



Perspective

Rate-potential decoupling: a biophysical perspective of electrocatalysis

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Abstract

In this perspective, the chemical physics of biological electron transfer are considered in relation to artificial electrocatalyst development. Nature's ability to access a wide range of chemical reactivities through a relatively narrow set of redox-active motifs, in part by decoupling electron transport rates from reaction driving forces, are suggested as a model for the future of electrocatalyst design and testing. Theoretical rationale and experimental precedents for this concept are put forth, outlining how emulating nature's ability to arbitrarily tune tunneling currents with respect to donor/acceptor redox potentials – reaction driving forces – may enhance our control over electrocatalyst selectivity.

Keywords: electrocatalysis, decoupling, bioinorganic, solar fuels, photoelectrochemistry, artificial photosynthesis, semiconductor-liquid junction

1. Canonical bioinorganic catalysis: the assumed centrality of structure-function relationships

Organometallic mimics of enzyme active sites, such as those of cytochrome P450 porphyrin centers and the oxygen evolving complex (OEC) of photosystem II, have substantially advanced our understanding of their underlying chemical mechanisms and spectroscopic features [1–3]. However, functional mimicry of redox enzymes, particularly through the use of organometallic complexes, has proven uniquely challenging to the field of bioinorganic chemistry. Attempts to link inorganic catalysis and enzyme biochemistry have generally focused on the development of homogeneous molecular catalysts, which act as structural mimics of biological active sites but typically ignore the higher-order aspects of protein design, particularly the roles played by secondary and tertiary structure [4, 5], solvent exclusion effects and regulated electron transport in enzyme catalysis [6, 7]. By and large, synthetic chemists have operated under an implicit assumption that structural mimicry of enzyme active sites should correspond to functional emulation of enzyme catalysis. However, this first approximation has often been found to be necessary but by itself insufficient, with molecular catalysts that superficially resemble protein active centers often failing to replicate enzyme reactivities.

While structure plays a critical role in facilitating enzyme catalysis, this clearly marks only one essential aspect of functional biomimicry. For example, while detailed molecular mimics of photosystem II's OEC have generally failed to fully reproduce the water oxidation chemistry it catalyzes, purely inorganic Mn oxide powders have been found to drive efficient water oxidation [8]. In the specific case of calcium-containing birnessite-like Mn oxides, these materials contain CaMn_2O_4 domains similar to the CaMn_4O_5 cubanes constituting the OEC, despite omitting the surrounding ligand framework present in both the OEC and organometallic model complexes [1, 8, 9]. The comparative success of inorganic powders (bearing little resemblance to protein active sites) relative to molecular catalyst active site mimics, signal that structural emulation alone is insufficient

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