Introduction: computational electrochemistry

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Editorial

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Advancing our understanding of electrochemical phenomena, and ultimately improving the operation and design of electrochemical systems, is aided by the use of advanced computational tools and theories that enable researchers to deconvolute complex, interacting phenomena at multiple length and time scales. Electrochemical phenomena range from atomistic and picosecond interactions and reactions within double layers up to meters and years for cells and lifetime predictions. Bridging these scales and complex nonlinear interactions is a daunting task, but advancements in understanding and computation have made it more attainable than ever. It is the hope that melding of computational power, data informatics, and experimental studies can become intertwined in the future, where virtual representations of real systems can greatly advance the scientific discovery process. In this thematic issue, experts from around the world discuss the current state of the art approaches and understanding in computational electrochemistry.

At the smallest length scales of individual reaction events and electron transfer, an understanding of the importance of electronic structure and, in particular, the electrochemical potential is critical. The review of Santos and Schmickler examines such electron transfer events at different electrode materials, elucidating the principles governing charge transfer. The review of Hammes-Schiffer and colleagues provides a similar level of insight on the critical issues of proton-coupled electron transfer, a process that is common in many molecular systems and an expanding area for electrochemical research in general. In a detailed review of atomistic simulations of the interface, Schwarz and colleagues explore ways to improve realism and accuracy and move toward more predictive simulations.

While new materials are continuously synthesized, computational approaches are now being leveraged to understand and guide such design, in both traditional and nontraditional electrochemical reactions. Examining specific electrochemically active materials, the review of Liu and colleagues explores atomistic understanding for the newly utilized two-dimensional electrocatalysts. This review is complemented by the multiscale analysis and techniques for solid-state polymer electrolytes provided by Paddison and colleagues. Here, it is shown again how the material and polymer interactions are important in such solid-state proton conductors, where true understanding and modeling of these complex systems is still relatively in its infancy.

There is a growing awareness of the important roles that electrolyte species and solvent play in electrochemical reactions. The reviews by Groß and Sakong and Ringe and colleagues both tackle the importance of solvent and water, but from different perspectives. Similarly, interest in the importance of the electrochemical interface, which drives almost all electrochemical reactions, is seeing a resurgence, especially in the formation of the double layer, which ions must traverse to react, under different conditions. The review by Wu examines double layer structures and behavior from a fundamental perspective and provides insight into the many coupled material interactions and processes. In a related review, Salanne and colleagues explore double layers in the context of capacitors, another heralded electrochemical energy storage device. This review explores the governing design criteria and equations and methodologies for describing their behavior.

Advances in theory have also enabled better approaches toward analyzing vast data sets, but physics-based understanding is critical. In the review by Franco and colleagues, they tackle the challenge of bridging scales and the question of whether artificial intelligence and machine learning are good approaches for the nuances of electrochemical devices and, specifically, batteries. In another multiscale analysis, Zhang and colleagues examine how different techniques can be used and their inherent trade-offs for electrolytes in rechargeable batteries.

Computational approaches at the continuum scale provide methods to examine multiple interactions and phenomena, and they predict and guide overall material and integration methodologies. While such models have been utilized for decades, especially in the battery and fuel-cell realms, only now are we being able to expand their reach into coupled length and time scales as well as more complex and nuanced interactions at interfaces and reactions. To this end, Weber and colleagues examine how modeling can be conducted for electrochemical synthesis flow cells in general. This review is complemented by the multiscale modeling review of López and colleagues, who look at electrochemical CO_2 reduction and provide a more focused view of the reaction pathways. Finally, critical for exploring electrochemical systems is the interrogation using such techniques as electrochemical impedance spectroscopy. The review by Orazem and colleagues explores how computational electrochemistry is

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critical for interpreting such complex signals and for a variety of different cases and conditions.

In conclusion, computational electrochemistry is more accessible than ever, but one needs to ensure that the correct approaches are being used to solve specific problems and questions. This thematic issue is timely in that newfound approaches toward machine learning, big and small data problems, and the importance of provenance and metadata are in a period of discovery and renaissance. It is our hope that the issue acts as a guide and reference for both those new to and those familiar with the field. Finally, we would like to give our heartfelt thanks to our colleagues that enthusiastically agreed to contribute excellent reviews to this thematic issue, even during the COVID pandemic. From our perspective, the future looks more promising than ever for computational approaches in elucidating electrochemical phenomena.

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