
II.F.1 Tailoring Hydrogen Evolution Reaction (HER) Catalysts for Operation at Specific pH Values

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We report the electrocatalytic activity of $[\text{HNi}(\text{DHMPE})_2][\text{BF}_4]$ (DHMPE= 1,2-bis(di-(hydroxymethyl)phosphino)ethane) (1) toward hydrogen production at pH 1, with a rate of $\sim 10^3 \text{ s}^{-1}$, Faradaic efficiency between 92–105%, and negligible decomposition after 18 h of electrolysis. We are currently pursuing new nickel diphosphine complexes modified to make stronger metal hydrides for the reduction of substrates such as CO_2 to other C1 fuels.

Abstract

Electrocatalysts that can generate renewable fuels under a wide range of pH conditions in water are necessary for renewable energy applications. Metal hydrides are critical intermediates involved in bond-breaking and bond-forming steps in relevant catalytic reactions. Our group relies on thermodynamic hydricity-pH relationships to target reaction conditions for selective hydride transfer for proton reduction to H_2 or CO_2 reduction to formate (HCO_2^-) in water. Although metal hydricities have been measured for hundreds of transition metal complexes in organic solvents, aqueous hydricities are relatively unexplored.