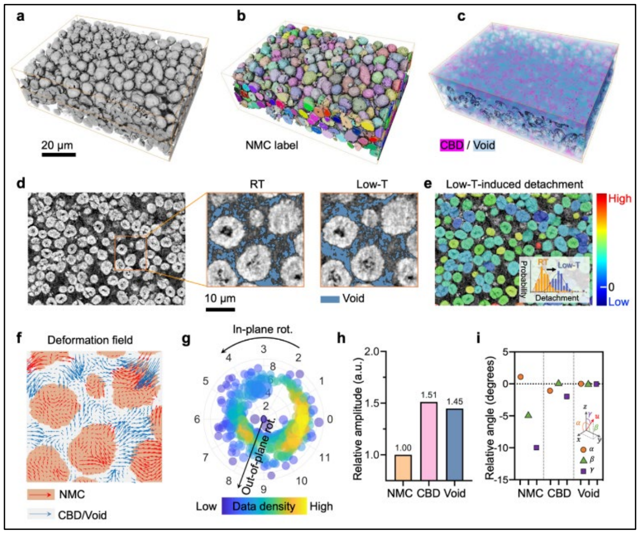

Figure 2. Nano-holo-tomography characterization of the composite cathode. (a) 3D visualization of the X-ray phase contrast nano-holo-tomographic data on the composite cathode with the identified NMC particles and segmented inactive phases, e.g. CBD and Void, shown in (b) and (c), respectively. (d) A representative lateral virtual slice with an enlarged region of interest compared before and after the low-temperature exposure. (e) The detachment caused by low temperature exposure is calculated particle by particle and the corresponding value is used to color code the map in panel (e). (f) The visualization of the low-temperature-induced structural deformation field over the region of interest shown in panel (d). (g) The statistical distribution of the deformation vectors' orientations (in degrees) of all NMC particles. (h) The relative amplitudes of different phases deformation normalized to that of the NMC particles. (i) The relative angle of different phases' deformation normalized to that of the void phase.

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