Using quantum methods to predict next-gen lithium-metal battery reactivity

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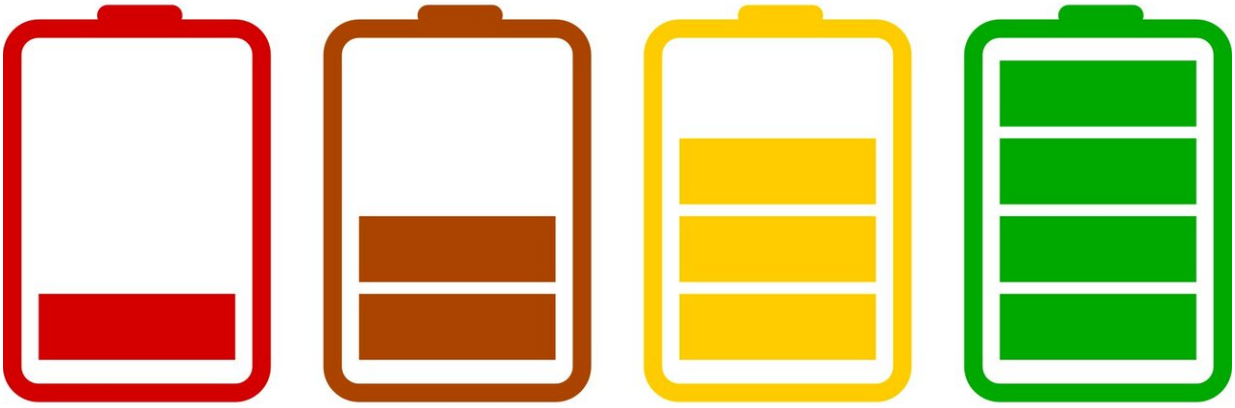

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Lithium-metal (Li-metal) batteries show great potential for packing more significant amounts of energy than the current lithium-ion batteries. For example, a Li-metal electric battery in a car could travel more miles, and a Li-metal phone battery could have longer battery life. However, the metal surface of Li-metal batteries is highly reactive, and there is limited understanding of the chemistry of these reactions.

Dr. Perla Balbuena, professor in the Artie McFerrin Department of Chemical Engineering at Texas A&M University, is using quantum chemical methods to track specific reactions that occur on the surfaces inside Li-metal batteries. Understanding Li-metal battery reactions and predicting products will enhance usability by decreasing their reactivity.

This research was recently published in the American Chemical Society's *ACS Applied Materials & Interfaces* journal and was co-authored by graduate student Dacheng Kuai from the Department of Chemistry at Texas A&M.

"We need to understand what type of reactions happen, how to slow down the reactions, what the components are, what the morphology of the evolving products is and how the ions and electrons move through the surface," said Balbuena. "Understanding these critical issues will allow us to commercialize Li-metal batteries in the near future."

When Li-metal batteries are manufactured, a thin film forms on the anode, commonly referred to as solid-electrolyte interphase (SEI). This film is made of multiple components and produced by electrolyte decomposition. The chemical makeup of the SEI is critical for ensuring peak performance from the battery and extending its lifespan. Through experimental efforts, theoretical predictions can reveal the details in this phenomenon at the atomistic and electronic levels.

In this study, the researchers targeted a polymer that develops due to electrolyte reactions on the battery's internal surfaces. Pinpointing this specific polymer reaction is challenging but necessary to optimize the SEI. The researchers simulated the interface at the atomistic level and solved accurate quantum chemical equations to map a time evolution of the polymer formation reaction.

"What differentiates this research is starting from the microscopic-level

description and letting the system evolve according to its electronic redistribution upon chemical reaction," Balbuena said. "There are many experimental techniques that can follow and monitor the reactions, but they're challenging. With this simulation, we can get new insights. We isolate the part of the system that is responsible for important chemical events. We follow that specific group of molecules and analyze the reactions spontaneously occurring at the surface of electrodes."

Unique to this research, the computational tools used can determine the minimum energy configurations and the arrangement of the molecules during the reaction, thus charting the reaction from beginning to end.

The researchers found that the species polymerizing in the SEI could be beneficial for Li-metal batteries because they can aid in controlling the level of reactivity of the battery materials.

"We are pleased about the results, as they provide insight into what could happen when using real electrodes," said Balbuena.

These findings illustrate the use of computational tools that can contribute to creating batteries that are more friendly to the environment, have longer lifespans and are cheaper to produce. As better chemistries evolve, Balbuena hopes the methodologies found in her research will be helpful for years to come.

"This research can be a driving force for batteries in a greener, more efficient direction," she said. "I know that this work will be helpful 10 years from now because 10 years ago, we made our initial contributions on Li-ion batteries and our findings helped on the development of today's successful technology. It is a cycle of continuous improvement."

More information: Dacheng Kuai et al, Solvent Degradation and Polymerization in the Li-Metal Battery: Organic-Phase Formation in

Solid-Electrolyte Interphases, *ACS Applied Materials & Interfaces* (2022). DOI: 10.1021/acsami.1c20487

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