Electronic Supplementary Material (ESI) for Dalton Transactions.

Supporting information

Multi-layered Heterogeneous Interfaces Created in $Co_{0.85}Se@Ni_3S_4$/NF to Enhance Supercapacitor Performances by Multi-step Alternating Electrodoposition

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Synthesis of PPy

Polypyrrole (PPy) was synthesized via an electrodeposition process by galvanostatic method carried out on a CHI660E electrochemical workstation with carbon cloth as the working electrode, an Ag/AgCl, KCl (saturated) electrode as the reference electrode and a Pt sheet (1.2 cm×1.2 cm) as the counter electrode. The typical deposition was performed in a mixed solution of 0.01 mmol SDBS (sodium dodecyl benzene sulfonate) and 0.15 mmol pyrrole monomer by galvanostatic method with a potential of 1 V for 800 seconds.

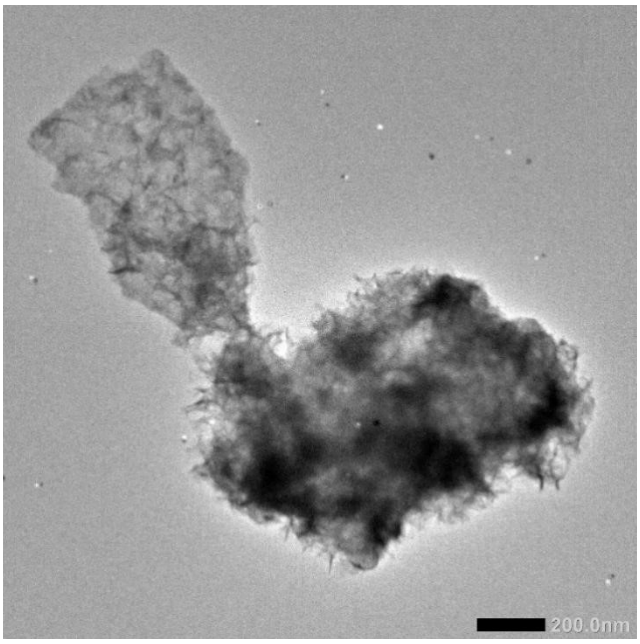

Fig. S1 The TEM image of 8L-$Co_{0.85}Se$@Ni_3S_4@NF

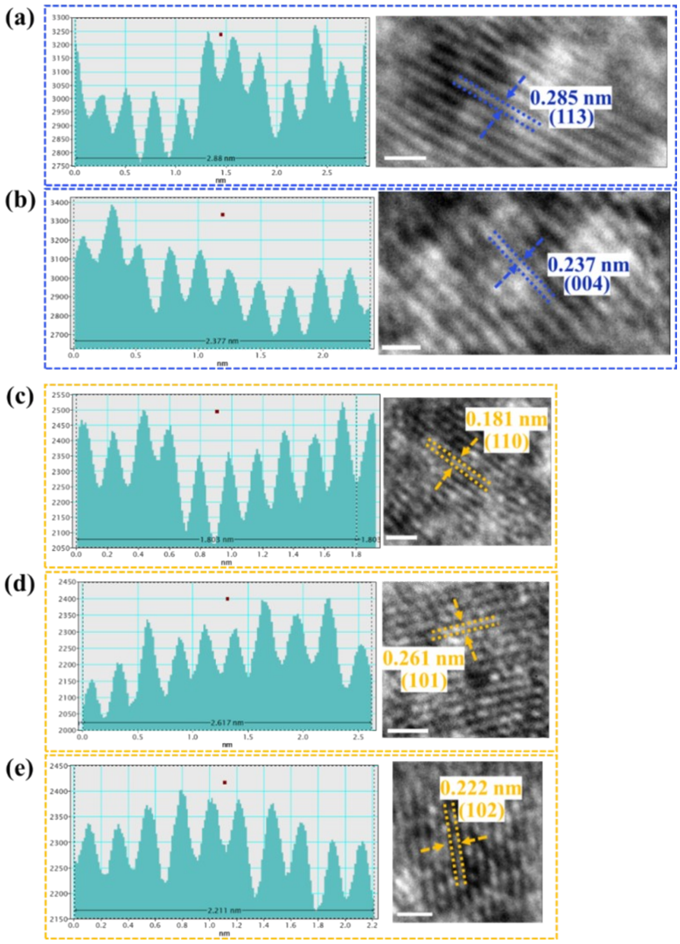

Fig. S2 HRTEM and corresponding IFFT images for Ni_3S_4 and $Co_{0.85}Se$ in Fig. 3b: (a) (113) and (b) (004) planes of Ni_3S_4; (c) (110), (d) (101) and (e) (102) planes of $Co_{0.85}Se$. The scale bar is 0.5 nm.

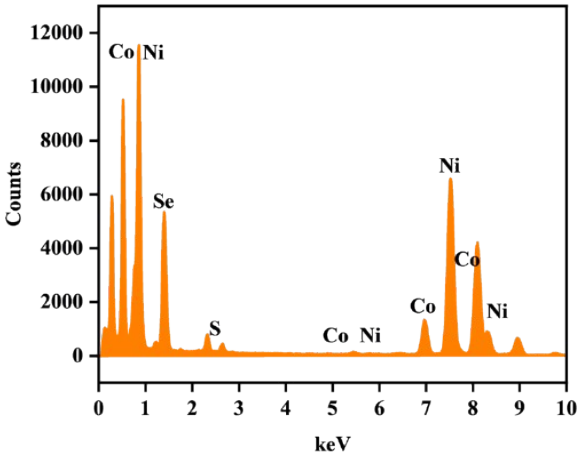

Fig. S3 EDS spectrum of 8L-$Co_{0.85}Se@Ni_3S_4$/NF

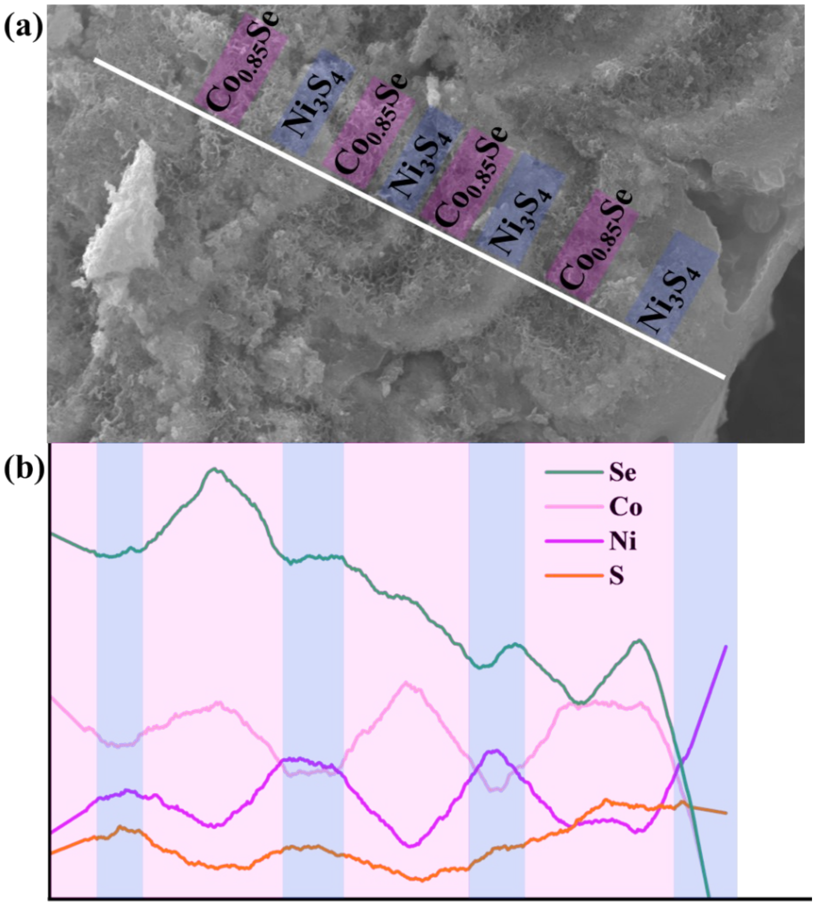

Fig. S4 SEM image and cross-section linear scanning EDS spectrum of 8L-$Co_{0.85}Se@Ni_3S_4$/NF.

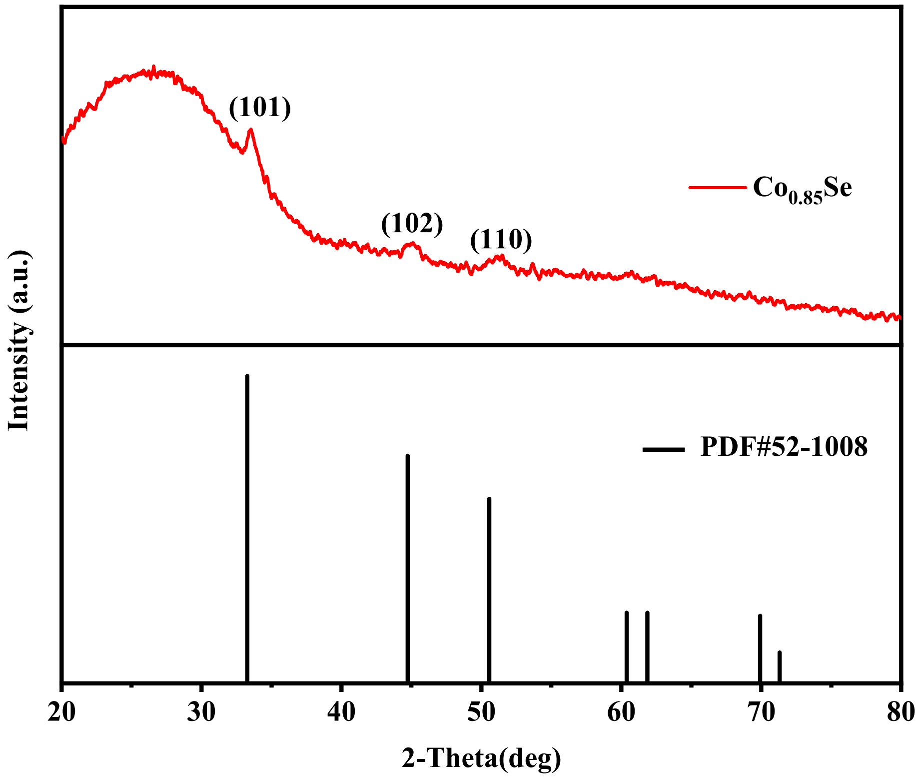

Fig. S5 XRD pattern of 1L-$Co_{0.85}Se$

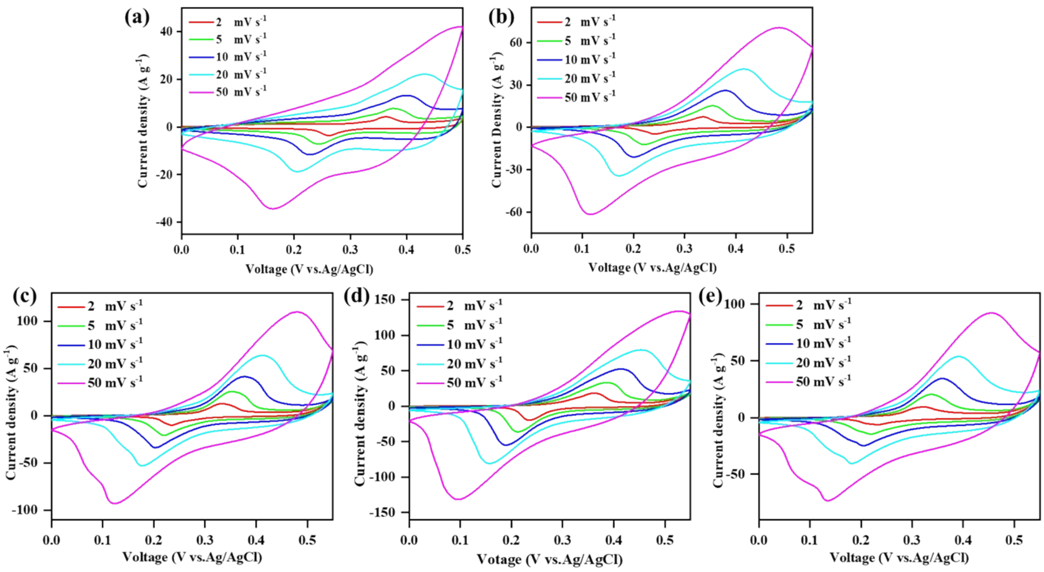

Fig. S6 CV curves of (a) 1L-$Co_{0.85}Se$@NF, (b) 2L-$Co_{0.85}Se$@Ni_3S_4/NF, (c) 4L-$Co_{0.85}Se$@Ni_3S_4/NF, (d) 6L-$Co_{0.85}Se$@Ni_3S_4/NF and (e) CoSeNiS/NF electrodes at scan rates from 2 to 50 mV s^{-1}.

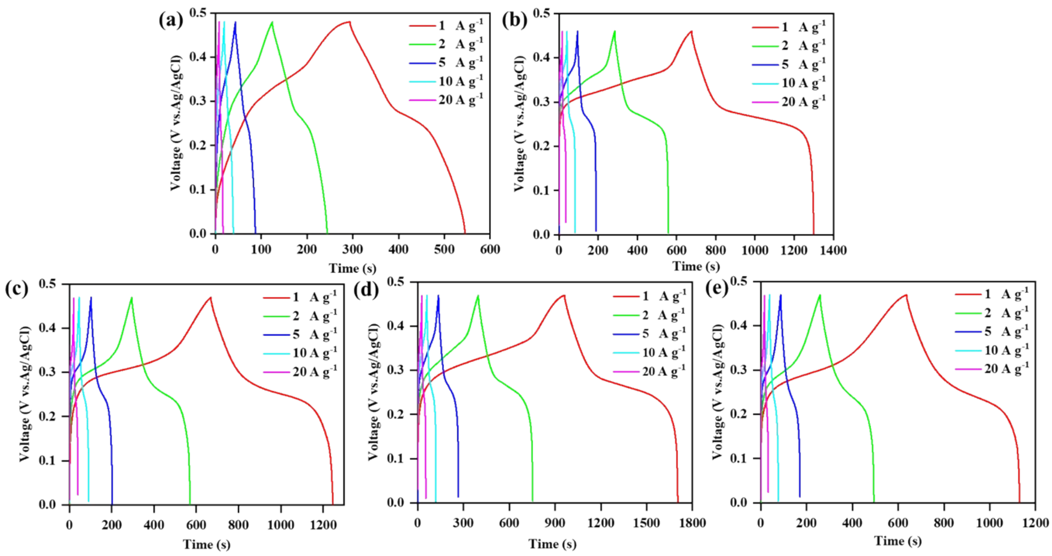

Fig. S7 GCD curves of (a) 1L-$Co_{0.85}Se$@NF, (b) 2L-$Co_{0.85}Se$@Ni_3S_4/NF, (c) 4L-$Co_{0.85}Se$@Ni_3S_4/NF, (d) 6L-$Co_{0.85}Se$@Ni_3S_4/NF and (e) CoSeNiS/NF electrodes at different current densities of 1 A g^{-1}, 2 A g^{-1}, 5 A g^{-1}, 10 A g^{-1} and 20 A g^{-1}.

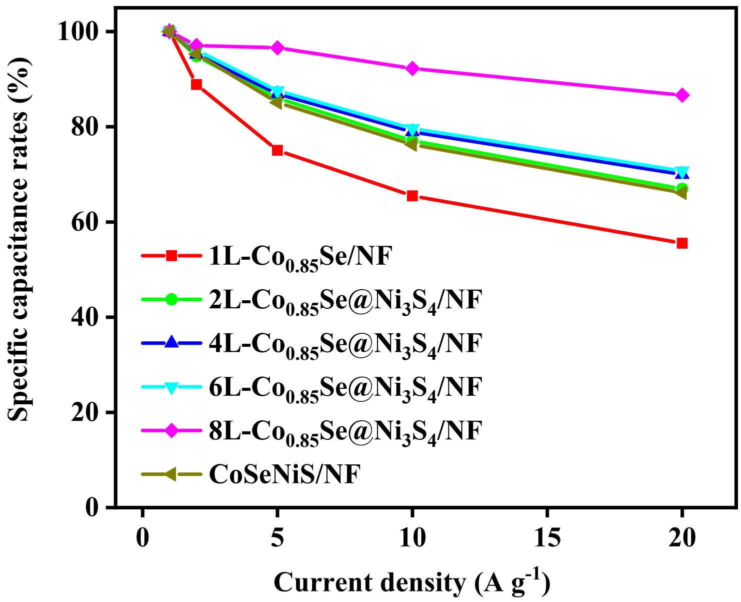

Fig. S8 Capacitance retentions rates of different electrodes at different current densities.

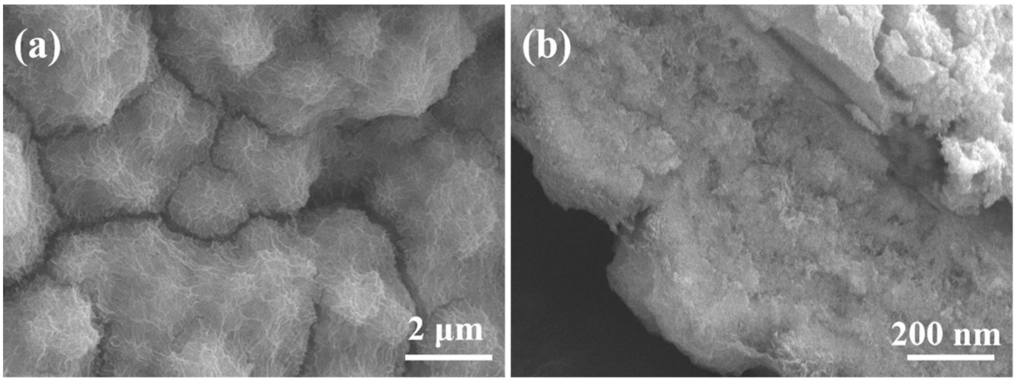

Fig. S9 SEM images of 8L-$Co_{0.85}Se$@Ni_3S_4@NF after cycling 5000 cycles.

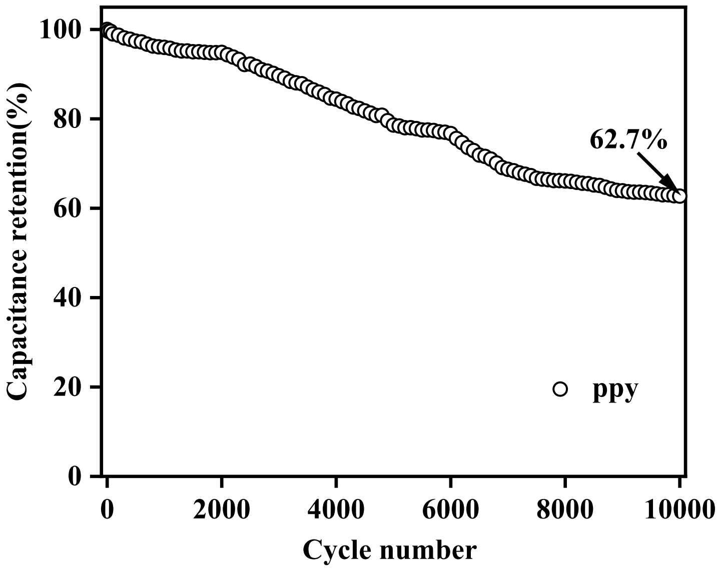

Fig. S10 Cycle retention test of ppy/NF

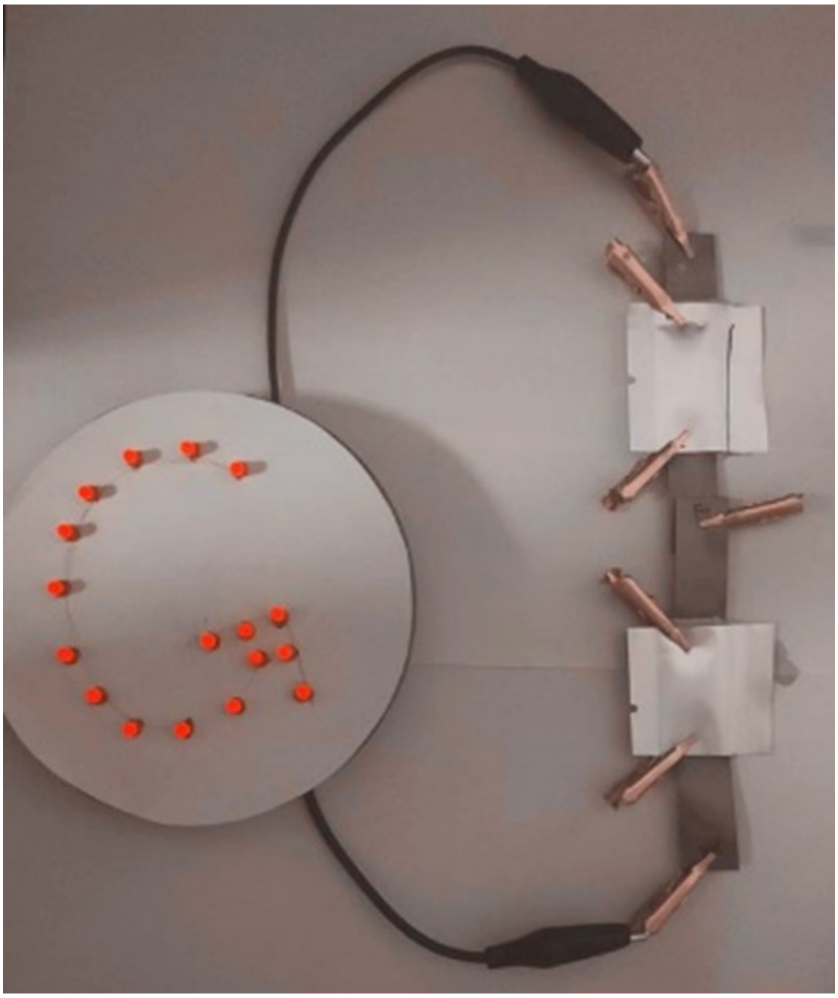

Fig. S11 Lighting 17 LED lights by two ASC devices connected in series.

Table S1 Element contents from EDS analysis of 8L-$Co_{0.85}Se@Ni_3S_4$/NF

Element	Weight（%）	Atomic
S	18.28	0.35
Co	22.09	0.23
Ni	24.87	0.26
Se	34.76	0.27

Table. S2 Electrochemical performances comparisons of 8L-$Co_{0.85}Se@Ni_3S_4$ with the recently reported CoSe-based and/or Ni_3S_4-based materials.

Materials	**Specific Capacitance**	**References**
8L-$Co_{0.85}Se@Ni_3S_4$/NF	1558.33 F g^{-1} at 1 A g^{-1}	This work
$CoFe_2Se_4$@CoNi-CH	1288.89 F g^{-1} at 1 A g^{-1}	*J. Colloid Interface Sci.* 2022, **621**, 149-159.
$Ni_9S_8@Ni_2B$	1555.33 F g^{-1} at 1 A g^{-1}	*J. Colloi Interface Sci.* 2023, **649**, 815-825.
$NiMo_3S_4$/BP	830 F g^{-1} at 1 A g^{-1}	*Small* 2024, **20**, 2310120
V-Ni_3S_2	1448.4 F g^{-1} at 1 A g^{-1}	*J. Colloid. Interface Sci.* 2023, **629**, 1049-1060.
$CoSe_2/NiSe_2$	1302.5 F g^{-1} at 1 A g^{-1}	*J. Electroanal. Chem.* 2021, **895**, 115479.
Ni_xSe_y	1025 F g^{-1} at 1 A g^{-1}	*J. Electroanal. Chem.* 2021, **895**, 115479.
(Ni,Co)Se_2-T	1412.5 F g^{-1} at 1 A g^{-1}	*Electrochim. Acta* 2021, **393**, 139049.
Ni_3S_2/Co_9S_8/C-2	1195 F g^{-1} at 1 A g^{-1}	*Appl. Surf. Sci.* 2022, **574**, 151727.

Table S3 Comparison table of the other identical ASCs with $Co_{0.85}Se@Ni_3S_4$/NF//PPy/NF ASC

Materials	Energy density and power density	Ref
$Co_{0.85}Se@Ni_3S_4$/NF//PPy/NF	76.98 Wh kg^{-1} at 775 W kg^{-1}	This work
$NiCo_2S_4$@HCs//AC	69.6 Wh kg^{-1} at 847 W kg^{-1}	*Adv. Funct. Mater.* 2023, **33**, 2210238
FCNS-2//AC	50 Wh kg^{-1} at 1.353 kW kg^{-1}	*Appl. Surf. Sci.* 2023, **611**, 155568
CoNiMn-S//RGO	42.1 Wh kg^{-1} at 750 W kg^{-1}	*Chem. Eng. J.* 2021, **405**, 126928
Ni_3S_{4-x}//AC	33.1 Wh kg^{-1} at 1.680 kW kg^{-1}	*Small* 2022, **18**, 2106074
$NiCo_2S_4$ //AC	21.4 Wh kg^{-1} at 1.663 kW kg^{-1}	*Mater. Res. Bull.* 2023, **157**, 112036
NiS/CNFs-2//AC	22.4 Wh kg^{-1} at 680 W kg^{-1}	*ACS Appl. Nano Mater.* 2022, **5**, 6192
$NiMoS_4/NiS_2$//NCO	38.6 Wh kg^{-1} at 958.6 W kg^{-1}	*Chem. Eng. J.* 2022, **435**, 135231
HCS//$Co_{0.85}Se@CoNi_2S_4$/GF	46.5 W h kg^{-1} at 750 W kg^{-1}	*J. Mater. Chem. A* 2018, **6**, 15630-15639.
Ni–Co–S/GF//PPy/GF	46.5 W h kg^{-1} at 825.0 W kg^{-1}	*Adv. Sci.* 2018, **5**, 1700375

Table S4 The cycle numbers corresponding to different electrochemical deposition layers

Layer numbers	**Sample/ cycle numbers**							
1	$Co_{0.85}Se$ /8							
2	$Co_{0.85}Se$ /4	Ni_3S_4 /4						
4	$Co_{0.85}Se$ /2	Ni_3S_4 /2	$Co_{0.85}Se$ /2	Ni_3S_4 /2				
6	$Co_{0.85}Se$ /2	Ni_3S_4 /2	$Co_{0.85}Se$ /1	Ni_3S_4 /1	$Co_{0.85}Se$ /1	Ni_3S_4 /1		
8	$Co_{0.85}Se$ /1	Ni_3S_4 /1	$Co_{0.85}Se$ /1	Ni_3S_4 /1	$Co_{0.85}Se$ /1	Ni_3S_4 /1	$Co_{0.85}Se$ /1	Ni_3S_4 /1