Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supplementary Information

An advanced and highly efficient Ce assisted NiFe-LDH electrocatalysts for overall water splitting

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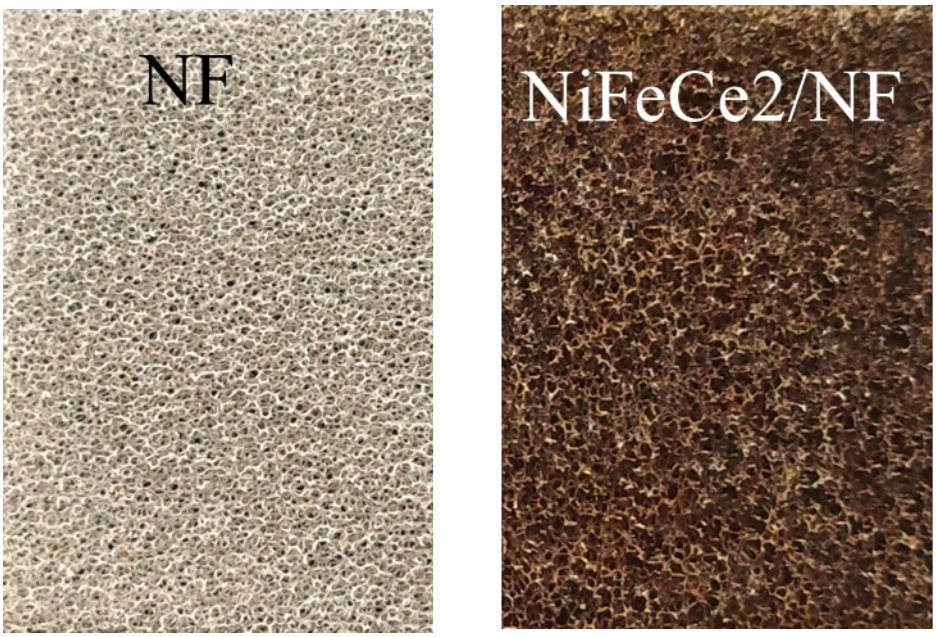

Figure S1. Photographs of (a) bare cleaned Nickel foam (NF) and (b) electrocatalysts deposited NF.

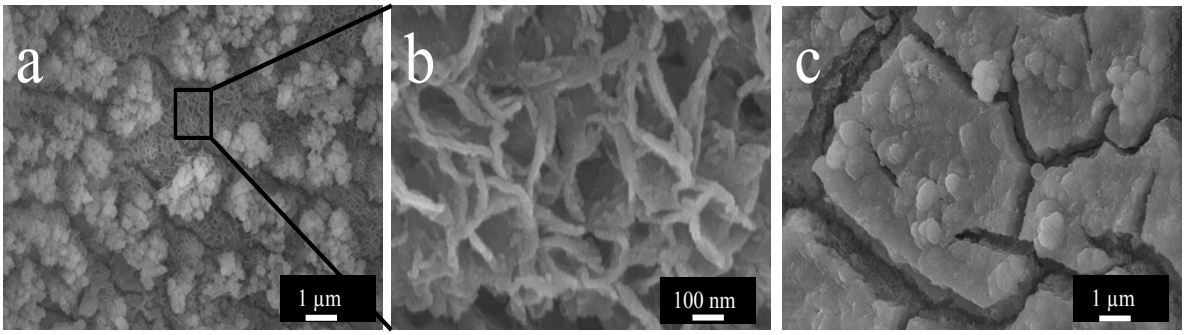

Figure S2. The FE-SEM images of (a) NiFe after 5 minute (b) its high magnified image (c) NiFe after 10 minute deposition time.

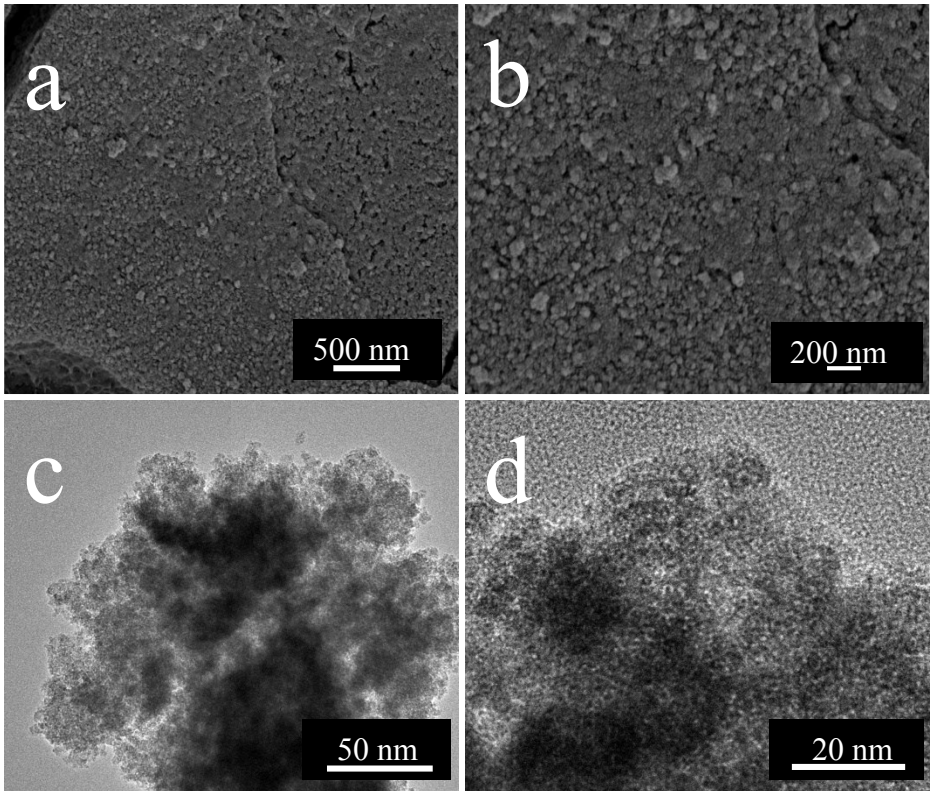

Figure S3 (a-b) FE-SEM images (c-d) TEM images of NiFe electrocatalysts at different magnification.

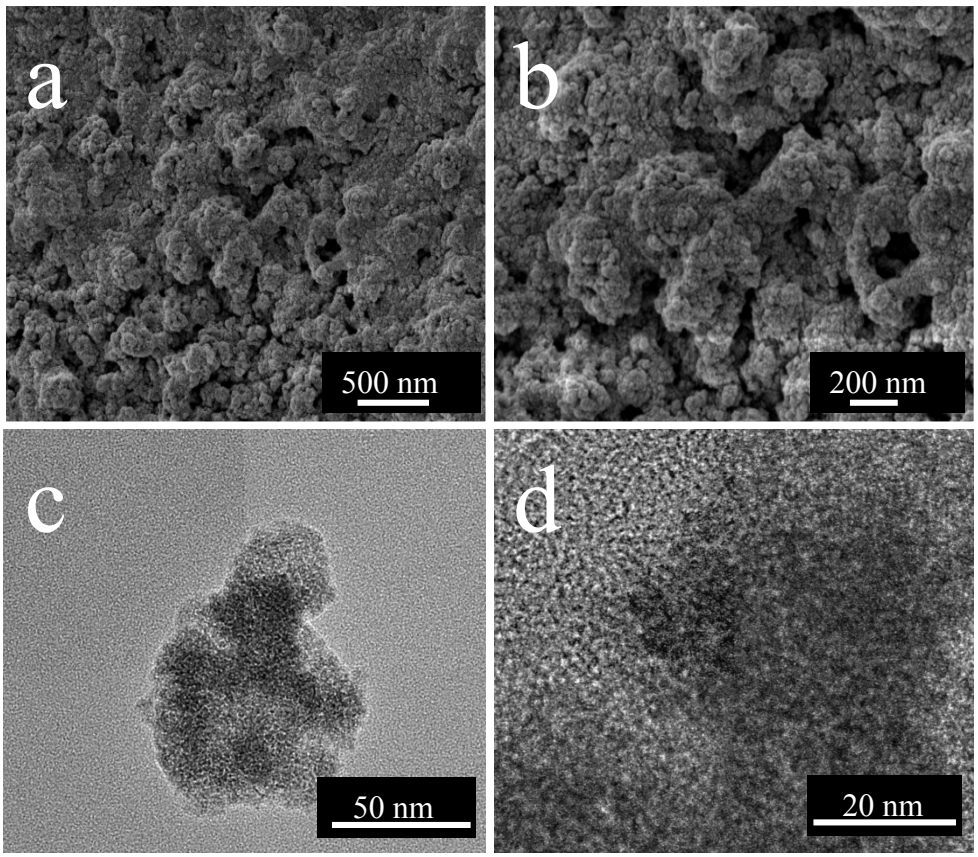

Figure S4: (a-b) FE-SEM images (c-d) TEM images of $NiFe_{0.9}Ce_{0.1}$ electrocatalysts at different magnification.

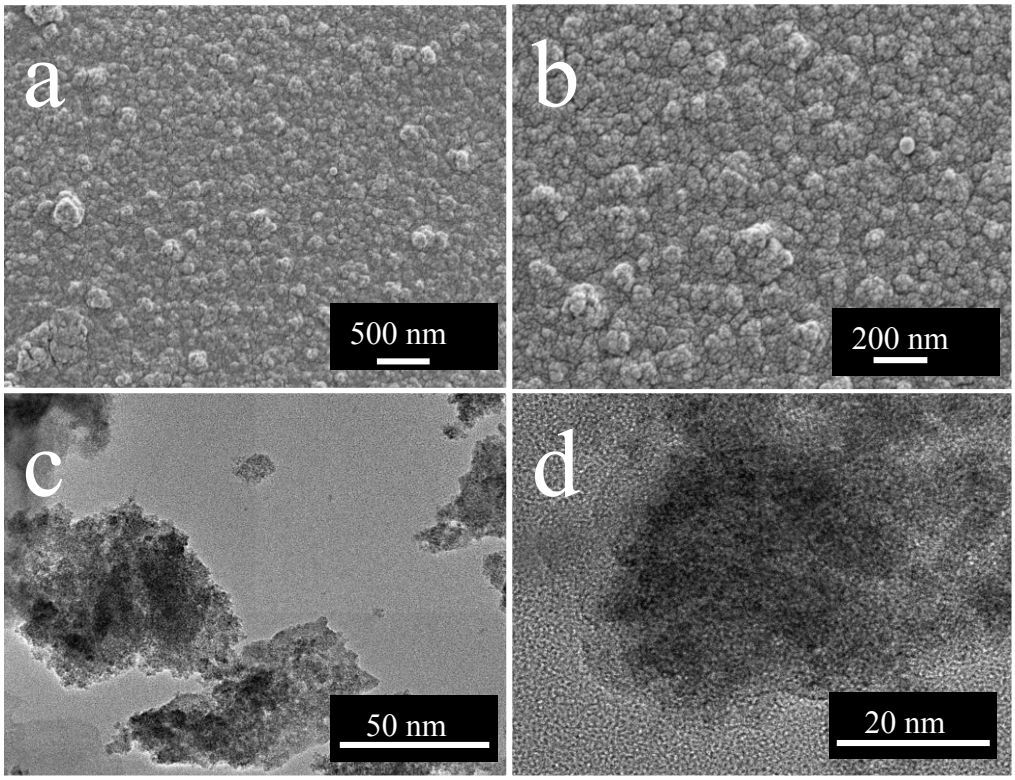

Figure S5: (a-b) FE-SEM images (c-d) TEM images of $NiFe_{0.7}Ce_{0.3}$ electrocatalysts at different magnification.

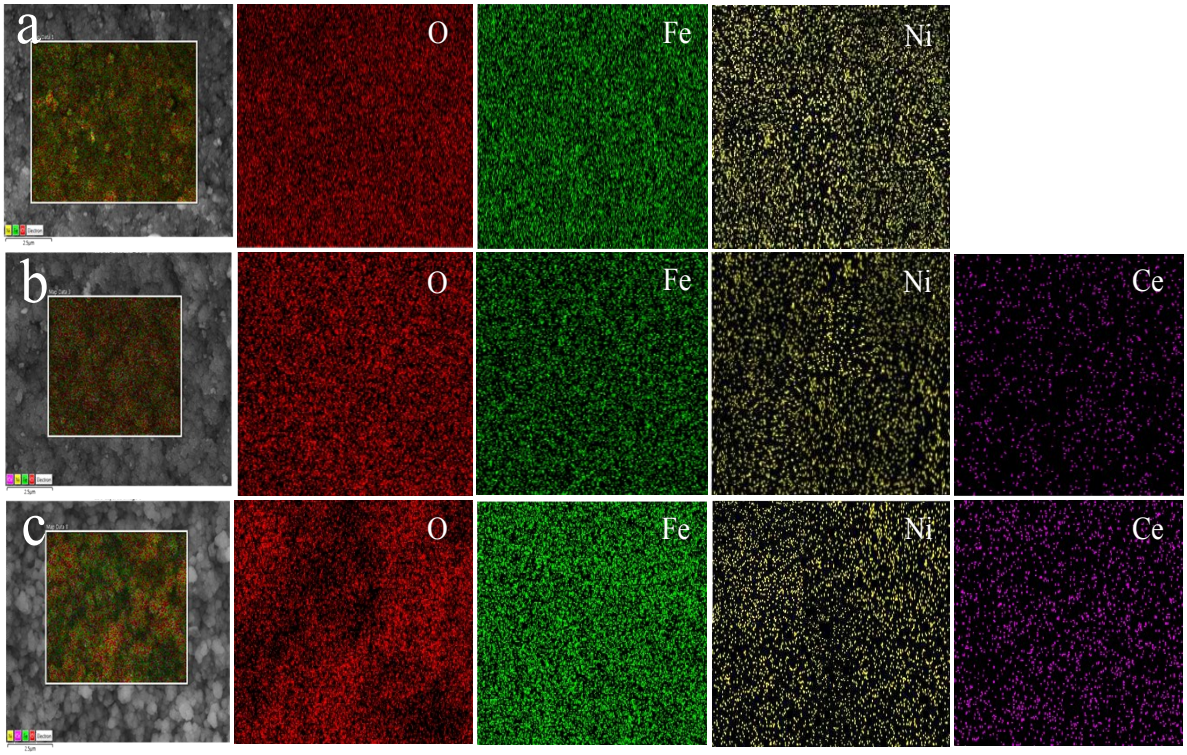

Figure S6: FE-SEM EDS elemental colour mapping for (a) NiFe (b) $NiFe_{0.8}Ce_{0.2}$ (c) $NiFe_{0.7}Ce_{0.3}$.

Table S1: The At% of Ni, Fe, O and Ce elements in electrocatalysts investigated in present study.

Electrocatalysts	**Ni(At%)**	**Fe(At%)**	**O(At%)**	**Ce(At%)**
NiFe	19.10	22.60	58.30	-
$NiFe_{0.9}Ce_{0.1}$	18.80	21.72	58.70	0.78
$NiFe_{0.8}Ce_{0.2}$	18.60	20.95	59.00	1.45
$NiFe_{0.7}Ce_{0.3}$	18.40	19.90	59.30	2.40

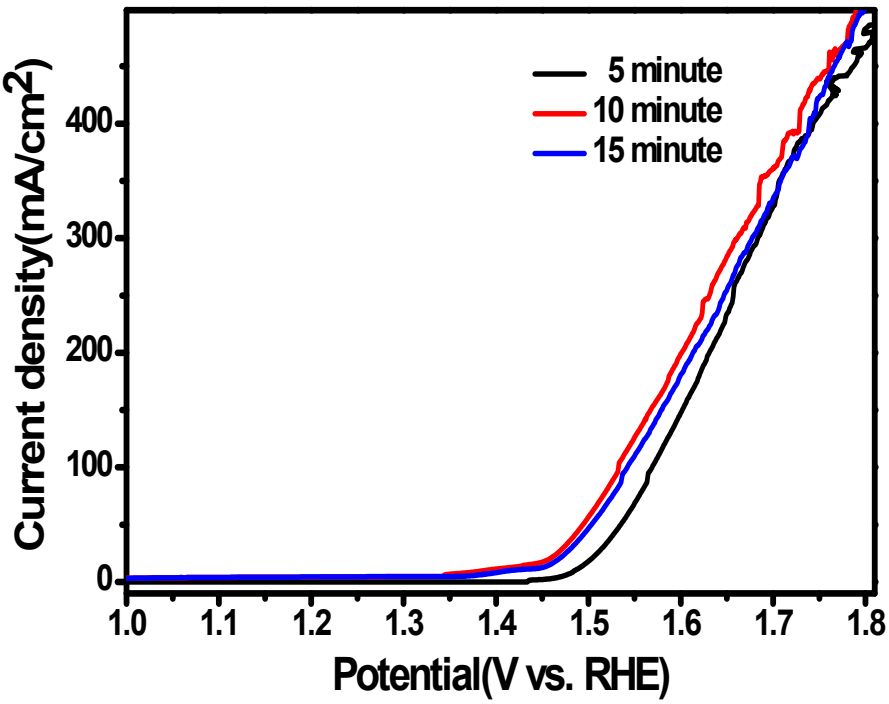

Figure S7: Polarization curves of NiFe electrocatalysts electrodeposited for different time.

Table S2: The OER overpotential required to achieve different current densities for various electrocatalysts tested in present study.

Electrocatalysts	**OER activity (@10 mA/cm^2)**	**OER activity (@50 mA/cm^2)**	**OER activity (@100 mA/cm^2)**
NiFe	197	260	304
$NiFe_{0.9}Ce_{0.1}$	185	254	284
$NiFe_{0.8}Ce_{0.2}$	175	245	273
$NiFe_{0.7}Ce_{0.3}$	205	279	314
RuO_2/C	230	320	390

Table S3: The literature survey of various reported electrocatalysts for OER catalytic activity (@10 mA/cm^2).

Electrocatalysts	**Support***	**OER activity (@10 mA/cm^2) mV**	**Stability (Hour)**	**Reference**
CoSe/NiFe	GF	250	10	10
Exfoliated NiFe-LDH	GC	290	12	31
NiFe-LDH nanosheets	NF	200	10	39
NiFe-LDH array	NF	224	50	44
NiFe-LDH/graphene	NF	205	1.5	50
Ni2/3Fe1/3-rGO	GC	210	10	51
Cu@NiFe-LDH	GF	199	48	52
Exfoliated NiFe-LDH/Defective graphene	GC	210	10	53
NiFe-OH/NiFeP	NF	199	24	54
NiFe-LDH	**NF**	**197**	**50**	**Present work**
$NiFe_{0.8}Ce_{0.2}$	**NF**	**175**	**50**	**Present work**

*Note: Nickel foam=NF

Glassy carbon disc electrode=GC

Exfoliated graphene foil=GF

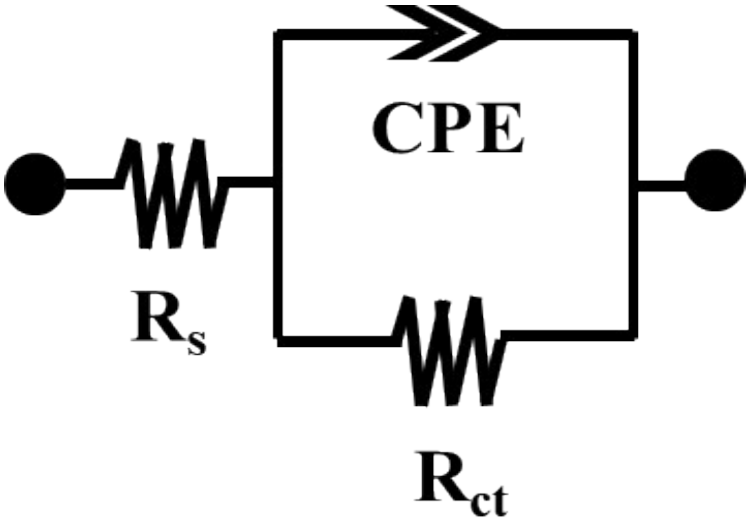

Figure S8: The equivalent circuit fitted for Nyquist plots.

Table S4: The summary of series resistance (R_s), and charge transfer resistance (R_{ct}) obtained after fitted the Nyquist plots.

Electrocatalysts	**R_s (Ω)**	**R_{ct} (Ω)**
NiFe	1.15	3.25
$NiFe_{0.9}Ce_{0.1}$	1.10	2.25
$NiFe_{0.8}Ce_{0.2}$	1.05	2.05
$NiFe_{0.7}Ce_{0.3}$	1.15	4.00
NF	1.40	150

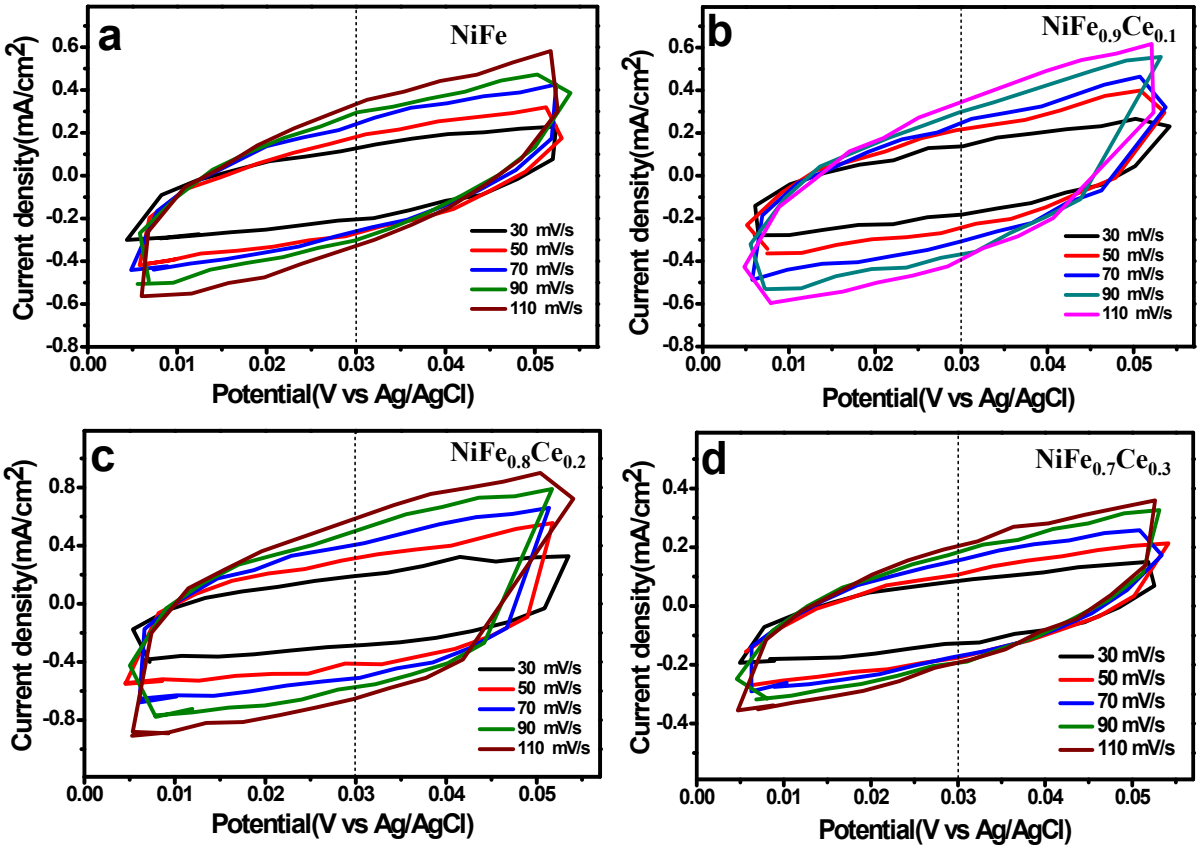

Figure S9: CV curves for different electrocatalysts carried out in non-faradic region at different scan rate in 1M KOH.

Table S5: The summarized double layer capacitance (DLC), electrochemical surface area (ECSA), roughness factor (RF) and active sites for various NiFe-based electrocatalysts tested in present study.

Electrocatalysts	**DLC (mF/cm^2)**	**ECSA (cm^2)**	**RF (cm^2)**	**Active sites Moles(10^{-5})**
NiFe	2.35	58.75	235	7.66
$NiFe_{0.9}Ce_{0.1}$	2.60	65.00	260	10.13
$NiFe_{0.8}Ce_{0.2}$	4.00	100	400	14.04
$NiFe_{0.7}Ce_{0.3}$	1.60	40	160	4.43

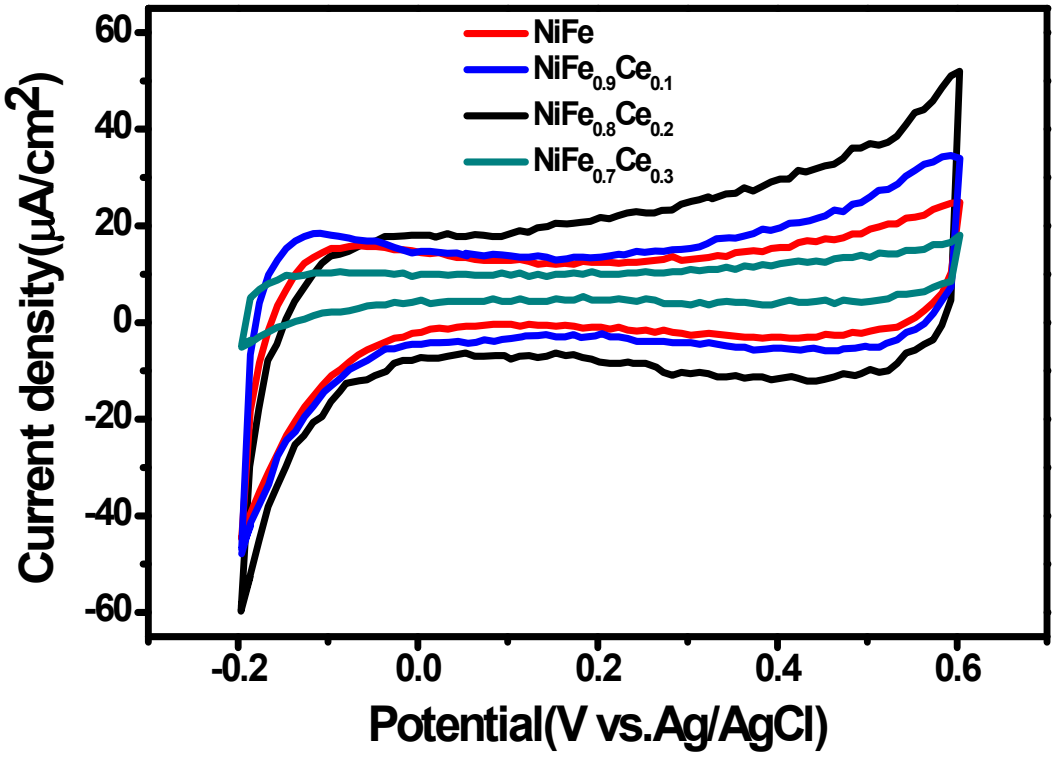

Figure S10: The CVs for various electrocatalysts were carried out in phosphate buffer solution (pH=7) at the scan rate of 50 mV.

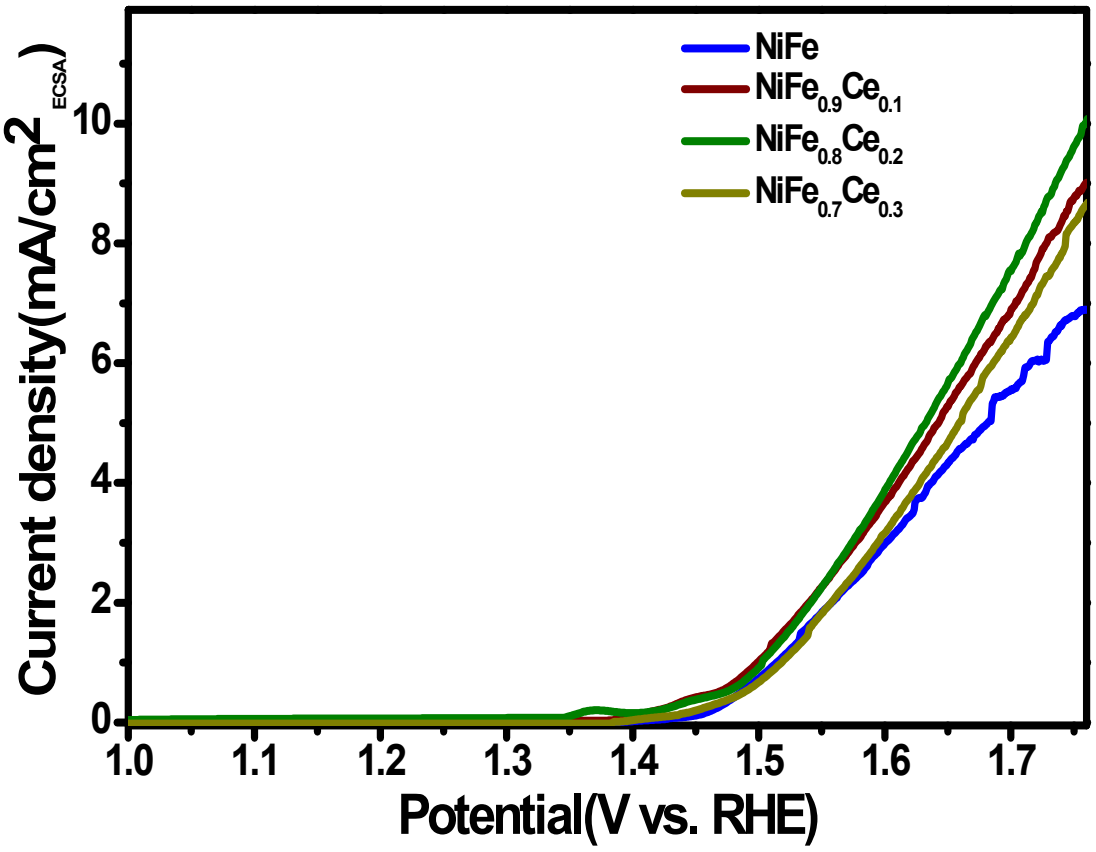

Figure S11: OER polarization curves with current density normalized by the calculated ECSA.

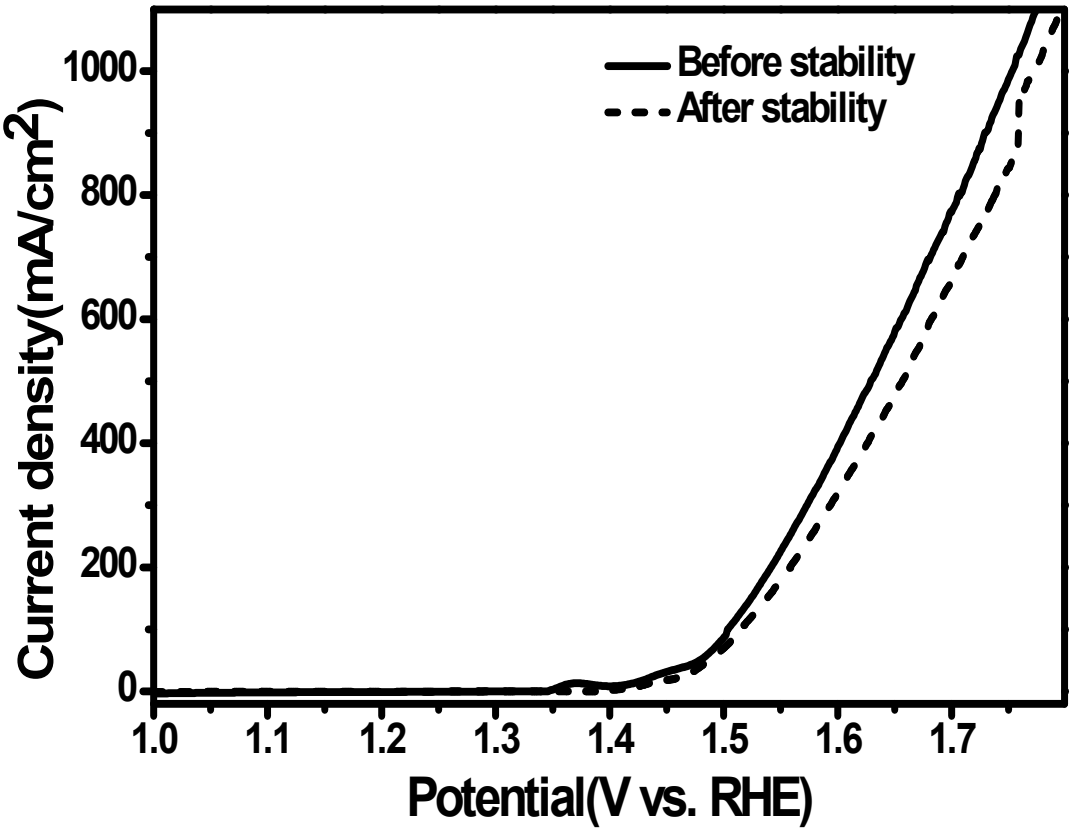

Figure S12: Polarization curves of $NiFe_{0.8}Ce_{0.2}$ electrocatalysts before and after 50 hour OER test.

Table S6: The HER overpotential required to achieve different current densities for various electrocatalysts tested in present study.

Electrocatalysts	**HER activity (@10 mA/cm^2)**	**HER activity (@50 mA/cm^2)**	**HER activity (@100 mA/cm^2)**
NiFe	175	298	369
$NiFe_{0.9}Ce_{0.1}$	167	257	321
$NiFe_{0.8}Ce_{0.2}$	147	254	313
$NiFe_{0.7}Ce_{0.3}$	195	296	374
Pt/C	55	155	209

Table S7: The literature survey of various reported NiFe-based electrocatalysts for HER catalytic activity (@10 mA/cm^2).

Electrocatalysts	**Support***	**HER activity (@10 mA/cm^2) mV**	**Stability (Hour)**	**Reference**
EG/$Co_{0.85}Se$/NiFe-LDH	GF	265	10	10
CoSe/NiFe-LDH	GF	260	-	10
NiFe-LDH NS/DG10	GC	210	5.5	53
NiFe/$NiCo_2O_4$	NF	105	10	56
Ni_3S_2/NF	NF	223	150	58
Cu@CoFe-LDH	CF	171	30	59
$Ni_{0.75}Fe_{0.125}V_{0.125}$	NF	125	15	60
$NiCo_2O_4$/NiFe-LDH	NF	192	10	61
NiFe-LDH	**NF**	**175**	**50**	**Present work**
$NiFe_{0.8}Ce_{0.2}$	**NF**	**147**	**50**	**Present work**

*Note: Nickel foam=NF

Copper foam=CF

Glassy carbon disc electrode=GC

Exfoliated graphene=GF

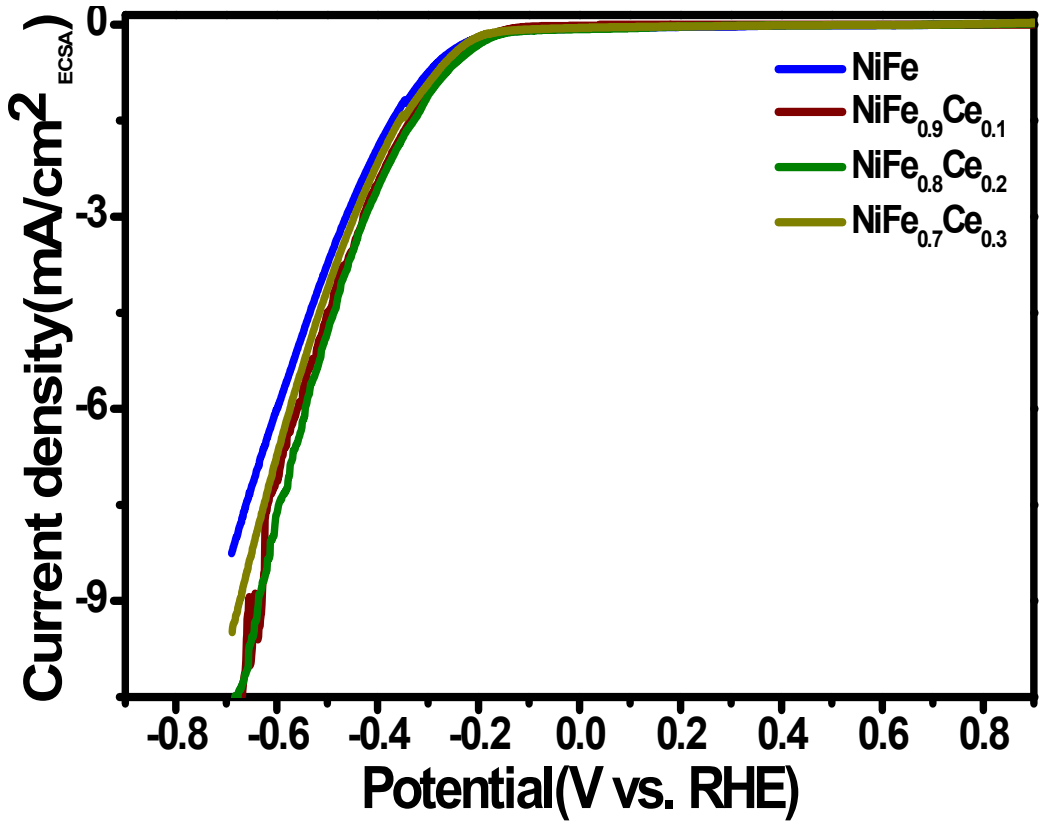

Figure S13: HER polarization curves with current density normalized by the calculated ECSA.

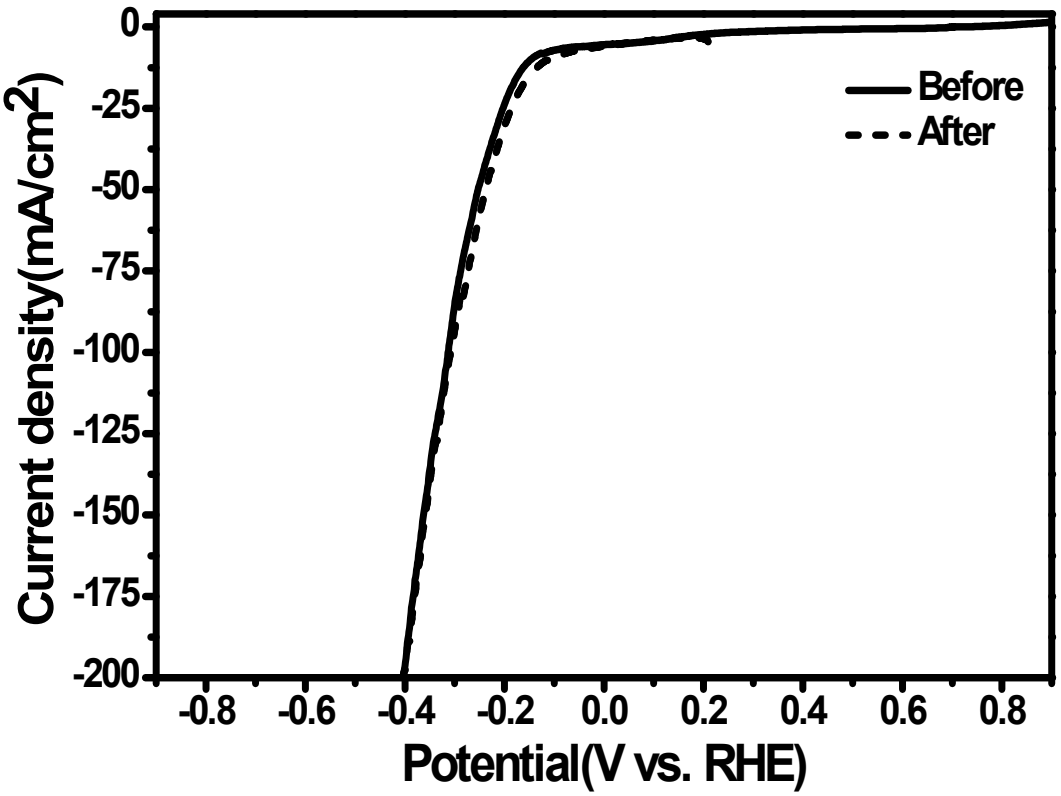

Figure S14: Polarization curves of $NiFe_{0.8}Ce_{0.2}$ electrocatalysts before and after 50 hour HER test.

Table S8: The literature survey of various reported electrocatalysts for overall electrochemical water splitting (Overpotential reported @10 mA/cm^2)

Electrocatalysts	**Support***	**Overpotential (@10 mA/cm^2) mV**	**Stability (Hour)**	**Reference**
EG/$Co_{0.85}Se$/NiFe-LDH	NF	1.65	10	10
Cu@NiFe-LDH	CF	1.54	48	52
NiFe/$NiCo_2O_4$	NF	1.60	12	61
$Ni_{0.75}Fe_{0.125}V_{0.125}$	NF	1.59	15	62
NiSe nanowires	NF	1.63	20	63
NiFe LDH	NF	1.70	3	64
NiMo HRNs	NF	1.64	10	65
Ni_3Se_2	CF	1.65	12	66
Fe doped CoP	TF	1.60	40	67
NiCoP	NF	1.58	24	68
$NiFe_{0.8}Ce_{0.2}$	**NF**	**1.59**	**50**	**Present work**

*Note: Nickel foam=NF

Copper foam=CF

Ti foil=TF

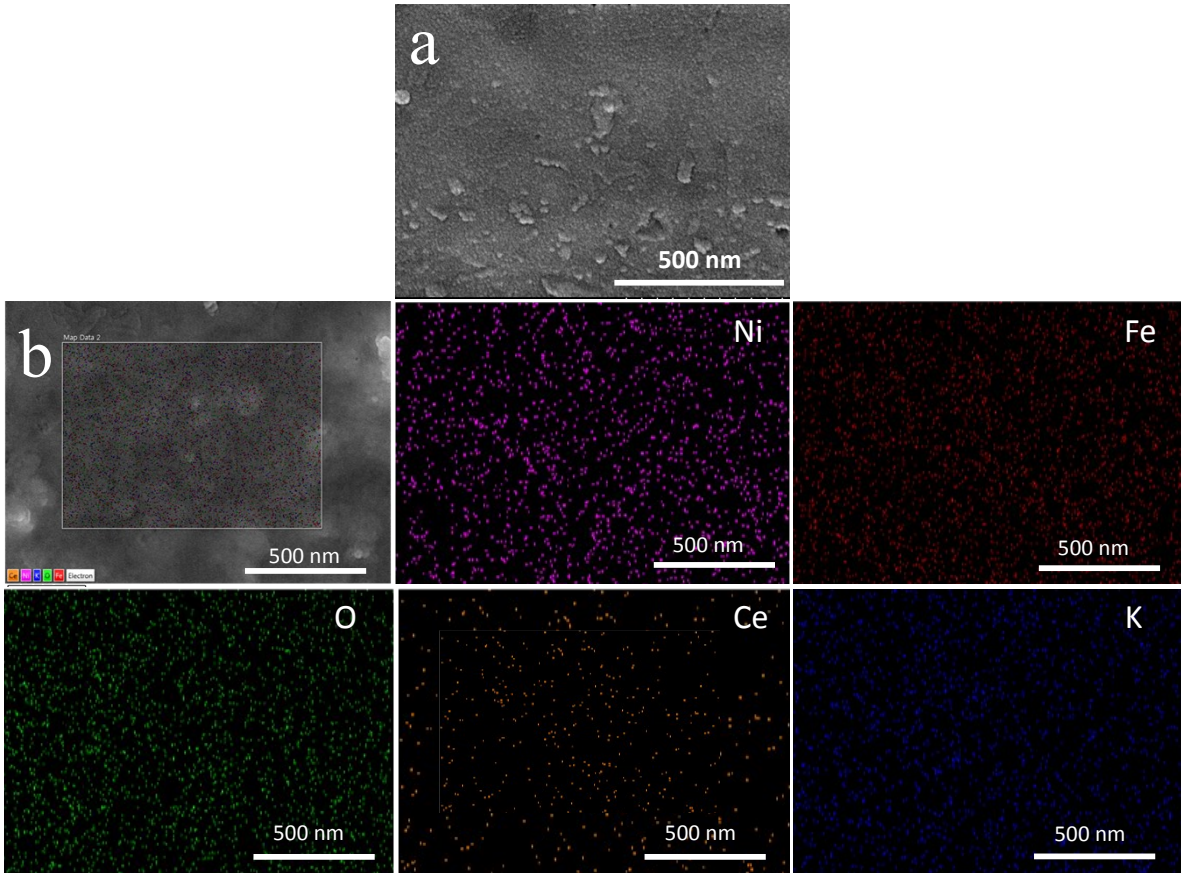

Figure S15: (a) FE-SEM image (b) EDS elemental colour mapping of anode carried out after 40 hour stability test (OER).

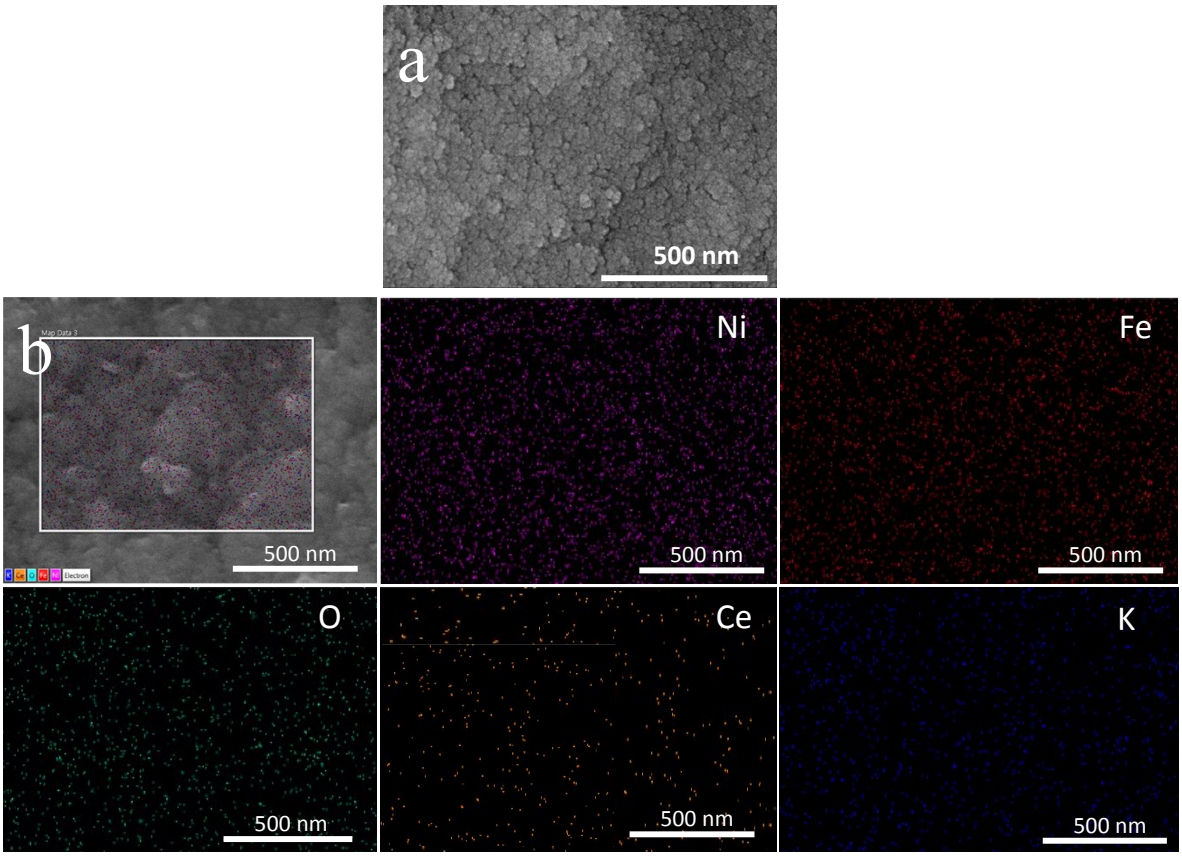

Figure S16: (a) FE-SEM image (b) EDS elemental colour mapping of cathode carried out after 40 hour stability test (HER).

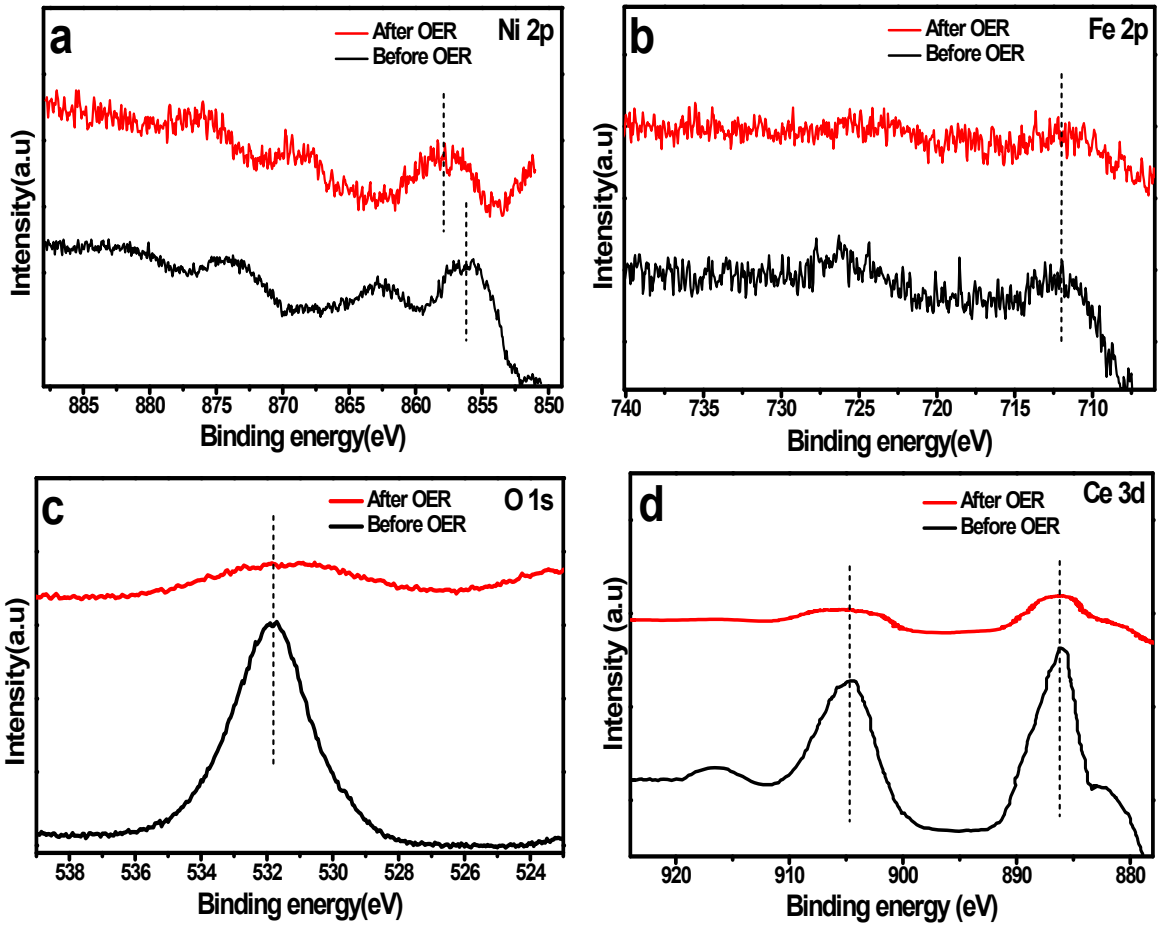

Figure S17: XPS analysis of $NiFe_{0.8}Ce_{0.2}$ electrocatalysts after 50 hour OER stability test.