

The heart of darkness (in heterogeneous catalysis)

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The science and practice of heterogeneous catalysis over the past century has been critically enabling for society – nearly every molecule of gasoline, diesel, and other energy carriers is prepared in molecular encounters on catalytic surfaces, >99.99% removal of NO_x from automotive exhaust is achieved by modern catalytic converters, and the production of practically all plastics, adhesives, paints, etc., relies on heterogeneous catalysts. Frequently, the transition from concepts and learnings to practice comes down to the faculty of catalytic processes associated with unselective or deactivation side paths. We illustrate recent advances in our understanding of the mechanisms and site requirements in catalysis by surfaces, with emphasis on concepts that tackle ubiquitous selectivity and lifetime challenges, based on case studies of two catalytic systems deployed in industrial practice.

The first part of this presentation will describe the mechanistic origins of over-oxidation and C-C bond scission products in the partial oxidation of propylene to acrolein on mixed metal oxide catalysts. We combine transient kinetic studies, co-feed experiments of aldehydes and carboxylic acids formed as byproducts in propylene oxidation, and isotopic-labeling studies to elucidate the reaction mechanisms, identify the existence and involvement of relevant surface intermediates, and develop an extensive reaction network describing the formation of all C₂ – C₆ products (> 20 C₂-C₆ products are formed in this chemistry at carbon selectivity as low as 0.001%), and illustrate the underlying mechanisms for C-C bond cleavage and formation reactions. These mechanistic insights provide guidance for process conditions and catalyst development to minimize the formation of undesired products.

The second part of this presentation will discuss parametric investigations of catalyst lifetime pursuant to changes in methanol space velocity and inlet methanol pressure and interpret trends of cumulative and transient selectivities to implicate formaldehyde, formed in transfer dehydrogenation reactions of methanol, as the key intermediate in transforming active olefin- and aromatic- chain carriers to inactive polycyclic intermediates in methanol-to-olefins catalysis. We also demonstrate efficacy of a bifunctional strategy via physical addition of rare earth oxides in improving the lifetime of methanol-to-olefins chabazite-type zeolite catalysts without disrupting the high selectivity to ethylene and propylene.



Vita:

Aditya Bhan received his Bachelor of Technology (B. Tech.) in Chemical Engineering from IIT Kanpur in 2000 and his PhD in Chemical Engineering from Purdue University in 2005. From January 2005 to August 2007, he was a postdoctoral scholar at the University of California at Berkeley and since then he has been on the Chemical Engineering and Materials Science faculty at the University of Minnesota. He leads a research group that focuses on mechanistic characterization of catalysts useful in energy conversion and petrochemical synthesis. His group at the University of Minnesota has been recognized with the Young Researcher Award from the Acid-Base Catalysis Society and the Ipatieff Prize from the American Chemical Society. He serves as Editor for Journal of Catalysis and as Chair of the ACS Catalysis Science & Technology Division.