Scientists reveal the root cause of rechargeable battery breakdown

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Battery research scientist Yaobin Xu inserts a sample into a transmission electron microscope to examine the function of a rechargeable battery. Credit: Andrea Starr | Pacific Northwest National Laboratory

For decades, researchers have assumed that the inevitable filmy buildup on electrodes inside rechargeable batteries is the driver of performance loss. Now, we know that view is backward.

The buildup of mossy or tree-like structured lithium metal deposits on battery electrodes is not the root cause of performance loss, but rather a side effect. The first direct measurement of the electrical properties at the boundary between the solid electrode and the liquid electrolyte inside a rechargeable battery is reported in *Nature Energy*.

The study, led by a research team at the Department of Energy's Pacific Northwest National Laboratory, shows that the so-called solid electrolyte interphase (SEI) is not an electronic insulator, as previously thought, but instead behaves like a semiconductor. The research solves the long-standing mystery of how SEI functions electrically during battery operation.

The findings have direct implications for designing longer-lasting batteries by tuning the physical and electrochemical properties of the liquid electrolyte, which is often referred to as the blood supply of an operating battery.

"A higher rate of electrical conductance induces a thicker SEI with intricate solid lithium forms, ultimately leading to inferior battery performance," said Chongmin Wang, a PNNL Laboratory Fellow and battery technology expert who co-led the study.

Micro-sized battery upends assumptions about how rechargeable batteries work

Researchers focus on this SEI layer, which is thinner than a sheet of tissue paper, because of its out-sized role in battery performance. This filmy mosaic selectively permits charged lithium ions to cross during discharge and controls movement of electrons that supply the battery's power.

When batteries are new, the SEI forms on the first charging cycle and ideally remains stable during the battery's expected lifespan. But a look inside an aging rechargeable battery often reveals substantial buildup of solid lithium on the negative electrodes. Battery researchers have assumed that this buildup causes the performance losses. Part of the reason for this guess work has been an inability to make measurements to test cause and effect.

In-situ transmission electron microscopy allows researchers to observe directly how the materials in a battery evolve at atomic and nanoscale, providing insight into rechargeable battery function. Credit: Andrea Starr | Pacific Northwest National Laboratory

Wang, along with co-lead of the study Wu Xu, a materials scientist of PNNL's Battery Materials and Systems Group, co-first authors Yaobin Xu and Hao Jia, and their colleagues at PNNL, Texas A&M University, and Lawrence Berkeley National Laboratory solved this problem by developing a new technique to directly measure electrical conduction across the SEI in an experimental system.

The team combined transmission electron microscopy with nanoscale manipulation of microfabricated metal needles inside the microscope. The researchers then measured the electrical properties of the SEI layer formed on either a copper or lithium metal with four different types of electrolytes.

The group's measurements revealed that as voltage increases in the battery, the SEI layer in all cases leaks electrons, making it semi-conductive.

Findings suggest carbon-containing molecules leak electrons, reducing battery life

Once they had recorded this semiconductor-like behavior, which had never been directly observed previously, they wanted to understand which components of the chemically complex SEI are responsible for the electron leakage.

"We found that the carbon-containing organic components of the SEI layer are prone to leaking electrons," Xu said.

The researchers concluded that minimizing the organic components in SEI would enable the batteries to have longer useful life.

"Even slight variations of the rate of conduction through the SEI can result in dramatic differences in efficiency and battery cycling stability," Wang added.

PNNL researchers Peiyuan Gao, Xia Cao, Phung M. L. Le, Mark H. Engelhard, Shuang Li and Ji-Guang Zhang also contributed to the research.

More information: Direct in situ measurements of electrical properties of solid–electrolyte interphase on lithium metal anodes, *Nature Energy* (2023). DOI: 10.1038/s41560-023-01361-1. www.nature.com/articles/s41560-023-01361-1

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