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Supporting Information

Solventless synthesis of nanospinel $Ni_{1-x}Co_xFe_2O_4$ ($0 \leq x \leq 1$) solid solutions for efficient electrochemical water splitting and supercapacitance

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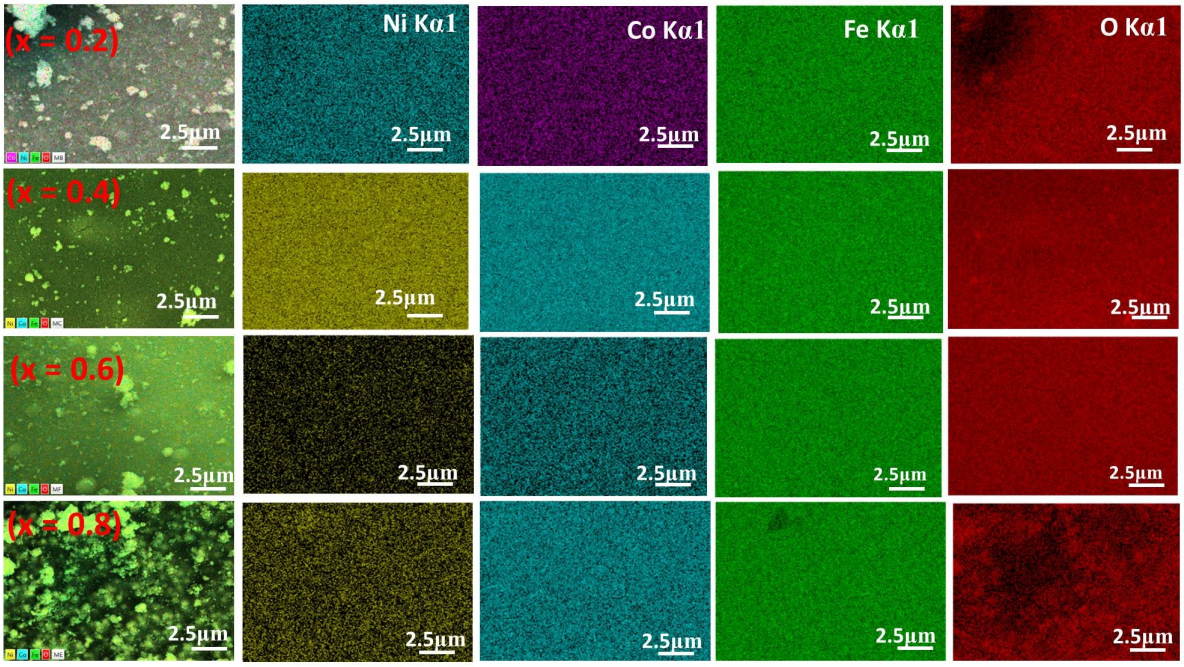

Fig. S1. EDX elemental mapping of $Ni_{1-x}Co_xFe_2O_4$ ($0.2 \leq x \leq 0.8$) solid solutions showing a uniform distribution of elements.

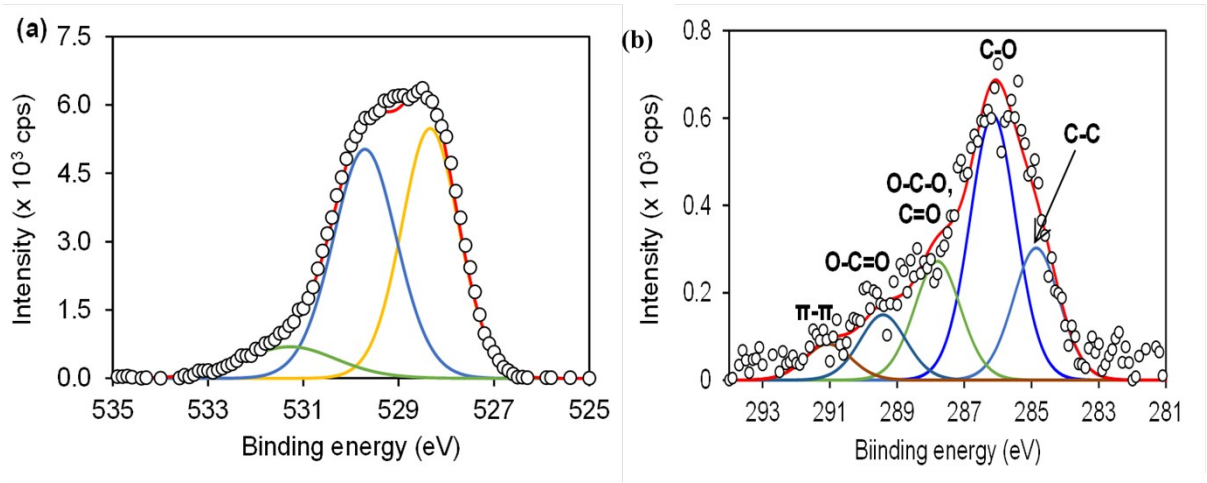

Fig. S2. XPS High resolution core-level spectra of (a) O 1s and (b) C 1s of the ternary $Ni_{1-x}Co_xFe_2O_4$ (x = 0.4).

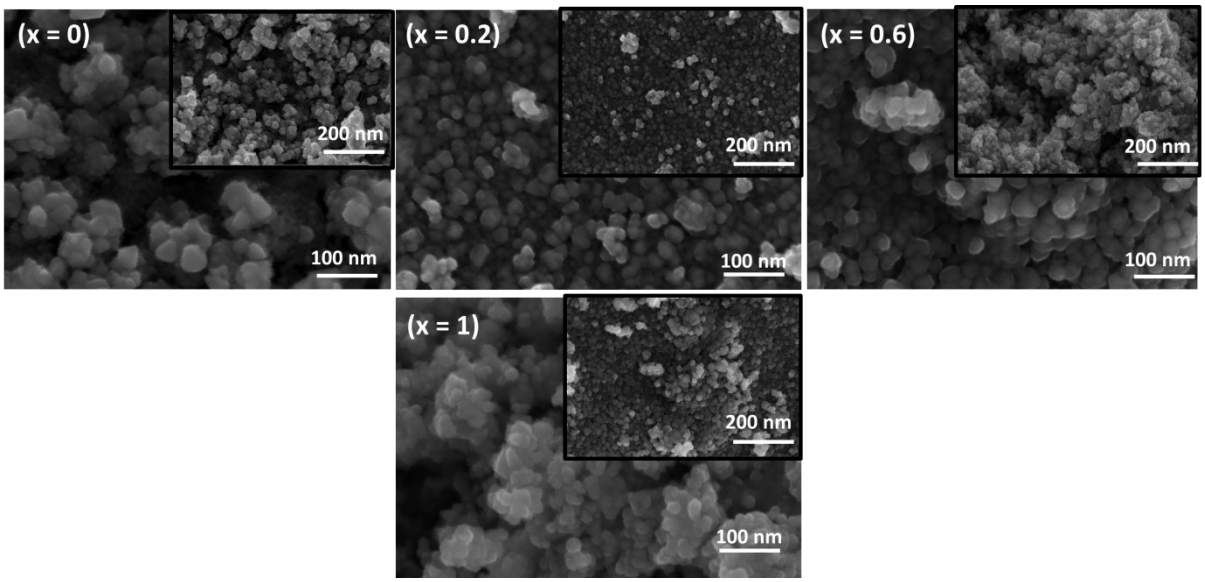

Fig. S3. SEM images of $Ni_{1-x}Co_xFe_2O_4$ (0, 0.2, 0.6 and 1) nanoparticles.

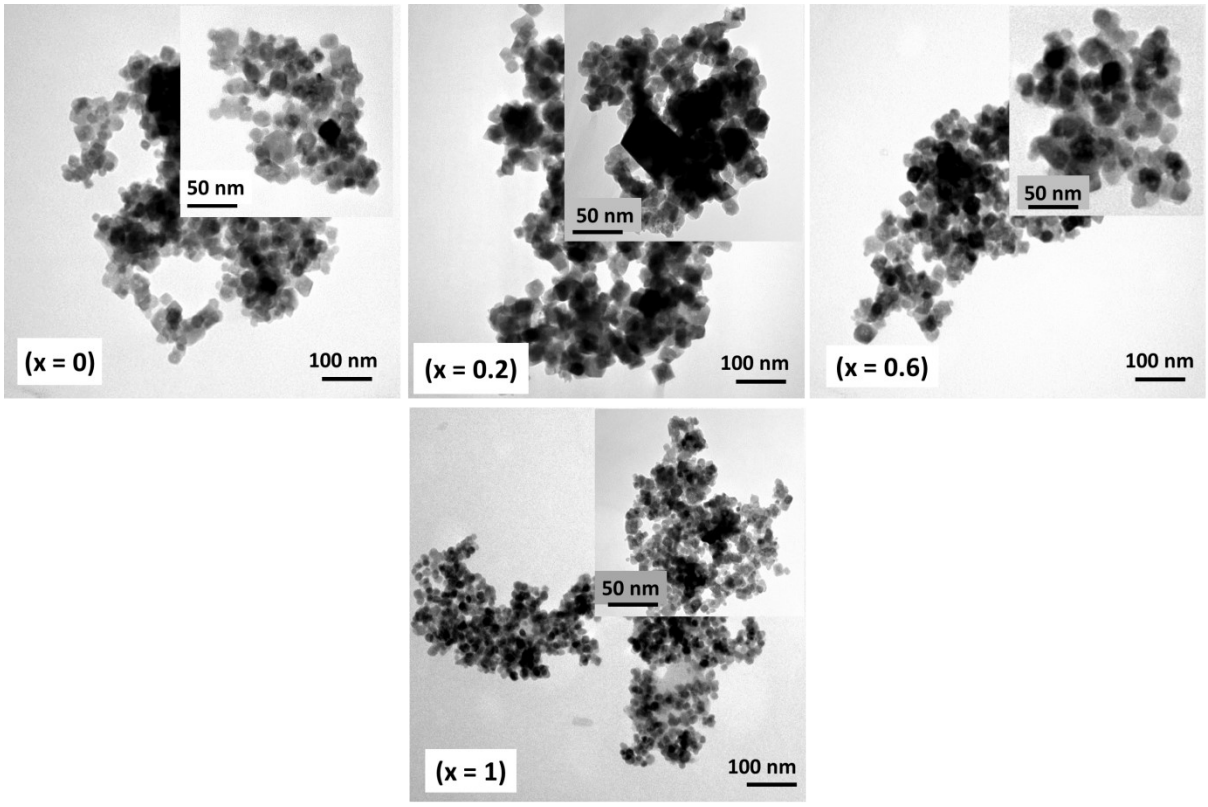

Fig. S4. TEM images of $Ni_{1-x}Co_xFe_2O_4$ (0, 0.2, 0.6 and 1) nanoparticles.

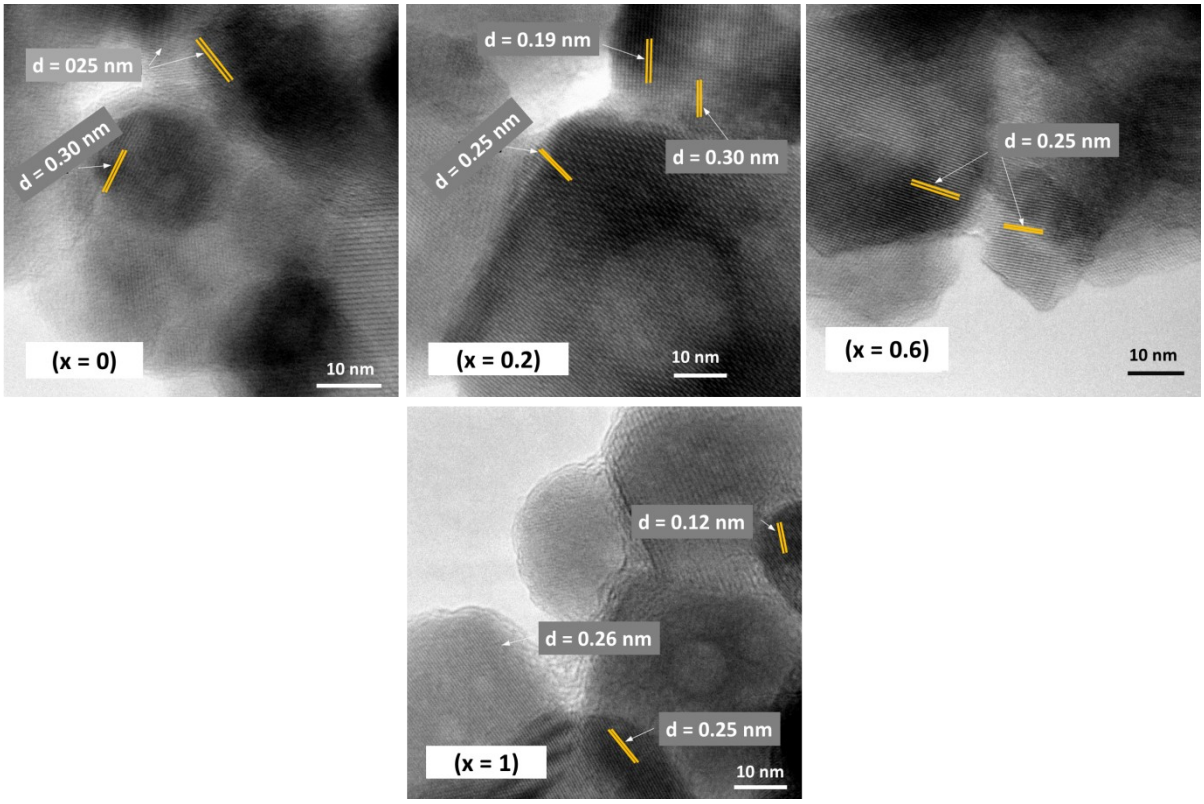

Fig. S5. HRTEM images of $Ni_{1-x}Co_xFe_2O_4$ (0, 0.2, 0.6 and 1) nanoparticles.

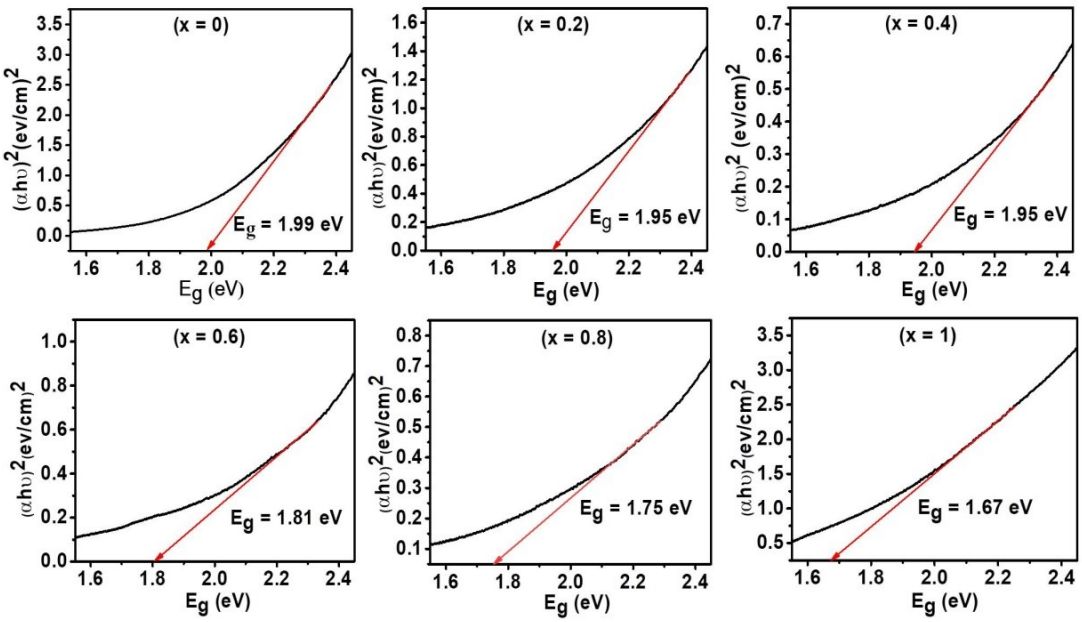

Fig. S6. Tauc plots of $(\alpha h\upsilon)^2$ versus energy for $Ni_{1-x}Co_xFe_2O_4$ ($0 \leq x \leq 1$) solid solutions.

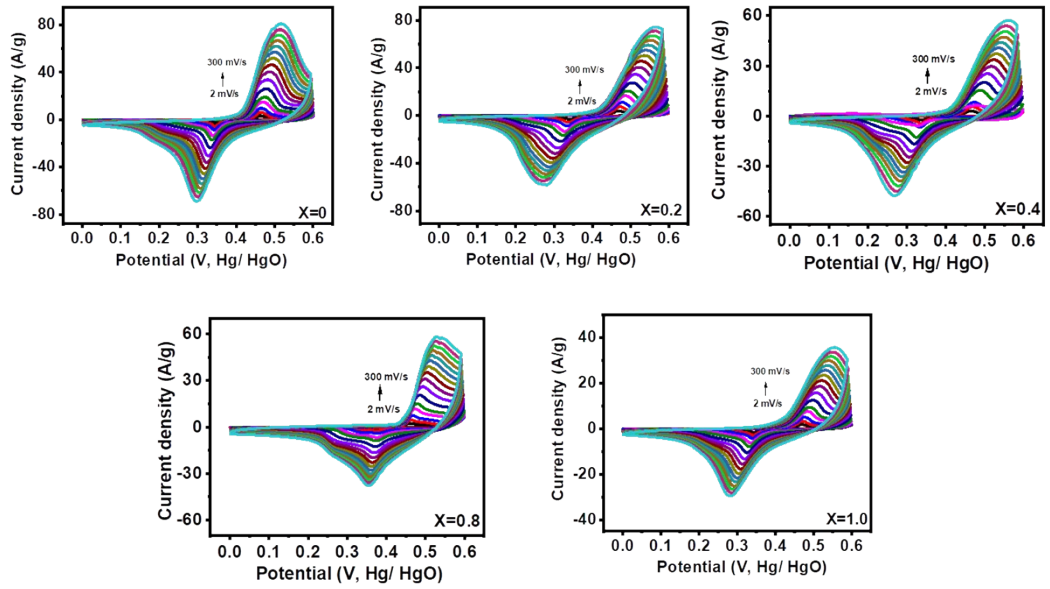

Fig. S7. CV curves of the $Ni_{1-x}Co_xFe_2O_4$ ($0 \leq x \leq 1$) samples at various scan rates.

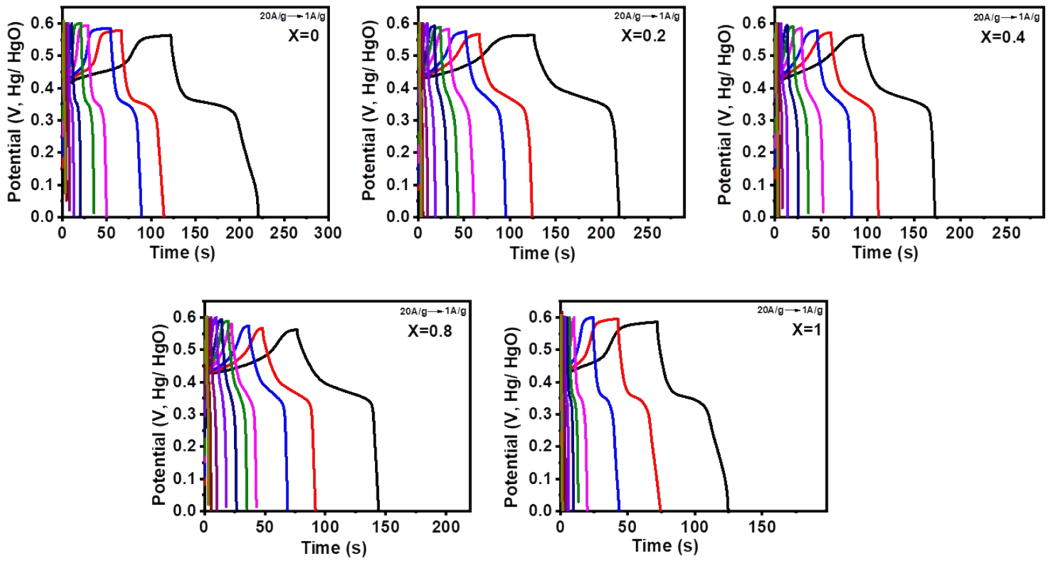

Fig. S8. Charge-discharge characteristics of $Ni_{1-x}Co_xFe_2O_4$ ($0 \leq x \leq 1$) samples at various current densities.

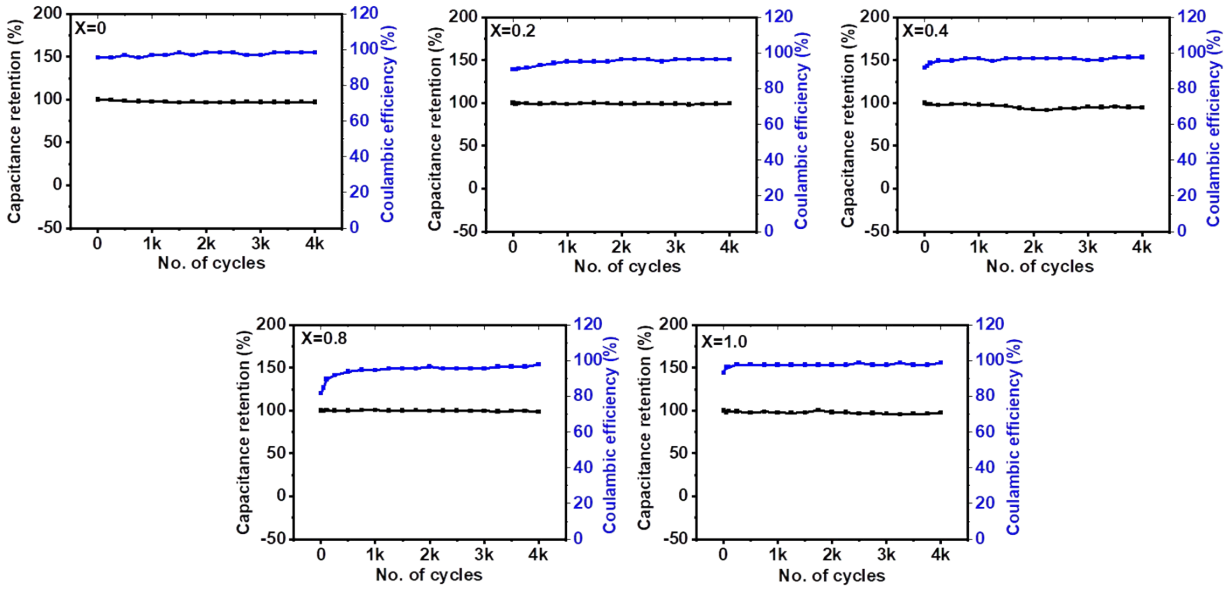

Fig. S9. Capacitance retention and columbic efficiency of various samples.

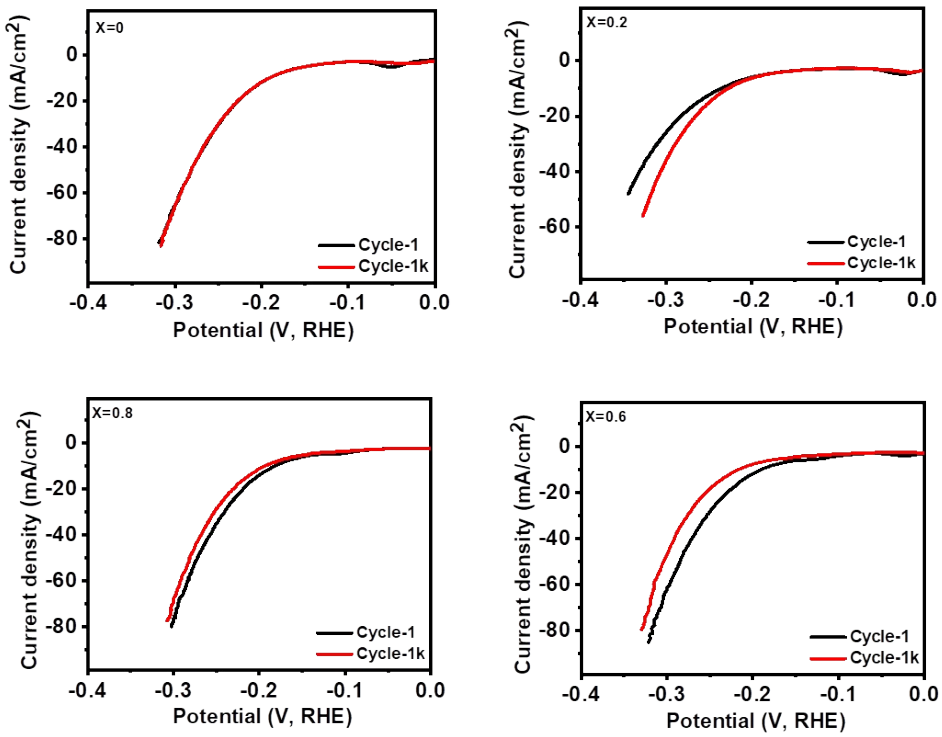

Fig. S10. HER polarization curves at various cycles for the various samples.

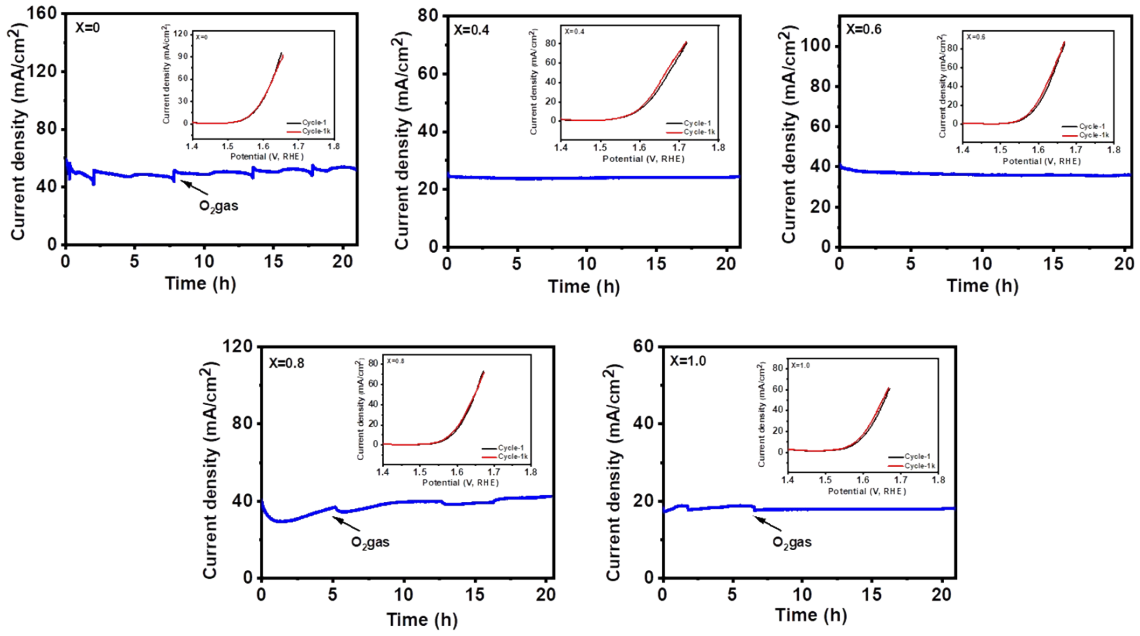

Fig. S11. Chronoamperometry characteristics of the various samples in 1M KOH (inset) OER polarization curves at various cycles for the various samples.

Table S1. Mass of precursors used in the synthesis of $Ni_{1-x}Co_xFe_2O_4$ ($0 \leq x \leq 1$) solid solutions.

(x)	**Target formula**	**Mass of $Ni(acac)_2$ (g)**	**Mass of $Co(acac)_3$ (g)**	Mass of $Fe(acac)_3$ (g)
0	$NiFe_2O_4$	0.100	-	0.275
0.2	$Ni_{0.8}Co_{0.2}Fe_2O_4$	0.080	0.028	0.275
0.4	$Ni_{0.6}Co_{0.4}Fe_2O_4$	0.059	0.055	0.275
0.6	$Ni_{0.4}Co_{0.6}Fe_2O_4$	0.039	0.083	0.275
0.8	$Ni_{0.2}Co_{0.8}Fe_2O_4$	0.019	0.111	0.275
1.0	$CoFe_2O_4$	-	0.100	0.198

Table S2. Comparison of the specific capacitance of $Ni_{0.4}Co_{0.6}Fe_2O_4$ electrode with other metal oxide-based electrodes.

Electrode material	**Synthesis route**	**Specific capacitance (F/g)**	**Current density (A/g)**	**Reference**
$Cu_{0.5}Co_{0.5}Fe_2O_4$	Sol-gel	76.9	1	[1]
$MnFe_2O_4$	Co-precipitation	173	1	[2]
$MgFe_2O_4$	Sol-gel	61	0.5	[3]
$Ni_{0.5}Co_{0.5}Fe_2O_4$	Sol-gel	50	1	[1]
Fe_3O_4	Solvothermal	97	3	[4]
$NiCo_2O_4$	Sol-gel	217	1	[5]
$Ni_{0.5}Cu_{0.5}Fe_2O_4$	Sol-gel	44	1	[1]
$MnFe_2O_4$ /graphene	Solvothermal	120	0.1	[6]
$CoMnFeO_4$	Sol-gel	150	1	[7]
$CdMn_2O_4$	Electrospinning	210	1	[8]
$Ni_{0.25}Mg_{0.75}Fe_2O_4$	Hydrothermal	133.95	0.5	[9]
$MgCr_2O_4$	Sol-gel	21	0.5	[10]
$Ni_{0.4}Co_{0.6}Fe_2O_4$	**Solventless**	**237**	**1**	**This study**

Table S3. Comparison of HER performance of the synthesized $Ni_{0.6}Co_{0.4}Fe_2O_4$ and $CoFe_2O_4$ with other reported Ni/Co-based electrocatalysts in alkaline electrolyte.

Catalyst	Synthetic method	η_{10} (mV in 1 M KOH	Tafel slope (mV/dec)	Reference
NiFe Sponges	Polyol-assisted chemical synthesis	190	82	[11]
NiCoP NW/CFP	Hydrothermal followed by sulfuration and phosphorization	170	73.0	[12]
$Ni_{1.5}Co_{1.5}S_4$NW/CFP		237	112.9	[12]
$CoFe_2O_4$/SWNTs	Sonochemical	263	46	[13]
Ni-$MoSe_2$	Hydrothermal	206	81	[14]
NiMnP	Colloidal	490	238	[15]
$FeSe_2/CoFe_2O_4$	Hydrothermal	231	88.76	[16]
CoCuZn/C	Electrodeposition	213	92	[17]
$NiCo_2S_4$/Ni foam	hydrothermal	210	-	[18]
$CoSe_2/MoSe_2$	Solvothermal	218	76	[19]
$NiCo_2O_4$/NiCoP	Solvothermal followed by phosphorization	198	91	[20]
$CoFe_2O_4$	**Solventless**	**169**	**113**	**This study**
$Ni_{0.6}Co_{0.4}Fe_2O_4$	**Solventless**	**168**	**120**	**This study**

Table S4. Comparison of OER performance of the synthesized $Ni_{0.8}Co_{0.2}Fe_2O_4$ with other reported Ni/Co-based electrocatalysts in alkaline electrolyte.

Catalyst	Synthetic method	η_{10} (mV) in 1 M KOH	Tafel slope (mV/dec)	Reference
NiS/Bi_2WO_6	Hydrothermal	527	238	[21]
$Ni_xCo_{3-x}O_4$/NF	Hydrothermal	320	38	[22]
$Ni_xCo_{3-x}O_4$ nonowires	Hydrothermal	337	75	[23]
Co/Fe-MOFs	Solvothermal	410	101	[24]
$MnO_2/NiCo_2O_4$/NF	Hydrothermal	340	139	[25]

$NiCo_2O_4$/NF	Solvothermal	465	137	[26]
NiCoP/C nanoboxes	MOF	330	96	[27]
Mn-Co oxyphosphide	Thermal oxidation and phosphidation	370	66	[28]
$CuCo_2S_4$	Colloidal	395	115	[29]
Mn-Co oxide	Thermal oxidation	420	60	[28]
$NiCo_2O_4$	Hydrothermal	346	94	[30]
$CoNi_{0.2}Fe_{0.05}$-Z-H-P	MOF and phosphidation	329	48.2	[31]
CoS	Electrodeposition	372	86.6	[32]
CoMnP nanoparticles	Solvothermal	330	61	[33]
CoP-PBSCF	*In-situ* exsolution	340	81.5	[34]
$Co_2Mo_3O_8$@NC	*In situ* pyrolysis	331	87.5	[35]
$NiCo_2O_4$	Hydrothermal	500	119	[25]
$Ni_{0.8}Co_{0.2}Fe_2O_4$	**Solventless**	**320**	**79**	**This study**

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