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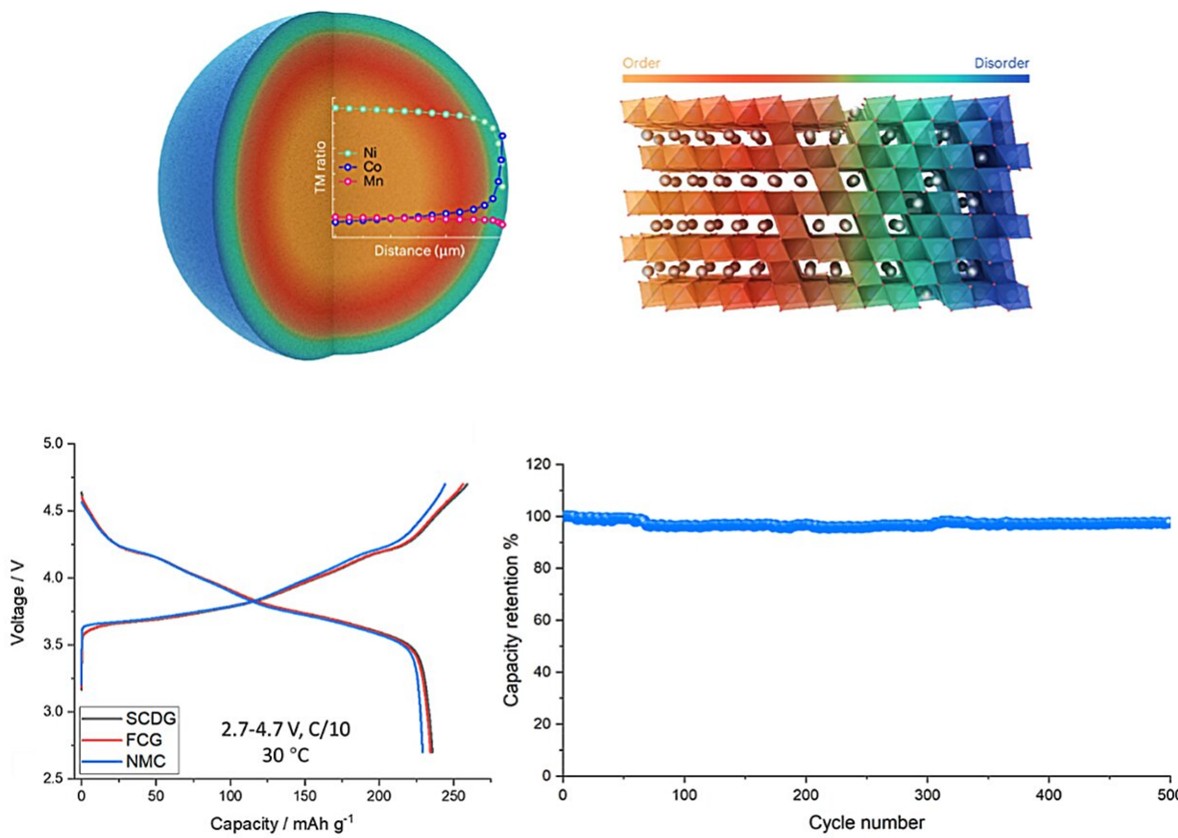
Ultra-stable layered oxide cathodes could boost battery performance

August 26 2024, by Ingrid Fadelli



a. Schematic diagram of the structure and composition dual gradient particle (SCDG) with gradient structure from order to disorder towards outer layer and gradient composition from Co-less bulk to Co-enriched surface. b. The capacity of SCDG compared to conventional layered NMC cathode and the 1st generation full concentration gradient cathode. c. The superior cycle stability of SCDG cathode with negligible capacity loss within 500 cycles in full cells. Credit: Liu et al.

Recent efforts aimed at developing more advanced battery technologies have in great part focused on designing novel cathode materials. This is because existing cathodes do not perform well at high voltages and can contribute to the rapid loss of battery capacity.

Layered oxide cathodes, a class of cathode materials with a layered crystal structure, have been found to be particularly promising for the development of next-generation batteries. Initial findings suggest that these materials could improve the performance of lithium-ion batteries, while also reducing their fabrication costs and limiting their environmental impact.

Researchers at Argonne National Laboratory recently designed novel ultra-stable NMCs cathodes, a type of layered oxide cathodes composed of nickel (Ni), manganese (Mn) and cobalt (Co). These newly designed materials, introduced in a paper in *Nature Energy*, were found to enable high performance in lithium-ion batteries without significant losses in capacity.

"To further advance NMC cathodes, our team developed a series of concentration gradient NMC cathodes to optimally harness the beneficial characteristics of Ni, Mn and Co," Dr. Khalil Amine, Argonne distinguished fellow and lead author of the paper, told Tech Xplore.

"In this concentration gradient cathode, the nickel concentration decreases linearly whereas the manganese concentration increases linearly from the center to the outer layer of each particle."

This full-gradient cathode design, which was patented by Dr. Amine in 2012, leverages the high energy density of Ni (found at the core of the cathodes), as well as the high thermal stability and long life of the Mn in the cathode's outer layers. Notably, this design has already been licensed to various battery technology and materials manufacturers.

"In pursuit of higher energy density and lower cost of next generation batteries, we pressed NMC cathodes for higher voltage operation (≥4.5 V) to achieve high capacity, which is over the voltage limitation of conventional layered structure and lead to fast capacity loss," said Dr. Amine.

"In addition, current bottlenecks in cobalt (Co) supply have negatively impacted commercial battery production and inspired the development of cathode materials that are less reliant on Co."

To overcome the limitations of existing NMC cathode designs, Dr. Amine and his colleagues set out to design a second updated version of their gradient cathodes. This second generation of cathodes is characterized by both concentration and structure-related gradients, which collectively address the shortcomings of existing cathodes with layered structures at high voltages.

Moreover, the researchers lowered the concentration of Co in the cathodes. This change in composition could significantly reduce both the cathode materials' manufacturing cost and their environmental impact.

"Previous layered cathodes suffer from a tradeoff between capacity, cyclability and safety. For example, increasing operation voltage could improve their capacity but at the expense of cycle life," explained Dr. Amine. "As a result, most batteries used in EVs are limited to being operated below 4.3V because the inherent structure tends to degrade at high voltages, leading to shortened lifetime and high safety risk."

The new cathodes introduced as part of this recent study have a unique composition and dual-gradient design, which address the voltage ceiling observed in other existing cathodes. By combining the advantages of different components and material structures into a single cathode, the team were able to attain outstanding performances.

"In detail, the bulk layered structure with high Ni content is able to deliver a high capacity and surface disorder rock-salt structure could withstand high voltage up to 4.7V without severe structure changes," said Dr. Tongchao Liu, co-author of the paper.

"Therefore, this dual gradient cathode could simultaneously achieve high capacity and superior cycle life when operating at high voltages (>4.5V). In addition, this design could reduce Co usage up to 1% and maximize its functionalities and reduce safety risk."

The researchers' newly introduced materials deviate from conventional cathode designs, which typically utilize a single structure and high Co concentrations. In initial experiments, the new cathodes were found to perform remarkably well, enabling the high-capacity and high-voltage operation of batteries at 4.5 V without any capacity losses, as well as negligible capacity fading when operating up to 4.7 V.

"By integrating the high energy density of the layered phase with the structural stability of the disordered rock-salt phase, our design addresses the longstanding tradeoff between capacity, cycle life, and safety," said Dr. Amine. "This innovation not only enhances the overall performance of the cathode but also broadens the research directions for cathode material design, allowing for the creation of new materials that far surpass existing ones."

This recent research effort opens new possibilities for the development of Li-ion batteries with lower Co concentrations that retain high capacities for longer periods of time, even while operating at high voltages. Moreover, the cathodes introduced by Dr. Amine and his colleagues could soon inspire other research teams to design similar materials with dual-gradient structures.

"The next steps for our research will involve further optimizing the dual-gradient design to further reduce Co and Ni usage while enhancing its energy density and scalability," said Dr. Amine. "We aim to explore additional material compositions and structural modifications to push the boundaries of energy density and stability even further."

As part of their future work, the researchers also plan to integrate their cathodes into full battery systems, as this will allow them to test their real-world performance and assess their compatibility with existing battery components. To run these tests, Dr. Amine patented his updated design and is initiating collaborations with battery manufacturers.

"In the long term, we envision our dual-gradient design inspiring a new generation of high-performance, cost-effective, and sustainable battery materials," added Dr. Liu. "By reducing reliance on cobalt and enhancing the structural integrity of cathodes at high voltages, our work could significantly impact the development of next-generation batteries for electric vehicles, portable electronics, and grid storage."

Argonne's Advanced Photon Source and Center for Nanoscale Materials (both part of the DOE Office of Science user facilities) and Brookhaven National Laboratory performed a series of experiments using X-ray, electron, and imaging techniques to characterize the new cathode material at rest and while operating.

These tests collectively assessed the material at the cathode, particle, and atomic levels and provided a comprehensive picture of its composition, structure, and performance.

More information: Tongchao Liu et al, Ultrastable cathodes enabled

by compositional and structural dual-gradient design, *Nature Energy* (2024). DOI: 10.1038/s41560-024-01605-8