"The Currents of Life: Electron Flow through Metalloproteins"

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Summary

Electron flow through proteins and protein assemblies in the respiratory and photosynthetic machinery commonly occurs between metal-containing cofactors that are separated by large molecular distances, often in the 10-25 angstrom range. Although these cofactors are weakly coupled electronically, the reactions are remarkably rapid and specific. We have investigated free energy, temperature, and distance dependences of electron transfer rates in Ru(diimine)-modified iron and copper proteins. Lessons learned about the control of electron flow through these and related molecules are aiding the design of sensitizer-modified proteins to generate hydrogen and oxygen from sunlight and water.

Professor Harry B. Gray (1935)

is the Arnold O. Beckman Professor of Chemistry and the Founding Director of the Beckman Institute at the California Institute of Technology. His main research interests focus on inorganic spectroscopy, photochemistry, and bioinorganic chemistry, with emphasis on understanding electron transfer in proteins. For his contributions to chemistry, which include over 700 papers and 17 books, he has received many prestigious awards, including the National Medal of Science from President Ronald Reagan (1986), the Wolf Prize in Chemistry (2004), the Benjamin Franklin Medal in Chemistry (2004) and the Priestley Medal (1991), as well as 16 honorary doctorates. He is a member of the National Academy of Sciences (USA), the Royal Society of Great Britain and many other distinguished learned societies.

Prof. Gray began his work in inorganic chemistry at Northwestern University, where he earned a Ph.D. in 1960. After a postdoctoral year at the University of Copenhagen, he joined the chemistry faculty at Columbia University, where his main interests centered on the electronic structures and reactions of inorganic complexes. During this period, he also employed molecular orbital theory to describe electronic structures of transition metal complexes. After moving to Caltech in 1966, he and coworkers continued their spectroscopic studies, which led them into investigations of solar photochemistry of metal complexes, including construction of donor-acceptor systems that mimic the early events of photosynthesis. In the 70's, Prof. Gray also attacked the problem of electron flow through biological molecules by examining kinetics of reactions of iron and copper proteins with inorganic complexes. Working with Ru-modified proteins in the early 1980s, he and coworkers demonstrated that electrons can tunnel rapidly over long molecular distances through folded polypeptide structures. Very recently, he has constructed photoactive electron tunneling wires to probe deeply buried active sites in cytochrome P450 and other redox enzymes, while the electron-transfer studies have been extended to protein crystals. Another recent research area pursued in Prof. Gray's laboratory concerns protein folding, whereby conformational charges are triggered by rapid electron transfer and monitored using fluorescent probes.

The research carried out by Prof. Gray and his team has influenced and inspired the field of inorganic chemistry since the 1960's and more recently has had a great impact on bioinorganic chemistry, biochemistry, biophysics and well beyond. For example, the work on protein folding is closely relevant to important medical issues such as the detection and treatment of Alzheimer's and Parkinson's diseases, while the understanding of electron transfer provides a foundation for the development of molecular electronic devices as well as systems for solar photochemical energy conversion.