Rothchild Lecture

Friday, April 22, 11:45 am

140 Hutchison Hall

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"From Synthetic Fe Nitrogenases to Opportunities in Reductive Electrochemical PCET (ePCET) Catalysis"

Abstract: Nitrogen reduction to NH_3 is a requisite transformation for life. While it is widely appreciated that the Fe-rich cofactors of nitrogenase enzymes facilitate this transformation, how they do so remains poorly understood. A central element of interest has been the site(s) of dinitrogen coordination and reduction, and the atomic level mechanism of this process. Fe is the only transition metal essential to all nitrogenases, and recent biochemical and spectroscopic data implicate Fe as the most likely site of N2 binding within the active site. These observations motivate the pursuit and study of synthetic nitrogenase catalysts as models for detailed study, examples of which were first discovered by our lab several years ago. In this lecture I will discuss our most recent efforts to understand how these Fe catalysts work. In particular, we have posited that concerted proton-electron-transfer (CPET) steps may be important under certain catalytic conditions, not just for iron but also for other (e.g., Mo) catalysts that convert nitrogen to ammonia, and perhaps even in nitrogenase enzymes. We are undertaking studies to place this idea on firm experimental footing via the study of ring-protonated metallocene intermediates featuring remarkably weak C-H bonds. Relatedly, we are exploring the development of electrochemical CPET (eCPET) mediators that mitigate hydrogen evolution and in turn enable reductive electrocatalysis of desirable substrates, including N_2, at comparatively mild potentials.

Zoom Meeting: https://rochester.zoom.us/j/95408612174

Website: https://events.rochester.edu/event/chemistry_2022_rothchild_peters

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