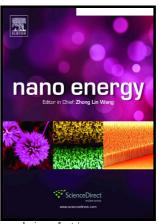
## Author's Accepted Manuscript

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## **ACCEPTED MANUSCRIPT**

# Iron-tuned Super Nickel Phosphide Microstructures with High Activity for Electrochemical Overall Water Splitting

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#### **Abstract**

Large-scale hydrogen production by electrolytic splitting of water is mainly governed by high-efficient yet cheap electrocatalysts that could be capable of accelerating the sluggish hydrogen evolution reaction (HER) and oxygen evolution reaction (OER). Herein, we report Fe-tuned Ni<sub>2</sub>P electrocatalysts with controllable morphology and structure by regulating atomic ratio of Ni/Fe, and reveal the Fe species-modulated electronic state behaviors and -boosted catalytic activity for water splitting. The electrocatalytic activity of Fe-tuned Ni<sub>2</sub>P nanosheets for both HER and OER can be further enhanced by assembling the nanosheets vertically on conductive 2D carbon fiber (CF) matrix to make hierarchical monolithic 3D electrode (Ni<sub>1.5</sub>Fe<sub>0.5</sub>P/CF), which features more accessible active sites and open structure that helps to speed up both the HER and OER. The improved electrocatalytic activity of Ni<sub>1.5</sub>Fe<sub>0.5</sub>P/CF is due to the combined synergistic effects of the high conductivity of CF matrix and the strong interaction between active species and the CF support, as evidenced by a low overpotential of 293 mV to achieve a high current density of 100 mA cm<sup>-2</sup> with superiorly long-term stability for OER. When the monolithic 3D Ni<sub>1.5</sub>Fe<sub>0.5</sub>P/CF electrodes were used as both anode and cathode for overall water splitting, a current density of 10 mA cm<sup>-2</sup> is

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<sup>&</sup>lt;sup>1</sup> The first two authors contributed equally to this work

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