Electronic Supplementary Material (ESI) for Inorganic Chemistry Frontiers.

Supporting Information

In-situ construction of $FeNi_2Se_4$-FeNi LDH heterointerfaces with electron redistribution for enhanced overall water splitting

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Supplementary Figures and Tables

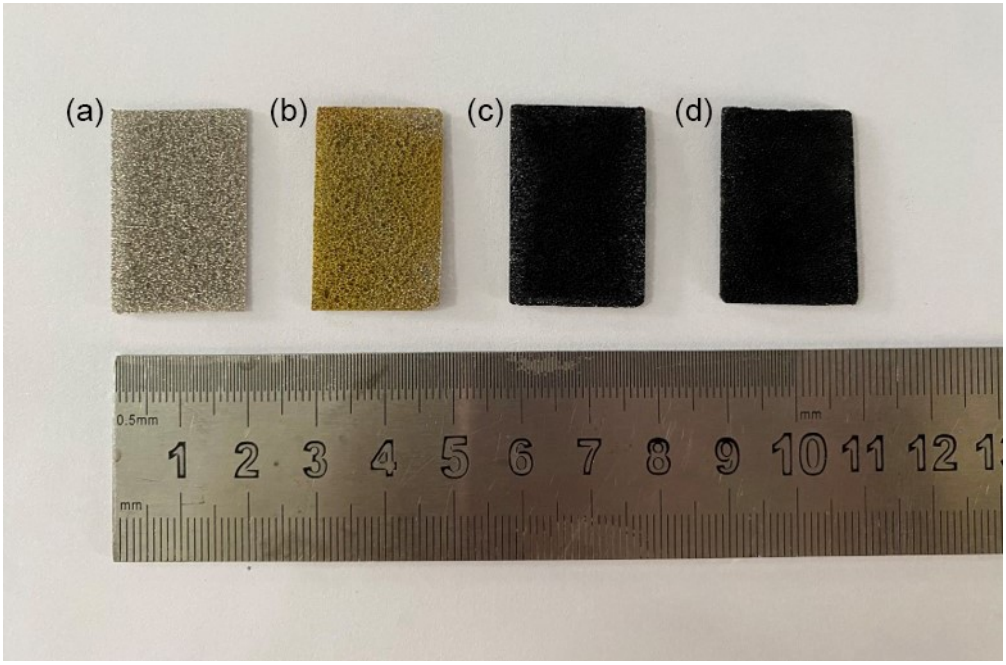

Fig. S1. Optical photograph of (a) bare nickel foam, (b) FeNi LDH/NF, (c) $FeNi_2Se_4$-FeNi LDH/NF and (d) $FeNi_2Se_4$/NF.

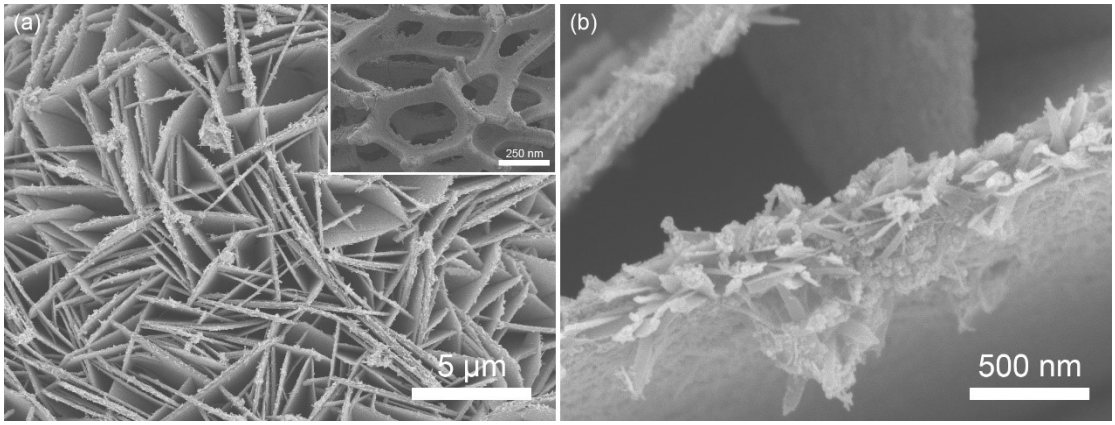

Fig. S2. SEM images of (a, b) $FeNi_2Se_4$-FeNi LDH/NF-90.

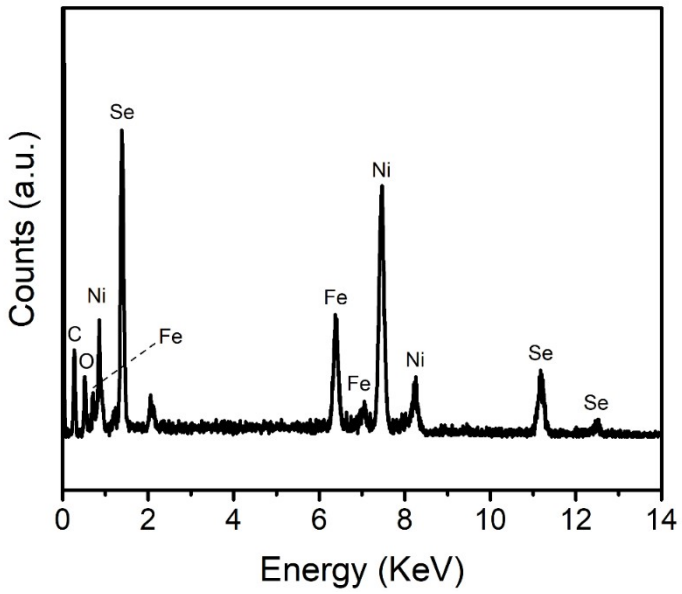

Fig. S3. EDX pattern of $FeNi_2Se_4$-FeNi LDH/NF.

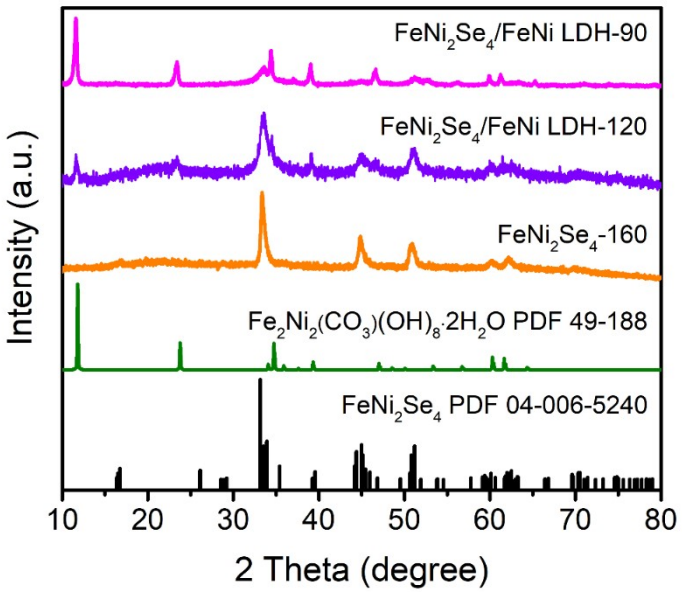

Fig. S4. XRD spectrum of FeNi LDH via different selenization temperatures.

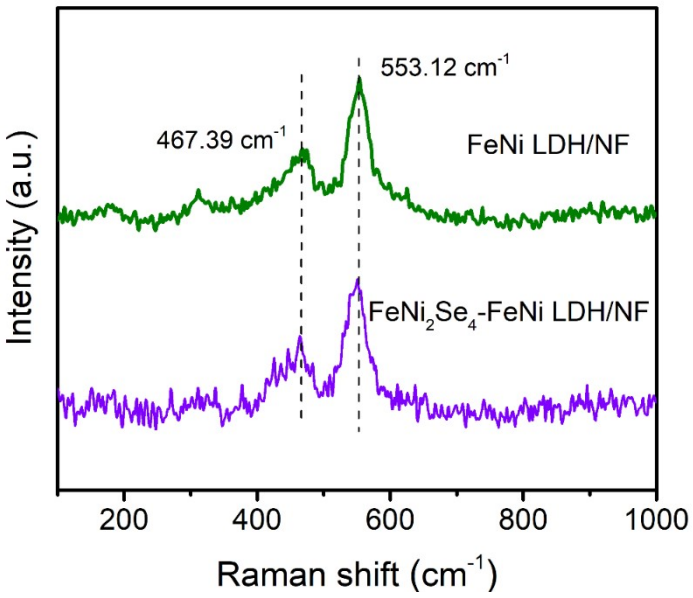

Fig. S5. Raman spectra of FeNi LDH and $FeNi_2Se_4$-FeNi LDH.

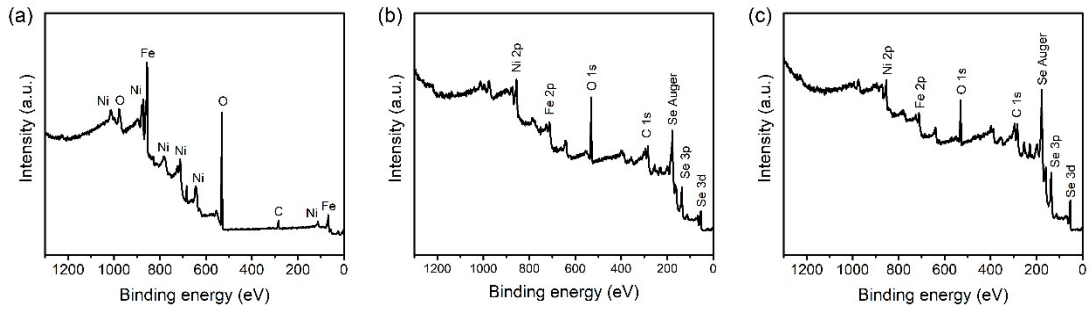

Fig. S6. XPS full spectra of (a) FeNi LDH, (b) $FeNi_2Se_4$-FeNi LDH and (c) $FeNi_2Se_4$.

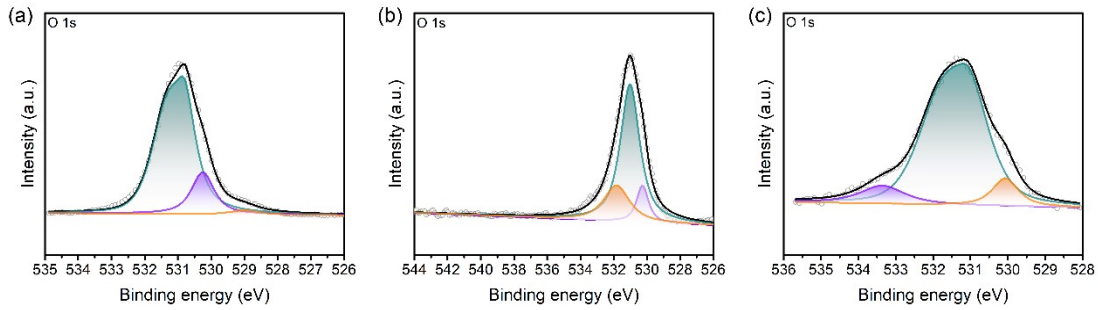

Fig. S7. O 1s of (a) FeNi LDH, (b) $FeNi_2Se_4$-FeNi LDH and (c) $FeNi_2Se_4$.

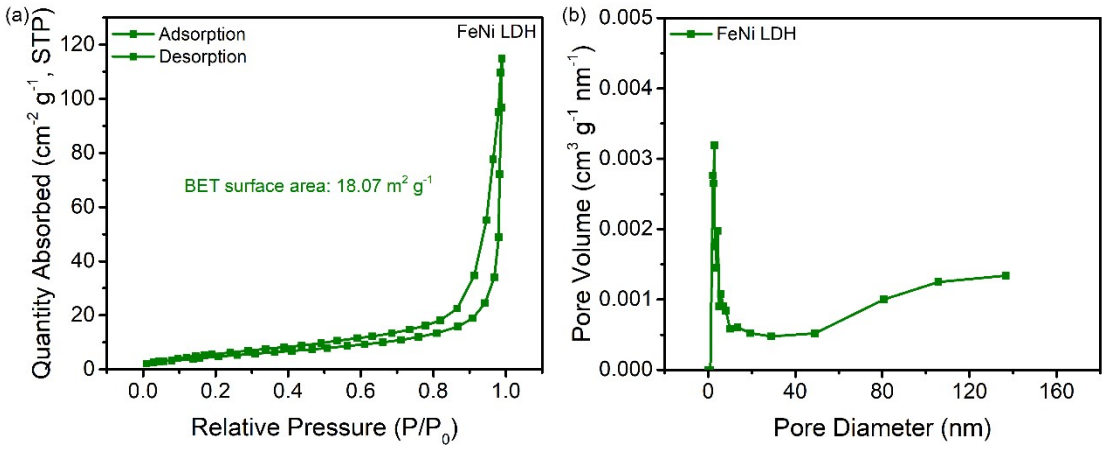

Fig. S8. (a) N_2 adsorption/desorption isotherm and (b) the corresponding pore size distribution of FeNi LDH.

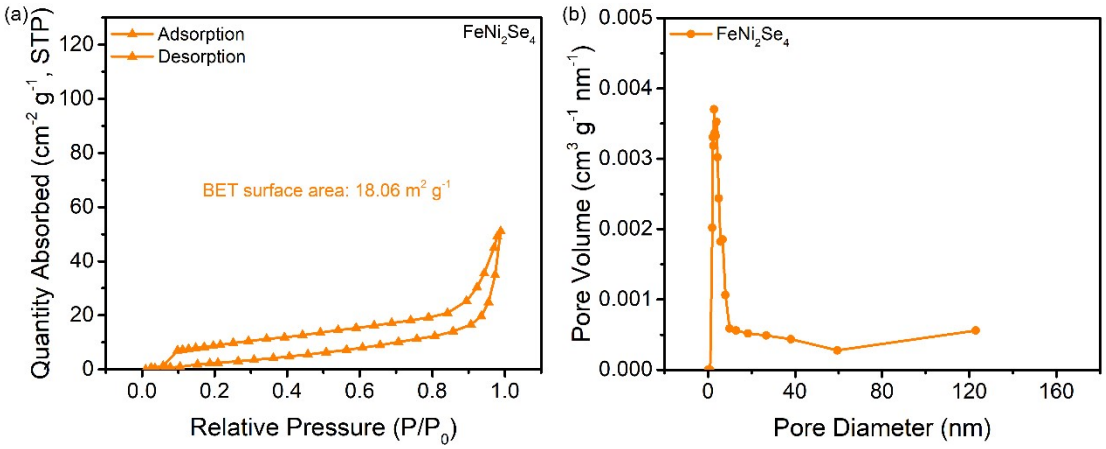

Fig. S9. (a) N_2 adsorption/desorption isotherm and (b) the corresponding pore size distribution of $FeNi_2Se_4$.

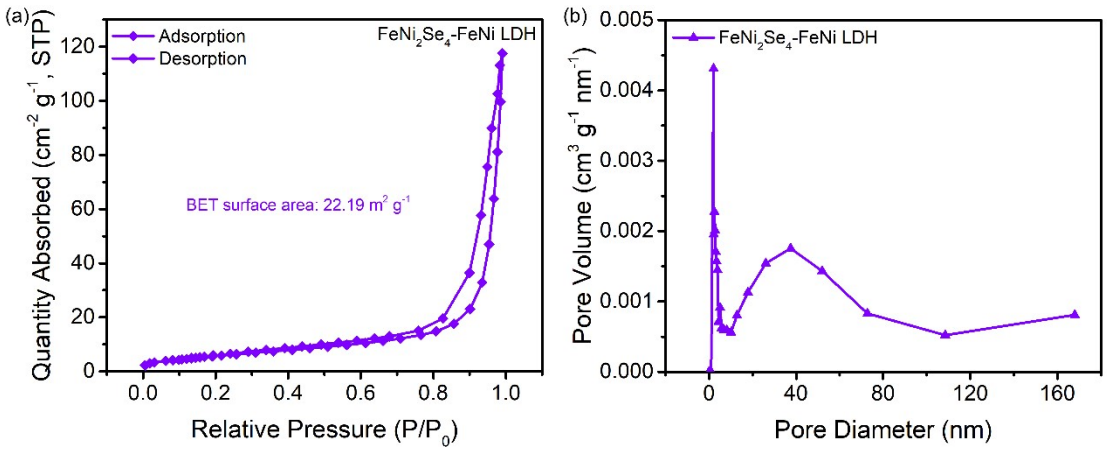

Fig. S10. (a) N_2 adsorption/desorption isotherm and (b) the corresponding pore size distribution of $FeNi_2Se_4$-FeNi LDH.

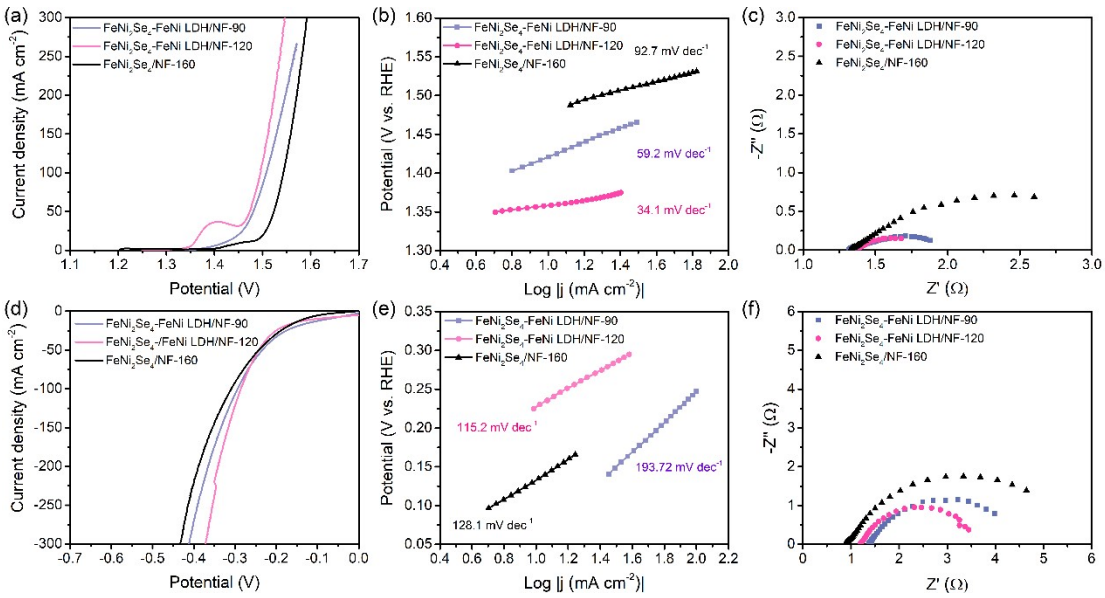

Fig. S11. (a) OER and (d) HER LSV curves of $FeNi_2Se_4$-FeNi LDH/NF-90, $FeNi_2Se_4$-FeNi LDH/NF-120 and $FeNi_2Se_4$/NF-160. (b, e) Tafel plots. (c, f) Nyquist plots.

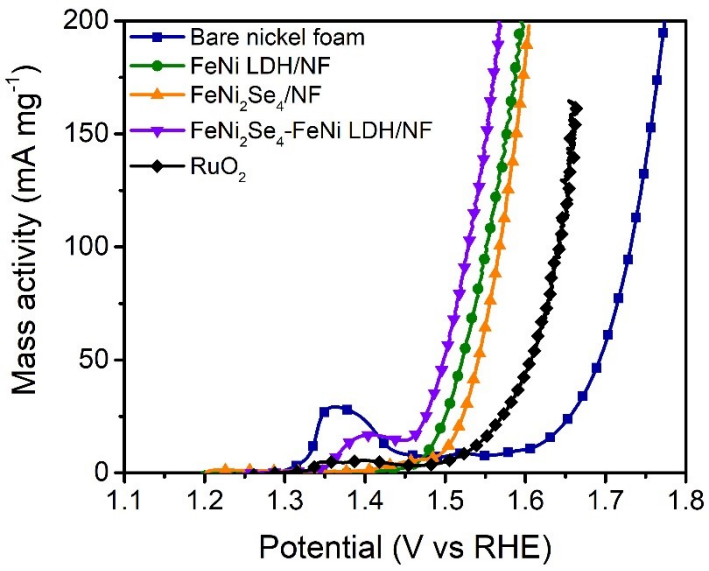

Fig. S12. OER LSV polarization curves normalized to catalyst loading of FeNi LDH, $FeNi_2Se_4$ and $FeNi_2Se_4$-FeNi LDH.

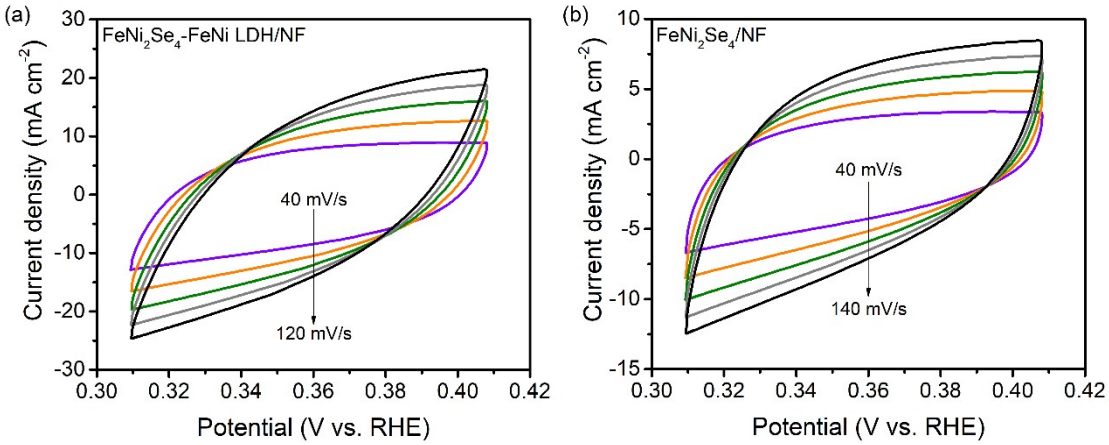

Fig. S13. Cyclic voltammograms of different samples from 20 mV s^{-1} to 140 mV s^{-1} between 0.31 V and 0.41 V.

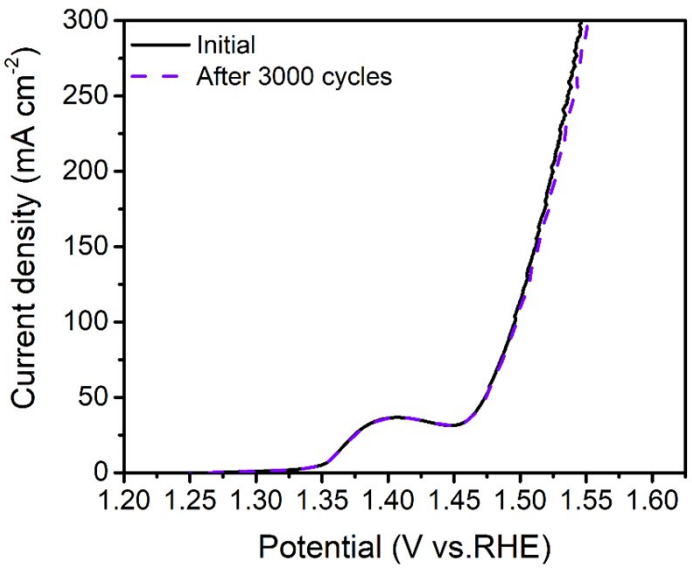

Fig. S14. OER LSV curves of $FeNi_2Se_4$-FeNi LDH/NF before and after 3000 CV scans.

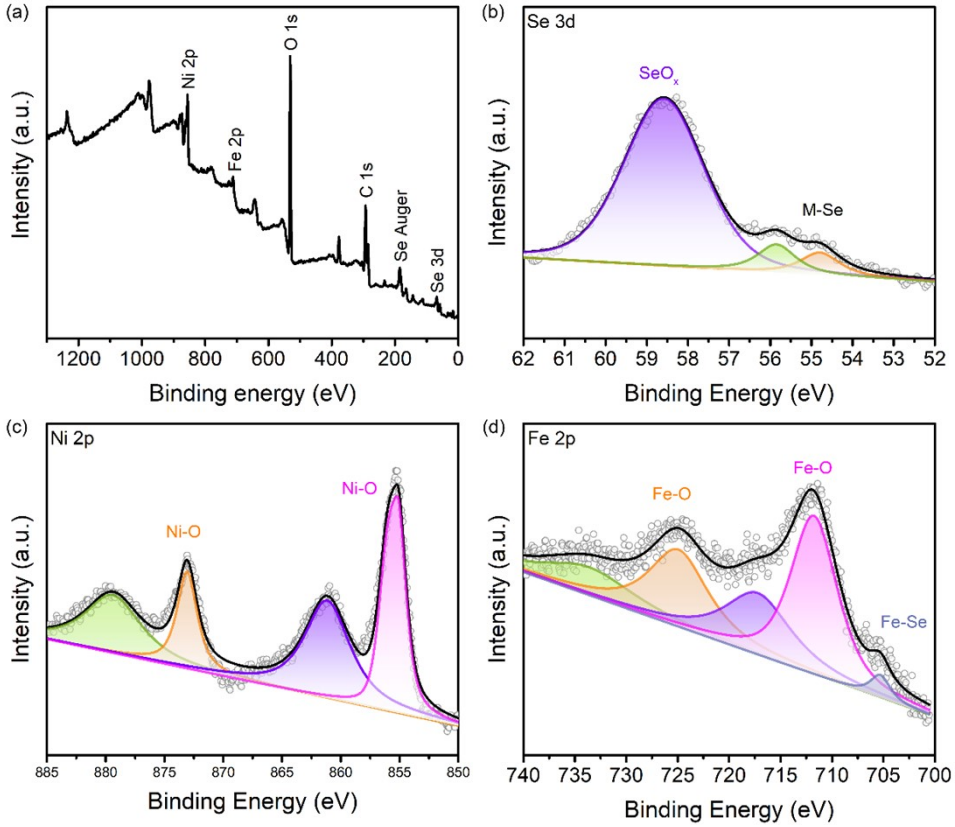

Fig. S15. (a) XPS full spectra of $FeNi_2Se_4$-FeNi LDH/NF after OER test, (b) Se 3d spectra, (c) Ni 2p spectra and (d) Fe 2P spectra after OER test.

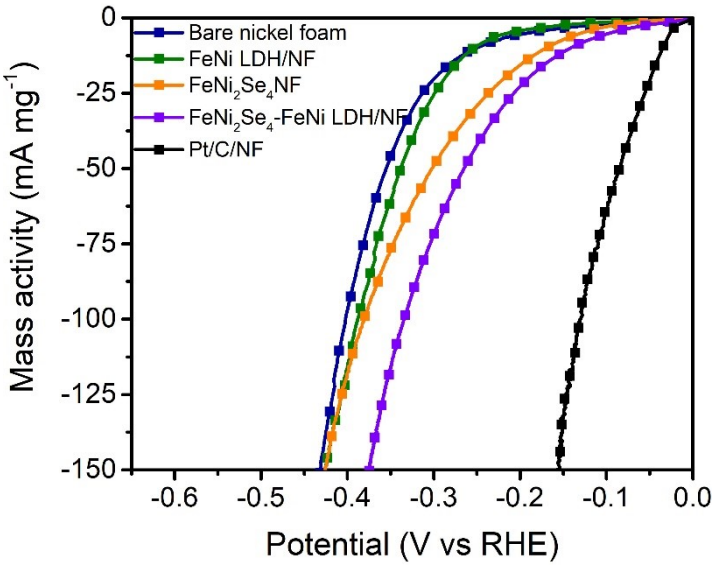

Fig. S16. HER LSV polarization curves normalized to catalyst loading of FeNi LDH, $FeNi_2Se_4$ and $FeNi_2Se_4$-FeNi LDH.

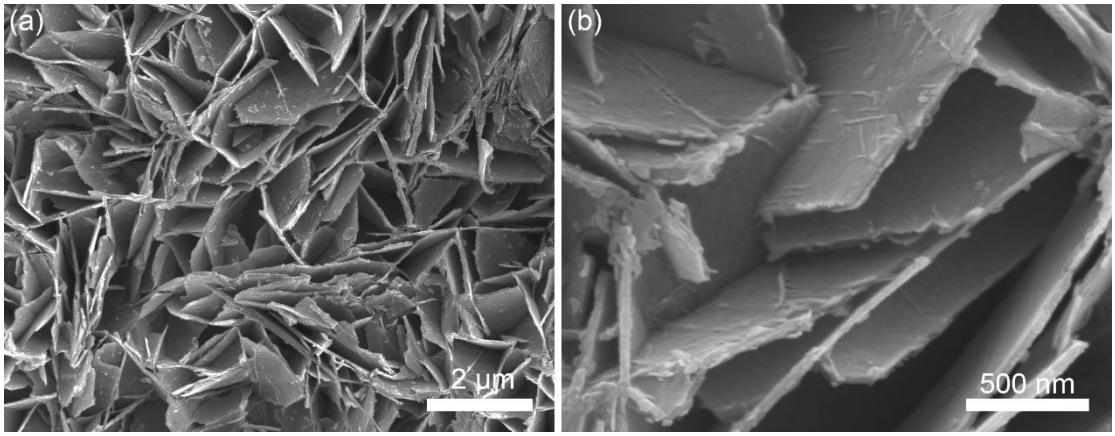

Fig. S17. SEM images of (a, b) $FeNi_2Se_4$-FeNi LDH/NF after the HER test.

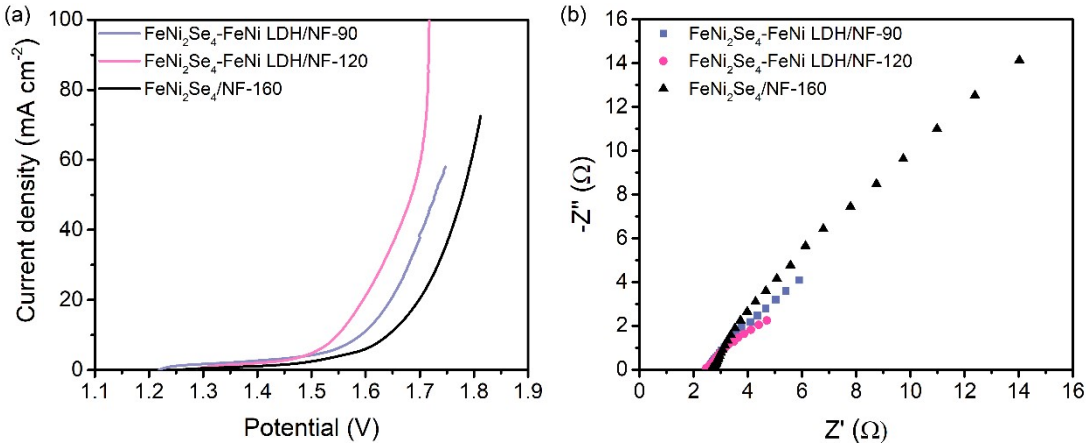

Fig. S18. Overall water splitting performance in 1.0 M KOH. (a) Polarization curves of $FeNi_2Se_4$-FeNi LDH/NF-90, $FeNi_2Se_4$-FeNi LDH/NF-120 and $FeNi_2Se_4$/NF-160 with iR correction. (b) Nyquist plots.

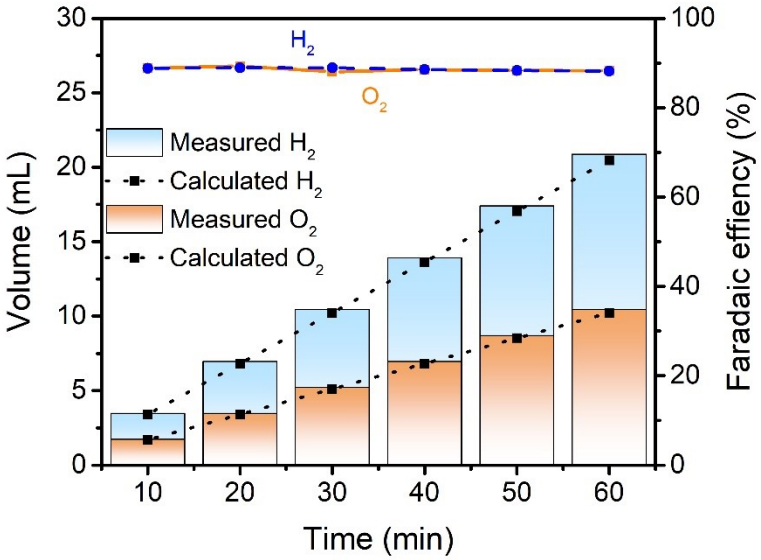

Fig. S19. Amount of hydrogen theoretically calculated and experimentally measured and the Faradaic efficiency.

Table S1. Comparison of η_{10} and Tafel slopes for OER between $FeNi_2Se_4$-FeNi LDH/NF and various reported catalysts.

Catalysts	Tafel slope (mV dec^{-1})	Overpotential (mV)	Reference
$FeNi_2Se_4$-FeNi LDH /NF	30.14	205 @10	This work
$MoS_2/Fe_5Ni_4S_8$/FeNi foam	28.1	204 @10	Adv. Mater. 2018, 30, 1803151
CoV-$Fe_{0.28}$	39.1	215 @10	Adv. Energy Mater. 2020, 10, 2002215
FeNi LDH/CoP/CC	33.5	225 @10	Angew. Chem. Int. Ed. 2019, 58,11903-11909
$(Ni_{0.77}Fe_{0.23})Se_2$	69	228 @10	J. Mater. Chem. A 2019, 7, 2831-2837
$H_2PO_2^-$/FeNi-LDH-V_2C	46	250 @10	Applied Catalysis B: Environmental 2021, 297, 120474
CoO-$CoSe_2$@N-CNTs/rGO	68	250 @10	Chemical Engineering Journal 2021, 422, 129982
$Fe_{0.09}Co_{0.13}$-$NiSe_2$	63	251 @10	Adv. Mater. 2018, 30, 1802121
Ni-Fe-$S_{3:1}$-160	40	245 @20	J. Mater. Chem. A 2019, 7, 12350-12357
$Fe_{7.4\%}$-NiSe	43	231 @50	J. Mater. Chem. A 2019, 7, 2233-2241
NiSe-$Ni_{0.85}Se$/CP	98	300 @10	Small 2018, 14, 1800763
FeOOH(Se)	54	287 @10	J. Am. Chem. Soc. 2019, 141, 7005-7013
$NiFe(OH)_x$/FeS		245 @50	Adv. Funct. Mater. 2019, 29, 1902180
MIL-59(FeNi)/Co NSs	38.46	216 @20	Chemical Engineering Journal 2020, 400, 125884

NF/NiSe@Fe_2O_3	36.9	220 @10	Science Bulletin 2021, 66, 52-61
M-CoO/CoFe LDHs	34	254 @10	Small 2018, 14, 1800195
CoFeV LDH/NF	57	242 @10	ACS Sustainable Chem. Eng. 2019, 7, 16828-16834
Ni_3Se_4@NiFe LDH/CFC	55.5	223 @10	Nanoscale Horiz. 2019, 4, 1132-1138
Fe-CoF_2-300	41.9	230 @10	Chemical Engineering Journal 2021, 425, 130686
Co_9S_8@NiFe LDH	52	220 @10	J. Mater. Chem. A 2021, 9, 12244-12254
$NiCo_2S_4$@NiFe LDH	86.4	287 @10	Applied Catalysis B: Environmental 2021, 286, 119869

Table S2. The comparison of water splitting performances of $FeNi_2Se_4$-FeNi LDH/NF and other catalysts in the literature.

Catalysts	Substrate	Voltage (V)	Reference
$FeNi_2Se_4$-FeNi LDH/NF	NF	1.56	This work
FeNi LDH/CoP/CC	CC	1.617	Angew. Chem. Int. Ed. 2019, 58,11903-11909
NiSe-$Ni_{0.85}Se$/CP	CP	1.62	Small 2018, 14, 1800763
$NiCo_2S_4$@NiFe LDH	NF	1.6	Applied Catalysis B: Environmental 2021, 286, 119869
hetero-Ni_3Se_4@NiFe LDH/CFC	CFC	1.54	Nanoscale Horiz. 2019, 4, 1132-1138
Co_9S_8@NiFe LDH	NF	1.63	J. Mater. Chem. A 2021, 9, 12244-12254
CCF LDH-60	Cu foam	1.681	Nano Energy 2017, 41 327-336
FeNi@FeNiB-700	FeNi foam	1.65	J. Mater. Chem. A 2019, 7, 19554-19564
$Fe_{7.4\%}$-NiSe	NF	1.585	J. Mater. Chem. A 2019, 7, 2233-2241
NiFe LDH@NiCoP	NF	1.57	Adv. Funct. Mater. 2018, 1706847

Table S3. OER Fitting results of EIS for FeNi LDH, $FeNi_2Se_4$ and $FeNi_2Se_4$-FeNi LDH.

Catalysts	Rs (ohm)	Rct (ohm)	Error (%)
FeNi LDH	1.275	11.3	0.807
$FeNi_2Se_4$	0.897	5.288	2.586
$FeNi_2Se_4$-FeNi LDH	1.209	2.445	0.816

Table S4. HER Fitting results of EIS for FeNi LDH, $FeNi_2Se_4$ and $FeNi_2Se_4$-FeNi LDH.

Catalysts	Rs (ohm)	Rct (ohm)	Error (%)
FeNi LDH	1.419	7.660	1.113
$FeNi_2Se_4$	1.390	1.818	0.204
$FeNi_2Se_4$-FeNi LDH	1.249	5.668	0.390