



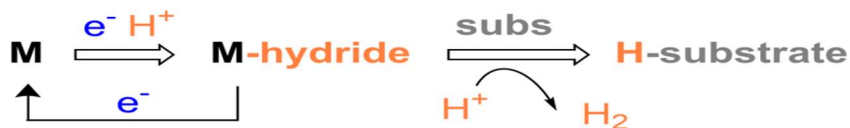
CHEMISTRY & BIOCHEMISTRY COLLOQUIUM: Organometallic chemistry for controlling proton coupled electron transfer catalysis

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About The Speaker:

Louise Berben was born in Sydney, Australia. She received a Bachelor of Science degree with 1st class honors from The University of New South Wales in 2000, and in 2005 was awarded a Ph.D. from the University of California Berkeley for research undertaken with Professor Jeffrey Long. In 2006 Louise began postdoctoral research with Professor Jonas Peters at the California Institute of Technology and in July 2007, moved with the Peters research group to the Massachusetts Institute of Technology. In July 2009, Louise joined the faculty at the University of California Davis where her research program focuses primarily on synthetic and physical inorganic chemistry.



Abstract:

I will present a seminar on the design and synthesis of organometallic molecules for controlling proton and electron transfer reactions for catalysis.

In part 1 I will talk about catalytic reactions which proceed through a metal-hydride intermediate. These usually suffer from trade off in selectivity and reaction rate which are determined by an interplay of reaction thermochemistry and kinetics. Selectivity for hydride transfer to substrate is lost to H₂ evolution when stronger metal-hydride donors are employed in efforts to enhance the reaction rate. This report describes a strategy to use a fast initial chemical step which promotes a pre-equilibrium formation of the catalyst-hydride intermediate. The result of this fast pre-equilibrium kinetics is to promote CO₂ electroreduction to formate formation at $5.26 \times 10^2 \text{ s}^{-1}$ at an overpotential of just $\sim 10 \text{ mV}$. Compared with molecular catalysts that have similar over-potential, this rate is enhanced by five orders of magnitude. As an alternative comparison, overpotential is lowered by 100 mV compared to catalysts with similar rate. The principles elucidated here for mechanism-guided catalyst design are general and can guide future and further improved electrocatalyst development in hydride transfer and X-H bond forming reactions (X = C, N, or other), for molecular and heterogeneous chemistry.

In part 2 redox active ligand complexes of Al(III) and Ga(III) are known to have metalloaromatic electronic structures and to support ligand-based proton transfer and electron transfer chemistry. In this talk I will discuss catalysis mediated by Al(III) complexes of the di(imino)pyridine ligand, including a series of catalytic processes that are all initiated by Al-ligand cooperative bond activation of the polar O-H and N-H bonds in substrates. Examples of the catalytic reactions include acceptorless dehydrogenation of amines and of formic acid, and transfer hydrogenation. The mechanistic details of these catalytic transformations will be discussed along with a discussion of the different Al(III) catalysts and how their electronic structures and coordination environment at the metal center influence the catalytic reaction chemistry.

Date:
9/16/2022

Time:
10:30 AM-11:50 AM

Location:
COB 110

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