

## **Supporting Information**

### **Enhanced Electrochemical Water Oxidation Activity by Structural Engineered Prussian Blue Analogue/rGO Heterostructure**

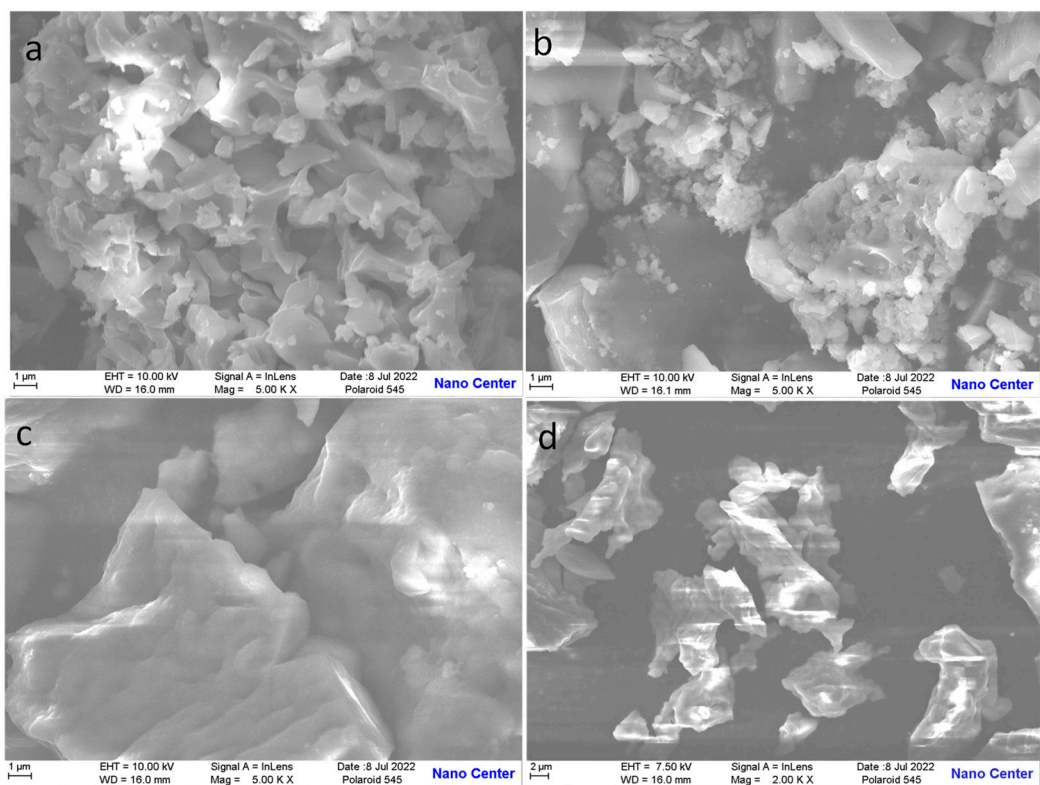
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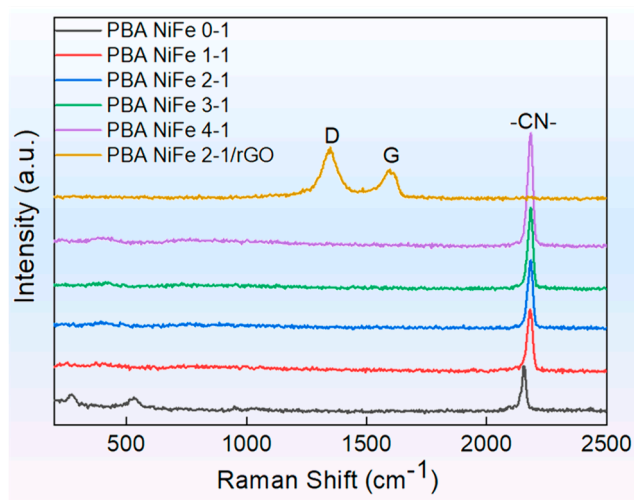
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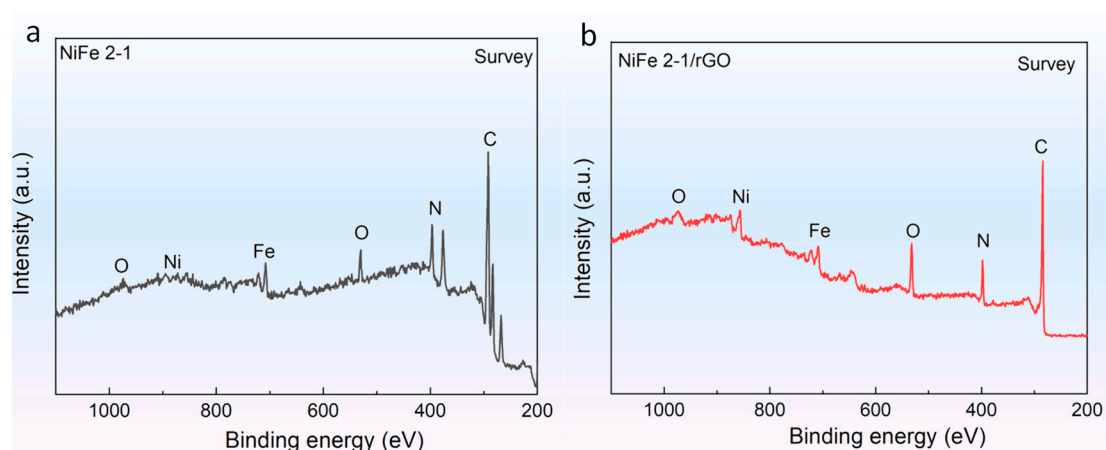
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**Figure S1.** SEM images of as-obtained PBA materials. (a) PBA 0-1; (b) PBA 1-1; (c) PBA 3-1; (d) PBA 4-1.



**Figure S2.** The Raman spectra of PBA materials.

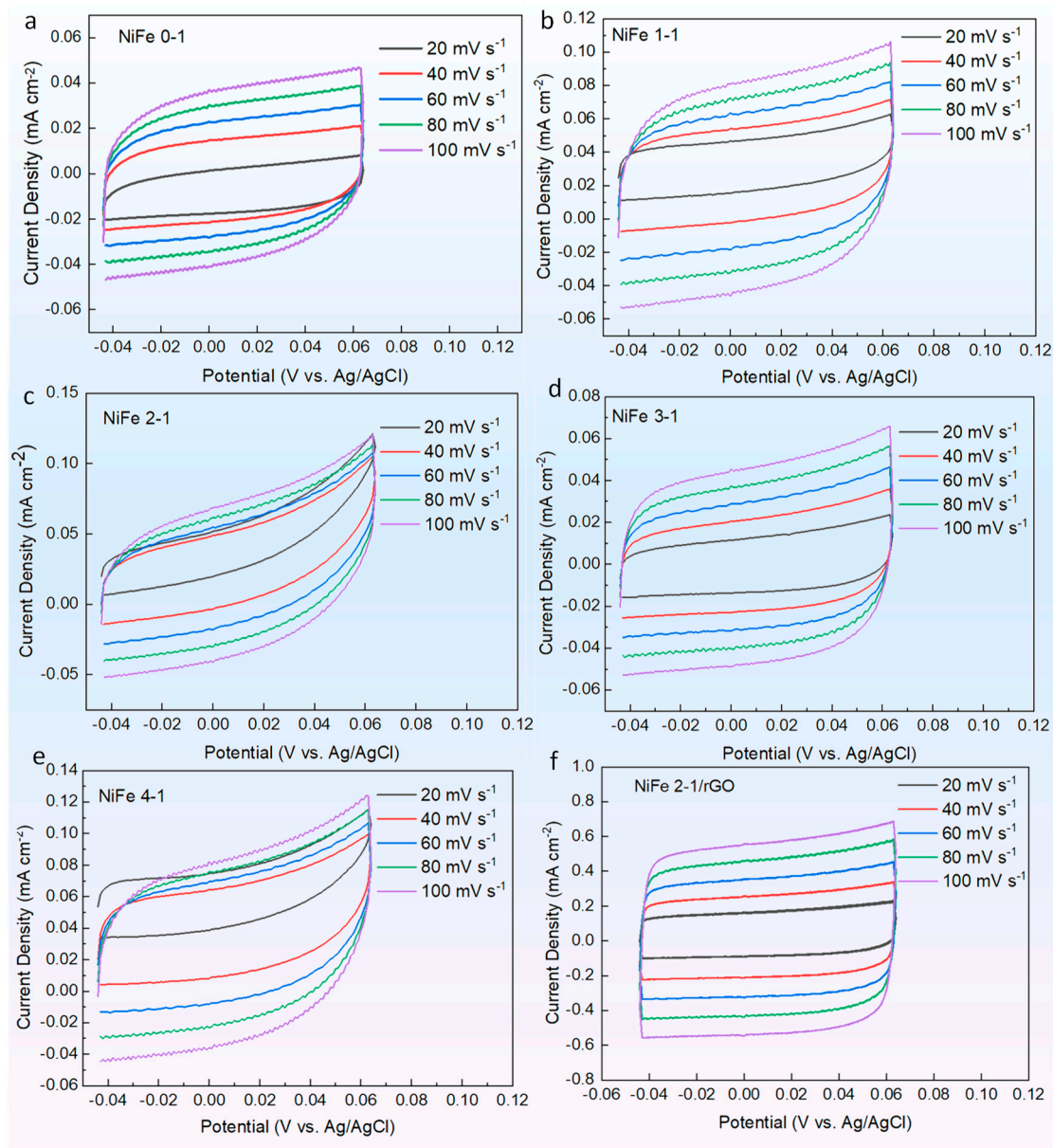


**Figure S3.** The XPS survey spectra of PBA materials before OER tests. **(a)** PBA NiFe 2-1; **(b)** PBA NiFe 2-1/rGO.

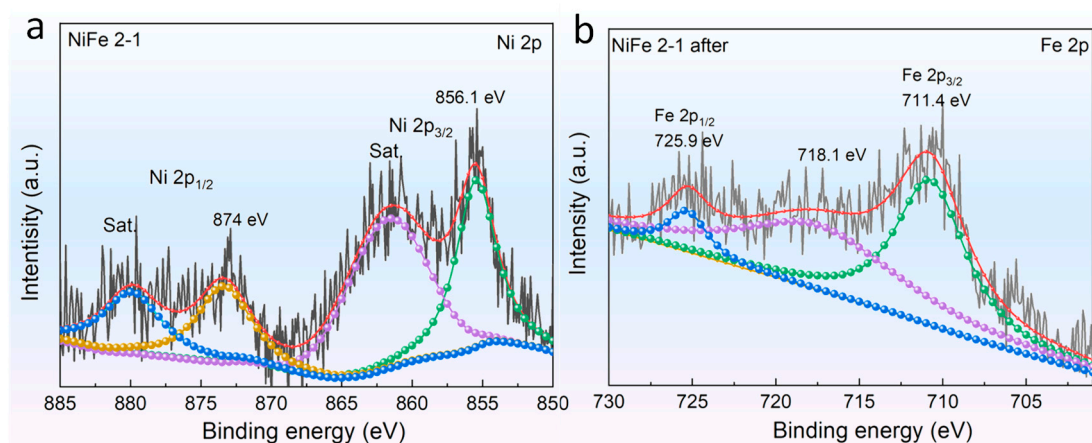
**Table S1.** Catalyst performance of analogous materials.

Materials	Methods	Mass loading /mg cm <sup>-2</sup>	Overpotential /mV (30 mAcm <sup>-2</sup> )	Tafel slopes /mV dec <sup>-1</sup>	Durability /h	Ref
NiHCF-200c	Sodium-ion electrochemical tuning method	0.5	300	41	24	[1]
FeCoNi PBAs polyhedrons	Oil bath reaction	0.5	300	52	—	[2]
Ni-Fe-K <sub>0.23</sub> MnO <sub>2</sub> CNFs-300	Sacrificial template strategy	2	290	42.3	24	[3]
NiCo@A-Ni Co-PBA-AA	Topological growth method	0.0006 (GCE)	410	79.1	40	[4]
(v-NiFe PBA@rGO	Rapid coprecipitation method	0.285	290	36.2	200	[5]
PBA NiFe 2-1/rGO	Coprecipitation method	0.5	331.5	57.9	40	This work

GCE: glassy carbon electrode.



**Figure S4.** CV curves of as-obtained PBA materials at different scan rates in non-Faraday potential range. (a) PBA NiFe 0-1; (b) PBA NiFe 1-1; (c) PBA NiFe 2-1; (d) PBA NiFe 3-1; (e) PBA NiFe 4-1; (f) PBA NiFe 2-1/rGO.



**Figure S5.** XPS spectra of PBA samples after OER measurement. High-resolution Ni 2p (**a**) and Fe 2p (**b**) spectra of PBA NiFe 2-1, respectively.

**Table S2.** The calculated Cdl ECSA values of as-obtained PBA materials.

Materials	C <sub>dl</sub> (mF cm <sup>-2</sup> )	ECSA (cm <sup>2</sup> )
PBA NiFe 0-1	0.4	10
PBA NiFe 1-1	0.6	15
PBA NiFe 2-1	0.5	12.5
PBA NiFe 3-1	0.4	10
PBA NiFe 4-1	0.5	12.5
PBA NiFe 2-1/rGO	5.3	132.5

## References

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2. Du, Y.; Ding, X.; Han, M.; Zhu, M., Morphology and Composition Regulation of FeCoNi Prussian Blue Analogues to Advance in the Catalytic Performances of the Derivative Ternary Transition-Metal Phosphides for OER. *ChemCatChem* **2020**, 12, 4339-4345.
3. Liao, H.; Guo, X.; Hou, Y.; Liang, H.; Zhou, Z.; Yang, H., Construction of Defect-Rich Ni-Fe-Doped K<sub>0.23</sub>MnO<sub>2</sub> Cubic Nanoflowers via Etching Prussian Blue Analogue for Efficient Overall Water Splitting. *Small* **2020**, 16, 1905223.
4. Zhang, H.; Li, P.; Chen, S.; Xie, F.; Riley, D. J., Anodic Transformation of a Core-Shell Prussian Blue Analogue to a Bifunctional Electrocatalyst for Water Splitting. *Adv. Funct. Mater.* **2021**, 31, 2106835.
5. Jo, S.; Kwon, J.; Choi, S.; Lu, T.; Byeun, Y.; Han, H.; Song, T., Engineering [Fe(CN)<sub>6</sub>]<sup>3-</sup> vacancy via free-chelating agents in Prussian blue analogues on reduced graphene oxide for efficient oxygen evolution reaction. *Appl. Surf. Sci.* **2022**, 574, 151620.