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## **Supporting Information**

## FeP/Ni<sub>2</sub>P Nanosheet Arrays as High-Efficiency Hydrogen

## **Evolution Electrocatalysts**

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Fig.S1 EDS elemental mapping images of FeP/Ni $_2$ P/CP. (a) overlay, (b) Fe element, (c) Ni element, (d) P element.



Fig.S2 SEM images of (a) FeP/CP-NA, (b) FeP/CP, (c) Ni<sub>2</sub>P/CP, (d) FeP/Ni<sub>2</sub>P/CP.



Fig.S3 XPS spectra of Ni(OH)<sub>2</sub>/CP and FeOOH/Ni(OH)<sub>2</sub>/CP precursor.



Fig.S4 (a) LSV curves of FeP/Ni<sub>2</sub>P/CP, FeP/CP, Ni<sub>2</sub>P/CP, FeP/CP-NA and FeP/Ni<sub>2</sub>P/CP-NA. (b) Corresponding  $\eta_{-10}$  and  $\eta_{-100}$ . (c) Plots of specific current densities (I<sub>s</sub>) of FeP/Ni<sub>2</sub>P/CP, FeP/CP and Ni<sub>2</sub>P/CP. The electrolyte is 0.5 M H<sub>2</sub>SO<sub>4</sub>.



Fig.S5 (a) LSV curves of FeP/Ni<sub>2</sub>P/CP, FeP/CP, Ni<sub>2</sub>P/CP, FeP/CP-NA and FeP/Ni<sub>2</sub>P/CP-NA. (b) Corresponding  $\eta_{-10}$  and  $\eta_{-100}$ . (c) Plots of specific current densities (I<sub>s</sub>) of FeP/Ni<sub>2</sub>P/CP, FeP/CP and Ni<sub>2</sub>P/CP. The electrolyte is 1.0 M KOH.



Fig.S6 simulated DOS of the Fe-3d orbital, Ni-3d orbital and P-3p orbital in FeP/Ni<sub>2</sub>P.



Fig.S7 Optimized structure of  $H_{ad}$  on (a) FeP(211) and (b)  $Ni_2P(111).$