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

Supporting Information for

$(Ni,Co)_{0.85}Se$ Nanosheets Array Derived From Layered Double Hydroxide toward Largely Enhanced Overall Water Splitting

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Supported Figures

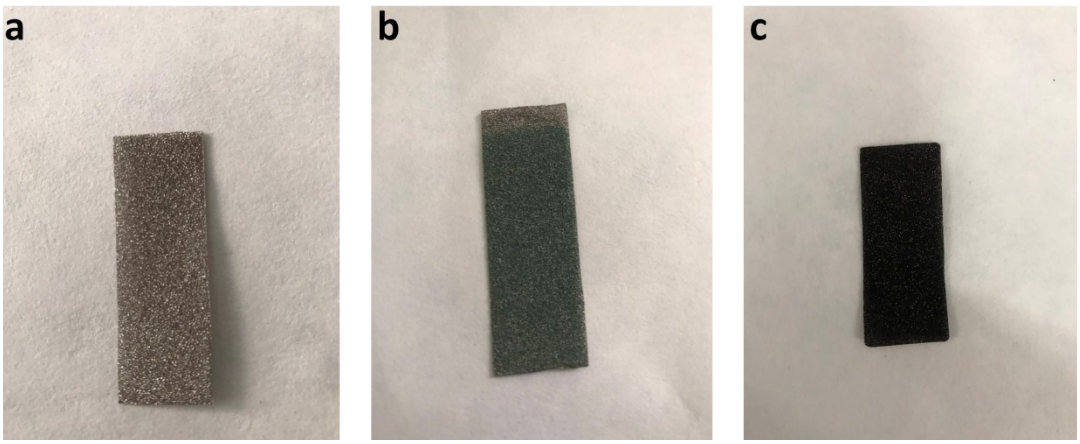

Fig. S1 The photographic images of (a) Ni foam, (b) NiCo-LDH, (c) $(Ni,Co)_{0.85}Se$ NSAs.

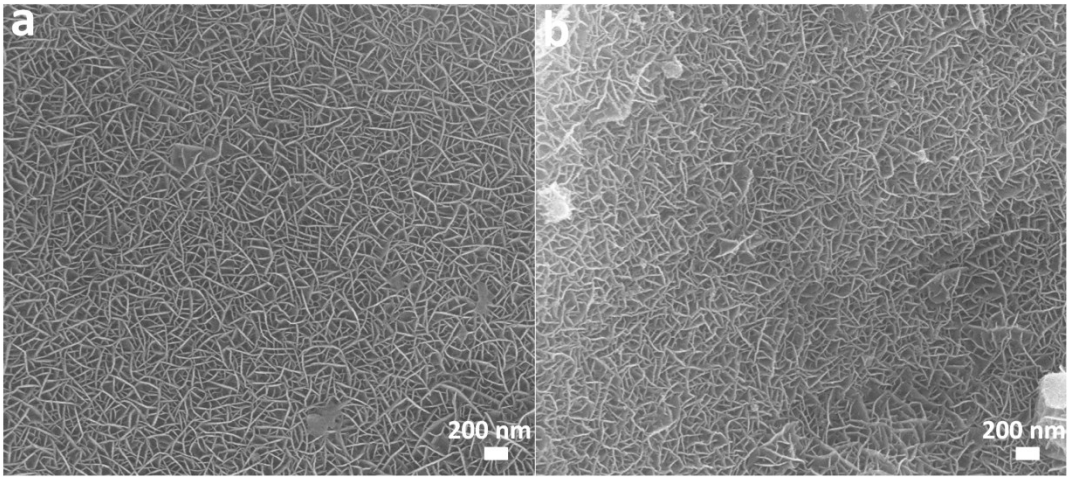

Fig. S2 Low magnification SEM images of (a) NiCo-LDH and (b) $(Ni,Co)_{0.85}Se$ NSAs.

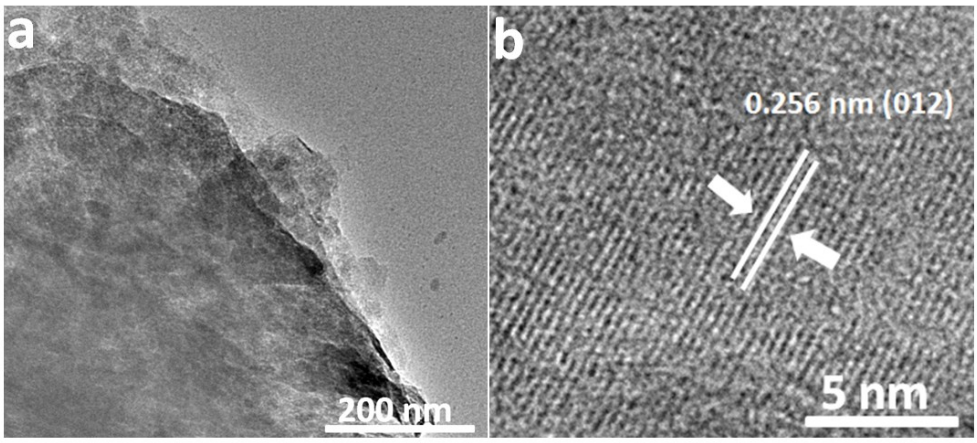

Fig. S3 (a) TEM and (b) HRTEM images of NiCo-LDH.

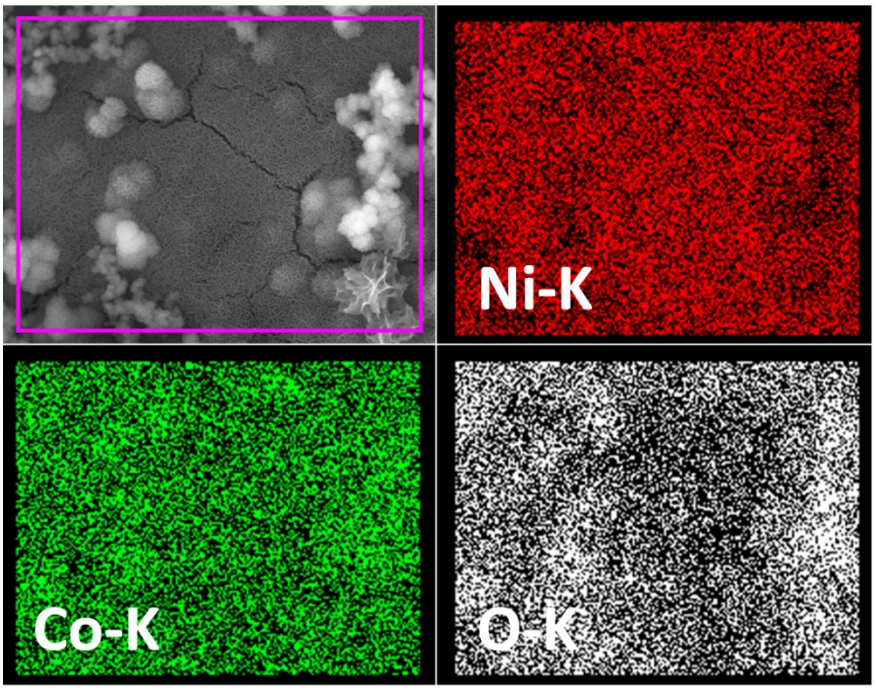

Fig. S4 EDS elemental mapping images of NiCo-LDH.

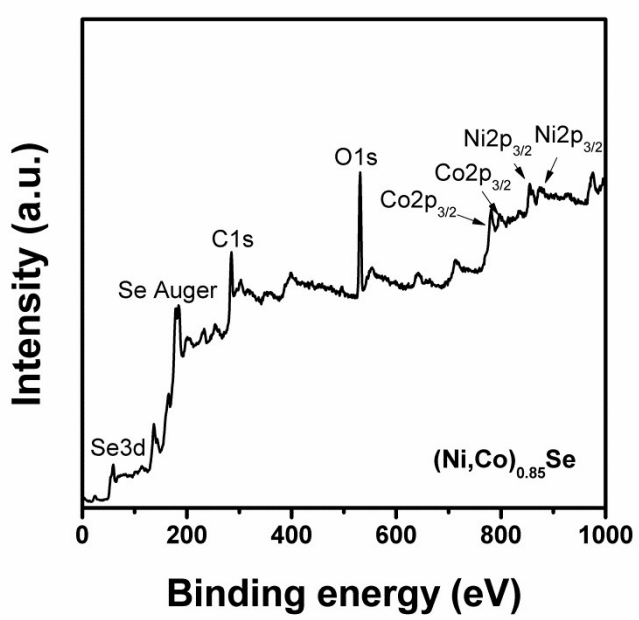

Fig. S5 XPS survey spectrum of $(Ni,Co)_{0.85}Se$ NSAs.

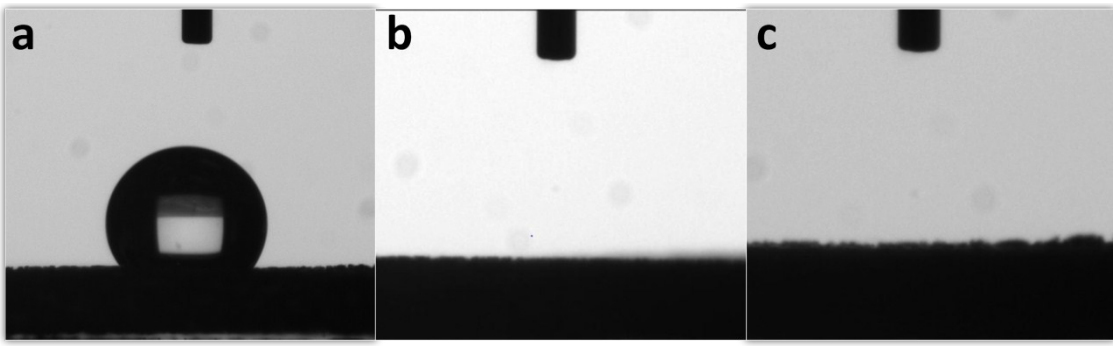

Fig. S6. Contact angel tests over (a) Ni foam, (b) NiCo-LDH and (c) $(Ni,Co)_{0.85}Se$ NSAs.

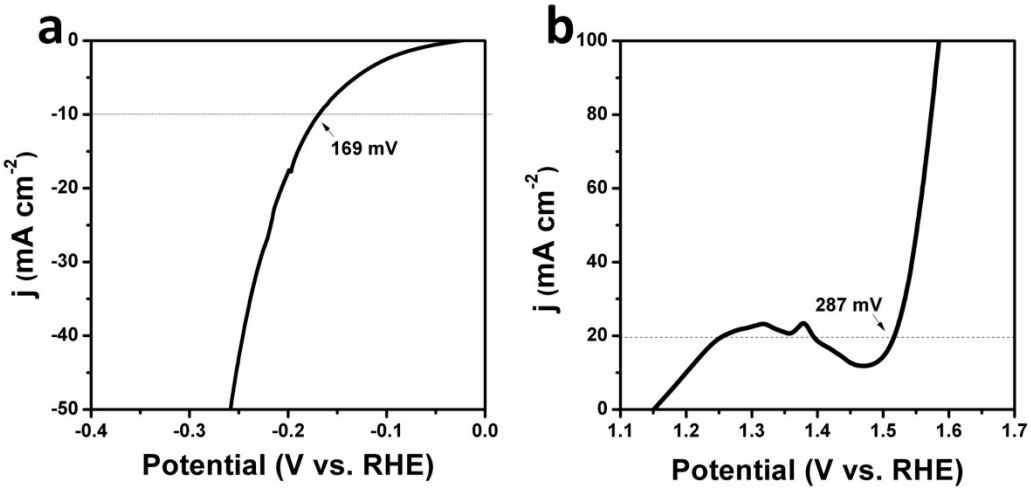

Fig. S7 Magnified polarization curves of $(Ni,Co)_{0.85}Se$ NSAs for (a) HER and (b) OER.

Table S1 Comparison of the HER activities of the $(Ni,Co)_{0.85}Se$ and other recently reported high-performance HER catalysts

Catalysis	Current density ($mA\ cm^{-2}$)	Overpotential (mV vs RHE)	Reference
$(Ni,Co)_{0.85}Se$ NSAs	10	169	This work
Electrosynthesis of NiCo-LDH	10	210	This work
$Ni_{0.85}Se$ films	10	200	29
NC-$Co_{0.85}Se$	30	210	30
$Co_{0.85}Se$/NiFe-LDH/graphene	10	260	31
Ni_3Se_2 nanoforest	10	203	32
MoSe-NiSe	10	210	33
$CoSe_2$/(NiCo)Se_2	17.5	200	34

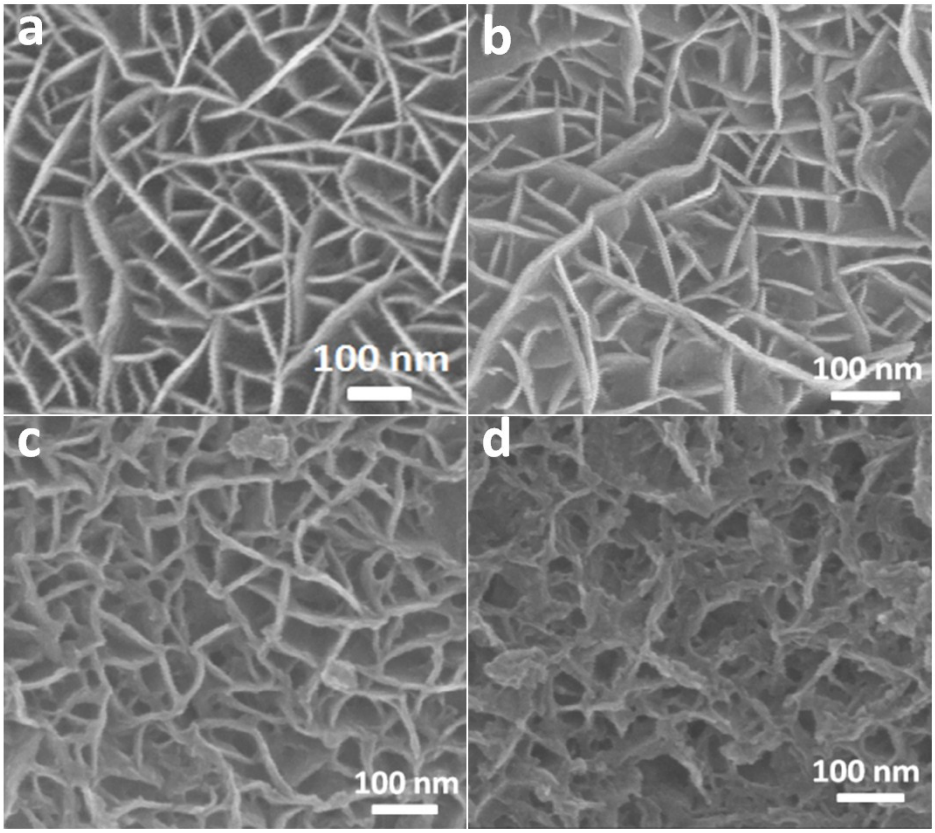

Fig. S8 SEM images of $(Ni,Co)_{0.85}Se$ NSAs with different selenylation time: (a) 1 h; (b) 3 h; (c) 8 h; (d) 24 h.

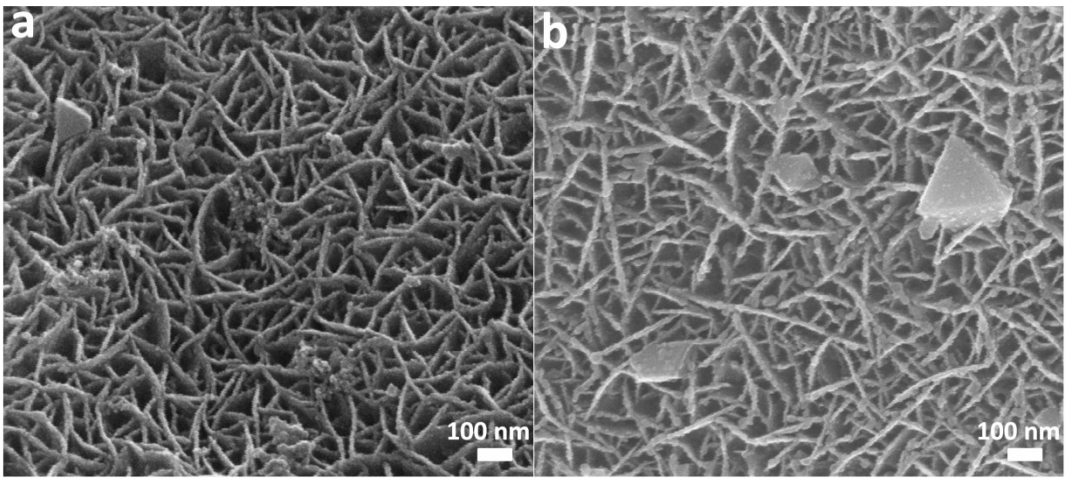

Fig. S9 SEM images of the $(Ni,Co)_{0.85}Se$ NSAs with (a) post-HER and (b) post-OER tests.

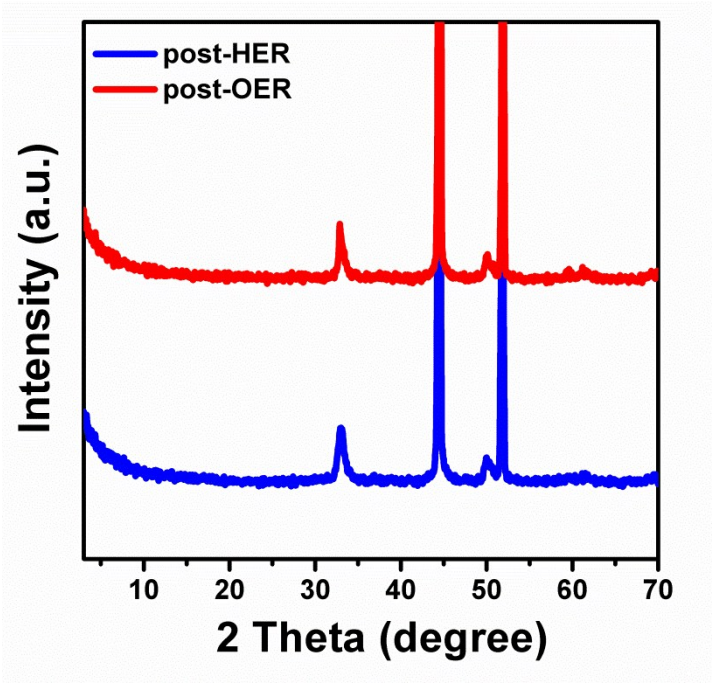

Fig. S10 XRD patterns of the $(Ni,Co)_{0.85}Se$ NSAs with (a) post-HER and (b) post-OER tests.

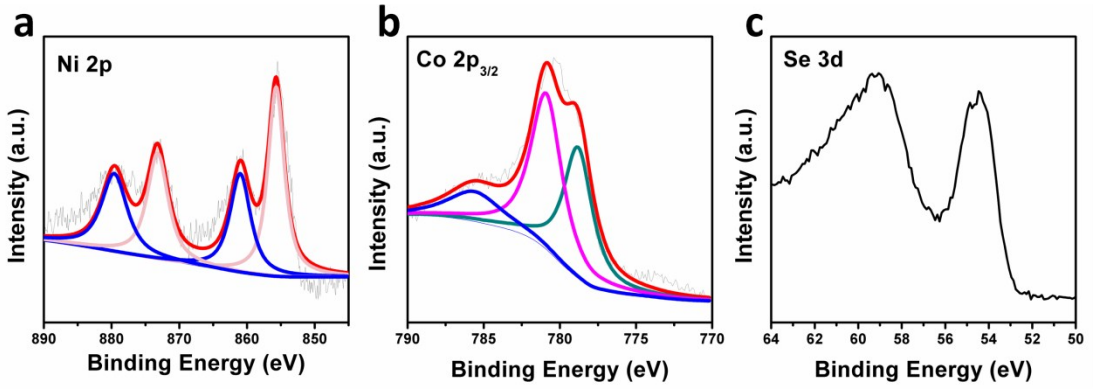

Fig. S11 High-resolution XPS spectra of post-HER $(Ni,Co)_{0.85}Se$ NSAs samples for (a) Ni 2p, (b) Co 2p and (c) Se 3d.

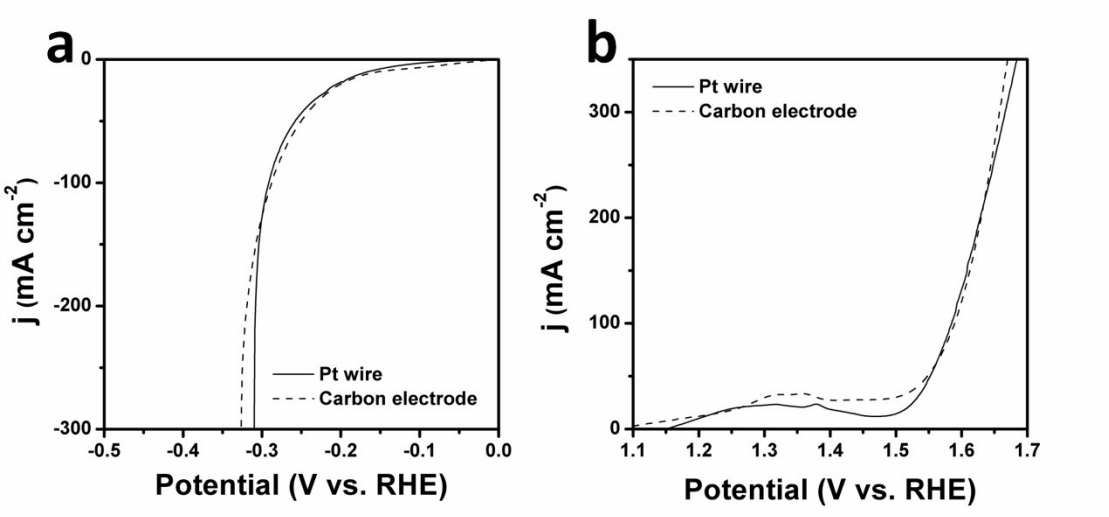

Fig. S12 Polarization curves of $(Ni,Co)_{0.85}Se$ with Pt wire and carbon electrode as counter electrode for (a) HER and (b) OER.

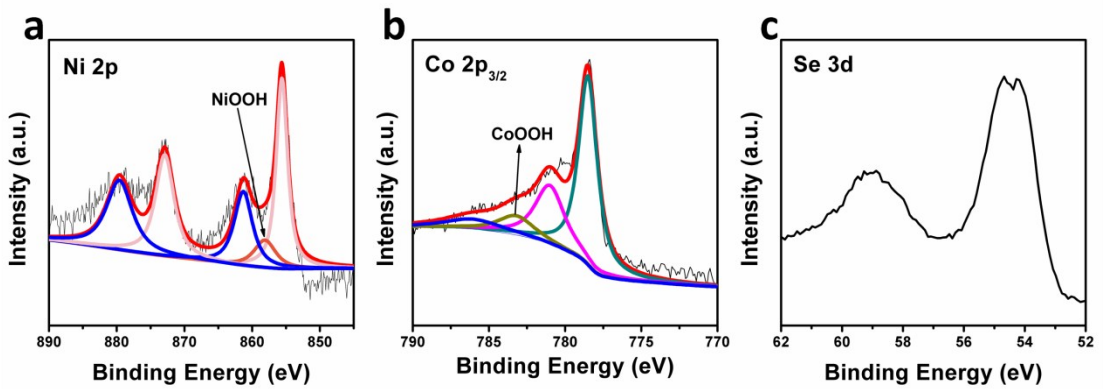

Fig. S13 High-resolution XPS spectra of post-OER $(Ni,Co)_{0.85}Se$ NSAs samples for (a) Ni 2p, (b) Co 2p and (c) Se 3d.

Table S2 Comparison of the OER activities of the $(Ni,Co)_{0.85}Se$ and other recently reported high-performance OER catalysts

Catalysis	Current density ($mA\ cm^{-2}$)	Overpotential (mV vs RHE)	References
$(Ni,Co)_{0.85}Se$ NSAs	20	287	This work
Electrosynthesis of NiCo-LDH	20	330	This work
$Co_{0.85}Se$@Ti mesh	29.6	570	37
$Ni_{0.85}Se$ films	10	302	29
$Co_{0.85}Se$@NC	10	320	38
Ni_3Se_2 films	10	310	21
$NiCo_2O_4$ hollow microcuboids	10	290	39
Core-Shell Ni-Co Nanowires	10	302	40

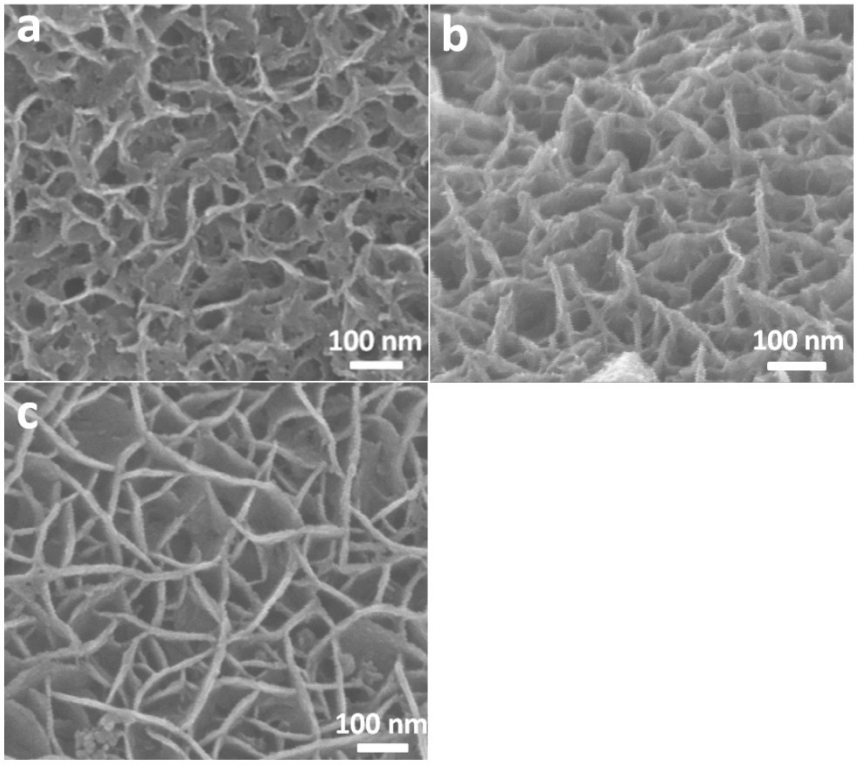

Fig. S14 SEM images of $(Ni,Co)_{0.85}Se$ NSAs with different feeding ratio: (a) 2:1; (b) 1:2; (c) 1:1.

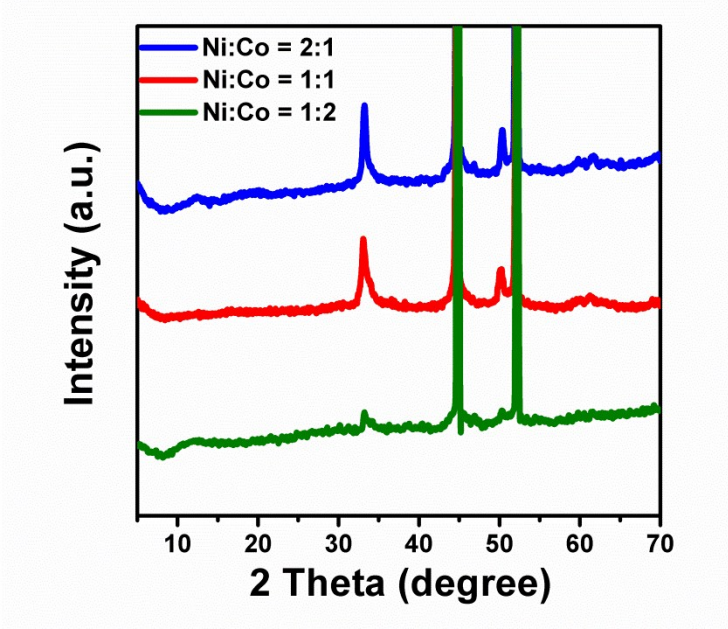

Fig. S15 XRD patterns of $(Ni,Co)_{0.85}Se$ NSAs with different feeding ratio.

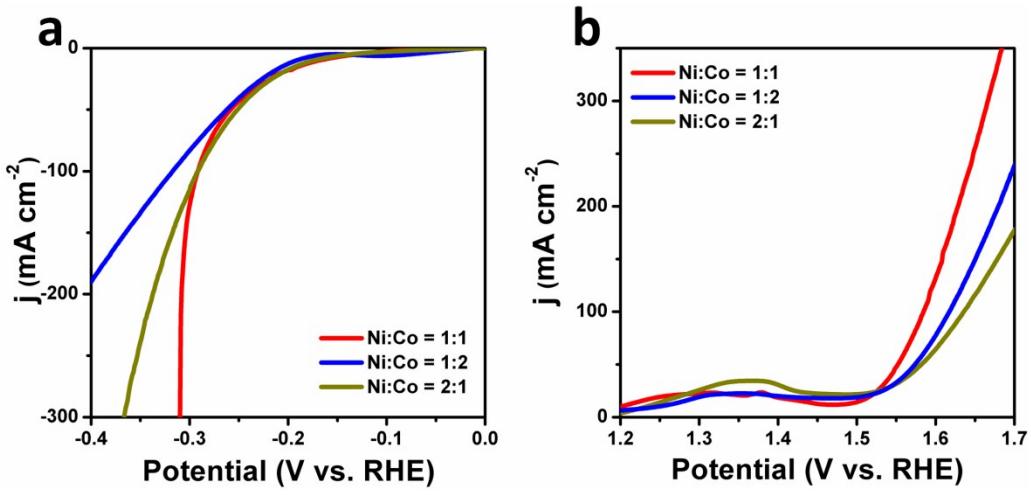

Fig. S16 Polarization curves of $(Ni,Co)_{0.85}Se$ NSAs with different feeding ratio for (a) HER and (b) OER.

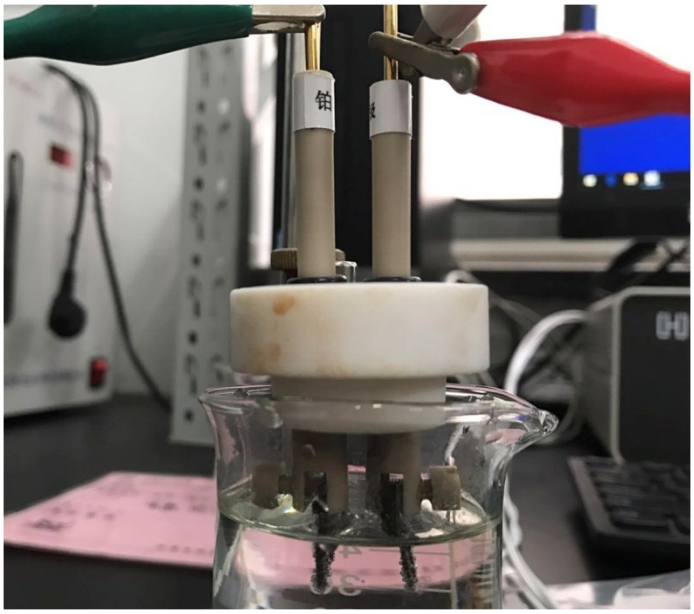

Fig. S17 The scene of two-electrode electrolyzer system for overall water splitting.

Table S3 Comparison of the electrochemical performance and durability of $(Ni,Co)_{0.85}Se$ and other recently reported high performance electrocatalysts for overall water splitting in 1 M KOH

Catalysis	Voltage at 10 mA cm^{-2} (V)	Durability (h)	Reference
$(Ni,Co)_{0.85}Se$ NSAs	1.65	50	This work
Electrosynthesis of NiCo-LDH	1.73	None	This work
$Co_{0.85}Se$/NiFe-LDH/graphene	1.67	10	35
$Co_{0.85}Se$@NC	1.76	10	42
Ni $_{2.3\%}$ -CoS_2	1.66	12	46
Ni_3Se_2@Cu foil	1.65	12	26
$NiCo_2S_4$ NWs	1.68	10	47
Hydrotalcite-like Ni(OH)	1.68	24	48

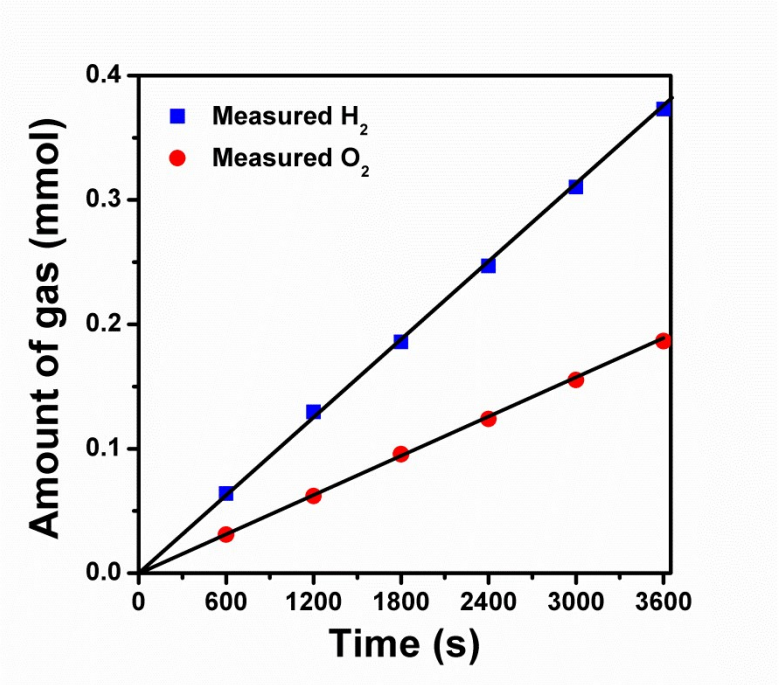

Fig. S18 The amount of gas theoretically calculated and experimentally measured versus time for overall water splitting of $(Ni,Co)_{0.85}Se$.

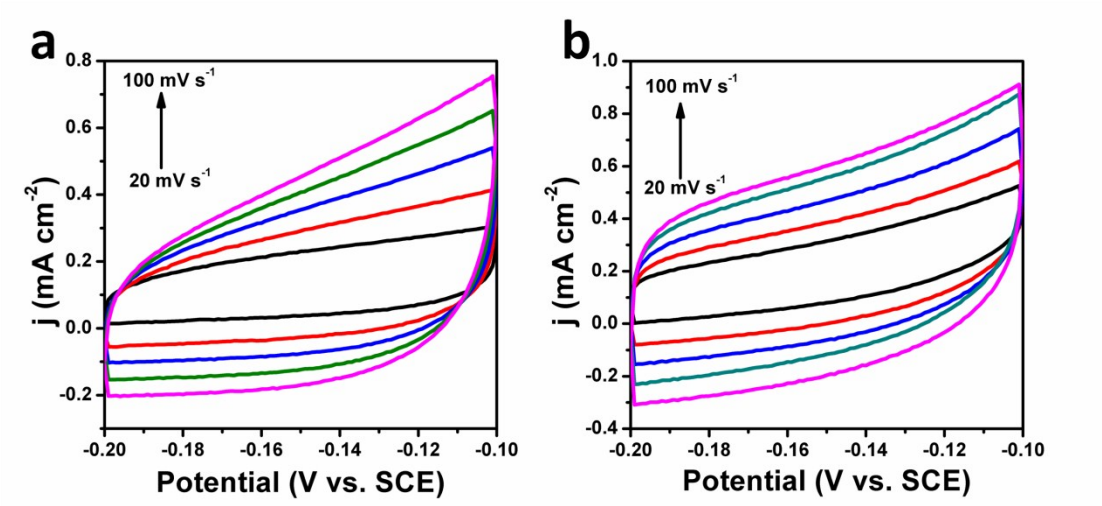

Fig. S19 CV curves at different scan rates for (a) NiCo-LDH and (b) $(Ni,Co)_{0.85}Se$ NSAs.

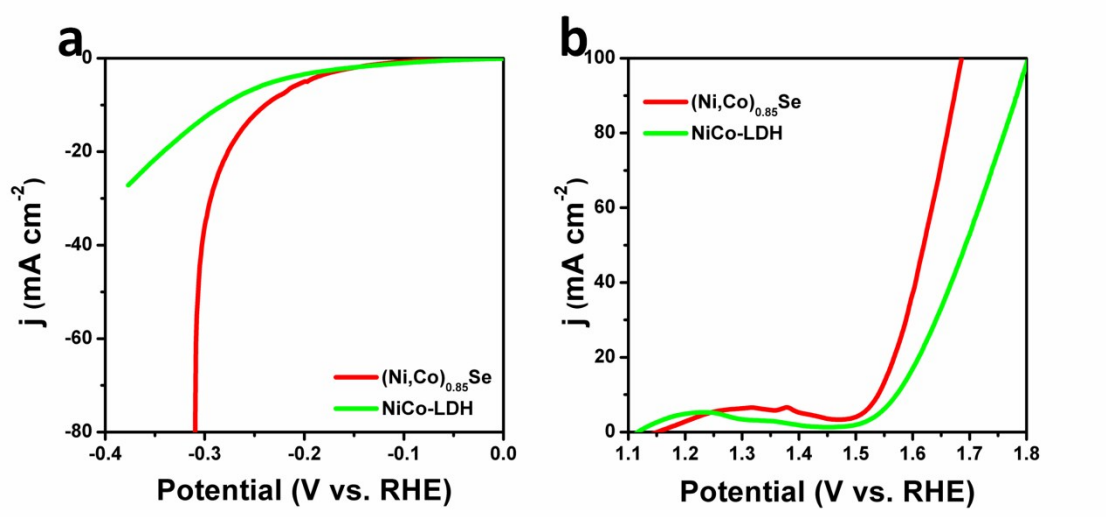

Fig. S20 Polarization curves versus normalized ESCA of $(Ni,Co)_{0.85}Se$ and NiCo-LDH for (a) HER and (b) OER.

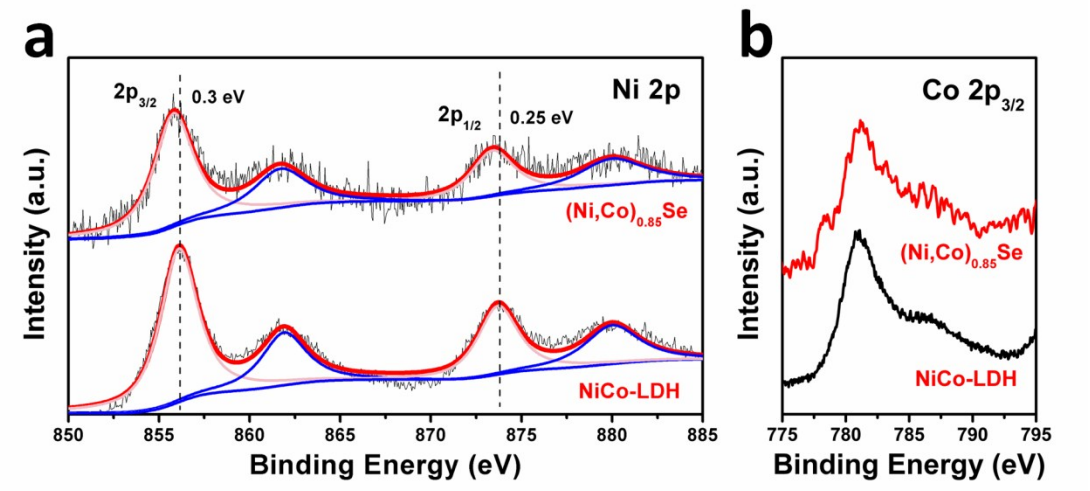

Fig. S21 Ni 2p (a) and Co $2p_{3/2}$ (b) orbital XPS spectra of NiCo-LDH and $(Ni,Co)_{0.85}Se$.

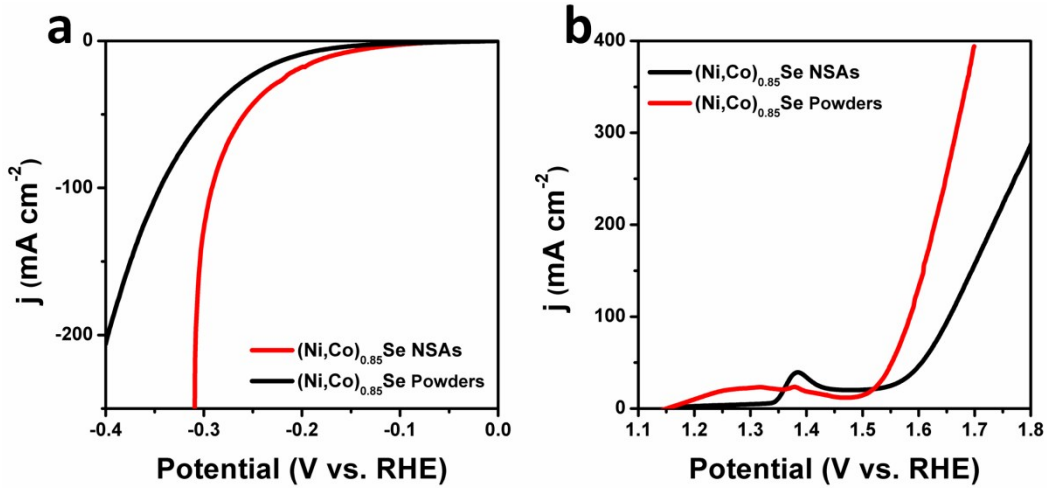

Fig. S22 Polarization curves of $(Ni,Co)_{0.85}Se$ NSAs and $(Ni,Co)_{0.85}Se$ powders for (a) HER and (b) OER.