Title: Designing $IrO_2(110)$ Surfaces for Selective Alkane Chemistry

Abstract:
Selective alkane conversion to high value fuels or chemicals, such as methanol or ethylene, has drawn increasing interest due to abundance of natural gas. Recently, we reported a combined temperature programmed reaction spectroscopy (TPRS) and density functional theory (DFT) study of below room temperature (T ~ 120 K) activation of methane on the stoichiometric $IrO_2(110)$ surface under ultrahigh vacuum (UHV) conditions. Subsequent work also showed that ethane to ethylene selectivity could be increased on $IrO_2(110)$ by creating some proportion of hydrogenated bridging oxygen (O_{br}-H) sites, which limit the oxidative capacity of the surface. This past work suggests that substitution of O_{br} sites with inert species may be an avenue to control selectivity between full and partial oxidation. We have used DFT to explore the effects of Cl substitution of O_{br}. Chlorine is isoelectronic to O_{br}-H and selective Cl substitution of O_{br} has been demonstrated on $RuO_2(110)$ by Over and co-workers. In this work, we apply DFT to explore selective conversion of ethane on Cl-doped $IrO_2(110)$. We find that the Cl-doped $IrO_2(110)$ is still facile in converting ethane to ethylene. In contrast, the ethylene selectivity step shows a strong effect due to Cl doping. Specifically, we find that H atom transfer from ethylene to the saturated O_{br} sites is unfavorable and therefore Cl doping will promote ethylene desorption over further reaction. Current efforts to develop a microkinetic model to explore ethylene selectivity under both TPRS and reaction conditions will be discussed.

Bio:
Dr. Aravind Asthagiri obtained his B.S. in Chemical Engineering with a minor in Mathematics (1998) from The Ohio State University, and a Ph.D. in Chemical Engineering from Carnegie Mellon University (2003). His research involves the application of first-principles based simulations to understand and rationally design novel catalysts for energy applications. He has co-edited a book titled “Computational Catalysis: Recent Advances in Methods and Applications” published in 2014 by the Royal Society of Chemistry. He has also served as the organizer for the Low Pressure group at the AIChE National Meetings and a co-chair of the executive committee for the 23rd North American Catalysis Meeting.