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, [FeII(TPA)(NCMe)2]2+ (TPA = tris(pyridyl-2-methyl)amine), capable of using H2O2 as oxidant to catalyze the stereo specific hydroxylation of alkanes as well as the epoxidation and cisdihydroxylation of olefins. Along the way, we discovered a family of nonheme FeIV=O complexes that could be trapped and characterized. The first part of this talk focuses on the properties of these novel FeIV=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 [FeIV(O)(TPA*)(NCMe)2]2+, namely [FeIV2(O)2(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 FeIV2(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 [FeIV2(O)2(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.

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