Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

A Co-doped Ni-Fe mixed oxide mesoporous nanosheet array with low overpotential and high stability towards overall water splitting

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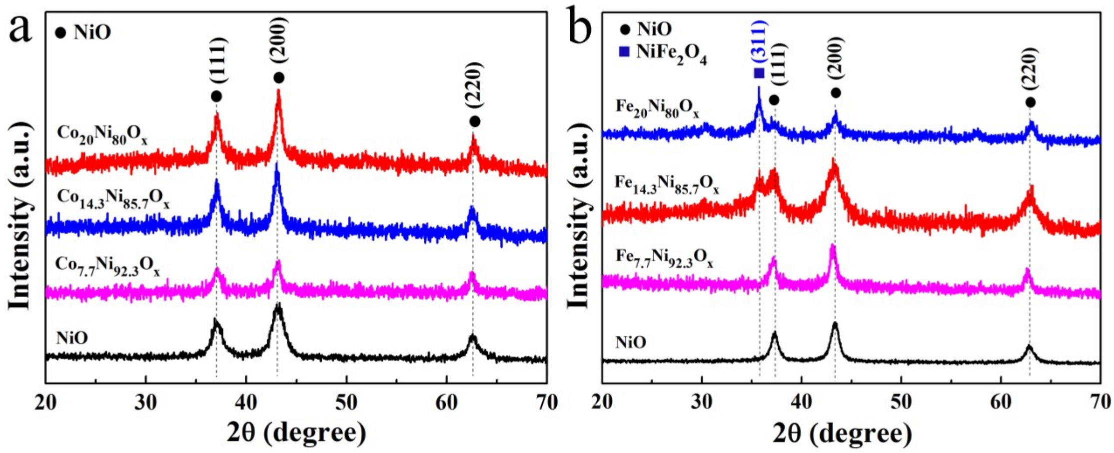

Fig. S1 The XRD patterns of the Co-doped NiO and NiFe mixed oxide samples. The measurements were taken with the powers ultrasonically detached from Ni foam. (a) From bottom to up, pure NiO, the Co-doped NiO samples with addition of 0.25, 0.5 and 0.75 mmol Co source, respectively. (b) From bottom to up, pure NiO, the NiFe mixed oxide samples with addition of 0.25, 0.5 and 0.75 mmol Fe source, respectively.

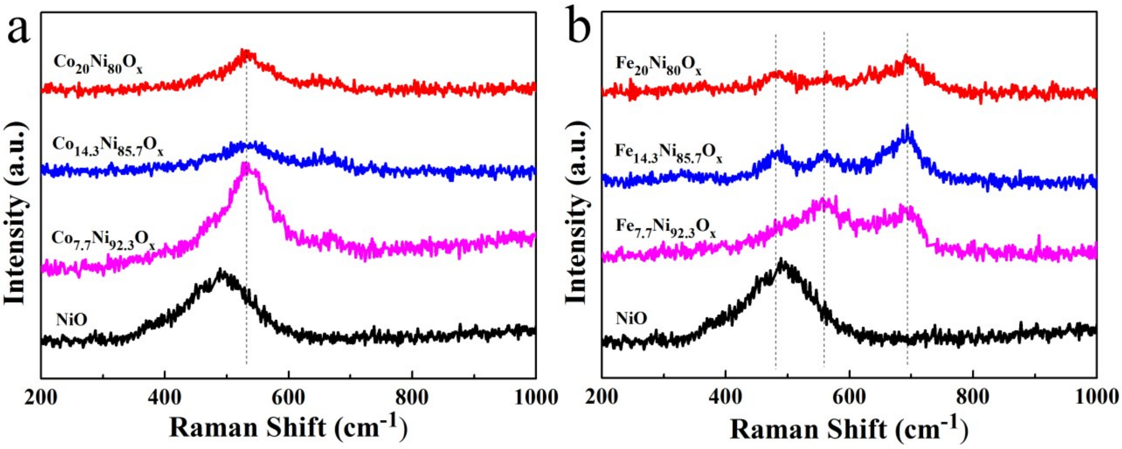

Fig. S2 The Raman spectra of the Co-doped NiO and NiFe mixed oxide samples. (a) From bottom to up, pure NiO, the Co-doped NiO samples with addition of 0.25, 0.5 and 0.75 mmol Co source, respectively. (b) From bottom to up, pure NiO, the NiFe mixed oxide samples with addition of 0.25, 0.5 and 0.75 mmol Fe source, respectively.

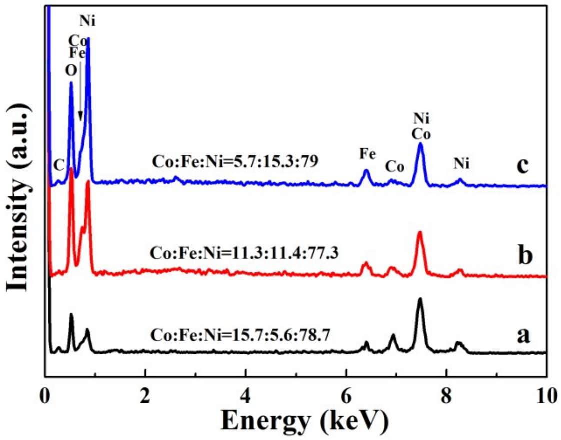

Fig. S3 The EDX results of the Co-doped NiFe mixed oxides with addition of Fe/Co ratio of 1:3 (a), 1:1 (b), and 3:1 (c), respectively.

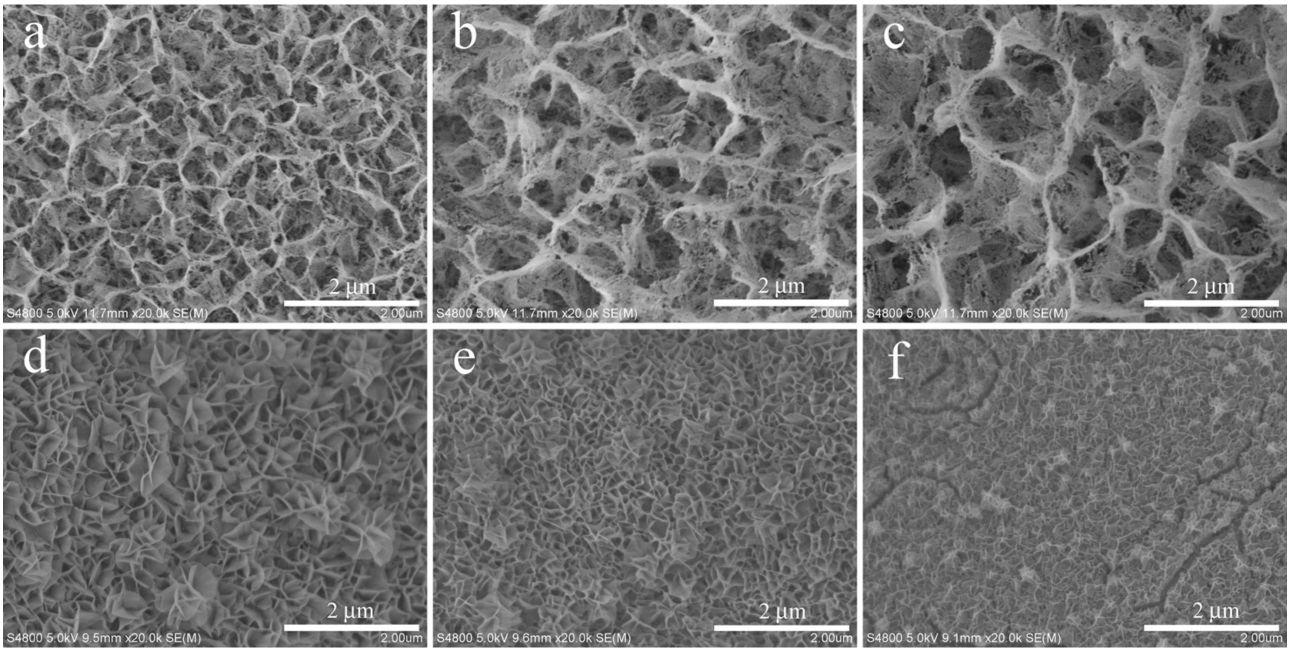

Fig. S4 The SEM images of the Co-doped NiO and NiFe mixed oxide samples. (a-c) The Co-doped NiO samples with addition of 0.25, 0.5 and 0.75 mmol Co source, respectively. (d-f) The NiFe mixed oxide samples with addition of 0.25, 0.5 and 0.75 mmol Fe source, respectively.

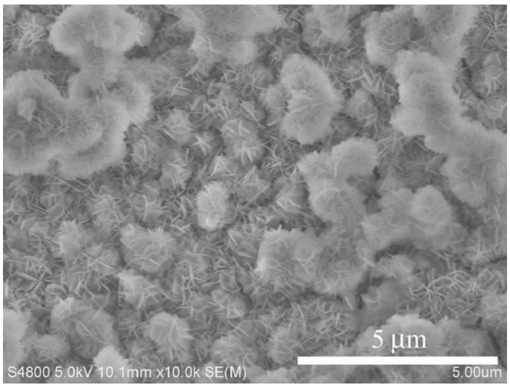

Fig. S5 The SEM image of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ precursor synthesized in 40 mL of deionized water.

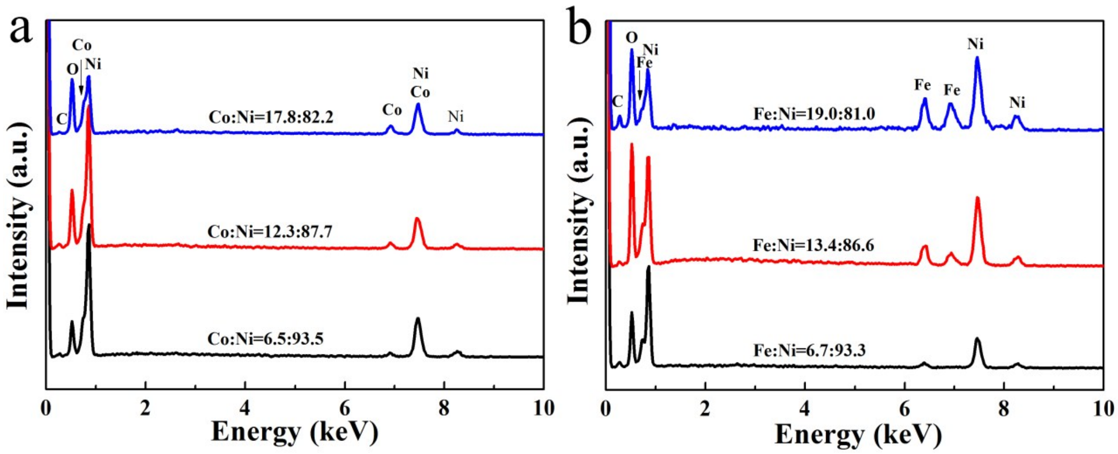

Fig. S6 The EDX results of the Co-doped NiO and NiFe mixed oxide samples. (a) From bottom to up, the Co-doped NiO samples with addition of 0.25, 0.5 and 0.75 mmol Co source, respectively. (b) From bottom to up, the NiFe mixed oxide samples with addition of 0.25, 0.5 and 0.75 mmol Fe source, respectively.

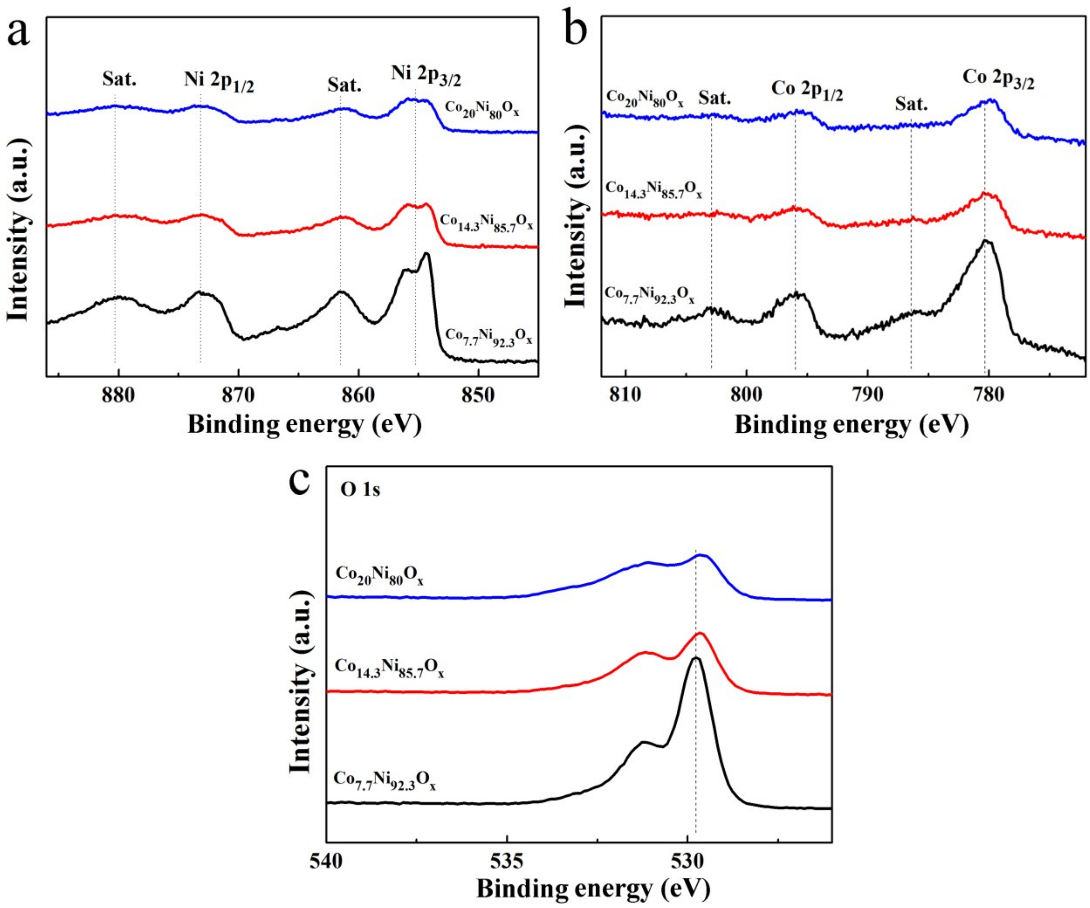

Fig. S7 The XPS spectra of Ni 2p (a), Co 2p (b), and O 1s (c) of Co-doped NiO samples.

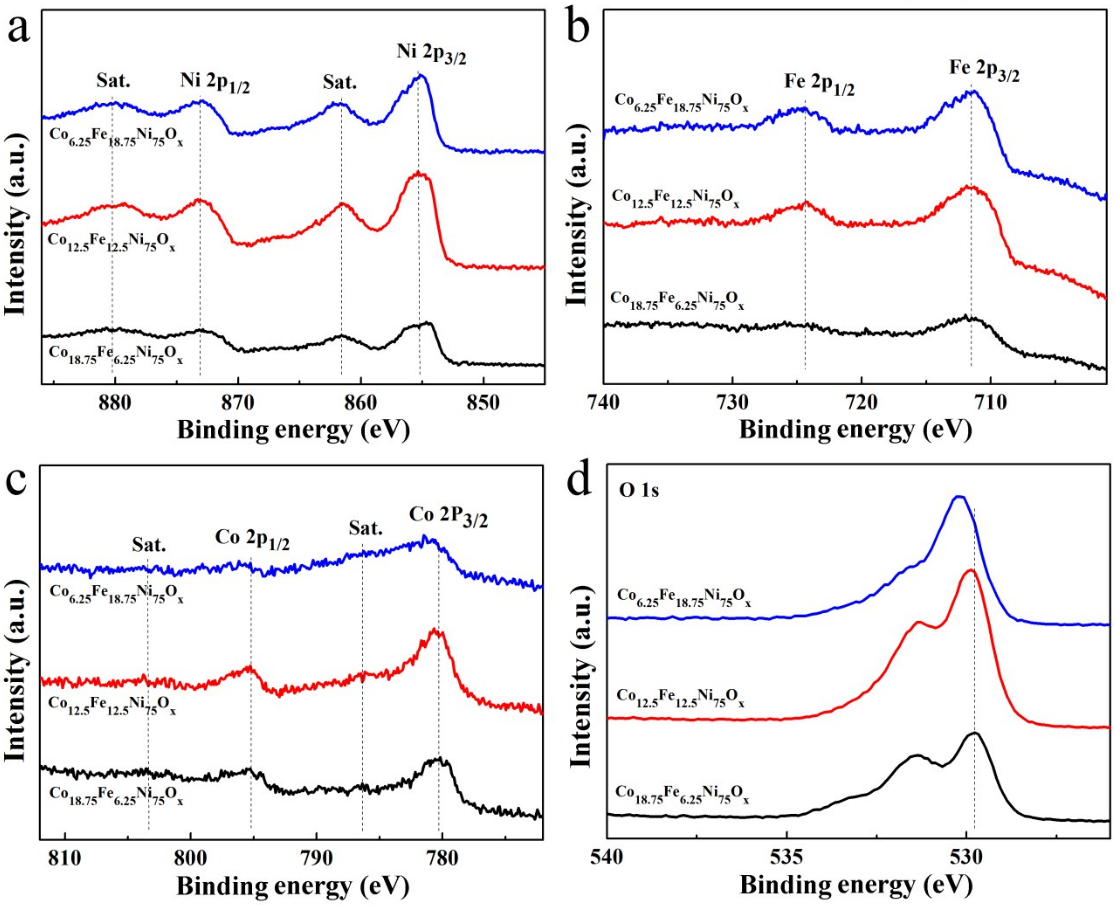

Fig. S8 The XPS spectra of Ni 2p (a), Fe 2p (b), Co 2p (c), and O 1s (d) of Co-doped $NiO/NiFe_2O_4$ samples.

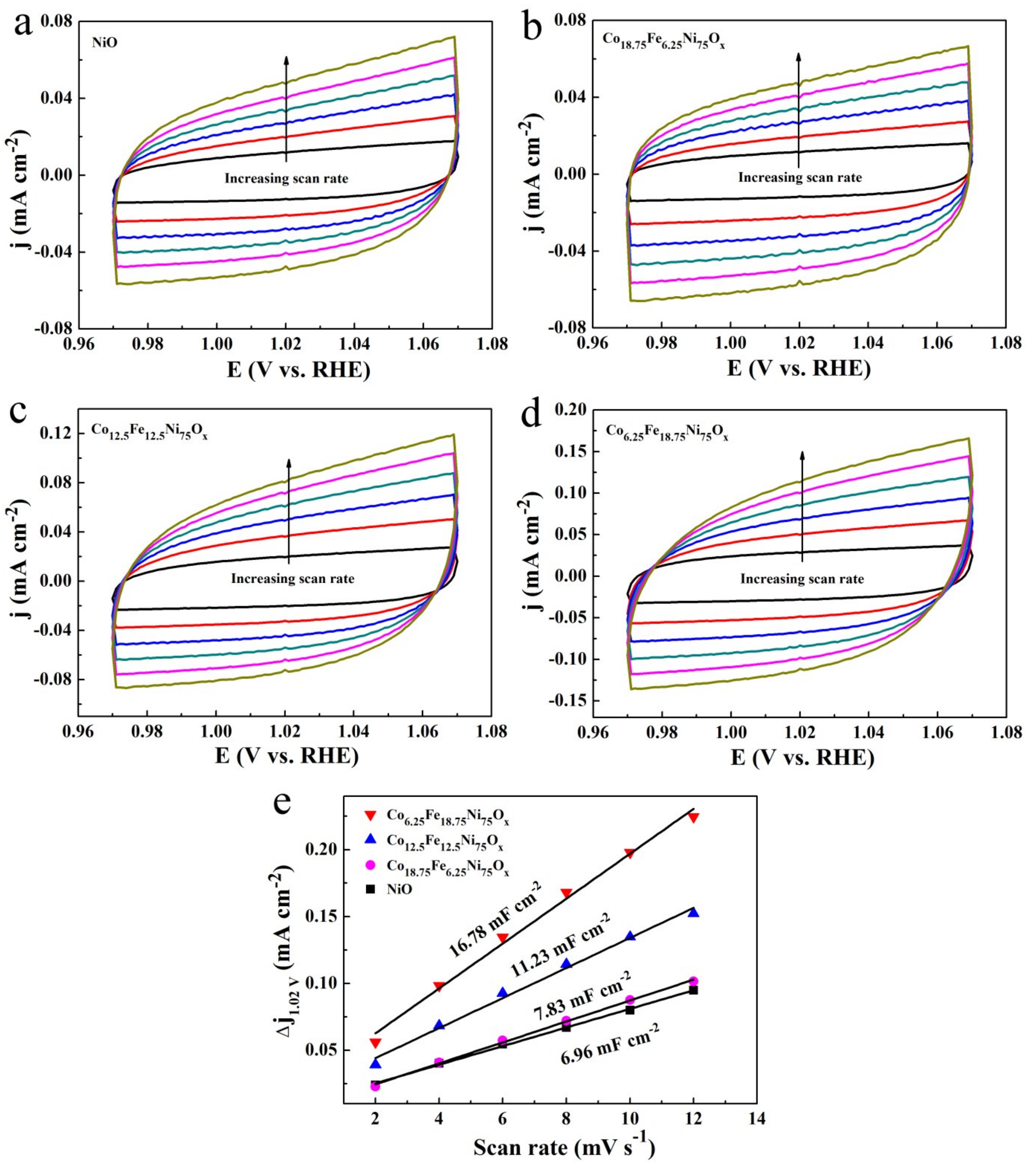

Fig. S9 The cyclic voltammograms of pure NiO (a) and the Co-doped NiFe mixed oxides (b-d) at different scan rates of 2, 4, 6, 8, 10 and 12 mV s^{-1}. (e) The capacitive currents at 1.02 V versus RHE with different scan rates.

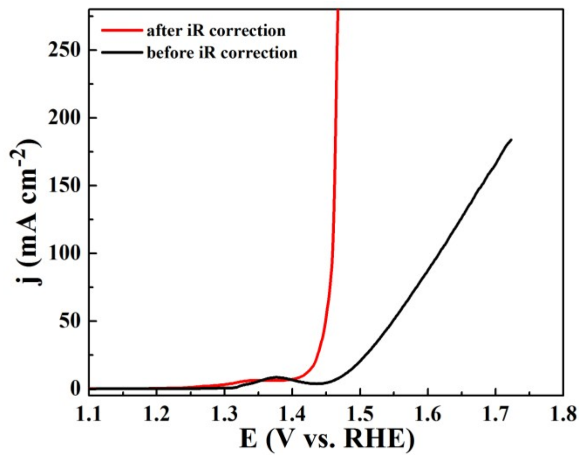

Fig. S10 The OER LSV curves of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ with and without iR correction.

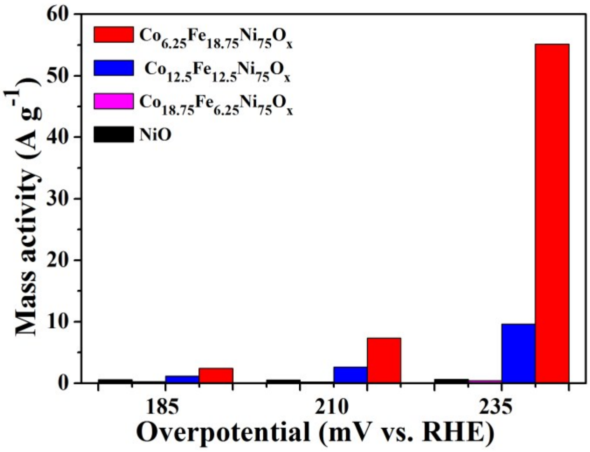

Fig. S11 The OER mass activity of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ at different overpotentials.

Table S1 The comparison of OER activity of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ with some representative Ni-based OER catalysts.

Catalyst	**Electrolyte**	**Loading (mg cm^{-2})**	**η(mV) @j (mA cm^{-2})**	**Tafel Slope (mV dec^{-1})**	**Electrode**	**Reference**
$Fe_{0.1}Ni_{0.9}O$	0.5 M KOH	–	297@10	37	Au/QCM	[1]
Co-Ni-Se/C	1 M KOH	1.5	275@30	63	Ni foam	[2]
$Fe_{0.2}Ni_{0.8}P_2$	1 M KOH	1.1	141@10	49.3	Carbon Paper	[3]
$NiSe_2$ nanoparticles	1 M KOH	2.5	295@20	82	Ti plate	[4]
Ni-Fe oxide nanotube array (Fe: 20%)	0.1 M KOH	–	380@5	105	ITO	[5]
Porous $NiFe_{0.15}O_x$	0.1 M KOH	–	328@10	42	Glassy Carbon	[6]
$NiSe_2$ NCs	1 M KOH	1	250@10	38	Glassy Carbon	[7]
Single-layer NiFe-LDH	1 M KOH	1	~300@10	40	Ni foam	[8]
NiFe-LDH nanoplatelet array	1 M KOH	1	224@10	52.8	Ni foam	[9]
NiCoFe-LDHs	0.1 M KOH	0.3	275@10	85	Ni foam	[10]
β-$Ni(OH)_2$ ultrathin nanomesh	1 M KOH	0.285	236@20	132	Glassy carbon	[11]
NiCo LDH	1 M KOH	–	367@10	40	Carbon Paper	[12]
$NiCo_2O_4$ nanowires	1 M KOH	–	360@10	60	Ti foil	[13]
$NiCo_2O_4/Co_{0.57}Ni_{0.43}$LMOs	0.1 M KOH	–	340@10	63	Carbon fiber papers	[14]
$Co_{6.25}Fe_{18.75}Ni_{75}O_x$	1 M KOH	~4	186@10	38.5	Ni foam	This work

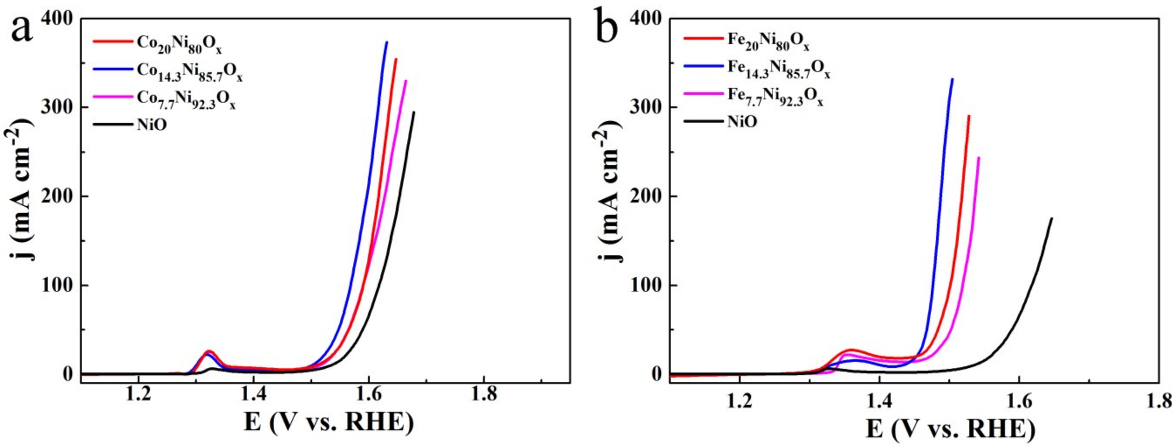

Fig. S12 The polarization curves for the Co-doped NiO (a) and NiFe mixed oxide samples (b) for OER.

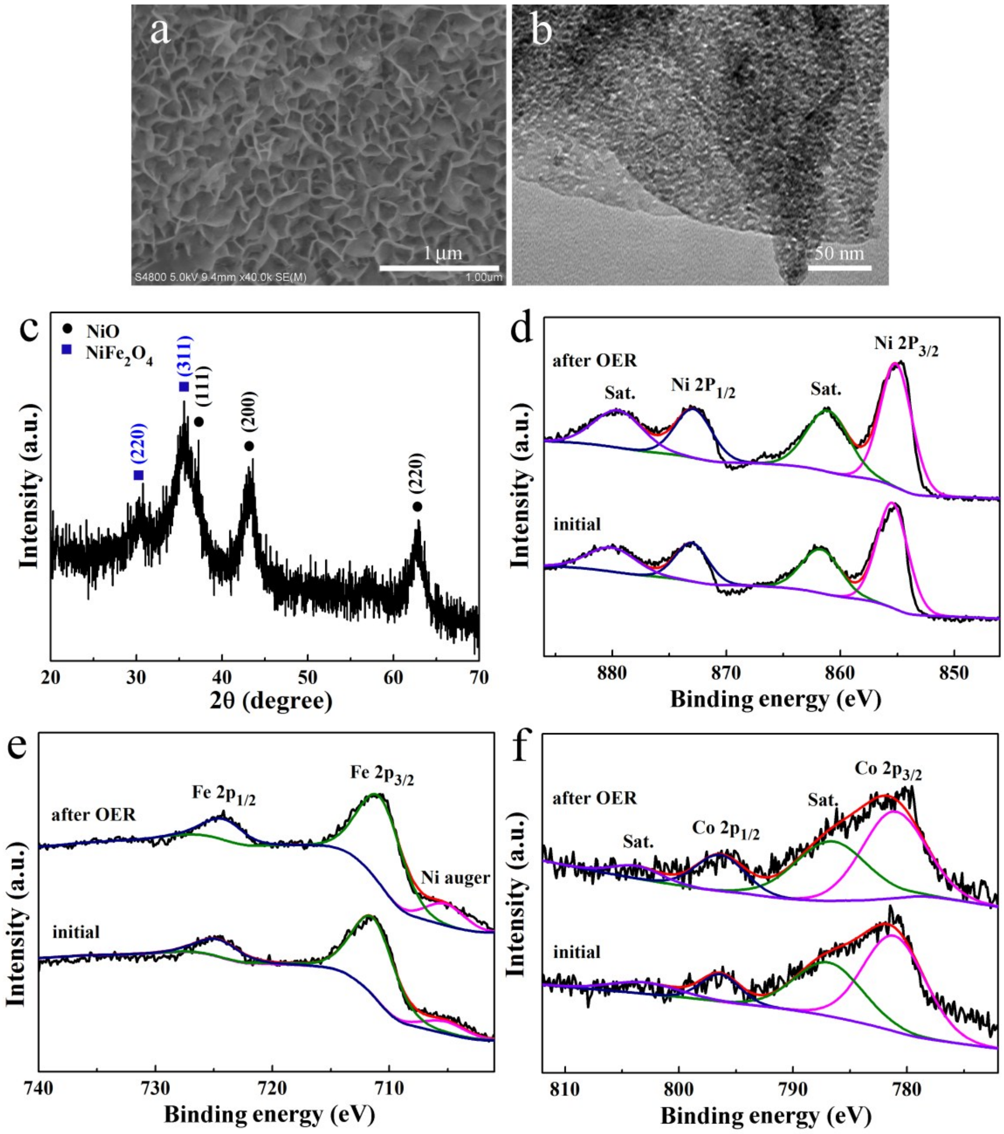

Fig. S13 The SEM image (a), the TEM image (b), the XRD pattern (c), the XPS spectra of Ni 2p (d), Fe 2p (e), Co 2p (f) of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ after 12 h OER chronopotentiometry test.

The FESEM image of the catalyst after 12 h OER chronopotentiometry test clearly revealed that the nanosheets array structure was remained (Fig. S13a), the TEM image of the nanosheet indicated the mesopores can still be identified (Fig. S13b), and the corresponding XRD pattern of the catalyst was barely changed after measurement (Fig. S13c). The XPS characterization revealed the position and shape of Ni 2p peaks after 12 h OER durable test were barely changed (Fig. S13d), indicating that electronic state of Ni does not change after OER durable test.[15] In addition, the post-Fe 2p (Fig. S13e) and post-Co 2p (Fig. S13f) peaks remained no changes. The results suggest the excellent structure stability of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ during the long-term OER measurement.

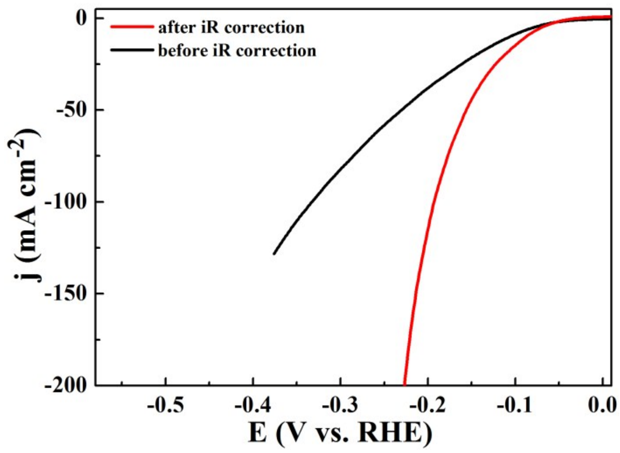

Fig. S14 The HER LSV curves of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ with and without iR correction.

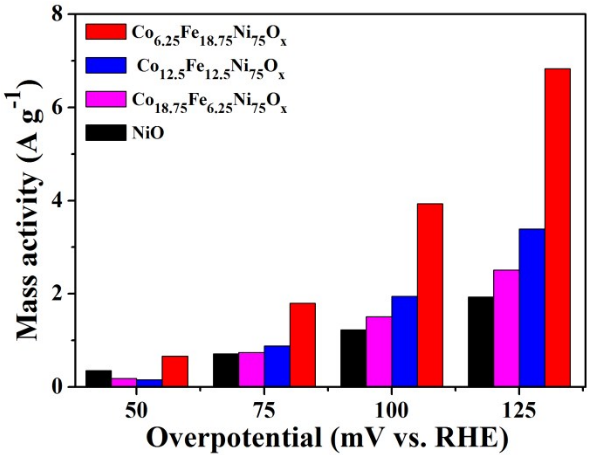

Fig. S15 The HER mass activity of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ at different overpotentials.

Table S2 The comparison of HER activity of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ with some representative Ni-based HER catalysts.

Catalyst	Electrolyte	Loading (mg cm^{-2})	η (mV) @j(mA cm^{-2})	Tafel Slope (mV dec^{-1})	Electrode	Reference
Co-Ni-Se /C	1 M KOH	1.5	90@10	81	Ni foam	[2]
$Fe_{0.2}Ni_{0.8}P_2$	1 M KOH	1.1	250@10	72.2	Carbon Paper	[3]
$NiSe_2$ nanoparticles	1 M KOH	2.5	96@10	82	Ti plate	[4]
NiCoFe-LDHs	0.1 M KOH	0.3	200@40	78	Ni foam	[10]
Porous $NiSe_2$ nanosheets	1 M KOH	~0.46	184@10	77	Carbon paper	[16]
NiSe Nanowire	1 M KOH	2.8	270@20	64	Ni foam	[17]
$Co_{0.13}Ni_{0.87}Se_2$	1 M KOH	1.67	64@10	63	Ti plate	[18]
Ni/NiO nanosheets	1 M KOH	0.3	145@10	43	Ni foam	[19]
NiFe (oxy)hydroxide/$NiCo_2O_4$	1 M KOH	–	105@10	88	Ni foam	[20]
$Ni(OH)_2$ Nanosheets	1 M KOH	2.9	127@10	140	Ni foam	[21]
$NiCo_2S_4$ nanoflakes	1 M KOH	–	65@10	84.5	Ni foam	[22]
Ni-Fe-P nanosheet array	1 M KOH	0.25	142@10	84.24	Ni foam	[23]
$Co_{6.25}Fe_{18.75}Ni_{75}O_x$	1 M KOH	~4	84@10	53.6	Ni foam	This work

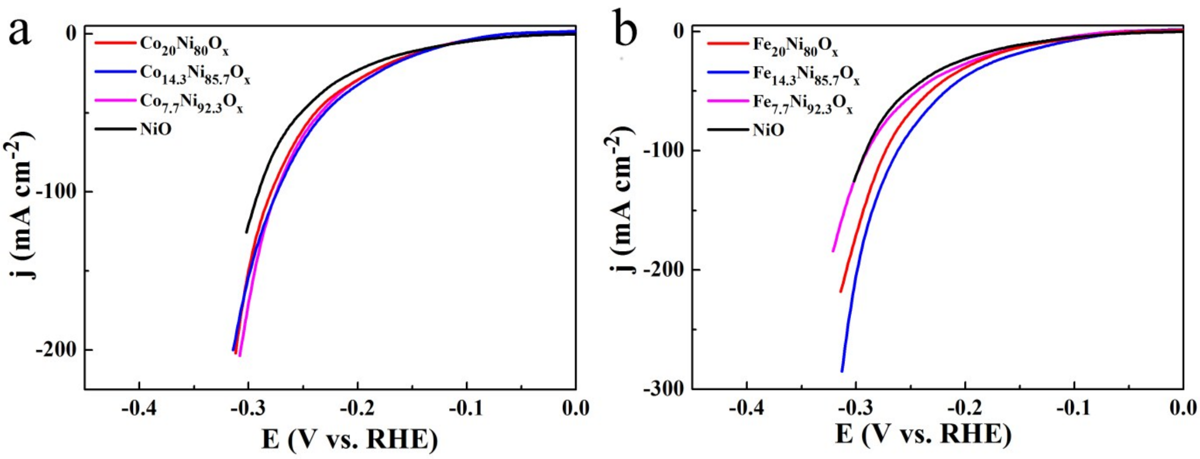

Fig. S16 The polarization curves for the Co-doped NiO (a) and NiFe mixed oxide samples (b) for HER.

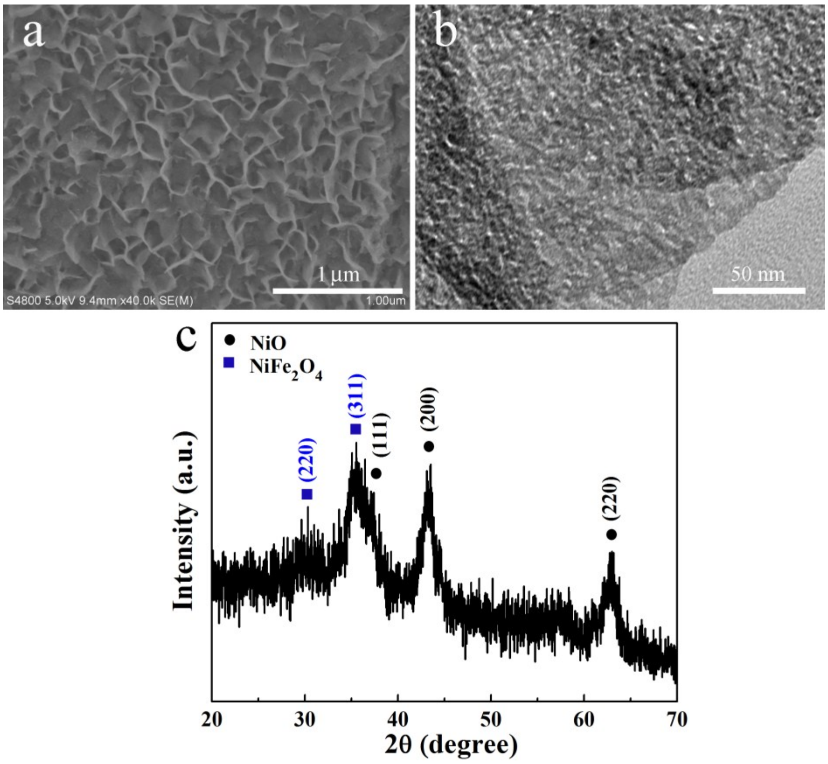

Fig. S17 The SEM image (a), the TEM image (b), and the XRD pattern (c) of $Co_{6.25}Fe_{18.75}Ni_{75}O_x$ electrode after 12 h HER chronopotentiometry test.

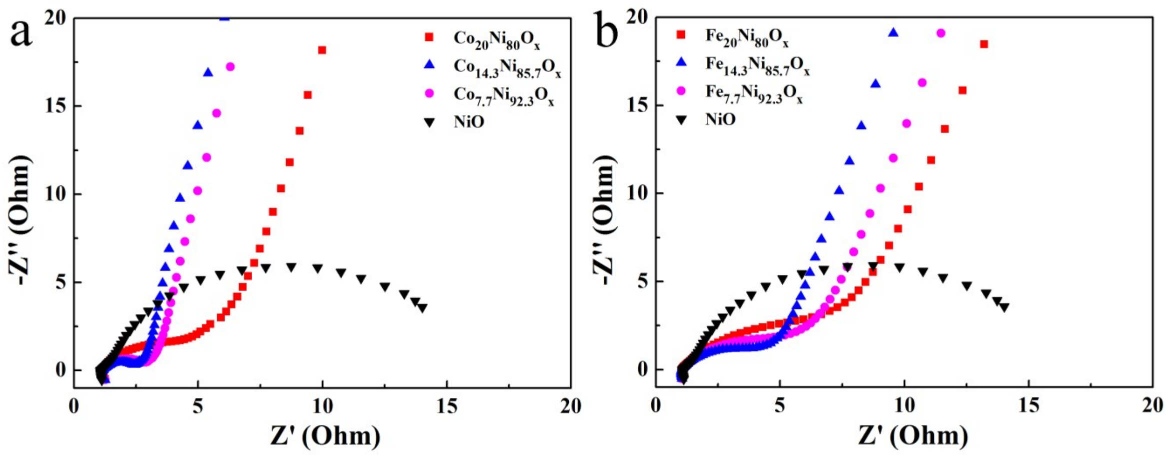

Fig. S18 The Nyquist plots of the Co-doped NiO (a) and NiFe mixed oxide electrodes (b). As a comparison, the data of NiO was also presented.

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