Solar and wind are carbon-neutral, sustainable energy sources, but they are intermittent, so energy storage is required. Catalysts that efficiently interconvert between electrical energy and chemical bonds (fuels) are needed for sustainable, secure energy. Electrocatalysts based on earth-abundant metals are needed since low-temperature fuel cells generally use platinum, an expensive, precious metal. We developed nickel(II) complexes for the electrocatalytic production of H$_2$ by reduction of protons. Pendant amines in the ligand function as proton relays, facilitating intramolecular and intermolecular proton mobility. Turnover frequencies up to $10^7$ s$^{-1}$ have been observed, though some have a high overpotential. Iron complexes with pendant amines in diphosphine ligands have been developed for the opposite reaction, oxidation of H$_2$ (1 atm). Our results illustrate the rational design of catalysts based on abundant, inexpensive metals as alternatives to precious metals.