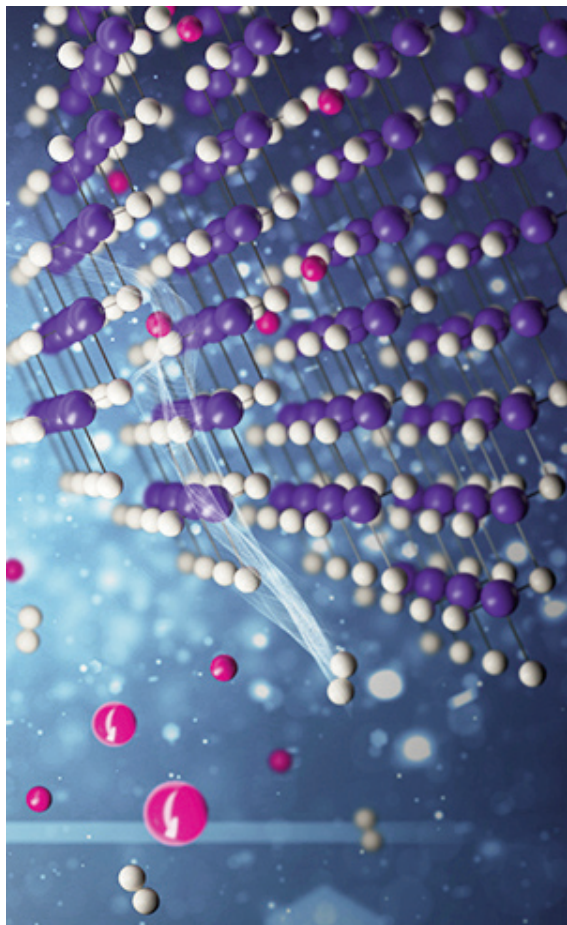


# THE ADVANCED PHOTON SOURCE

## A Cozy Graphene Coat Helps LNOs Reach Their Full Potential



A schematic of the high-voltage degradation that occurs at the electrode-electrolyte interface in cobalt-free  $\text{LiNiO}_2$  lithium-ion battery cathodes.

Lithium-ion batteries (LIBs) are the preferred storage solution because of their energy capacity and extended recyclability. Those qualities can be enhanced by the choice of cathode materials. One of the strongest candidates for improving cathode performance is lithium nickel oxide,  $\text{LiNiO}_2$  or LNO, which features high capacity, reduced costs, and avoids the ethical issues associated with the cobalt supply chain. However, the utility and practicality of LNO cathodes is hampered by poor stability and electrochemical degradation, especially when cycled at higher operating voltages over 4 volts.

Using the U.S. Department of Energy's Advanced Photon Source (APS), researchers found that oxygen stacking lattice changes are the primary culprit. Ni-rich layered oxides such as LNO tend to lose surface oxygen during charging to high levels, which creates instability and drives stacking transitions. The formation of oxygen vacancies coupled with the oxygen stacking changes leads to a degradation in the LNO structure.

With repeated cycles, this combination of faults causes increasing particle deformation at the microscale with cracking, bending, and creep, compromising electrical connections and resulting in steady capacity loss.

Given these observations, the investigators reasoned that preventing or suppressing initial surface oxygen loss might forestall the subsequent degradation cascade and preserve LNO capacity at high voltages. They tested this hypothesis by coating LNO powder particles with a hermetic layer of graphene, known to suppress oxygen evolution in cathode materials. Comparison of coated and uncoated LNO particles under scanning electron microscopy and cyclic voltammetry demonstrated that the phase transition is definitively affected in the graphene-coated particles.

This work provides not only detailed insight into the source of nagging technical problems of limited capacity and cycle life that have hampered the widespread adoption of high Ni-content layered oxide cathodes such as LNO for high-voltage uses but also points to a practical solution. By suppressing oxygen evolution at the initial electrochemical phases with the simple expedient of a graphene-based coating, the superior capacity of LNO cathodes can be preserved at high voltage, unlocking the full commercial potential of these valuable materials. – Mark Wolverton

See: Kyu-Young Park, Yizhou Zhu, Carlos G. Torres-Castanedo, Hee Joon Jung, Norman S. Luu, Ozge Kahvecioglu, Yiseul Yoo, Jung-Woo T. Seo, Julia R. Downing, Hee-Dae Lim, Michael J. Bedzyk, Christopher Wolverton, and Mark C. Hersam, "Elucidating and Mitigating High-Voltage Degradation Cascades in Cobalt-Free  $\text{LiNiO}_2$  Lithium-Ion Battery Cathodes," *Adv. Mater.* **34**, 2106402 (2022). DOI: 10.1002/adma.202106402

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