Achieving high-rate and high-capacity Zn metal anodes via a three-in-one carbon protective layer

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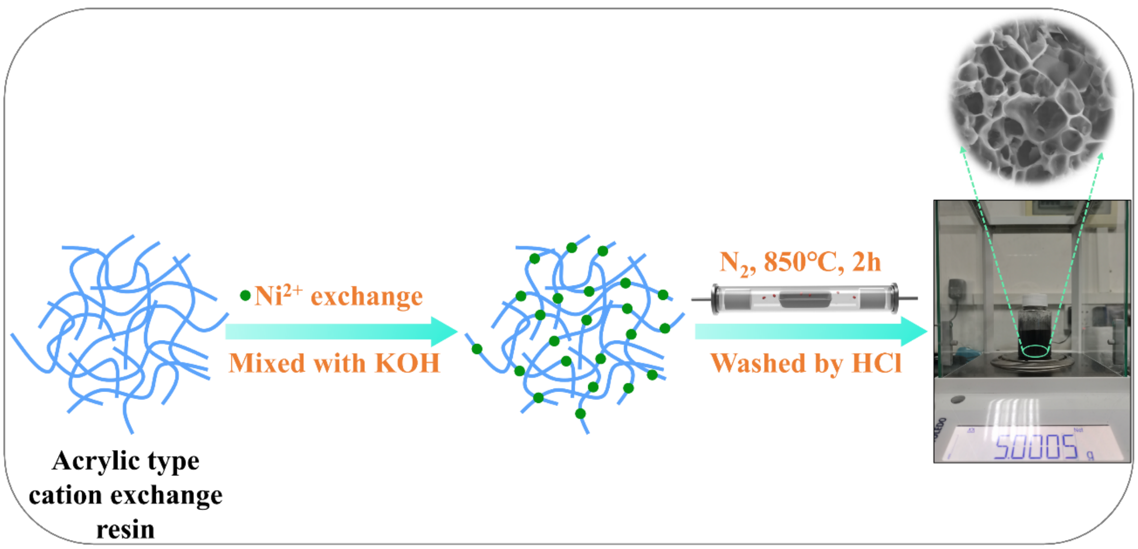

Figure S1. Synthetic process of 3D-NC powder with a 5 g yield per batch.

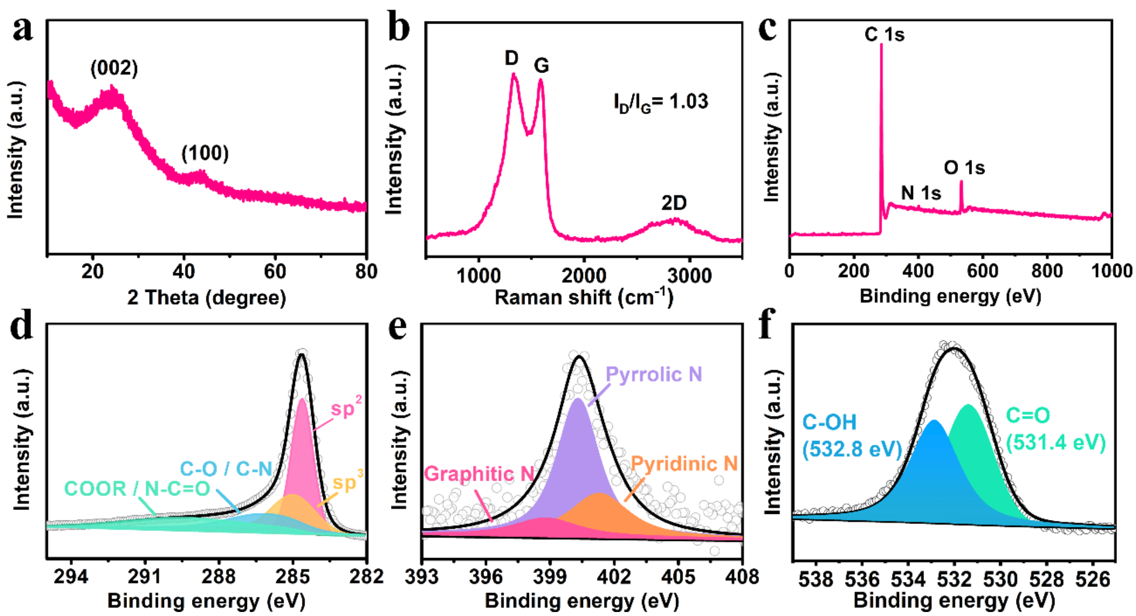

Figure S2. Physical characterization of 3D-NC. a) XRD patterns, b) Raman spectrum, and c) XPS survey spectrum of 3D-NC, XPS spectra of d) C 1s, e) N 1s and f) O1s.

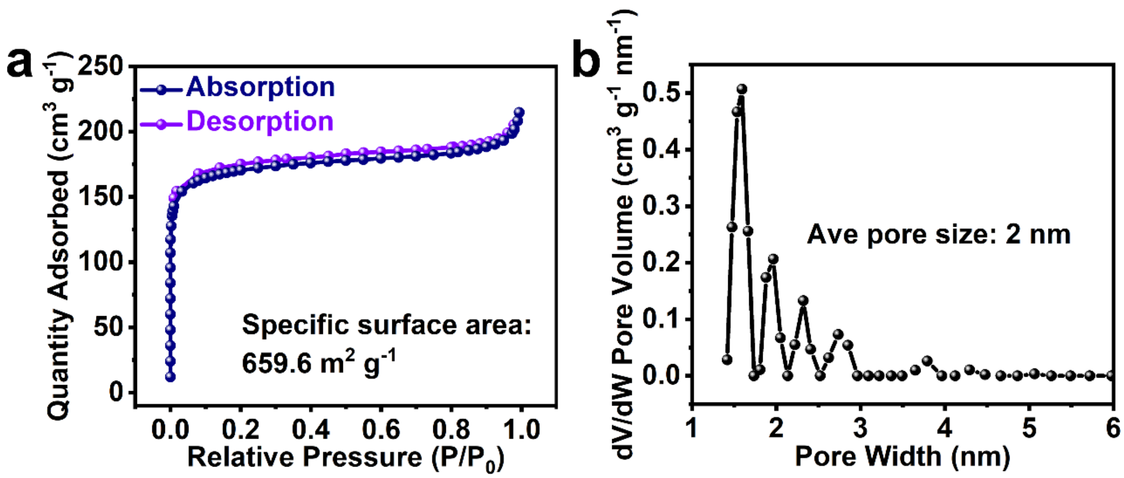

Figure S3. a) Nitrogen adsorption/desorption isotherms and b) Corresponding pore size distribution of the 3D-NC.

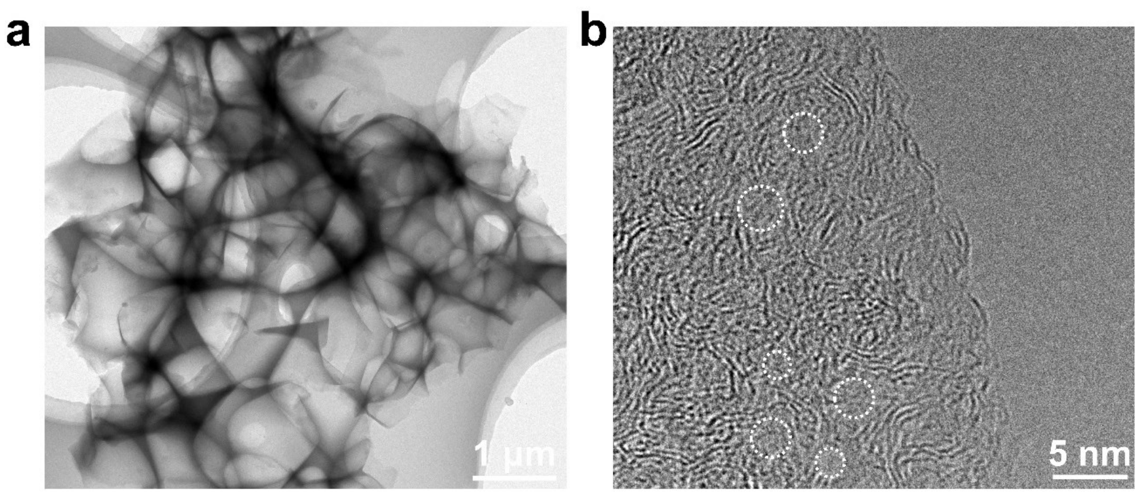

Figure S4. TEM (a) and high-resolution TEM (b) images of 3D-NC.

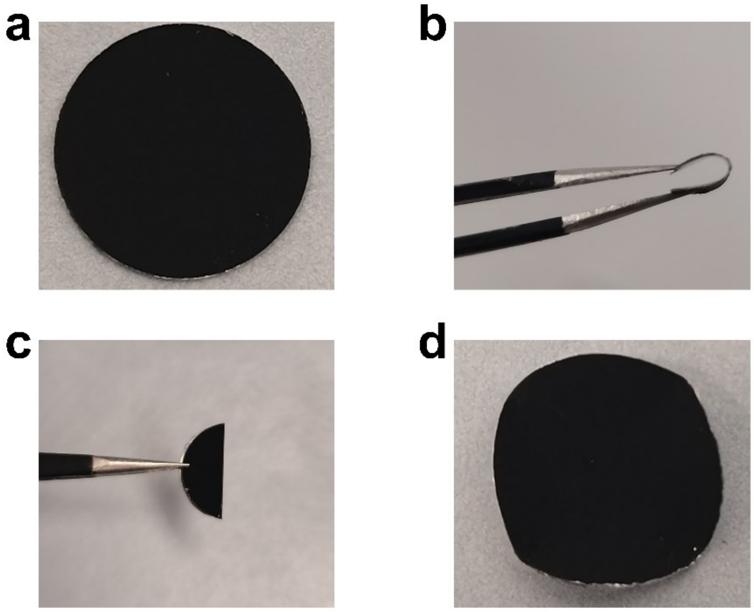

Figure. S5 Optical images of 3D-NC@Zn before (a), after bending (b,c) and repeated bending (d).

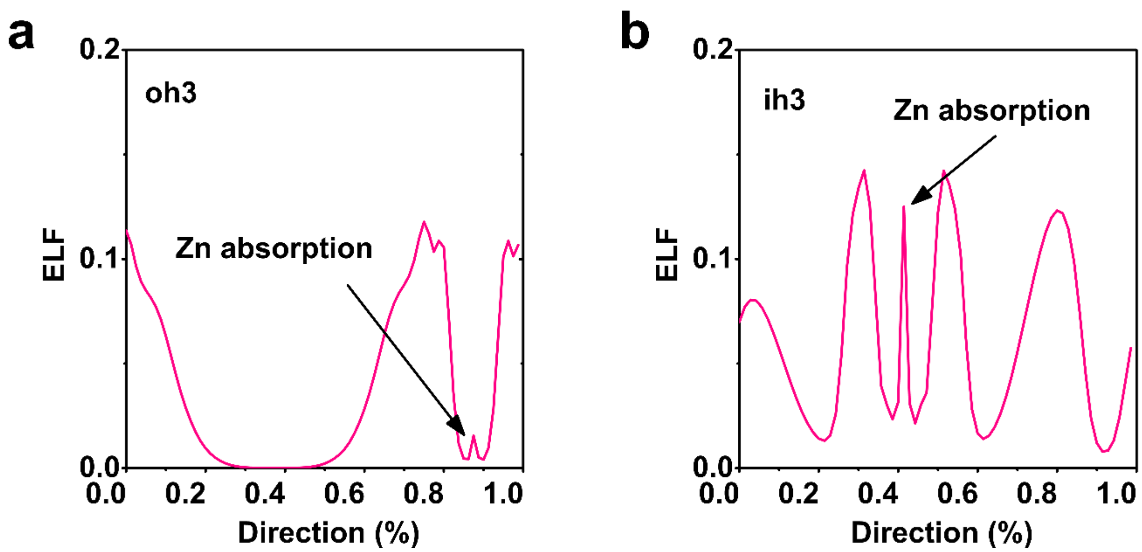

Figure S6. ELF curves of Zn absorbed on different sites of oh3 and ih3.

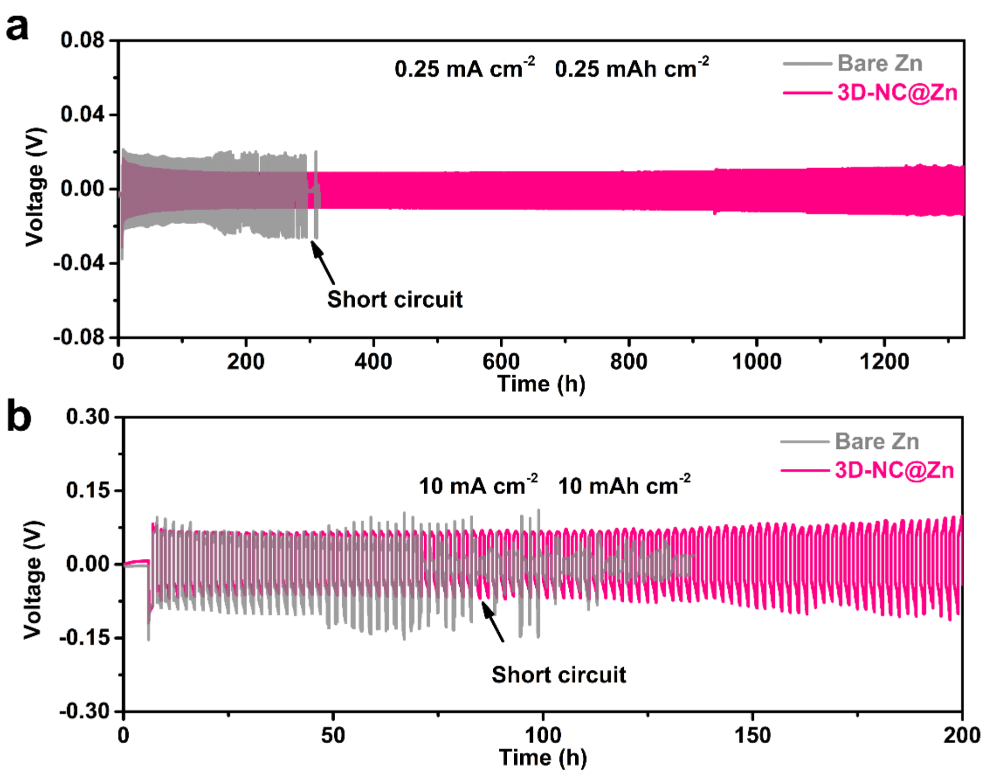

Figure S7. Galvanostatic cycling performance of symmetric cells at a) 0.25 mA cm^{-2}, 0.25 mAh cm^{-2} and b) 10 mA cm^{-2}, 10 mAh cm^{-2}.

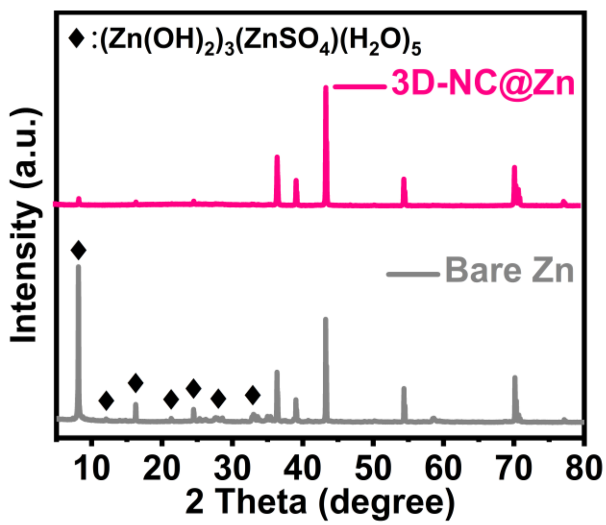

Figure S8. XRD patterns of Zn anode after cycled at 0.25 mA cm^{-2}, 0.25 mAh cm^{-2}.

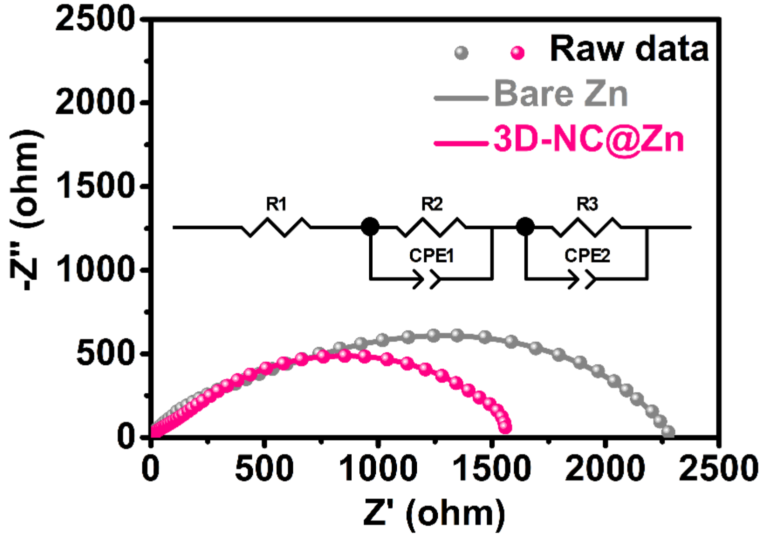

Figure S9. Nyquist plots of symmetric cells after cycled 1000 h at 10 mA cm^{-2}, 1 mAh cm^{-2}.

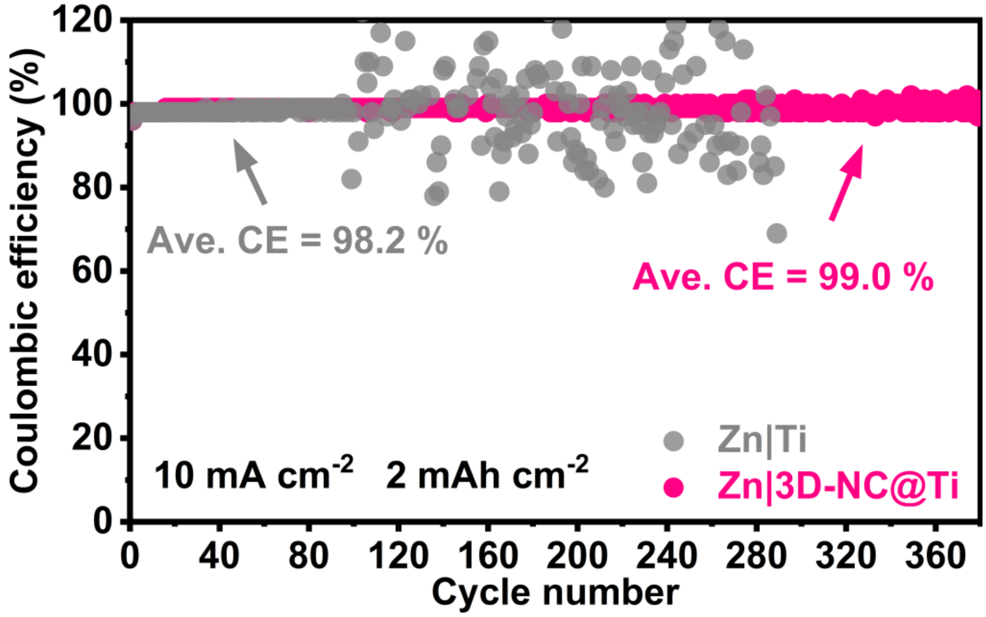

Figure S10. Coulombic efficiency of bare Zn|Ti and Zn|3D-NC@Ti asymmetric cell at 10 mA cm^{-2}, 2 mAh cm^{-2}.

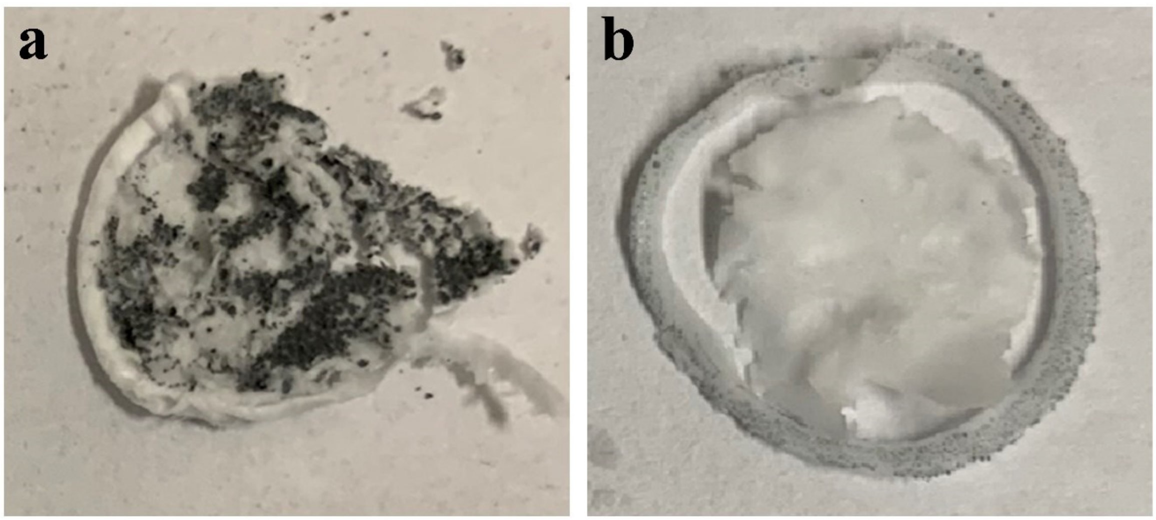

Figure S11. Optical images of the glass fibers recovered from a) bare Zn symmetric cell and b) 3D-NC@Zn after cycled 1000 h at 10 mA cm^{-2}, 1 mAh cm^{-2}.

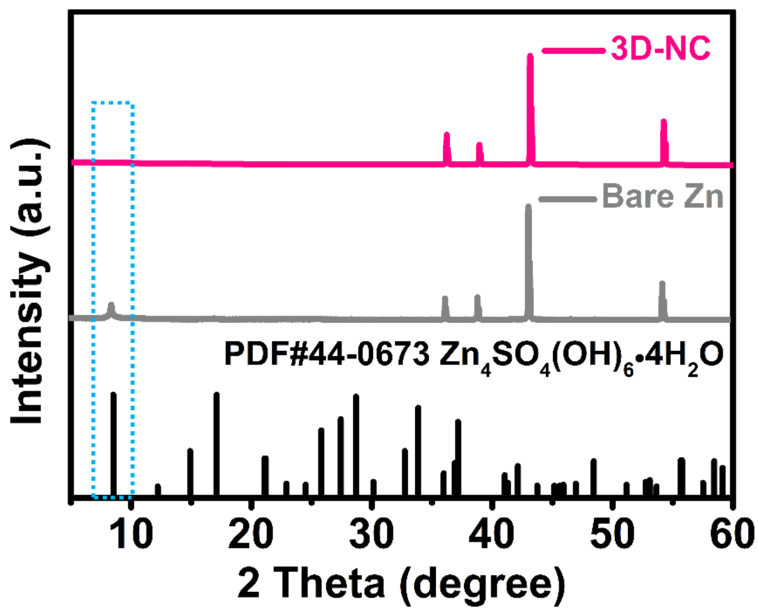

Figure S12. XRD patterns of bare Zn and 3D-NC@Zn after being soaked in the 2 M $ZnSO_4$ electrolyte for 7 days.

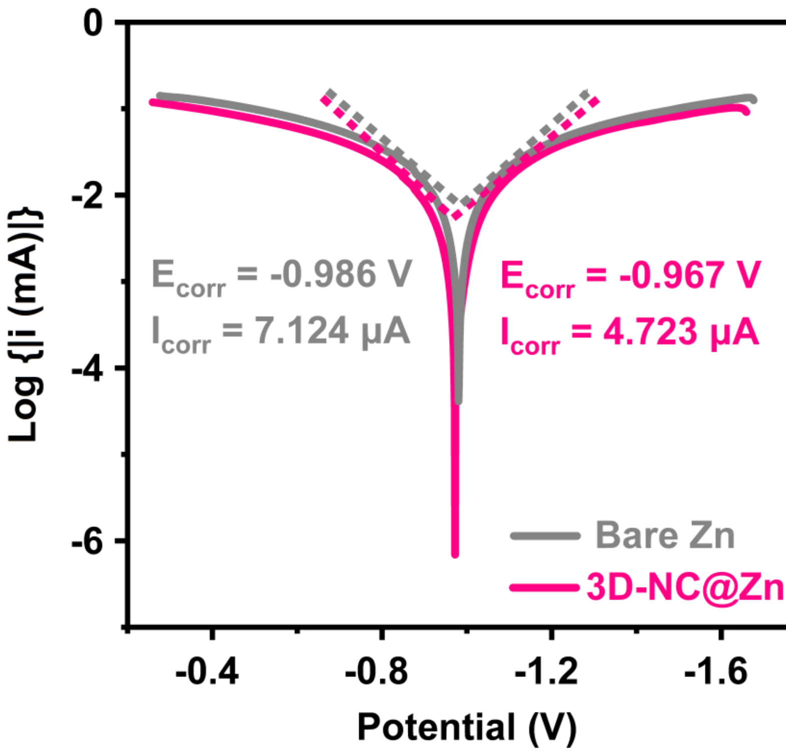

Figure S13. The linear polarization curves of bare Zn and 3D-NC@Zn electrodes.

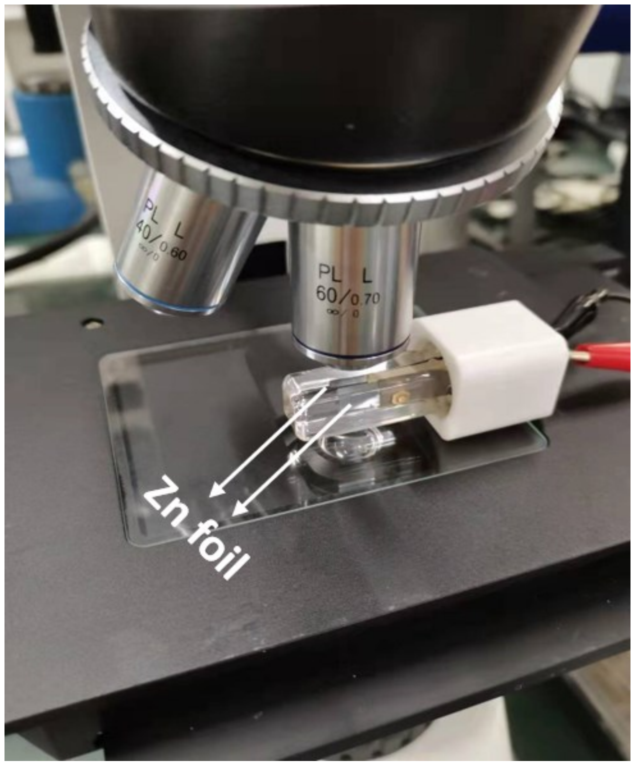

Figure S14. Exhibition of *in-situ* optical microscopy equipped with a transparent electrochemical cell.

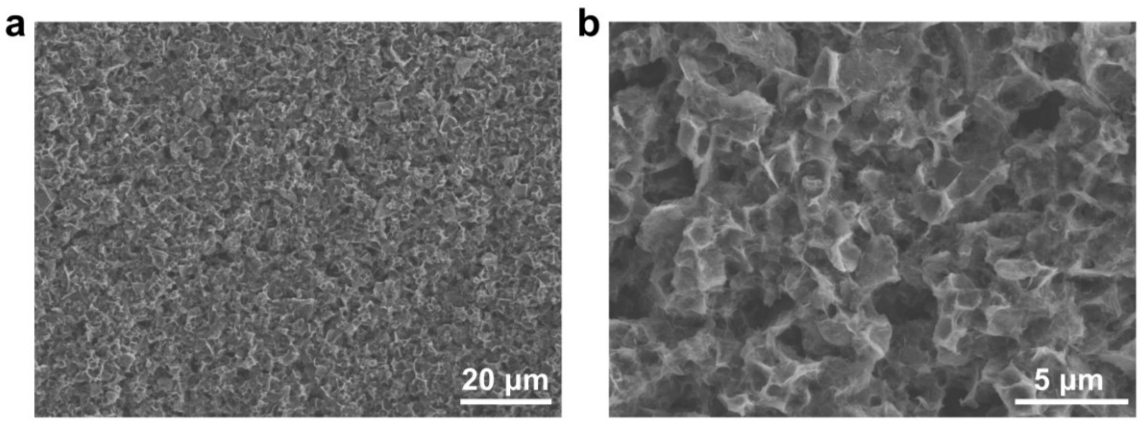

Figure S15. Structure observation of the 3D-NC layer after stripping for 10 mAh cm^{-2} at 1 mA cm^{-2}.

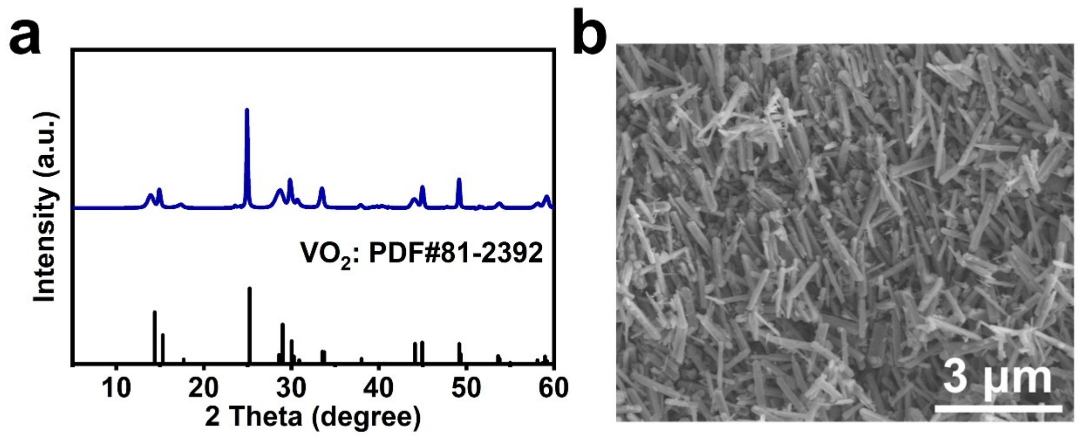

Figure S16. a) XRD patterns, and b) SEM images of the synthesized VO_2.

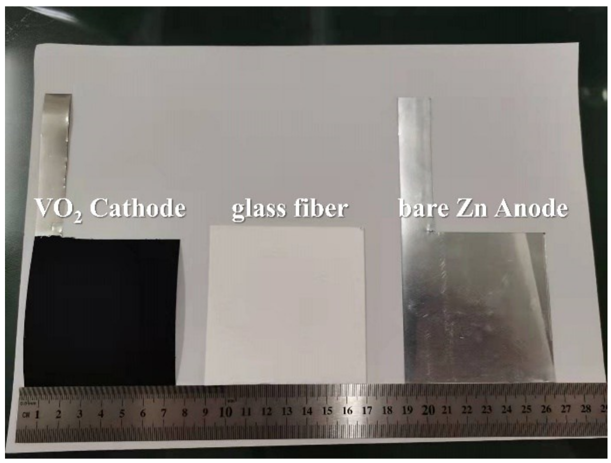

Figure S17. Optical images of the VO_2 cathode, glass fiber and bare Zn anode for the single stack in the bare Zn|VO_2 pouch cell.

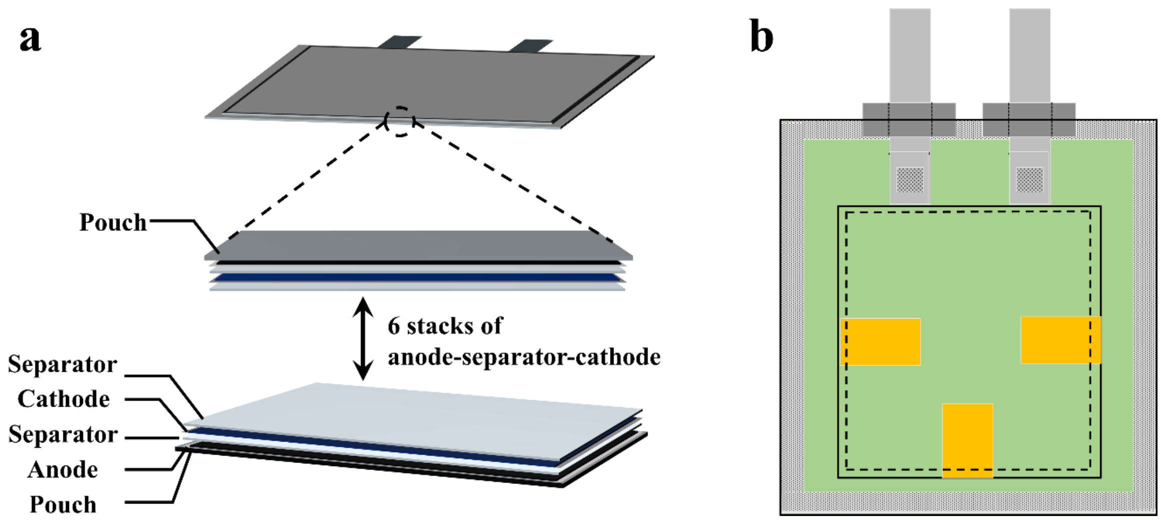

Figure S18. a) Illustration of the internal configuration for pouch cell. b) Schematic of the pouch cell (the green area is under compression stress by a fixture when the battery is cycling).

Table S1. Comparison of the cumulative plating capacity of 3D-NC@Zn with recently reported Zn metal anodes with surface modification.

Electrode	**Current density (mA cm^{-2})**	**Capacity (mAh cm^{-2})**	**Voltage hysteresis (mV)**	**Cycle life (h)**	**Depth of discharge (DOD %)**	**Cumulative plating capacity (mAh cm^{-2})**	**Ref.**
Sn@Zn	1	1	~60	2200	0.68	1100	44
HsGDY @Zn	2	0.1	~140	2400	/	1200	46
ZCO-30@Zn	2	2	~80	1400	/	1400	49
Sb@Zn	3	1	~60	1000	1.71	1500	47
BTO@Zn	5	2.5	~40	1500	14.3	3750	43
ZnSe@Zn	2	1	~100	1500	/	1500	48
SIR@Zn	2	2	~60	3500	0.21	3500	34
BN@Zn	5	2.5	~80	1600	0.43	4000	50
CNG@Zn	1	0.5	84	2956	4.75	1478	45
3D-NC@Zn	**1**	**10**	**36.6**	**1300**	**17.1**	**650**	**This work**
	10	**1**	**125**	**1300**	**1.71**	**6500**	

Table S2. Fitted EIS results of Zn|Zn symmetric cells before cycled.

Electrode	R_{SEI} (Ω)	R_{ct} (Ω)
Bare Zn	250	788
3D-NC@Zn	18.1	439.9

Table S3. Fitted EIS results of Zn|Zn symmetric cells after cycled 1000 h at 10 mA cm^{-2}, 1 mAh cm^{-2}.

Electrode	R_{SEI} (Ω)	R_{ct} (Ω)
Bare Zn	450.9	1841
3D-NC@Zn	90.8	1519

Table S4. Fitted corrosion parameters results from LSV curve.

Electrode	E_{corr} (V)	I_{corr} (μA)
Bare Zn	-0.986	7.124
3D-NC@Zn	-0.967	4.723

Table S5. Comparison of DOD values of reported Zn metal in full cells.

Anode	Cathode	Current density (A g^{-1})	DOD of Zn (%)	Life cycle	Ref
Ag@Zn	MnO_2	1	0.13	4000	15
Al_2O_3@Zn	MnO_2	1	0.37	1000	13
In@Zn	AC	2	0.24	5000	58
N@Zn	MnO_2	1	0.079	2000	59
Cu-Zn@Zn	MnO_2	3.08	0.18	500	60
C-750@Zn	MnO_2	0.3	0.33	5000	61
SIR@Zn	VS_2	5	0.25	1000	34
3D-NC@Zn	**VO_2**	**1**	**0.42**	**1000**	**This work**

Table S6. The specifications of the Zn|VO_2 punch cell with a designed capacity of 1.12 Ah

	Specification	Value	Units	Notes
Cathode (VO_2@Ti foil)	Reversible capacity	244	mAh g^{-1}	at 0.1 Ag^{-1}
	Voltage	0.8	V	*vs*. Zn^{2+}/Zn
	Active material ratio	70%	/	VO_2: Carbon black: PVDF = 7: 2: 1
	Electrode thickness	180	μm	double sides
	Mass loading	20	mg cm^{-2}	double sides
	Number of stacks	6	/	/
	Electrode area	54.76	cm^2	7.4 cm × 7.4 cm
	Area capacity	4.10	mAh cm^{-2}	double sides
	Total capacity	1122.4	mAh	double sides
	VO_2 powder amount	5.06	g	10% excess for slurry making in case of waste
Anode (Zn foil)	Theoretical capacity	820	mAh g^{-1}	/
	Voltage	0	V	*vs*. Zn^{2+}/Zn
	Thickness	100	μm	/
	Number of stacks	6	/	/
	Electrode area	57.76	cm^2	7.6 cm × 7.6 cm
Separator (glass fiber)	Thickness	300	μm	Whatman, GF/D
	Number of stacks	7	/	/
	Area	64	cm^2	8.0 cm × 8.0 cm
Electrolyte (2M $ZnSO_4$)	Volume	11	mL	~10 mL/Ah, flooded