Supplementary Information

Facile and Simple Microwave-Assisted Synthesis Method for Mesoporous Ultrathin Iron Sulfide Nanosheets as an Efficient Bifunctional Electrocatalyst For Overall Water Splitting

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Material Characterization

X-ray diffractometry (PANalytical XRD, with Cu Ka radiation) was used for the investigation of crystallographic phase. Microstructures and morphology of samples were observed via field emission scanning electron microscopy (FESEM, JEOL JEM-2100 F) fortified with energy-dispersive X-ray spectroscopy (EDS), transmission electron microscopy (TEM, JSM- 2100F, 200 kV), and high-resolution TEM (HRTEM, FEI Tecnai G2 F20, 200 kV). Using Veeco instrument atomic force microscopy (AFM) had been performed. Brunauer-Emmett-Teller surface areas (BET) was used for investigation of the specific surface area. PHI Quanteral II (Japan) with an Al K= 280.00 eV excitation source was used for the measurements of X-ray photoelectron spectroscopy (XPS).

Electrochemical Measurements

Electrochemical work station (CHI-660E) had been utilized for HER and OER in a three electrode system. Two electrode schemes has been used for overall water splitting, and Ag/AgCl and Pt foil had obtained as reference and counter electrodes for electrochemical measurements.

In the engineering of working electrode, a water-resistant insulator mask was firmly taped on uncontaminated rectangular carbon fiber paper (CFP) (0.5 cm x 5 cm) to characterize a 3 mm diameter circular active area and 3.5 mg of Fe_3S_4 was uniformly dissolved in water/ethanol solution (1/0.880 ml) and set it on sonication for 30 minutes, 60 µl of Nafion solution (Sigma Aldrich, 5wt%) were uniformly mixed and sonicated for 30 min. After that, catalytic electrode had been made through drop wise 4µl of the slurry on CFP and desiccated it at room temperature. Reversible Hydrogen Electrode used as a standard for all potentials as follows:

$$E(RHE) = E(Ag/AgCl) + 0.059\ pH + Eo$$

Overall water electrolyzer is prepared with 7µl of the ink had been coated on 0.05 cm × 1 cm active area of CFP electrode, earlier than testing dried out at room temperature.

Proposed reaction mechanism for OER

$$MX+OH^- \longrightarrow MOOH+OX^-$$

$$MOOH+OH^- \longrightarrow MO(OH)_2+e^-$$

$$M(OH)_2+OH^- \longrightarrow [MO(OH)_2]^++e^-$$

$$[M(OH)_2]^++2OH^- \longrightarrow [MO]^++O_2+2H_2O+2e^-$$

$$[MO]^++OH^- \longrightarrow MOOH$$

Here M is considered for Fe and X is considering for S.

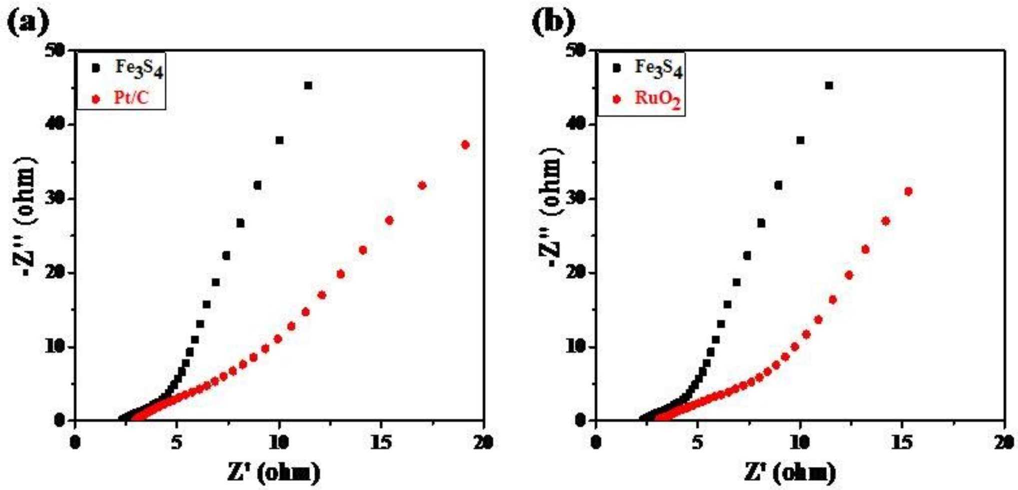

Figure S1.(a) Nyquist Plots of Pt/C and Fe_3S_4, (b) Nyquist Plots of RuO_2 and Fe_3S_4

Table S1. Comparison of the HER performances of Fe_3S_4 with the best-reported iron sulfde and other reported non-precious HER electrocatalysts.

Catalyst	Morphology	Electrolyte	Over potential (η10)(mV)	Tafel Slope (mV dec^{-1})	Ref.
Fe_3S_4	Nanosheets	1 M KOH	103	95	This work
FeS_2/C/NF	Nanoparticles	1 M KOH	202	98	1
Fe@$FeO_xS_yFeS_2$	Nanoparticles	1 M KOH	243 (η100)	77	2
FeS_2-rGO	Nanoparticles	0.5 M H_2SO_4	139	66	3
FeS_2-RGO	Lemellar Structure	0.5 M H_2SO_4	226	61	4
$FeNi_{0.20}S_2$	Nanosheet	0.5 M H_2SO_4	183	78.63	5
FeS_2-doped Mo S_2	Nanoflower	0.5 M H_2SO_4	136	82	6
Fe_3S_4	Particles	1 M KOH	279 (η50)	87.40	7
$Fe_{0.9}Co_{0.1}S_2$/CNT	Nanosheet	0.5 M H_2SO_4	120(η20)	46	8
$CoNi_2S_4$@CoS_2/NF	Microspheres	1 M KOH	173	45	9

$NiCo_2S_4$	Nanoparticles	1 M KOH	282	-	[10]
Co_9S_8	Naonowires	1 M KOH	217	110	[11]
$N-Ni_3S_2/NF$	Naonowires	1 M KOH	105	108	[12]
CoS_2	Nanospheres	1 M KOH	193	100	[13]
Co_3S_4-L	Nanosheet	1 M KOH	270	124.5	[14]
$Co(OH)_2/NiCoS$	Nanotubes	1 M KOH	148	88	[15]
Ni-S-B	Microspheres	30%wt KOH	240	121.2	[16]
$N-Ni_3S_2$@C/NF	Nanoflakes	1 M KOH	113	90	[17]
NiS_2	Nanospheres	1 M KOH	147	105	[18]
CoS_x	Nanosheets	1M KOH	127	123	[19]
$NiSe_2$	Nanosheets	1M KOH	184	184	[20]
Co_9S_8@MoS_2	Octahedrons/CNFs	1M KOH	190	110	[21]
CoP	Nanowires/CC	1M KOH	110	129	[22]
Ni_2P	Nanosheets	1M KOH	96	94	[23]
MoS_2	Film	0.5M H_2SO_4	260	50	[24]
WS_2	Nanosheets	0.5M H_2SO_4	250	60	[25]

Table S2. Comparison of the OER performances of Fe3S_4 with the best-reported iron sulfide and other reported non-precious OER electrocatalysts.

Catalyst	Morphology	Electrolyte	Over potential (η10) mV	Tafel Slope (mV dec^{-1})	Ref.
Fe_3S_4	Nanosheet	1 M KOH	230	50	This work
FeS_2/C/NF	Nanoparticles	1 M KOH	240	92	[26]
Fe@FeO_xS_y	Nanosheet	1 M KOH	238	82.7	[27]
Fe_3S_4	Particles	1 M KOH	255(η50)	46	[7]

$FeNiS_2$	Nanosheet	0.1 M KOH	310	46	[9]
$CoNi_2S_4@CoS_2/NF$	Peapod	1 M KOH	173	51	[11]
Co_9S_8	Naonowires	1 M KOH	299	67	[13]
CoS_2	Nanospheres	1 M KOH	290	57	[14]
Co_3S_4-L	Nanosheet	1 M KOH	350	84.7	[15]
$Co(OH)_2$/NiCoS	Nanotubes	1 M KOH	300	64	[17]
N-Ni_3S_2@C/NF	Nanoflakes	1 M KOH	310(η100)	75	[28]
NiS_2	Nanospheres	1 M KOH	271	65	[29]
Ni/NiS/NC	Nanoparticles	1 M KOH	310	75	[30]
CoS	Thin Film	1 M KOH	300	57	[31]
CoS/CNT	Flower	1 M KOH	330	142	[32]
Co_9S_8	Nanosheets	1M KOH	288	79	[33]
CoP	Film	1M KOH	345	47	[34]
Ni_2P	Nanosheets	1M KOH	255	57	[23]
NiMnCoS@rGO	Nanoparticles@sheets	1M KOH	320	53	[35]
CoP	Nanosheets	1M KOH	265	63	[36]

Table S3. Comparison of overall water splitting performances of $Fe_3S_4||Fe_3S_4$ with the best reported bi-functionalelectrocatalysts in the basic electrolyte.

Material	Morphology	Electrolyte	E at j= 10 mA cm^{-2} (V)	Ref.
Fe_3S_4	Nanosheet	1 M KOH	1.43	This work
FeS_2/C/NF	Nanoparticles	1 M KOH	1.72	[26]
Fe@FeO_xS_y	Nanosheet	1 M KOH	1.65	[2]
Fe_3S_4	Crumbled sheets	1 M KOH	1.68	[9]
Co_9S_8	Naonowires	1 M KOH	1.66	[14]
CoS_2	Nanospheres	1 M KOH	1.68	[15]
Co_3S_4-L	Nanosheet	1 M KOH	1.63	[17]
$Co(OH)_2$/NiCoS	Nanotubes	1 M KOH	1.62	[15]
N-Ni_3S_2@C/NF	Nanoflakes	1 M KOH	1.57	[29]
NiS_2	Nanospheres	1 M KOH	1.66	[18]
Ni/NiS/NC	Nanoparticles	1 M KOH	1.61	[31]
NiCoP	Hollow nanobricks	1 M KOH	1.62	[37]
Ni_2P	Nanosheets	1 M KOH	1.47	[23]
CuO@Ni/NiFe$(OH)_2$	Nanoarrays	1 M KOH	1.73	[38]
CoP/NF	Crystalline nanomaterial	1 M KOH	1.65	[39]

CoFeZrO	Nanosheet	1 M KOH	1.63	40
Co_9S_8	Nanosheets	1M KOH	1.55	19

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