

Physics Colloquium

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“Elucidating active sites and reaction mechanisms in heterogeneous catalytic conversion”

Heterogeneous catalysis is a prominent means to upgrade various carbon sources to chemicals and energy carriers. Designing an optimal catalyst is a multifaceted problem requiring analysis and decision making at multiple levels - from understanding molecular events to elucidating complex reaction mechanisms, identifying suitable catalysts, and optimizing reactor performance.

Set in this context, my talk will showcase two examples of how we apply ab initio methods (in particular density functional theory, or DFT) coupled with machine learning and cheminformatics to understand the active sites (i.e. where the reaction occurs on a catalyst) and the mechanism (the sequence of steps the reactants undergoes to form the desired product) of different catalytic reaction systems. In the first example, we will consider the reduction of carbon dioxide on molybdenum sulfide catalyst in the presence of hydrogen sulfide. Shale gas and natural gas reserves worldwide contain varying amounts of acidic gases such as CO₂ and H₂S. Current methods involve energy-intensive separation of CO₂ and H₂S before further downstream processing of these wastes (and CO₂ is often released to the atmosphere); our vision here is to develop techniques to convert CO₂ to more value-added products using a catalyst that is sulfur tolerant so that the costly separation step is avoided. In collaboration with our experimental partners (Baltrusaitis group at Lehigh), we employed DFT to understand how the catalyst evolves with varying process conditions and that the active site concentration is directly dependent on the chemical potential of the bulk sulfur and hydrogen. We employed this information in explaining experimental observations that H₂S inhibits CO₂ reduction and further identified the plausible reaction mechanism for this chemistry.

Several catalytic processes tend to be complex in that the underlying reaction system comprises of several hundreds to thousands of species and reactions, thereby precluding a manual analysis that is comprehensive and error-free. In the second example, I will present our rule-based computational tool, Rule Input Network Generator (RING), to construct and analyze the mechanism of such complex reaction networks. RING can construct an exhaustive network of all plausible reactions and species of a system and identify reaction pathways forming a specific product through rule-based queries and “prune” out energetically infeasible pathways. I will demonstrate the utility of this tool through examples involving mechanism identification in polyol conversion over transition metals.

Srinivas Rangarajan is an Assistant Professor and the Dolores and William Schiesser Faculty Fellow in Chemical and Biomolecular Engineering at Lehigh University. His area of expertise is in computational catalysis and materials design. He employs a variety of computational tools, ranging from ab initio to machine learning, microkinetic modeling, optimization, graph theory, and cheminformatics to elucidate catalytic reaction mechanisms and design new functional materials. Srinivas's research is (and has been) supported by the National Science Foundation (including the CAREER award), the ACS Petroleum Research Fund (especially the Doctoral New Investigator Award), and the Commonwealth of Pennsylvania (PITA).

Thursday, February 3, in LL 316 at 4:25 PM
For Zoom participation, please see information below:
Meeting ID: 972 1274 7894
Passcode: 631869