Increasing Green Hydrogen Production Accessibility: Derived Magnetite Nanosheets on Nickel Foam as a Highly Active Electrocatalyst for Oxygen Evolution

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Hydrogen has emerged as a promising medium for zero-carbon energy storage; however, due to the cost and complexity of electrocatalyst synthesis for alkaline water splitting (AWS), approximately 95% of hydrogen production still relies on fossil fuels. Here, we report a facile, scalable route to a novel, low-cost, self-supported magnetite electrode for the oxygen evolution reaction (OER). The electrocatalyst is fabricated via in situ transformation of sulfate-electrodeposited oxyhydroxide nanosheets into magnetite nanosheets through pseudomorphic replication, achieved by annealing followed by controlled hydrogen reduction via the Aerobic Hydroxide Conversion (AHC) process. This in situ synthesis drives sequential phase transitions—from mixed orthorhombic, through rhombohedral, to inverse spinel structures. The resulting electrode delivers an overpotential of only 273 mV at 10 mA cm?² and outperforms RuO2 in stability, owing to its binder-free architecture and high conductivity arising from delocalized electrons in magnetite. Post-operando characterization reveals the formation of an amorphous FeOOH surface layer and a maghemite intermediate atop crystalline Fe3O4, substantially increasing the density of active sites. Density functional theory calculations demonstrate that cation-vacancy-induced surface anisotropy in maghemite, coupled with superexchange spin-pinning between ferromagnetic domains, generates an intrinsic magnetic field that significantly enhances spin polarization and OER kinetics. Synthesized from earth-abundant precursors via an industrially viable process, this magnetite-based electrode offers a compelling path toward globally accessible, cost-effective green hydrogen production.

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