

Chemical & Biomolecular Seminar Series



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10:00—11:00 a.m.

102 Colburn Lab

Fabio H. Ribeiro is currently the R. Norris and Eleanor Shreve Professor of Chemical Engineering at the Davidson School of Chemical Engineering, Purdue University. He received his Ph.D. degree from Stanford University in 1989, worked for Catalytica, Inc. in Mountain View, California, held a post-doctoral fellowship at the University of California – Berkeley, and was on the Worcester Polytechnic Institute faculty before joining Purdue University in August 2003. His research interests are centered on the kinetics of heterogeneous catalytic reactions and catalyst characterization under reaction conditions. He has over 130 publications in scholarly journals. He was Chair for the American Institute of Chemical Engineer's Catalysis and Reaction Engineering Division (2010) and is editor for Journal of Catalysis. His honors include the NSF CAREER award (1997-2002), the Excellence in Catalysis Award from the Catalysis Society of Metropolitan New York (2005), the Henry J. Albert Award from the International Precious Metals Institute (2012), the Purdue College of Engineering Faculty Award of Excellence on Research (2014) and the Herman Pines Award from The Chicago Catalysis Club (2015).

On the Reaction Mechanism and the Nature of the Active Site for Standard Selective Catalytic Reduction of NO_x on Cu/SSZ-13 Zeolites

Cu/SSZ-13 zeolites are used commercially in mobile applications for the selective reduction of NO_x with ammonia. Our data suggest that the catalytic cycle is a redox one and occurs on a single Cu site. Complete reduction to Cu(I) was observed only in the absence of O₂ and with both NO+NH₃ present, which led to the formation of mobile diamminecopper(I) species. The order with respect to NH₃ was ~0, indicating strong adsorption of ammonia on Cu(I) and Cu(II). The effect of NO partial pressure indicates that NO was involved in both reduction and oxidation half-reactions.

X-ray absorption spectra for all samples at ambient conditions showed the presence of hydrated Cu²⁺ ions with a coordination number of ~4. Brønsted acid titrations and FTIR spectroscopy revealed that the ion exchanged Cu preferentially exchanged as isolated Cu²⁺ ions charge-balanced by two framework Al atoms followed by [CuOH]⁺ ions charge-balanced by one framework Al atom. Exposure to different gases such as He at 400 °C or H₂ at 250 °C revealed differences between these two sites and showed that the [CuOH]⁺ sites were more reducible compared to the Cu²⁺ sites. In spite of this apparent chemical difference, the two sites had similar standard SCR reaction kinetics, while *operando* XAS experiments showed similar Cu(I) – Cu(II) fractions at steady state, demonstrating their similarity for SCR redox chemistry. The chemical state of Cu was dependent on the gas treatments used. Consequently, it is essential that the catalyst be studied under differential reaction conditions with known concentration of reactants and products.