Study shows similarity between solid state and liquid state electrolytes used in batteries

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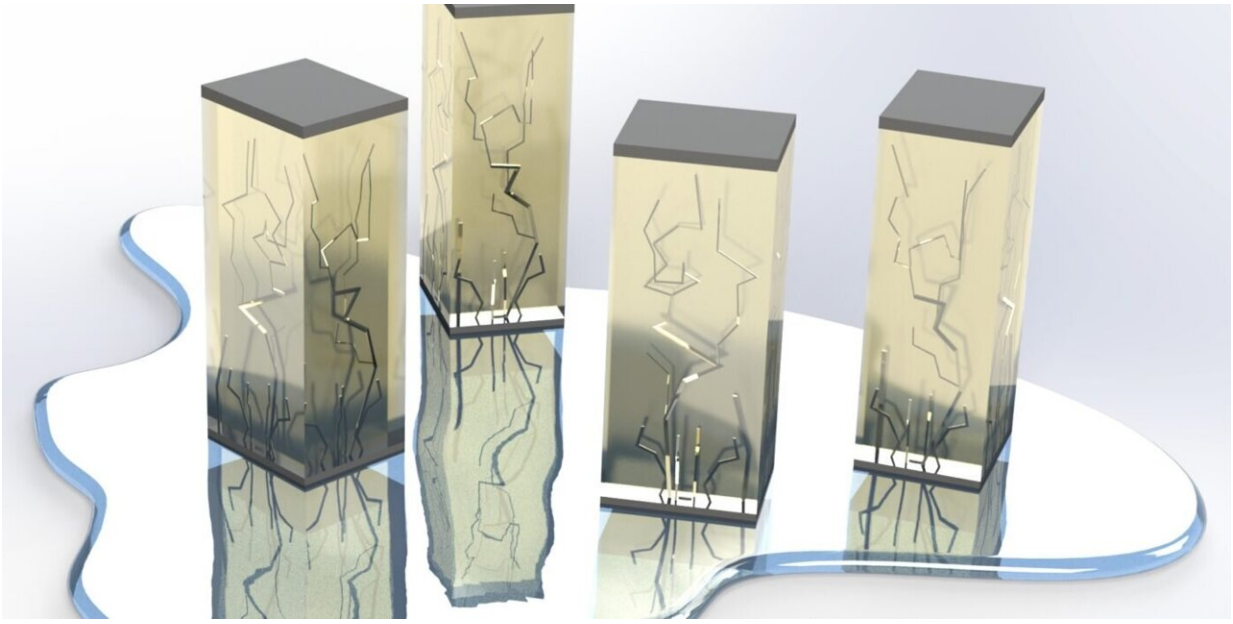

Ceramic solid electrolyte cells (vertical rectangular shapes) with dendrites (the lightning-like dark structures inside the rectangles) growing inside them from the bottom to the top. These solid electrolytes are floating on a liquid (blue puddle) which represents a liquid electrolyte. The reflections of the solid electrolytes in the blue liquid, particularly the dark dendrites, show the similarity in the dendrite initiation process in both the liquid and solid. Credit: Rajeev Gopal/Bai lab

New research at the McKelvey School of Engineering at Washington University in St. Louis is the first to show that a solid-state electrolyte has a high level of similarity to liquid electrolytes, which is good news

for designing safer and more efficient solid-state batteries based on reliable mechanistic knowledge.

"Our results reveal surprising similarities between the liquid and the solid electrolytes, and that allows us to borrow some ideas from the successful liquid electrolytes to help our design of the solid electrolytes," said Peng Bai, an assistant professor of energy, environmental and chemical engineering. "Before our work, solid electrolytes, at least the ceramic ones we studied here, are considered distinctly different from their liquid counterparts."

Batteries power so much of our lives, so finding new improvements will have a drastic societal impact, Bai said.

A promising route is the development of a full solid-state battery. A key component is the electrolyte in the center of the battery, which allows ion movement between the electrodes. Here, the traditionally used liquid electrolyte is replaced with a solid and coupled to a metal electrode. This not only boosts the amount of energy stored but also leads to a potentially safer battery.

However, an increasing number of reports about solid-state batteries tell a story of a key barrier, known as the critical current density (CCD), beyond which small tree-like structures called dendrites grow, resulting in battery failure. These reported CCDs are relatively low, which hinders fast charging and compromises further development of solid-state batteries.

"The CCD of solid-state electrolytes is a mystery. We are working to reveal why it exists, what its true physics are and how it changes under different operating conditions," said Bai, the principal investigator of this project and corresponding author of the paper published April 12 in *ACS Energy Letters*.

"Our discovery shows that the magnitude of CCD is linked to the thickness of the solid electrolyte, similar to the limiting current in liquid electrolytes that are well-known to have thickness dependence," he said. "If you can make the solid electrolyte thin enough, we can get away from this CCD issue, therefore avoiding dendrite growths and the internal short-circuiting."

Experimental innovations involved in this study include taking a standard pellet, a small circular disc fully densified by controlled sintering of ceramic powders, and precisely cutting it into multiple smaller pieces. The pieces were then tested using an electroanalytical technique different from those commonly used.

"These child samples from the same mother pellet were almost identical," Bai said. "Testing hundreds of these ideally consistent miniature samples made the results we obtained more reliable and the statistics more meaningful."

Rajeev Gopal, a doctoral student in Bai's lab and first author of the paper, said this study can be a real difference-maker.

"Our work can shed light on the mysterious phenomenon of dendrite initiation at the CCD," Gopal said. "The statistical trends we unveiled here will help predict and ultimately mitigate this type of growth, increasing the feasibility of these electrolytes in real-world batteries."

More information: Rajeev Gopal et al, Transient Polarization and Dendrite Initiation Dynamics in Ceramic Electrolytes, *ACS Energy Letters* (2023). DOI: 10.1021/acsenergylett.3c00499

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