

## Supporting Information

### **Tough and single lithium-ion conductive nanocomposite electrolytes based on PAES-g-PEG and POSS-PEG for lithium-sulfur battery**

*Yunho Shin, Anh Le Mong, Chi Nguyen Thi Linh, and Dukjoon Kim\**

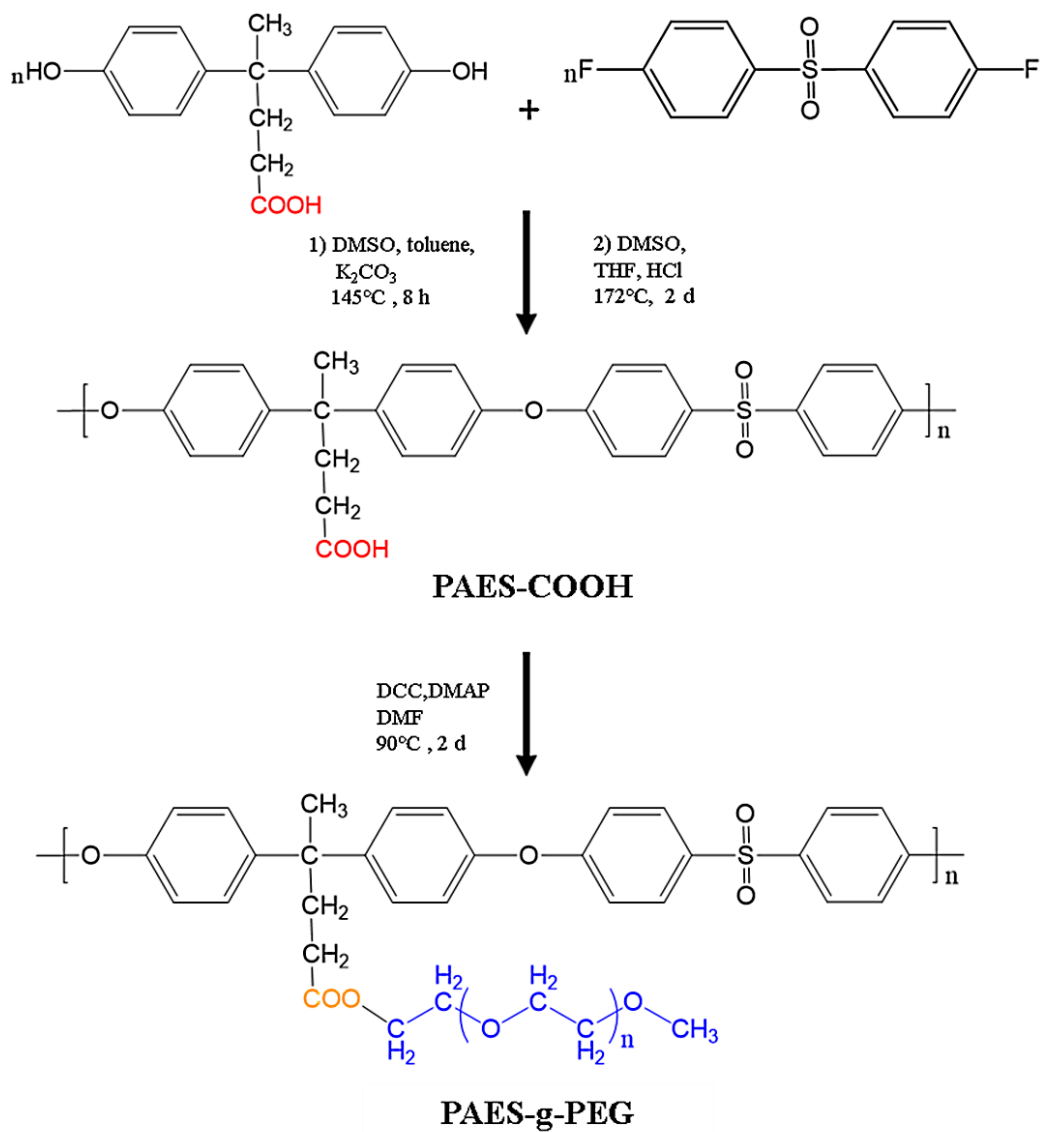
Y. Shin, A.L. Mong, C.N.T. Linh, Prof. D. Kim

School of Chemical Engineering, Sungkyunkwan University, Gyeonggi, Suwon, 16419,  
Republic of Korea.

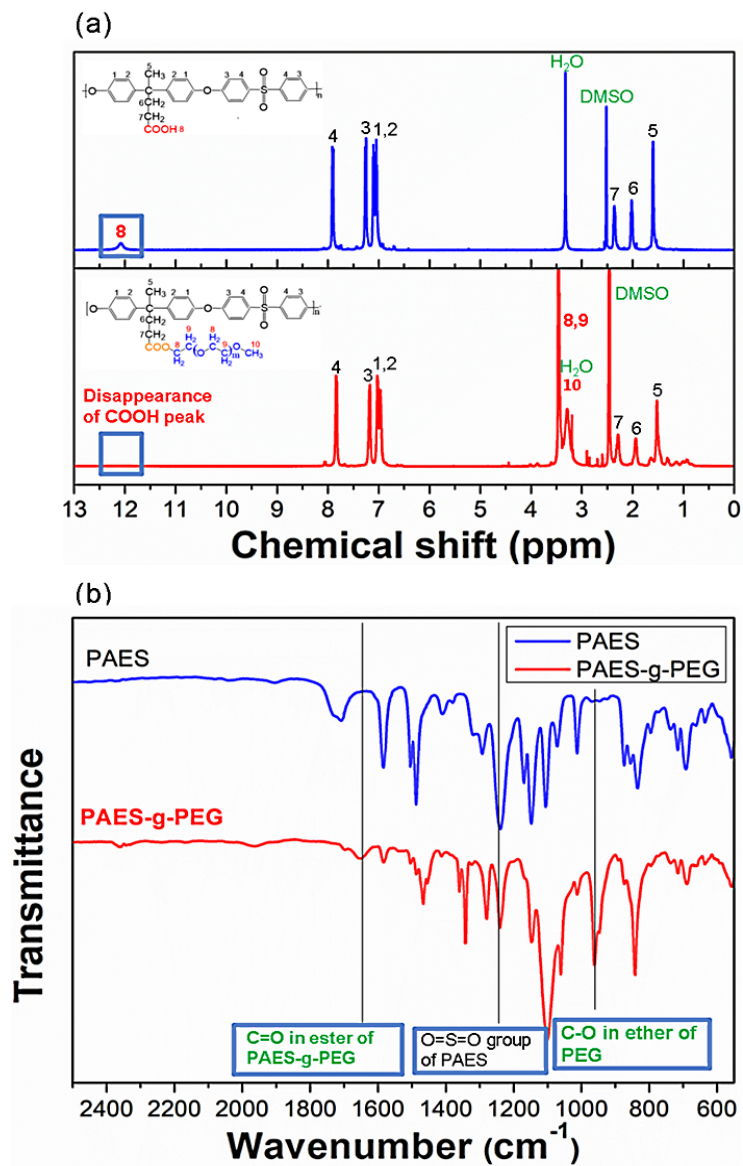
E-mail: [djkim@skku.edu](mailto:djkim@skku.edu)

Number of page: 22

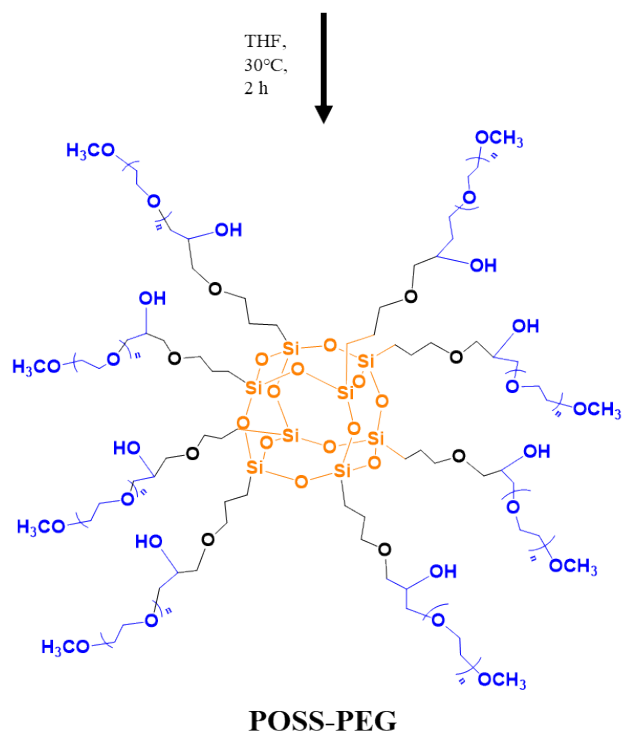
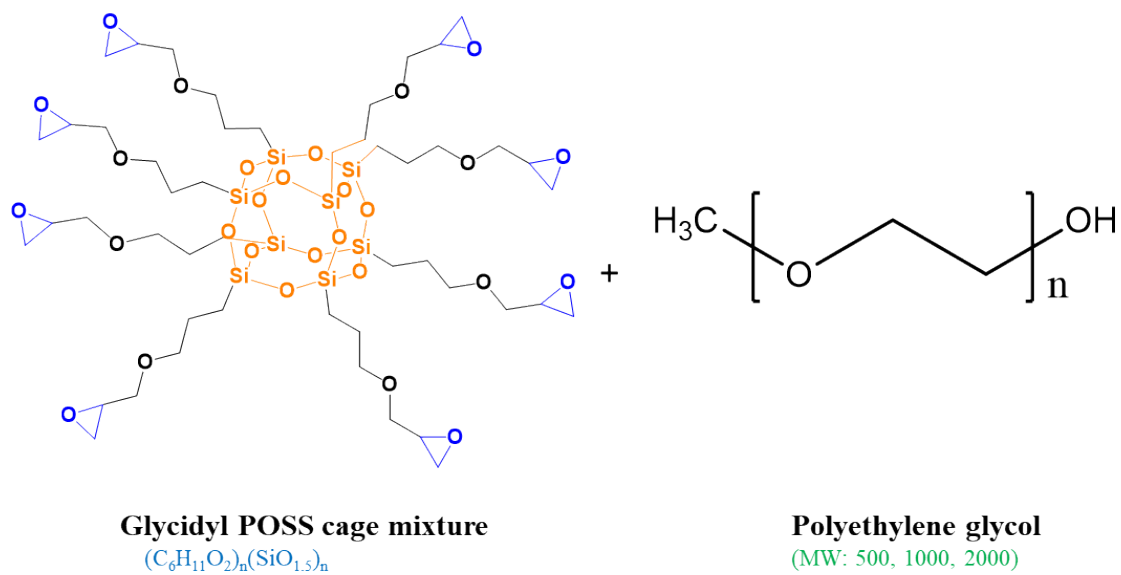
Number of figure: 15



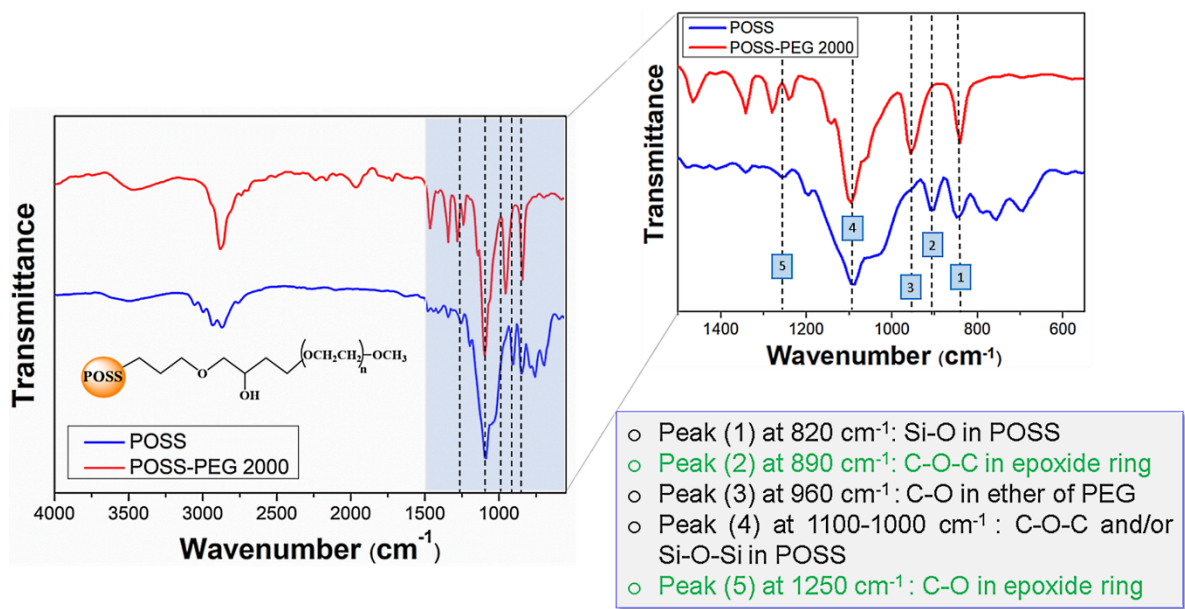
**Figure S1.** Synthetic scheme of PAES-COOH and PAES-g-PEG.



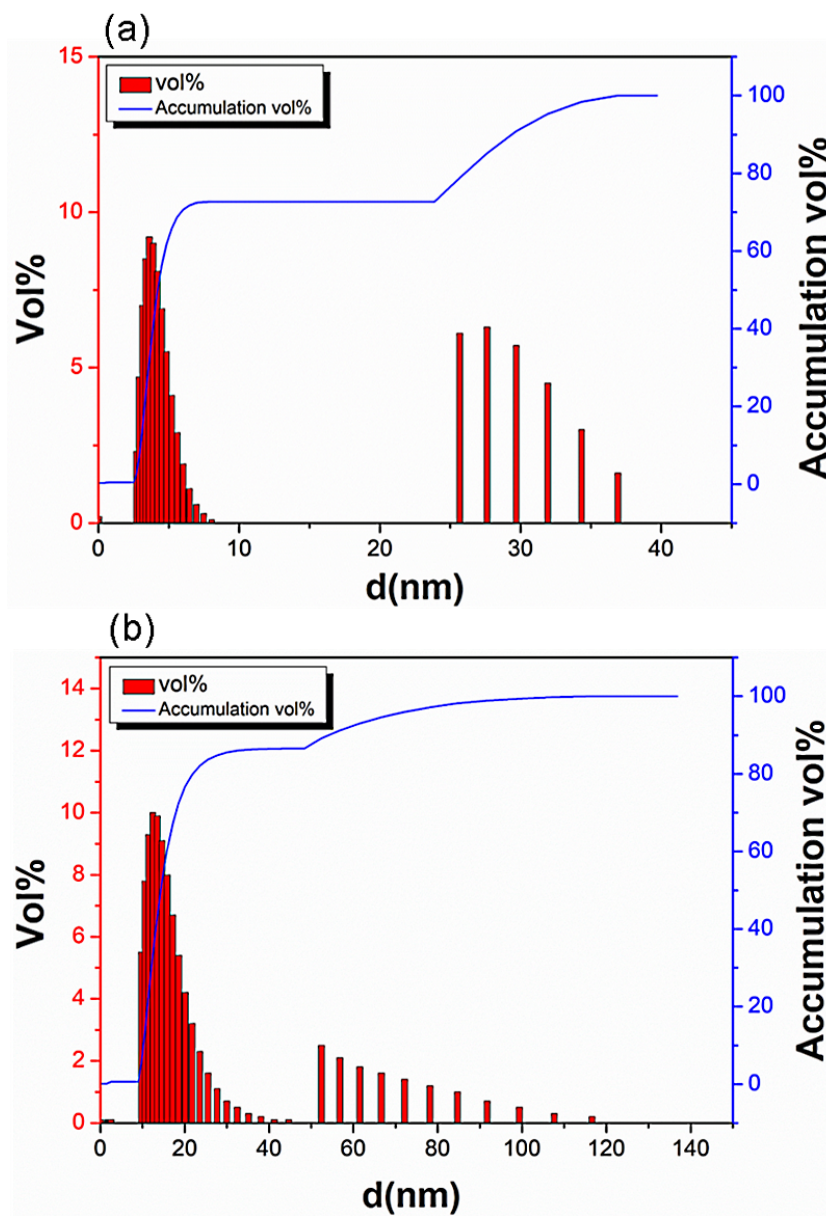
**Figure S2.** (a)  $^1\text{H-NMR}$ ; and (b) FTIR spectra of PAES-COOH and PAES-g-PEG.



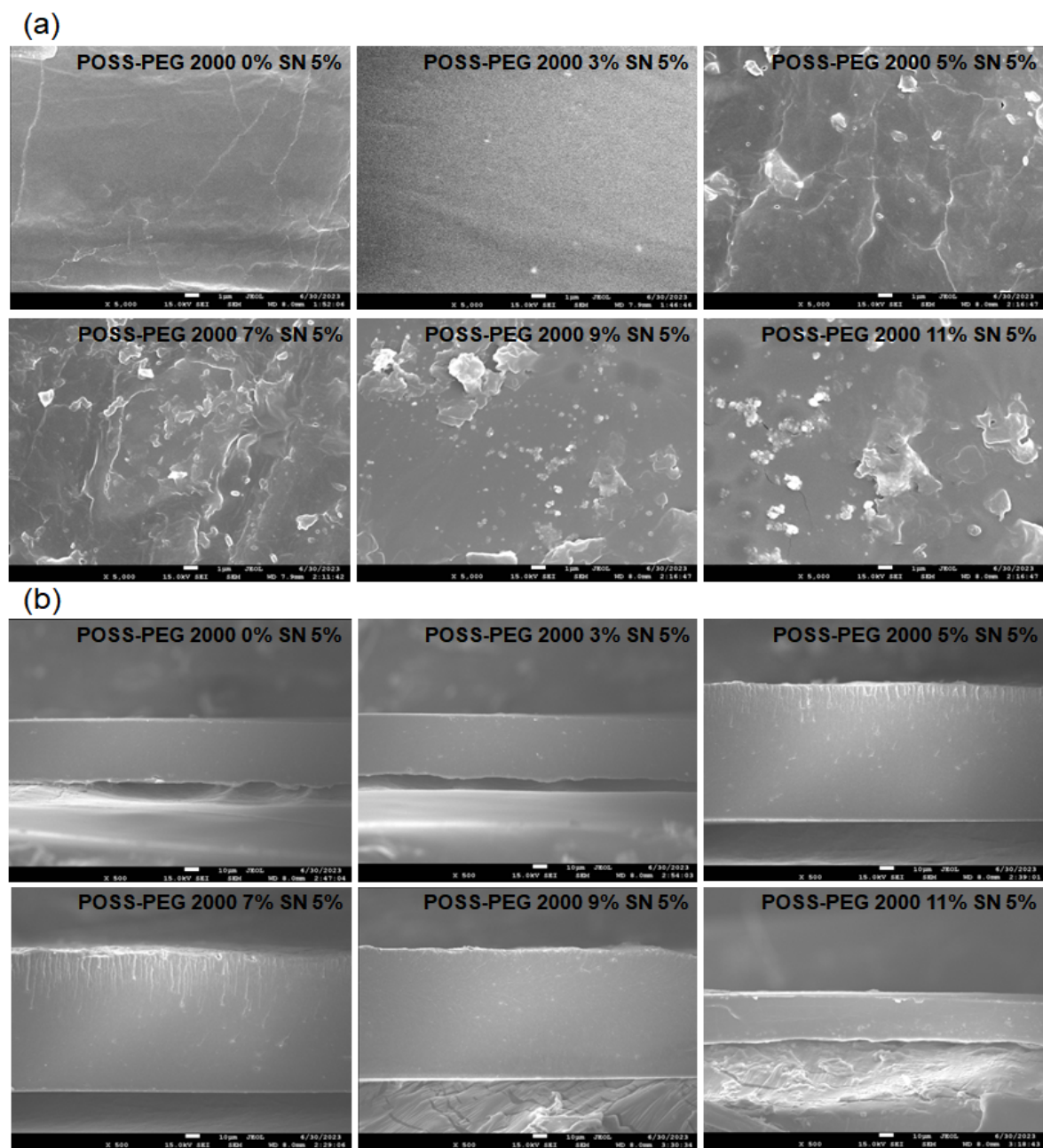
**Figure S3.** Synthetic scheme of POSS-PEG.



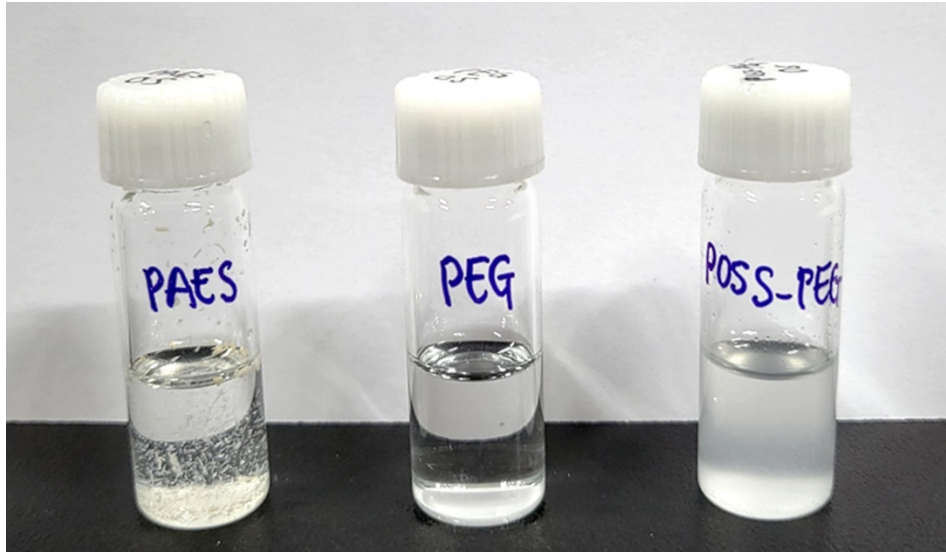
**Figure S4.** FTIR spectra of POSS and POSS-PEG 2000 nanoparticles.



**Figure S5.** Nanoparticle size of (a) POSS and (b) POSS-PEG 2000 using DLS analysis.

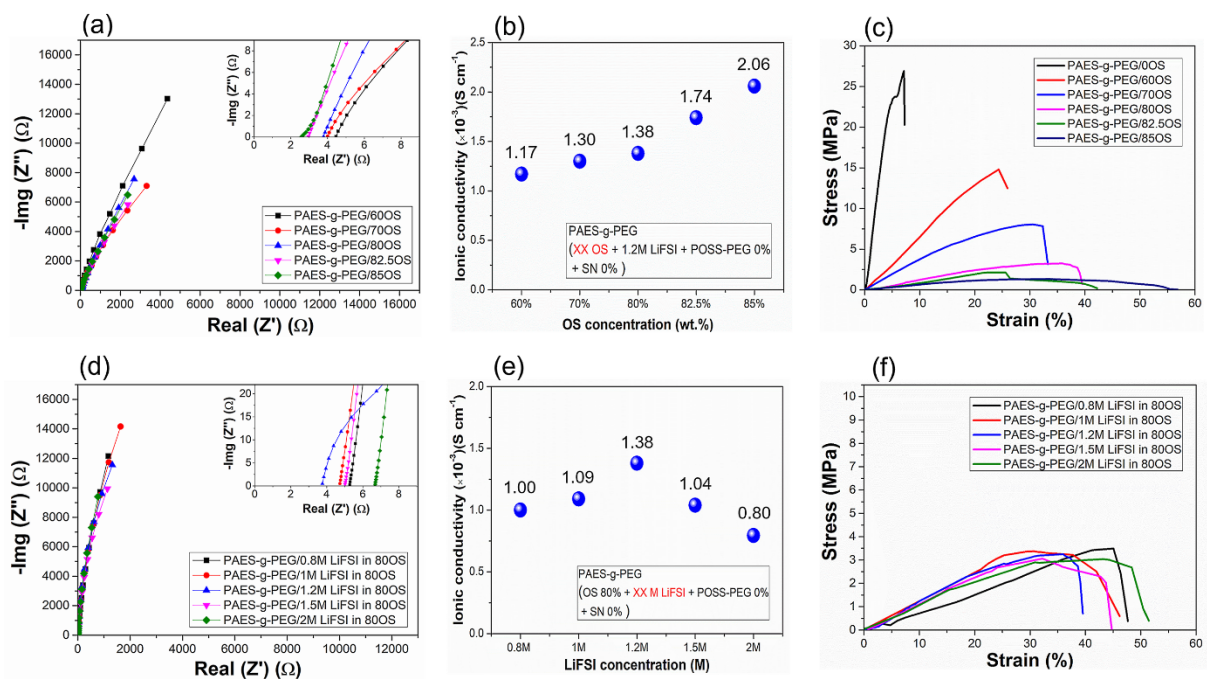


**Figure S6.** FESEM images of PAES-g-PEG membranes containing different amounts of POSS-PEG 2000 at (a) surface; and (b) cross-section.

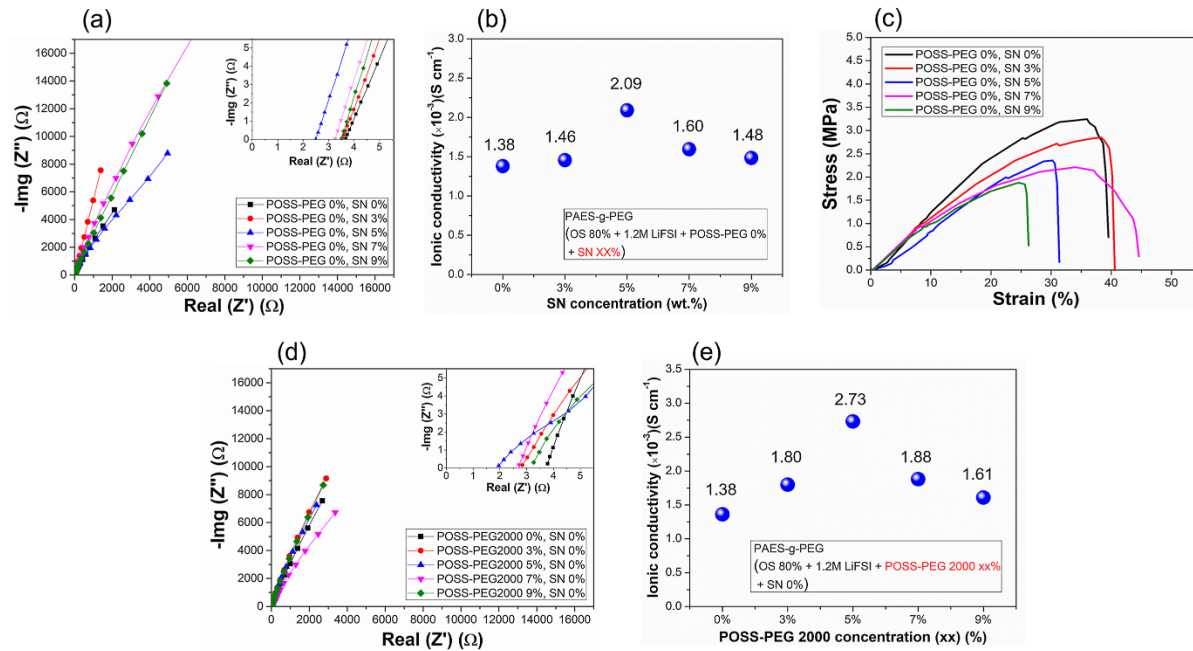


**Figure S7.** Miscibility of PAES, PEG, and POSS-PEG in OS.

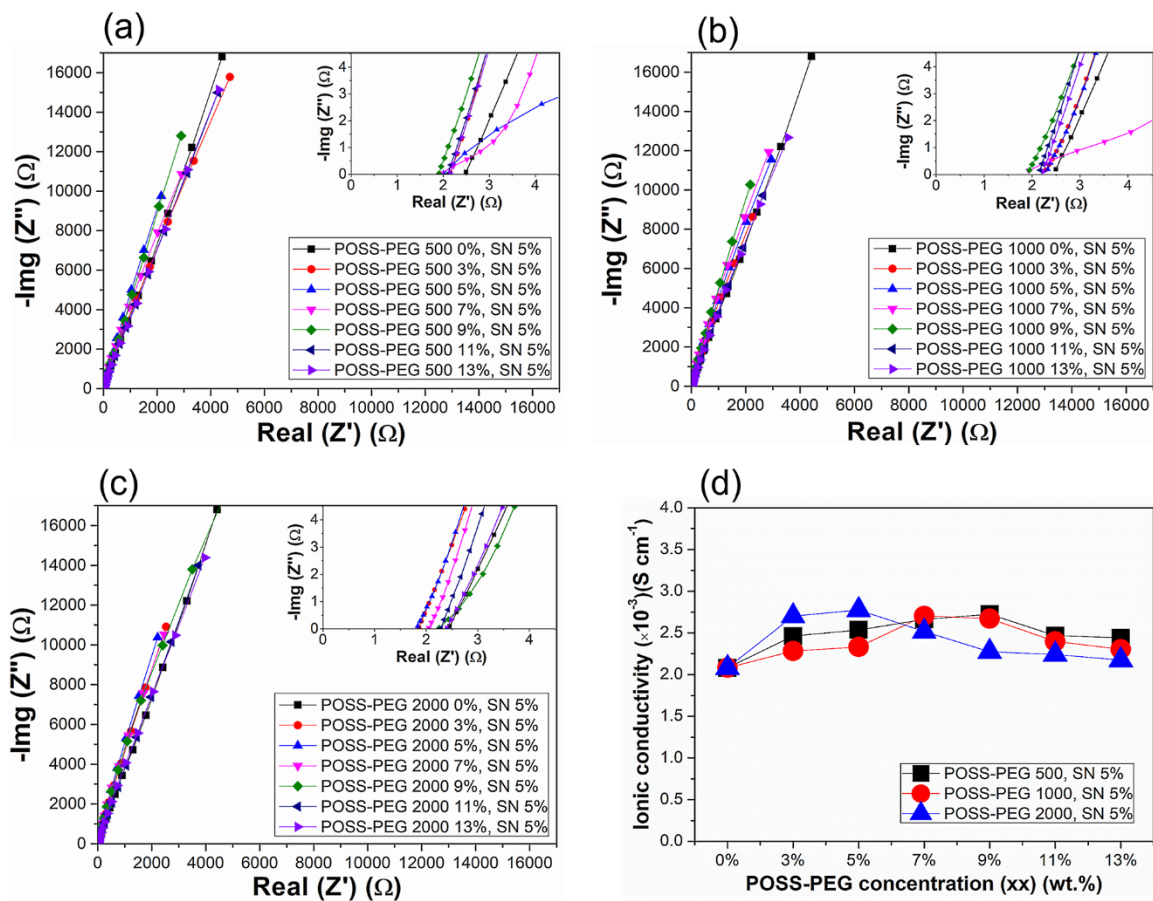




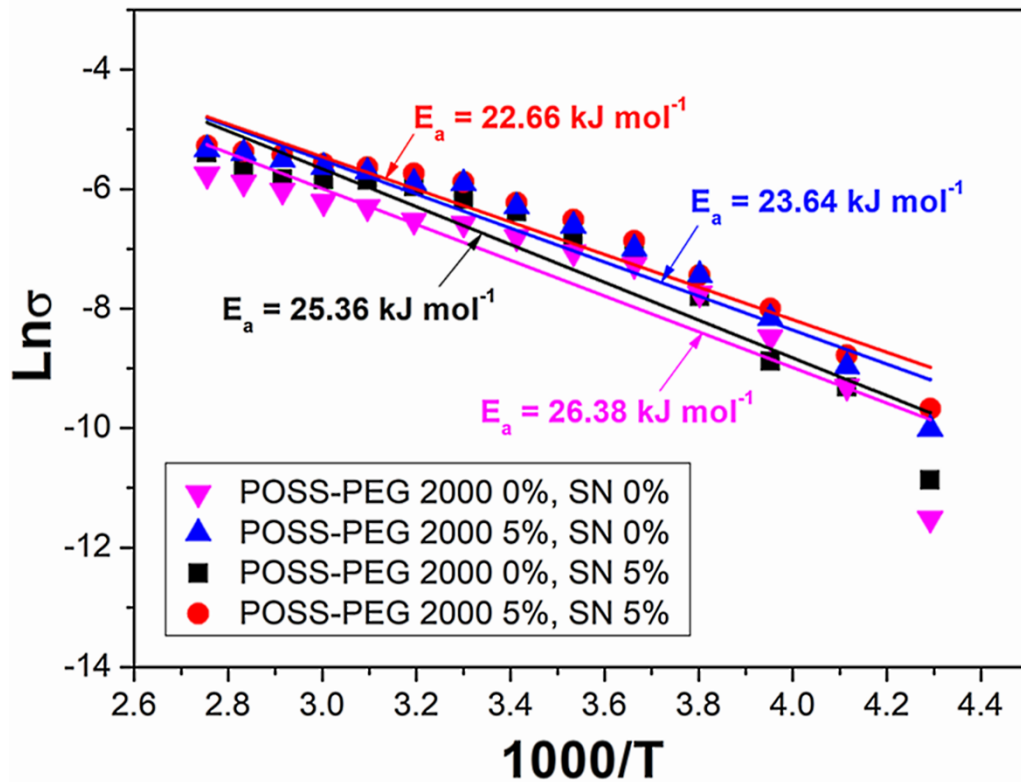
**Figure S8.** Effect of (a-c) OS concentration; and (d-f) LiFSI concentration on ionic conductivity and mechanical property of PAES-g-PEG membrane.



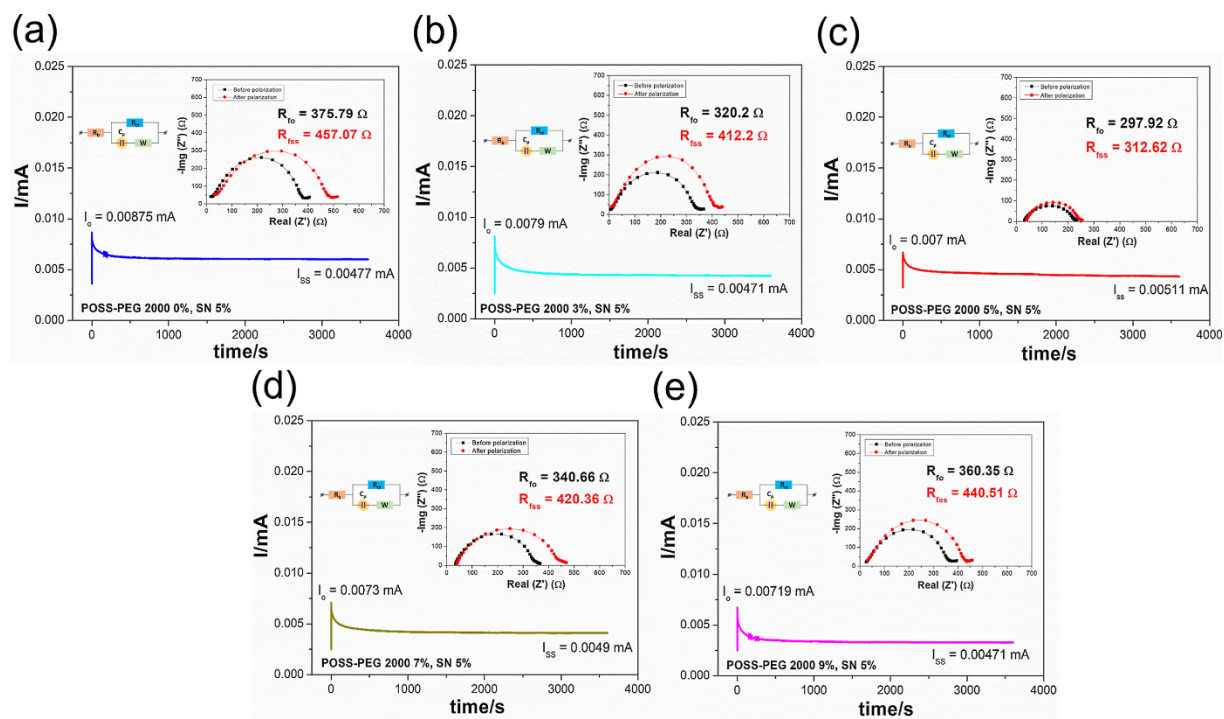
**Figure S9.** Effect of (a-c) SN concentration on ionic conductivity and mechanical property of PAES-g-PEG membrane; and (d, e) POSS-PEG 2000 concentration on ionic conductivity of PAES-g-PEG membrane without SN additive.



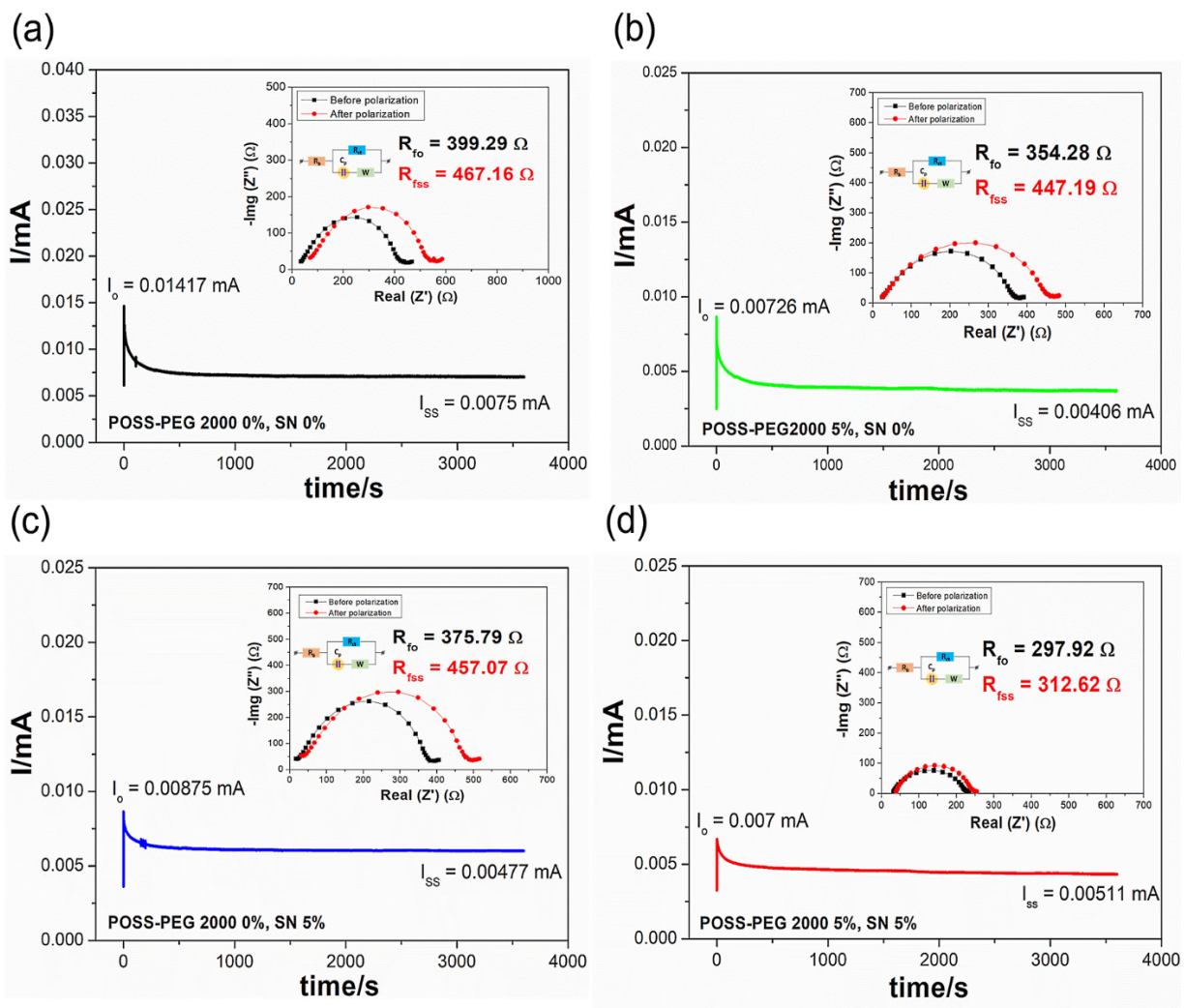
**Figure S10.** (a-c) Nyquist plot; and (d) ionic conductivity of PAES-g-PEG membranes containing POSS-functionalized with various PEG side chain lengths.



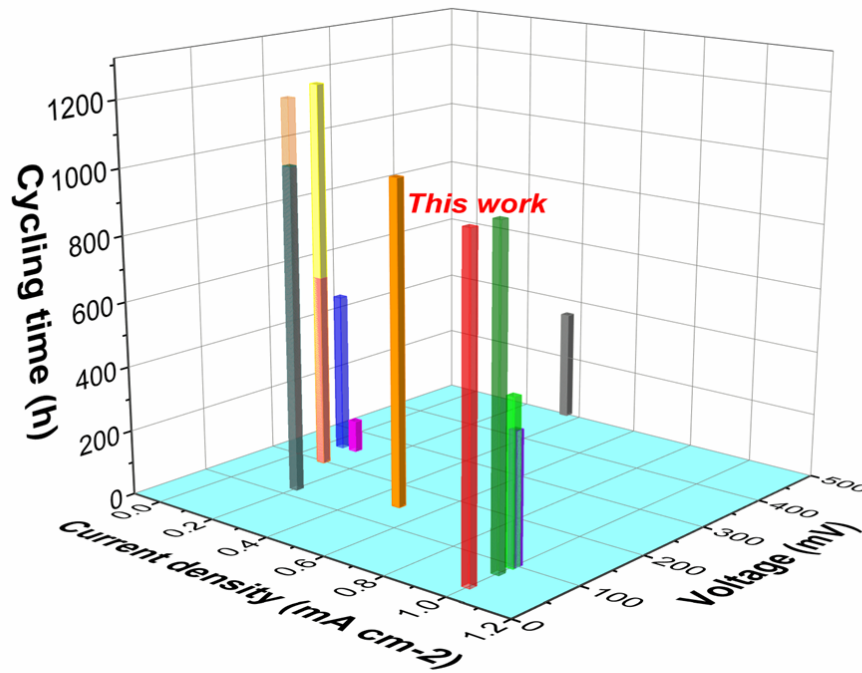
**Figure S11.** Thermal activation energy of PAES-g-PEG membranes containing different amounts of POSS-PEG 2000 and SN.



**Figure S12.** Chronoamperometry curves and interfacial resistance before and after polarization of PAES-g-PEG membranes containing various POSS-PEG 2000 contents at 5 wt.% SN.

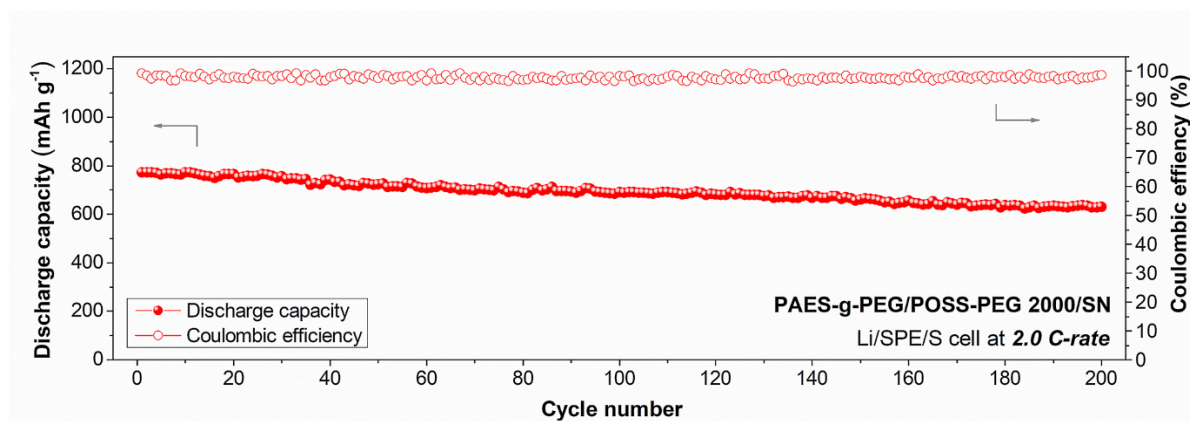


**Figure S13.** Chronoamperometry curves and interfacial resistance before and after polarization of PAES-g-PEG membranes containing different concentrations of POSS-PEG 2000 and SN.



- |                                        |                                                                                      |             |
|----------------------------------------|--------------------------------------------------------------------------------------|-------------|
| <span style="color: green;">■</span>   | (PETA-PDAALi) coated glass fiber                                                     | [1]         |
| <span style="color: red;">■</span>     | PAES-g-PEG/POSS-PEG/SN                                                               | [This work] |
| <span style="color: teal;">■</span>    | P(VDF-HDP)/(PIL+EMIM-TFSI)                                                           | [2]         |
| <span style="color: yellow;">■</span>  | PEGMA-co-HFBMA/ LiTFSI                                                               | [3]         |
| <span style="color: grey;">■</span>    | Flourinated polyoxalate                                                              | [4]         |
| <span style="color: blue;">■</span>    | PVA-UPy/PEG750                                                                       | [5]         |
| <span style="color: orange;">■</span>  | PME/ LiTFSI                                                                          | [6]         |
| <span style="color: green;">■</span>   | PEO/LLZTO composite                                                                  | [7]         |
| <span style="color: purple;">■</span>  | Li <sub>2</sub> Se/LiCl ceramic                                                      | [8]         |
| <span style="color: pink;">■</span>    | Gc-Li <sub>3.2</sub> P <sub>0.8</sub> Sn <sub>0.2</sub> S <sub>4</sub> glass ceramic | [9]         |
| <span style="color: orange;">■</span>  | PEO/TEGDME-cross-TMPTA                                                               | [10]        |
| <span style="color: magenta;">■</span> | PVT-EMITFSI                                                                          | [11]        |

**Figure S14.** Comparison of interfacial stability of symmetric Li/Li cells containing PAES-g-PEG/POSS-PEG 2000/SN with that of Li/Li cells using other solid electrolytes recently published. <sup>1-11</sup>



**Figure S15.** Cycling performance of Li/SPE/S cell assembled with PAES-g-PEG membrane containing POSS-PEG 2000 5% and SN 5% at 2.0 C-rate.



**Table S1.** Comparison of ionic conductivity ( $\sigma$ ), tensile strength and electrochemical stability window of PAES-g-PEG/POSS-PEG 2000/SN membrane with those of other electrolytes recently reported.<sup>12-24</sup>

Electrolyte membranes	$\sigma$ (mS cm <sup>-1</sup> )	TS <sup>a)</sup> (MPa)	ESW <sup>b)</sup> (V)	[Ref]
GPE-PI10	6.22	*	5.5	[12]
Poly(PEG-co-BTA)/zwitterion	4.79	0.05	4.5	[13]
<b>PAES-g-PEG/POSS-PEG 2000/SN</b>	<b>2.78</b>	<b>3.7</b>	<b>4.9</b>	<b>This work</b>
PIL-UPy/ LiTFSI (DOL+DME)	1.57	0.047	5.3	[14]
PAES-g-2PEG/IL-EC	1.15	0.9	4.95	[15]
ultrathin dual-salt PEO	0.57	2.4	4.8	[16]
PEO/LiTFSI/In <sub>2</sub> O <sub>3</sub>	0.527	5.1	4.6	[17]
PVAC/LLZTO	0.496	5.97	5.4	[18]
PI/PVDF/LiTFSI	0.41	6.1	5.1	[19]
PVDF+CA/BPSO-LiTFSI	0.4	6.8	4.7	[20]
PZE <sub>w</sub> -50%	0.275	*	4.94	[21]
F-PMIA@ZIF-8-PEO-LiTFSI	0.239	8.39	5.1	[22]
PEO/LiTFSI/ox-PIL@GO	0.101	0.43	5.28	[23]
PEO/POSS-PEG1000	0.08	0.406	5.08	[24]

<sup>a)</sup> Tensile strength and <sup>b)</sup> electrochemical stability window.

**Table S2.** Comparison of the S/SPE/Li cell performance based on PAES-g-PEG/POSS-PEG 2000/SN membranes with those based on some other electrolytes recently reported.<sup>25–35</sup>

Electrolyte membranes	Discharge capacity (mAh g <sup>-1</sup> )				Capacity retention (%) (C /cycle number)	[Ref]
	0.2C	0.5C	1.0C	2.0C		
PEO/ PIM-8% <i>solid electrolyte</i>	1200	1100	910	600	66.3% (0.5C/100 <sup>th</sup> )	[26]
SO <sub>3</sub> Li-g-UIO(MOF)/LiTFSI +IL	985	890	749	-	84% (0.2C/250 <sup>th</sup> )	[27]
PEO-Li <sub>4</sub> (BH <sub>4</sub> ) <sub>3</sub> I/ SiO <sub>2</sub> (5%wt)	950	817	613	583	62.2% (0.1C/75 <sup>th</sup> )	[25]
<b>PAES-g-PEG/POSS-PEG/SN</b>	<b>980.1</b>	<b>929.9</b>	<b>865.9</b>	<b>772.8</b>	<b>85.3% (0.2C/200<sup>th</sup>)</b> <b>82.0%(2.0C/200<sup>th</sup>)</b>	<b>This work</b>
PETT-DA/(PEO+PVDF-HFP)	910	766	624	543	87.1% (2.0C/300 <sup>th</sup> )	[28]
<i>In-situ</i> S-DCBQ organosulfur	890	795	750	600	74% (0.2C/100 <sup>th</sup> )	[29]
PEO/(LLZO+MWCNT)	873	810	500	400	72.6% (0.2C/60 <sup>th</sup> )	[30]
PETEA+DVA/(DOL+TEGDM)	779	621	325	220	70% (0.5C/300 <sup>th</sup> )	[31]
PEO/ (P <sub>2</sub> S <sub>5</sub> + LiTFSI)	750	450	-	-	55% (0.2C/350 <sup>th</sup> )	[32]
PEO/ Li <sub>1.3</sub> Al <sub>0.3</sub> Ti <sub>1.7</sub> (PO <sub>4</sub> ) <sub>3</sub> / PEO)	692.9	428.4	362.3	-	75% (0.1C/100 <sup>th</sup> )	[33]
Polydopamine-coated Li <sub>6</sub> PS <sub>5</sub> Cl	552.8	226.4	-	-	72% (0.2C/100 <sup>th</sup> )	[34]
PEO/(TCM+ LiTFSI)	450	300	-	-	85% (0.1C/50 <sup>th</sup> )	[35]

## Reference

- 1 D. Zhou, A. Tkacheva, X. Tang, B. Sun, D. Shanmukaraj, P. Li, F. Zhang, M. Armand and G. Wang, *Angewandte Chemie International Edition*, 2019, **58**, 6001–6006.
- 2 M. Yao, H. Zhang, K. Dong, B. Li, C. Xing, M. Dang and S. Zhang, *J. Mater. Chem. A*, 2021, **9**, 6232–6241.
- 3 M. Jia, P. Wen, Z. Wang, Y. Zhao, Y. Liu, J. Lin, M. Chen and X. Lin, *Advanced Functional Materials*, 2021, **31**, 2101736.
- 4 H. Sun, X. Xie, Q. Huang, Z. Wang, K. Chen, X. Li, J. Gao, Y. Li, H. Li, J. Qiu and W. Zhou, *Angewandte Chemie International Edition*, 2021, **60**, 18335–18343.
- 5 Y. H. Jo, B. Zhou, K. Jiang, S. Li, C. Zuo, H. Gan, D. He, X. Zhou and Z. Xue, *Polym. Chem.*, 2019, **10**, 6561–6569.
- 6 H. Li, Y. Du, X. Wu, J. Xie and F. Lian, *Advanced Functional Materials*, 2021, **31**, 2103049.
- 7 R.-A. Tong, L. Chen, G. Shao, H. Wang and C.-A. Wang, *Journal of Power Sources*, 2021, **492**, 229672.
- 8 D. Lee, S. Sun, H. Park, J. Kim, K. Park, I. Hwang, Y. Jung, T. Song and U. Paik, *Journal of Power Sources*, 2021, **506**, 230158.
- 9 F. Zhao, S. H. Alahakoon, K. Adair, S. Zhang, W. Xia, W. Li, C. Yu, R. Feng, Y. Hu, J. Liang, X. Lin, Y. Zhao, X. Yang, T.-K. Sham, H. Huang, L. Zhang, S. Zhao, S. Lu, Y. Huang and X. Sun, *Advanced Materials*, 2021, **33**, 2006577.
- 10 F. Fu, W. Lu, Y. Zheng, K. Chen, C. Sun, L. Cong, Y. Liu, H. Xie and L. Sun, *Journal of Power Sources*, 2021, **484**, 229186.

- 11X. Tian, P. Yang, Y. Yi, P. Liu, T. Wang, C. Shu, L. Qu, W. Tang, Y. Zhang, M. Li and B. Yang, *Journal of Power Sources*, 2020, **450**, 227629.
- 12H. Zhang, J. Chen, J. Liu, X. Zhang, J. Yang, Y. Nuli, H. Ma and J. Wang, *Energy Storage Materials*, 2023, **61**, 102885.
- 13K. Deng, S. Zhou, Z. Xu, M. Xiao and Y. Meng, *Chemical Engineering Journal*, 2022, **428**, 131224.
- 14P. Guo, A. Su, Y. Wei, X. Liu, Y. Li, F. Guo, J. Li, Z. Hu and J. Sun, *ACS Appl. Mater. Interfaces*, 2019, **11**, 19413–19420.
- 15A. L. Mong and D. Kim, *Energy Storage Materials*, 2022, **47**, 394–407.
- 16F. Fu, Y. Zheng, N. Jiang, Y. Liu, C. Sun, A. Zhang, H. Teng, L. Sun and H. Xie, *Chemical Engineering Journal*, 2022, **450**, 137776.
- 17X. Zhang, H. Zhang, Y. Geng, Z. Shi, S. Zhu, Q. Xu and Y. Min, *Chemical Engineering Journal*, 2022, **444**, 136328.
- 18K. Fan, X. Lai, Z. Zhang, L. Chai, Q. Yang, G. He, S. Liu, L. Sun, Y. Zhao, Z. Hu and L. Wang, *Journal of Power Sources*, 2023, **580**, 233342.
- 19Y. Ma, Y. Jiao, Y. Yan, W. Chen, Y. Li, M. Zhou, D. Chen and J. Zhu, *Journal of Power Sources*, 2022, **548**, 232034.
- 20L. Chen and L. Fan, *Energy Storage Materials*, 2018, **15**, 37–45.
- 21Y. Tao, W. Wei, Q. Gu, X. Jiang and D. Li, *Materials Letters*, 2023, **351**, 134934.
- 22S. Luo, N. Deng, H. Wang, Q. Zeng, Y. Li, W. Kang and B. Cheng, *Chemical Engineering Journal*, 2023, **474**, 145683.
- 23W. Bao, Z. Hu, Y. Wang, J. Jiang, S. Huo, W. Fan, W. Chen, X. Jing, X. Long and Y. Zhang, *Chemical Engineering Journal*, 2022, **437**, 135420.

- 24 J. Ma, M. Zhang, C. Luo, M. Li, X. Guan, F. Chen and X. Ma, *Solid State Ionics*, 2021, **363**, 115606.
- 25 X. Zhang, T. Zhang, Y. Shao, H. Cao, Z. Liu, S. Wang and X. Zhang, *ACS Sustainable Chem. Eng.*, 2021, **9**, 5396–5404.
- 26 Y. Ji, K. Yang, M. Liu, S. Chen, X. Liu, B. Yang, Z. Wang, W. Huang, Z. Song, S. Xue, Y. Fu, L. Yang, T. S. Miller and F. Pan, *Advanced Functional Materials*, 2021, **31**, 2104830.
- 27 P. Chiochan, X. Yu, M. Sawangphruk and A. Manthiram, *Advanced Energy Materials*, 2020, **10**, 2001285.
- 28 X. Wang, X. Hao, Z. Hengjing, X. Xia and J. Tu, *Electrochimica Acta*, 2020, **329**, 135108.
- 29 S. Huang, Y. Wang, J. Hu, Y. Von Lim, D. Kong, L. Guo, Z. Kou, Y. Chen and H. Y. Yang, *NPG Asia Materials*, 2019, **11**, 55.
- 30 F. Chen, Y. Zhang, Q. Hu, S. Cao, S. Song, X. Lu and Q. Shen, *Journal of Solid State Chemistry*, 2021, **301**, 122341.
- 31 H. Du, S. Li, H. Qu, B. Lu, X. Wang, J. Chai, H. Zhang, J. Ma, Z. Zhang and G. Cui, *Journal of Membrane Science*, 2018, **550**, 399–406.
- 32 S. Chen, B. Ding, Q. Lin, Y. Shi, B. Hu, Z. Li, H. Dou and X. Zhang, *Journal of Electroanalytical Chemistry*, 2021, **880**, 114874.
- 33 Y. Wang, G. Wang, P. He, J. Hu, J. Jiang and L.-Z. Fan, *Chemical Engineering Journal*, 2020, **393**, 124705.
- 34 G. Liu, J. Shi, M. Zhu, W. Weng, L. Shen, J. Yang and X. Yao, *Energy Storage Materials*, 2021, **38**, 249–254.

35A. Santiago, J. Castillo, I. Garbayo, A. Saenz de Buruaga, J. A. Coca Clemente, L. Qiao, R. Cid Barreno, M. Martinez-Ibañez, M. Armand, H. Zhang and C. Li, *ACS Appl. Energy Mater.*, 2021, **4**, 4459–4464.