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Boosting overall water splitting by incorporating sulfur into NiFe (oxy) hydroxide

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ABSTRACT

Developing highly active and cost-effective electrocatalysts for enhancing the hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) is a significant challenge for overall water splitting. Sulfur-incorporated nickel iron (oxy)hydroxide (S-NiFeOOH) nanosheets were directly grown on commercial nickel foam using a galvanic corrosion method and a hydrothermal method. The incorporation of sulfur into NiFeOOH enhanced the catalytic activity for the HER and OER in 1 M KOH electrolyte. The enhanced catalytic activity is attributed to the change in the local structure and chemical states due to the incorporation of sulfur. High performance for overall water splitting was achieved with an alkaline water electrolyzer. This was realized by employing S-NiFeOOH as a bifunctional electrocatalyst, thereby outperforming a water electrolyzer that requires the usage of precious metal electrocatalysts (i.e., Pt/C as the HER electrocatalyst and IrO2 as the OER electrocatalyst). Moreover, when driven by a commercial silicon solar cell, an alkaline water electrolyzer that uses S-NiFeOOH as a bifunctional electrocatalyst generated hydrogen under natural illumination. This study shows that S-NiFeOOH is a promising candidate for a large-scale industrial implementation of hydrogen production for overall water splitting because of its low cost, high activity, and durability. In addition, the solar-driven water electrolyzer using S-NiFeOOH as a bifunctional electrocatalyst affords the opportunity for developing effective and feasible solar power systems in the future.

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1. Introduction

Electrochemical water splitting is an attractive approach for generating hydrogen by utilizing renewable electricity from solar or wind energy [1–3]. Water splitting involves the hydrogen evolution reaction (HER, $2H_2O + 2e^- \rightarrow 2OH^- + H_2$) and the oxygen evolution reaction (OER, $4OH^- \rightarrow 2H_2O + 4e^- + O_2$) [4,5]. However, the scalable industrial application of water splitting is still hampered by the enormous energy penalty caused by the sluggish kinetics of the two water electrolysis half-reactions (i.e., HER and OER) [6–8]. Although precious metal-based electrocatalysts show high activity for the HER (Pt-based) and OER (IrO₂-based), their utilization is limited because of their cost and scarcity. The development of highly active bifunctional electrocatalysts for water splitting affords the integrated advantages of simplifying device fabrication

and reducing costs [9]. In order to overcome the aforementioned obstacles, several types of earth-abundant transition metal (e.g., Ni, Mn, Fe, Cu, Co)-based electrocatalysts (i.e., oxides [10-13], phosphides [14-17], sulfides [18-20], selenides [21-24] and hydroxides [25-28]) have been developed, which show high catalytic activity for overall water splitting. Transition metal-based hydroxides (hydroxides, (oxy)hydroxides, and layered double hydroxides) are considered particularly promising bifunctional electrocatalysts because of their high catalytic activity and excellent durability [29-35]. However, in many previous studies, transition metal-based hydroxides showed high catalytic activity for the OER, but showed relatively low catalytic activity for the HER, demonstrating the limited efficacy of transition metal-based hydroxides for overall water splitting [36–38]. Further, the intrinsically low electrical conductivity and lack of electrochemically active surfaces of transition metal-based hydroxides are serious problems [39-41]. Moreover, these transition metal hydroxides require the use of poorly conductive polymeric binders (i.e., Nafion,

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