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Electronic Supplementary Information

One-pot synthesis of Mn-Fe bimetallic oxide heterostructure as bifunctional electrodes for efficient overall water splitting

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Fig. S1. (a) OER polarization curves and (b) HER polarization curves of Mn_3O_4/Fe_2O_3 -NF complex, $Mn_3O_4+Fe_2O_3$ -NF complex, Fe_2O_3 -NF, and Mn_3O_4 -NF in 1.0 M KOH. (c) XRD pattern and (d) SEM image of Mn_3O_4/Fe_2O_3 (Fe:Mn ratio of 1:1).



Fig. S2. SEM and magnified SEM images of MnO₂-NF (a, b), MnFeO-NF-0.2 (c, d), MnFeO-NF-0.4 (e, f), MnFeO-NF-0.6 (g, h), MnFeO-NF-0.8 (i, j), and MnFeO-NF-1 (k, l), respectively.



Fig. S3. EDS data of (a) MnFeO-NF-0.2, (b) MnFeO-NF-0.4, (c) MnFeO-NF-0.6, (d) MnFeO-NF-0.8, and (e) MnFeO-NF-1. Inset image is a table of atomic percent of Mn and Fe.



Fig. S4. (a) XPS survey of MnFeO-NF-0.2, MnFeO-NF-0.4, MnFeO-NF-0.6, MnFeO-NF-0.8, and MnFeO-NF-1. (b–d) High resolution XPS spectra of Mn 2p, Fe 2p, and O 1s.



Fig. S5. (a) Polarization curves and (b) Tafel plots of MnFeO-NF-0.2, MnFeO-NF-0.4, MnFeO-NF-0.6, MnFeO-NF-0.8, and MnFeO-NF-1 in 1.0 M KOH.



Fig. S6. EIS of MnFeO-NF-0.2, MnFeO-NF-0.4, MnFeO-NF-0.6, MnFeO-NF-0.8, MnFeO-NF-1, MnO₂-NF, Mn₃O₄-NF, and Fe₂O₃-NF at 1.533 V *vs.* RHE.



Fig. S7. CV curves at different scan rates for catalysts: (a) MnFeO-NF-0.2, (b) MnFeO-NF-0.4, (c) MnFeO-NF-0.6, (d) MnFeO-NF-0.8, (e) MnFeO-NF-1, (f) MnO₂-NF, (g) Mn₃O₄-NF, and (h) Fe₂O₃-NF, (i) Plots of capacitive currents with different scan rates for various catalysts to determine C_{dl} .



Fig. S8. (a) SEM image of MnFeO-NF-0.4 after stability test. (b–d) High resolution XPS spectra of Mn 2p, Fe 2p, and O 1s after stability test.



Fig. S9. (a) Polarization curves and (b) Tafel plots of MnFeO-NF-0.2, MnFeO-NF-0.4, MnFeO-NF-0.6, MnFeO-NF-0.8, and MnFeO-NF-1 in 1.0 M KOH.



Fig. S10. EIS of MnFeO-NF-0.2, MnFeO-NF-0.4, MnFeO-NF-0.6, MnFeO-NF-0.8, MnFeO-NF-1, MnO₂-NF, Mn₃O₄-NF, and Fe₂O₃-NF at -0.177 V *vs.* RHE.



Fig. S11. CV curves at different scan rates for catalysts: (a) MnFeO-NF-0.2, (b) MnFeO-NF-0.4, (c) MnFeO-NF-0.6, (d) MnFeO-NF-0.8, (e) MnFeO-NF-1, (f) MnO₂-NF, (g) Mn₃O₄-NF, and (h) Fe₂O₃-NF. (i) Plots of capacitive currents with different scan rates for various catalysts to determine C_{dl} .



Fig. S12. (a) SEM image of MnFeO-NF-0.8 after stability test. (c-d) High resolution XPS spectra of Mn 2p, Fe 2p, and O 1s after stability test for MnFeO-NF-0.8.



Fig. S13. Electrolyzation installation diagram of the MnFeO-NF-0.8 || MnFeO-NF-0.4.

 Table S1. Comparison of the overpotentials for OER of different catalysts in this work.

Catalyst	$\eta_{10}(\mathrm{mV})$	$\eta_{100}(\mathrm{mV})$	$\eta_{500}(\mathrm{mV})$
MnFeO-NF-0.2	227	274	295
MnFeO-NF-0.4	157	225	257
MnFeO-NF-0.6	220	285	312
MnFeO-NF-0.8	243	319	356
MnFeO-NF-1	258	340	405
MnO ₂ -NF	342	433	464
Mn ₃ O ₄ -NF	339	438	481
Fe ₂ O ₃ -NF	318	432	
NF	363	482	538

Table S2.	Comparison of the ov	erpotentials for	HER of different	ent catalysts in this
work.				

Catalyst	$\eta_{10}(\mathrm{mV})$	$\eta_{100}(\mathrm{mV})$	η ₅₀₀ (mV)
MnFeO-NF-0.2	186	295	347
MnFeO-NF-0.4	162	286	348
MnFeO-NF-0.6	153	235	314
MnFeO-NF-0.8	63	139	192
MnFeO-NF-1	143	231	280
MnO ₂ -NF	151	239	300
Mn ₃ O ₄ -NF	186	308	415
Fe ₂ O ₃ -NF	181	294	392
NF	171	270	326

Electrocatalyst	η for OER at target j (mV@mA cm ⁻²)	η for HER at target j (mV@mA cm ⁻²)	Voltage for OWS at target j (mV@mA cm ⁻²)	Ref.
δ -MnO ₂ /NF	320@10	196@10		1
(Fe _{1-x} ,Mn _x)OOH	246@10			2
Ni ₁₁ (HPO ₃) ₈ (O		84@100		3
H) ₆ /Mn ₃ O ₄				
NiCoFe	240@10	231@10	1.52@10	4
phosphate NSs-				
C/NF				
NiFe/NiCo ₂ O ₄ /	340@1200	105@10	1.67@10	5
Ni Foam				
Hierarchical	290@10	110@10	1.65@10-	6
NiCo ₂ O ₄				
Co@Co ₃ O ₄	391@10	221@10		7
NiCo-nitrid	183@10	71@10	1.68@20	8
es/NiCo ₂ O ₄ /GF				
NiCo ₂ S ₄	260@10	210@10	1.63@10	9
NW/NF				
NiCo ₂ S ₄ /Ni ₃		119@10		10
S ₂ /NF				
Mn ₃ O ₄ /Fe ₂ O ₃ /	213@50	63@10	1.59@10	This
NF				work

Table S3. Comparison of activity for HER, OER, and overall water splitting of $Mn_3O_4/Fe_2O_3/NF$ with other electrocatalysts in 1.0 M KOH.

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