Design and properties of a bio-inspired catalyst/electrode system for electrocatalytic H_2 production from water*

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Photo- or electro-catalytic production of hydrogen from water has attracted much attention in recent years, prompting efforts to design an active catalyst comprised of earth-abundant elements. Insight into the design of such a catalyst can be gained from nature as, e.g., the di-iron hydrogenases of certain anaerobic bacteria produce hydrogen efficiently, with turnover numbers approaching 10^4/sec. In this talk, I shall discuss the design of a stable, efficient electrocatalyst for H_2 production starting from the di-iron hydrogenase active center. The cluster, attached directly to an FeS_2 (100) surface, forms a catalyst/electrode complex with a high density of reactive sites. Moreover the direct link between the active center and the surface ensures faster electron transfer than obtainable by attaching the entire enzyme to the surface. This complex could be an efficient, inexpensive hydrogen producer if it avoids the O_2 poisoning of the hydrogenase active center. We have investigated the reaction of solvated oxygen with the cluster/electrode complex in water via Car-Parrinello molecular-dynamics simulations. We find that the same changes that improve the stability of the cluster in water make also the catalyst *oxygen tolerant*, reducing to water any oxygen it binds and surviving without structural change for times far longer than those of practical concern.

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