Elucidating catalytic events: from molecular interactions to complex reaction chemistries

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Heterogeneous catalysis is a prominent means to upgrade carbon sources to chemicals and energy carriers. Designing an "optimal" catalytic system is an open multi-faceted challenge requiring analysis and decision making at many levels – from understanding molecular events to elucidating complex reaction networks, identifying suitable catalysts, and optimizing reactor performance. My research, to this end, brings disparate yet complementary tools from computational chemistry, process systems engineering, and cheminformatics.

In this talk, I will focus on these multiple facets. First, I will show, using density functional theory, how studying molecular events such as adsorption can be used to derive structure-property relationships that elicit details of catalytic reactivity and selectivity. Specifically, I will discuss how electronic structure calculations of adsorption of organonitrogen and organosulfur compounds present in vacuum gasoil on the active sites of industrial hydrotreating catalysts gives insights into the inhibition of hydrodesulfurization (HDS) chemistry and site requirements for a maximum-HDS-minimum-inhibition catalyst. Second, I will discuss how designing optimal catalysts require mathematical tools to identify catalytic parameters in conjunction with inputs from (and feedback to) experiments and computational chemistry. I will present new methods that leverage nonlinear optimization approaches to (i) rigorously identify active sites and surface environment of catalysts, and (ii) identify "optimal" energetic parameters that maximize observable catalytic properties such as rate, selectivity, etc. Third, I will present a new rule-based computational tool, Rule Input Network Generator (RING), to construct and analyze the mechanisms of complex reaction networks. Such networks are ubiquitous and comprise of several hundreds to thousands of reactions and species. 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.

Bio

Srinivas is a postdoctoral researcher working with Profs. Manos Mavrikakis and Christos Maravelias at University of Wisconsin-Madison. He obtained his PhD in Chemical Engineering at University of Minnesota in 2013 under the advisorship of Profs. Prodromos Daoutidis and Aditya Bhan. Srinivas is originally from India and did his undergraduate studies at Indian Institute of Technology Madras. His research interests are in the areas of computational catalysis, complex reaction networks, and nonlinear optimization.