Parallel modeling of both enzyme active sites and biomimetic complexes provides closer coupling of these two areas and a better understanding of their similarities and differences. Our recent computational work has developed along several lines: (1) thermodynamic properties such as acidity constants (pKa) and redox potentials (E0) of both enzyme active sites and model complexes; (2) explorations of the mechanism and thermodynamics of reactions that produce oxygenated complexes, particularly the question of S=O vs. mu-Ofe2; (3) electronic structure and rearrangement mechanisms of oxidized, reduced, and protonated complexes and active sites; (4) correlation of IR frequencies as a means of identifying the structure and state of active sites; and (5) the electronic structure of new diiron nitrosyl complexes, where replacement of CO by NO+ in some ways parallels the effect of oxidizing the diiron complex.