Supporting Information for

Durable modulation of Zn (002) plane deposition via reproducible zincophilic carbon quantum dots towards low N/P ratio Zinc-Ion Batteries

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Supplementary Figures and Tables

Table S1 Comparison in depth of discharge (DOD) and cycling lifespan for Zn||Zn symmetric cells with different modified strategies.

Modified strategies	Zn foil thickness (μm)	Current density (mA cm^{-2})	Areal capacity (mAh cm^{-2})	DOD (%)	Lifespan (h)	Ref.
N,S-CDs additive	***10***	***1.95***	***3.9***	***67***	***103***	***This work***
CNF-SO_3Zn separator	10 μm on one side and 130 μm on the other	2	3	50	100	[1]Adv. Funct. Mater., 2022
		1	5	80	60	
$Zn_3(PO_4)_2$/ZnF_2-rich in situ SEI	10	3	3	50	250	[2]Energy Environ. Sci., 2021
		4.7	4.7	80	120	
N,S-CDs additive	***20***	***1.95***	***7.8***	***67***	***250***	***This work***
	20	***3.9***	***7.8***	***67***	***200***	
Zwitterionic hydrogel electrolyte	~20	20	10	80.9	100	[3]Adv. Energy Mater., 2022
Polyamide coating	20	10	10	85	150	[4]Energy Environ. Sci., 2019
MX-TMA coating	20	10	10	85	450	[5]Energy Storage Mater., 2022
ZGL coating	20	1	5	42.7	560	[6]Energy Storage Mater., 2022
		1	10	85.5	250	
TiO_2 & PVDF coating	25	8.85	8.85	60	250	[7]Adv. Funct. Mater., 2021
Glass fiber gasket	~26	50	10	66.7	100	[8]Angew. Chem., Int. Ed., 2022

CO_2-purged electrolyte	30	10	10	57	100	[9]ACS Nano, 2022
NH_2-$PSiO_x$ coating	30	20	10	57	~300	[10]Energy Storage Mater., 2023
PSN coating	30	10	10	60	250	[11]Adv. Funct. Mater., 2021
2-propanol composite electrolyte	~50	15	15	51.2	500	[12]Adv. Mater., 2022
Cellulose film separator	~50	2	20	69.7	300	[13]Energy Storage Mater., 2022
ZP coating	50	25	17.67	65	200	[14]Angew. Chem., Int. Ed., 2023
		25	19.03	70	138	
		26.55	21.74	80	60	
Janus separator	~85	28.3	28.3	56	220	[15]Adv. Mater., 2022
CuHCF coating	100	30	30	51.3	200	[16]Small, 2022
Acetone additive	100	50	50	73.5	800	[17]Adv. Funct. Mater., 2023
Montmorillonite coating	100	10	45	77	>1000	[18]Adv. Energy Mater., 2021
ZnHAP/BC separator	100	1	29.1	50	1025	[19]Small, 2023
		1	46.5	80	611	
ZnP coating	100	20	30	51	300	[20]Adv. Funct. Mater., 2021
		15	48	82	100	

Table S2 Electrochemical performance of the full cells with different modified strategies.

Modified strategies	Zn foil	Cathode		Full cell performance		Cathode area specific capacity (mAh cm^{-2})	N/P ratio	Energy density (Wh kg^{-1})		Ref.
	Thickness (μm)	Material	Mass loading (mg cm^{-2})	Specific capacity (mAh g^{-1})	Current density			Based on active material	Including electrolyte	
Nafion-Zn-X coating	300	VS_2	3	~ 175	0.5 A g^{-1}	0.53	513.00	0.95	0.60	[21]Angew. Chem., Int. Ed., 2020
Cellulose film separator	200	α-MnO_2	2	~ 300	0.2 A g^{-1}	0.60	189.92	5.80	0.15	[13]Energy Storage Mater., 2022
F-GQDs additive	100	MnO_2	1	~ 330	0.2 A g^{-1}	0.33	189.92	6.38	NA	[22]Chem. Eng. J., 2023
$Zn_3(PO_4)_2/ZnF_2$-rich in situ SEI	100	α-MnO_2	1	~ 270	0.1 A g^{-1}	0.27	189.92	5.22	2.36	[2]Energy Environ. Sci., 2021
In^{3+} additive	100	V_2O_5	0.65	~ 540	0.2 A g^{-1}	0.35	152.81	3.90	NA	[23]Adv. Funct. Mater., 2022
TiO_2 & PVDF coating	80	MnO_2	1	244	2 C	0.24	151.94	5.88	1.94	[7]Adv. Funct. Mater., 2021
Mxene in situ coating	50	α-MnO_2	1	~ 300	0.2 A g^{-1}	0.30	94.96	11.44	NA	[24]Angew. Chem., Int. Ed., 2021
ZIF-8 in situ coating	50	$LaVO_4$	1.9	~ 95	10 mA cm^{-2}	0.18	72.96	3.84	NA	[25]Adv. Sci., 2020
rGO in situ coating	110	$V_3O_7 \cdot H_2O$	5.13	245	1.5 A g^{-1}	1.26	47.09	6.95	NA	[26]ACS Appl. Mater. Interfaces, 2018
Liquefied Gas Electrolytes	100	$Na_2V_6O_{16} \cdot 1.63H_2O$	~ 3	~ 260	0.02 A g^{-1}	0.78	38.65	8.39	NA	[27]ACS Energy Lett., 2021
Gelatin coating	50	V_6O_{13}	~ 2	~ 375	0.1 A g^{-1}	0.75	35.06	15.92	NA	[28]Adv. Energy Mater., 2021
Montmorillonite coating	100	MMT-MnO_2	8.5	~ 270	2 C	2.30	22.34	40.21	NA	[18]Adv. Energy Mater., 2021
1.4-dioxane additive	100	MnO_2	9.4	~ 130	0.05 A g^{-1}	1.22	20.20	21.17	NA	[29]ACS Appl. Mater. Interfaces, 2021

PVB coating	10	MnO_2	1.13	~ 250	1 C	0.28	16.86	47.71	NA	[30]Adv. Funct. Mater., 2020
1.4-dioxane additive	20	$NH_4V_4O_{10}$	13.3	~ 160	2 A g^{-1}	2.13	2.44	52.00	11.71	[31]ACS Nano, 2023
FCOF coating	NA	MnO_2	16	NA	3 mA cm^{-2}	~ 2.00	2.00	130.00	55.00	[32]Nat. Commun., 2021
	NA	MnO_2	8	~ 180	4 mA cm^{-2}	NA	5.00	113.00	NA	
	NA	MnO_2	8	~ 160	4 mA cm^{-2}	NA	10.00	74.00	NA	
GQDs additive	NA	NVO (PDF#49-0996)	1.5	350	0.1 A g^{-1}	0.53	NA	NA	NA	[33]Nano Energy, 2022
N,S-CDs additive	***10***	***$Na_2V_6O_{16}\cdot 3H_2O$***	***6.56***	***345***	***0.2 A g^{-1}***	***2.27***	***1.83***	***132.34***	***9.26***	***This work***
N,S-CDs additive (lean electrolyte)	***10***	***$Na_2V_6O_{16}\cdot 3H_2O$***	***11.52***	***294***	***0.2 A g^{-1}***	***3.38***	***1.05***	***144.98***	***39.34***	***This work***
	20	***$Na_2V_6O_{16}\cdot 3H_2O$***	***16.47***	***303***	***0.2 A g^{-1}***	***4.99***	***1.46***	***129.75***	***26.34***	

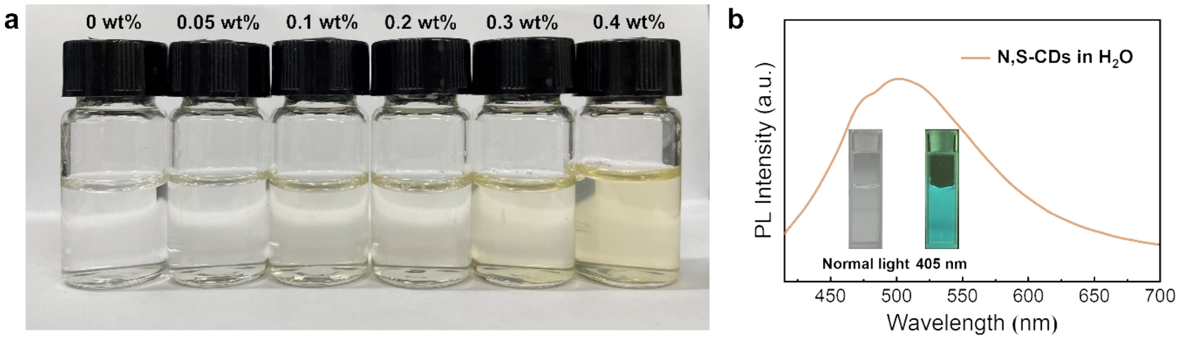

Fig. S1 (**a**) Digital image of 2 M $ZnSO_4$ electrolyte solution containing various amounts (0, 0.05, 0.1, 0.2, 0.3, and 0.4 wt %) of N,S-CDs additives. (**b**) Photoluminescence spectra of N,S-CDs in aqueous solutions under 405 nm excitation. Inset: optical images of the N,S-CDs aqueous solution under nature light and excited by 405 nm.

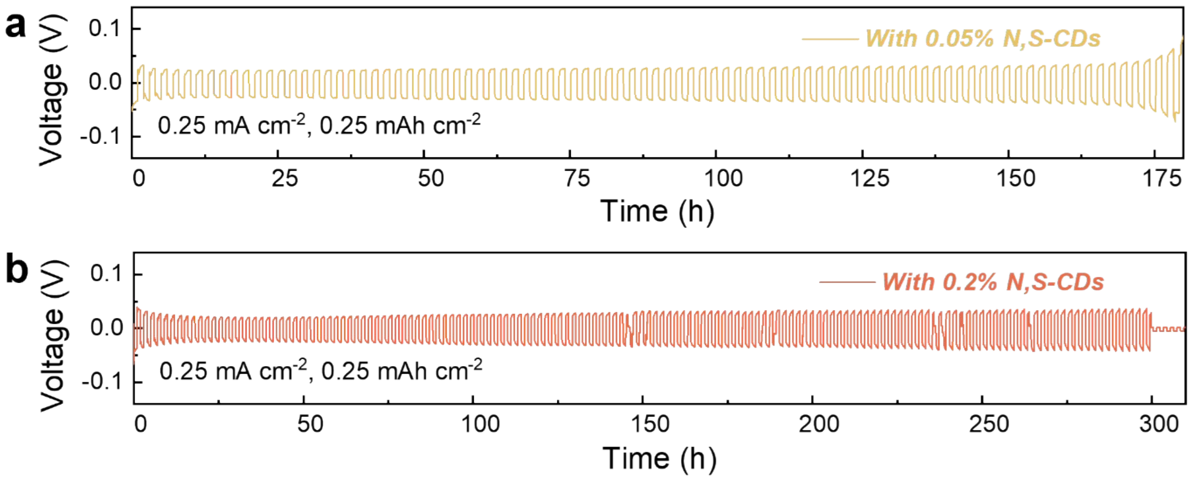

Fig. S2 Long-term cycling performance of the Zn||Zn symmetric cells using electrolyte with (**a**) 0.05% N,S-CDs and (**b**) 0.2% N,S-CDs at 0.25 mA cm^{-2} for 0.25 mAh cm^{-2}.

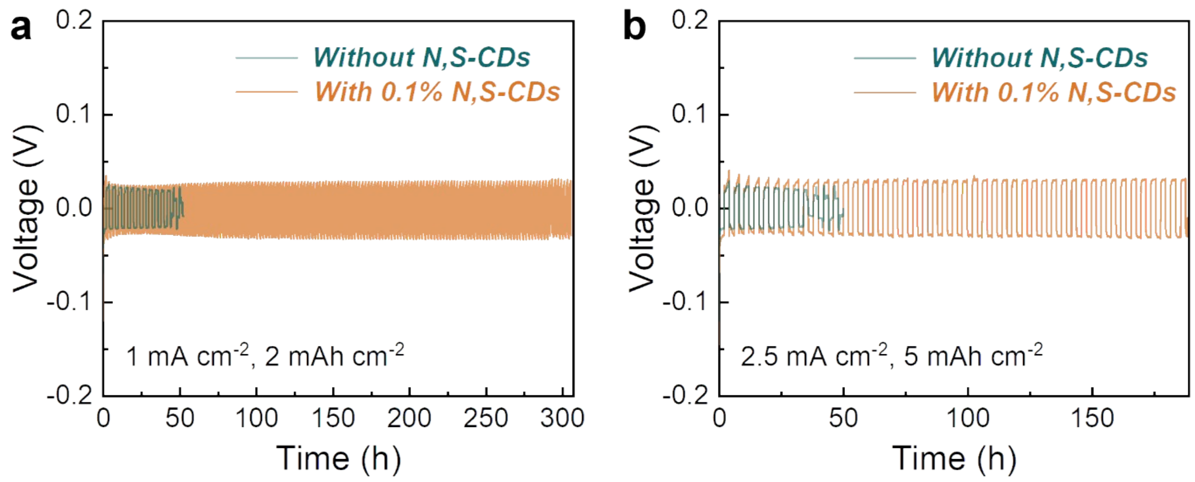

Fig. S3 Long-term cycling performance of the Zn||Zn symmetric cells using pristine $ZnSO_4$ electrolyte and electrolyte with 0.1% N,S-CDs at (**a**) 1 mA cm^{-2} for 2 mAh cm^{-2} and (**b**) 2.5 mA cm^{-2} for 5 mAh cm^{-2}.

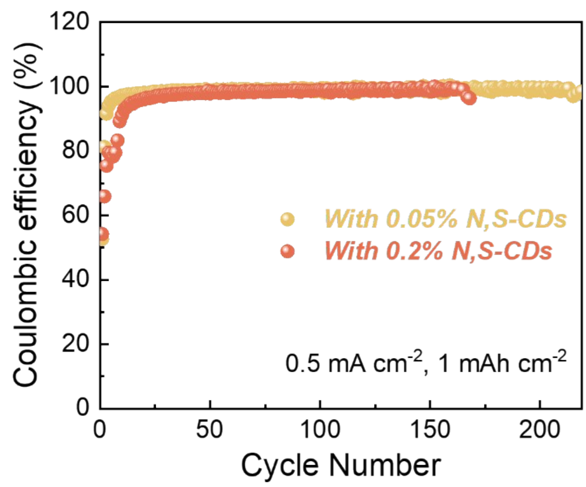

Fig. S4 Coulombic efficiency of Zn||Cu cells using electrolyte with 0.05% N,S-CDs and 0.2% N,S-CDs at 0.5 mA cm^{-2} for 1 mAh cm^{-2}.

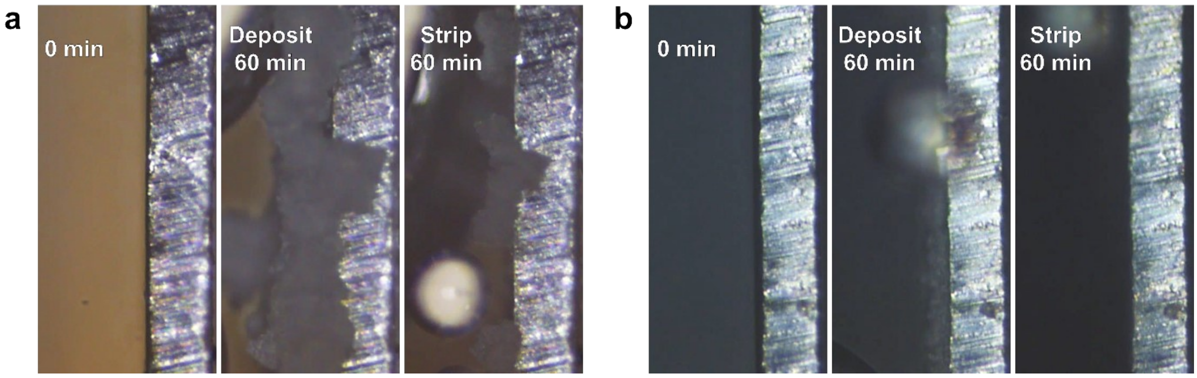

Fig. S5 In situ OM images of the anodes deposited at 5 mA cm^{-2} for 60 minutes and then stripped at 1 mA cm^{-2} for 60 minutes in the electrolyte (**a**) without and (**b**) with 0.1% N,S-CDs.

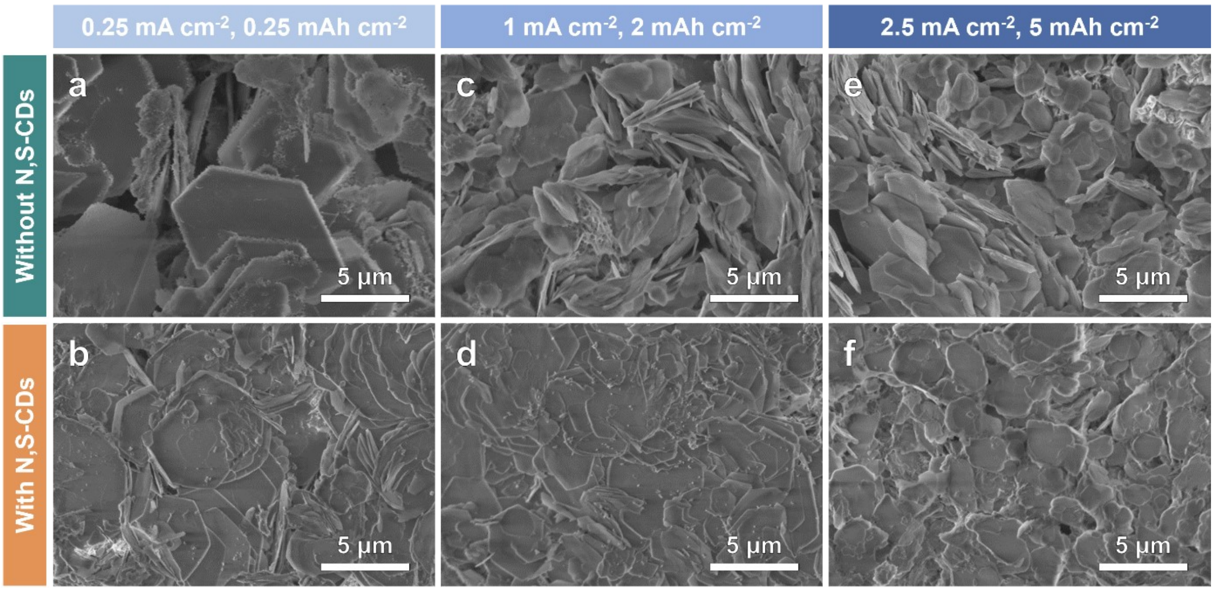

Fig. S6 Top-view SEM images of the anodes in the Zn||Zn symmetric cells using pristine $ZnSO_4$ electrolyte and electrolyte with 0.1% N,S-CDs at (**a, b**) 0.25 mA cm^{-2} for 0.25 mAh cm^{-2}, (**c, d**) 1 mA cm^{-2} for 2 mAh cm^{-2} and (**e, f**) 2.5 mA cm^{-2} for 5 mAh cm^{-2} after 15 cycles.

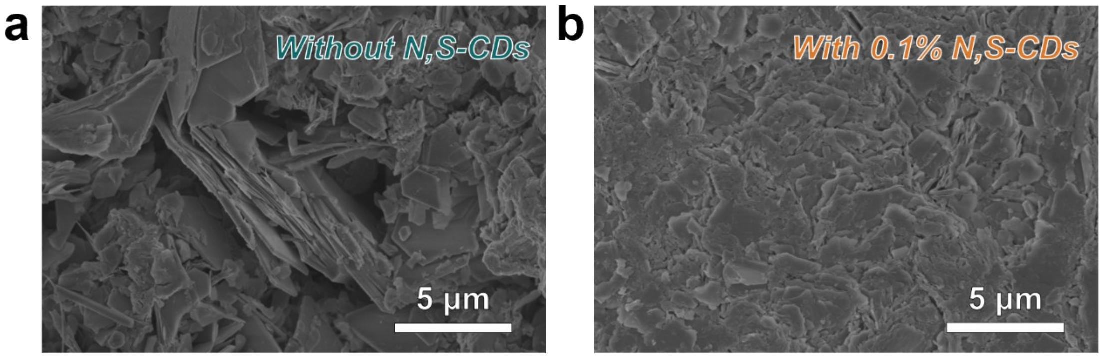

Fig. S7 Top-view SEM images of the anodes in the Zn||Zn symmetric cells after rate test (**a**) without and (**b**) with 0.1% N,S-CDs.

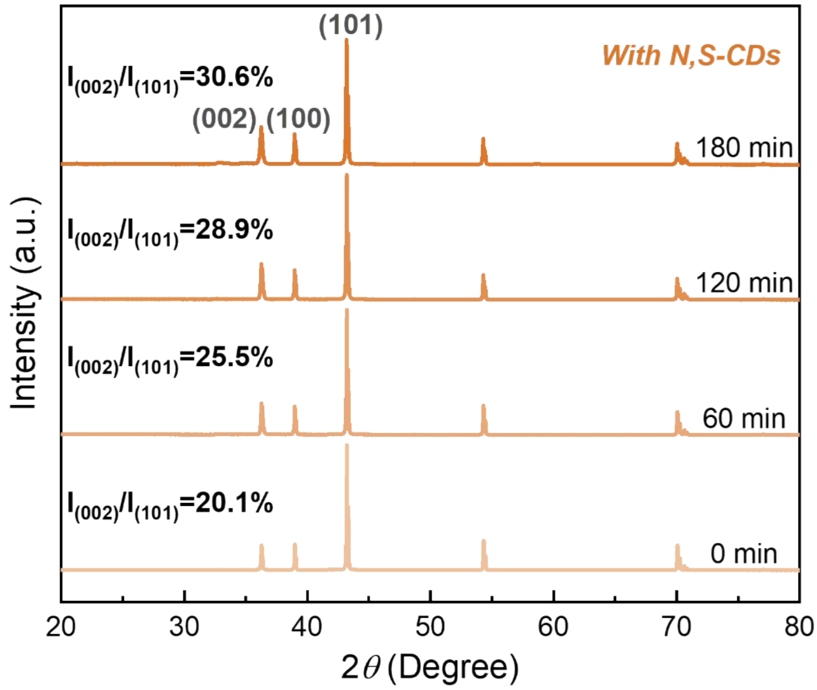

Fig. S8 XRD patterns of the Zn deposits with deposition time from 0 to 180 minutes in the electrolyte with 0.1% N,S-CDs.

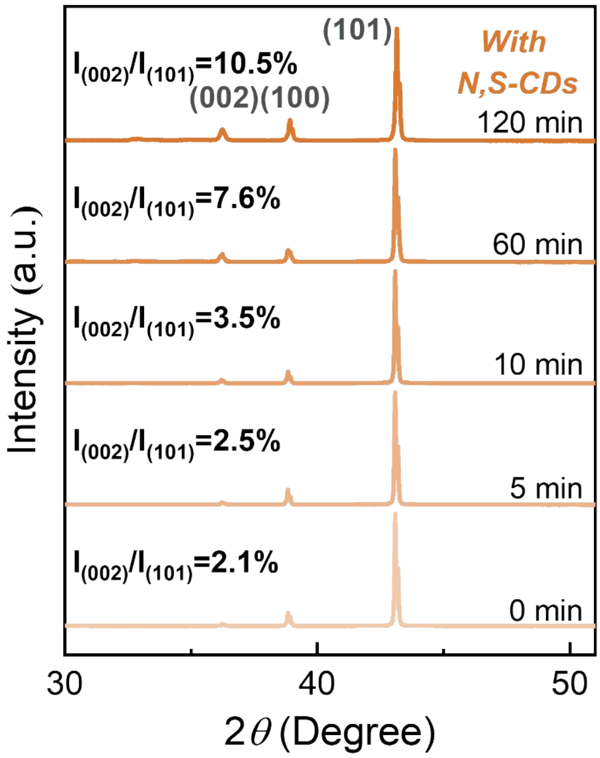

Fig. S9 XRD patterns of the Zn anode (with a lower initial $I_{(002)}/I_{(101)}$ ratio) using electrolyte with 0.1% N,S-CDs at various deposition time ranging from 0 to 120 min.

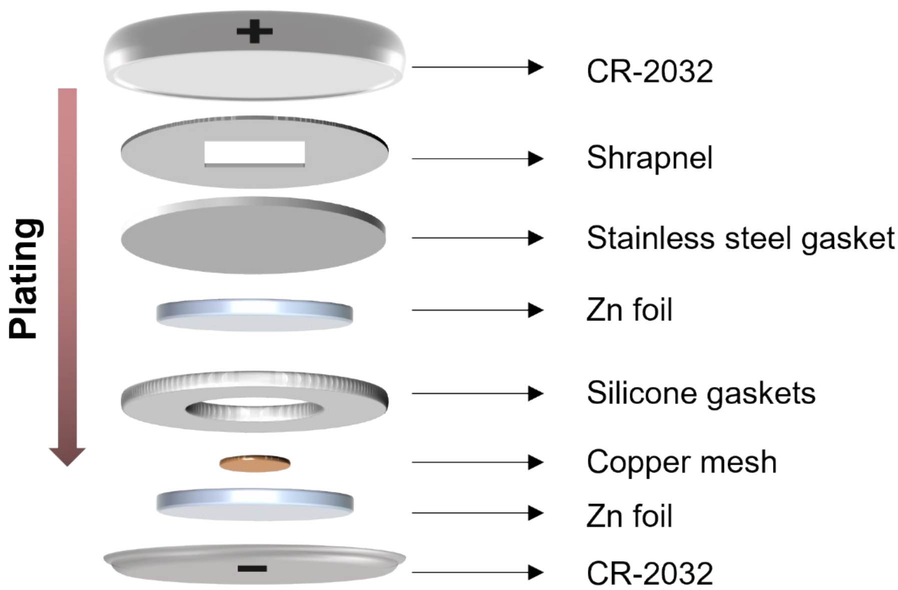

Fig. S10 Schematic illustration of the special-design cell configuration to obtain the in situ generated Zn deposit on a Cu mesh.

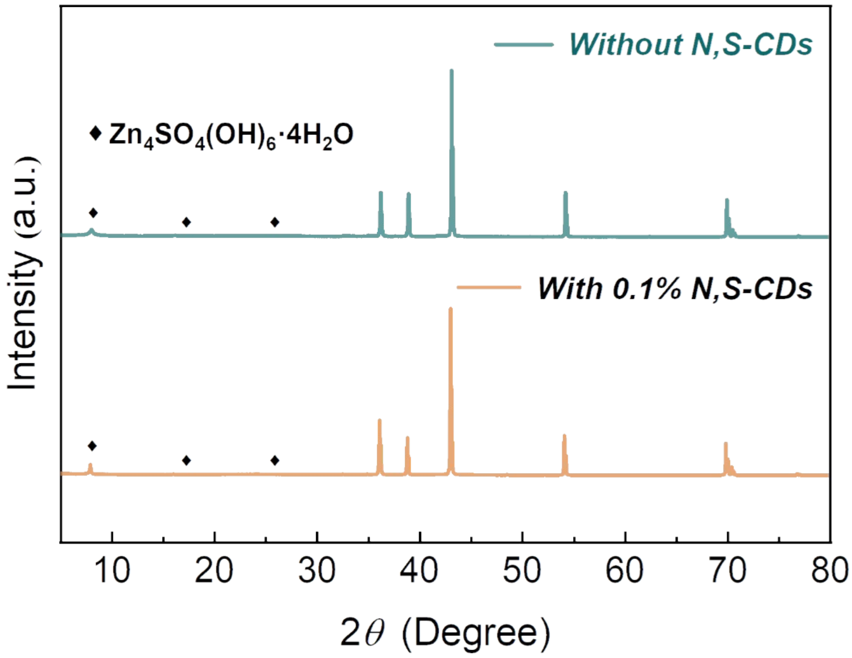

Fig. S11 XRD patterns of the cycled Zn anode using the electrolyte without and with 0.1% N,S-CDs.

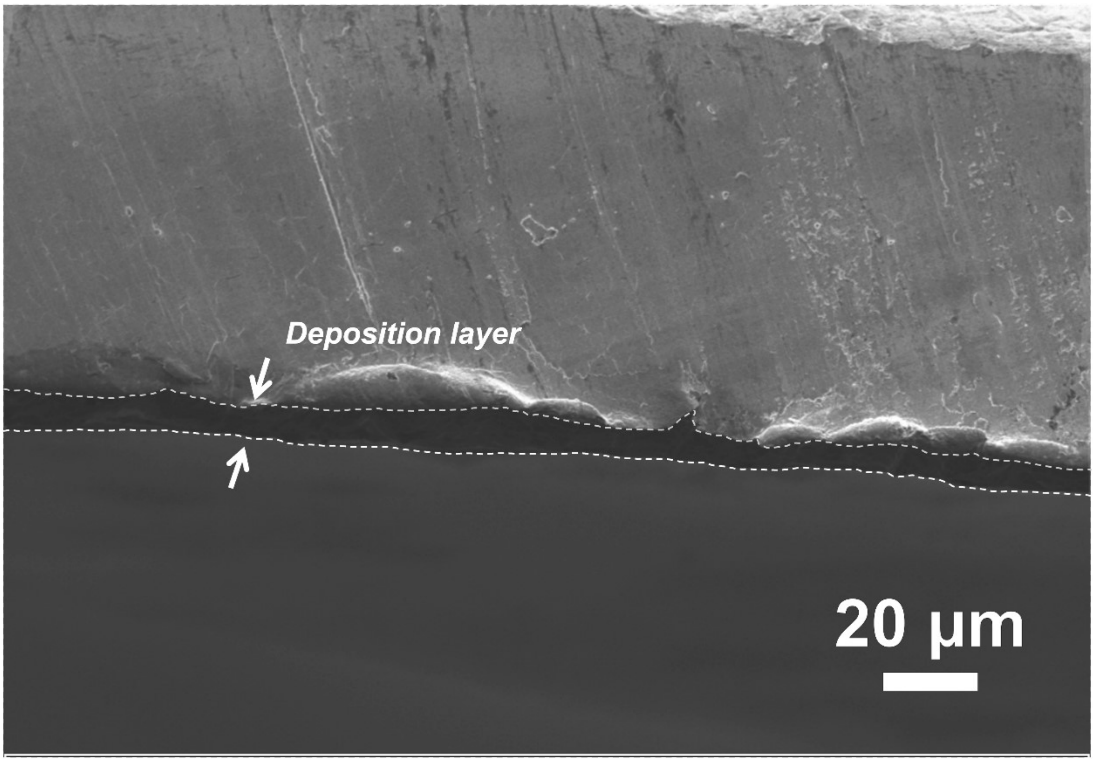

Fig. S12 Cross-section SEM image of the anodes deposited at 1 mA cm^{-2} for 1 mAh cm^{-2} in the electrolyte with 0.1% N,S-CDs.

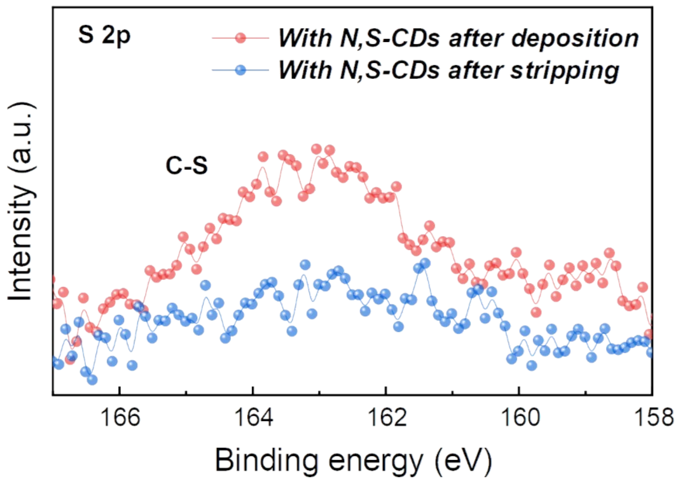

Fig. S13 XPS high-resolution spectra of S 2p of the anode surface after deposition (deposited at 1 mA cm^{-2} for 60 minutes) and stripping (deposited at 1 mA cm^{-2} for 60 minutes and then stripped at 1 mA cm^{-2} for 60 minutes) in the electrolyte with 0.1% N,S-CDs.

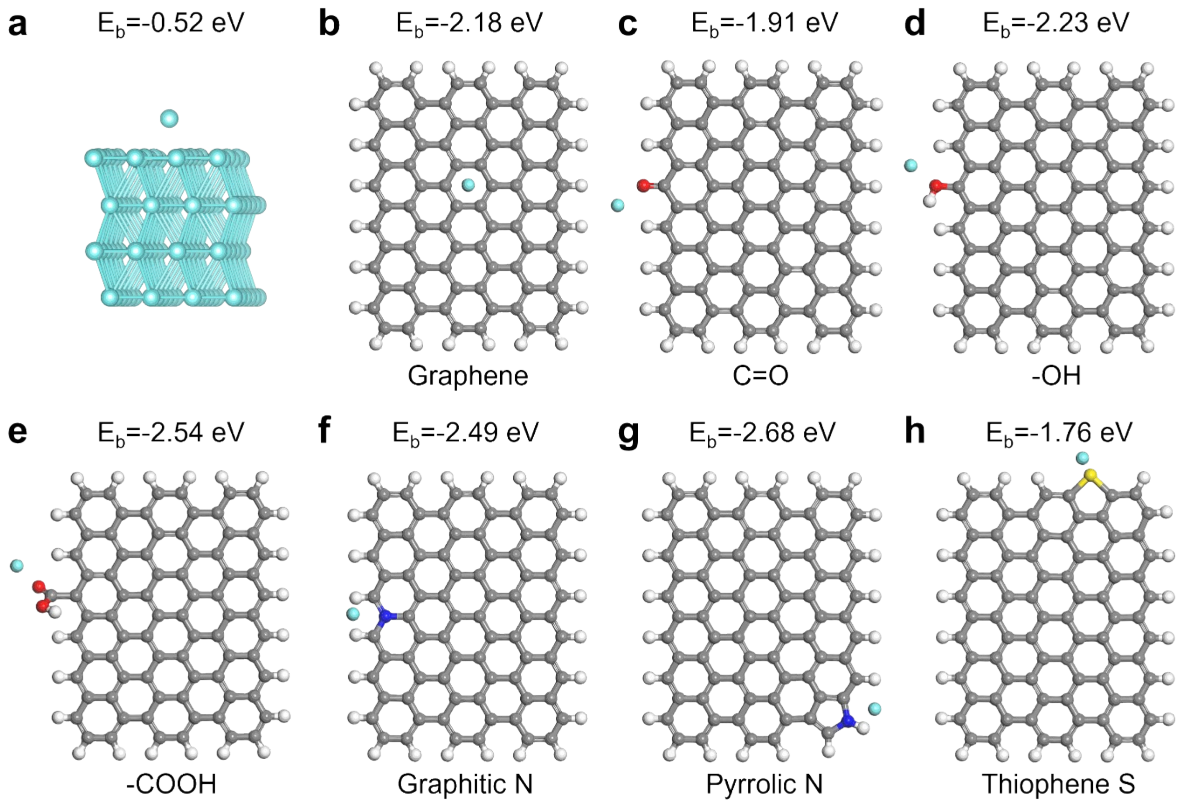

Fig. S14 (**a**) Atomic model structure of binding energy between Zn^{2+} ions and Zn substrate. (**b–h**) Atomic model structure of binding energy between Zn^{2+} ions and graphene quantum dots with different functional groups.

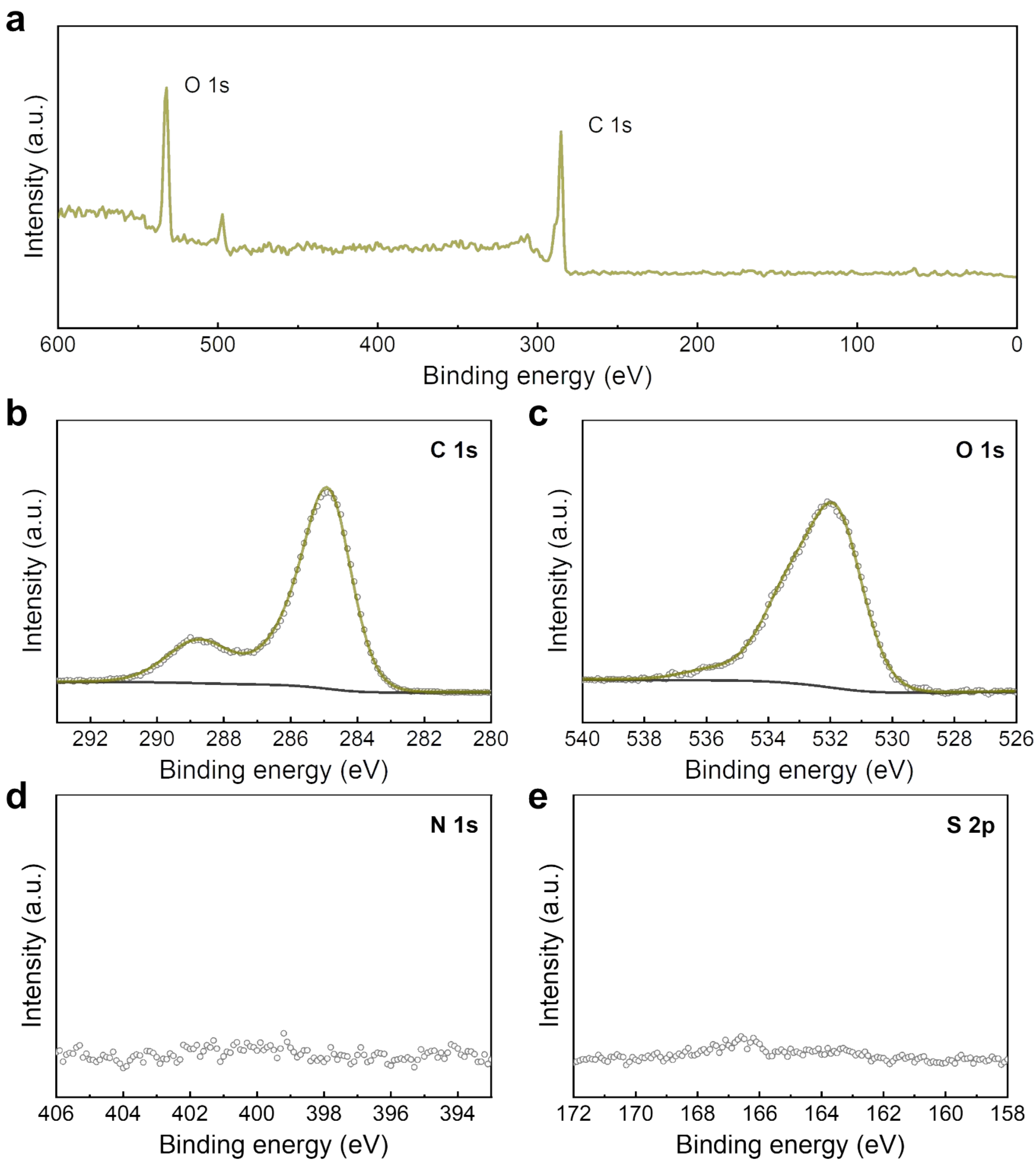

Fig. S15 (**a**) The XPS full spectra of the Undoped-CDs. The XPS high-resolution spectra of (**b**) C 1s, (**c**) O 1s, (**d**) N 1s, and (**e**) S 2p of the Undoped-CDs, respectively.

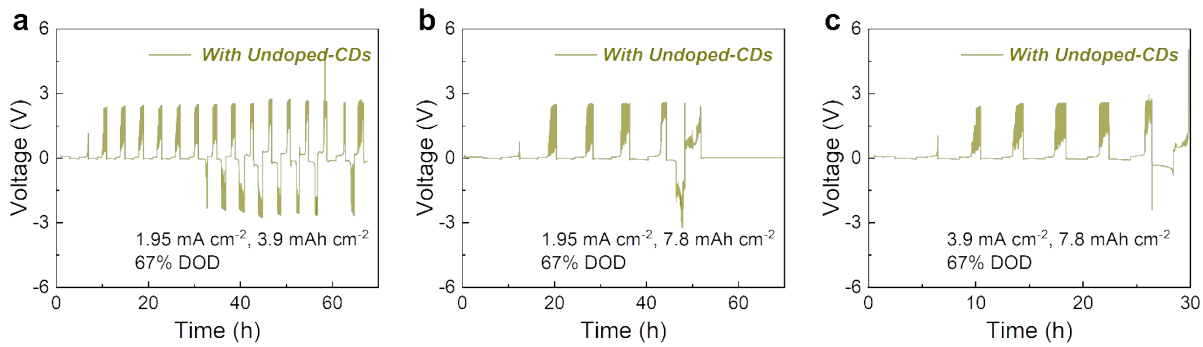

Fig. S16 (**a**) Long-term cycling performance of the Zn||Zn (thickness: 10 μm) symmetric cells using electrolyte with 0.1% Undoped-CDs at 1.95 mA cm^{-2} for 3.9 mAh cm^{-2}. Long-term cycling performance of the Zn||Zn (thickness: 20 μm) symmetric cells using electrolyte with 0.1% Undoped-CDs at (**b**) 1.95 mA cm^{-2} for 7.8 mAh cm^{-2} and (**c**) 3.9 mA cm^{-2} for 7.8 mAh cm^{-2}.

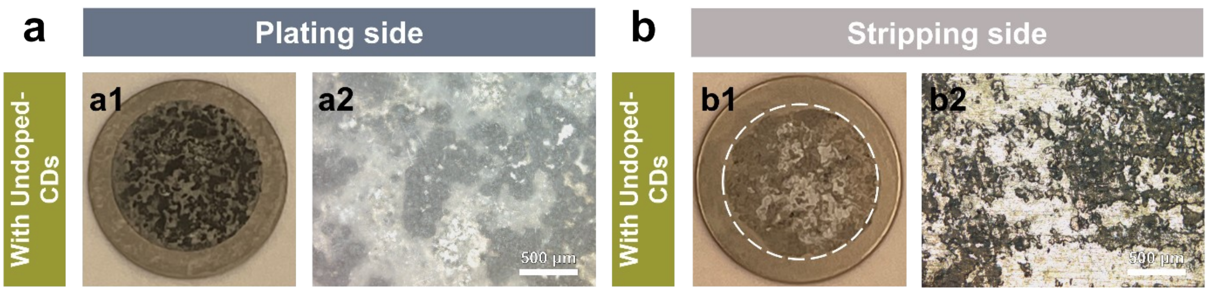

Fig. S17 Digital images and top-view OM images of the Zn foil surface after (**a**) the second plating and (**b**) the second stripping in the Zn||Zn (thickness: 10 μm) symmetric cells using electrolyte with 0.1% Undoped-CDs at 1.95 mA cm^{-2} for 3.9 mAh cm^{-2}.

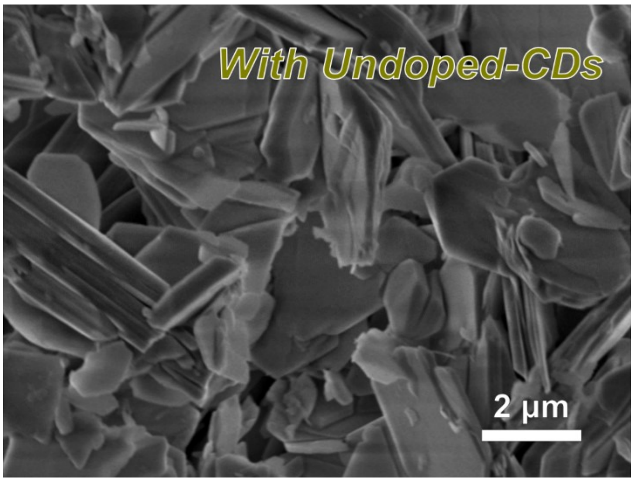

Fig. S18 Top-view SEM images of the cycled Zn anodes using electrolyte with Undoped-CDs.

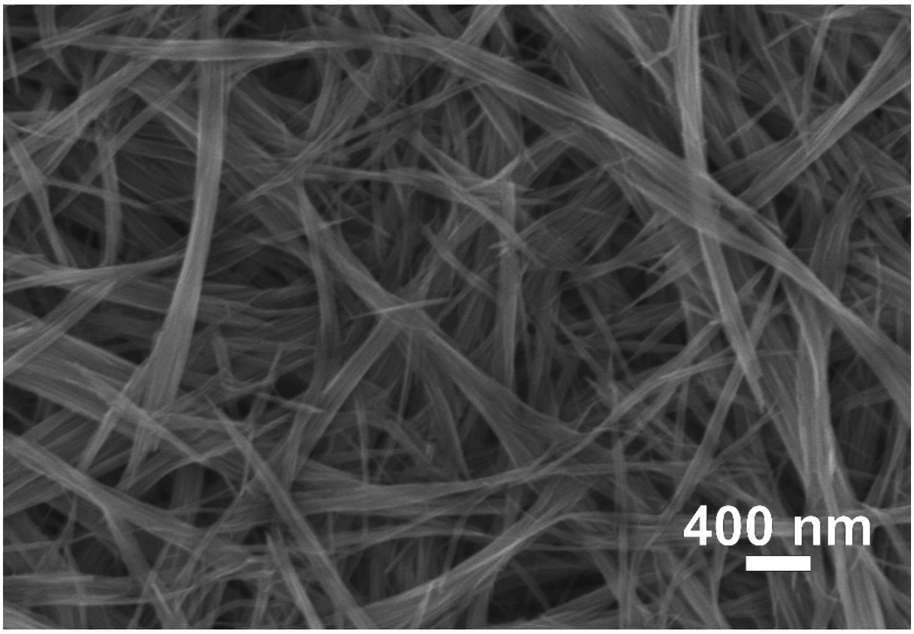

Fig. S19 Top-view SEM image of the NVO nanobelts.

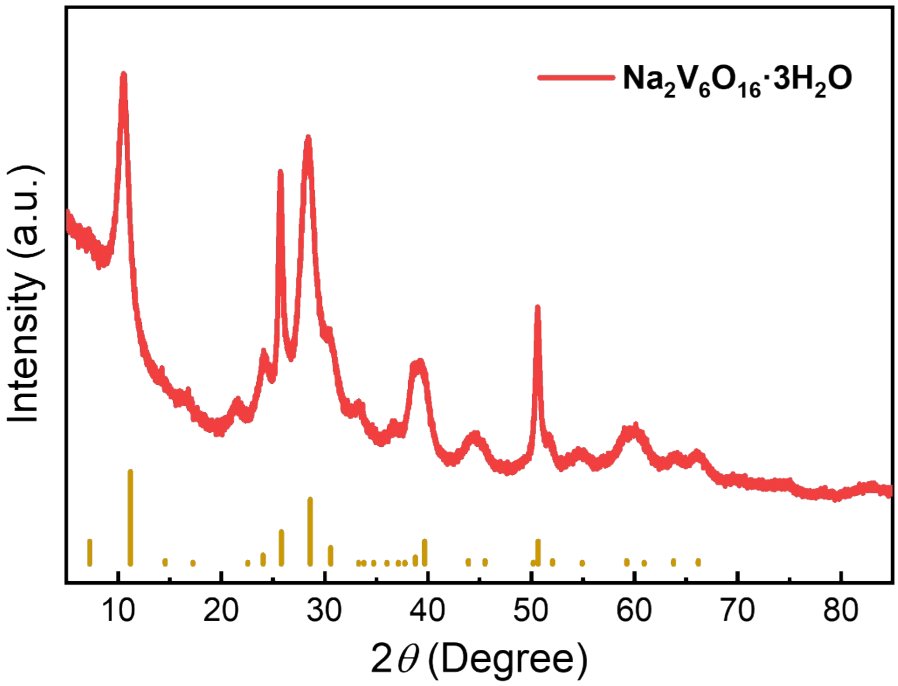

Fig. S20 XRD pattern of the synthesized NVO material.

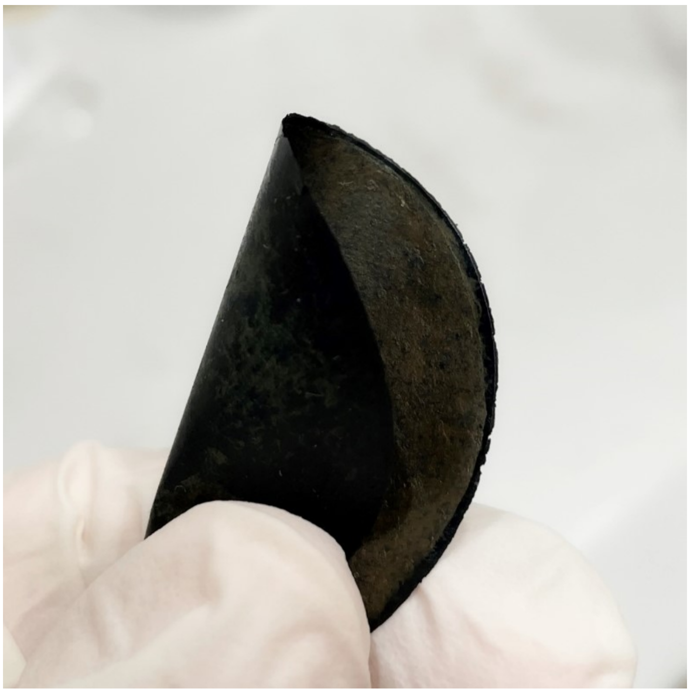

Fig. S21 Digital image of the self-supporting cathode membrane.

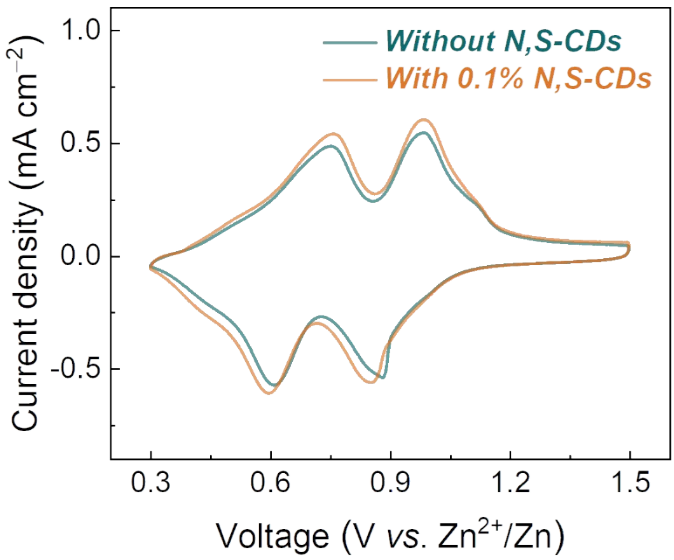

Fig. S22 Cyclic voltammetry (CV) curves of Zn||NVO cells in a voltage range of 0.3 V to 1.5 V at 0.1 mV s^{-1}.

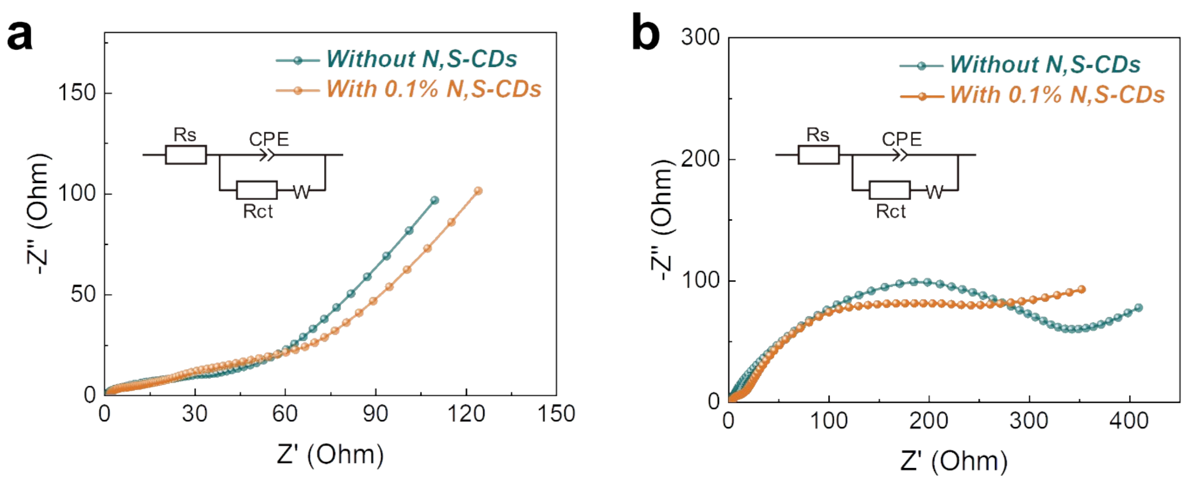

Fig. S23 Electrochemical impedance spectroscopy (EIS) plots of Zn||NVO cells (**a**) before and (**b**) after the CV test.

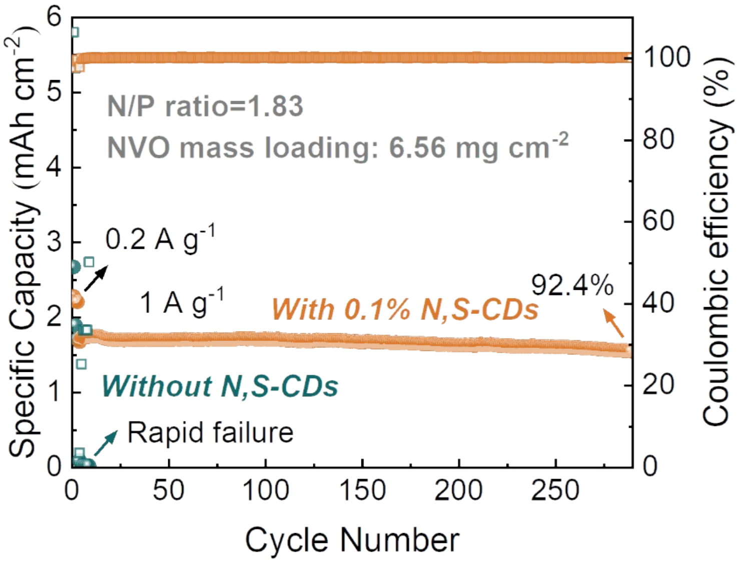

Fig. S24 Cycling performance of the Zn||NVO full cells with low N/P ratio of 1.83.

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