

Supporting Information

Defect-enriched Active Self-supported Coralline-like High-Entropy Spinel Oxide (FeNiCoMnCr)₃O₄ electrodes for High-stability Oxygen Evolution Reaction

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Table S1 Specific surface areas of the catalysts.

Catalysts	HEO@NF	HEO-H400@NF	HEO-H500@NF	HEO-H600@NF
$S_{\text{BET}}(\text{m}^2/\text{g})$	17.486	24.164	42.515	38.778

Table S2 XPS Analysis of Ni Elements on nickel foam self-supported electrode after H_2 treatment at different temperatures

Sample	Ni(852.5 eV)	Ni^{2+} (854.85 eV)	Ni^{3+} (856.60 eV)
HEO@NF	--	41.13	58.87
HEO-H400@NF	16.31	44.37	39.32
HEO-H500@NF	16.30	48.08	35.62
HEO-H600@NF	33.83	25.63	40.55

Table S3 XPS Analysis of HEO@NF and HEO-H500@NF

Element	HEO@NF	HEO-H500@NF
Fe^{2+} (710.5 eV)	69.43	49.69
Fe^{3+} (712.5 eV)	30.57	50.31
Ni (852.5 eV)		16.30
Ni^{2+} (854.85 eV)	41.13	48.08
Ni^{3+} (856.60 eV)	58.87	35.62
Co^{2+} (782.2 eV)	48.15	40.75
Co^{3+} (780.2 eV)	51.85	59.25
Mn^{3+} (641.7 eV)	60.74	76.54
Mn^{4+} (644.8 eV)	39.26	23.46
Cr^{3+} (576.2 eV)	64.12	55.81
Cr^{6+} (578.3 eV)	35.88	44.19

Table S4 TOF values for all samples

Sample	j at overpotential of 300 mV.(mA/cm ²)	TOF(s ⁻¹)
HEO@NF	11	3.31
HEO-400@NF	22	6.62
HEO-500@NF	270	81.33
HEO-600@NF	105	31.63
HEO-500+NF	44	13.25

Table S5 Overpotential and tafel slope of spinel metal oxides reported in this paper and literature at 100 mA cm⁻² (All electrolytes were 1 M KOH)

Catalyst	Overpotential at 100 mA cm ⁻² (mV)	Tafel Slope (mV dec ⁻¹)	Reference
HEO-H500@NF	280	40.3	This work
(FeNiCoCuV) ₃ O ₄	308	48.5	[1]
(FeCoNiMnZn) ₃ O ₄	422	53	[2]
(FeCoNiCrMn) ₃ O ₄	300	41.2	[3]
S-(FeNiCoMnCr) ₃ O ₄	285	32.3	[4]
(FeNiCoMnCr) ₃ O ₄	340	53.7	[5]
(FeCoNiCrMn) ₃ O ₄	300	52.4	[6]
(Co Mn ₃ NiFeZn) ₃ O ₄	410	53.5	[7]

Table S6 Overpotential and tafel slope of self-supporting high entropy electrode reported in this paper and literature at 100 mA cm⁻²(All electrolytes were 1 M KOH)

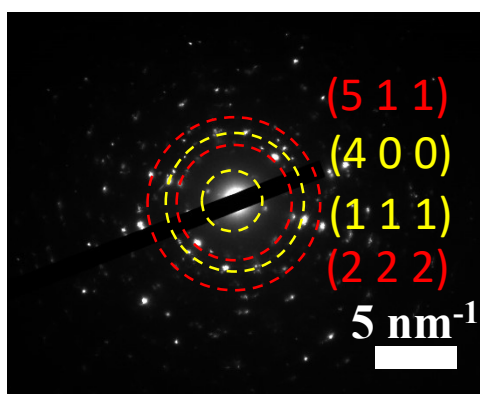
Catalyst	Overpotential at 100 mA cm ⁻² (mV)	Tafel Slope (mV dec ⁻¹)	Reference
HEO-H500@NF	280	40.3	This work
(CrMnFeCoCu) ₃ O ₄ @CC	380	59.4	[8]
(FeCoMnZnMg) ₃ O ₄ @NF	390	59	[9]
FeCoNiCrMn HEA@NF	>320	64.3	[10]
AlFeCoNi ₃ Mo _{0.2} HEA	370	72	[11]
AlCoCrFeNi HEA	366	53.4	[12]
NiCoFeMnCrO@NF	294	46.7	[13]
CoCrFeNiNb HEA	304	42.7	[14]
FeCoNiCrMn HEA@NF	355	49.29	[15]

Table S7 Equivalent Circuit Parameters for Samples of HEO-HX@NF

Sample	Rs(Ω)	Rct (Ω)	Cdl-T	Cdl-P
HEO@NF	1.329	224.9	0.093277	0.74671
HEO-H400@NF	1.378	22.39	0.17609	0.82518
HEO-H500@NF	1.315	11.26	0.10813	0.82345
HEO-H600@NF	1.157	12.59	0.0931	0.83714

Table S8 XPS Analysis of HEO-H500@NF before and after OER

Element	Before OER (at.%)	After OER (at.%)
Fe ²⁺ (710.5 eV)	45.19	49.69
Fe ³⁺ (712.5 eV)	54.81	50.31
Ni(852.5 eV)	16.30	--
Ni ²⁺ (854.85 eV)	48.08	38.18
Ni ³⁺ (856.60 eV)	35.62	68.12
Co ²⁺ (782.2 eV)	40.75	16.35
Co ³⁺ (780.2 eV)	59.25	83.65
Mn ³⁺ (641.7 eV)	76.54	66.43
Mn ⁴⁺ (644.8 eV)	23.46	33.57
Cr ³⁺ (576.2 eV)	55.81	35.30
Cr ⁶⁺ (578.3 eV)	44.19	64.70

**Fig. S1** SEAD image of HEO-H500@NF electrode.

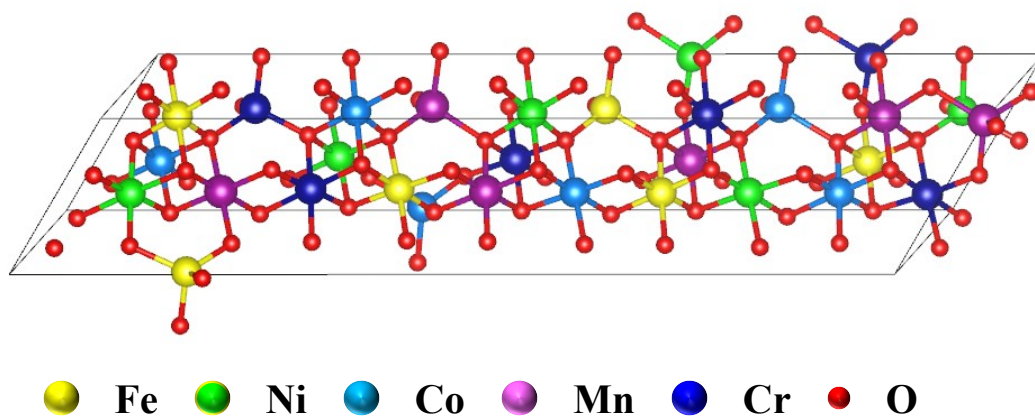


Fig. S2 XRD calculation model of $(\text{FeNiCoMnCr})_3\text{O}_4$

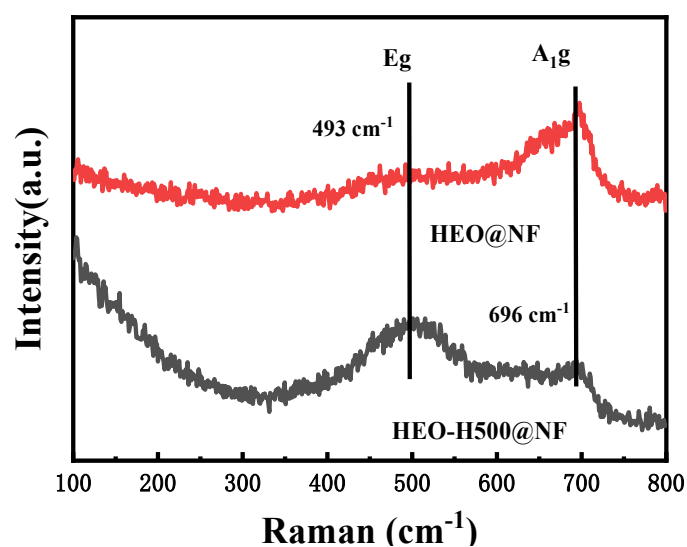


Fig. S3 Raman of HEO@NF and HEO-H500@NF

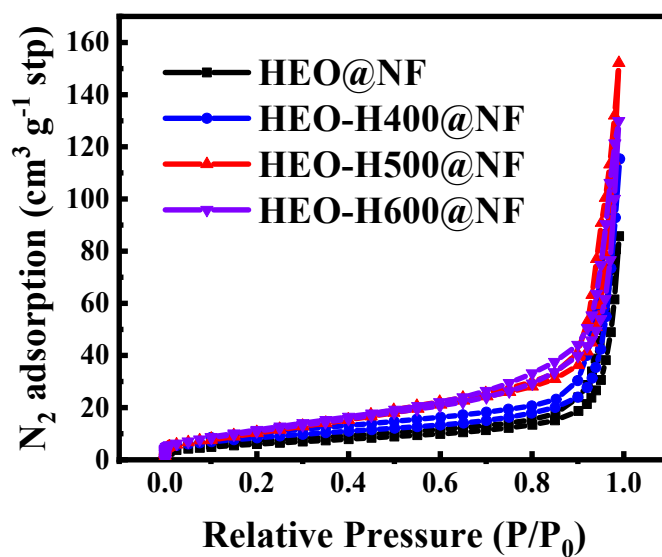


Fig. S4 Nitrogen adsorption-desorption isotherms of self-supported foam nickel electrode after H_2 treatment at different temperatures

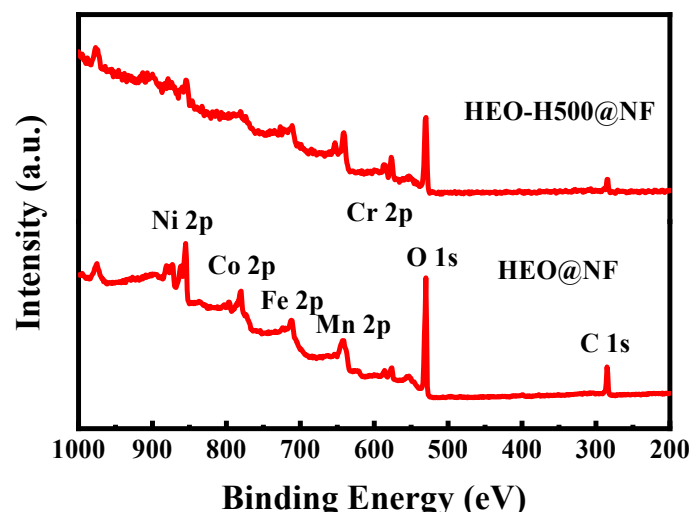


Fig. S5 XPS full spectra of HEO@NF and HEO-H500@NF

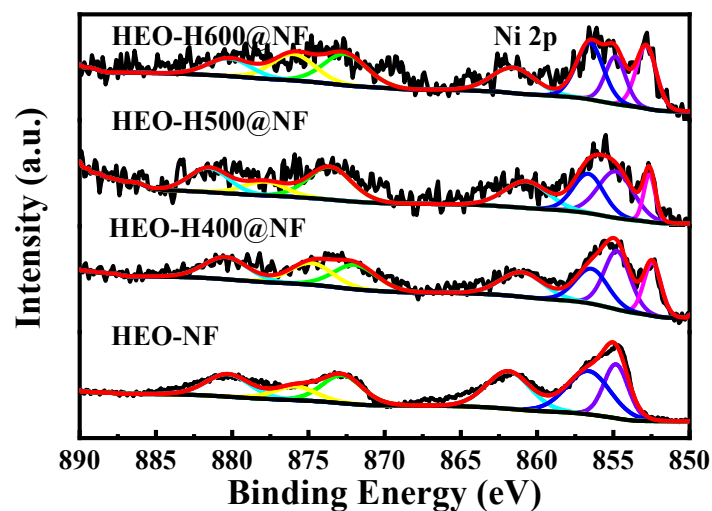


Fig. S6 High resolution Ni 2p spectra of nickel foam self-supported electrode after H₂ treatment at different temperatures

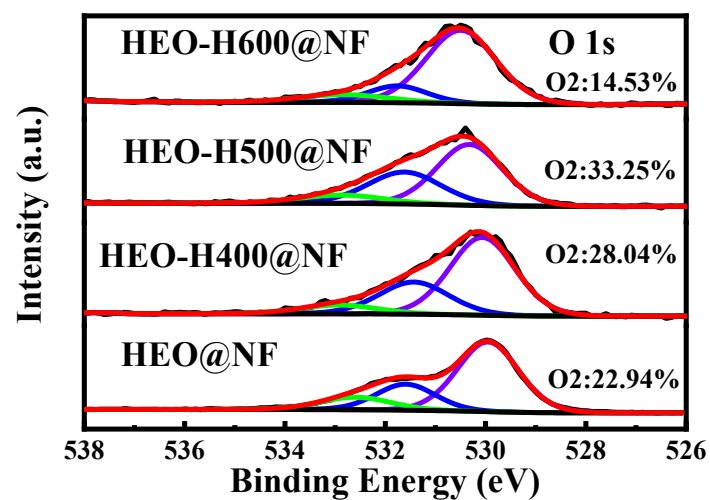


Fig. S7 High resolution O 1s spectra of nickel foam self-supported electrode after H₂ treatment at

different temperatures

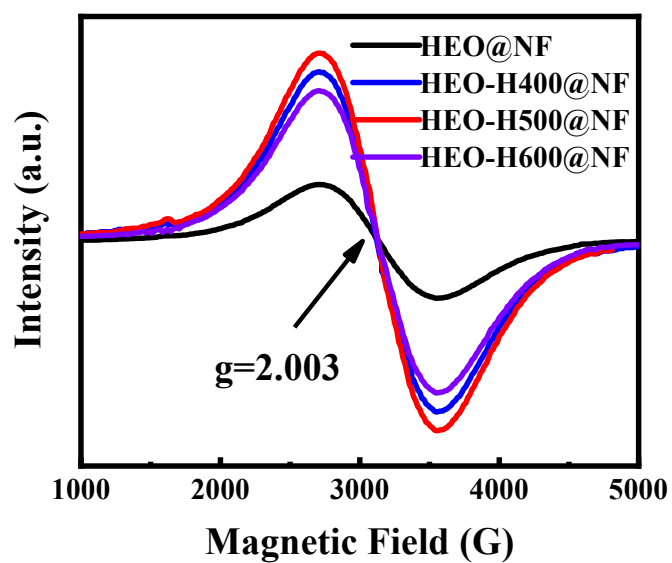


Fig. S8 ESR spectra of nickel foam self-supported electrode after H_2 treatment at different temperatures

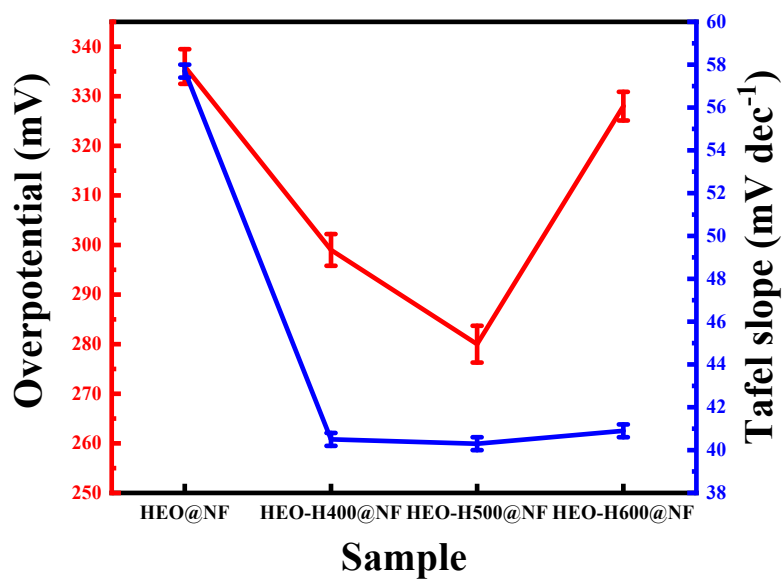


Fig. S9 Over potential(100 mA/cm²) and Tafel slopes of the as-synthesized electrodes in 1.0 m KOH.

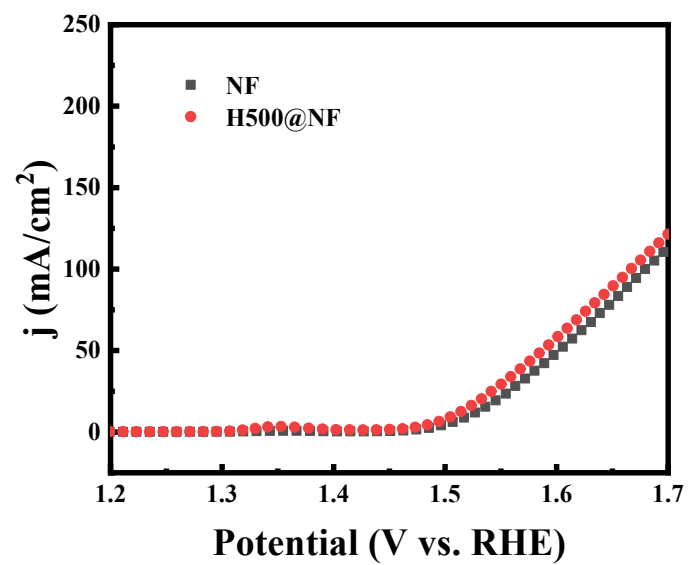


Fig. S10 Electrochemical performances of NF and H500@NF

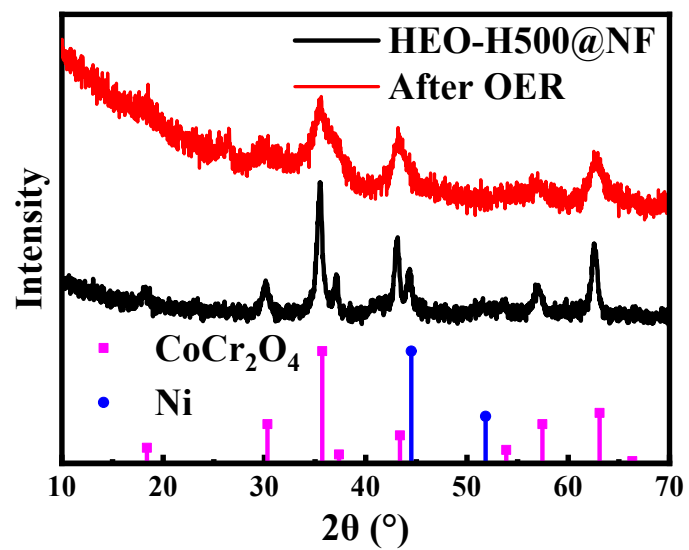


Fig. S11 XRD of HEO-H500@NF before and after activation

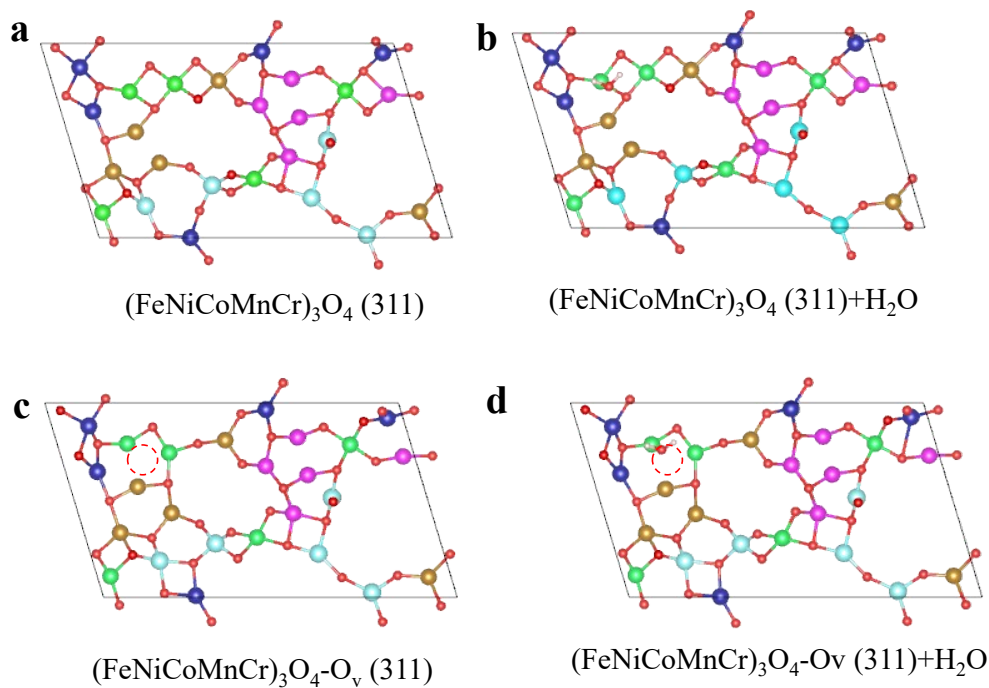


Fig. S12 The adsorption model of H₂O onto the (3 1 1) of (a, b) pristine (FeNiCoMnCr)₃O₄ and (c, d) (FeNiCoMnCr)₃O₄-O_v. (C-axis projection)

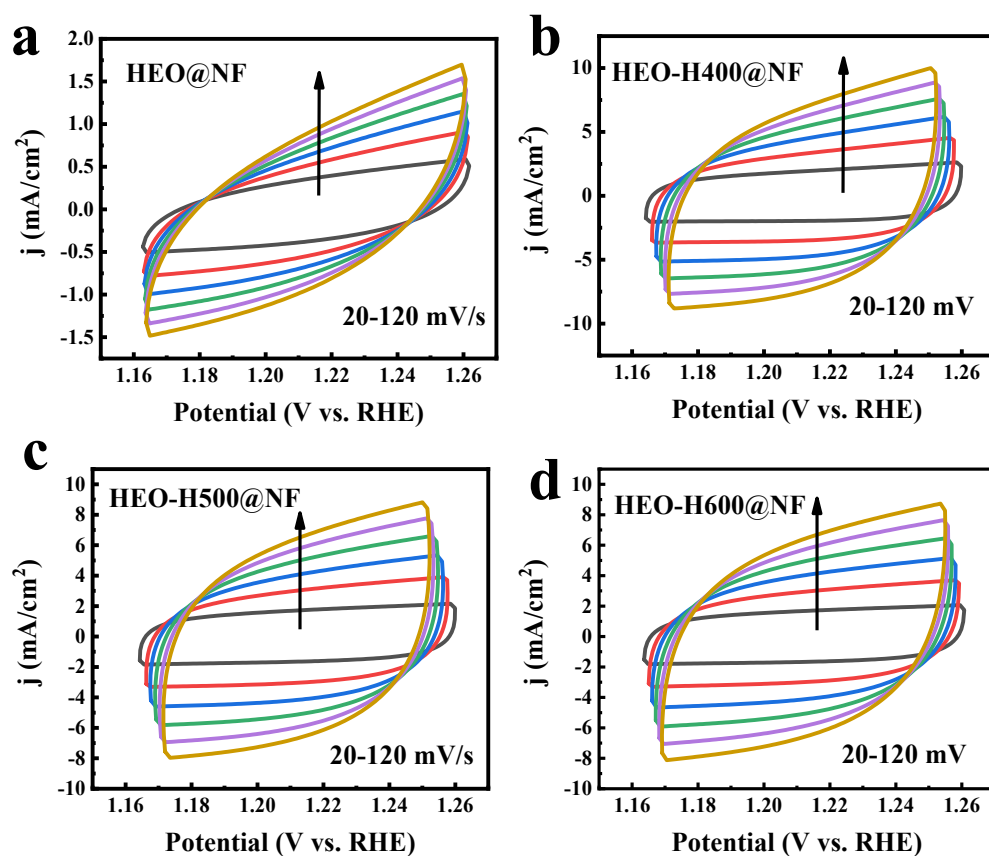


Fig. S13 The CV Cure of nickel foam self-supported electrode after H₂ treatment at different temperatures

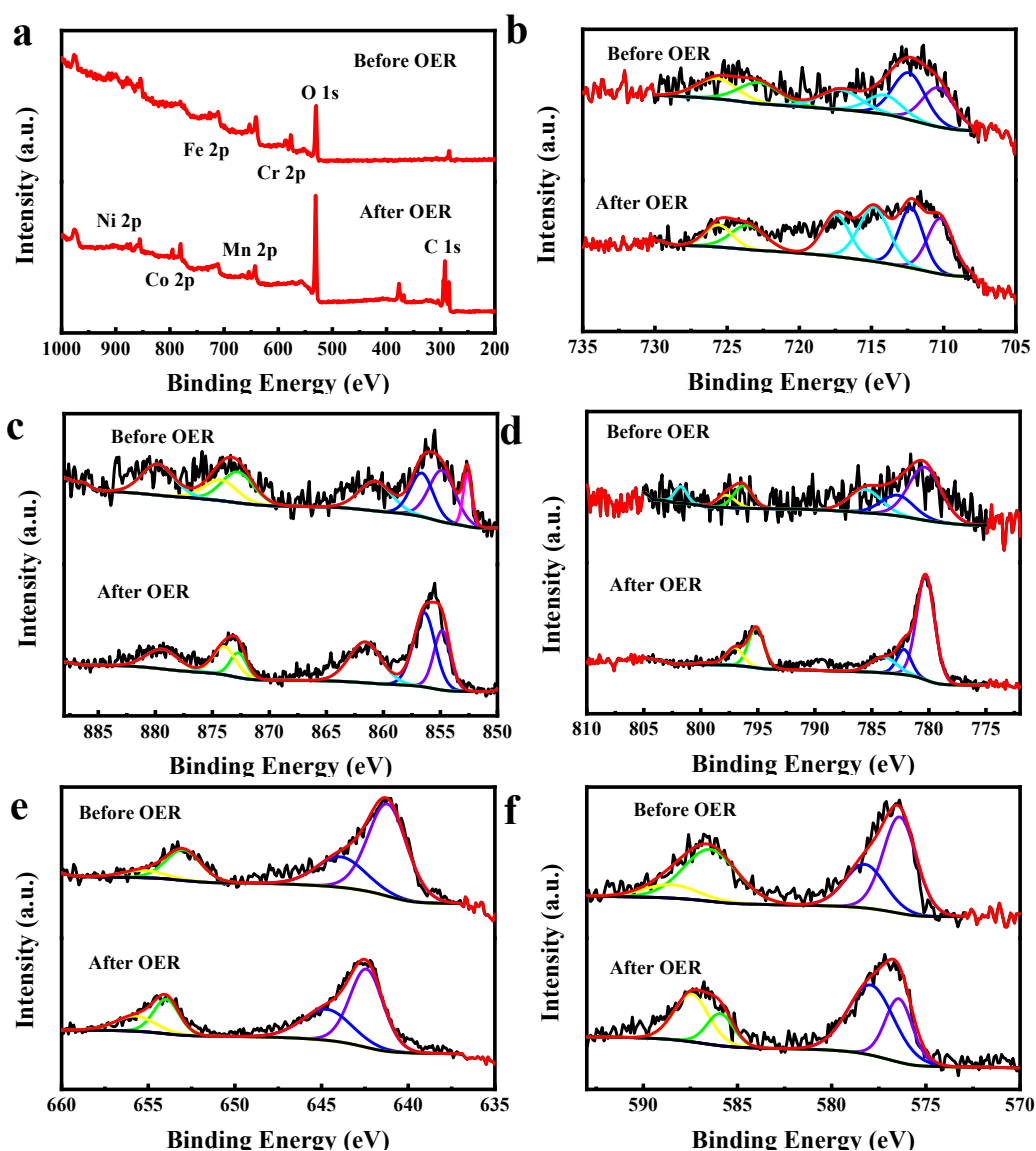


Fig. S14 XPS of HEO-H500@NF Before and after OER (a) full spectra (b) Fe 2p spectra (c) Ni 2p (d) Co 2p spectra (e) Mn 2p spectra (f) Cr 2p spectra

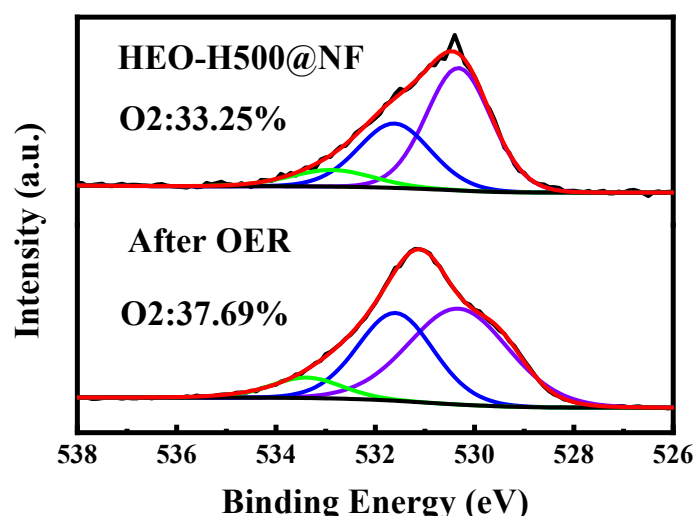


Fig. S15 Before and after OER O1s XPS of HEO-H500@NF

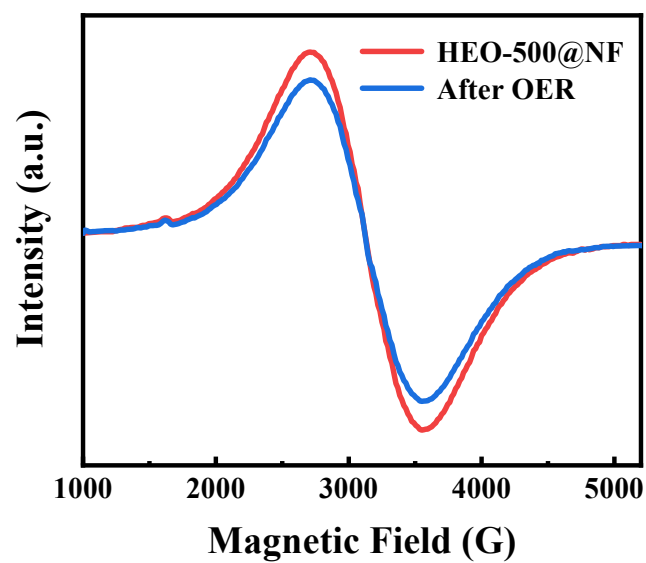


Fig. S16 ESR spectra of HEO-H500@NF before and after OER

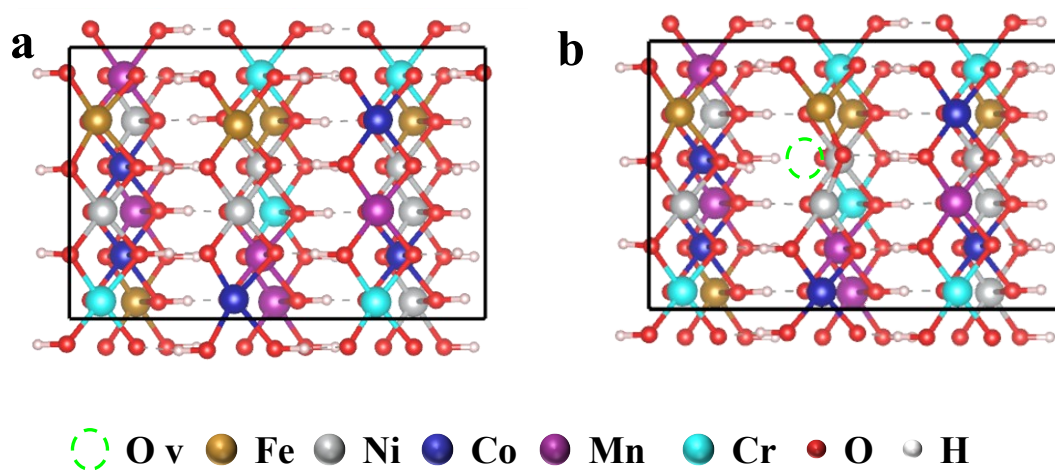


Fig. S17 The calculation model (a) $(\text{FeNiCoMnCr})(\text{OH})_x$ (b) $(\text{FeNiCoMnCr})(\text{OH})_x\text{-Ov}$

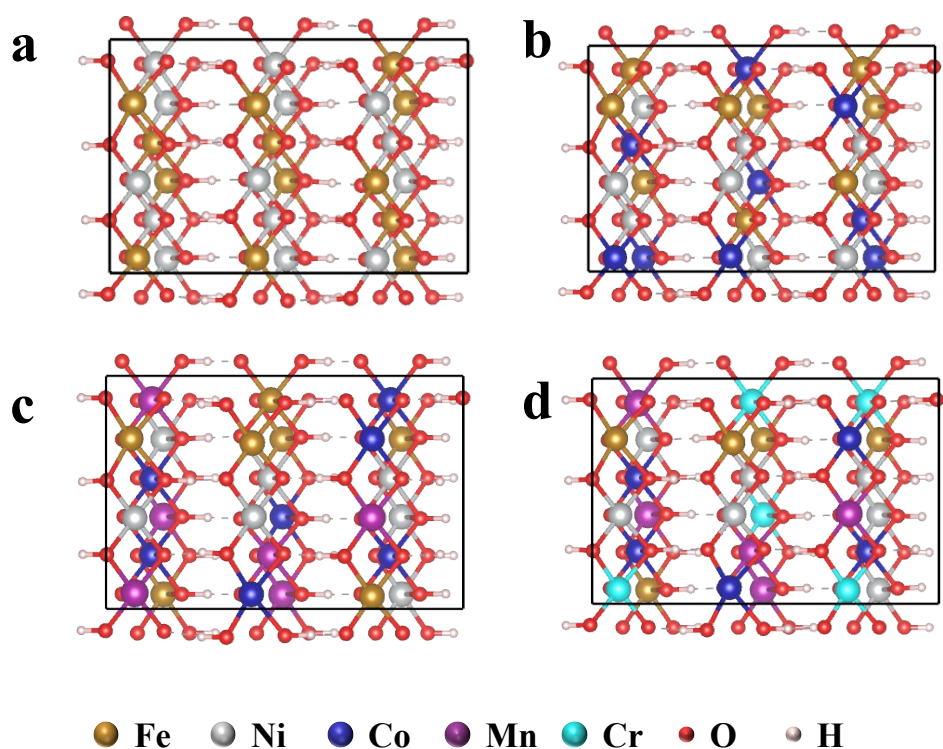


Fig. S18 The calculation model (a)(NiFe)(OH)_x (b)(NiFeCo)(OH)_x (c) (NiFeCoMn)(OH)_x (d)(FeNiCoMnCr) (OH)_x

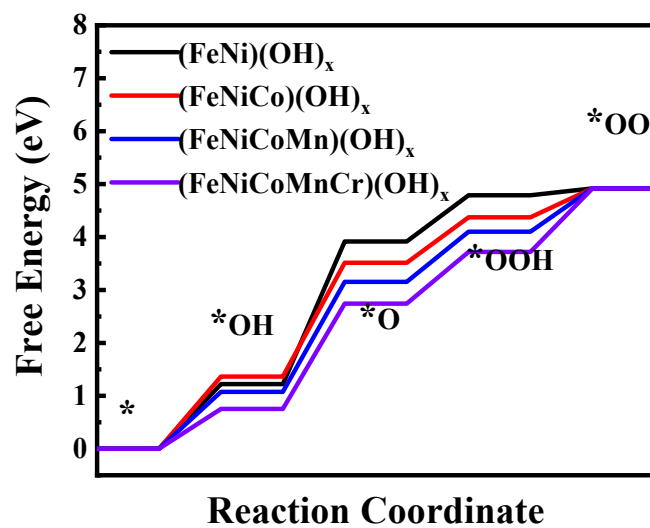


Fig. S19. Gibbs free energies diagram for the OER process catalyzed by (FeNi)(OH)_x, (FeNiCo)(OH)_x, (FeNiCoMn)(OH)_x and (FeNiCoMnCr)(OH)_x.

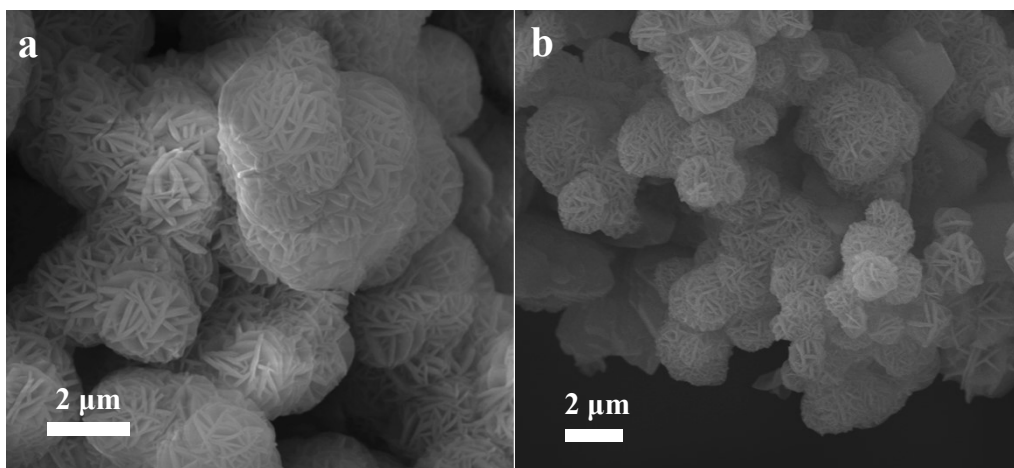


Fig. S20 SEM of HEO-H500@NF before and after activation (a)before OER (b)after OER

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