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DEPARTMENT OF CHEMISTRY AND BIOCHEMISTRY COLLOQUIUM UTAH STATE UNIVERSITY

Exploring the Nonheme High-Valent Iron-Oxo Landscape for Hydrocarbon Oxidations

Twenty years ago, we initiated efforts to mimic the oxidation chemistry of nonheme iron enzymes. In this effort, we discovered the first example of a nonheme iron, $[\text{Fe}^{\text{II}}(\text{TPA})(\text{NCMe})_2]^{2+}$ (TPA = tris(pyridyl-2-methyl)amine), capable of using H_2O_2 as oxidant to catalyze the stereo specific hydroxylation of alkanes as well as the epoxidation and cis-dihydroxylation of olefins. Along the way, we discovered a family of nonheme $\text{Fe}^{\text{IV}}=\text{O}$ complexes that could be trapped and characterized. The first part of this talk focuses on the properties of these novel $\text{Fe}^{\text{IV}}=\text{O}$ complexes and what they teach us about how to break C-H bonds. The second part of this talk shifts attention to a dimer of $[\text{Fe}^{\text{IV}}(\text{O})(\text{TPA}^*)(\text{NCMe})_2]^{2+}$, namely $[\text{Fe}^{\text{IV}}_2(\text{O})_2(\text{TPA}^*)_2]^{4+}$ (TPA* = TPA having methyl and methoxy substituents on the pyridine rings to increase their basicity). On the basis of Raman experiments by Proshlyakov and Lipscomb, such an $\text{Fe}^{\text{IV}}_2(\text{O})_2$ “diamond core” is found in intermediate Q of soluble methane monooxygenase (sMMO-Q), the enzyme intermediate directly responsible for methane hydroxylation. We have now learned how to synthesize the $[\text{Fe}^{\text{IV}}_2(\text{O})_2(\text{TPA}^*)_2]^{4+}$ complex and characterized its properties. In fact, we have used three independent approaches to make this intriguing complex. What we have learned about its chemistry will be discussed.

4-5PM (MDT) | Zoom

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