Scientists explore electronic properties of liquid electrolytes for energy technologies

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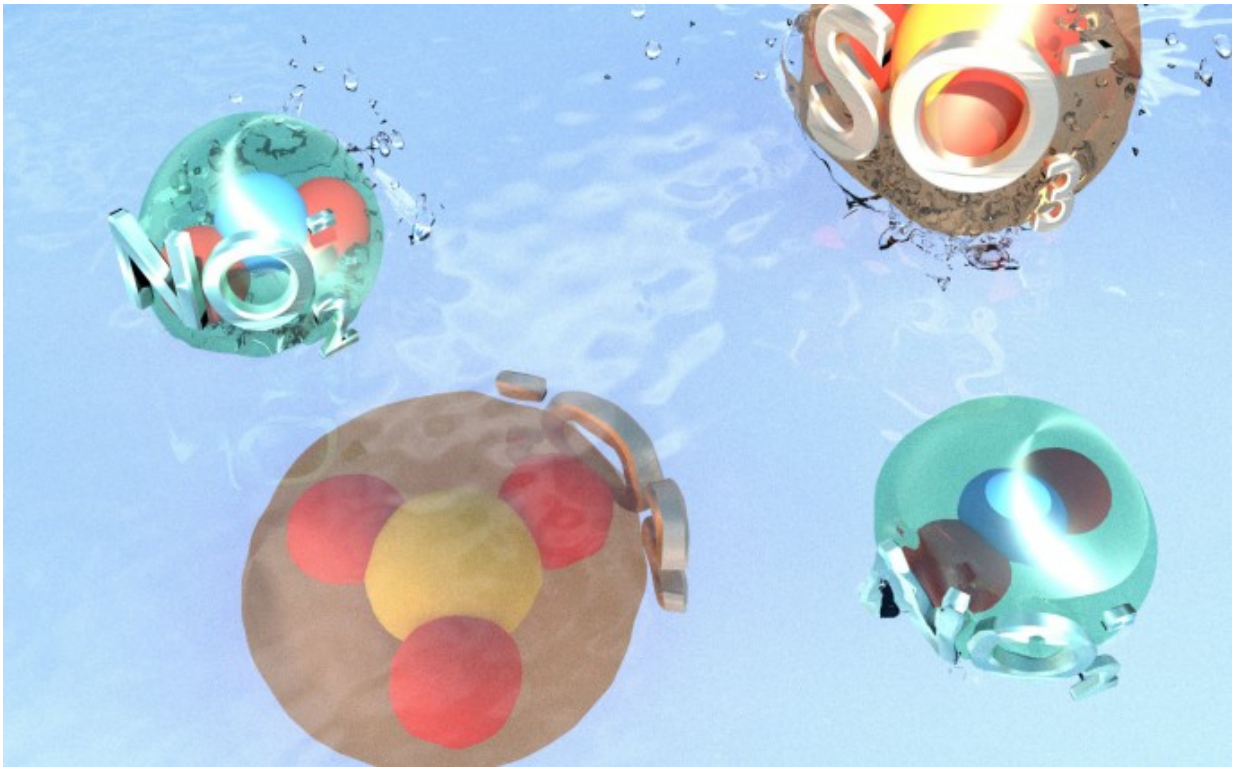

This artist's impression shows various solvated ions in liquid water. Credit: Nicholas Brawan/Institute for Molecular Engineering, University of Chicago and Tuan Anh Pham/LLNL

Liquid electrolytes are essential components in a variety of emerging energy technologies, including batteries, supercapacitors and solar-to-fuel devices.

"To predict and optimize the performance of these devices, a detailed understanding of the electrolytes, particularly their electronic properties such as the ionization potential and electron affinity, is critical," said Anh Pham, a Lawrence Livermore National Laboratory (LLNL) Lawrence Fellow in the Quantum Simulations Group, and the lead author of a paper in the June 23 edition of Science Advances.

As an example, Pham pointed to how proper energy alignment at the electrode-electrolyte interface of photoelectrochemical (PEC) cells is key to achieving efficient hydrogen production.

Pham, along with LLNL researcher Eric Schwegler, Robert Seidel and Steven Bradforth from the University of Southern California, and Marco Govoni and Giulia Galli from the Argonne National Laboratory and the University of Chicago, have presented an experimentally validated simulation strategy for computing the electronic properties of aqueous electrolytes.

The team directly simulated and measured the electronic excitation of various solvated ions in liquid water. By combining first-principles molecular dynamics simulations with state-of-the-art electronic structure methods, the team could predict the excitation energies of the solvents and solutes, such as the ionization potentials of the solvated ions. The team also demonstrated that the coupling of this theoretical framework with advanced spectroscopy techniques provides a powerful tool for the identification of chemical species and reactions that occur in solutions.

The new method opens up the possibility to predict the electronic response in complex electrolytes for a range of applications. For example, the research provided a theoretical foundation for understanding and engineering the electronic properties of liquid electrolytes in PEC cells for hydrogen production and ionic liquid for batteries.

"The proposed computational framework is general and applicable to non-metallic liquids, offering great promise in understanding and engineering solutions and liquid electrolytes for a variety of important energy technologies," Pham said.

In a broader sense, the new simulation capability represents the first step toward a unified method for the simulation of realistic, heterogeneous interfaces in electrochemical systems.

More information: Tuan Anh Pham et al. Electronic structure of aqueous solutions: Bridging the gap between theory and experiments, *Science Advances* (2017). DOI: 10.1126/sciadv.1603210

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