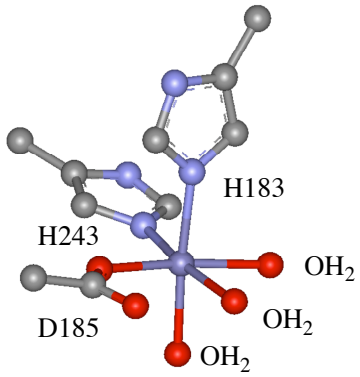
The Use of Sterically Bulky Caboxylates to Model Mononuclear Non-Heme Iron(II) Active Sites

Seth J. Friese, David P. Klein, Lawrence Que, Jr., and William B. Tolman

Department of Chemistry and Center for Metals in Biocatalysis, University of Minnesota, Minneapolis Minnesota (friese@chem.umn.edu)

Carboxylate ligands play an important role in the active sites of many metalloproteins. A large family of dioxygen activating mononuclear non-heme iron(II) enzymes has emerged in the last decade, all of which share a common a 2-His-1-carboxylate facial triad that binds the metal center (e.g. deacetoxycephalosporin C synthase). Despite the similarity in metal sites, the enzymes catalyze a broad range of oxidative transformations. We have initiated an effort to synthesize models for such sites in order to understand how sterics and supporting ligands can affect structure and reactivity. Our strategy involves the use of sterically bulky carboxylate ligands to control the nuclearity of the iron complexes and obtain complexes that achieve close fidelity to the 2-His-1-carboxylate facial triad. The synthesis of these mononuclear iron(II) complexes and their reactivity towards various oxygen donors will be presented.



Active Site:
Deacetoxycephalosporin C Synthase (DAOCS)