Oxygen Redox Mechanisms in Commercial Lithium-Ion Battery Cathodes Revealed through RIXS and sXAS

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Lithium-ion batteries are the cornerstone of modern energy storage, powering mobile devices and electric vehicles. However, their theoretical energy density remains underutilized due to the instability of cathode materials at high states of charge. While oxygen redox has been suspected to play a role in this instability, indirect evidence has upheld the dominance of the transition metal-based redox paradigm. In this study, we uncover direct evidence of oxygen involvement in the redox process, identifying distinct oxygen species through resonant inelastic X-ray spectroscopy (RIXS).[1] Complementary insights from soft X-ray absorption spectroscopy (sXAS) further demonstrate oxygen's role in charge compensation. Additionally, we investigate the stability of oxygen redox as influenced by cathode chemical composition, revealing the detrimental impact of unstably oxidized lattice oxygen at high states of charge.[2] Importantly, we show that dopants can mitigate these effects, stabilizing oxygen redox and enhancing cathode performance. This work lays the foundation for developing high-energy-density cathodes that maximize specific capacity while addressing degradation challenges, paving the way for next-generation energy storage solutions.

Keywords: Lithium-ion battery, resonant inelastic X-ray scattering, X-ray absorption, redox mechanism, materials discovery.

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