

# Department of Chemical and Biomolecular Engineering

## FALL 2015

### Graduate Seminar Series



Seminar  
3:30 - 4:30 pm

Eck Visitors Center  
Auditorium

## Computational Design of Highly Selective Transition Metal Catalysts Encapsulated by Metal-Organic Frameworks for Butane Oxidation to 1-Butanol

Tuesday, September 22, 2015, 3:30 pm

Eck Visitors Center Auditorium

Catalysts are one of the most important technologies in society today, with catalytic processes accounting for nearly 20% of the US GDP. A key focus of catalysis research is designing catalysts that convert feedstocks into higher value products. One of the greatest challenges is selectivity, i.e., production of a desired product over an undesired one, and this is particularly challenging when the desired product is less thermodynamically stable.

The aim of this project is to design a metal nanoparticle catalyst encapsulated within a metal-organic framework (MOF) for this purpose. MOFs are porous crystalline solids comprised of metal-based nodes connected by organic "linker" molecules. The appropriate MOF for this system has small pores that force the surface/molecule interaction to occur at the molecule's terminus. The appropriate metal nanoparticle optimally balances dehydrogenation, hydrogenation, and oxidation processes. In this work, we use molecular simulations to map out several possible pathways for  $2 \text{C}_4\text{H}_{10} + \text{O}_2 \rightarrow 2 \text{C}_4\text{H}_9\text{OH}$  on oxygen-covered transition metal catalyst surfaces, using "featureless" rings comprised of He to simulate the steric restrictions imposed by the MOF pores. Our results suggest that C-H bond activation proceeds through an "oxygen assisted" mechanism, that a key intermediate in the reaction is the 1-butoxy  $\text{C}_4\text{H}_9\text{O}^*$  radical, and that over-dehydrogenation of both the terminal and secondary C is possible. This is true even in MOFs where the pores have diameters that are similar to the kinetic diameter of butane, emphasizing the challenge in designing highly selective catalysts.

*Rachel B. Getman is an Asst. Prof. in Chemical and Biomolecular Engineering at Clemson University. Her group uses quantum chemical calculations and Monte Carlo and molecular dynamics simulations to investigate molecular-level phenomena at fluid/solid interfaces. She is particularly interested in catalytic processes that occur under aqueous conditions and in catalysis involving metal-organic frameworks (MOFs). Dr. Getman earned her PhD from the University of Notre Dame in 2009 where she worked with Professor William F. Schneider.*



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