

Understanding and Controlling Electrochemistry for Fuel Cells and Batteries

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This talk addresses both the Oxygen Reduction Reaction (ORR) – a central reaction important in fuel cells and reactivity associated with battery electrodes. In the former, we develop design rules for effective new ORR catalysts and actualize these by synthesizing and characterizing Cu-based catalysts for this reaction. We show that ligands which are effective to make good ORR catalysts also have a positive role in other chemistries, particularly those involving CO₂ reduction and the oxygen evolution reaction. Finally, we develop a new platform to more generally address proton-coupled electron transfer (PCET) reactions, of which the ORR is one. We use this platform to vary the rate of proton transfer to an ORR catalyst, and show that product distribution from the ORR can be tuned by proton availability, a feature which has general implications for other catalysts performing this reaction

Relevant to batteries, we present a new technique to probe the electrochemically-induced mechanics of electrodes by calculating the electrochemical stiffness of electrodes via coordinated *in situ* stress and strain measurements. Through the electrochemical stiffness, we elucidate inherent and rate-dependent mechanical responses of graphitic battery electrodes. We find that stress and strain are asynchronous. In particular, stress development is found to lead strain development as different graphite-lithium intercalation compounds are formed. These measurements provide a new paradigm for understanding mechanical effects in intercalation systems, such as batteries. Electrochemical stiffness measurements provide new insights into the origin of the rate-dependent chemo-mechanical degradations, and provide a probe to evaluate advanced battery electrodes.