

Internal Currents in Lithium Batteries after Fast Charging



Three-dimensional tomographic data from a lithium-ion battery electrode, a few minutes after fast charging (start of rest) and about 20 minutes later (end of rest). The blue layer shows metallic lithium that has plated on the electrode surface, while the black layer shows underlithiated graphite particles in the electrode itself. By analyzing changes in this data over time, researchers were able to quantify ionic currents generated by the movement of lithium ions from the plated lithium to the underlithiated graphite.

The mystery of thermal runaway at rest

How likely is an electric-vehicle (EV) battery to self-combust and explode? The chances of that happening are actually pretty slim: Some analysts say that gasoline vehicles are nearly 30 times more likely to catch fire than EVs. But recent news of EVs catching fire while parked have left many consumers—and researchers—scratching their heads over how these rare events could possibly happen.

Researchers have long known that high electric currents can lead to "thermal runaway"—a chain reaction that can cause a battery to overheat, catch fire, and explode. But without a reliable method to measure currents inside a resting battery, it has not been clear why some batteries go into thermal runaway when an EV is parked. Now, by using operando x-ray microtomography at the ALS, scientists have shown that the presence of large local currents inside batteries at rest after fast charging could be one of the causes behind thermal runaway.

A source of local ionic current

In a lithium-ion battery, the anode is mostly made of graphite. When a healthy battery is charged slowly, lithium ions from the cathode weave themselves between the layers in graphite particles, which can then reach a full state of charge. In contrast, when the battery is charged rapidly, the lithium ions have a tendency to plate on the surface of the graphite in the form of lithium metal.

Previous microtomography experiments at the ALS indicated that these deposits cast lithiation "shadows"—regions of poor



In lithium batteries after fast charging, researchers measured the persistence of internal currents and found that large local currents continue even after charging has stopped.

Significance and Impact

The work uses hard x-ray 3D imaging at the Advanced Light Source (ALS) in a novel way and sheds light on the causes of thermal runaway and the catastrophic failure of lithium batteries at rest.

lithiation at the back of the graphite electrode. When this happens, even though the battery is at rest, there is a potential difference between the plated lithium metal and the underlithiated particles that can drive local ionic currents. Measuring these time-dependent currents requires a nondestructive operando technique that's also spatially resolved, because the lithium plating is not uniform across the electrode.

Peering inside a battery's undercurrents

At ALS Beamline 8.3.2, the researchers were able to tomographically measure slight expansions in the graphite particle sizes as lithiation increased. This provided them with information about the degree of lithiation in the graphite at specific times after fast charging. They also measured changes in the volume of the plated lithium



layer at the same time intervals. With this data, the researchers were able to determine the spatially resolved ionic current densities in the lithium and graphite (i_L and i_G , respectively).

The results revealed that, after charging the battery in 10 minutes, the average current densities decreased from 1.5 to 0.5 mA/cm² in about 20 min after charging stopped. Surprisingly, however, the range of the lithium current density was independent of time, with outliers generating alarming current densities as high as 25 mA/cm². In comparison, the current density required to charge the battery in 10 minutes was 18 mA/cm².

The persistence of these outliers provides a clue as to the origin of catastrophic failure in batteries at rest. However, much more work is needed before this approach can be used to develop improved safety protocols.



Top: Lithium-stripping current densities (i_L) —the ionic-current source—obtained from changes in the thickness of the plated lithium layer over time. Bottom: Corresponding graphitelithiation current densities (i_G) —the ionic-current sink—obtained from changes in the graphite state of charge.

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Publication: A.S. Ho, D.Y. Parkinson, S.E. Trask, A.N. Jansen, and N.P. Balsara, "Large Local Currents in a Lithium-Ion Battery during Rest after Fast Charging," *ACS Nano* **17**, 19180 (2023), doi:10.1021/acsnano.3c05470.

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Funding: US Department of Energy (DOE), Office of Energy Efficiency and Renewable Energy, Vehicle Techologies Office; and the National Science Foundation. Operation of the ALS is supported by the US DOE, Office of Science, Basic Energy Sciences program.

