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Li-ion and Li-air Batteries: Harnessing Oxygen Redox and Understanding Interfacial Reactivity in High Energy Batteries

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Abstract: Multiple directions in battery research are now being pursued with the goal of advancing beyond the specific energy limits imposed by current Li-ion batteries. When considering the design of new high-energy storage systems, new materials, processes, or chemistries are introduced that are inherently more unstable than conventional Li-ion battery materials, resulting in limited battery cycle life and safety. Two such examples of high energy battery chemistries, high voltage operation of Ni-rich Li[Ni, Mn, Co]O_2 Li^+ insertion electrodes (Ni-rich NMC) and Li-O_2 electrochemistry, will be discussed in this presentation. Previous observations of high-voltage instabilities include NMC surface reconstruction, transition metal dissolution, electrolyte decomposition, and formation of solid surface species. However, the picture of these processes is still incomplete, with the dependence on electrolyte and NMC composition not yet fully understood. I will present results in which isotopic labeling of ^{18}O in Ni-rich NMCs is combined with quantitative gas evolution analysis to identify key contributions to these high voltage instabilities, including instabilities related to solid-state anionic (oxygen) redox and the surprising impact of residual solid lithium carbonate (Li_2CO_3) on electrolyte and electrode degradation. These results are reminiscent of similar issues with Li_2CO_3 formation during Li-O_2 battery operation, where large overpotentials are observed during battery charging as a result of parasitic interfacial carbonate formation. This presentation will emphasize the need to accurately quantify these minor parasitic side reactions to fully understand their large influence on battery performance.