# Supporting Information

## Construction of a stable lithium sulfide membrane to greatly confine polysulfides for high performance lithium–sulfur batteries

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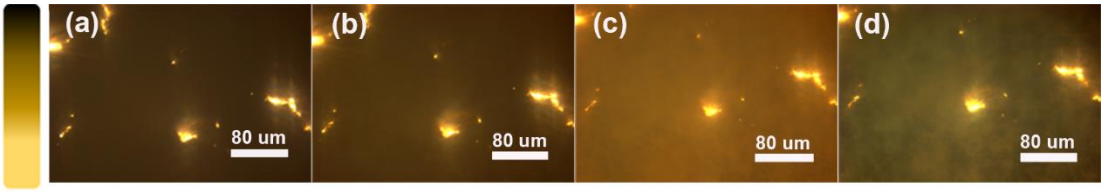


**Fig. S1** The in-situ optical microscopy images of the different stages in an electrochemical charge/discharge process at 0.1 mV $s^{-1}$ scan rate and 1.7-2.7 V voltage window. (a), A initial state. (b), discharge to 2.2 V. (c), discharge to 1.7 V and (d), re-charge to 2.7 V. (The shape guide shows the color variation of cathode surface from black to yellow).

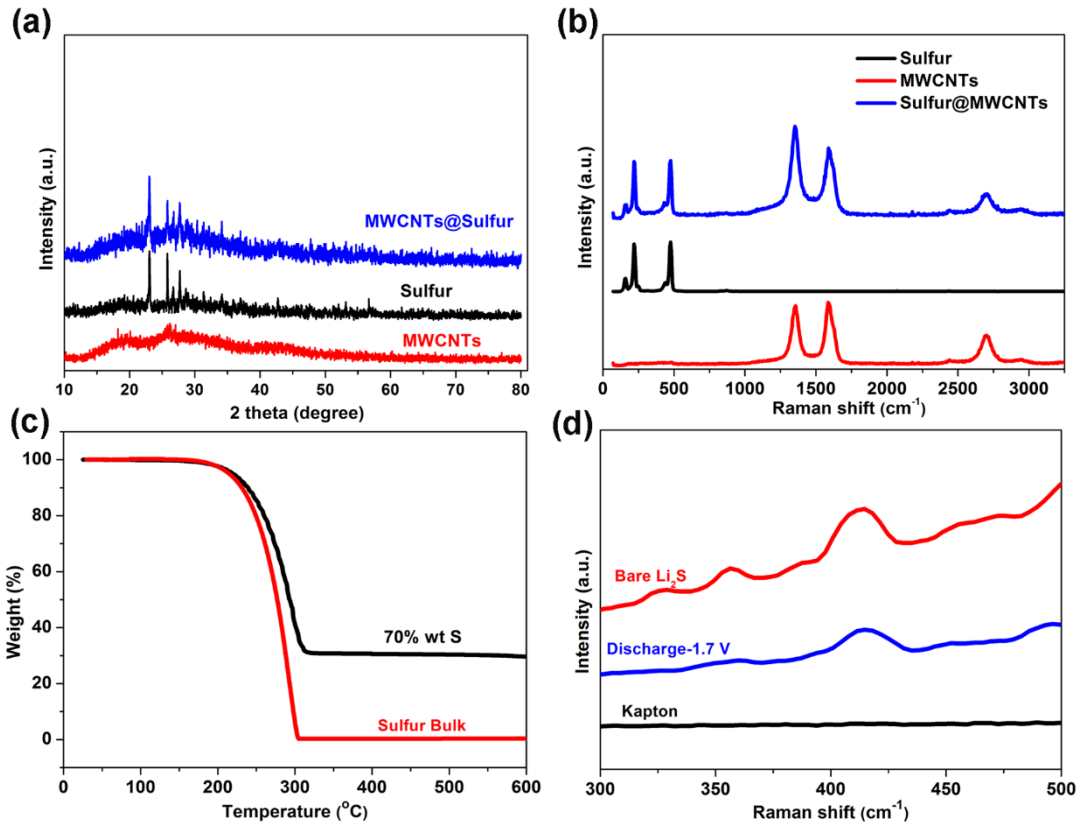


**Fig. S2** (a) XRD and (b) Raman spectrum includes MWCNTs@sulfur, sulfur and MWCNTs. (c) TGA curve shows 70% wt. sulfur in composites.(d) Raman spectrum of commercial $Li_2S$ refer to electrode (discharge to 1.7 V), all of them had been protected by kapton.

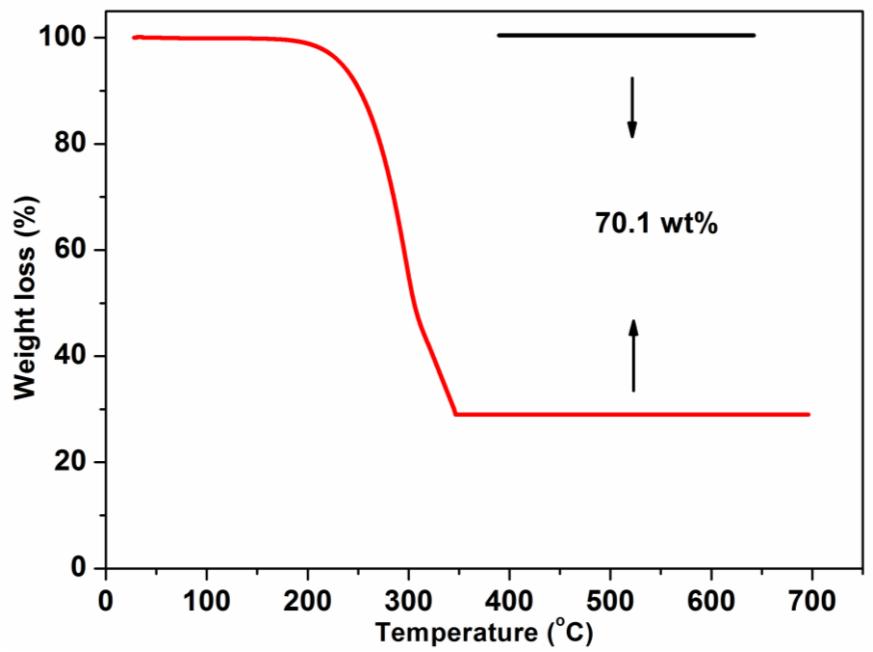


**Fig. S3** TGA curve shows 70.1% wt. sulfur in KCNTs.

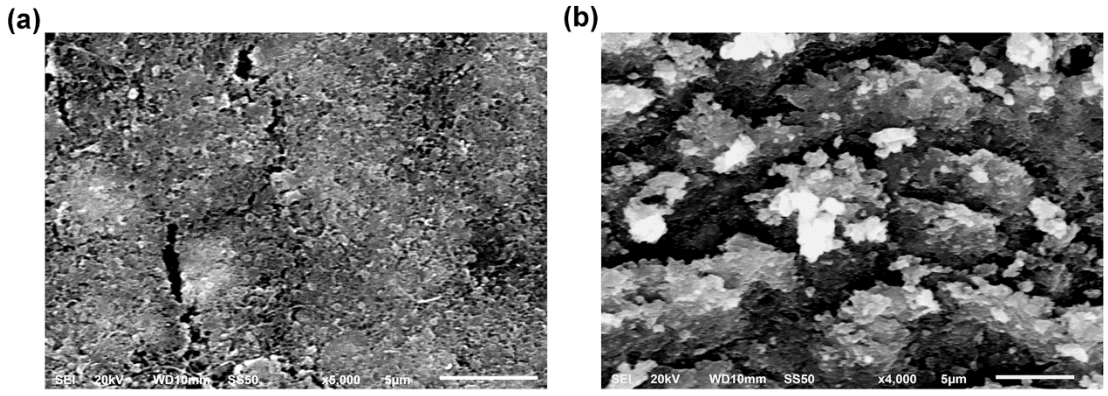


**Fig.S4** (a) the SEM image of KCNTs and (b) Ketjen black and super P with sulfur as the electrode materials after the cycling.

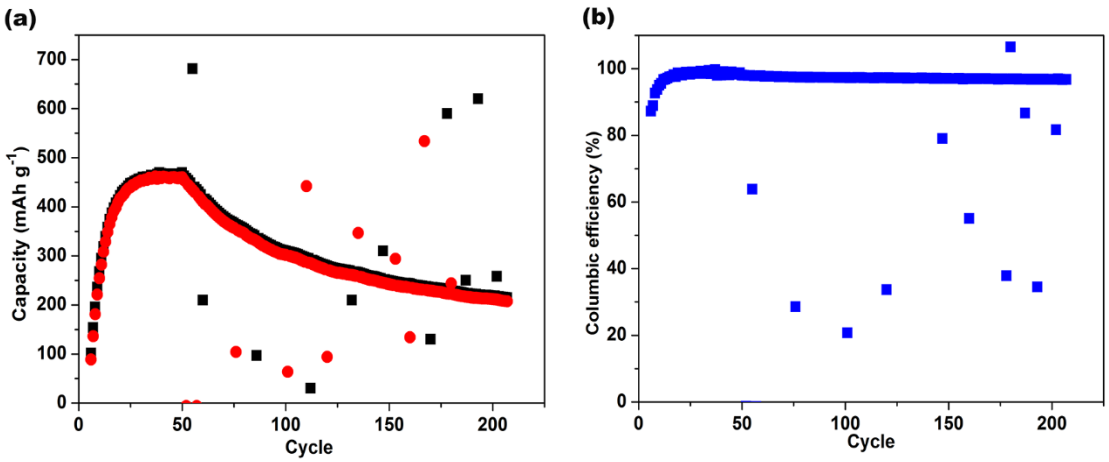


**Fig.S5** The KCNTs@S electrode without LS membrane has cycling performance (a) and columbic efficiency (b) at 1C current density.

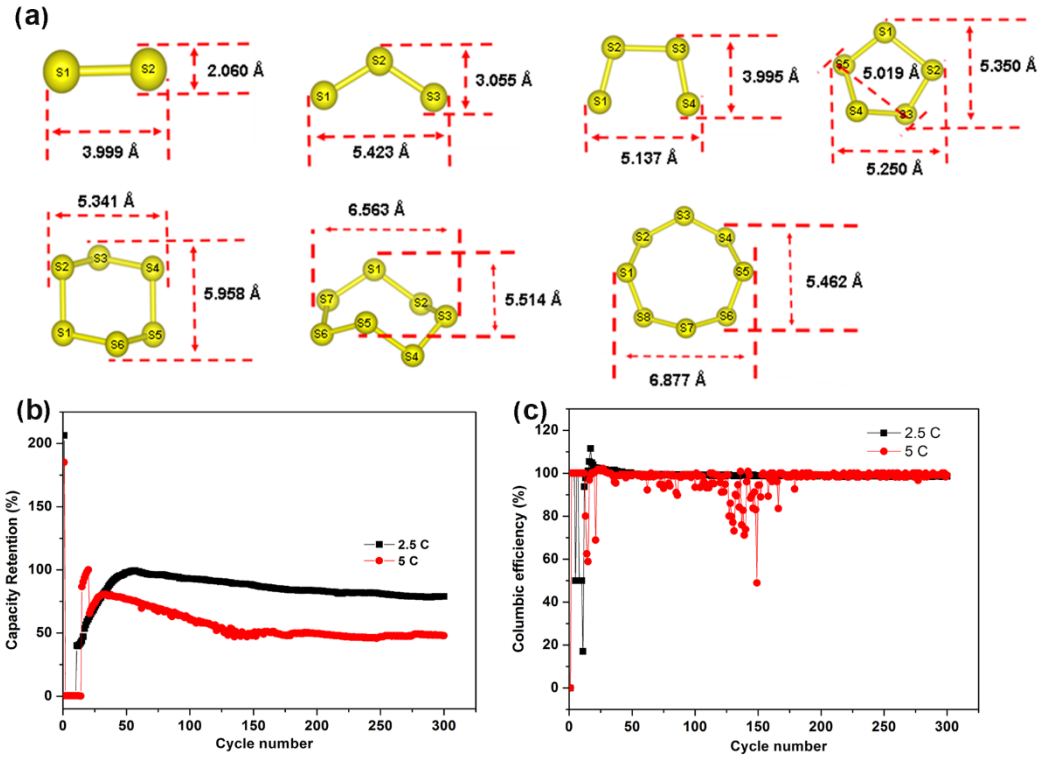

**Fig. S6** (a) the theoretical calculation of various polysulfide intermediates space size. (b) The cycling performance of LS membrane improved cathode (70% wt. S) at 2.5 C and 5 C. (c) columbic efficiency of LS membrane improved cathode (70% wt. S) at 2.5 C and 5 C.

Table 1 | Summary the performance of MWCNTs/Sulfur as cathode for Li–S battery systems.

| Approach | Voltage window (V) | Rate performance | Sulphur loading in electrode | Total cycle number | Degradation rate per cycle | Reference |
|---|---|---|---|---|---|---|
| **nano-sulfur/MWCNTs composite** | **1.5~3** | **N/A** | **48%** | **30** | **1.1% (300** | 1 |
| **S-coated-MWCNTs cathode** | **1.5~2.5** | **N/A** | **68%** | **60** | **N/A** | 2 |
| **polyacrylonitrile-sulfur@MWCNT composite** | **1~3** | **450 (4 C)** | **48%** | **50** | **0.3% (C/10)** | 3 |
| Dual core–shell structured sulfur cathode composite | **1.5~2.8** | **665 (1.2 C)** | **47.8%** | **200** | **0.177% (1500)** | 4 |
| **hierarchical S/MWCNT nanomicrosphere** | **1.7~2.7** | **600 (1 C)** | **45.6%** | **200** | **0.27% (1000 )** | 5 |

| | | | | | | |
|---|---|---|---|---|---|---|
| **Self-weaving sulfur–carbon composite** | **1.5~2.8** | **1012 (4 C)** | **40%** | **100** | **0.323% (1 C)** | 6 |
| **Aligned carbon nanotube/sulfur composite** | **1.6~3** | **280 (1 C)** | **76.5%** | **80** | **~0.43% (0.1 C)** | 7 |
| **Graphene/CNT @Porous Carbon** | **1.5~3** | **970 & 617 (5 C)** | **45% & 69.3%** | **150** | **0.12% & 0.18%(1 C)** | 8 |
| **Nitrogen-Doped Aligned Carbon Nanotube/Graphene** | **1.6~3** | **770 (5 C)** | **44.7%** | **80** | **0.3% (1 C)** | 9 |
| **Sulfur Nanocrystals Confined in Carbon Nanotube Network** | **1.9~2.6** | **934 (5 C)** | **50%** | **100** | **0.164% (1 C)** | 10 |
| **This work** | **1.7~2.7** | **560 (5 C)** | **56%** | **210** | **0.031% (1 C)** | |

Table 2 | Characteristic of various carbon-sulfur composites with high sulfur loading on the electrode (>60 wt%)

| **Approach** | **Sulfur loading density (mg cm$^{-2}$)** | **$LiNO_3$ additive** | **Cycling performance (mAh g$^{-1}$)** | **Capacity retention rate** | **Reference** |
|---|---|---|---|---|---|
| **Electrocatalysis of polysulfide conversion by sulfur-deficient MoS2 nanoflakes for lithium–sulfur batteries** | **0.9** | **2 wt%** | **1159.9~628 (0.5 C, 600 cycles)**<br>**1159.9~819.9 (0.5 C, 150 cyles)** | **54%**<br>**70%** | 11 |
| **A Sulfur-Rich Copolymer@CNT Hybrid Cathode with** | **1.2~1.58** | **0.2 m** | **818~647 (1 C,~450 cycles)** | **79%** | 12 |

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|---|---|---|---|---|---|
| **Dual-Confinement of Polysulfides** | | | | | |
| **Graphitized porous carbon materials with high sulfur loading for lithium-sulfur batteries** | **4** | **0.5 m** | **908~739**<br>**(0.1C, 100 cycles)** | **81%** | 13 |
| **Sole Chemical Confinement of Polysulfides on Nonporous Nitrogen/Oxygen Dual-Doped Carbon at the Kilogram Scale for Lithium–Sulfur Batteries** | **1.2~1.5** | **2 wt%** | **587~529**<br>**(1C ,180 cycles)** | **90%** | 14 |
| **This work** | **3** | **2 wt%** | **756~685**<br>**(1 C, 330 cycles)** | **90%** | |

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