

Supporting Information

Enhanced cyclic stability of Li-sulfur battery composites by low pressure penetration of PEDOT:PSS coating

Qiang Huang¹, Guojun Zha^{2*}, Zhaoyu Hu¹, Hangzhong Liu¹, Seema Agarwal³, Haoqing Hou^{1*}

¹College of Chemistry and Chemical Engineering, Jiangxi Normal University, Nanchang, China.

²School of new energy science and engineering, Xinyu University, Xinyu, China.

³Macromolecular Chemistry and Bavarian Centre for Battery Technology, University of Bayreuth, Universitätsstrasse 30, 95440 Bayreuth, Germany.

*Email: zhaguojun_8@163.com *Email: haoqing@jxnu.edu.cn

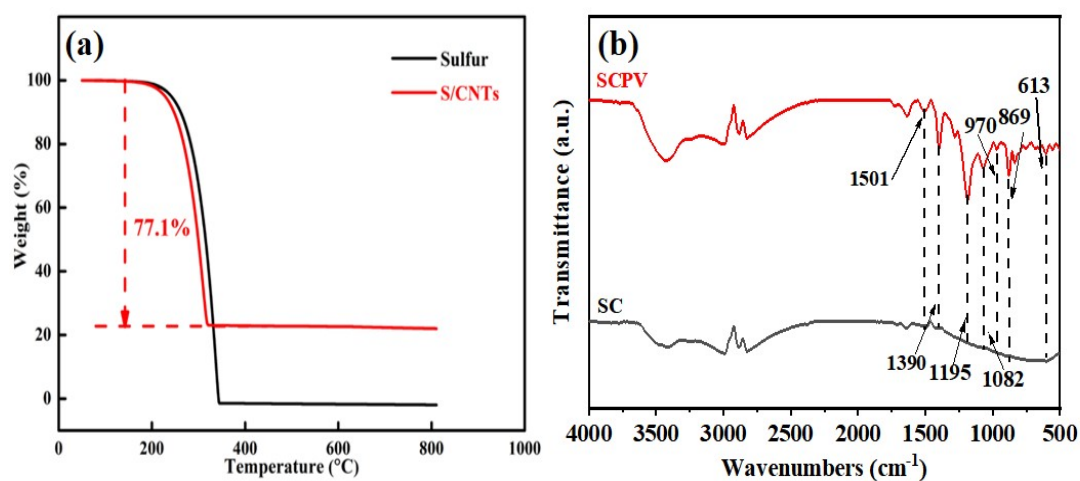


Fig.S1. (a)TGA curves of S and S/CNTs and (b)FTIR Spectrum of SC and SCPV

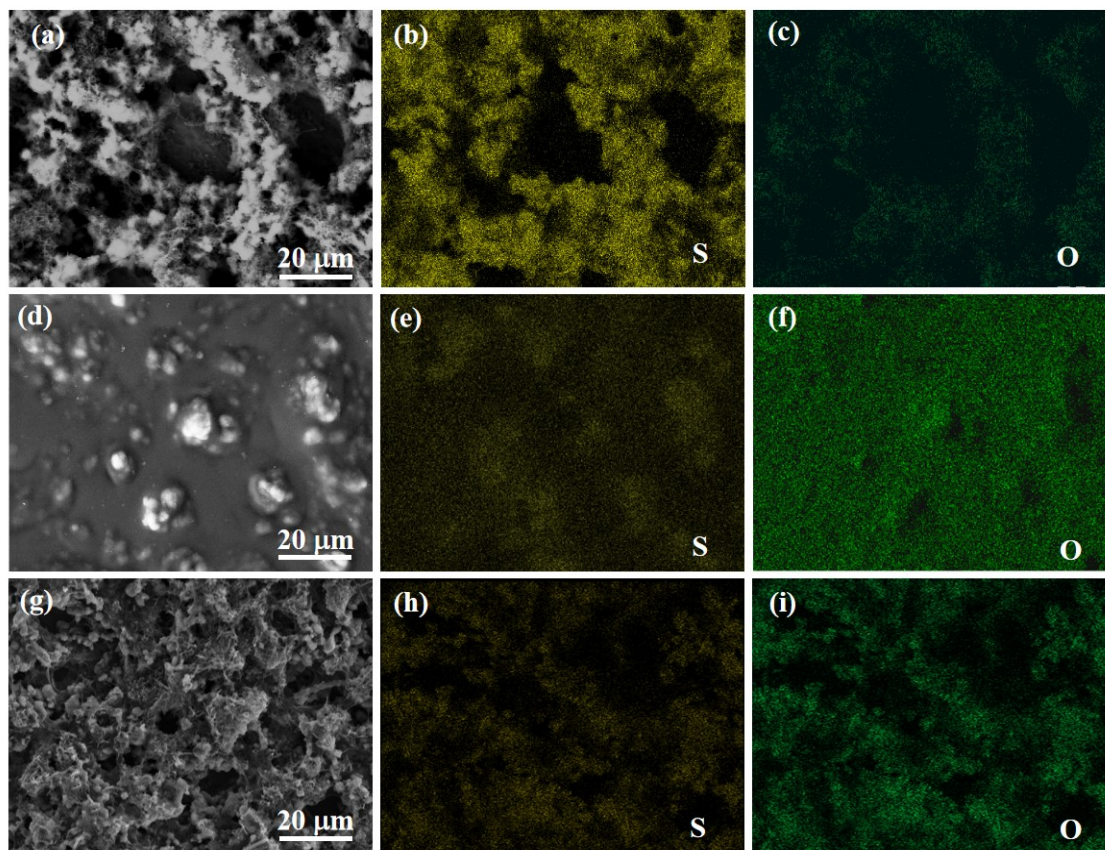


Fig.S2. SEM image of the SC (a) electrode and the EDS mapping of S (b) and O (c) elements on the (a) electrode surface, SEM image of the SCP (d) electrode and the EDS mapping of S (e) and O (f) elements on the (d) electrode surface, SEM image of the SCPV (g) electrode and the EDS mapping of S (h) and O (i) elements on the (g) electrode surface.

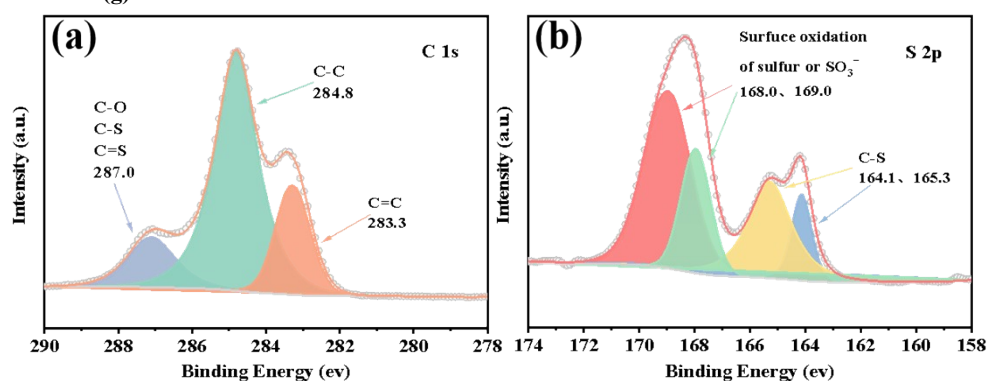


Fig.S3. XPS spectra of (a) C1s, (b) S2p for PEDOT:PSS.

One of the parameters affecting the rate performance of lithium-sulfur battery electrode materials is the Li^+ diffusion coefficient. The GITT method was used to determine the Li^+ diffusion coefficient of SC, SCP and SCPV. The formula of Li^+ diffusion coefficient is as follows (eq 1) [1,2].

$$D_{\text{Li}^+} = \frac{4(m_B V_M)}{\pi(M_B S)} \left\{ \frac{\Delta E_S}{\tau(dE_\tau/d\sqrt{\tau})} \right\}^2 \left(\tau \ll L^2/D_{\text{Li}^+} \right) \quad (\text{Seq 1})$$

V_M ($\text{cm}^3 \text{mol}^{-1}$) represents the molar volume of the sample. M_B (g mol^{-1}) represents the molar mass of the cathode material and m_B (g) represents the mass of the cathode material. S (cm^2)

represents the area of the pole piece. The eq 2 can be further simplified as follows [1,2] in the area where the curve conforms to the linear relationship between the square root of time ($\tau^{1/2}$) and potential (E):

$$D_{Li^+} = \frac{4}{\pi\tau} \left(\frac{m_B V_M}{M_B S} \right)^2 \left(\frac{\Delta E_S}{\Delta E_\tau} \right)^2 \quad (\text{Seq 2})$$

According to the $M_B = V_M \rho_B$, eq 3 can be further simplified as follows:

$$D_{Li^+} = \frac{4}{\pi\tau} \left(\frac{m_B}{\rho_B S} \right)^2 \left(\frac{\Delta E_S}{\Delta E_\tau} \right)^2 \quad (\text{Seq3})$$

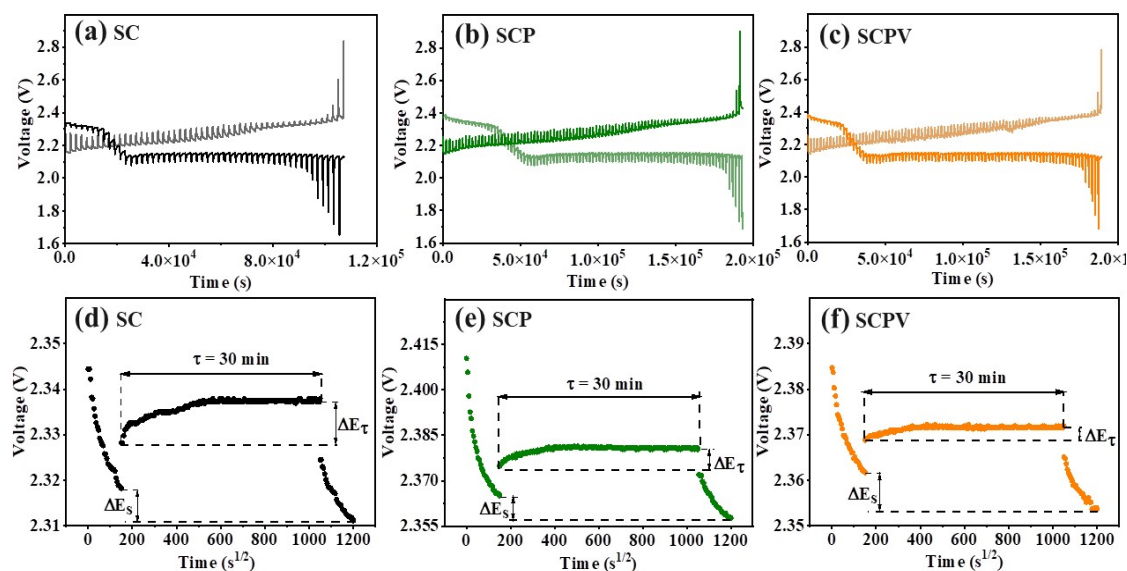


Fig.S4. GITT curves of (a) SC, (b) SCP and (c) SCPV electrodes at second discharging-charging process. The single titration presents linear correlation both the square root of time and the potential for (d) SC, (e) SCP, (f) SCPV samples.

Tab.S1. Comparison of the electrochemical performance of PEDOT: PSS modified Lithium-sulfur battery in this work with other literatures reported in the literature.

| Materials | Measurement Conditions | | | Electrochemical Performance | | | Ref. |
|---------------------------------------|--------------------------------|------|---------------------------------------|---|--------------|----------------|-----------|
| | Voltage Range (V) | T(K) | Current Density (mA g ⁻¹) | Initial Capacity (mAh g ⁻¹) | Cycle Number | Retention Rate | |
| PEDOT:PSS modified LSB | 1.7-2.7 vs. Li/Li ⁺ | 298 | 0.5C (1C=1675) | 996.8 | 200 | 91.87% | This Work |
| | | | 0.1C | | / | / | |
| | | | PVDF-HFP modified LSB | 1.7-2.7 vs. Li/Li ⁺ | 298 | 0.5C | |
| | 2.8-4.3 vs. Li/Li ⁺ | 2C | 408 | / | | / | |
| PEDOT:PSSCNT inter layer modified LSB | 1.7-3.0 vs. Li/Li ⁺ | 303 | 0.5C | 921 | 200 | 70.9% | [4] |
| PEDOT:PSS modified LSB | 1.5-3.0 vs. Li/Li ⁺ | 303 | 0.2 C | 1051 | 100 | 79% | [5] |

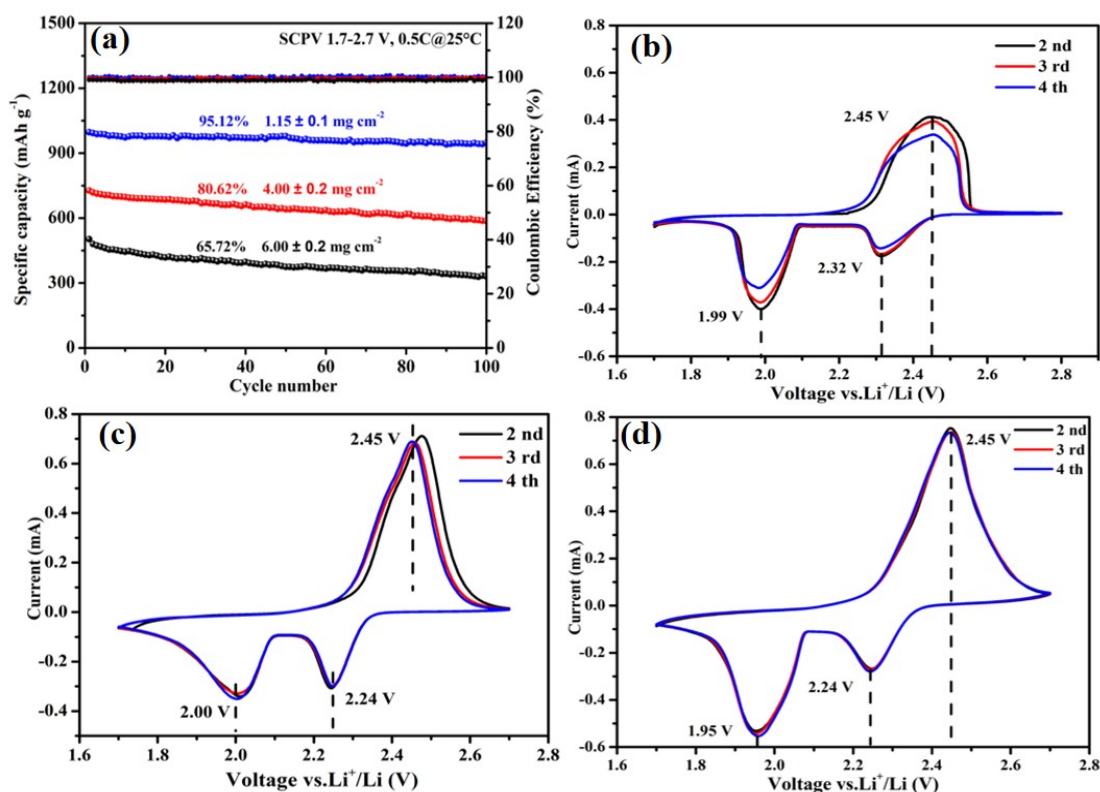


Fig.S5. (a)cycling performance under different loads of sulfur. CV curves of (b) SC, (c) SCP and (d) SCPV at a scan rate of 0.1 mV s⁻¹.

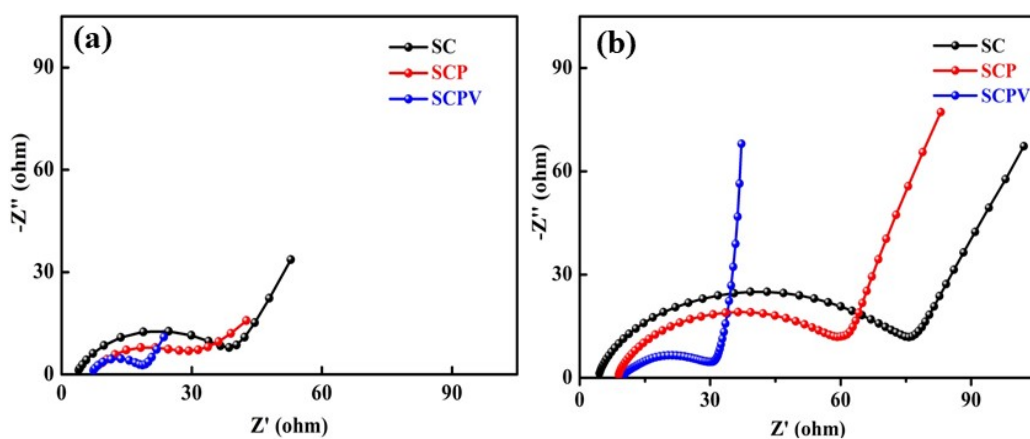


Fig.S6. EIS spectra of SC, SCP, and SCPV electrodes (a) before (b) after cycle.

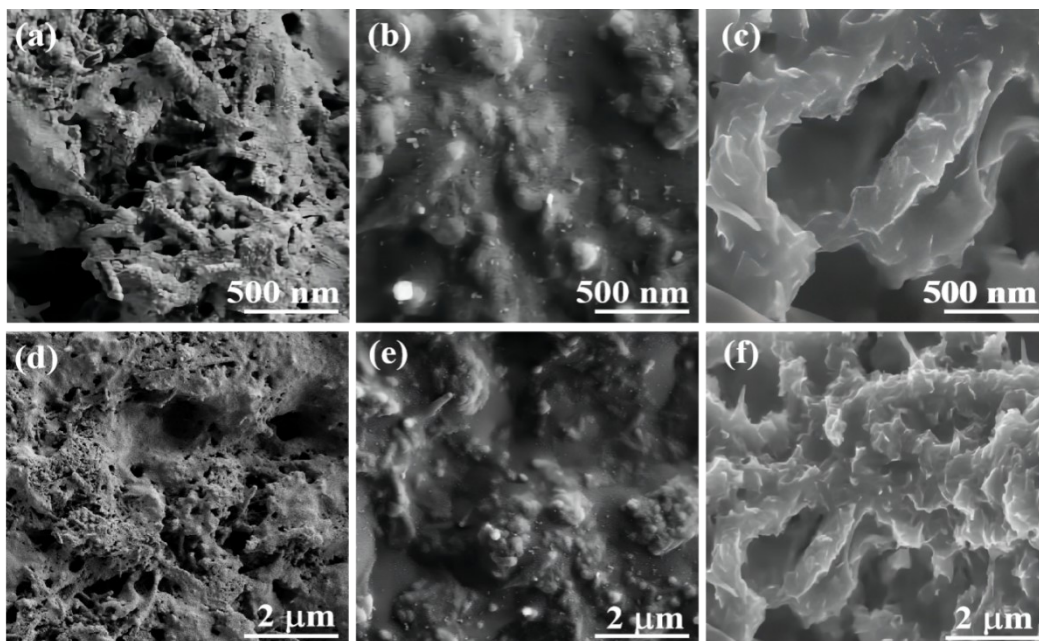


Fig.S7. SEM image of (a, d) SC, (b, e) SCP and (c, f) SCPV electrodes after 200 cycles at 0.5 C.

References

- [1] G. Zha, Y. Luo, N. Hu, C. Ouyang, H. Hou, Surface modification of the $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ cathode material by coating with FePO_4 with a yolk-shell structure for improved electrochemical performance, *ACS Appl. Mater. Inter.* 12 (2020), 36046-36053.
- [2] Y. S. Bai, X.Y. Wang, X.Y. Zhang, H. B. Shu, X. K. Yang, B. N. Hu, Q. L. Wei, H. Wu, Y. F. Song, The kinetics of Li-ion deintercalation in the Li-rich layered $\text{Li}_{1.12}[\text{Ni}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}]_{0.89}\text{O}_2$ studied by electrochemical impedance spectroscopy and galvanostatic intermittent titration technique, *Electrochim. Acta* 109 (2013) 355-364.
- [3] J. Yu, S. Liu, G. Duan, H. Fang, H. Hou, Dense and thin coating of gel polymer electrolyte on sulfur cathode toward high performance Li-sulfur battery, *Compos. Commun.* 19 (2020) 239-245.
- [4] A. Wang, G. Xu, B. Ding, Z. Chang, Y. Wang, H. Dou, X. Zhang, Highly conductive and lightweight composite film as polysulfide reservoir for high-performance lithium-sulfur batteries, *ChemElectroChem* 4 (2017) 362-368.
- [5] Y. Yang, G. Yu, J. J. Cha, H. Wu, M. Vosgueritchian, Y. Yao, Z. Bao, Y. Cui, Improving the performance of lithium-sulfur batteries by conductive polymer coating, *ACS Nano* 5 (2011) 9187-9193.