TCSUH Bi-Weekly Seminar

Opportunities and Limits of Dynamically Enhanced Catalysis

Prof. Lars Grabow

Dan Luss Professor, William A. Brookshire Department of Chemical and Biomolecular Engineering, University of Houston, and PI, Texas Center for Superconductivity at the University of Houston

Wednesday, October 11, 2023

In Person – Room 102, Houston Science Center, 12:00 p.m. – 1:00 p.m.

Sandwiches will be provided on a first-come, first-served basis.

ABSTRACT: Catalyst design has traditionally focused on tuning binding energies of surface intermediates to balance the kinetic requirements of multiple, often parallel chemical processes to achieve optimal static catalytic activity as described by the Sabatier principle. Recently, researchers at the Center for Programmable Energy Catalysis (CPEC) have shown that periodic modulation of binding energies of surface intermediates can be leveraged to enhance the catalyst turnover beyond the Sabatier limit by several orders of magnitude for a relatively simple model reaction. Practically, modulation of binding energies can be achieved using a catalytic condenser, a novel device that changes the electron density at the catalyst surface via applied potentials in a programmable pattern.

To explore the opportunities and limits of dynamically enhanced catalysis, we have studied steam methane reforming (SMR) because dynamic SMR operation in portable modular reactors represents a promising strategy to capture fugitive methane (CH_4) emissions from distributed and consequently intractable. Our DFT results on Ru over a range of applied surface charges reveal that the binding energies of important SMR intermediates exhibit unique scaling relationships. Static simulation responses show that applying small positive charges to Ru can greatly enhance the steady-state turnover frequencies (TOFs) by up to four orders of magnitude. Such large improvements in TOFs make positive charging of the Ru surface an effective strategy to realize high catalytic activities at relatively low temperatures. For the dynamic case, we observed significant improvements in the time-averaged TOFs beyond the static activities at the square wave endpoints over a wide band of resonance frequencies (10^6 Hz – 10^{11} Hz). In some cases, forced oscillation of charge resulted in average TOFs exceeding the static maximum by 27.3%. Based on sensitivity analyses performed for both TKMs, we propose that dynamic rate improvement is only possible when the system oscillates between two kinetic regimes that are uniquely controlled by distinct elementary steps. We speculate that further improvements are possible but may require more complex programs.

BIO: Prof. Lars Grabow is the Dan Luss Professor in the William A. Brookshire Department of Chemical and Biomolecular Engineering at the University of Houston. He received his Ph.D. in Chemical Engineering from the University of Wisconsin in 2008, followed by postdoctoral appointments at the Technical University of Denmark and Stanford University. His expertise is the application of electronic structure calculations, kinetic modeling, data science and transient kinetic characterization to problems in heterogeneous catalysis, surface science and electrochemical energy storage. His papers have been cited more than 7,300 times (Google Scholar) and he was elected into the 2018 Class of Influential Researchers by *Industrial and Engineering Chemistry (IE&C) Research*. Prof. Grabow won the prestigious U.S. Department of Energy (DOE) Early Career Award (2014), the NSF CAREER Award (2015), and most recently, the Andrea Prosperetti Research Computing Faculty Award from the Cullen College of Engineering at the University of Houston (2021). He currently serves as Editor of *Surface Science* and on the International Advisory Board of *ChemCatChem*.