

Title:

Atomic cation vacancy engineering of NiFe-LDH nanosheets towards oxygen evolution reaction.

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Abstract:

Electrochemical oxygen evolution reaction (OER) is the efficiency-determining process of water splitting, which is considered one of the most promising strategies for generating hydrogen. Although some progress has been made in the development of transition metal-based OER catalysts, most are still inferior to their noble metal counterparts. Another drawback is that most of them cannot be used as bifunctional electrocatalysts for both OER and hydrogen evolution reaction (HER). Layered double hydroxides based on Ni and Fe (NiFe LDH) have been reported to exhibit relatively high OER activity, while the enhancement of intrinsic activity and specific surface area is still needed to further improve the catalytic performance. In our previous work, efficient strategies such as morphology design^[1], composition tuning^[2], and heterostructure^[3] were explored to meet the needs. Recently, vacancy engineering has been proposed to be a promising protocol for improving the electrocatalytic activity of transition metal LDHs.

In the current work, Ni²⁺-Fe³⁺ LDHs were synthesized using a homogeneous precipitation method in the presence of hexamethylenetetramine (HMT, C₆H₁₂N₄) and anthraquinone-2-sulfonate (AQS, C₁₄H₇O₅S).^[1] Herein, doping with a designated amount of Zn²⁺ into Ni²⁺-Fe³⁺ LDHs was carried out. After successfully exfoliating the bulk Ni_{2-x/3}Zn_{x/3}Fe_{1/3} LDHs (x = 0-0.2) into single-layer nanosheets, vacancies were generated through chemical etching out Zn²⁺ in a strong alkali solution (Fig.1a). The Ni^VFe-LDH nanosheets was then flocculated with anionic CO₃²⁻ to produce catalysts in powder form for OER measurements. The results show that vacancies in the resultant Ni^VFe-LDH nanosheets may help significantly improve the intrinsic activity for OER (Fig.1c). Furthermore, these nanosheets can be hetero-assembled with HER-active molybdenum disulfide (MoS₂) nanosheets to serve as a bifunctional catalyst for water splitting (Fig.1d).

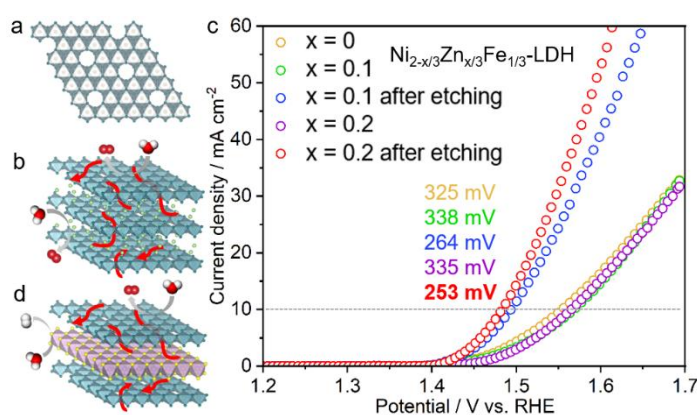


Fig.1. Schematic models of (a) vacancy containing Ni^VFe-LDH nanosheets, (b) CO₃²⁻-flocculated Ni^VFe-LDH, (c) OER activity of CO₃²⁻-flocculated Ni^VFe-LDH samples with different composition, Schematic models of (d) Ni^VFe-LDH/MoS₂

References:

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