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Chemistry

Influence of the Secondary Coordination Sphere and Electronic Effects on Metal Oxo Complexes

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Transition metal oxo complexes are ubiquitous in natural and synthetic systems and are key intermediates invoked in a wide array of oxidative transformations including C-H bond activation, desaturation, halogenation, and dioxygen evolution. Due to the nature of these systems, such as those bound within a protein scaffold or heterogeneous catalyst, there are often challenges associated with identifying the key reactive sites and mechanisms of their operation. To address this problem, model compounds have been used to generate and study metal oxo species in discrete, molecular platforms. In this presentation, I will discuss two methods that I have used during my doctoral and postdoctoral work to stabilize metal oxo species in order to study their physical and chemical properties. I will first show how hydrogen-bonding motifs can aid in the stabilization of an Fe(IV)-oxo complex and also aid in the movement of protons to study aspects of iron-containing enzyme active sites. I will also discuss studies towards understanding cobalt-based heterogeneous catalysts including attempts to synthesize a terminal Co(IV)-oxo complex via Co(II)-iodosylarene adducts which were shown to perform C-H bond activation chemistry. Finally I will discuss the synthesis and characterization of the first and, to date, only example of a Co(III)-oxo complex. This novel Co(III)-oxo complex was shown to perform C-H bond activation by what has recently been called an asynchronous PCET mechanism. These studies demonstrate that metal oxo model compounds can be stabilized via careful control of the primary and secondary coordination sphere in order to gain a better understanding of their properties and by extension, those of catalytic and enzymatic systems.

Jan. 24th | Lopez Hall | Room 106 | @ 12pm