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Seminar Series

Presents

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Quantum and Classical Modeling Approaches to Catalyst Design

Innovative catalytic systems will shape our energy and environmental future, as such systems will play a central role in improving the efficiency of low-carbon technologies. Successful heterogeneous catalysts employed in these technologies inevitably feature complex surface morphologies, as multiple functionalities are required to achieve conversion that is both active and selective. To aid the design of such catalysts, computational guidance derived from both quantum and classical mechanics is essential for bridging design challenges that span electronic and atomistic scales. This presentation will demonstrate the application of quantum and classical simulation techniques to unravel the interdependence of surface stability and activity impacting the performance of two important classes of catalytic systems. The first class features metal nanoclusters on oxide supports, which are ubiquitously present in technologies seeking to effectively utilize fossil resources. This study investigates the interaction between Pd nanoclusters and CeO_2 support morphologies, as the formation of mixed Pd-O-Ce surface phases are linked to rapid hydrocarbon activation. Quantum and classical simulations demonstrate that Pd mixing in the lattice structure of the CeO_2 support creates meta-stable $Pd4+$ surface sites that are highly reactive toward C-H bond scission. The second class of catalysts features semiconductor surfaces that enable the photo-electrochemical reduction of CO_2, which has recently received significant attention as a promising route for generating carbon-neutral fuels via sunlight. Experimental evidence suggests that pyridine (Py) is an effective co-catalyst during CO_2 reduction over numerous semiconductor materials. Efforts to identify the responsible Py-catalyzed mechanism are complicated by surface reconstructions that occur during operation, which alter both the nature of adsorption sites present on the surface and the band edge alignment of the semiconductor. This energy alignment determines the direction of electron transfer at the electrode/electrolyte interface, and consequently determines which reaction steps can occur spontaneously at the catalyst surface. Quantum-based computational techniques are used to predict surface reconstructions that are likely to occur during operation, thus establishing the surface structure and corresponding energy level alignment under operating conditions. Knowledge of this alignment is then used to evaluate the thermodynamic feasibility of proposed elementary steps, thus lending insight into the Py-catalyzed mechanism. Together, these studies demonstrate the power of both quantum and classical approaches for assessing simultaneous interactions between multiple components in complex catalytic systems.

About the Speaker

Thomas P. Senftle completed his BSc in Chemical Engineering at Notre Dame, where he conducted research on ionic liquids for CO_2 capture under the guidance of Prof. William Schneider. He completed his PhD in Chemical Engineering at Penn State with joint advisors Prof. Michael Janik and Prof. Adri van Duin, where his thesis included the computational design, at both quantum and classical levels, of oxide-supported noble metal catalysts. He is presently a postdoctoral associate at Princeton University working in the research lab of Prof. Emily Carter, with a focus on the ab initio design of semiconductors for photo-electrochemical applications. His current research interests include both the development and application of hybrid simulation techniques for modeling multi-component catalytic systems at quantum and classical scales.

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