Synthetic Cluster Models of Biological and Heterogeneous Oxygen Evolving Catalysts

Research focuses on developing molecular solutions to problems related to energy, materials, and health. Having nature as a source of inspiration, we develop and study systems displaying interactions between multiple molecular centers (metals, Lewis acids, hydrogen bonding functionalities, etc.) to facilitate a variety of chemical processes.

Abstract

Redox-inactive metals are found in biological and heterogeneous water oxidation catalysts, but their roles are currently not well understood. Targeting synthetic model clusters of these catalysts, triphenylbenzene moieties appended with pyridine and alkoxide donors were utilized as multinucleating ligands for first-row transition metals. Complexes of Mn^{2+}, Fe^{2+}, Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+} were synthesized and found to display trinuclear cores supported by bridging alkoxides. Trimanganese complexes were used as precursors for more elaborate metal oxide clusters. Tetranuclear complexes displaying Mn₄, Mn₃Ca and other Mn₃M motifs (M=Na⁺, Sr²⁺, Zn²⁺, Sc³⁺, Y³⁺) with varied number of bridging oxo ligands were prepared and studied. A significant variation of reduction potentials depending on the nature of the redox inactive metal was observed. Implications to the function of the oxygen evolving complex of photosystem II and synthetic oxygen production catalysts will be discussed.