

**Electronic Supplementary Information for
Intrinsic poor-crystallized Fe₅O₇(OH)•4H₂O: A highly efficient oxygen
evolution reaction electrocatalyst under alkaline conditions**

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Experimental Section

Characterizations

Powder X-ray diffraction (XRD) data were obtained on a RigakuD/MAX 2550 diffractometer with Cu K α radiation ($\lambda=1.5418\text{ \AA}$). Scanning electron microscopy (SEM) measurements were conducted on a Hitachi S-4800 field emission scanning electron microscope at an accelerating voltage of 20 kV. Transmission electron microscopy (TEM) measurements were made on a Hitachi H-8100 electron microscopy (Hitachi, Tokyo, Japan) with an accelerating voltage of 200 kV. X-ray photoelectron spectroscopy (XPS) measurements were carried out on an ESCALABMK II X-ray photoelectron spectrometer using Mg as the exciting source. The electron spin resonance (ESR) spectra were collected on a Bruker A300 Paramagnetic spectrometer, which were performed at room temperature in vacuum.

Electrochemical measurements

Electrochemical measurements were performed with a CHI 660E electrochemical analyzer (CH Instruments, Inc., Shanghai) in a typical three electrode system, with Fe₅O₇(OH)•4H₂O/NF as working electrode, graphite as counter electrode and Hg/HgO as reference electrode. All tests were carried out at room temperature.

TOF calculation

To calculate TOF, the surface concentration of active sites related to the redox species should be first obtained. On the basis of the electrochemical CV curves (Fig. S4a and S4c), the oxidation peak current of redox species demonstrates linear change on scan rates (Fig. S4b and S4d). The slope of the line can be calculated using the following equation:

$$slope = \frac{n^2 F^2 A \tau_0}{4RT}$$

n is the number of electrons transferred, F is Faraday's constant, A is the surface area of the electrode, τ_0 is the surface concentration of active sites (mol cm^{-2}), and R and T are the ideal gas constant and the absolute temperature, respectively. TOF values can be finally calculated based on the formula:

$$TOF = \frac{JA}{4FM}$$

J is the current density at certain overpotential, A is the area of the electrode, 4 indicates the mole of electrons consumed for evolving one mole of O_2 from water, F is Faraday's constant and m is the number of moles for active sites.

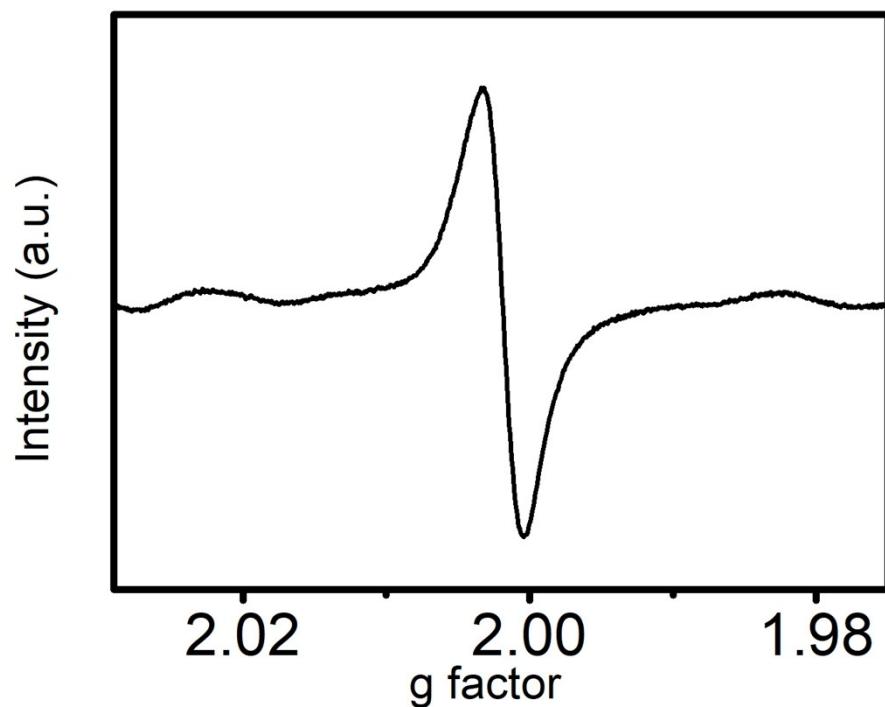


Fig. S1. ESR spectrum of $\text{Fe}_5\text{O}_7(\text{OH})\bullet 4\text{H}_2\text{O}$ nanosheet

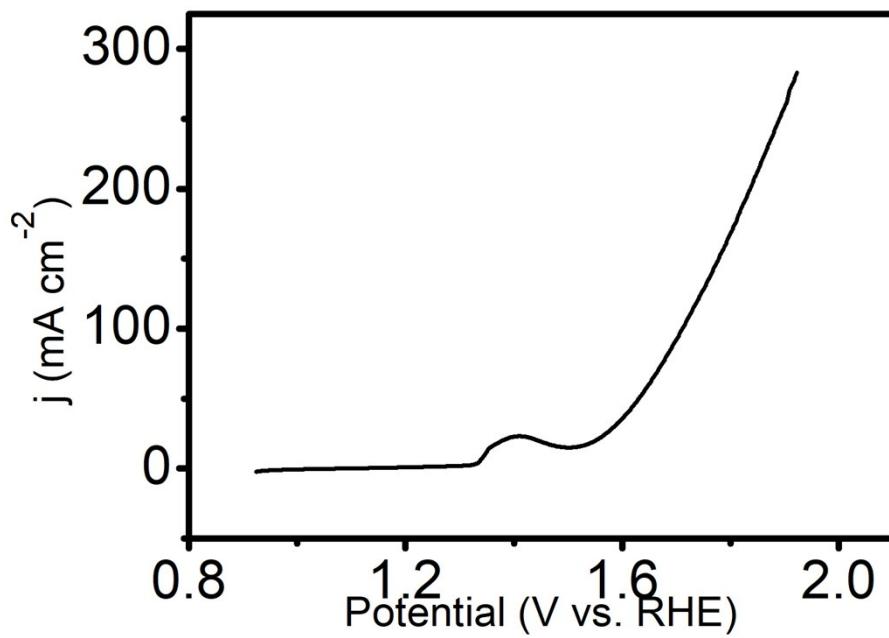


Fig. S2. LSV spectrum of $\text{Fe}_5\text{O}_7(\text{OH})\bullet 4\text{H}_2\text{O}/\text{NF}$ after high-temperature treatment.

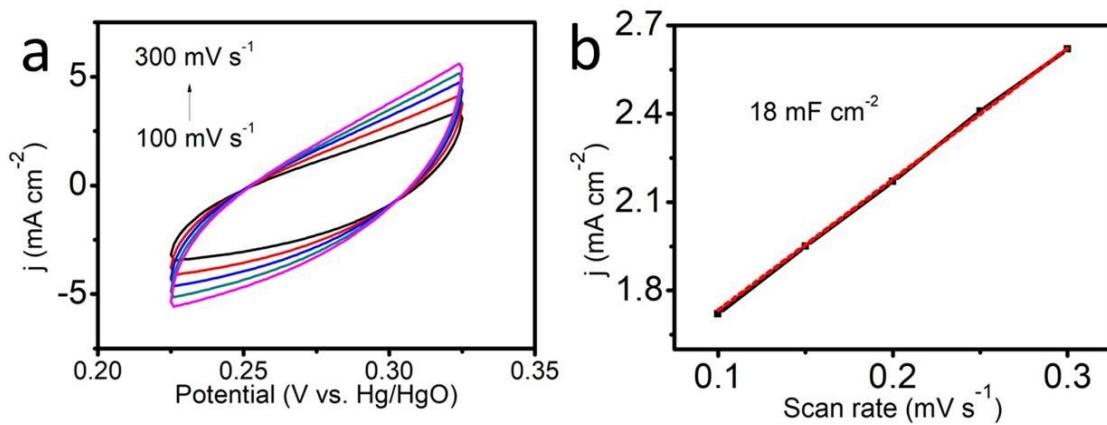


Fig. S3. Cyclic voltammograms of (a) $\text{Fe}_5\text{O}_7(\text{OH})\bullet 4\text{H}_2\text{O}/\text{NF}$ in the non-faradaic capacitance current range at scan rates of 100, 150, 200, 250 and 300 mV s^{-1} . (d) The linear dependence of capacitive current at 0.27 V vs. Hg/HgO on the scan rate for $\text{Fe}_5\text{O}_7(\text{OH})\bullet 4\text{H}_2\text{O}/\text{NF}$.

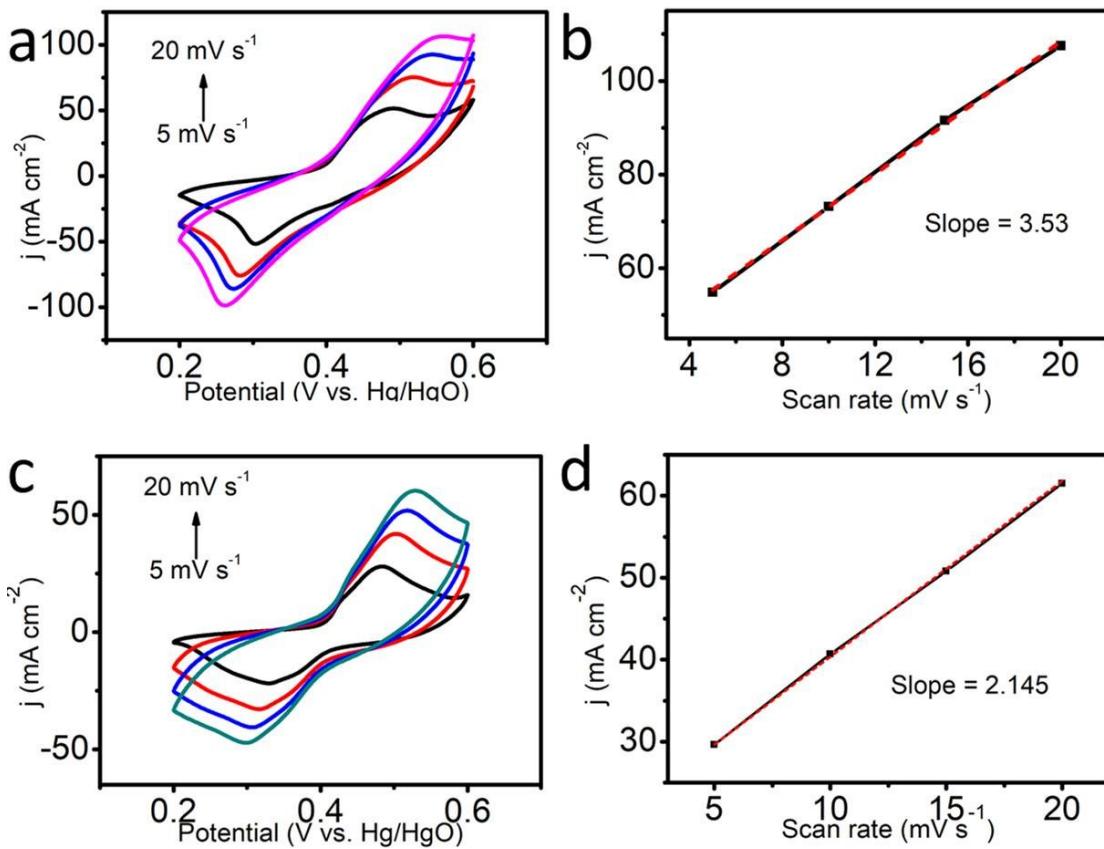


Fig. S4. Cyclic voltammograms of (a) $\text{Fe}_5\text{O}_7(\text{OH}) \bullet 4\text{H}_2\text{O}/\text{NF}$ and (c) $\text{Fe}_5\text{O}_7(\text{OH}) \bullet 4\text{H}_2\text{O}/\text{NF}$ after heat treatment at scan rates of 5, 10, 15, and 20 mV s^{-1} . (b, d) The linear plot related to the oxidation currents for redox species as a function of scan rates for (b) $\text{Fe}_5\text{O}_7(\text{OH}) \bullet 4\text{H}_2\text{O}/\text{NF}$ and (d) $\text{Fe}_5\text{O}_7(\text{OH}) \bullet 4\text{H}_2\text{O}/\text{NF}$ after heat treatment.

Table S1. Comparison of catalytic performance for $\text{Fe}_5\text{O}_7(\text{OH}) \bullet 4\text{H}_2\text{O}/\text{NF}$ with other Fe-based OER electrocatalysts under alkaline conditions.

Catalyst	j (mA cm^{-2})	η (mV)	Electrolyte	Ref.
$\text{Fe}_5\text{O}_7(\text{OH}) \bullet 4\text{H}_2\text{O}/\text{NF}$	100	269	1.0 M KOH	This work
NiFe_2O_4	10	262	1.0 M KOH	1
$\text{NF}@\text{NC}-\text{CoFe}_2\text{O}_4$	100	290	1.0 M KOH	2
$\text{NiO}/\text{NiFe}_2\text{O}_4$	10	303	1.0 M KOH	3
Fe_xN	100	290	1.0 M KOH	4
$\text{Ni}@\text{FeOOH}$	100	370	1.0 M KOH	5

$\text{Fe}_{1.3}\text{Mn}_{0.3}\text{P}$	10	490	1.0 M KOH	6
FeP	100	420	1.0 M KOH	7
$(\text{Co}_{0.47}\text{Fe}_{0.53})_2\text{P}$	10	290	1.0 M KOH	8
Fe-O ₂ cat	100	537	1.0 M KOH	9
Iron oxyhydroxide	20	510	1.0 M KOH	10
NiFe LDH/NF	10	269	1.0 M KOH	11
NiFe-LDH/CNT	10	250	1.0 M KOH	12
Ni-Fe (oxy) hydroxide	10	275	1.0 M KOH	13
NiFe-LDH	10	309±2	0.1 M KOH	14
Fe-doped Ni ₂ P/NF	50	230	1.0 M KOH	15
Fe ₃ C/NF	10	262	1.0 M KOH	16
FeS-Ni ₃ S ₂ /NF	100	290	1.0 M KOH	17
Fe-MOF/NF	100	270	1.0 M KOH	18

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