Supporting Information

Optimized trimetallic selenides heterostructures as high-performance trifunctional electrodes for self-sustained hydrogen production

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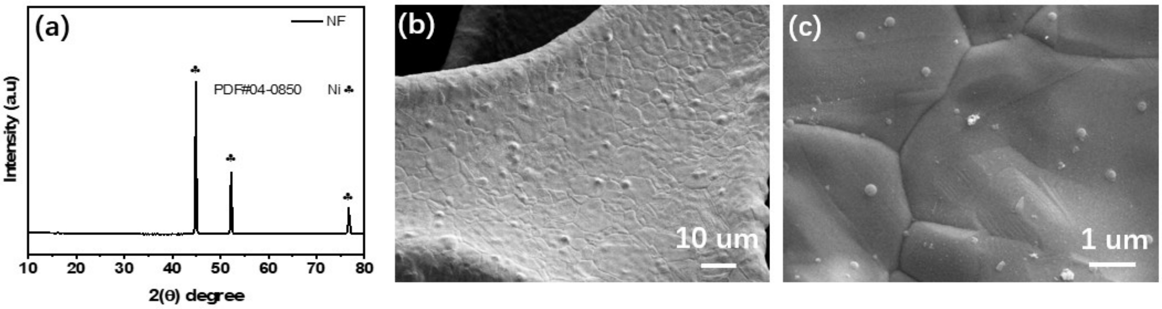

Figure S1. (a) XRD and (b-c) SEM images of pristine NF.

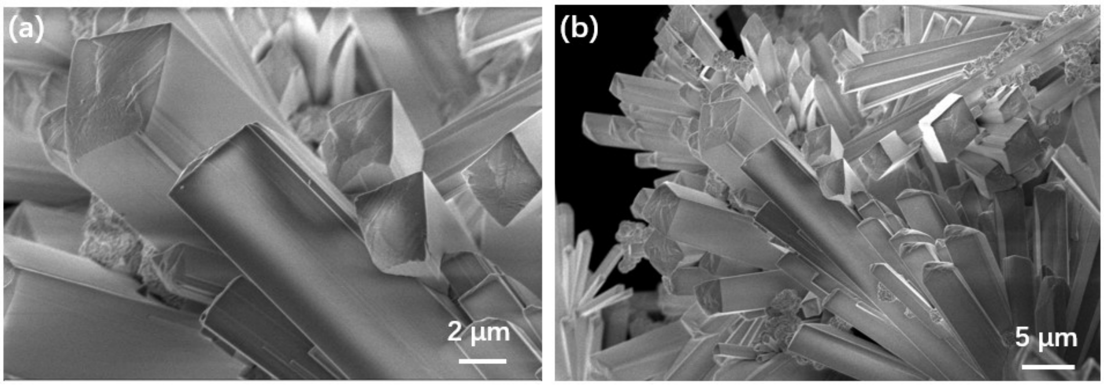

Figure S2. (a-b) SEM images of Co_xMo_y precursors.

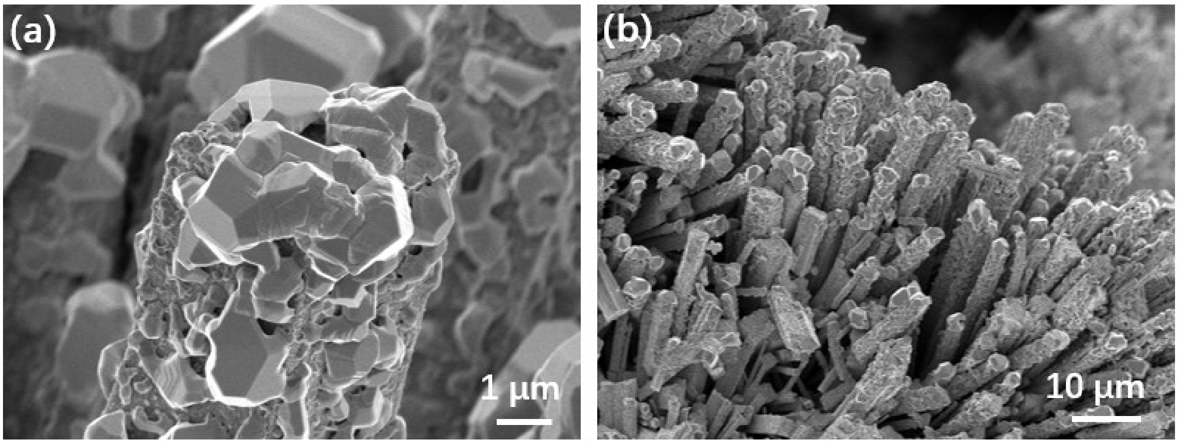

Figure S3. (a-b) SEM images of N-NCM-Se.

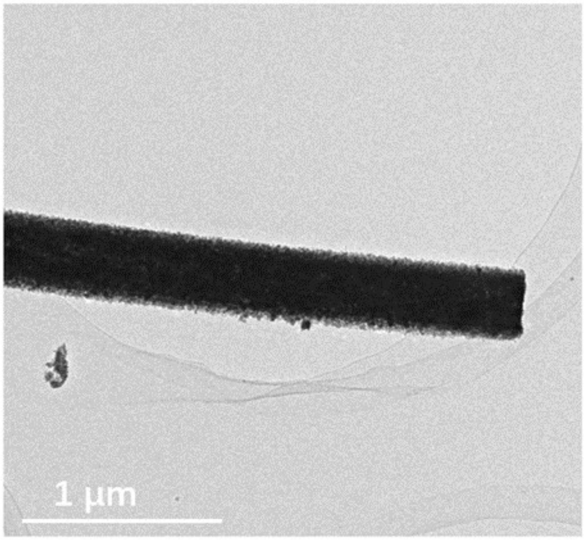

Figure S4. TEM image of N-NCM-Se with selenium particles anchored on the surface.

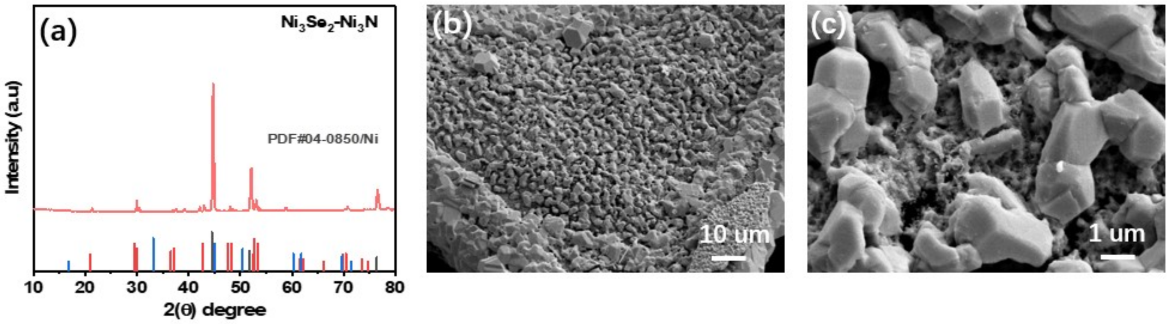

Figure S5. (a) XRD spectrum (b-c) SEM images of Ni_3Se_2-Ni_3N.

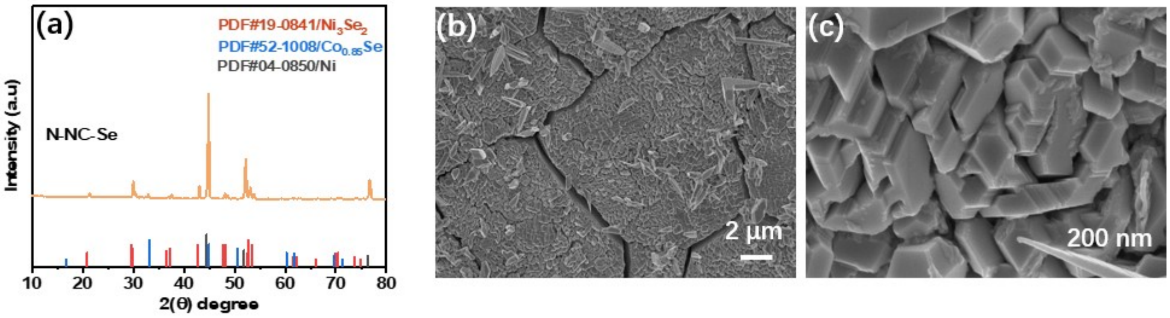

Figure S6. (a) XRD spectrum (b-c) SEM images of N-NC-Se.

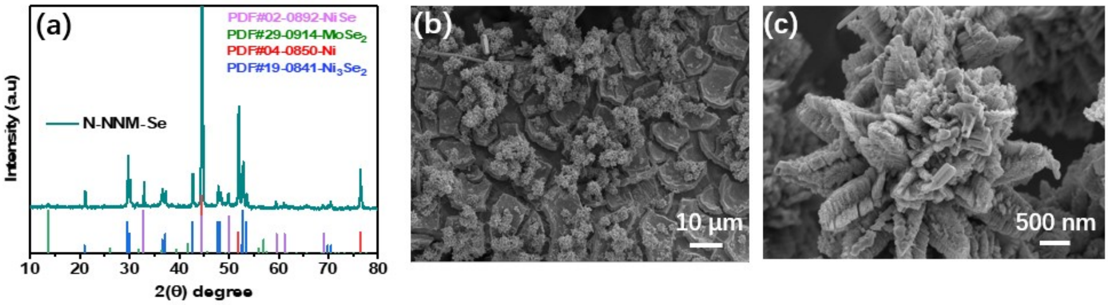

Figure S7. (a) XRD spectrum (b-c) SEM images of N-NNM-Se.

Table S1. HER performance comparison of N-NCM-Se with previous reports on selenides-based electrocatalysts.

S/No.	Electrode	Overpotential (mV) @10 mA cm^{-2}	Tafel slope (mV dec^{-1})	References
1	N-NCM-Se	14.4 mV	66.38	Our work
2	$Co_{0.13}Ni_{0.87}Se_2$/Ti)	64mV	107	Nanoscale, 2016, 8, 3911–3915
3	CoNiSe/NC-3	100 mV	66.5	J. Energy Chem, 34 (2019) 161-170
4	$Co_{0.75}Ni_{0.25}Se$/NF	106mV	74	Nanoscale, 2019, 11, 7959–7966
5	$CoSe_2$@$NiSe_2$	162 mV	43.24	Int. J. Hydrog. Energy, 45 (2020) 30611-30621
6	$NiSe_2$	143mV	49	Nano Energy, (2016) 20, 29–36
7	NiSe/NF	96mV	120	Angew. Chem. Int. Ed, 2015, 54, 9351-9355
8	NiSe/$CoSe_2$	117mV	257.4	J. Alloys Compd 976 (2024) 173092
9	$MoSe_2$–$NiSe_2$–$CoSe_2$/ PNCF	38 mV	38	Nano Energy, 71 (2020) 104637
10	$NiSe_2$/$MoSe_2$@N-BCCSs	119mV	89	Adv. Funct. Mater, 2024, 34, 2314226
11	$Mo_{0.6}$ doped $NiSe_2$	172	115	Electrochim Acta, 475 (2024) 143683
12	5% Mo/Se$NiSe_2$	89mV	57	Int. J. Hydrog. Energy 49 (2024) 25-36
13	Mo_3Se_4-NiSe	84.4mV	59.62	Adv. Mater, 2024, 36, 2305813
14	VSe-$CoSe_2$/$MoSe_2$	74mV	52.4	J. Colloid. Interface Sc, 654 (2024) 1040-1053
15	Ni-$MoSe_x$@$CoSe_2$ CSNs	37mV	68	J. Chem. Eng, 383 (2020) 123129

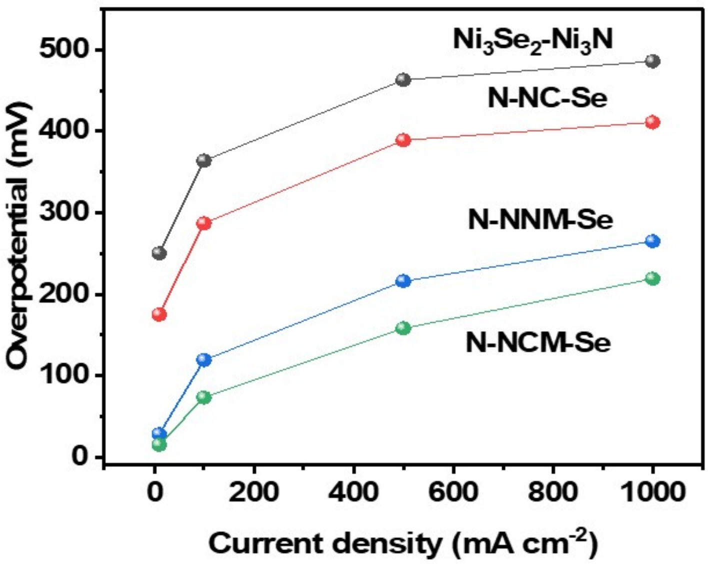

Figure S8. HER overpotential for the as-prepared electrocatalsyts at varying current densities.

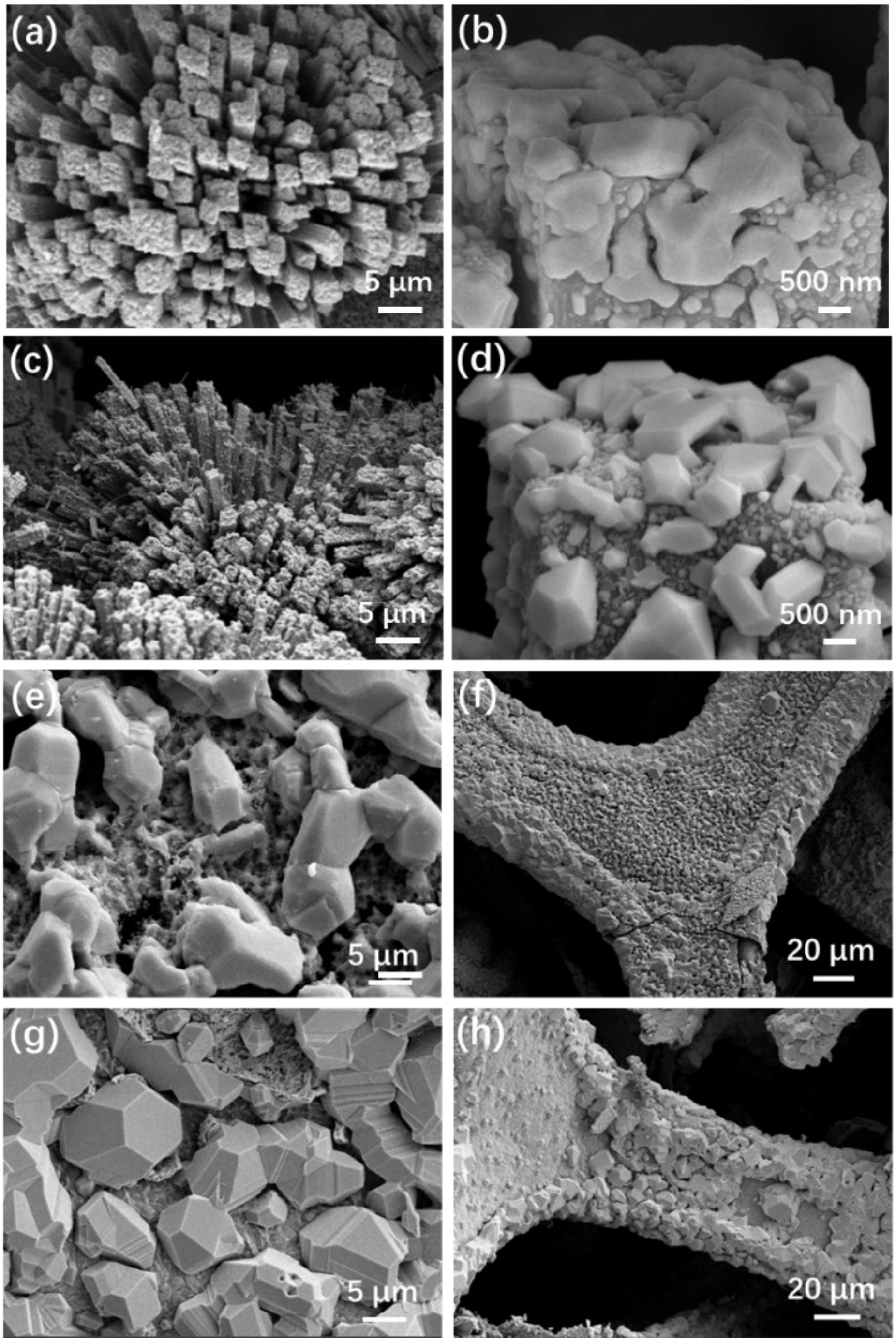

Figure S9. SEM images of (a-b) N-NCM-Se/100 and (c-d) N-NCM-Se/300 derived from selnization of the Co_xMo_y precursors. SEM images of (e-f) Ni_3Se_2-Ni_3N/100 and (g-h) Ni_3Se_2-Ni_3N/300 electrodes derived from direct selenization of the pristine NF.

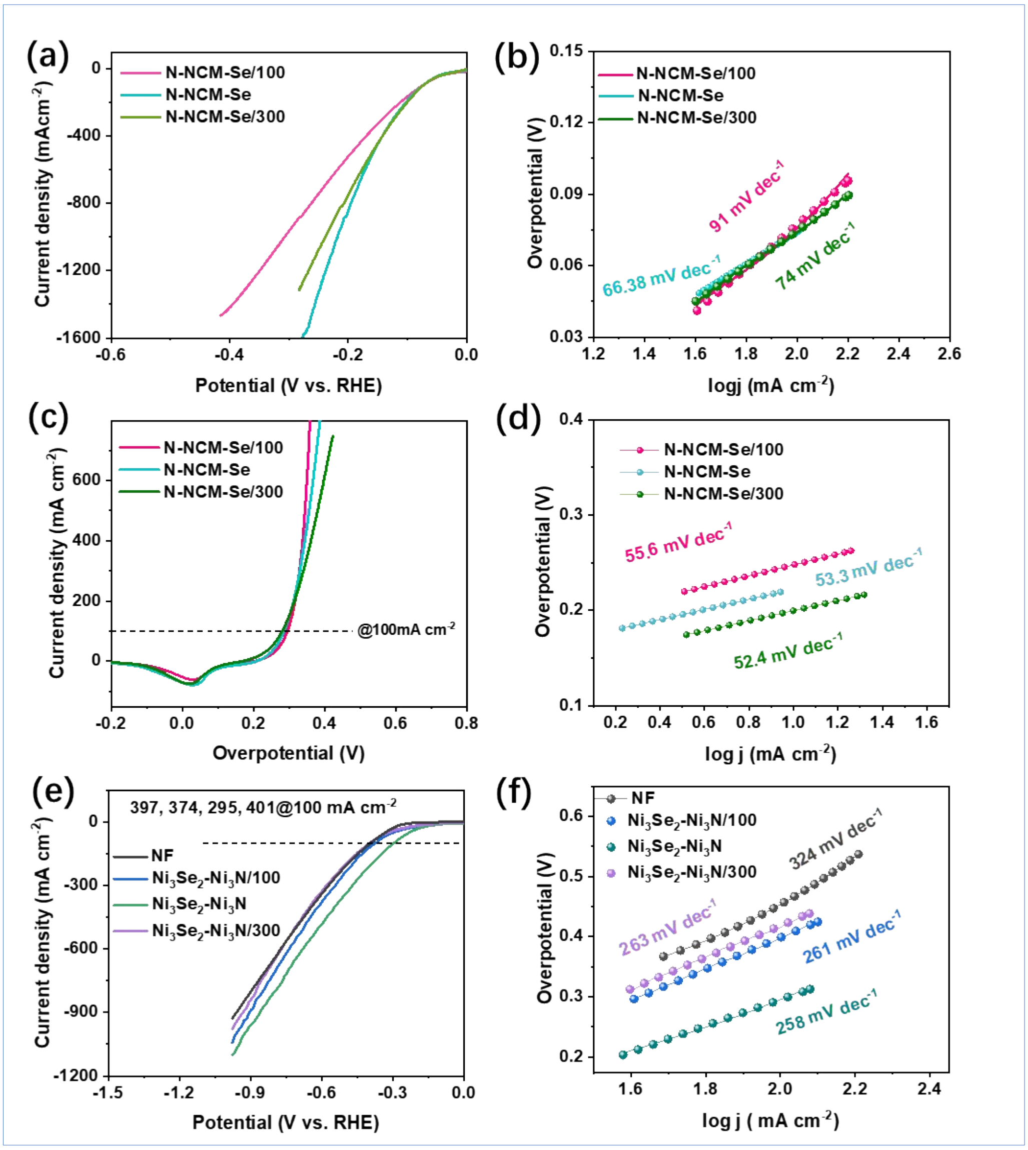

Figure S10. (a) HER LSV curves, (b) HER Tafel slopes, (c) OER LSV curves, and (d) OER Tafel slopes of the N-NCM-Se electrodes with different selenium concentration. (e) HER LSV curves and (f) HER Tafel slopes of Ni_3Se_2-Ni_3N electrodes with different selenium concentration.

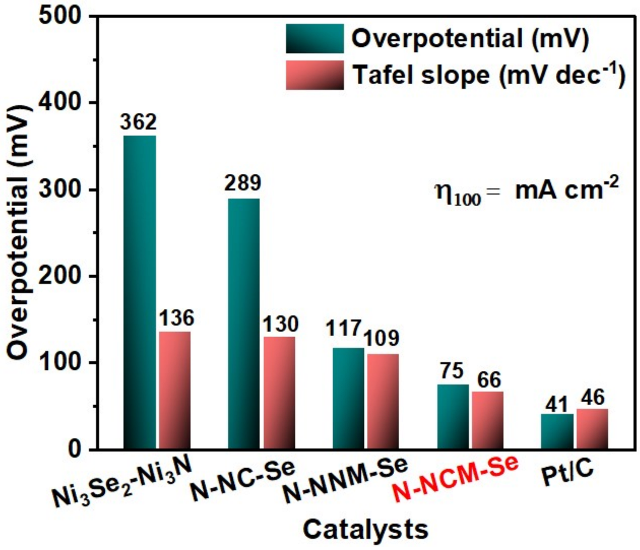

Figure S11. Comparison bar graph of the overpotential and Tafel slopes of our prepared N-NCM-Se with other control samples.

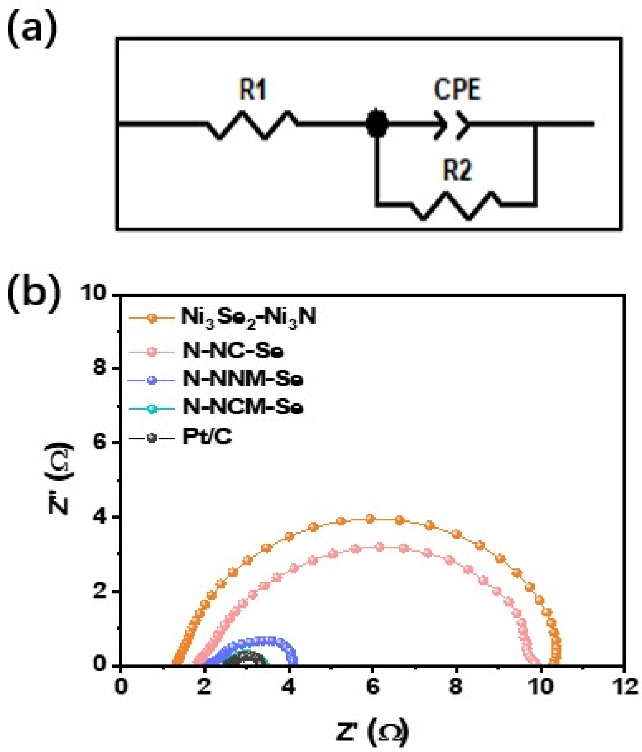

Figure S12. (a) EIS fitted circuit diagram of (a) N-NCM-Se. (b) Nyquist plots of the different electrodes.

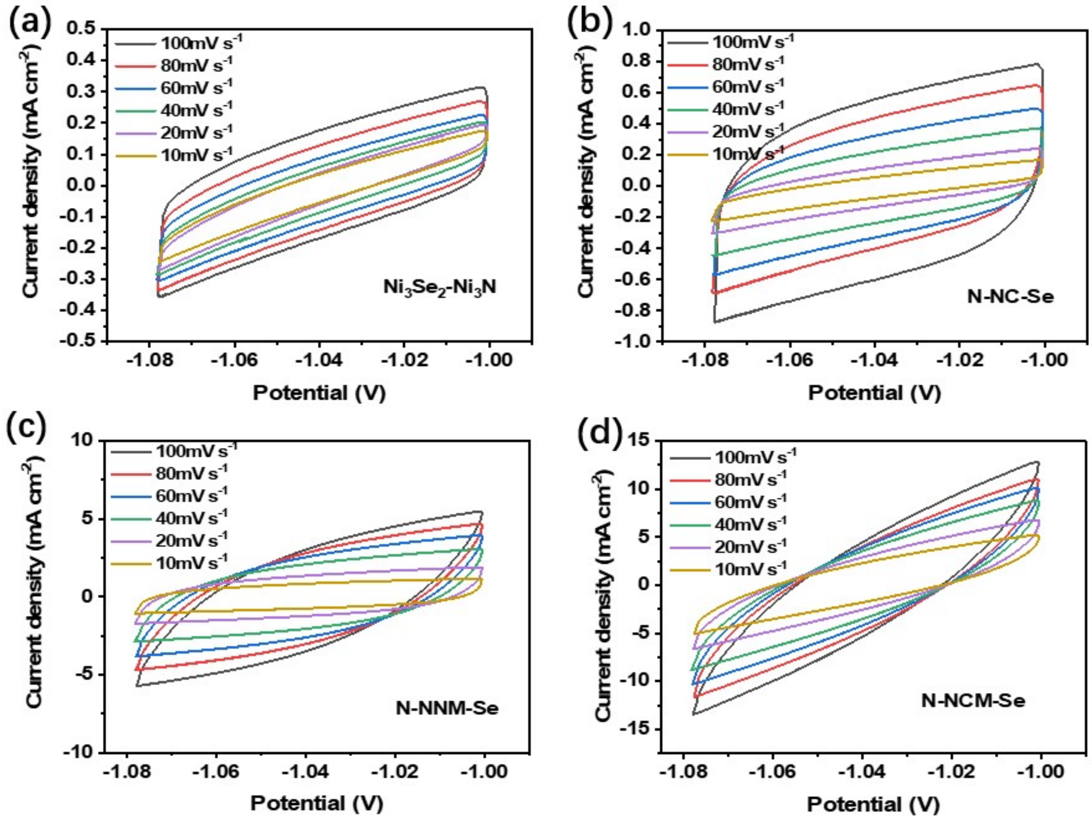

Figure S13. CV curves of (a) Ni_3Se_2-Ni_3N, (b) N-NC-Se, (c) N-NNM-Se, and (d) N-NCM-Se at different scan rates at the non-faradic region for HER.

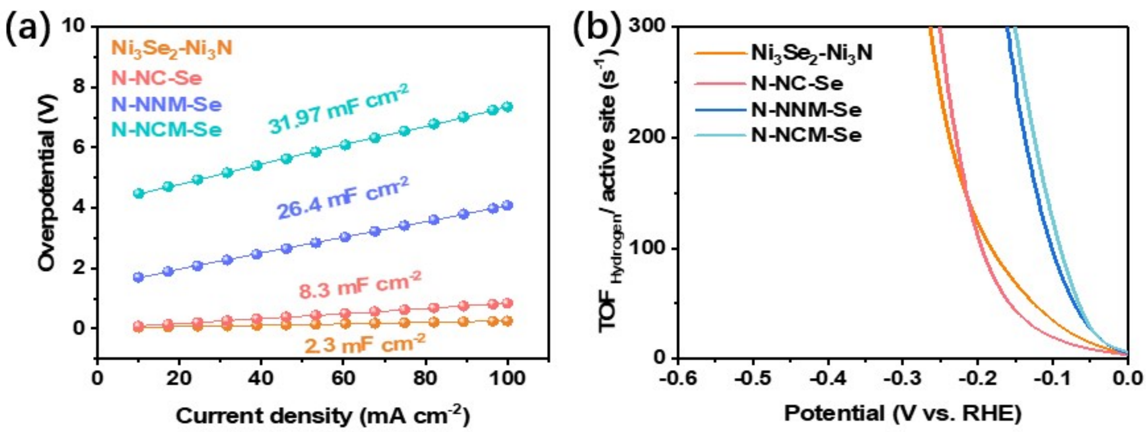

Figure S14. (a) HER C_{dl} values derived from the CV curves in Figure S13, and (b) normalized LSV curves after TOF calculation.

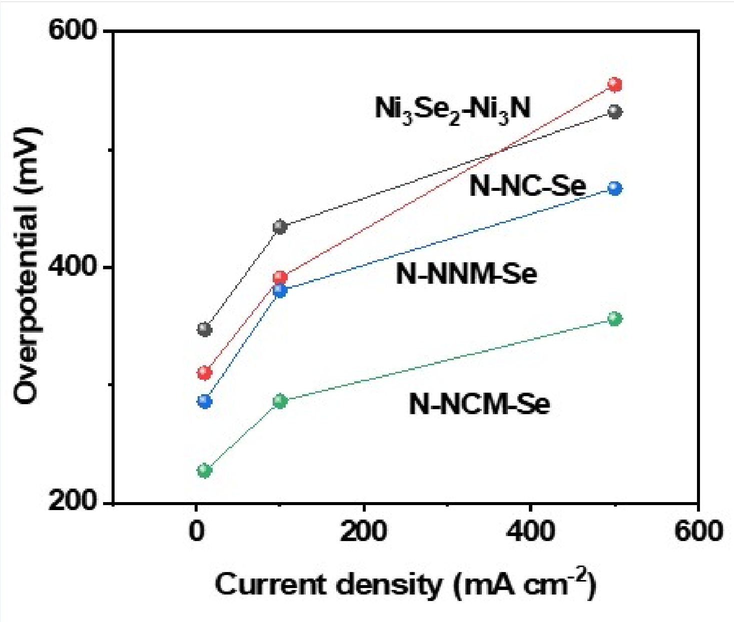

Figure S15. OER overpotentials for the as-prepared electrodes at different current densities.

Table S2. OER performance comparison of N-NCM-Se with previous reports on selenides-based electrocatalysts.

Electrode	Overpotential (mV) @10 mA cm^{-2}	Tafel slope (mV dec^{-1})	References
N-NCM-Se	**226 mV**	**53.33**	**Our work**
$NiSe_2/CoSe_2$	226mV	53	Adv. Mater. 2020, 32, 2000607
$CoSe_2$@pMF-R	378 mV	81.1	Int. J. Hydrog. Energy, 49 (2024) 995-1004
$Ni_{0.27}Co_{0.28}Fe_{0.30}Se$@CNT	240mV	44	J. Mater. Chem. A, 2021, 9, 24261-24267
$Co_{0.21}Ni_{0.25}Cu_{0.54})_3Se_2$	272 mV	54	J. Mater. Chem. A, 2019, 7, 9877-9889
$Fe_{0.48}Co_{0.38}Cu_{0.14})Se$	256mV	40.8	ACS Sustain. Chem. Eng. 2019, 7, 9588-9600
Cu_2Se	270mV	48	ACS Appl. Energy Mater. 2018, 1, 4075-4083
$MnSe_2$@3 wt%	270mV	160	Appl. Surf. Sci, 605 (2022) 154804
$CoMn)Se_2$	274 mV	39	Appl. Catal. B: Environ. 236 (2018) 569-575
Ni_3Se_2	290mV	89	Energy Environ. Sci, 2016, 9, 1771-1782
$Zn_{0.1}Co_{0.9}Se_2$	340mV	43.2	Electrochim Acta 349 (2020) 136336
MnSe@MWCNT	290mV	54	J. Mater. Chem. A, 2022, 10, 6772-678

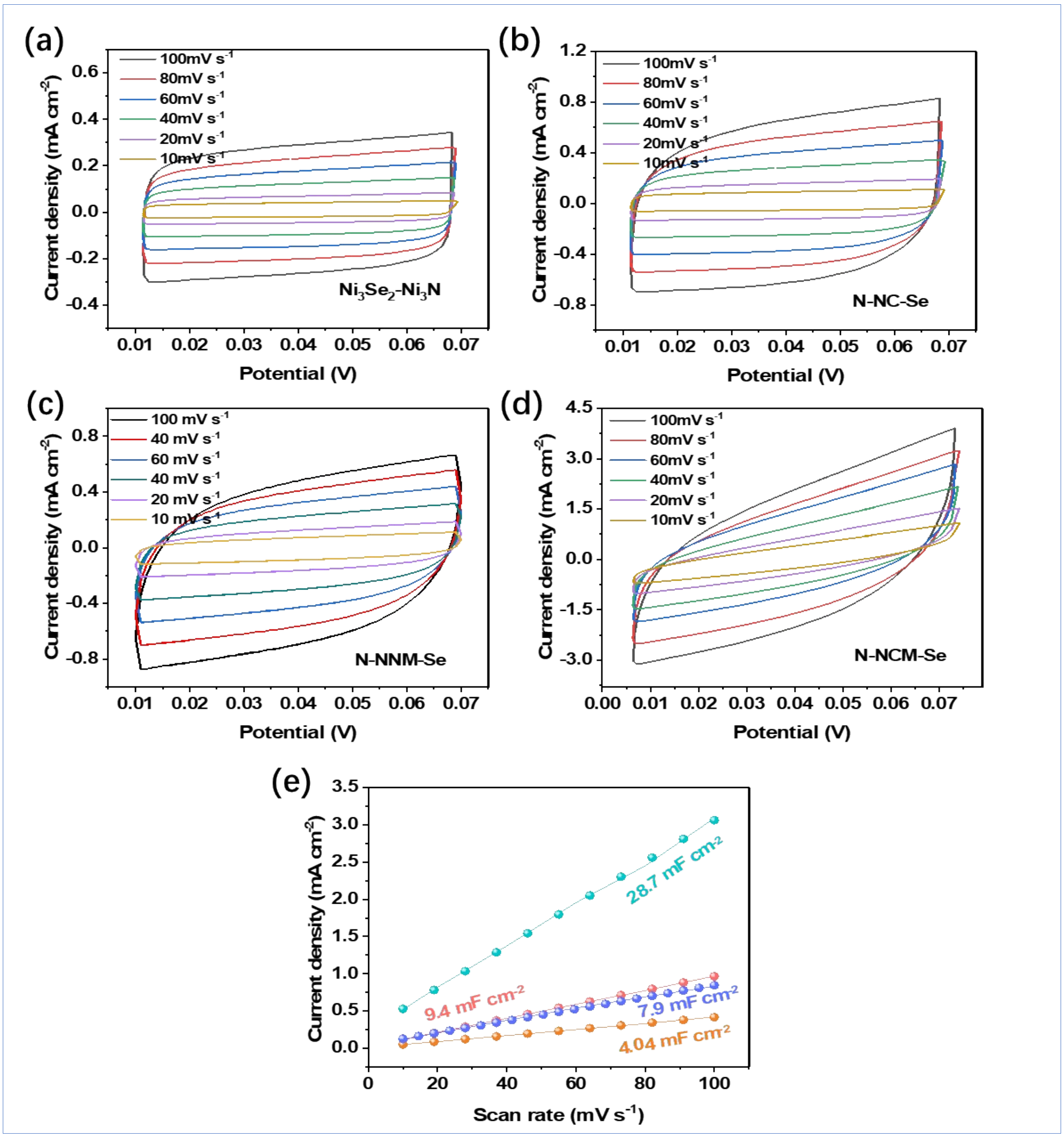

Figure S16. CV curves of (a) Ni_3Se_2-Ni_3N, (b) N-NC-Se, (c) N-NNM-Se, and (d) N-NCM-Se at different scan rates at the non-faradic region for OER. (e) The Corresponding C_{dl} values for different samples.

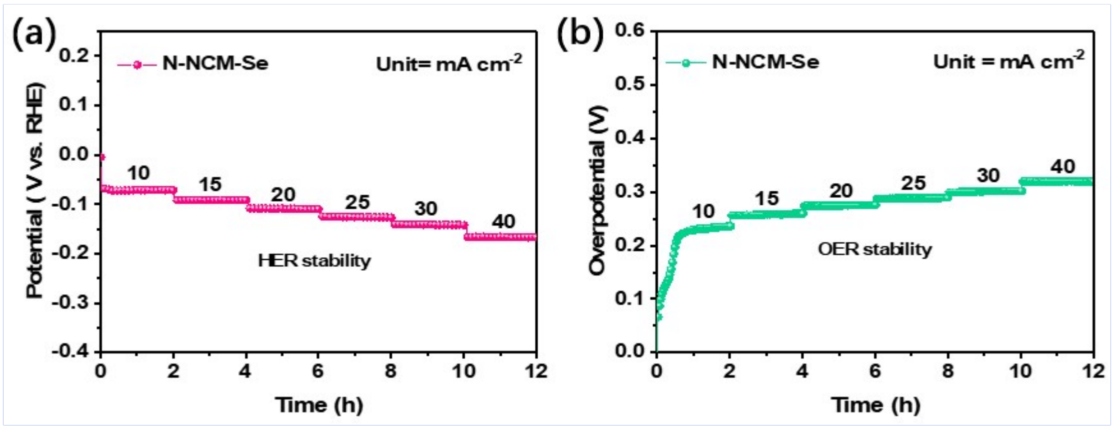

Figure S17. (a) HER and (b) OER stabilities of N-NCM-Se at different current densities.

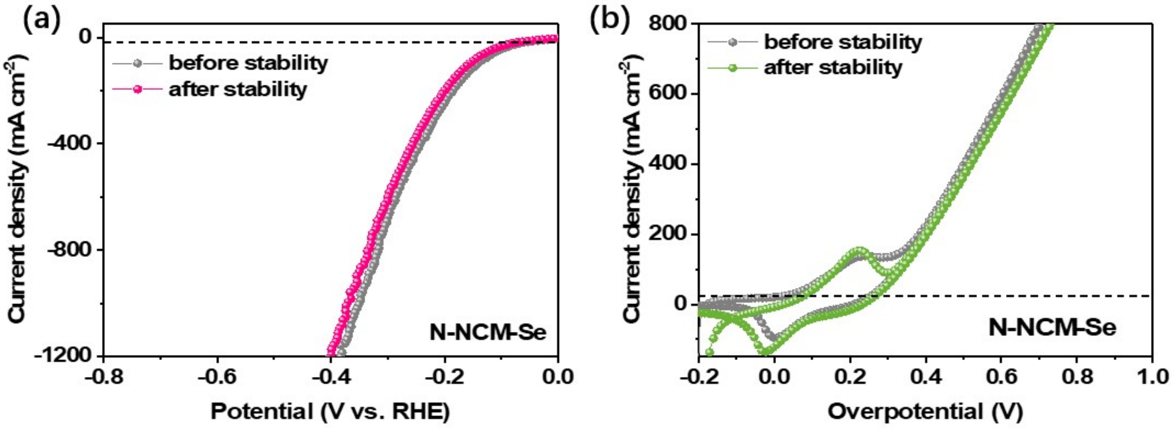

Figure S18. (a) HER LSV curves and (b) OER CV curves of N-NCM-Se before and after 60 h long term stability test.

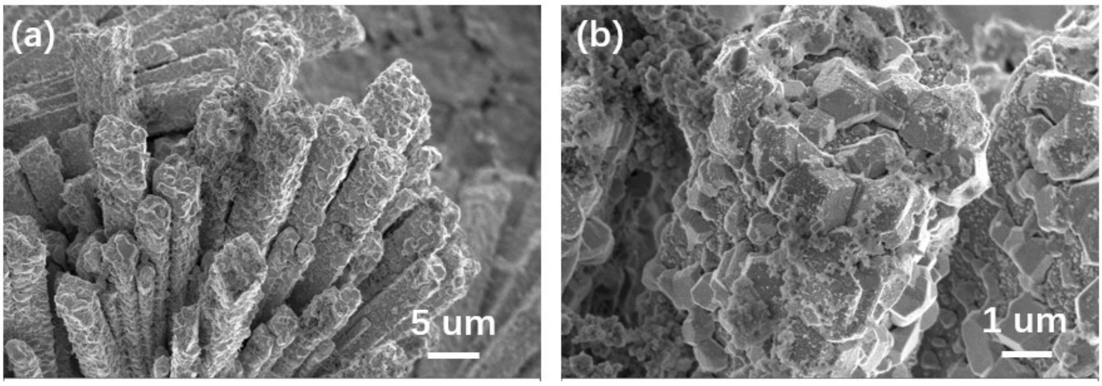

Figure S19. SEM images of N-NCM-Se after 60 h HER stability test.

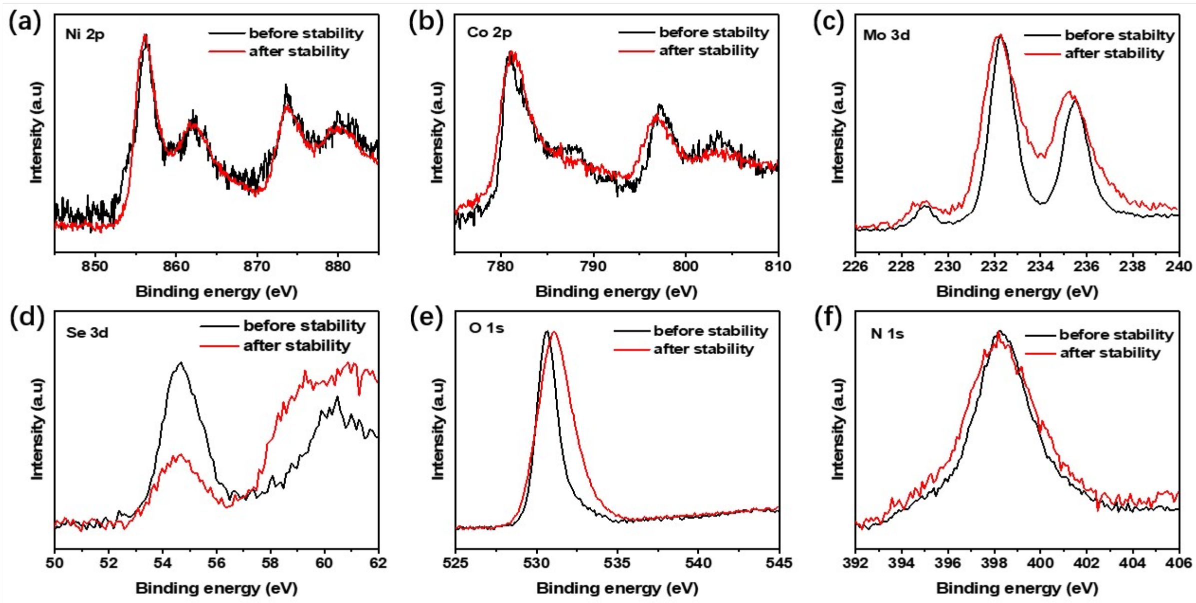

Figure S20. (a) Ni 2p, (b) Co 2p, (c) Mo 3d, (d) Se 3d, (e) O 1s, and (f) N 1s XPS spectra of N-NCM-Se before and after 60 h HER stability test.

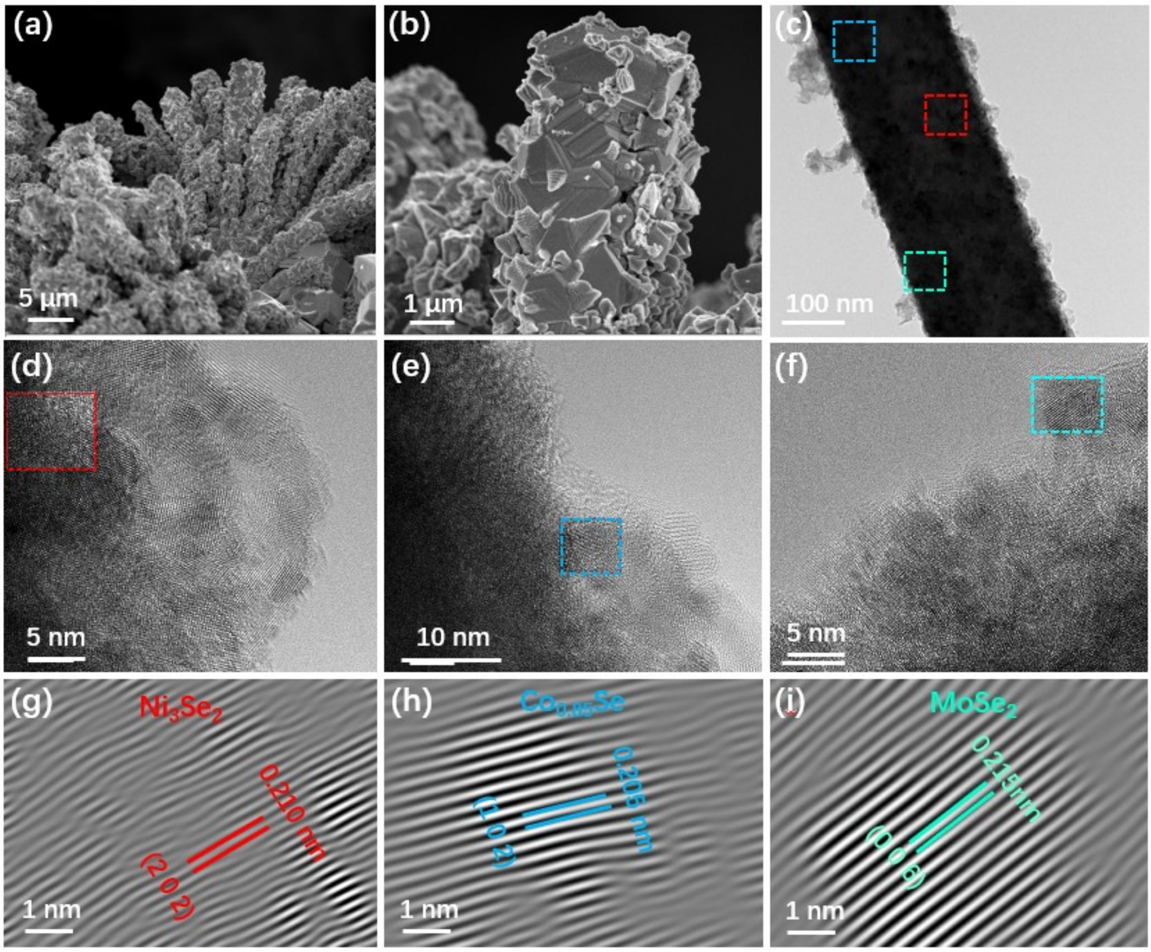

Figure S21. Structural stability of N-NCM-Se after 60 h OER stability. (a-b) SEM and (c) TEM images of N-NCM-Se. (d-f) HRTEM images for different constituents of the composite and (g-i) the corresponding lattice fringes.

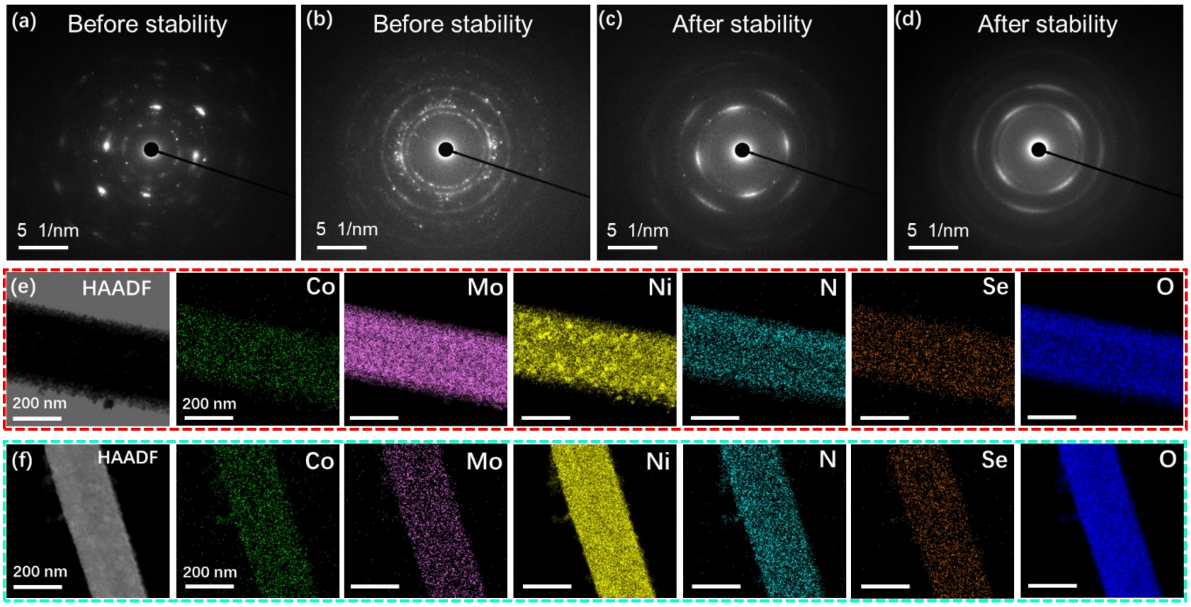

Figure S22. (a-b) SAED of N-NCM-Se before stability. (c-d) SAED of N-NCM-Se after stability and their corresponding HAADF/TEM elemental mapping (e) before and (f) after OER stability.

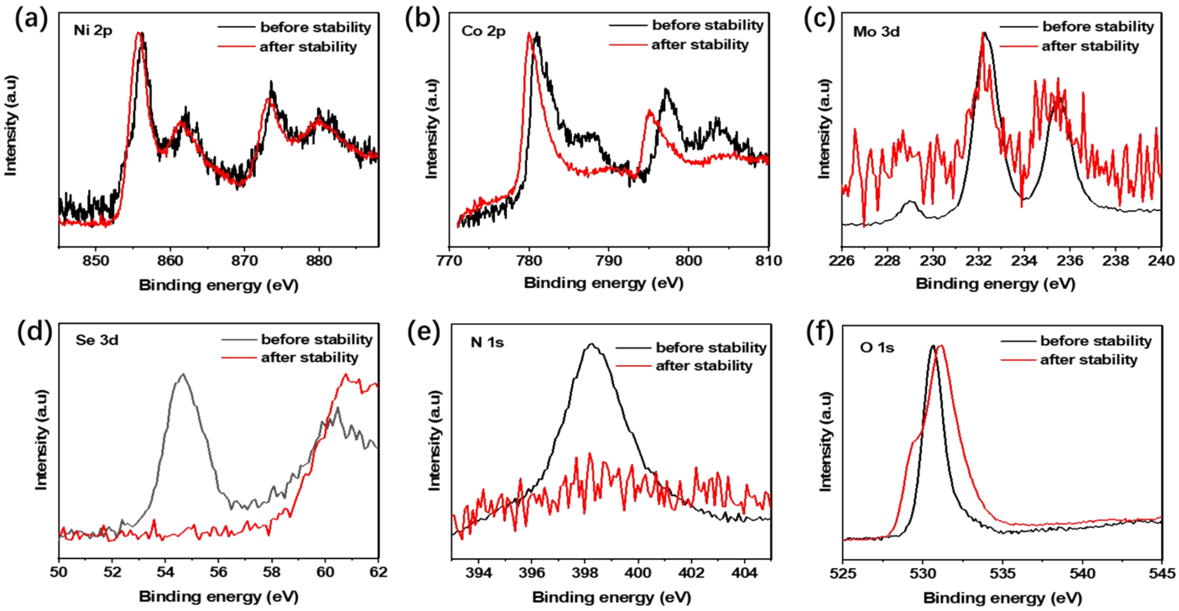

Figure S23. (a) Ni 2p, (b) Co 2p, (c) Mo 3d, (d) Se 3d, (e) N 1s, and (f) O 1s XPS spectra of N-NCM-Se before and after 60 h OER stability test.

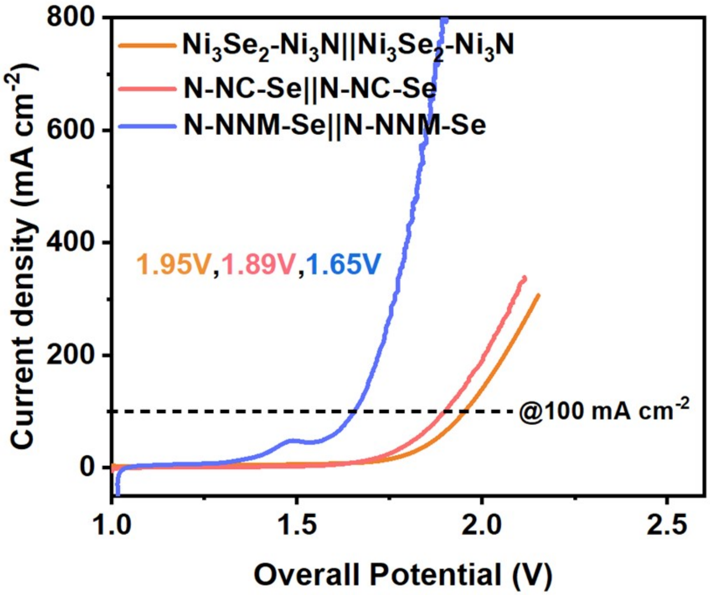

Figure S24. OWS LSV curves for Ni_3Se_2-Ni_3N, N-NC-Se and N-NNM-Se electrodes using both cathode and anode.

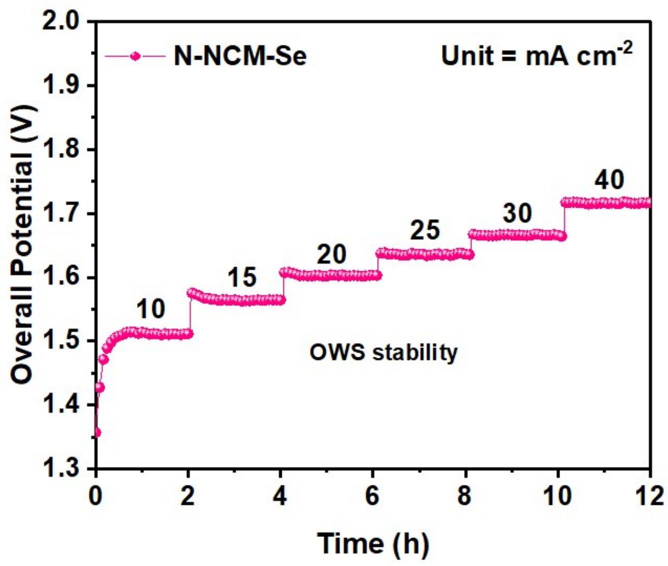

Figure S25. OWS stability of N-NCM-Se||N-NCM-Se for 12 h at different densities.

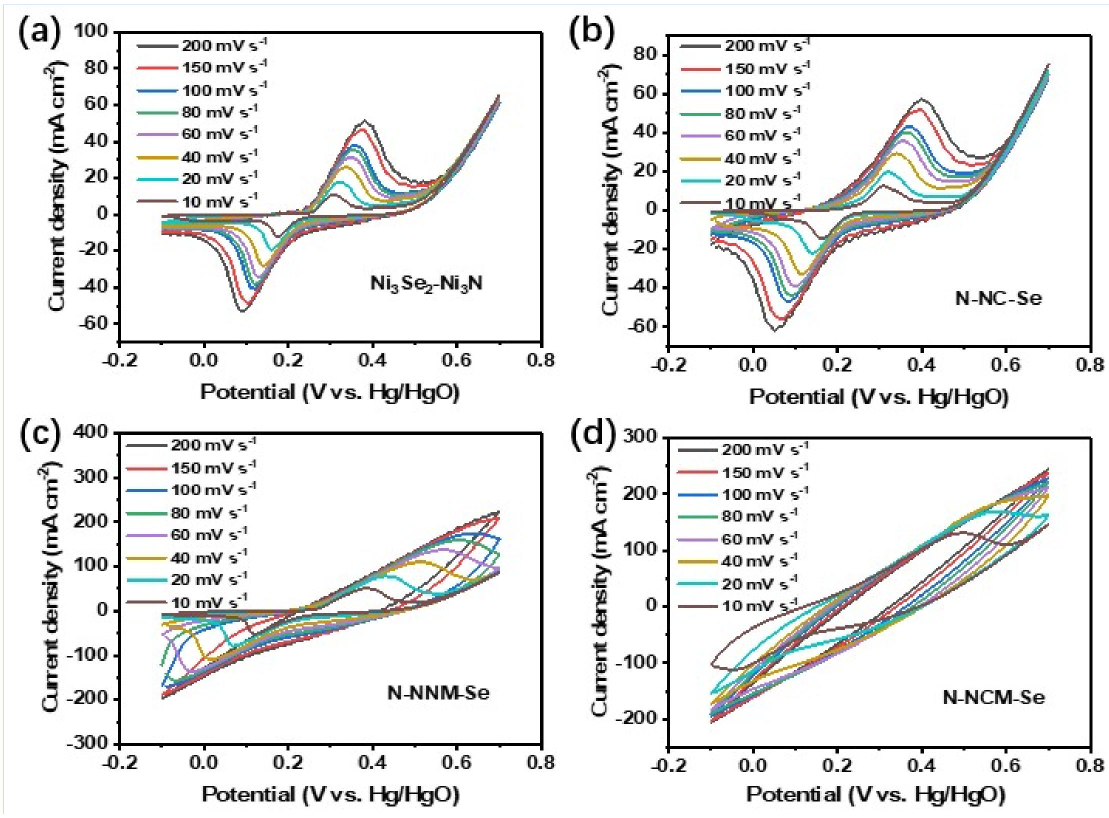

Figure S26. CV curves of (a) Ni_3Se_2-Ni_3N, (b) N-NC-Se (c) N-NNM-Se and (d) N-NCM-Se at different scan rates for the SC three electrode system.

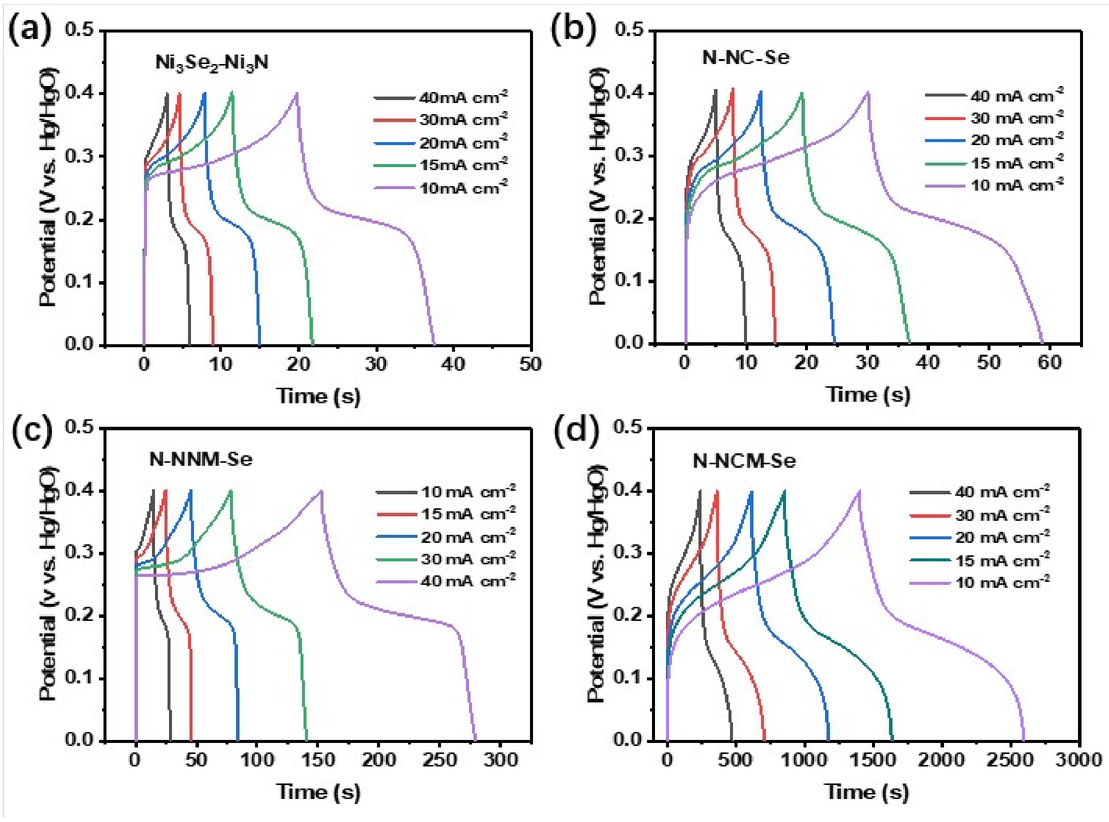

Figure S27. GCD curves of (a) Ni_3Se_2-Ni_3N, (b) N-NC-Se (c) N-NNM-Se and (d) N-NCM-Se at different current densities for the SC three electrode system.

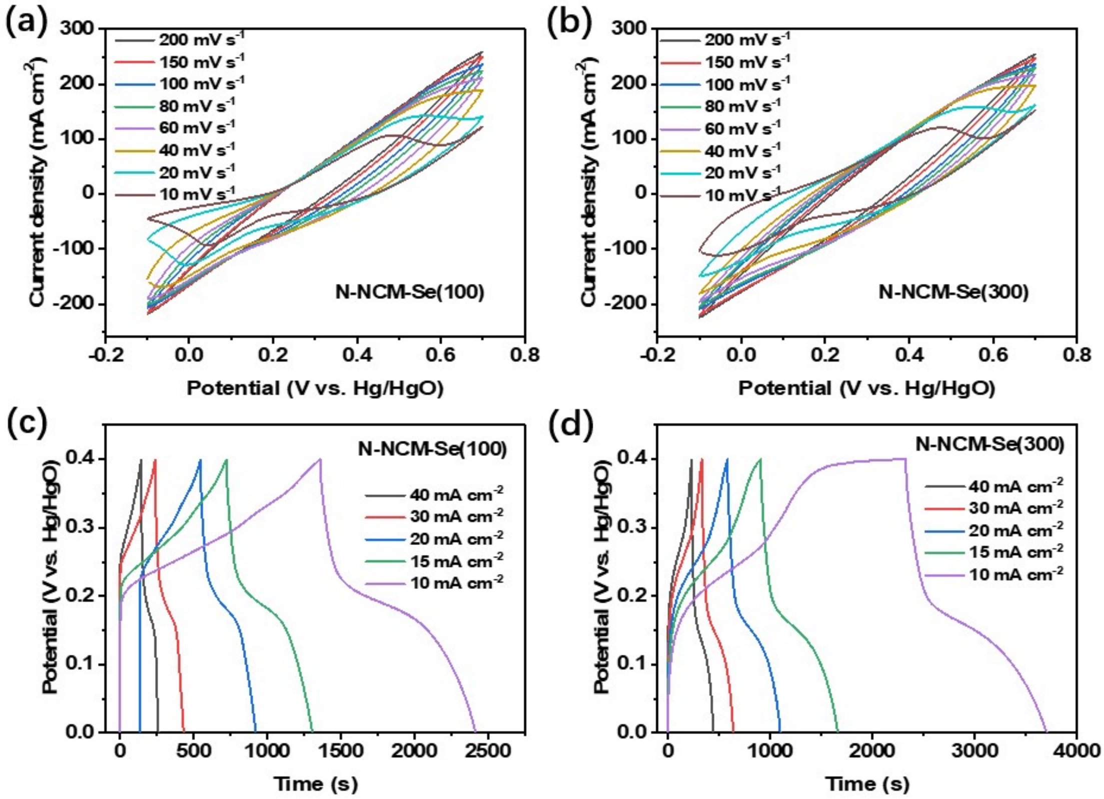

Figure S28. CV curves of (a) N-NCM-Se/100 and (b) N-NCM-Se/300 at different scan rates for the SC three electrode system. (c-d) GCD curves of (c) N-NCM-Se/100 and (d) N-NCM-Se/300 at different current densities for the SC three electrode system.

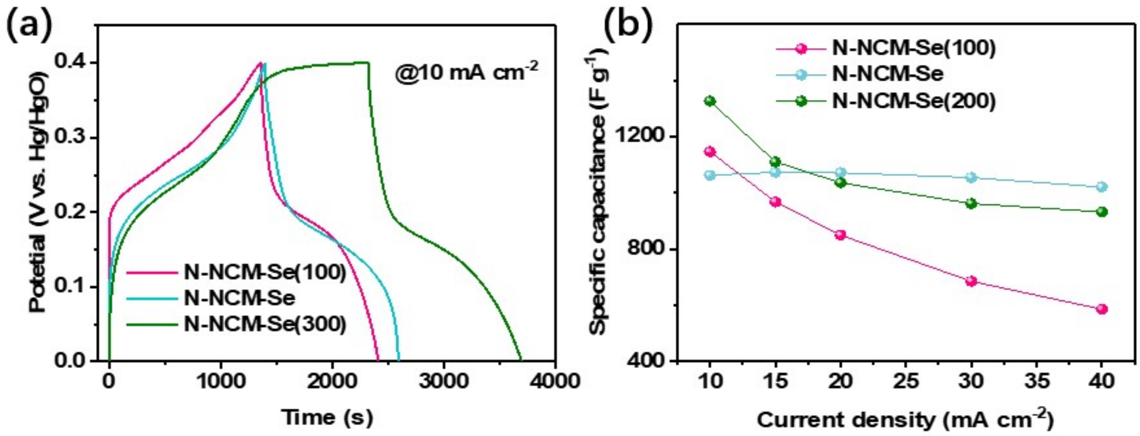

Figure S29. Comparisons of the (a) GCD curves at 10 mA cm^{-2} and (b) specific capacitances of N-NCM-Se electrodes with different selenium concentrations.

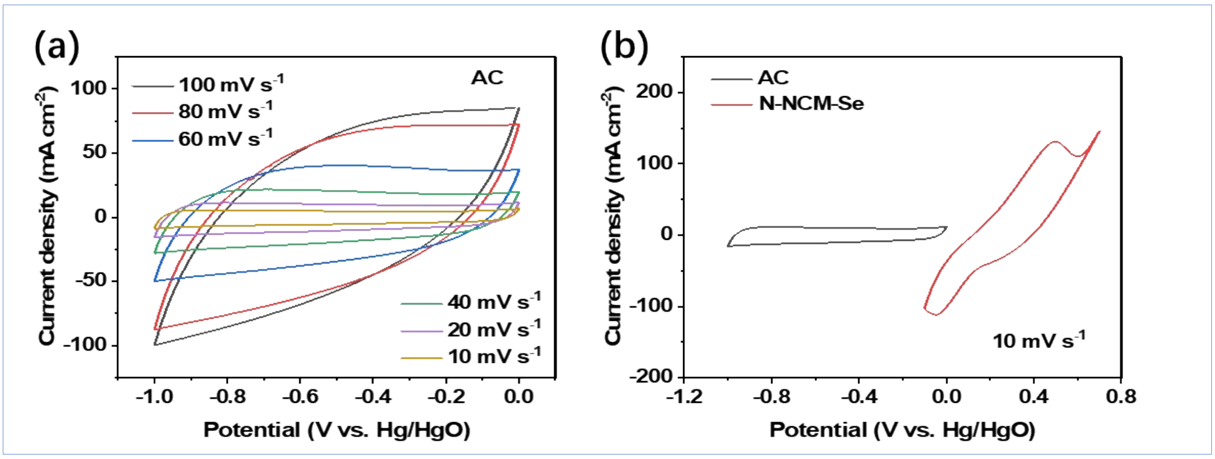

Figure S30. CV curves for (a) AC anode and (b) combined CVs of AC anode and N-NCM-Se cathode.

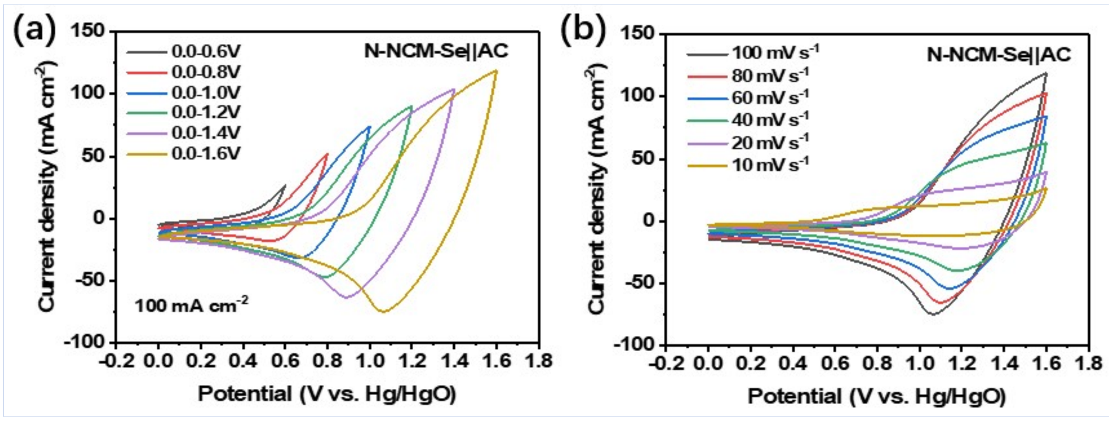

Figure S31. (a) CV curves of N-NCM-Se||AC ASC device using different electrochemical window at fixed scan rate of 100 mV s^{-1}. (b) CV curves of N-NCM-Se||AC ASC device at wide potential window at different scan rates using fixed potential range of 0 - 1.6 V.

Table S3. N-NCM-Se||AC performance comparison with other recent reports on selenides-based electrode in ASC.

C =Specific capacitance; E= Energy density; and P= Power density

Materials	Electrolyte	C (F g-1)	E (Wh kg^{-1})	P (W kg^{-1})	References
M-Ni_3Se_2@CoSe||AC	**3MKOH**	**206 @0.6 A g^{-1}**	**73**	**1838**	**Our work**
3DG/ZnSe-$SnSe_2$||AC	3MKOH	80.5 @1 A g^{-1}	25.3	750	ACS Appl. Nano Mater. 2024, 7, 13434–13446
$CoSe_2$@NiMn-LDH@$Cu_{1.8}$Se||AC	6M KOH	102 @0.2A g^{-1}	36.6	760.2	J. Colloid Interface Sc, 655 (2024) 273-285
3DG/(Co,Ni)Se_2 CNWs||AC	6M KOH	78.1 @ 1Ag^{-1}	28	800	Colloid Surf A: Phys. Eng, 698 (2024) 134595
SnO_2-SnSe||AC	3M KOH	93.8 @ 1Ag^{-1}	33.4	800.4	Nano Energy, 126 (2024) 109690
Bi_2Se_3||$NiCo_2S_4$	3M KOH	139 @ 0.5 A g^{-1}	48	800	J. Energy Storage,75, 2024, 109662
$NiCo_2Se_4$||AC	6M KOH	120 @ 2 A g^{-1}	42.4	1600	J. Alloys Compd 973 (2024) 172913
$MnCoSe_2$||rGO	1M KOH	71 @ 1A g^{-1}	32	227	Surf. Interfaces 42 (2023) 103358
CoSe-Se@Ni||AC	3.0 M KOH	106.73@0.5A g^{-1}	37.94	475.30	Ceramics International 47 (2021) 15293–15306
V-(Ni,Co)Se_2@Nb_2CTx||AC	6 M KOH	110.8 @ 2 A g^{-1}	45.6	168.4	J. Chem. Eng. 484 (2024) 149440
CuCoSe@NiS/NF||AC	3.0M KOH	117.4 @ 1 A g^{-1}	41.8	800	J. Mater. Chem. A, 2024,12, 13818-13829