

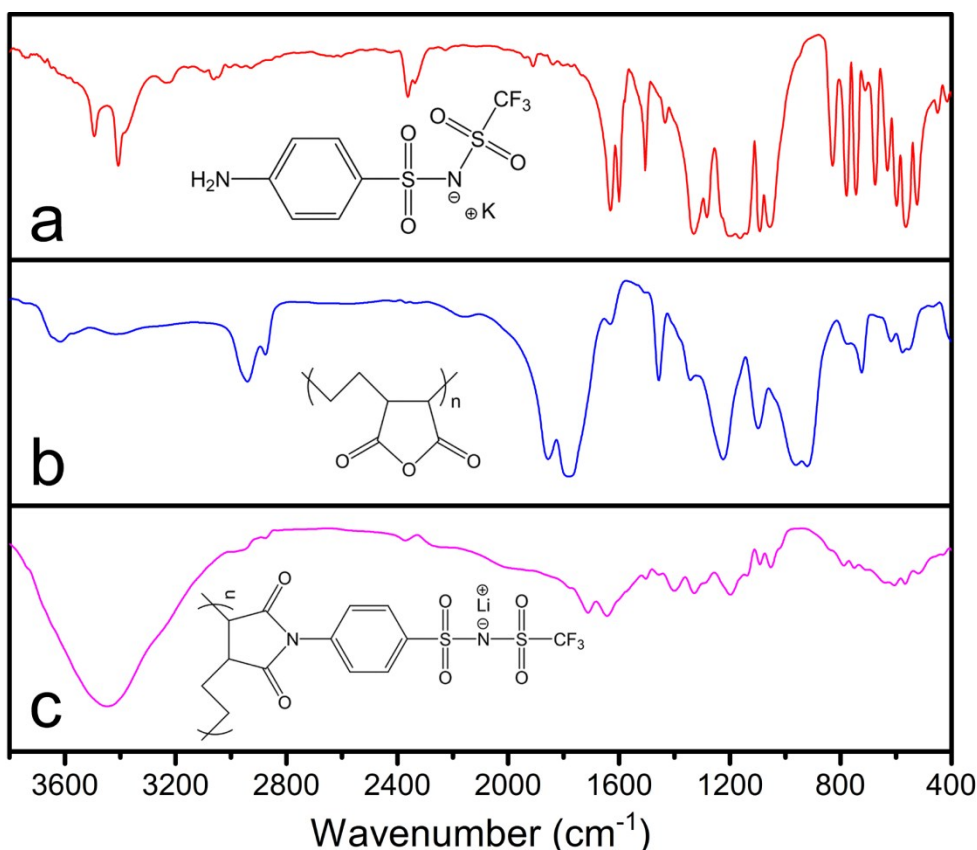
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## Electronic Supplementary Information

### Single ion conducting lithium sulfur polymer batteries with improved safety and stability

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**Fig. S1** FT-IR spectra of (a) KATFSI, (b) PEMA and (c) PEMA-graft-LiATFSI.

The FT-IR spectra of the KATFSI, PEMA and PEMA-graft-LiATFSI are shown in **Fig. S1**. For KATFSI (**Fig. S1a**),  $3494\text{ cm}^{-1}$  ( $\nu_{\text{N-H}}$ , *as*,  $-\text{NH}_2$ ),  $3408\text{ cm}^{-1}$  ( $\nu_{\text{N-H}}$ , *s*,  $-\text{NH}_2$ ),  $1632\&1601\text{ cm}^{-1}$  ( $\delta_{\text{N-H}}$ ,  $-\text{NH}_2$ ),  $1506\text{ cm}^{-1}$  ( $\nu_{\text{C-F}}$ ,  $-\text{CF}_3$ ),  $1328\&1282\text{ cm}^{-1}$  ( $\nu_{\text{S=O}}$ , *as*, two types of  $-\text{SO}_2-$ ),  $1092\&1053\text{ cm}^{-1}$  ( $\nu_{\text{S=O}}$ , *s*, two types of  $-\text{SO}_2-$ ). For PEMA (**Fig. S1b**),  $1855\text{ cm}^{-1}$  ( $\nu_{\text{C=O}}$ , *s*, anhydride),  $1780\text{ cm}^{-1}$  ( $\nu_{\text{C=O}}$ , *as*, anhydride),  $1224\text{ cm}^{-1}$  ( $\nu_{\text{C-O-C}}$ , anhydride). For PEMA-graft-LiATFSI (**Fig. S1c**),  $1712\text{ cm}^{-1}$  ( $\nu_{\text{C=O}}$ , *s*, anhydride),  $1641\text{ cm}^{-1}$  ( $\nu_{\text{C=O}}$ , *as*, anhydride),  $1502\text{ cm}^{-1}$  ( $\nu_{\text{C-F}}$ ,  $-\text{CF}_3$ ),  $1399\&1327\text{ cm}^{-1}$  ( $\nu_{\text{S=O}}$ , *as*, two

types of  $\text{-SO}_2\text{-}$ ),  $1091\&1051\text{ cm}^{-1}$  ( $\nu_{\text{S=O}}$ , s, two types of  $\text{-SO}_2\text{-}$ ).



Fig. S2 The photo image of the dry and wet single ion conducting electrolyte membrane.

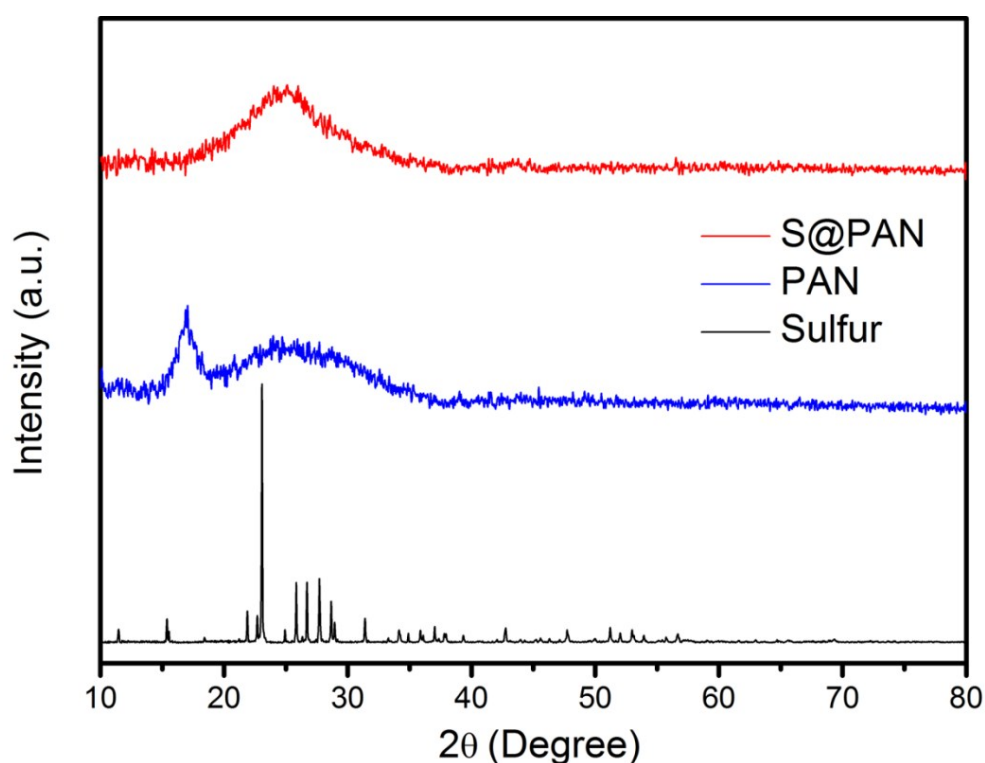


Fig.S3 XRD patterns of sulfur, PAN and S@PAN.

The XRD patterns of sulfur, PAN and S@PAN are shown in Fig. S3. Similar to the results reported in several previous studies,<sup>1-5</sup> the XRD pattern of the S@PAN obtained by the pyrolysis reaction of sulfur and polyacrylonitrile only shows a broad peak at  $2\theta=25^\circ$  with no diffraction peak of sulfur indexed to the *Fddd* orthorhombic structure (JCPDS: 08-0247).<sup>1-3</sup> Moreover, the diffraction peak at  $2\theta=17^\circ$  ascribed to the (110) plane of the crystal of polyacrylonitrile cannot be found in S@PAN.<sup>4,5</sup> The results indicate that sulfur is homogeneously embedded in the backbone

of the pyrolyzed polyacrylonitrile and becomes amorphous.<sup>1-4</sup>

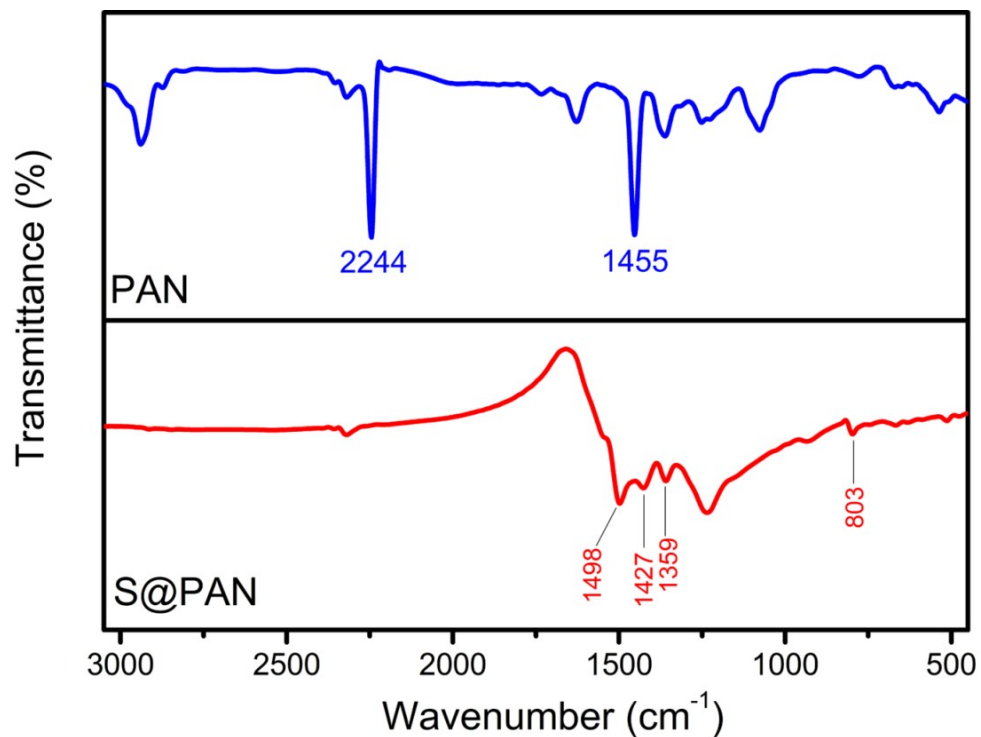


Fig. S4 FT-IR spectra of PAN and S@PAN.

**Fig. S4** displays the FT-IR spectra of PAN and S@PAN. The peaks at 1455 cm<sup>-1</sup> and 2244 cm<sup>-1</sup> ascribed to the -CH<sub>2</sub> and -CN groups of PAN disappear after the pyrolysis reaction.<sup>6</sup> Several new peaks emerge in the FT-IR spectra of S@PAN. The peaks at 1359 cm<sup>-1</sup> and 1498 cm<sup>-1</sup> indicate the existence of -CH deformation and C=C double bonds, respectively. The peaks at 803 cm<sup>-1</sup> and 1427 cm<sup>-1</sup> correspond to the formation of a heterocyclic structure in the pyrolysis process between sulfur and polyacrylonitrile.<sup>7</sup>

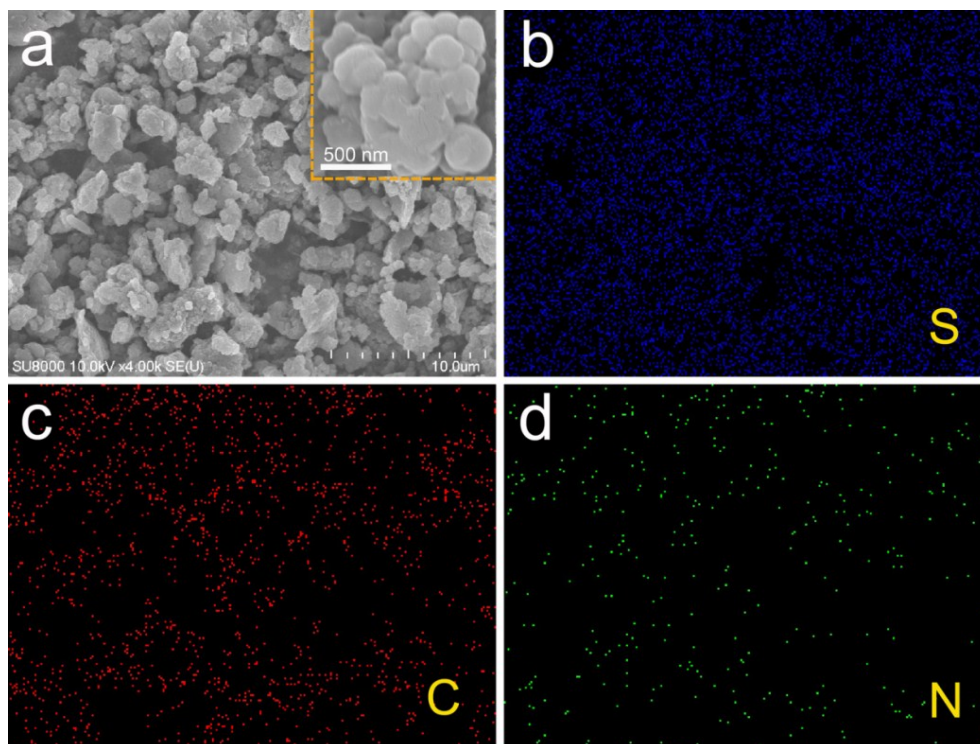
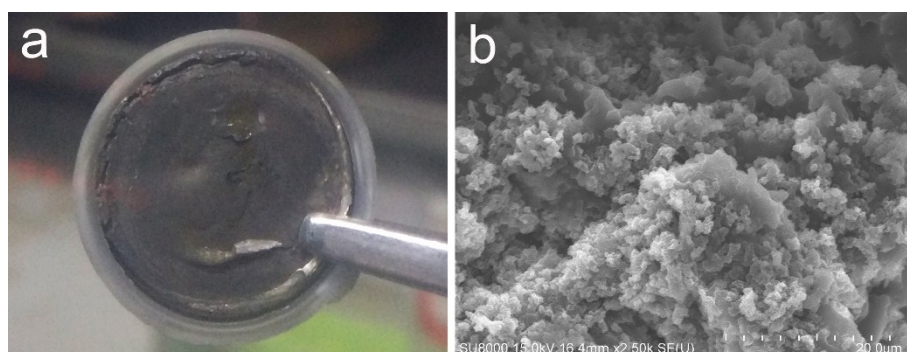
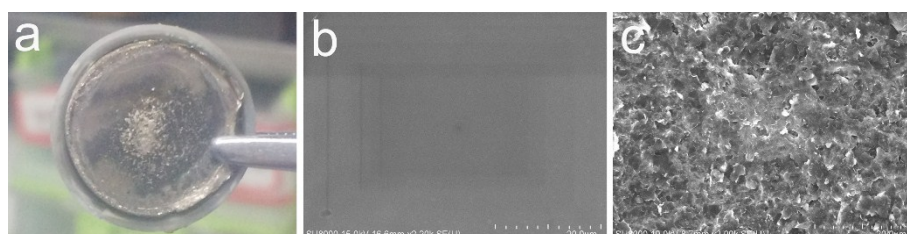


Fig. S5 Micromorphology and elemental distribution by EDS.

S@PAN represents the irregular micron-sized secondary particles made of nano-sized aggregates as shown in Fig. S5a. And the elemental S (Fig. S5b), C (Fig. S5c) and N (Fig. S5d) are distributed uniformly in the sample.



**Fig. S6** The morphology of lithium metal with commercial dual ion electrolyte after 1000 cycles at 1 C. (a) Photo image; (b) FE-SEM image.



**Fig. S7** The morphology of lithium metal with single ion conductor after 1000 cycles at 1 C. (a) Photo image; (b) FE-SEM image of the surface with metallic shining; (c) FE-SEM image of the dark area.

## References

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