

Electronic Supplementary Information

**Revisiting Polymeric Single Lithium-Ion Conductors as an Organic Route for  
All-Solid-State Lithium Ion and Metal Batteries**

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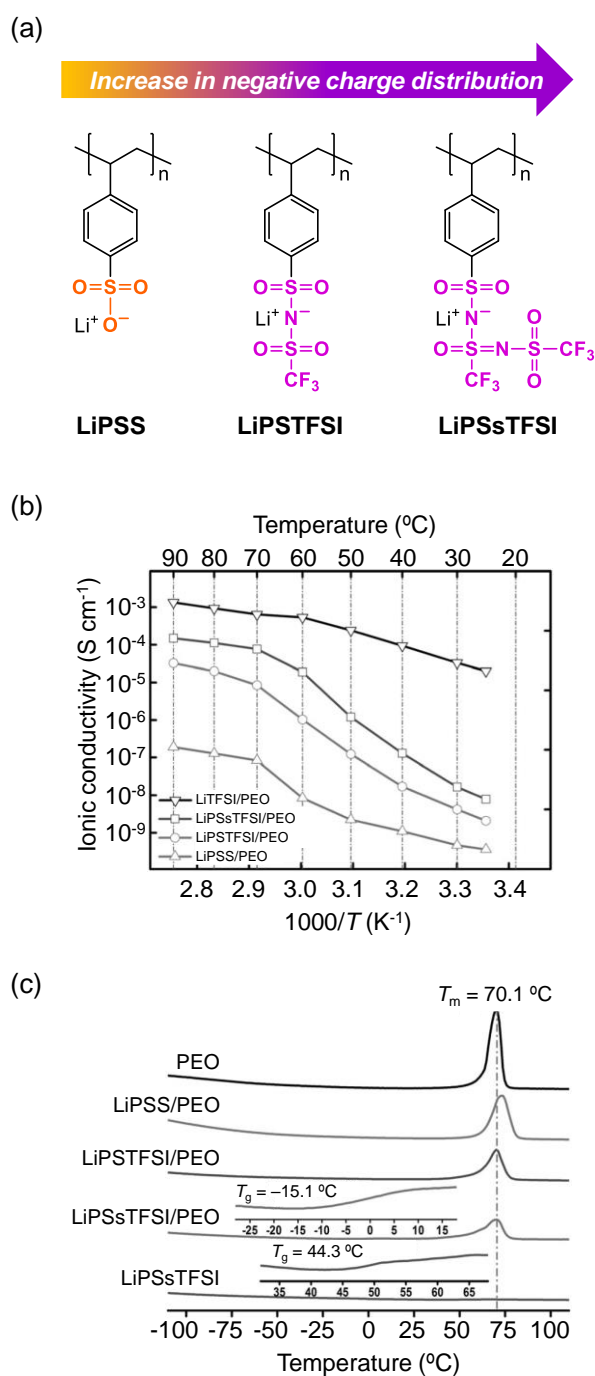
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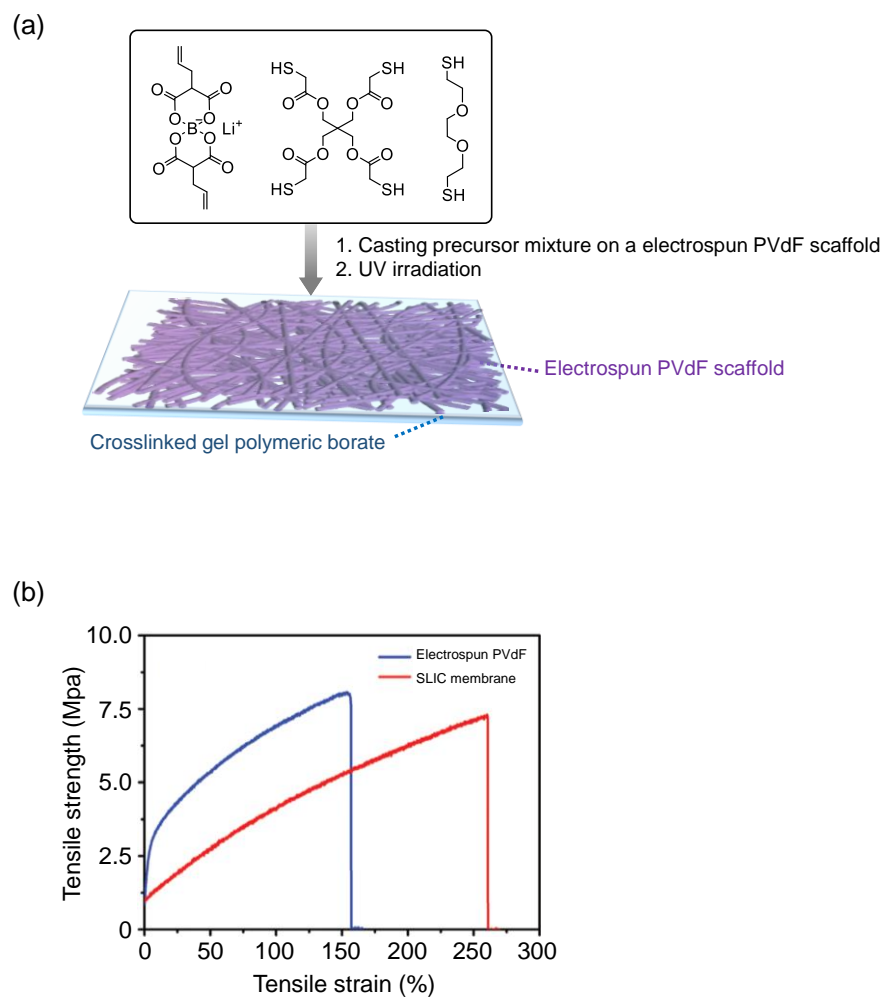
**Fig. S7** Change in interfacial resistance as a function of time for the Li|Li symmetric cells with/without CBP additive.....S6

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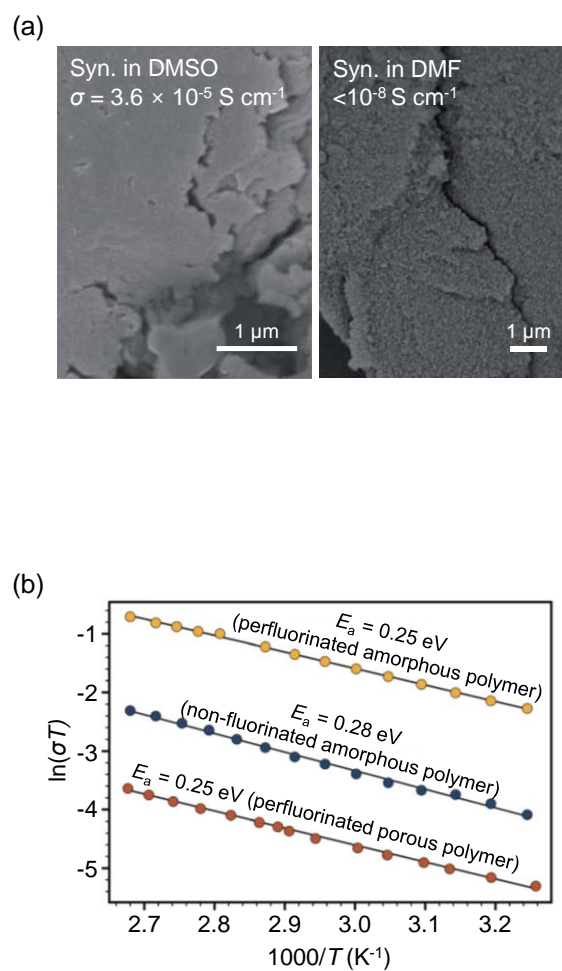
## Figures



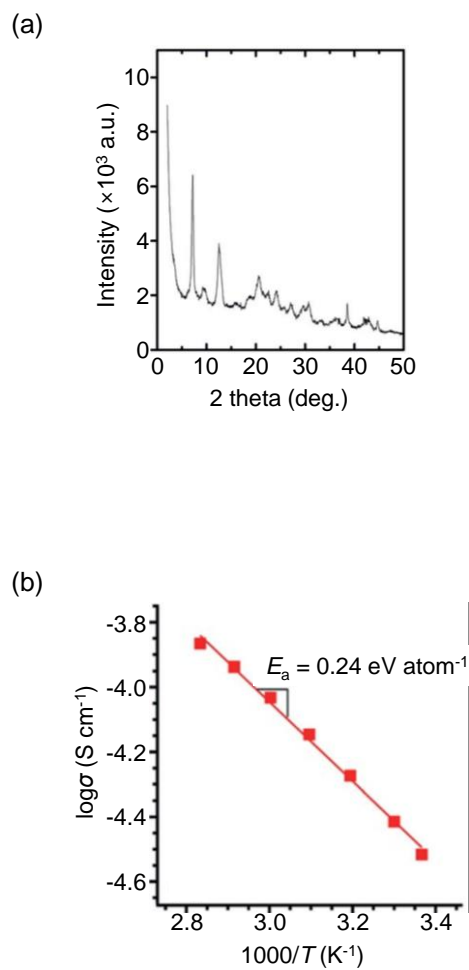
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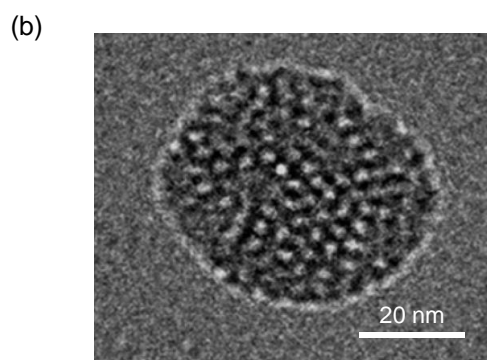
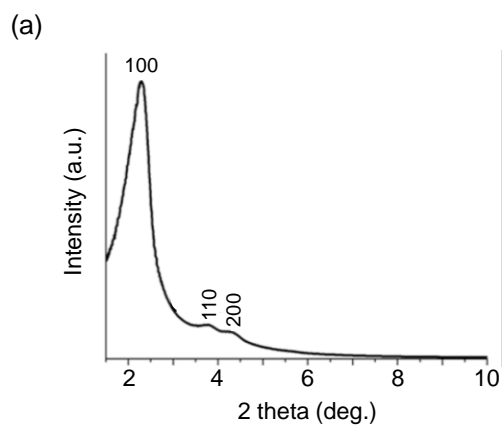
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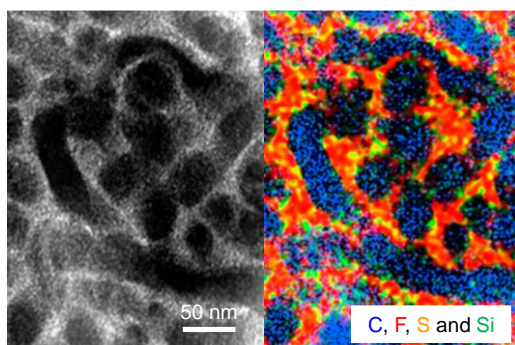
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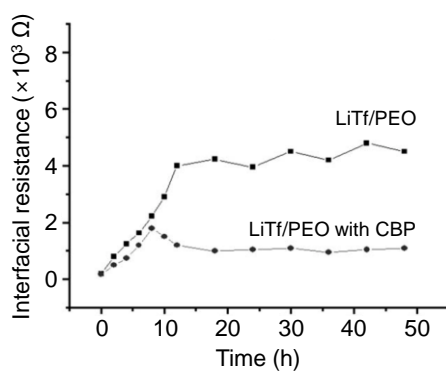
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**Fig. S6** STEM image of the silsesquioxane-based hybrid/SEO blend electrolyte along with composite elemental mapping for carbon (C; blue), fluorine (F; red), sulfur (S; yellow) and silicon (Si; green). Reprinted with permission from ref. S6. Copyright 2017 American Chemical Society.



**Fig. S7** Change in interfacial resistance as a function of time for the Li|Li symmetric cells with/without CBP additive. Reprinted with permission from ref. S7. Copyright 2010 Wiley Periodicals, Inc.



## References

- S1 Q. Ma, H. Zhang, C. Zhou, L. Zheng, P. Cheng, J. Nie, W. Feng, Y.-S. Hu, H. Li, X. Huang, L. Chen, M. Armand and Z. Zhou, *Angew. Chem. Int. Ed.*, 2016, **55**, 2521-2525.
- S2 K. Deng, J. Qin, S. Wang, S. Ren, D. Han, M. Xiao and Y. Meng, *Small*, 2018, **14**, 1801420.
- S3 J. F. Van Humbeck, M. L. Aubrey, A. Alsbaiee, R. Ameloot, G. W. Coates, W. R. Dichtel and J. R. Long, *Chem. Sci.*, 2015, **6**, 5499-5505.
- S4 Y. Du, H. Yang, J. M. Whiteley, S. Wan, Y. Jin, S.-H. Lee and W. Zhang, *Angew. Chem. Int. Ed.*, 2016, **55**, 1737-1741.
- S5 Y. Kim, S. J. Kwon, H. Jang, B. M. Jung, S. B. Lee and U. H. Choi, *Chem. Mater.*, 2017, **29**, 4401-4410.
- S6 I. Villaluenga, S. Inceoglu, X. Jiang, X. C. Chen, M. Chintapalli, D. R. Wang, D. Devaux and N. P. Balsara, *Macromolecules*, 2017, **50**, 1998-2005.
- S7 A. M. Stephan, T. Prem Kumar, N. Angulakshmi, P. S. Salini, R. Sabarinathan, A. Srinivasan and S. Thomas, *J. Appl. Polym. Sci.*, 2011, **120**, 2215-2221.