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GUEST EDITORIAL

Electrochemical Energy Storage

Paul Barbara, a Senior Editor of *Accounts of Chemical Research* for over 15 years was fascinated with light–matter interactions (see the Nano and Molecular Science and Technology Special Issue Honoring Paul Barbara, November 20, 2012). Throughout his career, he sought to utilize both experimental and theoretical methods to understand the scientific underpinnings of interfacial charge separation and transfer processes of nanostructured materials for energy needs. Paul believed that the mechanistic understanding of interfacial charge separation and transfer process in nanomaterials was in its infancy, without a broadly accepted theoretical description. In 2009, he led a successful effort for a DOE Energy Frontier Research Center, of which he became the director. It was at this time that Paul encouraged us to organize a special issue of *Accounts* focused on Electrochemical Energy Storage (EES) that highlighted new multidisciplinary approaches for the study of ion-coupled electron transfer processes at interfaces with a closer pairing between theory and modeling that are vital to moving this field forward. Today we are at the beginning of full-scale adoption of the most promising EES systems for hybrid electric vehicle (HEV), plug-in hybrid electric vehicle (PHEV), and electric vehicle (EV) applications. Advanced EES systems are also vital for achieving the full potential of renewable energy sources (e.g., solar and wind) as part of our electric distribution grid. Energy stored in these EES systems occurs through the movement of ions between the cathode and the anode electrodes. Both the anode and cathode are actually complex composites containing not only active materials for ion storage but also other components, namely, a polymeric binding agent and conductive carbon filler, which help facilitate ion and electron transfer processes. EES devices constructed from these advanced materials offer exceptional promise for higher energy density (W·h/kg) and power density (W/kg) applications compared with existing commercial technologies. Depending on the specific choice of materials for the EES system the voltage, capacity, life, and safety can vary dramatically. Unfortunately, the practical energy density of the most promising materials is approximately 25–50% of its theoretical value due to inefficiencies related to the mass or volume of the inert components (binders and conductive fillers), irreversible losses associated with phase and volume changes, and incomplete utilization of the active material due to poor ionic or electronic communication. A key requirement for advancing EES technology is to disentangle the factors that govern ion-coupled electron transfer at these complex interfaces and to clarify stability and degradation processes.

In this Special Issue, a collection of 17 Accounts on EES systems provide a detailed examination of a wide variety of research activities and methods within the field. These include the synthesis and exploration of nanostructured materials as they offer enticing new prospects for discovering breakthrough materials and transforming energy storage concepts. The development of new spectroscopic characterization tools is highlighted in an effort to elucidate factors that govern charge transfer reactivity in model EES materials by exploring the influence of size, morphology, and electrode configuration on ion-coupled electron transfer, volume expansion/contraction processes, and phase formation mechanisms. Innovative computational and atomic-level theoretical methods are provided in the context of understanding transport dynamics and electronic structure.

As will be evident from the Accounts, there have been tremendous advances in revealing the basic atomistic and electronic mechanisms of EES systems, both with the development of truly exceptional experimental and computational methods and with the ever expanding collection of characterization tools. We hope that these Accounts will present new strategies for enhancing the storage and transport properties in existing EES systems, as well as provide the conceptual basis for resolving outstanding fundamental questions and ultimately lead to next-generation materials with higher energy and power densities that will revolutionize the field of electrochemical energy storage.

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