

CENTER FOR CATALYTIC SCIENCE AND TECHNOLOGY

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Bill Schneider's expertise is in chemical applications of density functional theory (DFT) simulations. After receiving his Ph.D. in Inorganic Chemistry from the Ohio State University, he began his professional career in the Ford Motor Company Research Laboratory working on a variety of problems related to the environmental impacts of automobile emissions. At Ford he developed an interest in the catalytic chemistry of NO_x for diesel emissions control, and he has published extensively on the chemistry and mechanisms of NO_x decomposition, selective catalytic reduction, trapping, and oxidation catalysis. In 2004, he joined the Chemical and Biomolecular Engineering faculty at the University of Notre Dame as an Associate Professor. At Notre Dame he has continued his research into the theory and molecular simulation of heterogeneous

catalysis, with particular emphasis on reaction environment effects on catalytic materials and their implications for mechanism and reactivity. He was promoted to Professor in 2010 and awarded the H. Clifford and Evelyn A. Brosey Chair in the College of Engineering in 2016. He has co-authored more than 180 papers and book chapters, is a Fellow of the American Association for the Advancement of Science, is a Senior Editor for The Journal of Physical Chemistry, and was the 2018 recipient of the Giuseppe Parravano Award of the Michigan Catalysis Society.

MODELING AND EXPLOITING NON-IDEALITIES IN NITROGEN CATALYSIS

Among the most significant developments in heterogeneous catalysis in the last 20 years is the emergence of microkinetic models, often parameterized from density functional theory (DFT) calculations, used to quantify observed catalyst performance and to guide the discovery of new catalytic materials. While DFT directly reports binding energies and elementary step activation energies, the free energies that enter into microkinetic models must be computed from additional approximations. Frequently these free energy approximations assume ideal behavior—that adsorbates do not interact with one another, or that adsorbates are immobile or vibrate harmonically about a binding site. These assumptions can and do have an impact on predicted catalyst performance and potentially even on predicted trends. In this presentation I discuss our work related to three categories of non-idealities, presented in the context of nitrogen oxidation and reduction catalysis. I describe our strategy for calculation of adsorbate translational free energy, the contribution least well described by conventional models, and compare the exact results to standard approximations. I briefly describe computational models of adsorbate-adsorbate interactions, examine the implications of their incorporation into explicit, lattice-based microkinetic models, and describe recent advances in mean-field descriptions of this coverage-dependence. Lastly, I describe recent work to explore the kinetic consequences of the most fundamental assumption of all—that of energy equipartition—in an effort to rationalize and guide plasma-enhanced catalysis.