

## Supporting Information

### **Zincophilic Sn Sites Induced the Local Ion Enrichment for the Compact and Homogenous Zn Biscuit Growth in the Long-Life Zn Metal Batteries**

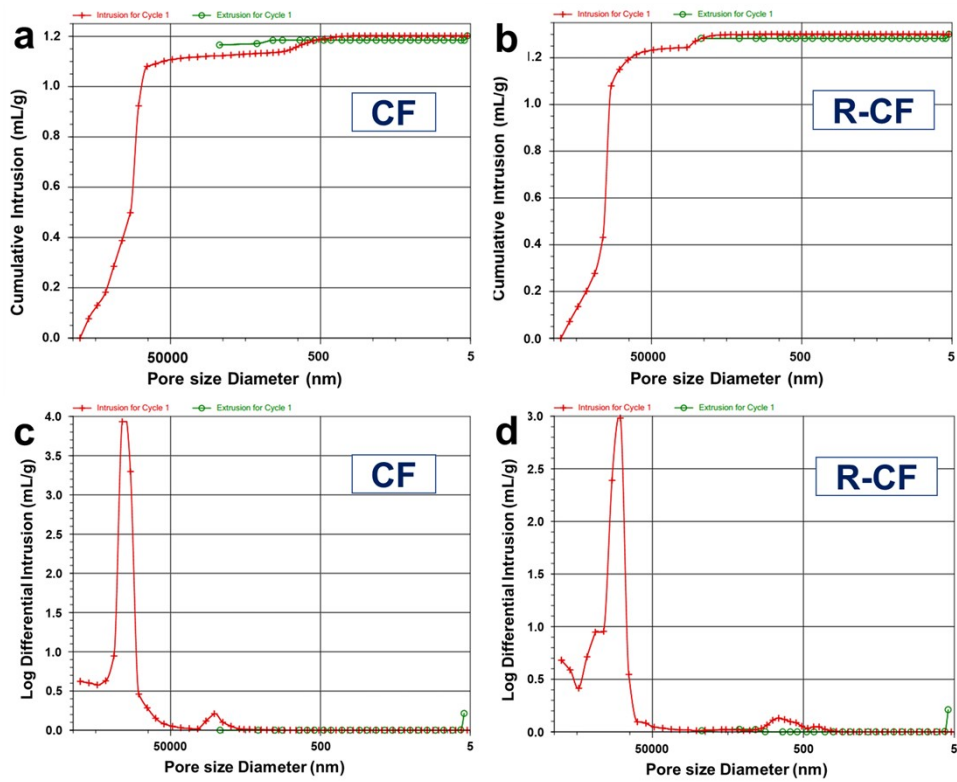
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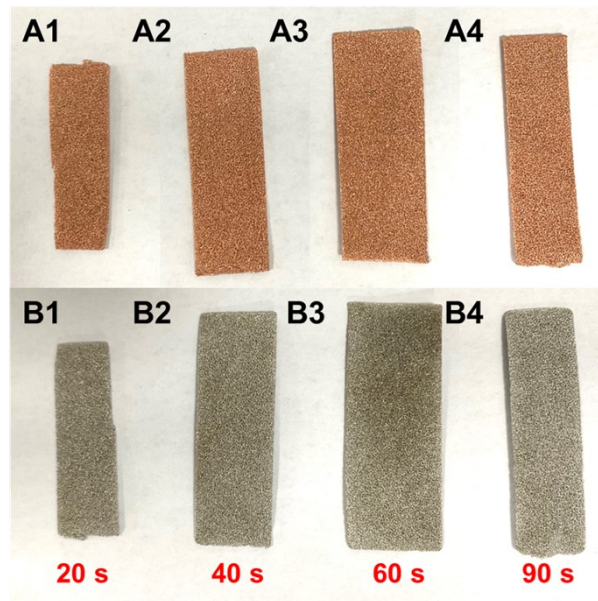
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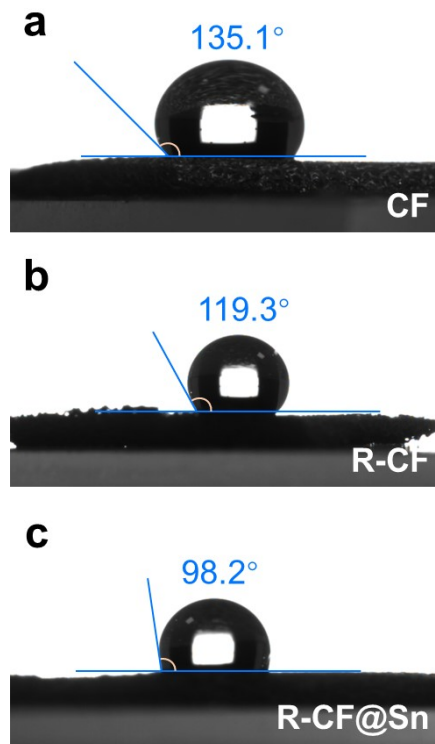
Email address: [liutc@usx.edu.cn](mailto:liutc@usx.edu.cn); [yongwang@shu.edu.cn](mailto:yongwang@shu.edu.cn); [zhixin.yu@usx.edu.cn](mailto:zhixin.yu@usx.edu.cn)



**Fig. S1** Intrusion and extrusion plot and pore size distribution of (a, c) bare CF and (b, d) R-CF substrate.



**Fig. S2** Optical images of R-CF (A1-A4) and related skeletons with Sn decoration at different immersion time (B1-B4).



**Fig. S3** Experiments about contact angle test on CF, R-CF and R-CF@Sn substrates.

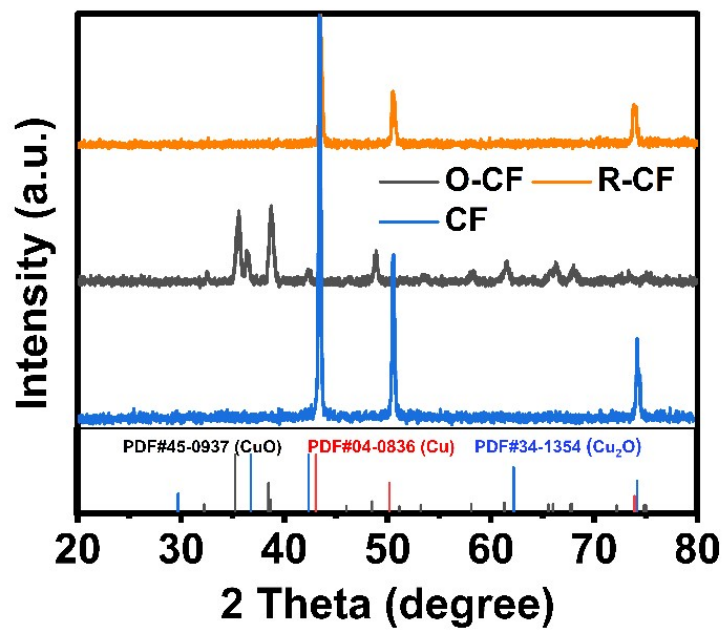
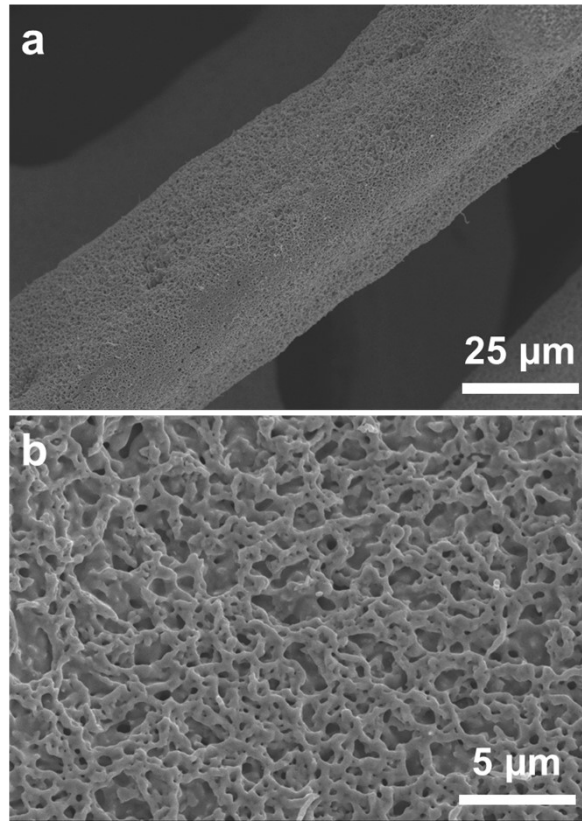
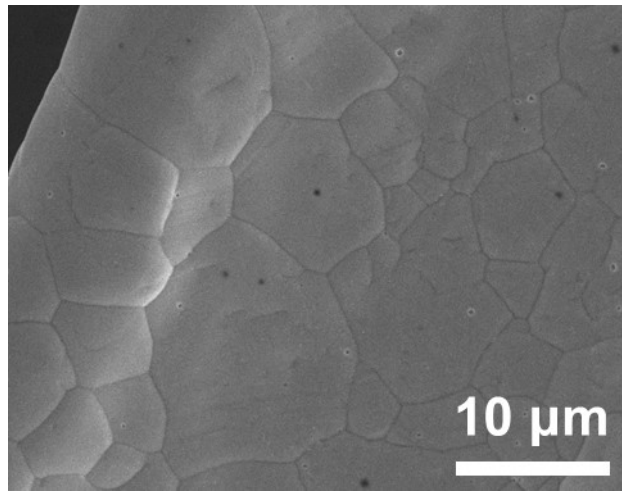


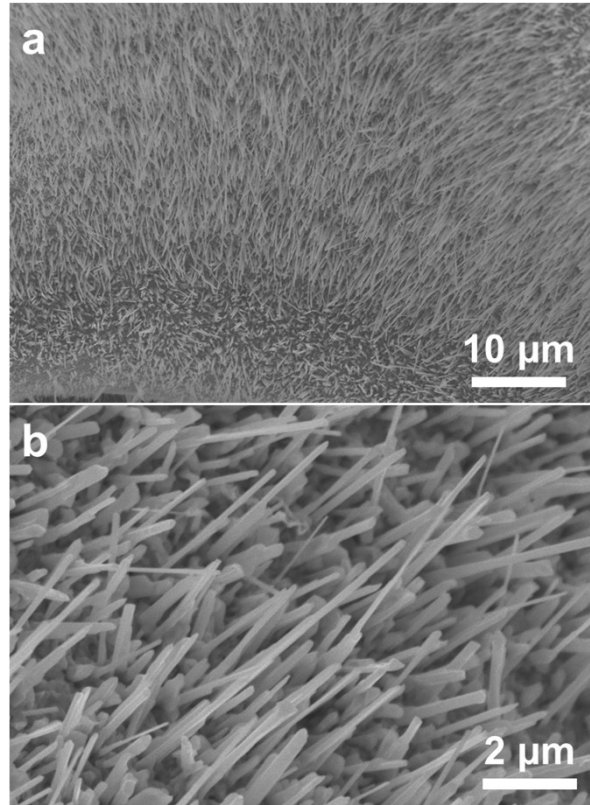
Fig. S4 XRD patterns of CF, O-CF and R-CF skeleton.



**Fig. S5** SEM image of obtained R-CF skeleton.

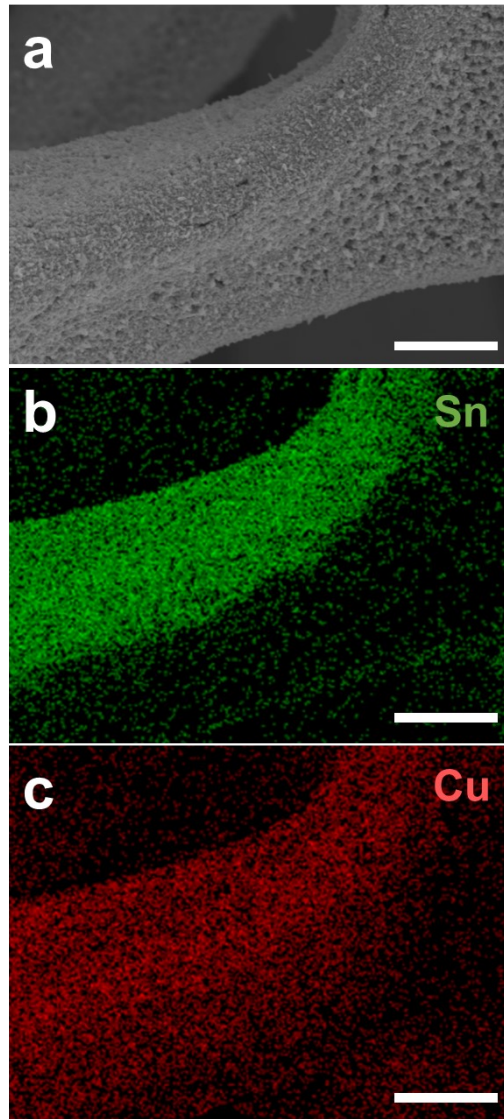


**Fig. S6** SEM image of cleaned CF skeleton.

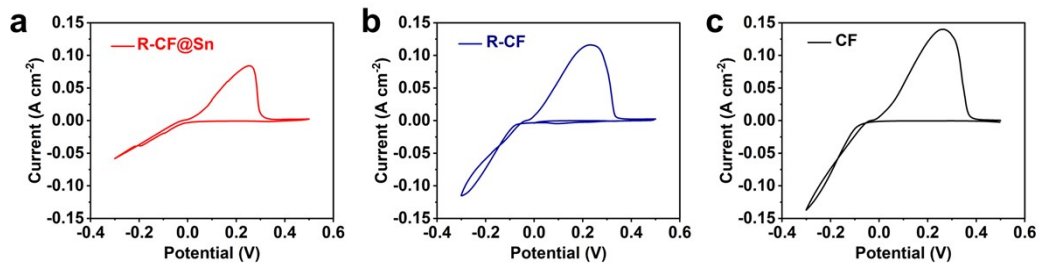


**Fig. S7** SEM image of irregular O-CF with numerous nanoneedles.

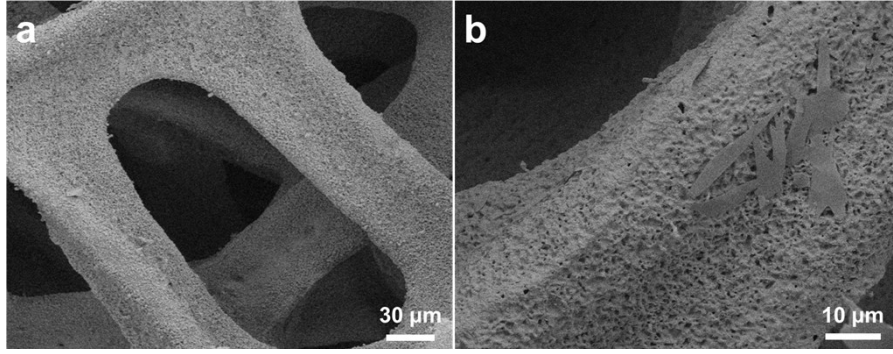




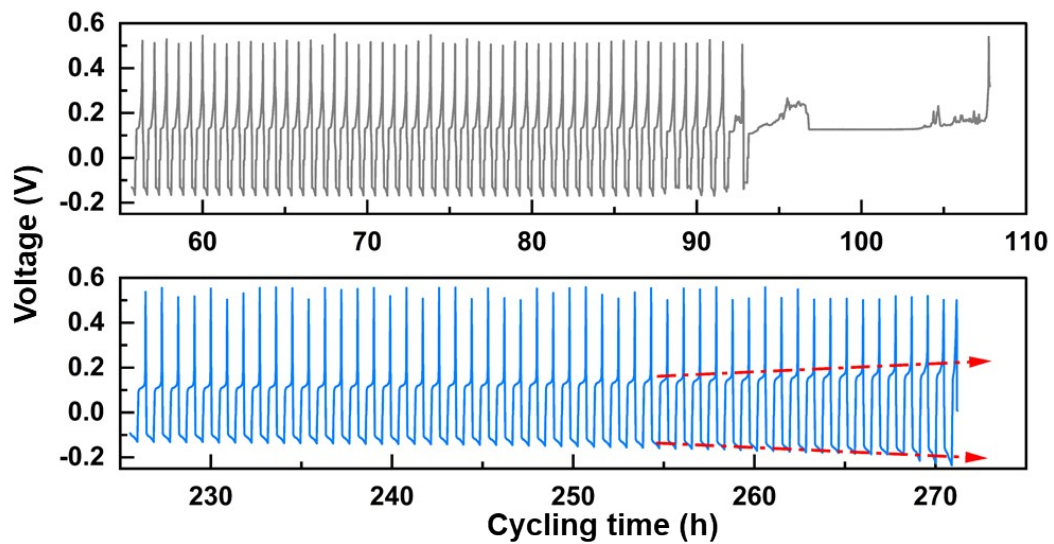
**Fig. S8** (a) SEM image of R-CF@Sn skeleton and related element mappings of (b) Sn and (c) Cu. Each bar is 25  $\mu\text{m}$ .



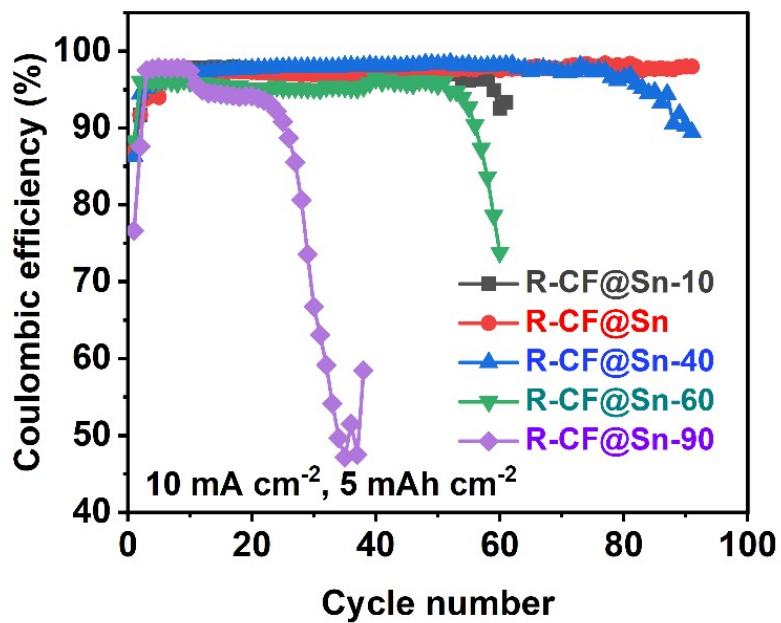
**Fig. S9** Cyclic voltammety curves of different electrodes.



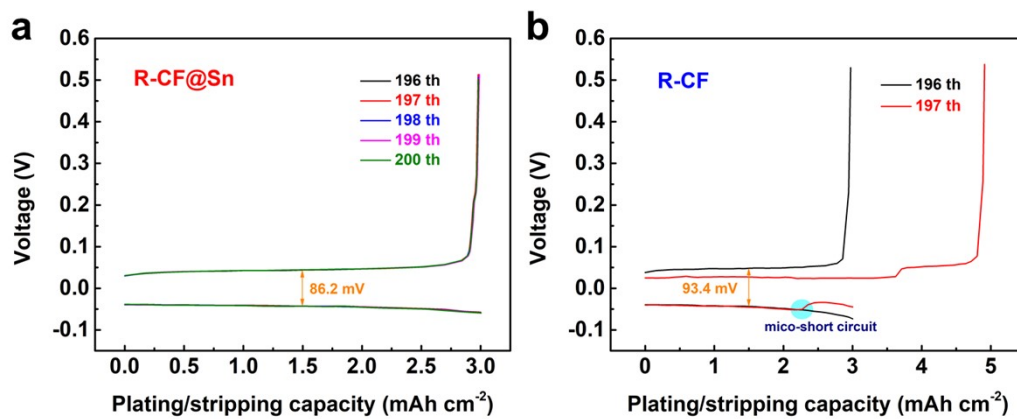
**Fig. S10** The morphology of the R-CF@Sn electrode after 300 cycles at the condition of  $5 \text{ mA cm}^{-2}$  and  $1 \text{ mAh cm}^{-2}$ .



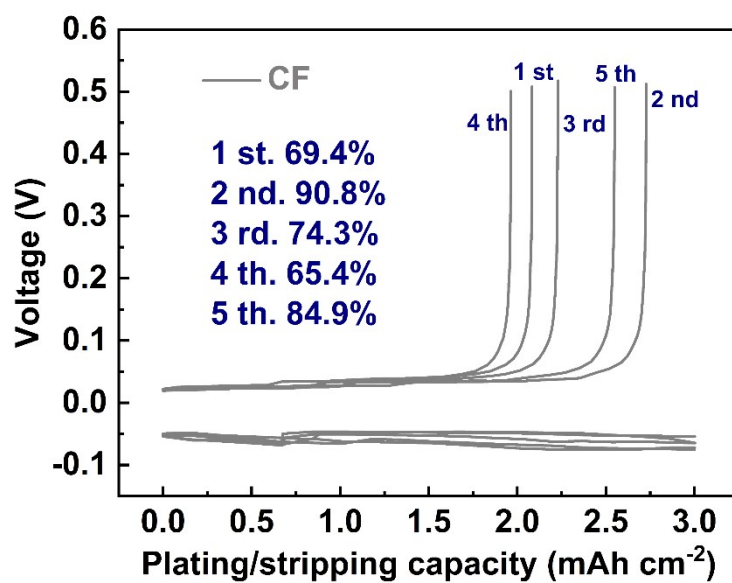
**Fig. S11** Time-voltage plots at the last 50 cycles of CF (grey color) and R-CF skeletons (blue color).



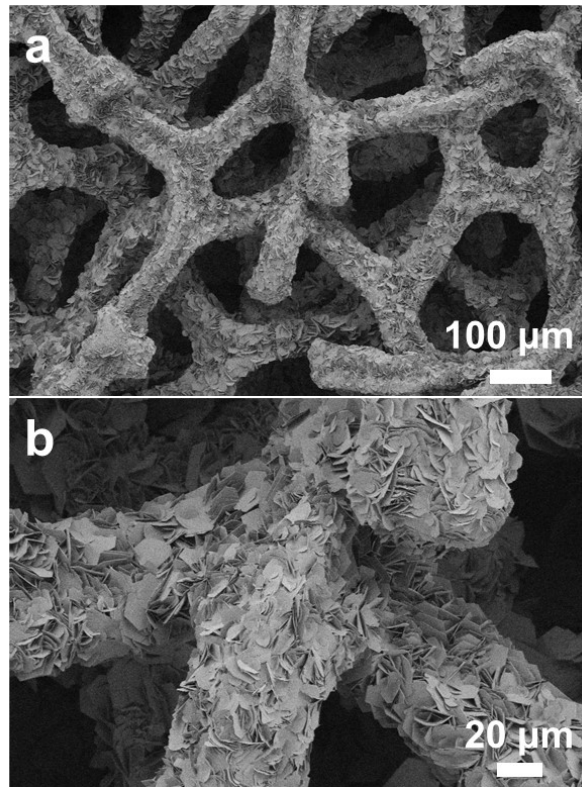
**Fig. S12** Coulombic efficiency comparison of various skeletons containing zincophilic Sn decoration under the reaction time of 10, 20, 40, 60 and 90 s.



**Fig. S13** Voltage-capacity plots of (a) R-CF@Sn (from 196<sup>th</sup> to 200<sup>th</sup> cycle) and (b) R-CF (196<sup>th</sup> and 197<sup>th</sup> cycle).

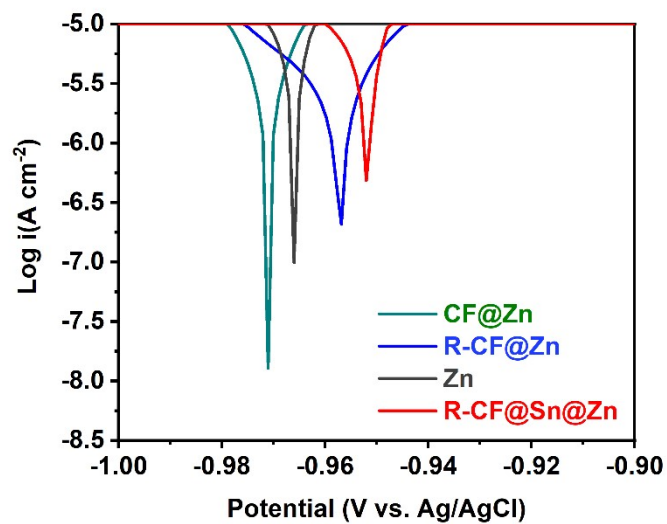


**Fig. S14** Related voltage-capacity plots of bare CF at last 5 cycles when cycled at 10 mA cm<sup>-2</sup> with 3 mAh cm<sup>-2</sup>.

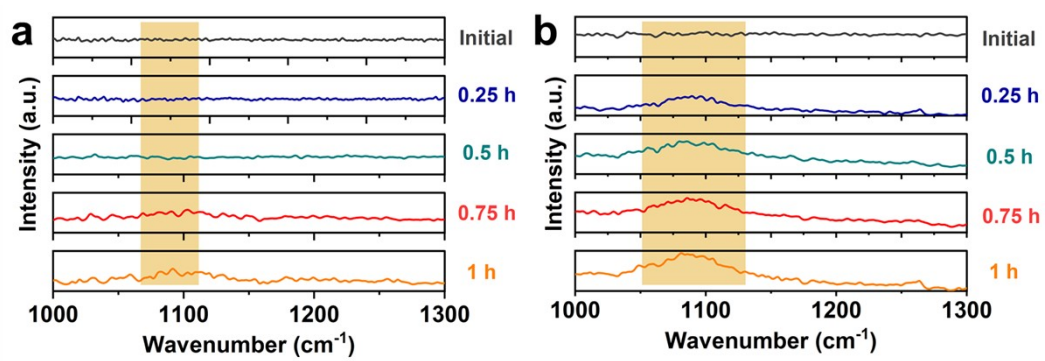


**Fig. S15** SEM images of (a-b) R-CF loading deposited Zn metal of  $3 \text{ mAh cm}^{-2}$ .

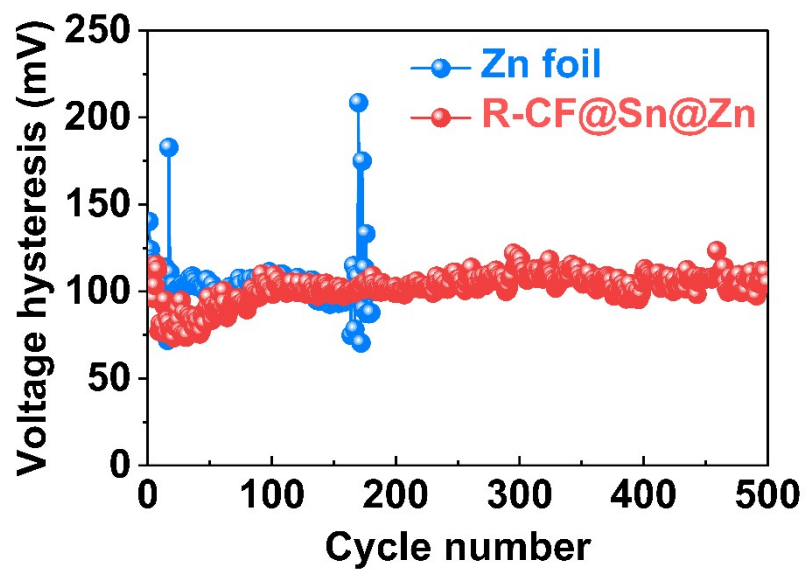




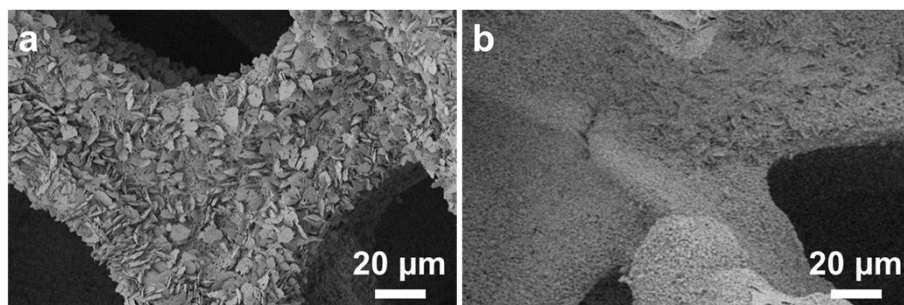
**Fig. S16** Tafel plots of different electrodes in a three-electrode system.



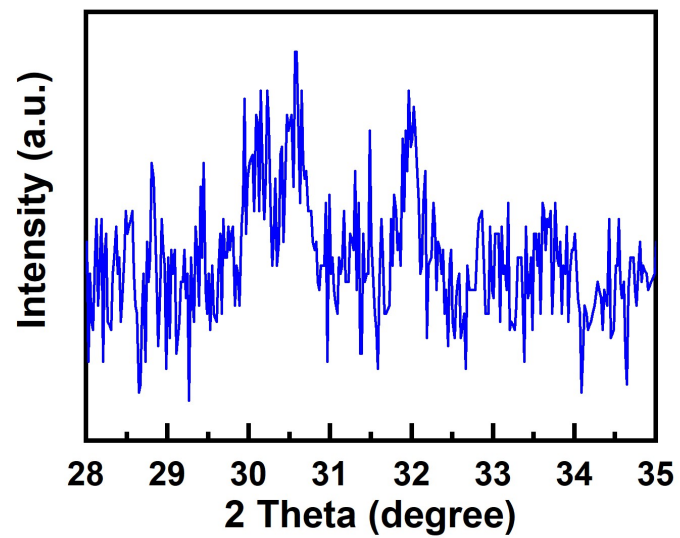
**Fig. S17** *In situ* FTIR spectra of continuous Zn plating on (a) R-CF@Sn and (b) bare CF substrate at the deposition current density of 3 mA cm<sup>-2</sup>.



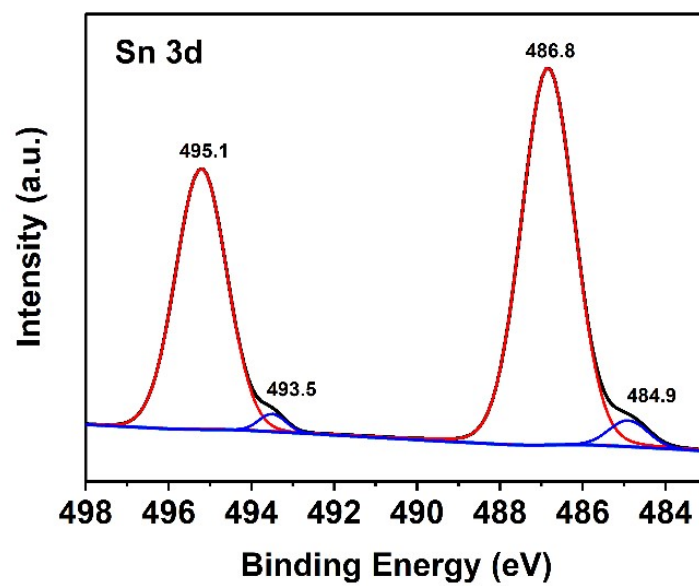
**Fig. S18** Results of voltage hysteresis of two cells based on Zn foil and R-CF@Sn@Zn electrodes.



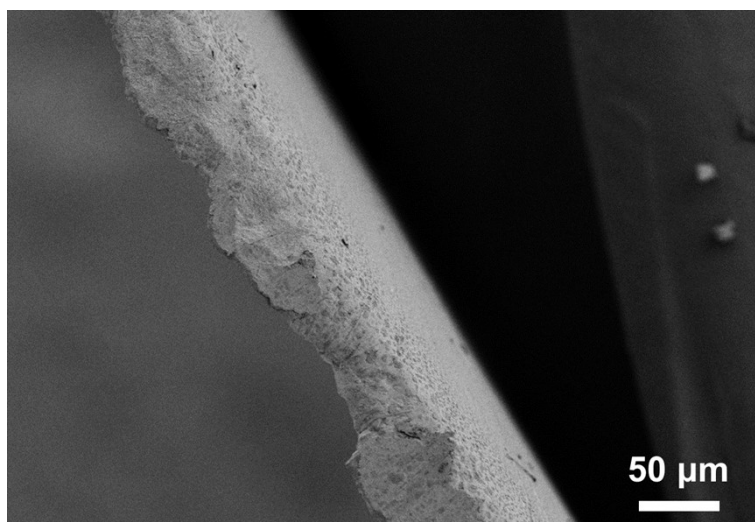
**Fig. S19** SEM images of R-CF@Sn@Zn electrodes after stripping Zn deposition of (a) 1 and (b) 3 mAh cm<sup>-2</sup>.



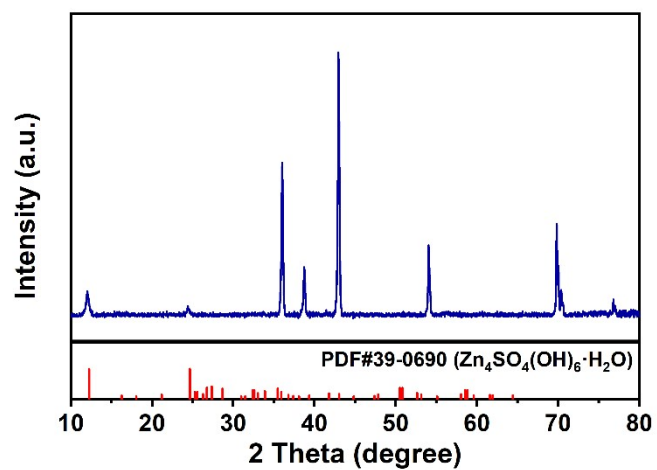
**Fig. S20** XRD pattern of R-CF@Sn@Zn electrode in a symmetric cell after 100 cycles.



**Fig. S21** High-resolution XPS of Sn 3d of cycled R-CF@Sn@Zn electrode.

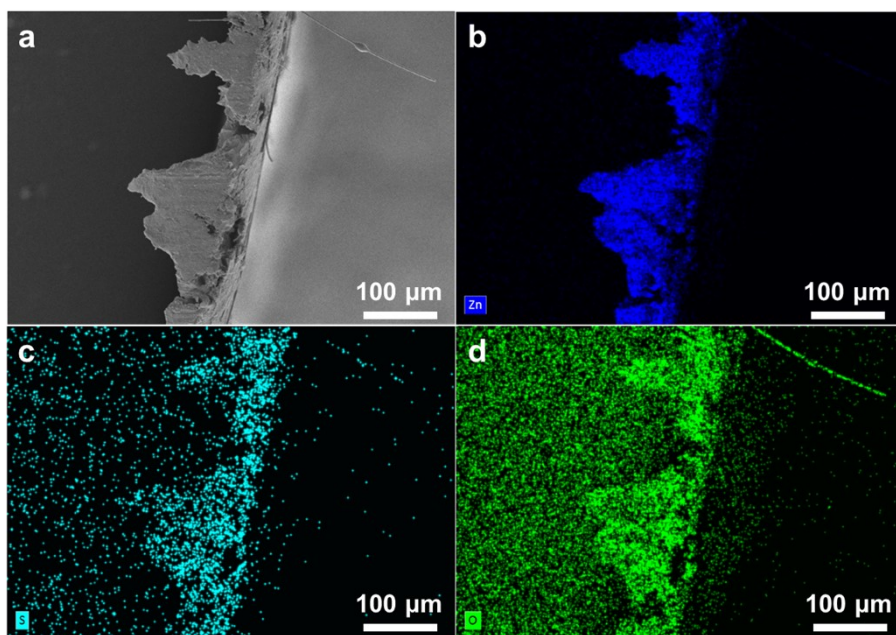


**Fig. S22** Cross-sectional SEM image of bare Zn foil.

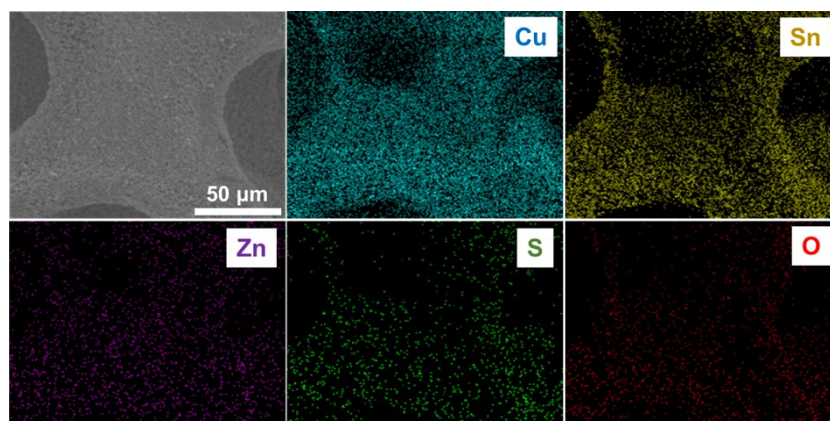


**Fig. S23** XRD pattern of cycled Zn electrode.

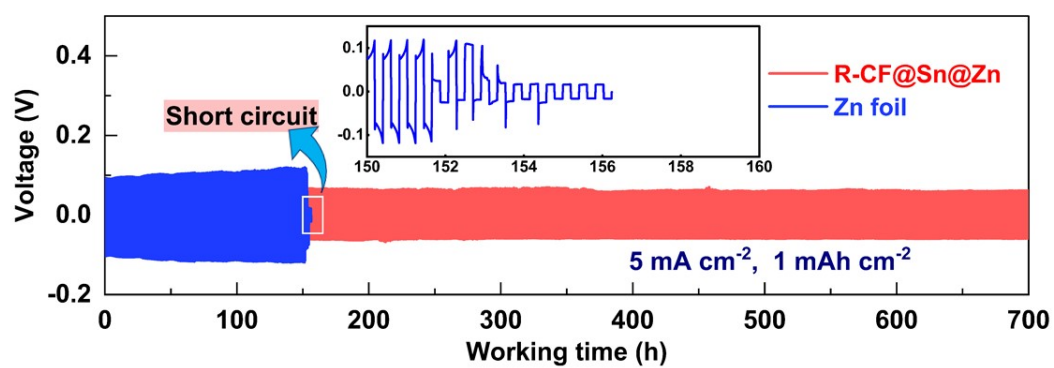




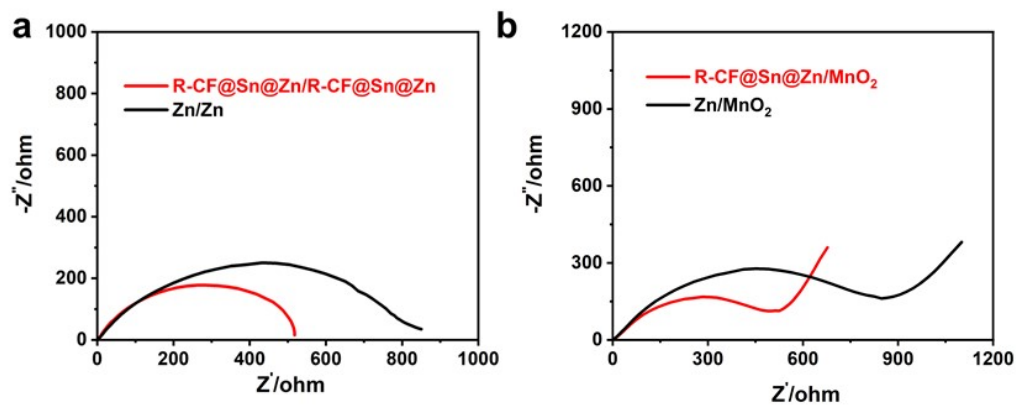
**Fig. S24** (a) Cross-sectional SEM image of Zn electrode after 100 cycles and corresponding element mappings of (b) Zn, (c) S and (d) O components.



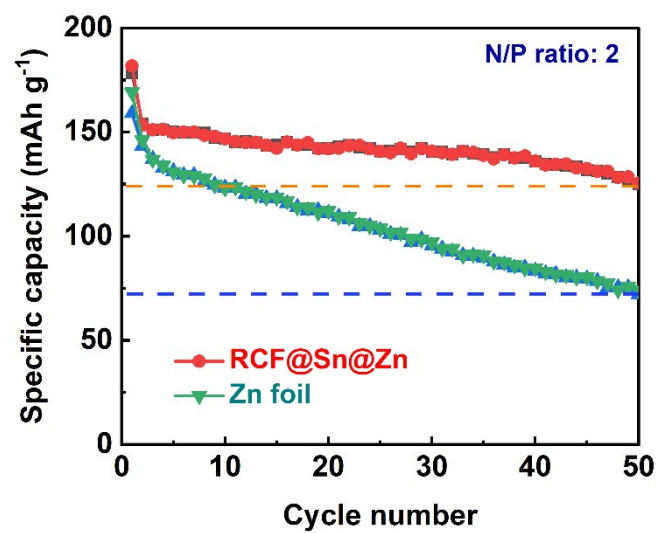
**Fig. S25** SEM image of R-CF@Sn electrode after 100 cycles and related element mappings of Cu, Sn, Zn, S and O.



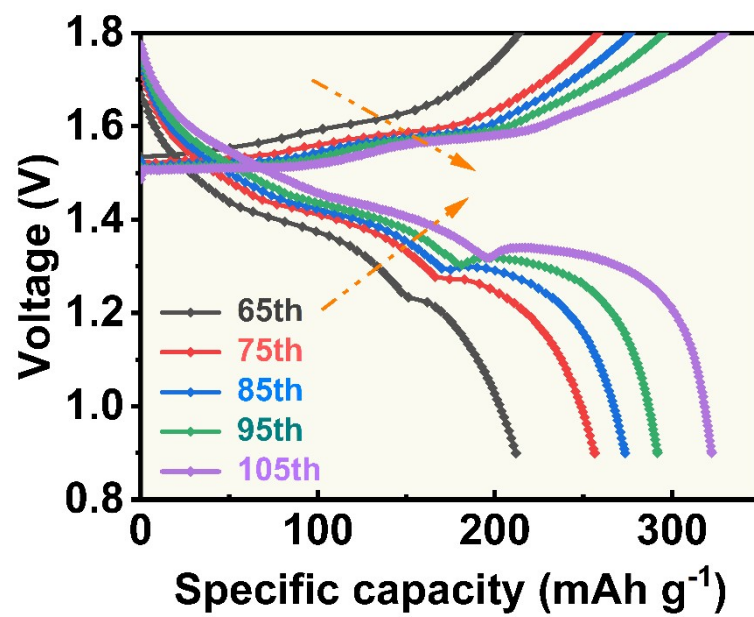
**Fig. S26** Galvanostatic charge-discharge profiles of symmetric cells with R-CF@Sn@Zn and Zn foil at 5 mA cm<sup>-2</sup> with 1 mAh cm<sup>-2</sup>.



**Fig. S27** Electrochemical impedance spectroscopy of R-CF@Sn@Zn and Zn electrode in (a) symmetric and (b) full cells.



**Fig. S28** Long-term cycling plots of cells with a low N/P ratio of 2 at 2 A g<sup>-1</sup>.



**Fig. S29** Voltage-capacity profiles at different cycles in the reversible rate process of the cells with R-CF@Sn@Zn.

**Table S1** Electrochemical properties comparison of the R-CF@Sn and relative 3D self-supporting and porous frameworks.

<b>Current collectors</b>	<b>Working condition</b>	<b>Lifespan</b>	<b>Ref.</b>
Sn@NHCF host	1 mA cm <sup>-2</sup> , 1 mAh cm <sup>-2</sup>	370 h	1
N-VG@CC	0.5 mA cm <sup>-2</sup> , 0.5 mAh cm <sup>-2</sup>	150 h	2
	1 mA cm <sup>-2</sup> , 1 mAh cm <sup>-2</sup>	70 h	
3D CNT framework	2 mA cm <sup>-2</sup> , 2 mAh cm <sup>-2</sup>	200 h	3
ACC-600@Cu <sup>2+</sup>	1 mA cm <sup>-2</sup> , 1 mAh cm <sup>-2</sup>	425 h	4
O, N-CC	1 mA cm <sup>-2</sup> , 1 mAh cm <sup>-2</sup>	320 h	5
PAN-Cu	2 mA cm <sup>-2</sup> , 1 mAh cm <sup>-2</sup>	275h	6
PCF	1 mA cm <sup>-2</sup> , 1 mAh cm <sup>-2</sup>	750 h	7
carbon foam	1 mA cm <sup>-2</sup> , 1 mAh cm <sup>-2</sup>	800 h	8
<b>This work</b>	<b>1 mA cm<sup>-2</sup>, 1 mAh cm<sup>-2</sup></b>	<b>1600 h</b>	
	<b>5 mA cm<sup>-2</sup>, 1 mAh cm<sup>-2</sup></b>	<b>700 h</b>	

### References for Table S1

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