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

Boosting Electrocatalytic Water Oxidation of NiFe Layered Double Hydroxide *via* the Synergy of 3d-4f Electron Interaction and Citrate Intercalation

Shuo Chen,[a] Zhicheng Zheng,[a] Qing ying Li,[a] Hao Wan,[b,] Gen Chen,[a] Ning Zhang,[a] Xiaohe Liu,[a,b,*] Renzhi Ma[c,*]*

[a] School of Materials Science and Engineering, Central South University, Changsha, Hunan 410083, P. R. China. E-mail: liuxh@csu.edu.cn

[b] Zhongyuan Critical Metals Laboratory, Zhengzhou University, Zhengzhou 450001, P. R. China. E-mail: wanhao@zzu.edu.cn

[c] International Center for Materials Nanoarchitectonics (WPI-MANA), National Institute for Materials Science (NIMS), Namiki 1-1, Tsukuba, Ibaraki 305-0044, Japan. E-mail: Ma.Renzhi@nims.go.jp

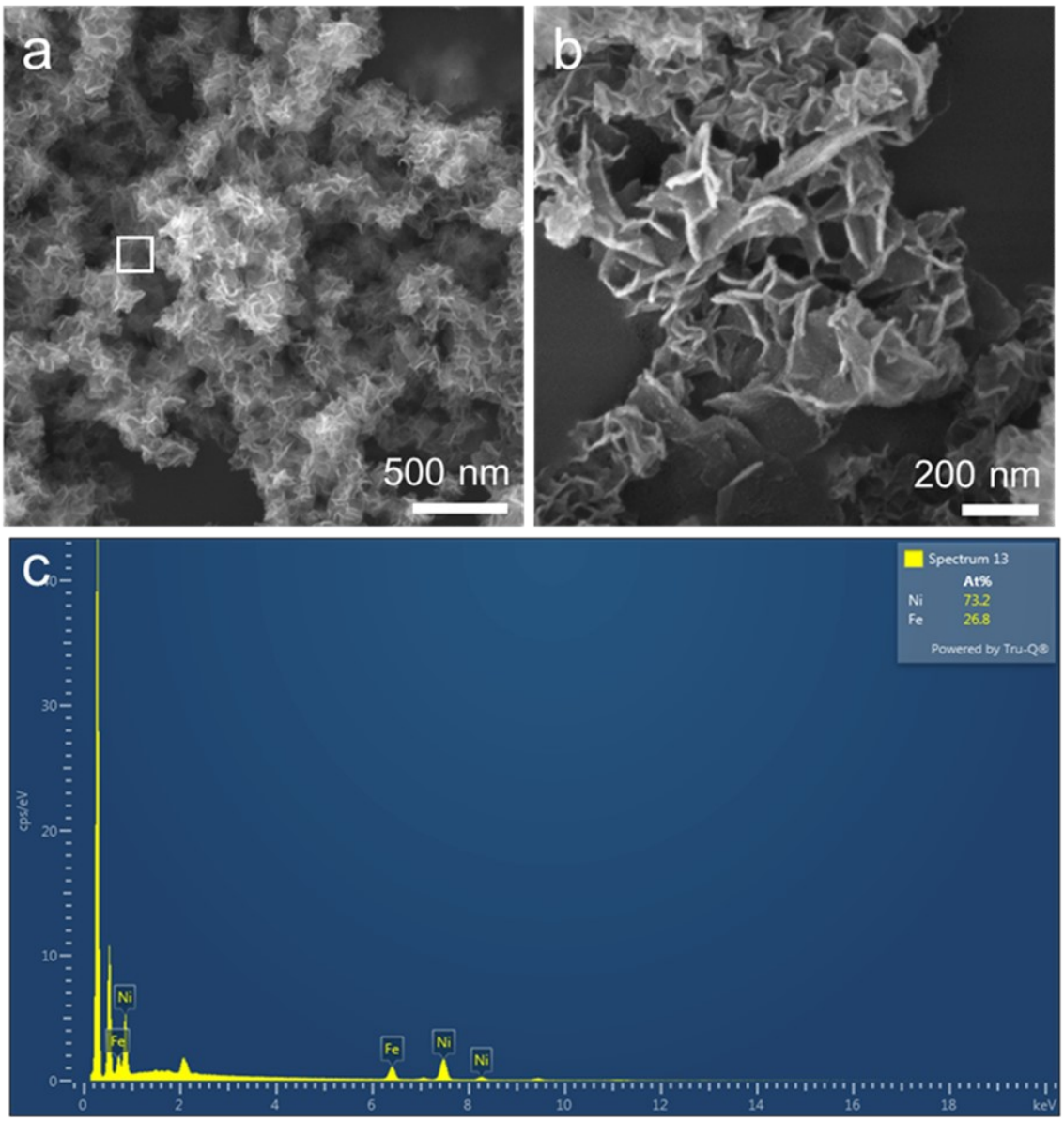

Figure S1. (a-b) SEM images and (c) EDX result of Ni_2Fe_1 PNs.

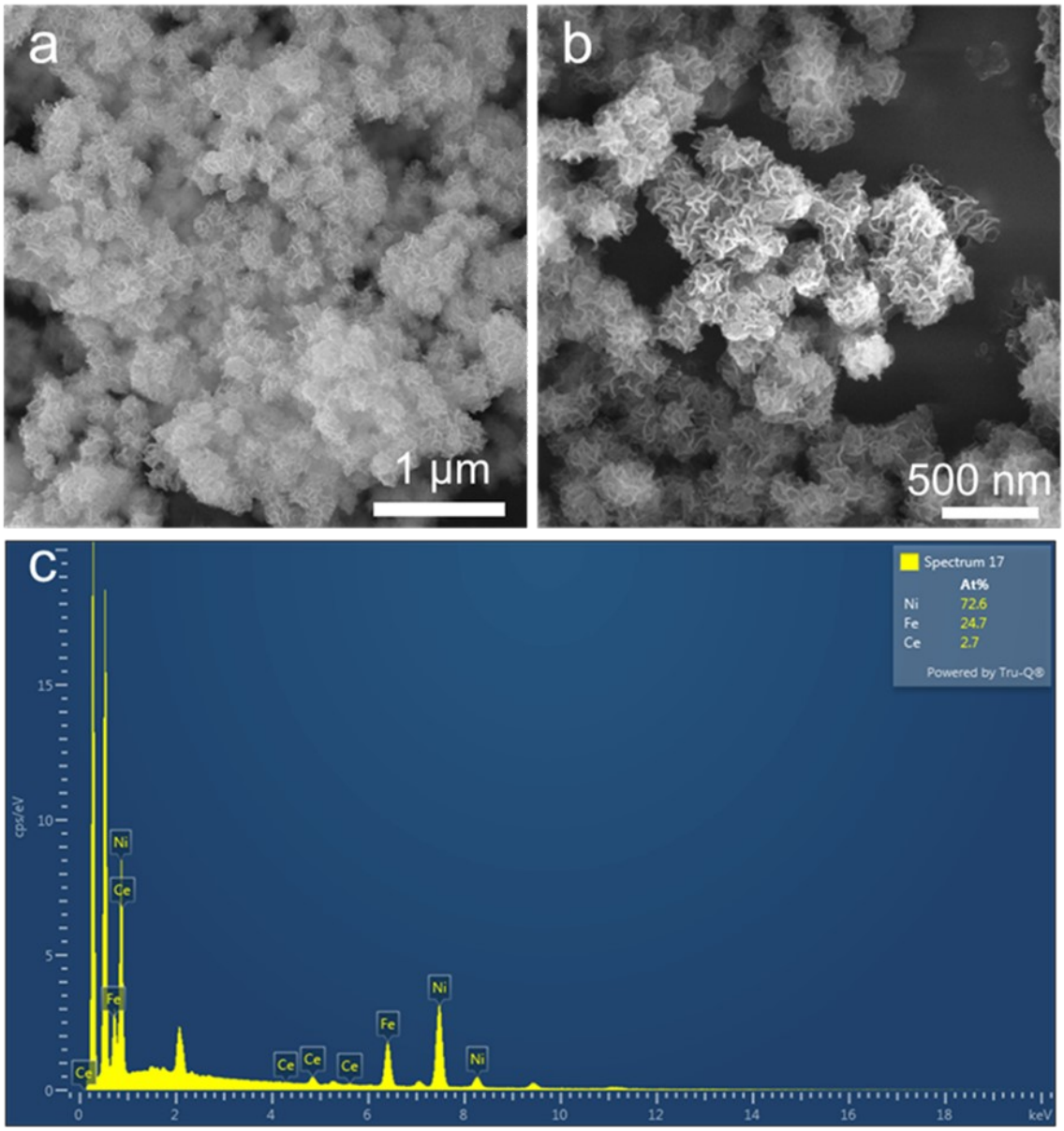

Figure S2. (a-b) SEM images and (c) EDX result of $Ni_2Fe_{0.9}Ce_{0.1}$ PNs.

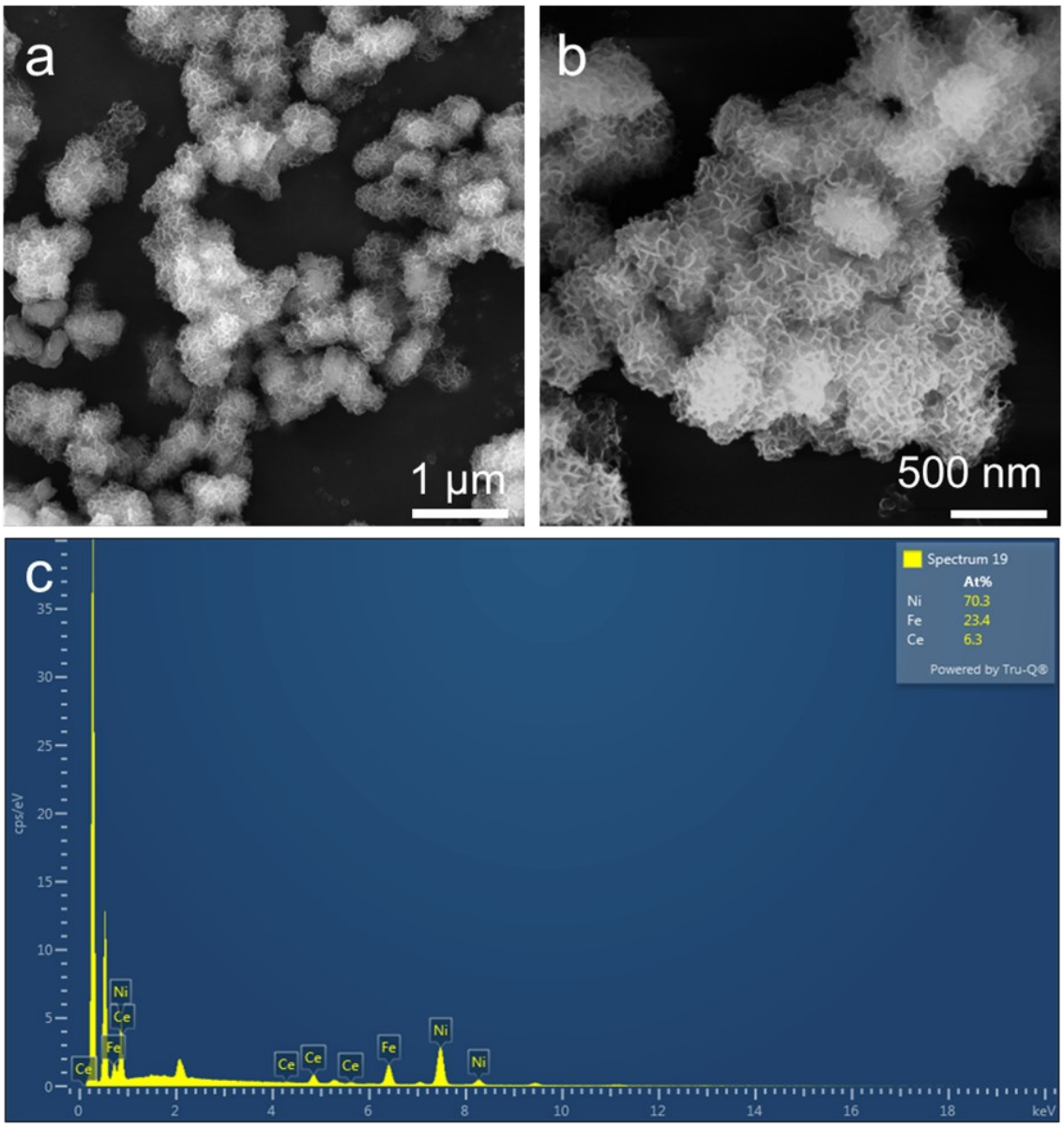

Figure S3. (a-b) SEM images and (c) EDX result of $Ni_2Fe_{0.8}Ce_{0.2}$ PNs.

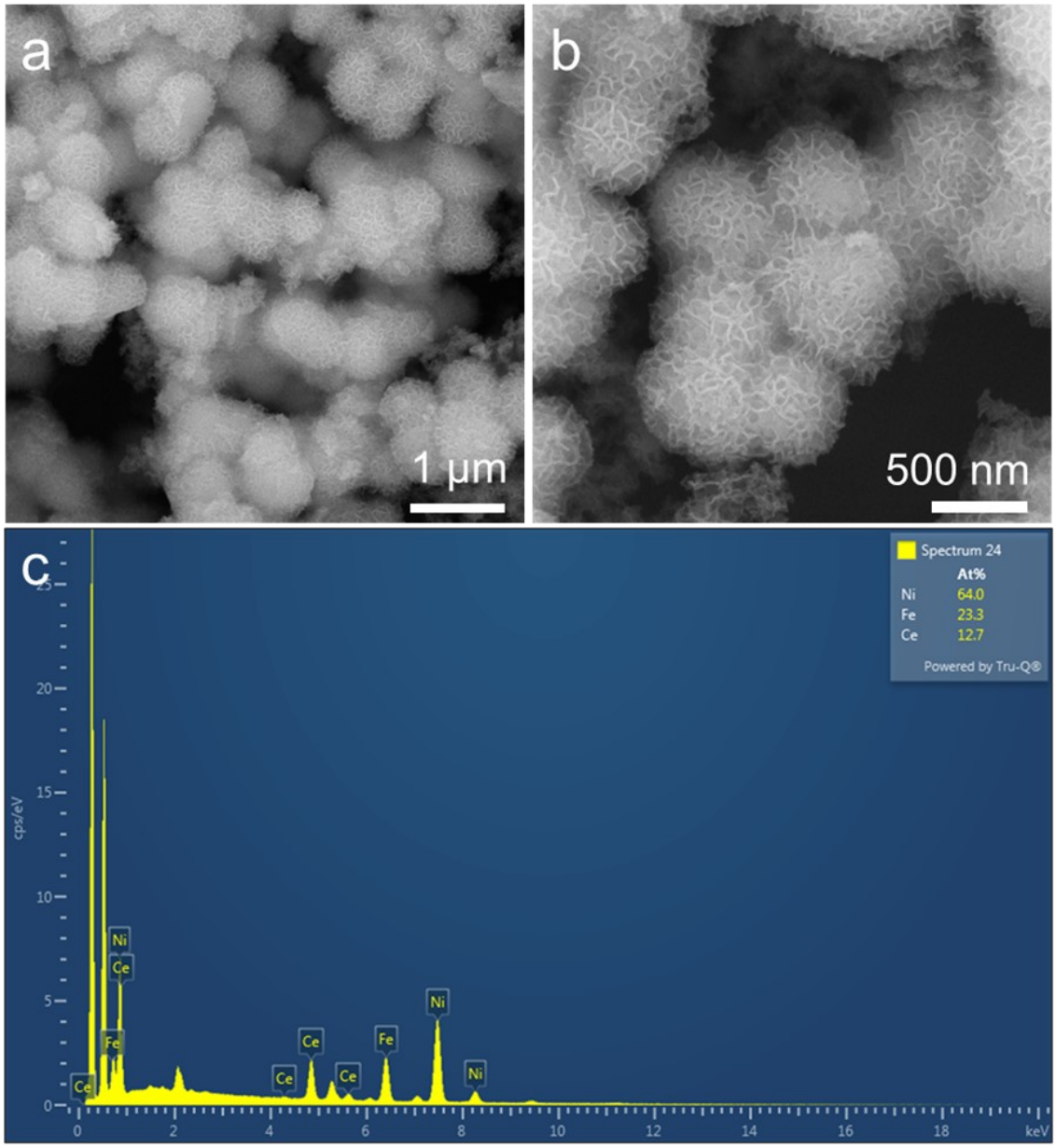

Figure S4. (a-b) SEM images and (c) EDX result of $Ni_2Fe_{0.7}Ce_{0.3}$ PNs.

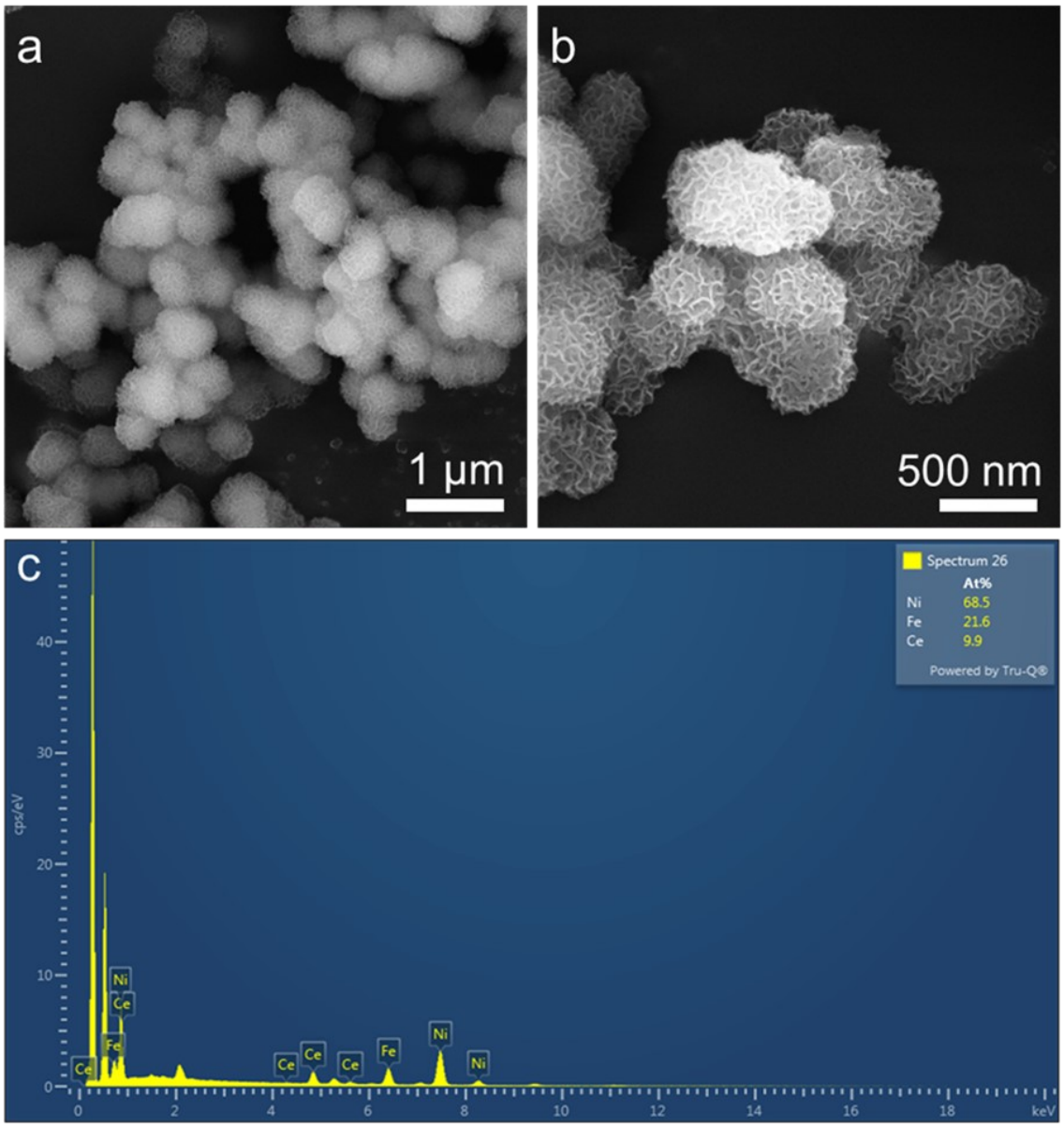

Figure S5. (a-b) SEM images and (c) EDX result of $Ni_2Fe_{0.6}Ce_{0.4}$ PNs.

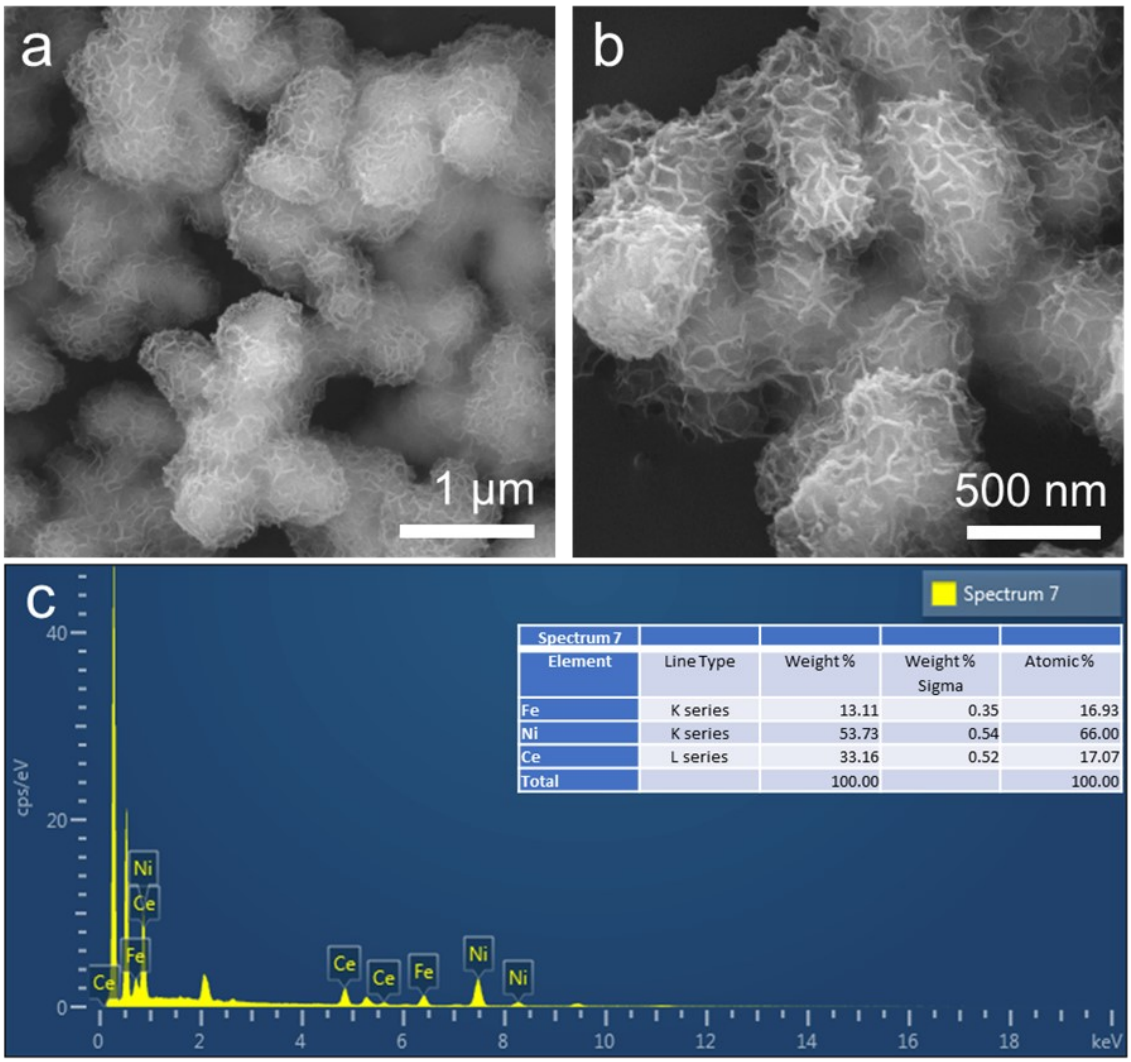

Figure S6. (a-b) SEM image and (c) EDX result of $Ni_2Fe_{0.5}Ce_{0.5}$ PNs.

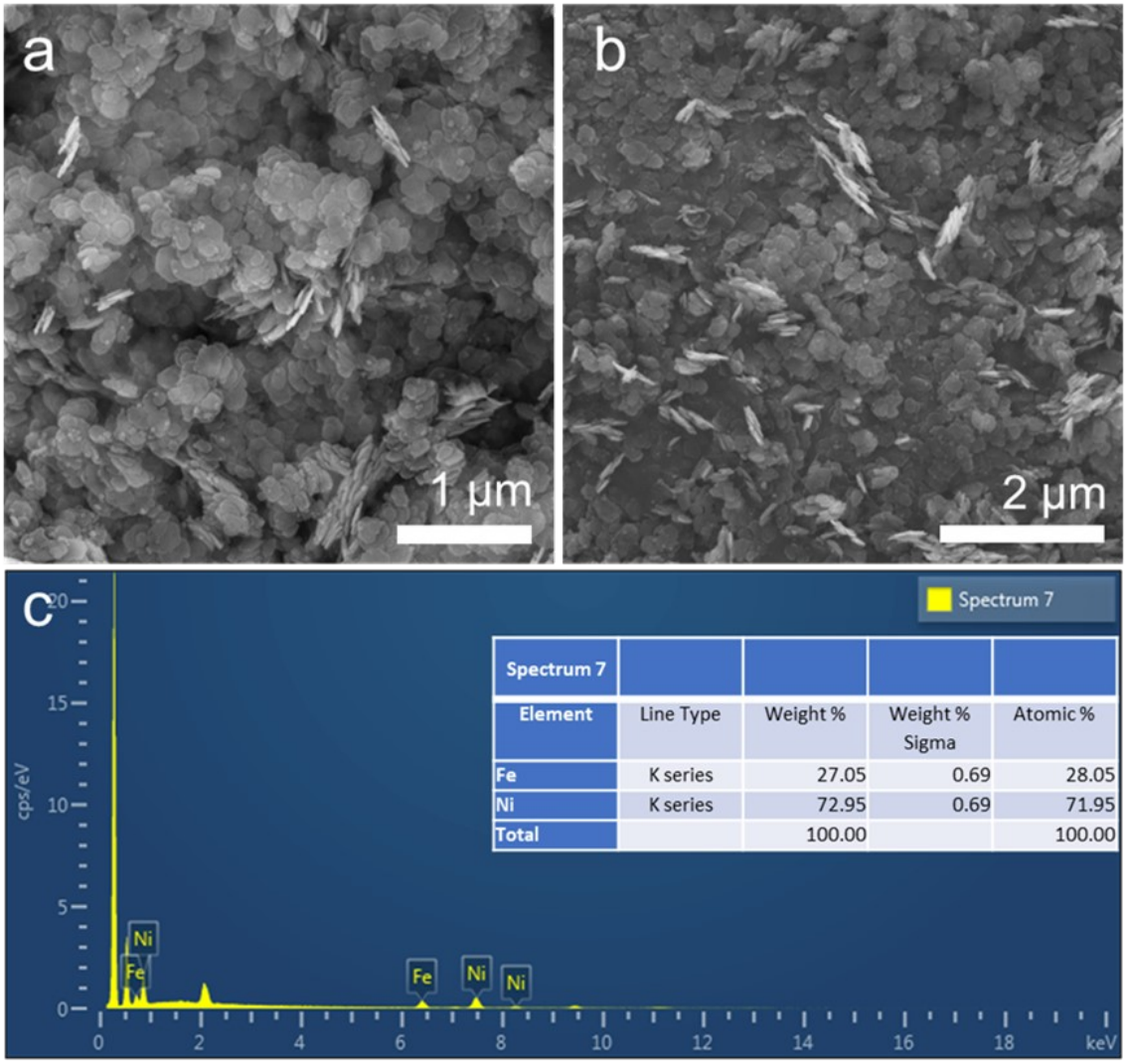

Figure S7. (a-b) SEM images and (c) EDX result of Ni_2Fe_1 Ns.

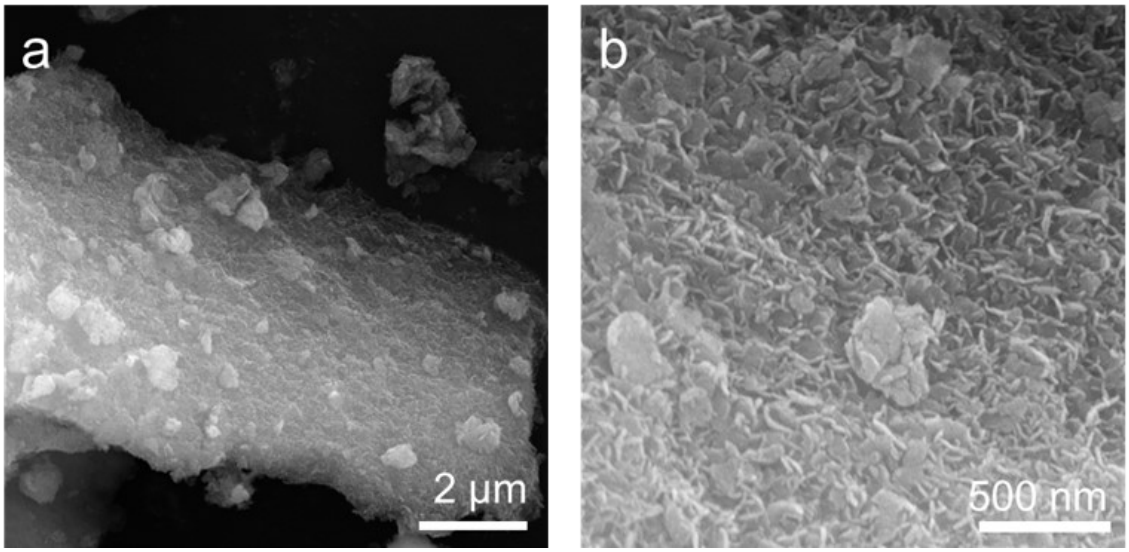

Figure S8. (a-b) SEM images of Ni_2Fe_1 LDH product prepared without citrate addictive.

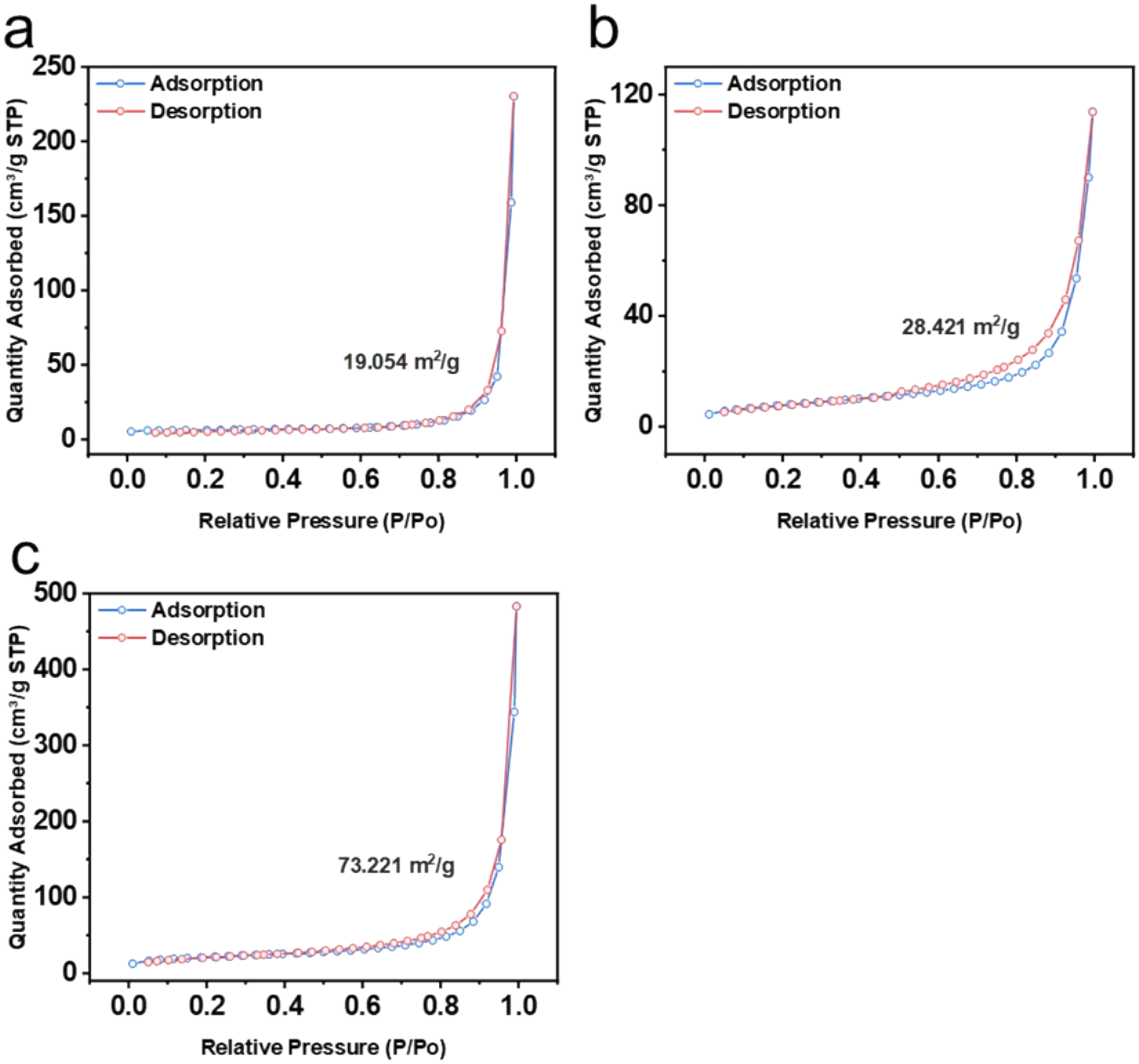

Figure S9. N_2 sorption/desorption isotherms of (a) Ni_2Fe_1 Ns, (b) $Ni_2Fe_{0.7}Ce_{0.3}$ PNs and (c) Ni_2Fe_1 PNs.

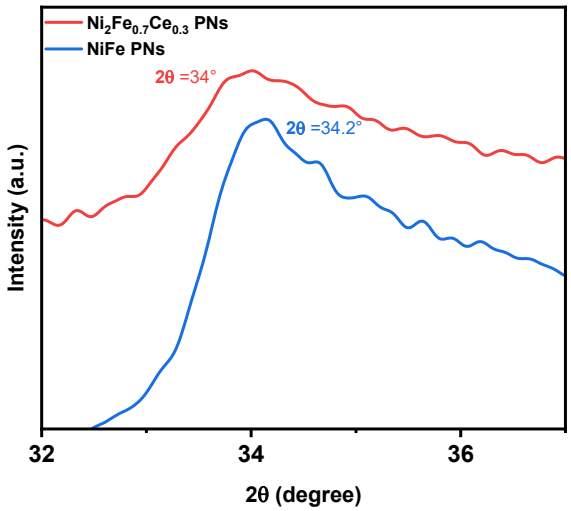

Figure S10. Magnified XRD pattern for (012) peak.

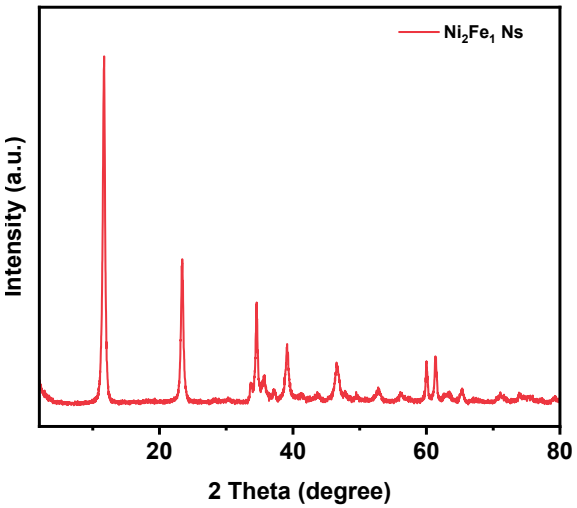

Figure S11. XRD patterns of Ni_2Fe_1 Ns.

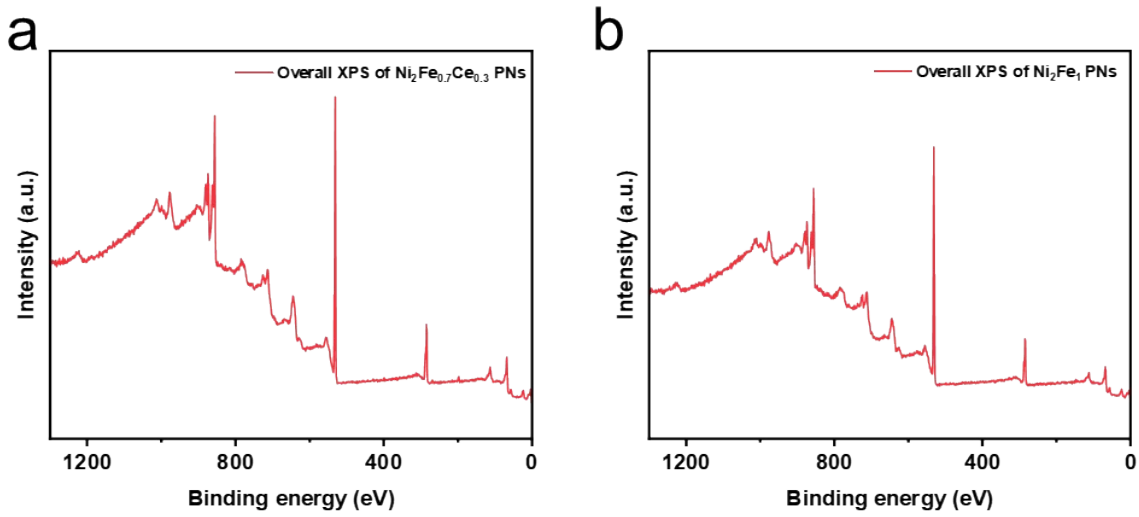

Figure S12. Overall XPS surveys of (a) $Ni_2Fe_{0.7}Ce_{0.3}$ PNs and (b) Ni_2Fe_1 PNs.

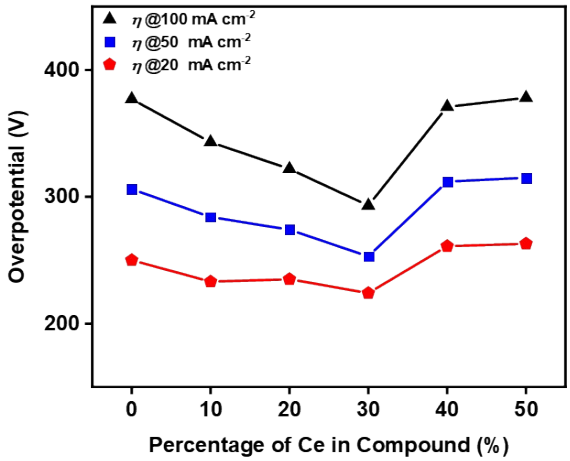

Figure S13. Overpotential at different current density.

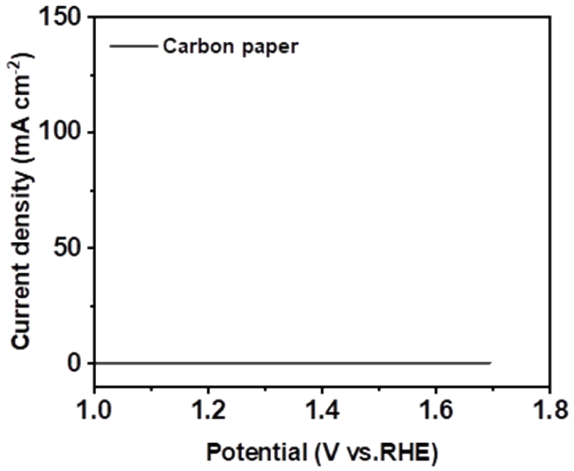

Figure S14. OER activity of carbon paper.

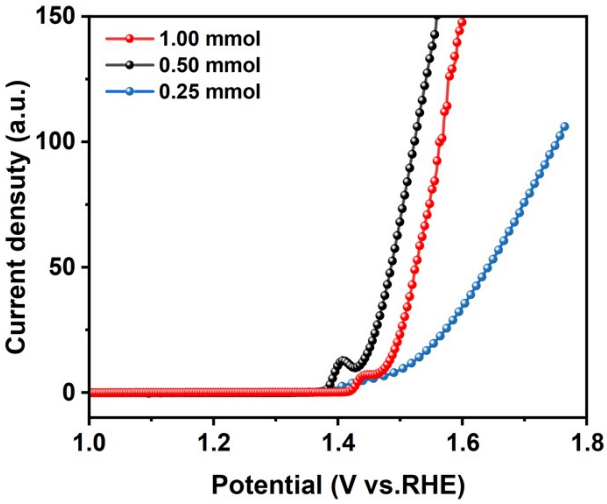

Figure S15. LSV curves of catalysts with different citrate additive amounts.

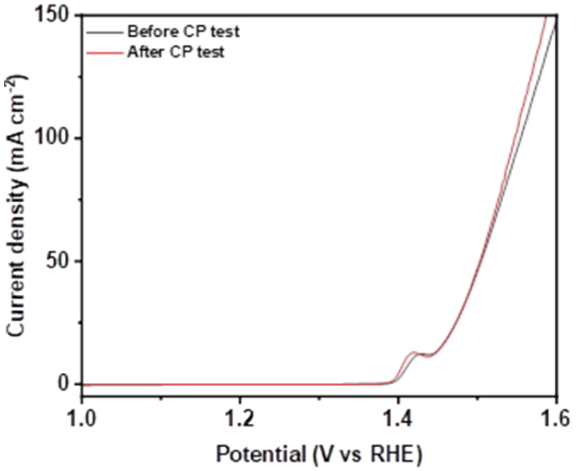

Figure S16. LSV curves of $Ni_2Fe_{0.7}Ce_{0.3}$ PNs without iR-correction before and after Chronopotentiometric test.

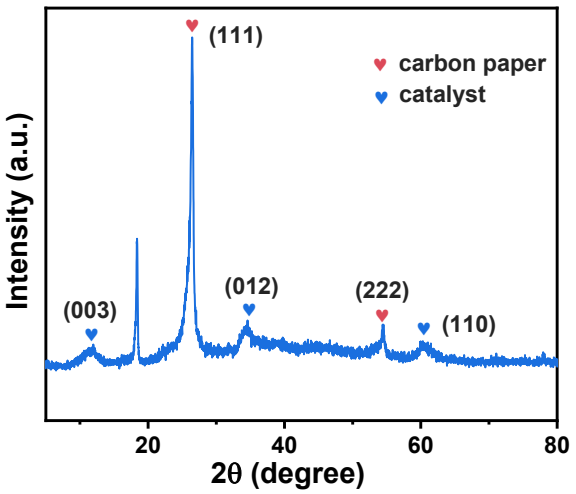

Figure S17. XRD pattern of $Ni_2Fe_{0.7}Ce_{0.3}$ PNs after chronopotentiometric test.

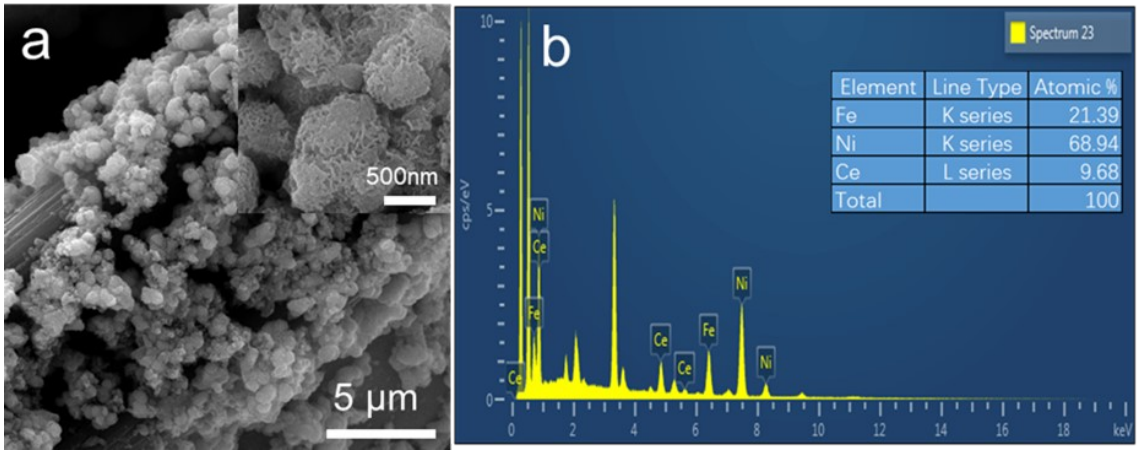

Figure S18. (a) SEM images and (b) EDX result after chronopotentiometric test.

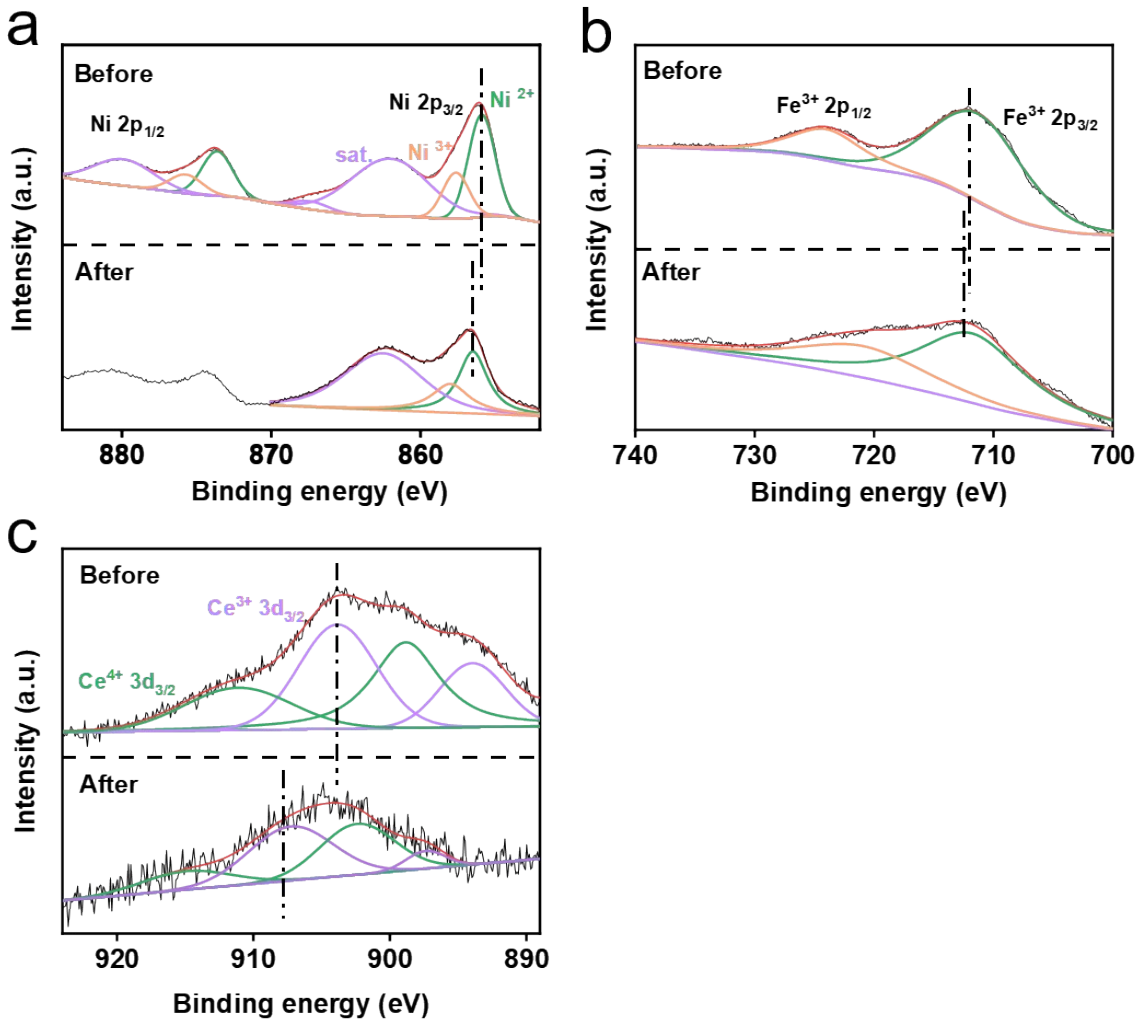

Figure S19. XPS spectra after the chronopotentiometric test. (a) Ni 2p, (b) Fe 2p and (c) Ce 3d.

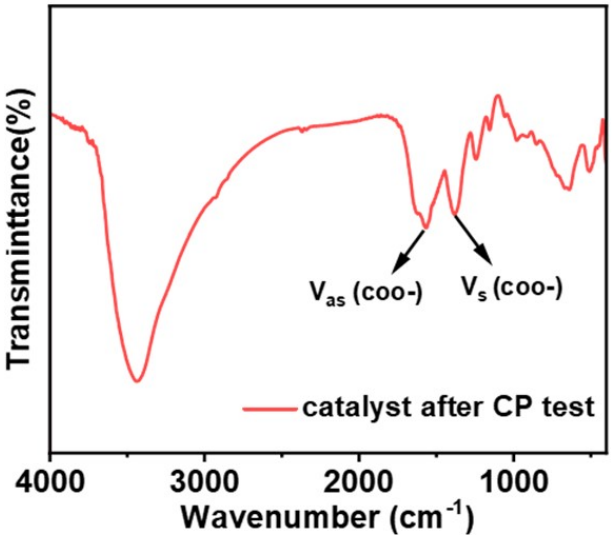

Figure S20. The FT-IR spectrum of $Ni_2Fe_{0.7}Ce_{0.3}$ PNs after the chronopotentiometric test.

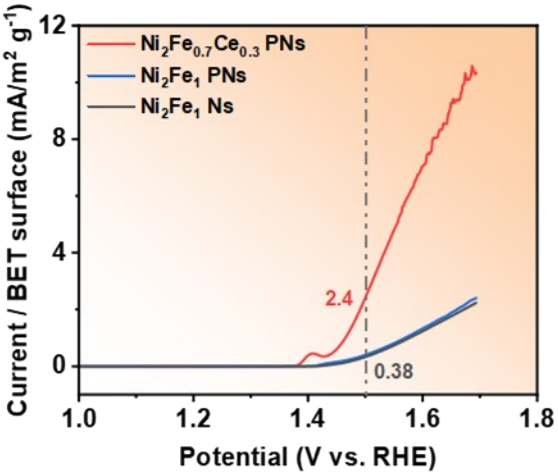

Figure S21. LSV curves normalized by BET surface areas.

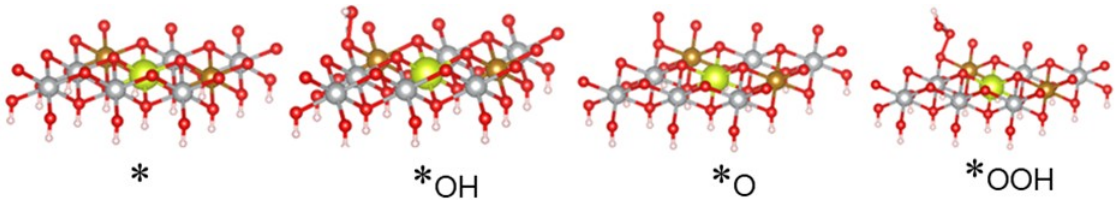

Figure S22. Adsorption models of each OER elementary step.

Table S1. Binding energies and corresponding fitting relative areas in XPS spectra

Orbits	$Ni_2Fe_{0.7}Ce_{0.3}$ PNs	Ni_2Fe_1 PNs
Fe^{3+} $2p_{3/2}$	711.43 eV /311657.4	712.50 eV/ 338187.2
Fe^{3+} $2p_{1/2}$	724.09 eV/ 155828.7	724.91 eV/ 169093.6
Ni^{2+} $2p_{3/2}$	856.07 eV/ 214567.7	855.69 eV/ 237217.3
Ni^{3+} $2p_{1/2}$	857.63 eV/ 96498.85	857.21 eV/ 83106.64
Ce^{3+} $3d_{3/2}$	904.33 eV/ 57828.39	/
Ce^{4+} $3d_{3/2}$	910.83 eV/ 70843.83	/

Table S2. Overpotential delivering different current densities

	20mA cm^{-2}	**50mA cm^{-2}**	**100mA cm^{-2}**
Ni_2Fe_1 PNs	250	306	377
$Ni_2Fe_{0.9}Ce_{0.1}$	233	284	343
$Ni_2Fe_{0.8}Ce_{0.2}$	235	274	322
$Ni_2Fe_{0.7}Ce_{0.3}$	224	253	293
$Ni_2Fe_{0.6}Ce_{0.4}$	261	312	371
$Ni_2Fe_{0.5}Ce_{0.5}$	263	315	378
Ni_2Fe_1 Ns	352	419	534
RuO_2	381		

Table S3. The values of parameters in the fitting circuit

	$Ni_2Fe_{0.5}Ce_{0.5}$ PNs	**$Ni_2Fe_{0.6}Ce_{0.4}$ PNs**	**$Ni_2Fe_{0.7}Ce_{0.3}$ PNs**	**$Ni_2Fe_{0.8}Ce_{0.2}$ PNs**	**$Ni_2Fe_{0.5}Ce_{0.5}$ PNs**	**Ni_2Fe_1 PNs**	**Ni_2Fe_1 Ns**
Rs (Ω)	3.161	2.941	2.915	2.877	2.863	2.783	2.821
CPE-1-T	0.0049445	0.011861	0.01443	0.015921	0.0065464	0.00025401	0.0020007
CPE-1-P	0.60365	0.54977	0.76804	0.81399	0.67977	0.87632	0.71344
R_{ct1} (Ω)	1.096	1.187	0.28386	0.24508	0.52489	0.64906	2.167
CPE-2-T	0.054842	0.041936	0.070164	0.06374	0.064373	0.051382	0.0078659
CPE-2-T	0.7567	0.78909	0.86533	0.85811	0.79748	0.65801	0.7131
R_{ct2} (Ω)	9.384	7.162	4.232	4.563	4.379	5.229	20.98

Table S4. OER activity of recently reported NiFe-based catalysts

Catalysts	Overpotential at 20 mA cm^{-2}	Supporter	Loading Capacity (mg cm-1)	References
$Ni_2Fe_{0.7}Ce_{0.3}$ PNs	224	CP	0.4	This work
NiFeS@Ti_3C_2 MXene/NF	290	NF	1.25	1
CoO@NiFe LDH/NF	225	NF	0.78	2
$Co_{1.98}$-NiFe LDH	236	CP	1	3
Fe_2P-NiCoP	239	NF		4
NiFe LDH-Ci/CC	240	CC	0.67	5
Ni–Fe LDH DSNCs	246	CP	0.22	6
NiFeW-LDH@CP	248	CP	0.8	7
$NiFe_2O_4$@N/rGO	252	NF	1	8
NiFe-Pi/P	255	NF	2	9
$(Ni_{0.5}Fe_{0.5})C_2O_4$ nanorods	266	NF		10
NiFe PBAs	267	rotating disk electrode (RDE)	0.15	11
CNT@NiFe-LDH NS	270	NF	0.5	12
MoNiFe-LDH	317	glass carbon		13

CP, CC, NF refer to carben paper, carbon cloth and nickel foam respectively.

References

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