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

Bianca Ceballos
Department of Chemistry
University of California, Irvine
Irvine, CA 92697
Phone: (949) 824-2509
Email: bceballo@uci.edu

DOE Manager: Mark Spitler
Phone: (301) 903-4568
Email: Mark.Spitler@science.doe.gov

We report the electrocatalytic activity of $[\text{HNi}(\text{DHMPPE})_2][\text{BF}_4]$ (DHMPPE= 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.