Engineered Cytochromes for Solar Fuels

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Abstract: The use of engineered heme proteins to perform the photocatalytic generation of hydrogen fuel from water will be presented. Light-absorbing and charge-transfer chromophores and hydrogen-producing catalysts are prepared from cytochromes *c*, proteins with an iron protoporphyrin IX (heme) group covalently attached to the polypeptide. To prepare modified cytochromes, a system has been developed for the biosynthesis of a wide range of proteins and peptides with covalently attached heme. The heme iron in the engineered cytochrome is substituted with other metals to impart the desired function on the construct. In addition, the protein or peptide sequence may be altered to tune reactivity and other properties. Zinc-cytochromes act as photoinduced electron donors, either to carbon nanotube acceptors for charge separation or directly to catalysts. Cobalt-substituted cytochromes are employed as catalysts for proton reduction in neutral-pH water under aerobic conditions with high turnover numbers. Use of protein and peptide engineering facilitates the optimization of catalyst stability and overpotential. This protein engineering-based approach to solar fuel production offers a convenient pathway to a wide range of water-soluble functional assemblies.