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Diiron(IV)oxo Crystal Structure Modeling Intermediate Q of Methane Monoxygenase

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Nonheme iron enzymes catalyze a wide variety of reactions by O2 activation, including the biosynthesis of the antibiotic chloramphenicol, the radical initiator in the production of deoxyribonucleotides from ribonucleotides by ribonucleotide reductase intermediate X (RNRX) in all eukaryotic life-forms and the selective oxidation of methane to methanol by the soluble form of methane monoxygenase intermediate Q (sMMOQ).[1] High-valent diiron active intermediates have been identified for both sMMOQ and RNRX and are formulated as a diiron(IV)-bis-µ-oxo and a mixed valent Fe(III)Fe(IV)-bis-µ-oxo respectively.[1] Previously, the crystal structure of a mixed-valent Fe(III)Fe(IV)-bis-µ-oxo model complex was reported.[2] A high-yielding synthesis of the reported diiron(IV)-bis-µ-oxo has allowed for the successful structural solution of a high-valent diiron(IV) model complex for sMMOQ.[3] This sMMOQ model complex, along with four other diiron-bis-µ-oxo model complexes in different oxidation and protonation states are compared to sMMOQ and RNRX and related to the mechanism of O2 activation by nonheme enzymes.


Keywords: High-valent Nonheme Iron, Methane Monoxygenase, Model Complex