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Competitive Coordination Strategy Towards Reversible Hybrid Hetero-Homogeneous Oxygen-Evolving Catalyst

Authors : Peikun Zhang, Chunhua Cui

Abstract : Photoelectrochemical (PEC) water splitting provides a promising pathway to convert solar energy into renewable fuels. However, the main and seemingly insurmountable obstacle is that the sluggish kinetics of oxygen evolution reaction (OER) severely jeopardizes the overall efficiency, thus exploring highly active, stable, and appreciable catalysts is urgently requested. Herein a competitive coordination strategy was demonstrated to form a reversible hybrid homo-heterogeneous catalyst for efficient OER in alkaline media. The dynamic process involves an in-situ anchoring of soluble nickel–bipyridine pre-catalyst to a conductive substrate under OER and a re-dissolution course under open circuit potential, induced by the competitive coordination between nickel–bipyridine and nickel-hydroxyls. This catalyst allows to elaborately self-modulate a charge-transfer layer thickness upon the catalytic on-off operation, which affords substantially increased active sites, yet remains light transparency, and sustains the stability of over 200 hours of continuous operation. The integration of this catalyst with exemplified state-of-the-art Ni-sputtered Si photoanode can facilitate a ~250 mV cathodic shift at a current density of 20 mA cm-2. This finding helps the understanding of catalyst from a “dynamic” perspective, which represents a viable alternative to address remaining hurdles toward solar-driven water oxidation.

Keywords : molecular catalyst, oxygen evolution reaction, solar energy, transition metal complex, water splitting
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