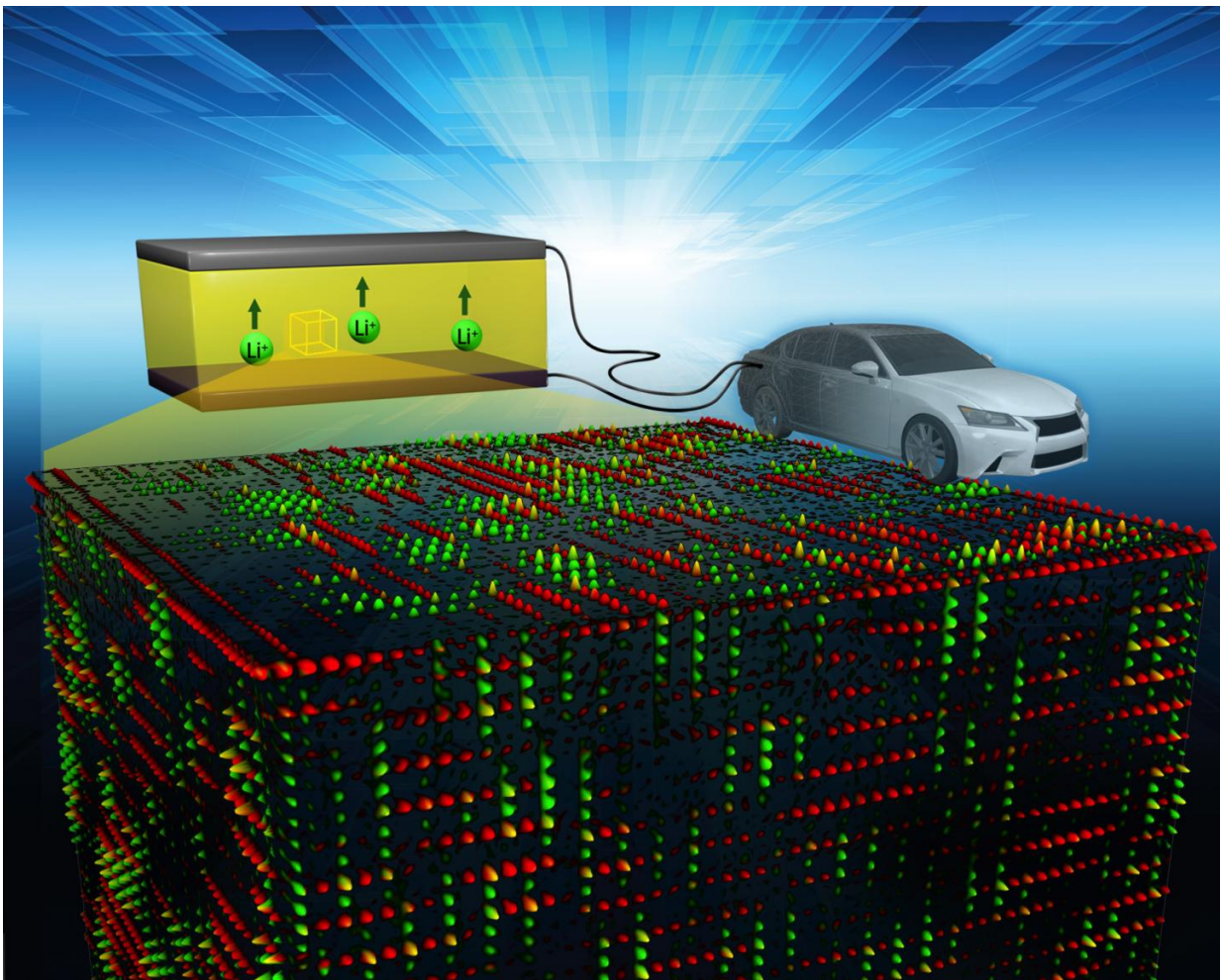


Speedy ion conduction in solid electrolytes clears road for advanced energy devices

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An ORNL-led research team found the key to fast ion conduction in a solid electrolyte. Tiny features maximize ion transport pathways, represented by red and green. Credit: Oak Ridge National Laboratory, U.S. Dept. of Energy

In a rechargeable battery, the electrolyte transports lithium ions from the negative to the positive electrode during discharging. The path of ionic flow reverses during recharging. The organic liquid electrolytes in commercial lithium-ion batteries are flammable and subject to leakage, making their large-scale application potentially problematic. Solid electrolytes, in contrast, overcome these challenges, but their ionic conductivity is typically low.

Now, a team led by the Department of Energy's Oak Ridge National Laboratory has used state-of-the-art microscopy to identify a previously undetected feature, about 5 billionths of a meter (nanometers) wide, in a solid electrolyte. The work experimentally verifies the importance of that feature to fast [ion transport](#), and corroborates the observations with theory. The new mechanism the researchers report in *Advanced Energy Materials* points out a new strategy for the design of highly conductive solid electrolytes.

"The solid electrolyte is one of the most important factors in enabling safe, high-power, high-energy, solid-state batteries," said first author Cheng Ma of ORNL, who conducted most of the study's experiments. "But currently the low conductivity has limited its applications."

ORNL's Miaofang Chi, the senior author, said, "Our work is basic science focused on how we can facilitate ion transport in solids. It is important to the design of fast ion conductors, not only for batteries, but also for other energy devices." These include supercapacitors and fuel cells.

To directly observe the atomic arrangement in the [solid electrolyte](#), the researchers used aberration-corrected scanning transmission electron microscopy to send electrons through a sample. To observe an extremely small feature in a three-dimensional (3D) material with a method that essentially provides a two-dimensional (2D) projection, they needed a

sample of extraordinary thinness. To prepare one, they relied on comprehensive materials processing and characterization capabilities of the Center for Nanophase Materials Sciences, a DOE Office of Science User Facility at ORNL.

"Usually the transmission electron microscopy specimen is 20 nanometers thick, but Ma developed a method to make the specimen ultra-thin (approximately 5 nanometers)," Chi said. "That was the key because such a thickness is comparable to the size of the hidden feature we finally resolved."

The researchers examined a prototype system called LLTO, shorthand for its lithium, lanthanum, titanium and oxygen building blocks. LLTO possesses the highest bulk conductivity among oxide systems.

In this material, lithium ions move fastest in the planar 2D pathways resulting from alternating stacks of atomic layers rich in either lanthanum or lithium. The ORNL-led team was the first to see that, without hurting this superior 2D transport, tiny domains, or fine features approximately 5 to 10 nanometers wide, throughout the 3D material provided more directions in which lithium ions could move. The domains looked like sets of shelves stacked at right angles to others. The smaller the shelves, the easier it was for ions to flow in the direction of an applied current.

ORNL's Yongqiang Cheng and Bobby Sumpter performed molecular dynamics simulations that corroborated the experimental findings.

Previously, scientists looked at the atomic structure of the simplest repeating unit of a crystal—called a unit cell—and rearranged its atoms or introduced different elements to see how they could facilitate ion transport. Unit cells are typically less than 1 nanometer wide. In the material that the ORNL scientists studied for this paper, the unit cell is

nearly half a nanometer. The team's unexpected finding—that fine features, of only a few nanometers and traversing a few unit cells, can maximize the number of ionic transport pathways—provides new perspective.

"The finding adds a new criterion," Chi said. "This largely overlooked length scale could be the key to fast ionic conduction."

Researchers will need to consider phenomena on the order of several nanometers when designing materials for fast ion conduction.

Ma agreed. "The prototype material has high ionic conductivity because not only does it maintain unit-cell structure, but also it adds this fine feature, which underpins 3D pathways," Ma said. "We're not saying that we shouldn't be looking at the unit-cell scale. We're saying that in addition to the unit cell scale, we should also be looking at the scale of several [unit cells](#). Sometimes that outweighs the importance of one unit cell."

For several decades, when researchers had no explanation for certain material behaviors, they speculated phenomena transcending one unit cell could be at play. But they never saw evidence. "This is the first time we proved it experimentally," Ma said. "This is a direct observation, so it is the most solid evidence."

The title of the paper is "Mesoscopic Framework Enables Facile Ionic Transport in Solid Electrolytes for Li-ion Batteries."

More information: Cheng Ma et al, Mesoscopic Framework Enables Facile Ionic Transport in Solid Electrolytes for Li Batteries, *Advanced Energy Materials* (2016). [DOI: 10.1002/aenm.201600053](https://doi.org/10.1002/aenm.201600053)

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