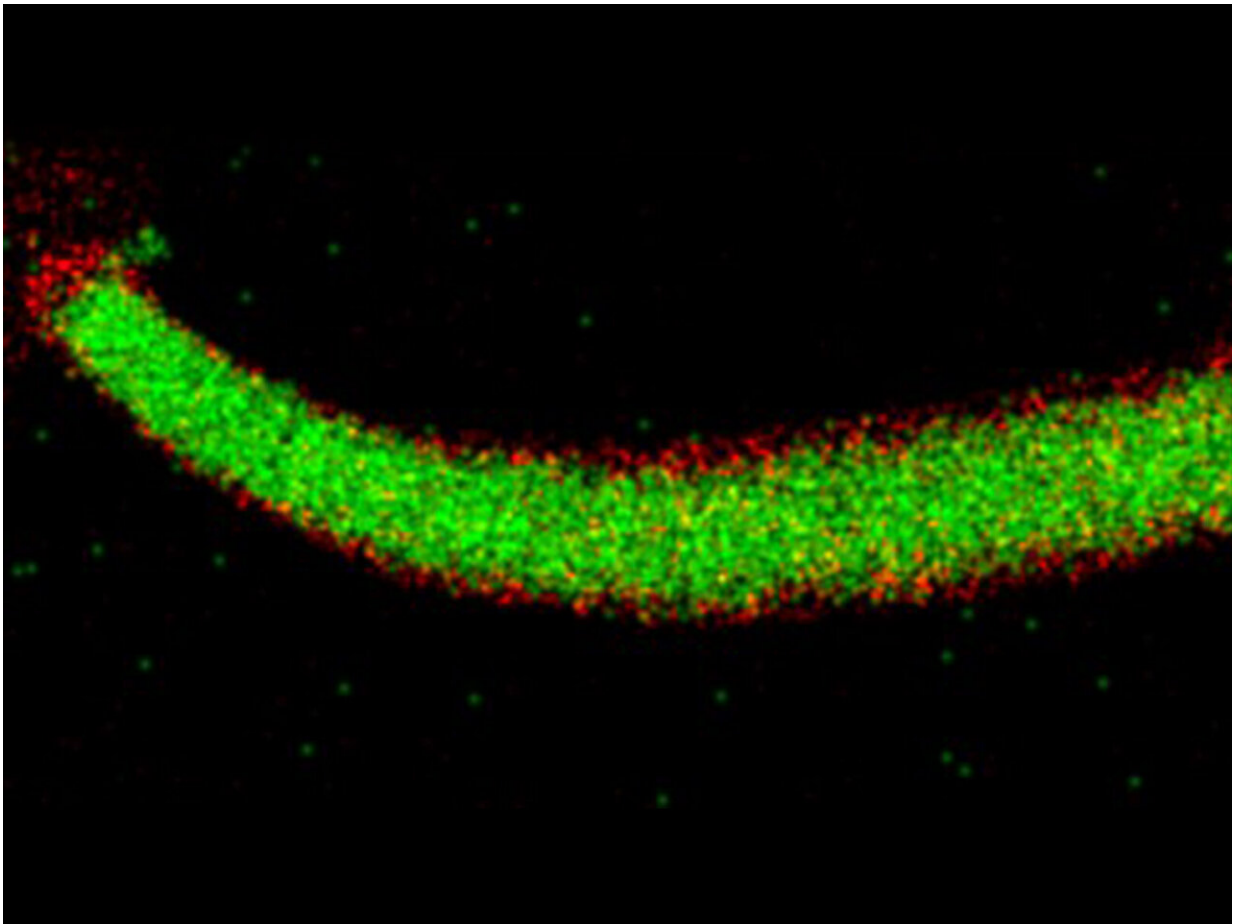


Silicon anodes muscle in on battery technology

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A silicon anode virtually intact after one cycle, with the silicon (green) clearly separate from a component of the solid electrolyte interphase (fluorine, in red).
Credit: Chongmin Wang | Pacific Northwest National Laboratory

Silicon is a staple of the digital revolution, shunting loads of signals on a device that's likely just inches from your eyes at this very moment.

Now, that same plentiful, cheap material is becoming a serious candidate for a big role in the burgeoning battery business. It's especially attractive because it's able to hold 10 times as much energy in an important part of a battery, the [anode](#), than widely used graphite.

But not so fast. While [silicon](#) has a swell reputation among scientists, the material itself swells when it's part of a battery. It swells so much that the anode flakes and cracks, causing the battery to lose its ability to hold a charge and ultimately to fail.

Now scientists have witnessed the process for the first time, an important step toward making silicon a viable choice that could improve the cost, performance and charging speed of batteries for electric vehicles as well as cell phones, laptops, smart watches and other gadgets.

"Many people have imagined what might be happening but no one had actually demonstrated it before," said Chongmin Wang, a scientist at the Department of Energy's Pacific Northwest National Laboratory. Wang is a corresponding author of the paper recently published in *Nature Nanotechnology*.

Of silicon anodes, peanut butter cups and packed airline passengers

Lithium ions are the energy currency in a [lithium-ion battery](#), traveling back and forth between two electrodes through liquid called electrolyte. When lithium ions enter an anode made of silicon, they muscle their way into the orderly structure, pushing the silicon atoms askew, like a stout airline passenger squeezing into the middle seat on a packed flight. This

"lithium squeeze" makes the anode swell to three or four times its original size.

When the lithium ions depart, things don't return to normal. Empty spaces known as vacancies remain. Displaced silicon atoms fill in many, but not all, of the vacancies, like passengers quickly taking back the empty space when the middle passenger heads for the restroom. But the lithium ions return, pushing their way in again. The process repeats as the lithium ions scoot back and forth between the anode and cathode, and the empty spaces in the silicon anode merge to form voids or gaps. These gaps translate to battery failure.

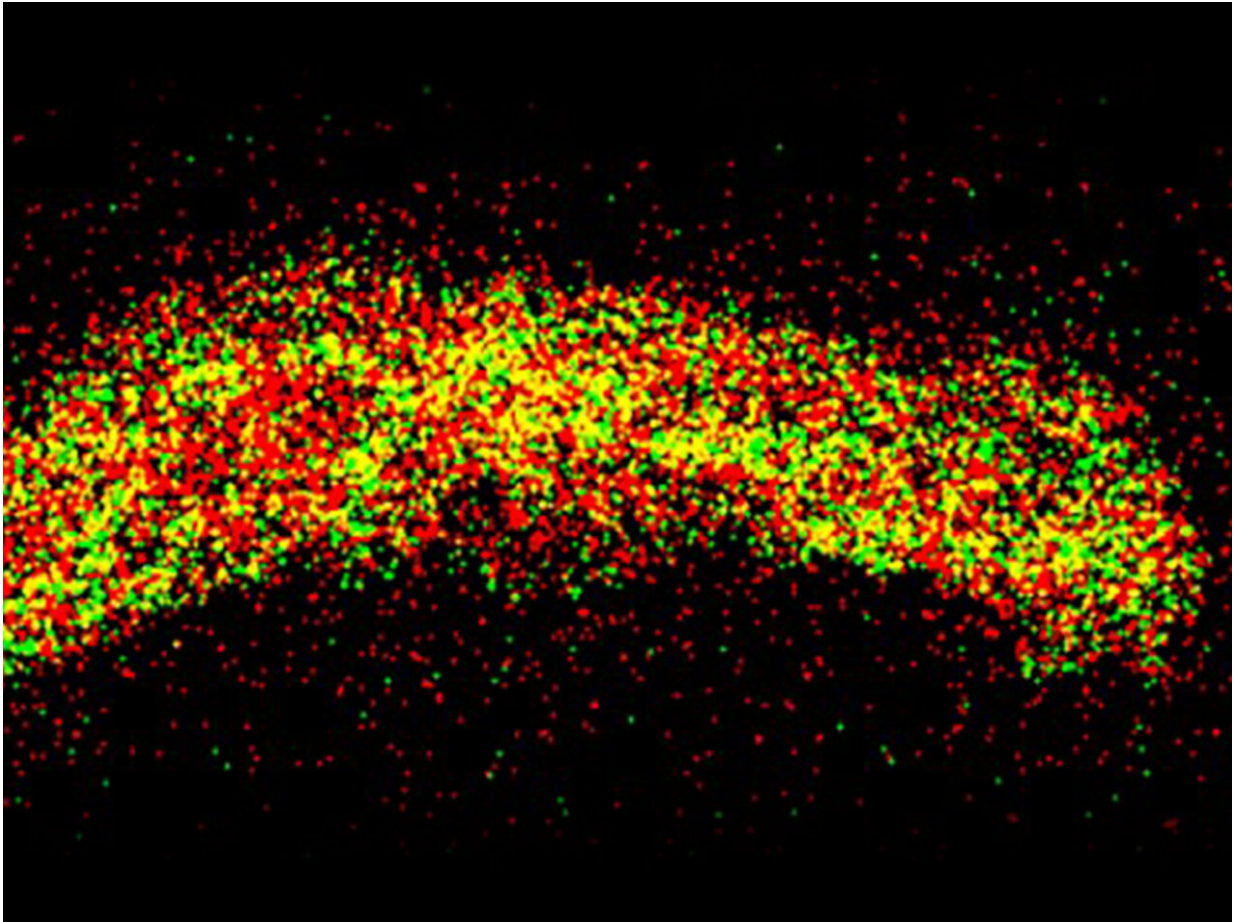
Scientists have known about the process for years, but they hadn't before witnessed precisely how it results in battery failure. Some have attributed the failure to the loss of silicon and lithium. Others have blamed the thickening of a key component known as the solid-electrolyte interphase or SEI. The SEI is a delicate structure at the edge of the anode that is an important gateway between the anode and the liquid electrolyte.

In its experiments, the team watched as the vacancies left by lithium ions in the silicon anode evolved into larger and larger gaps. Then they watched as the liquid electrolyte flowed into the gaps like tiny rivulets along a shoreline, infiltrating the silicon. This inflow allowed the SEI to develop in areas within the silicon where it shouldn't be, a molecular invader in a part of the battery where it doesn't belong.

That created dead zones, destroying the ability of the silicon to store lithium and ruining the anode.

Think of a peanut butter cup in pristine shape: The chocolate outside is distinct from the soft peanut butter inside. But if you hold it in your hand too long with too tight a grip, the outer shell softens and mixes with the soft chocolate inside. You're left with a single disordered mass whose

structure is changed irreversibly. You no longer have a true peanut butter cup. Likewise, after the electrolyte and the SEI infiltrate the silicon, scientists no longer have a workable anode.



A silicon anode after 100 cycles: The anode is barely recognizable as a silicon structure and is instead a mix of the silicon (green) and the fluorine (red) from the solid electrolyte interphase. Credit: Chongmin Wang | Pacific Northwest National Laboratory

The team witnessed this process begin immediately after just one battery cycle. After 36 cycles, the battery's ability to hold a charge had fallen

dramatically. After 100 cycles, the anode was ruined.

Exploring the promise of silicon anodes

Scientists are working on ways to protect the silicon from the electrolyte. Several groups, including scientists at PNNL, are developing coatings designed to act as gatekeepers, allowing lithium ions to go into and out of the anode while stopping other components of the electrolyte.

Scientists from several institutions pooled their expertise to do the work. Scientists at Los Alamos National Laboratory created the silicon nanowires used in the study. PNNL scientists worked together with counterparts at Thermo Fisher Scientific to modify a cryogenic transmission electron microscope to reduce the damage from the electrons used for imaging. And Penn State University scientists developed an algorithm to simulate the molecular action between the liquid and the silicon.

Altogether, the team used electrons to make ultra-high-resolution images of the process and then reconstructed the images in 3-D, similar to how physicians create a 3-D image of a patient's limb or organ.

"This work offers a clear roadmap for developing silicon as the anode for a high-capacity battery," said Wang.

More information: Chongmin Wang et al, Progressive growth of the solid–electrolyte interphase towards the Si anode interior causes capacity fading, *Nature Nanotechnology* (2021). [DOI: 10.1038/s41565-021-00947-8](https://doi.org/10.1038/s41565-021-00947-8)

Provided by Pacific Northwest National Laboratory

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