

Abstract:

Dinuclear copper assemblies are encountered in the active centers of various oxidases involved in the oxidation of organic substrates by dioxygen. The role of the protein backbone in these metalloproteins is to hold the two Cu-centers in close proximity as to allow a two-electron oxidation of the Cu (I) rest state to the Cu (II) catalytically active form. The objective is to synthesize a dinuclear copper complex by different approaches using a pyrazole ligand and introducing substituents on the bipy site. Furthermore, the investigation of the reactivity of the Cu (I) complex towards oxidants other than dioxygen and the reactivity of Cu (II) complex towards organic substrates follows along the same line of work. At the next stage, reactions of the copper complexes with substrates will be monitored by spectroscopic methods (UV-Vis and NMR) and the reaction products will be characterized by standard and mass spectroscopic methods. Electrochemical studies (cyclic voltammetry) will be employed to define redox stability and redox reactivity of the copper complexes. The reactivity results will be used to direct further tuning of the chemical properties of the copper complexes by the introduction of substituents. The new copper complexes thus prepared will be structurally characterized by X-ray crystallography. The coordination geometries around the Cu-centers of both complexes are significantly different than those of known oxygenases and their synthetic model compounds. This creates the expectation of new chemistries that may be mediated by the new copper complexes. The oxygenation of aromatic and aliphatic hydrocarbons, if accomplished, will constitute an enormous success of the project. This would be a great way of controlling biological processes, creating catalyst substituents for organic reactions that are very slow in absence of catalyst, and even considering the role of these metalloproteins in infectious and neurodegenerative diseases.