Supplementary Information

Unraveling the Mechanism of Enhanced Oxygen Evolution Reaction using $NiO_x@Fe_3O_4$ decorated on surface-modified carbon nanotubes

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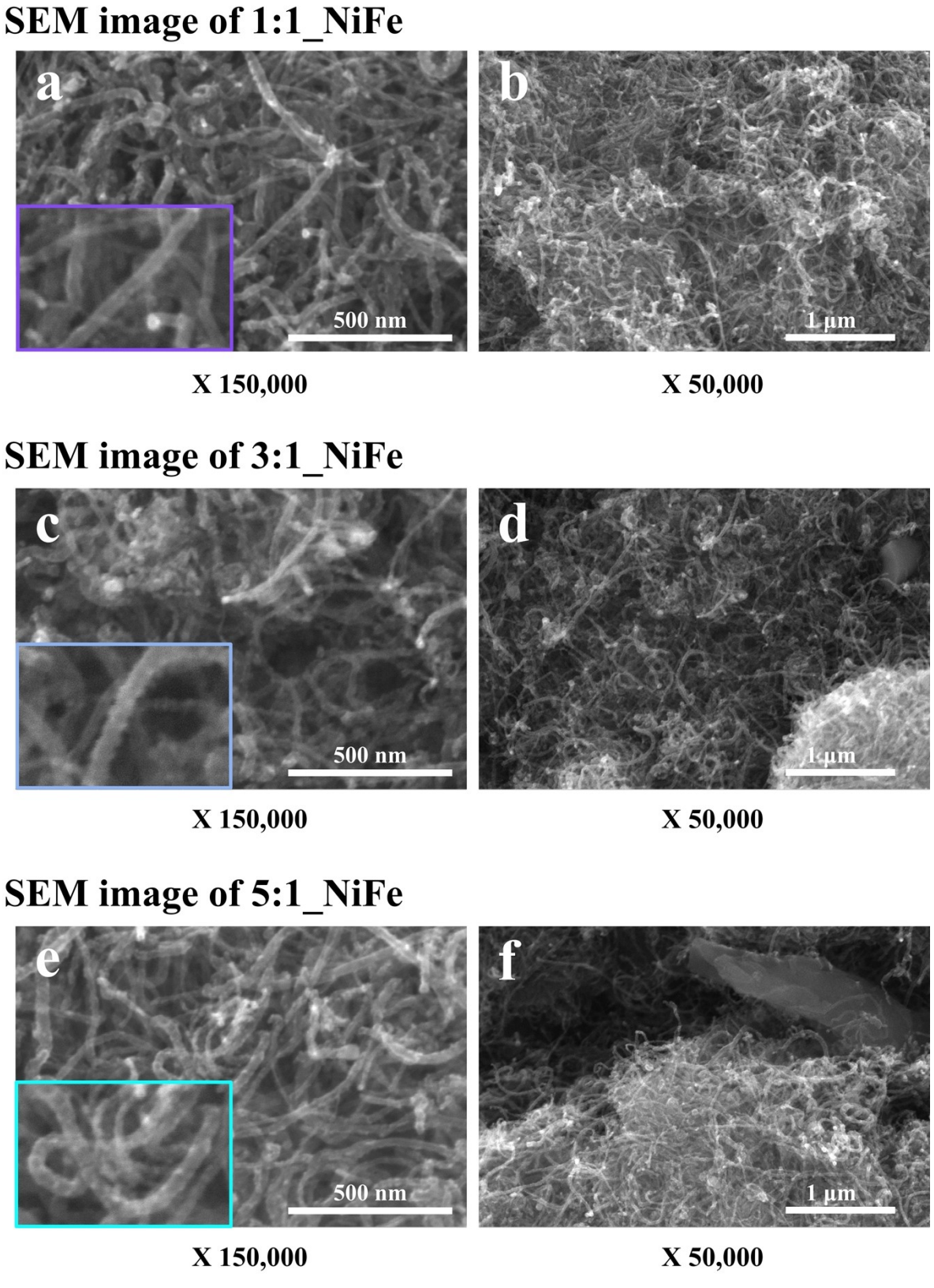
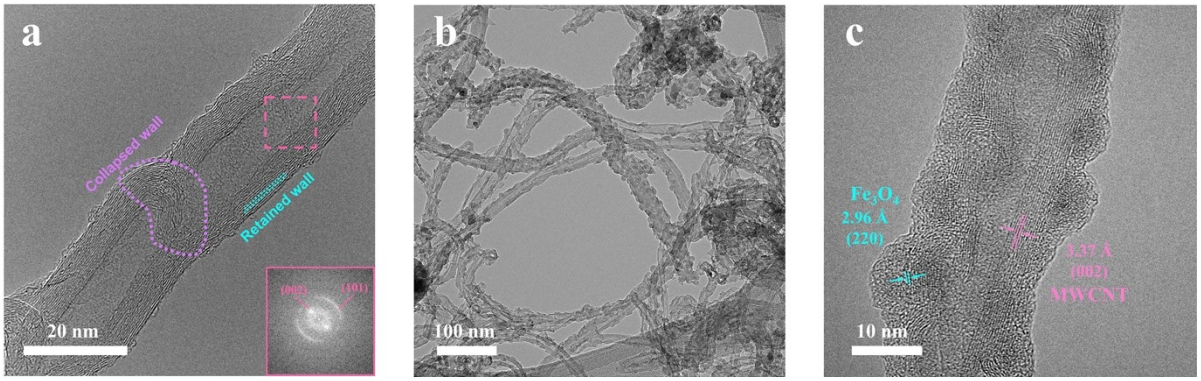
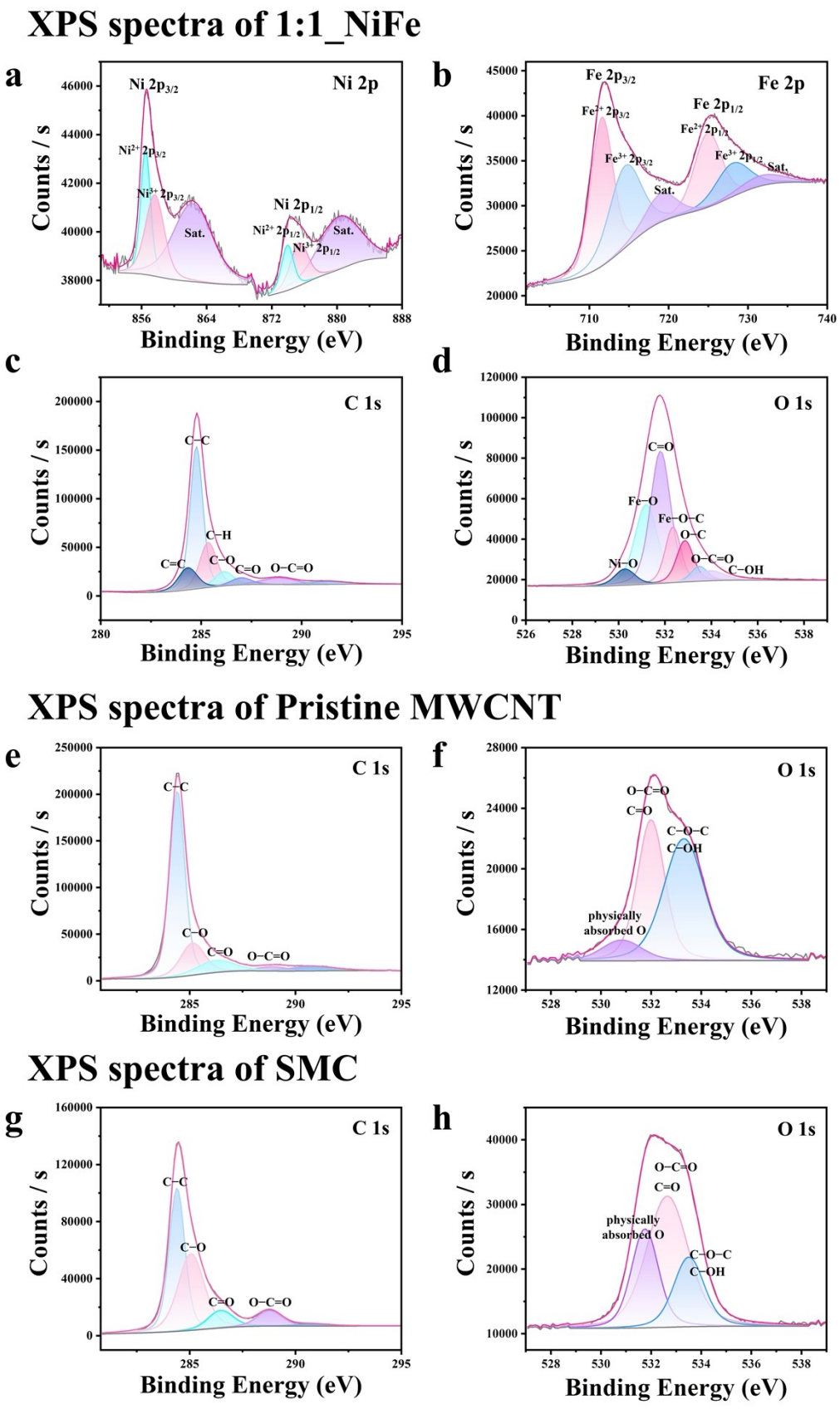


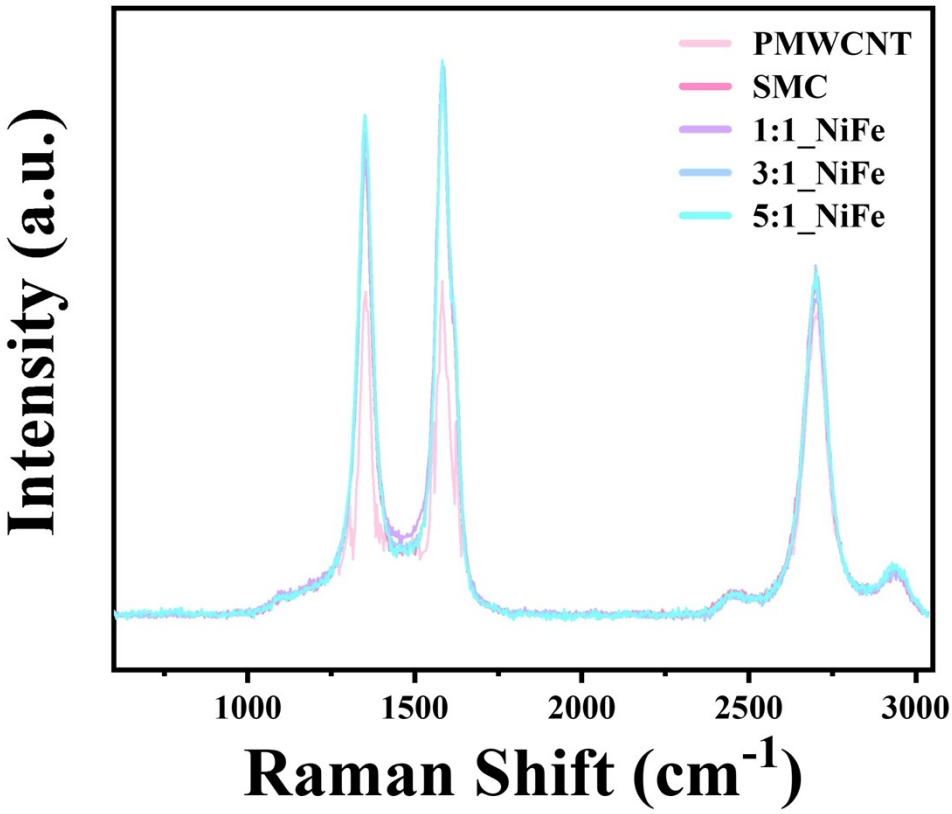
Supplementary Figure 1. SEM images. **a, b** 1:1_NiFe **c, d** 3:1_NiFe **e, f** 5:1_NiFe.



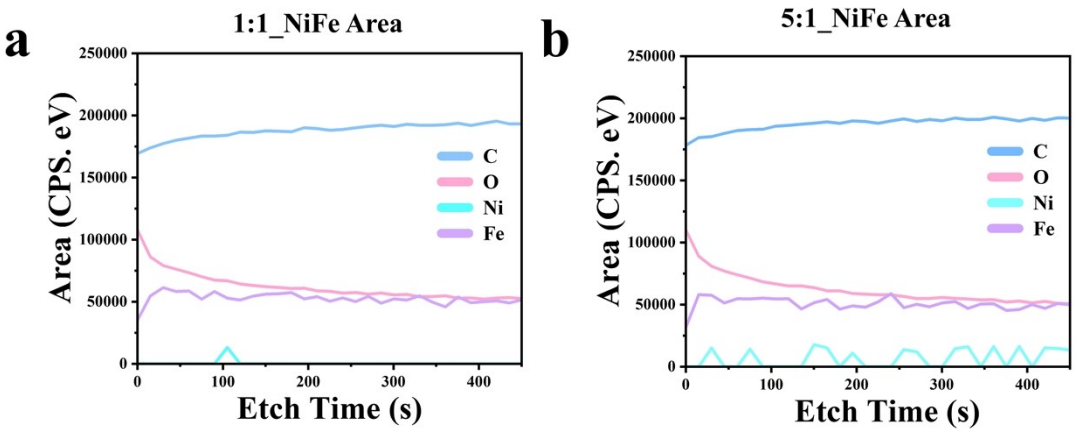
Supplementary Figure 2. TEM images. **a** Collapsed and retained wall SMC. **b** Low resolution image of 5:1_NiFe **c** 1:1_NiFe.



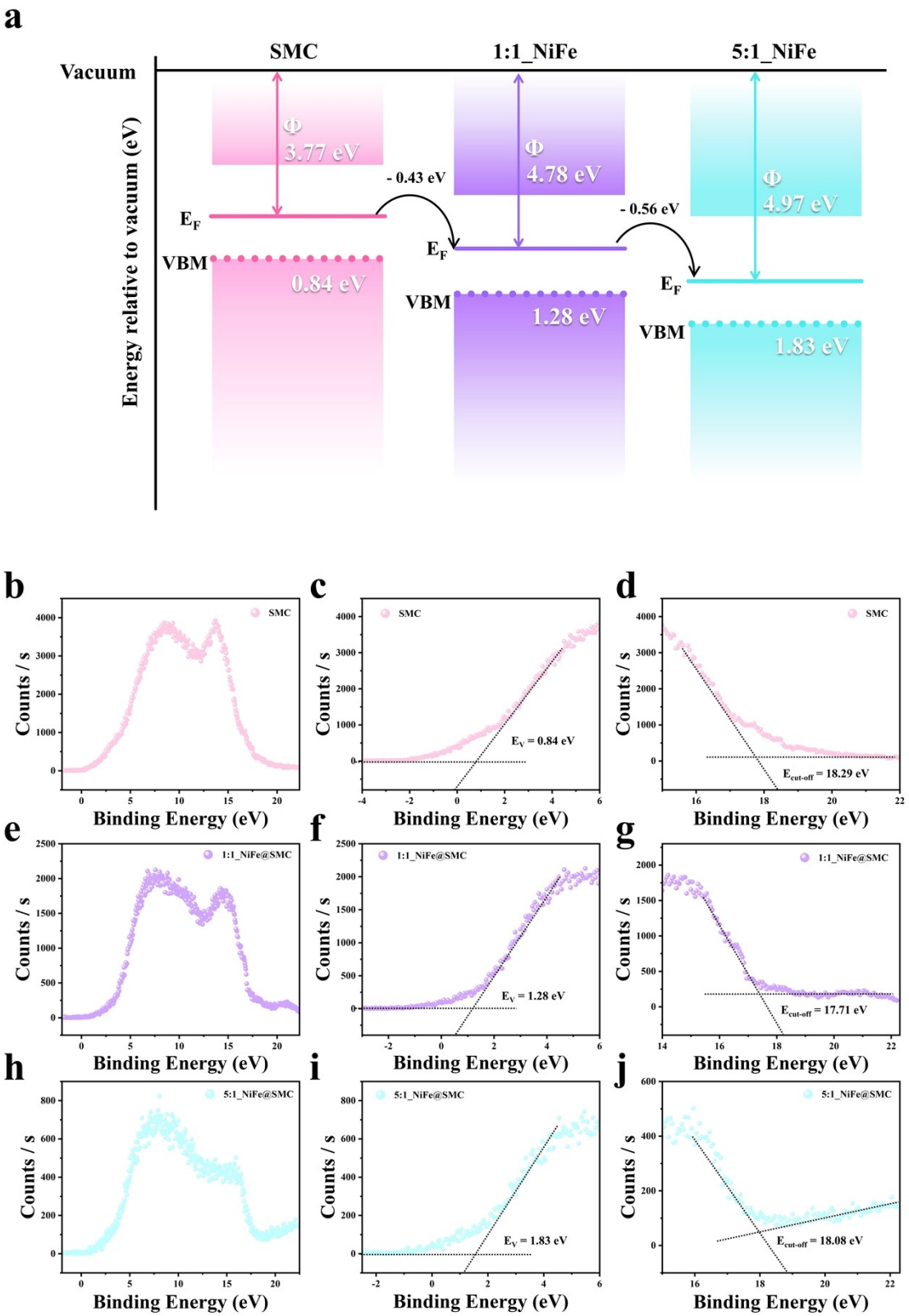
Supplementary Figure 3. XPS spectra of 1:1_NiFe: **a** Ni 2p, **b** Fe 2p, **c** C 1s, and **d** O 1s. XPS spectra of pristine MWCNT: **e** C 1s and **f** O 1s. XPS spectra of SMC: **g** C 1s and **h** O 1s.



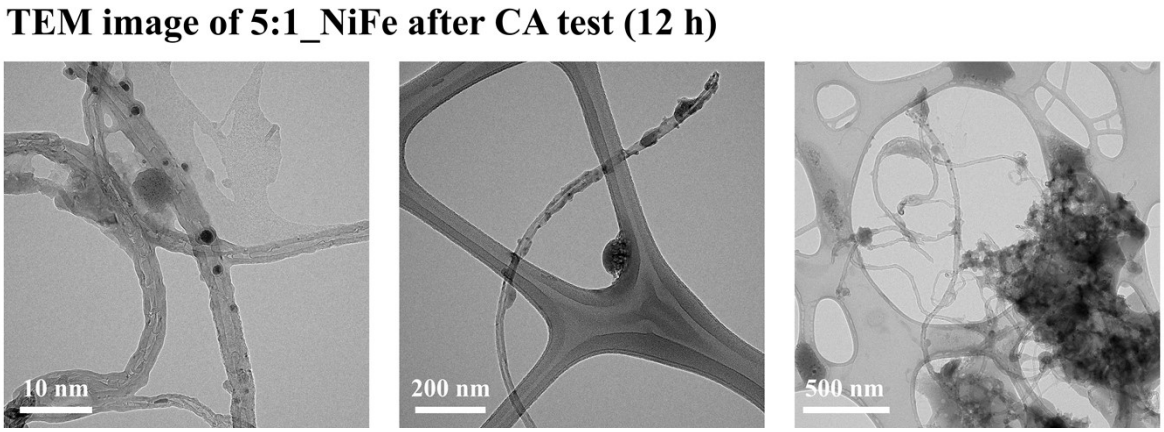
Supplementary Figure 4. Raman spectra of PMWCNT, SMC, 1:1_NiFe, 3:1_NiFe, and 5:1_NiFe.



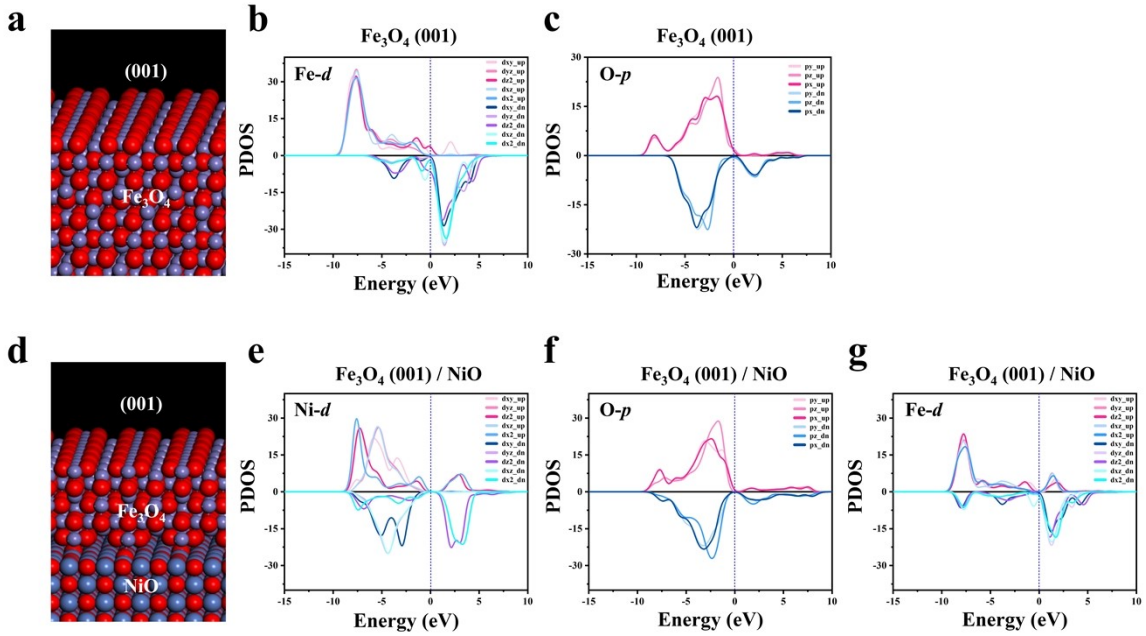
Supplementary Figure 5. Peak area of XPS depth profile: **a** 1:1_NiFe and **b** 5:1_NiFe.



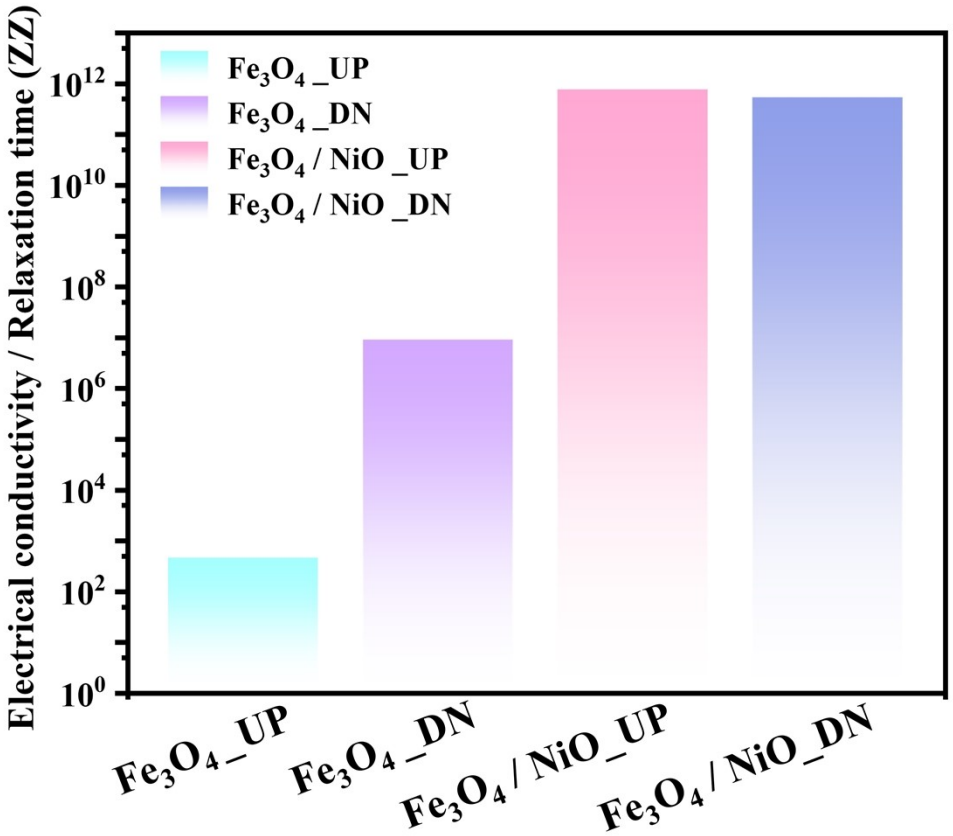
Supplementary Figure 6. a Schematic illustration of the band structures of SMC, 1:1_NiFe, and 5:1_NiFe. UPS spectra of **b** SMC, **e** 1:1_NiFe, and **h** 5:1_NiFe. Enlarged UPS spectra for calculating WFs: **c, d** SMC, **f, g** 1:1_NiFe **i, j** 5:1_NiFe.



Supplementary Figure 7. Low-resolution TEM images of 5:1_NiFe after CA test (12 h).



Supplementary Figure 8. a surface slab model of Fe_3O_4(001) and corresponding PDOS of the **b** Fe-*d*, **c** O-*p* orbital, **d** surface slab model of Fe_3O_4(001)/NiO(001) and corresponding PDOS of the **e** Ni-*d*, **f** O-*p*, **g** Fe-*d*.



Supplementary Figure 9. Calculated electrical conductivity/relaxation time along z axis with up- and down-spin components of Fe-*d* orbital for Fe_3O_4 and Fe_3O_4/NiO.

XPS spectra of 1:1_NiFe				XPS spectra of 5:1_NiFe			
Core level	**Position [eV]**	**FWHM**	**Area**	**Core level**	**Position [eV]**	**FWHM**	**Area**
Ni 2p	856.4869	1.45301	8968.2	**Ni 2p**	856.6089	1.62168	11642.5
	857.6247	2.74222	11027.4		858.0758	1.99213	4888.7
	862.2747	5.65502	21783.3		862.3449	6.53418	25629.2
	873.9263	1.76049	3917.4		874.2601	2.11441	5580.4
	875.3649	2.58965	5463.6		875.8505	2.73322	3606.7
	880.3237	5.97129	15185.5		880.3876	5.3074	12253.9
Fe 2p	711.5805	3.32038	65799.5	**Fe 2p**	711.7844	3.30673	77283.4
	714.5154	4.80872	52482.5		714.7857	4.63599	55052.4
	719.0264	4.64257	17542.1		719.15	4.88029	21970.1
	724.9465	4.04612	40132		725.0329	3.93529	43600.4
	728.0539	4.98635	22199.5		727.7854	4.52997	20888.1
	731.7826	6.51174	8907		730.6401	7.37617	19056.7
O 1s	529.3975	1.13729	13817.8	**O 1s**	529.354	1.22782	21137.1
	530.2526	1.11585	54862.3		530.1972	1.13979	88129.8
	530.8492	1.03566	79947.6		530.6792	0.965672	55327.7
	531.1948	0.819101	24316.3		531.1198	0.972646	39339.6
	531.5157	0.898503	20207.8		531.5754	0.90247	18304.9
	532.1509	0.880186	7014.9		532.1459	0.890923	12089.2
	532.9328	1.3943	6874.2		532.8037	0.93061	5305.2
C 1s	284.3383	1.10223	29979.8	**C 1s**	284.2222	0.93716	29914.2
	284.7647	0.745764	131150.4		284.7769	0.709016	147558
	285.348	0.993179	55216.2		285.211	0.930601	50591
	286.1361	1.08299	18820.8		285.7428	1.34262	40641
	287.003	1.17573	9974.2		286.8214	1.36339	15368
	288.8894	2.00339	15834.3		288.9455	2.20279	17452.5
	291.2894	2.16573	7715.1		291.3123	1.91351	7592.1

Supplementary Table 1. Quantitative XPS information of 1:1_NiFe and 5:1_NiFe.

	Pristine MWCNT	SMC	1:1_NiFe	3:1_NiFe	5:1_NiFe
I_D/I_G	0.77	0.81	0.82	0.87	0.90

Supplementary Table 2. Ratios of the I_D/I_G band intensity of the different samples.

Catalysts	Overpotential [mV vs. RHE]	Tafel slope [mV dec^{-1}]	Rct [Ω]	Reference
5:1_NiFe	286	32	39.33	This work
Co@Co_3O_4/NC-1	420	91.5	-	[S1]
$LaNiO_3$	460	96	63.49	[S2]
Ni_1Fe_2-O	316	47	25.52	[S3]
$Zn_{0.35}Co_{0.65}O$	322	42.6	-	[S4]
$CoNiO_2$-8	269	56.9	-	[S5]
$NiMoO_4$@Co_3O_4	282	58	1.1	[S6]
N, S-CNT	360	56	-	[S7]
Co_3O_4@MWCNT (vs. Ag/AgCl)	474	51	4.2	[S8]
MnO_2/carbon fiber paper	467	111.7	-	[S9]

Supplementary Table 3. The OER activities of recently reported electrocatalysts for overall water splitting.

	SMC	1:1_NiFe	5:1_NiFe
$E_{cut-off}$ [eV]	18.29	17.71	18.08
E_{VBM} [eV]	0.84	1.28	1.83
Work Function [Φ]	3.77	4.78	4.97

Supplementary Table 4. $E_{cut-off}$, E_{VBM}, and WFs of SMC, 1:1_NiFe, and 5:1_NiFe based on UPS data.

OER energy [eV] \ Model	DBT		SCV	
	Fe_3O_4	Fe_3O_4/NiO	Fe_3O_4	Fe_3O_4/NiO
R 1	0.779	0.385	1.427	1.233
R 2	2.636	1.093	3.303	1.717
R 3	0.972	1.949	0.274	0.850
R 4	1.135	2.096	0.519	1.723

Supplementary Table 5. Calculated energy barriers of the four steps of the OER considering DBT and SCV structures of the Fe_3O_4(001) and Fe_3O_4(001)/NiO(001).

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