A technique to guide the development of faster and longer lasting next-generation batteries

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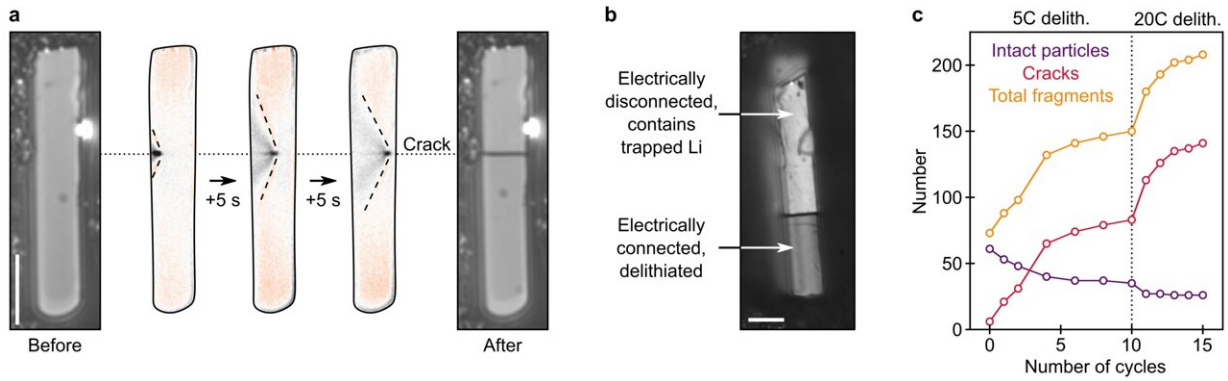

a) Optical images showing crack formation in a single rod-shaped particle of $Nb_{14}W_3O_{44}$. Black dashed lines highlight Li-ion fronts propagating from the crack. b) Optical image of a fractured particle, following 20 charge-discharge cycles. The more brightly scattering fragment has a higher Li-content, suggesting it has become inactive. Scale bars are 5 μm. c) Extent of particle fracture in a fixed population of active particles, over 15 charge-discharge cycles. Credit: Research Team, Cavendish Laboratory, Department of Physics, University of Cambridge

Clean and efficient energy storage technologies are essential to establishing a renewable energy infrastructure. Lithium-ion batteries are already dominant in personal electronic devices, and are promising candidates for reliable grid-level storage and electric vehicles. However,

further development is needed to improve their charging rates and usable lifetimes.

To aid the development of such faster-charging and longer-lasting batteries, scientists need to be able to understand the processes occurring inside an operating battery, to identify the limitations to battery performance. Currently, visualizing the active battery materials as they work requires sophisticated synchrotron X-ray or electron microscopy techniques, which can be difficult and expensive, and often cannot image quickly enough to capture the rapid changes occurring in fast-charging electrode materials. As a result, the ion dynamics on the lengthscale of individual active particles and at commercially-relevant fast-charging rates remains largely unexplored.

Researchers at the University of Cambridge have overcome this problem by developing a low-cost lab-based optical microscopy technique to study lithium-ion batteries. They examined individual particles of $Nb_{14}W_3O_{44}$, which is among the fastest charging anode materials to-date. Visible light is sent into the battery through a small glass window, allowing the researchers to watch the dynamic process within the active particles, in real time, under realistic non-equilibrium conditions. This revealed front-like lithium-concentration gradients moving through the individual active particles, resulting in internal strain which caused some particles to fracture.

Particle fracture is a problem for batteries, since it can lead to electrical disconnection of the fragments, reducing the storage capacity of the battery. "Such spontaneous events have severe implications for the battery, but could never be observed in real time before now," says co-author Dr. Christoph Schnedermann, from Cambridge's Cavendish Laboratory.

The high-throughput capabilities of the optical microscopy technique

enabled the researchers to analyze a large population of particles, revealing that particle cracking is more common with higher rates of delithiation and in longer particles. "These findings provide directly-applicable design principles to reduce particle fracture and capacity fade in this class of materials," says first author Alice Merryweather, a Ph.D. candidate at Cambridge's Cavendish Laboratory and Chemistry Department.

Moving forward, the key advantages of the methodology—including the rapid data acquisition, single-particle resolution, and high throughput capabilities—will enable further exploration of what happens when batteries fail and how to prevent it. The technique can be applied to study almost any type of battery material, making it an important piece of the puzzle in the development of next-generation batteries.

The research was published in *Nature Materials*.

More information: Alice J. Merryweather et al, Operando monitoring of single-particle kinetic state-of-charge heterogeneities and cracking in high-rate Li-ion anodes, *Nature Materials* (2022). DOI: 10.1038/s41563-022-01324-z

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