**Supplementary Material**

# Anti-freezing, high-voltage, and ultra-stable micro-supercapacitors enabled by phase engineering of 2D $SiP_2$ in LiCl-DMSO electrolyte

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**Table S1** Comparison of electrochemical performance of representative nano-electrode materials for MSCs

| Electrode materials* | $C_A$ (mF $cm^{-2}$) | Energy density (μWh $cm^{-2}$) | Power density (mW $cm^{-2}$) | Capacitance retention (%) | Cycle stability | Ref. |
|---|---|---|---|---|---|---|
| $B_4C$@C | 7.33 | 1.02 | 1.09 | 100.94 | 50,000 | 1 |
| PG-MSCs | 9.8 | - | - | 89.5 | 2000 | 2 |
| LIG-MSCs | 7.12 | 0.63 | - | 100 | 10,000 | 3 |
| $MoS_2$ | 8 | - | - | 92 | 1000 | 4 |
| PEDOT MSCs | 9 | - | - | 80 | 10,000 | 5 |
| G-CNTs | 9.81 | 1.36 | 0.25 | 95.5 | 10,000 | 6 |
| 3D GA | 12.6 | - | - | - | - | 7 |
| MXene@EG | 3.26 | - | - | 82 | 2500 | 8 |
| TP@EG | 3.42 | - | - | 86 | 10,000 | 9 |
| Graphene | 1.324 | 0.64 | 20 | 98 | 10,000 | 10 |
| (Ta, Ti, Nb)C/C | 41.6 | 5.8 | 0.04 | 92.6 | 9000 | 11 |
| LSG/Ni-CAT MOF | 15.2 | 4.1 | 7 | 87 | 5000 | 12 |
| rGO/Ag-NW | 8.6 | 0.669 | 0.73 | - | - | 13 |
| Pnma $SiP_2$ | 12.39 | 6.86 | 18.26 | 97.89<br>82.37 | 20,000<br>30,000 | This work |

*PG: Phosphorene and graphene, LIG: Laser-induced graphene, PEDOT: Poly(3,4-

ethylenedioxythiophene) electrodes, G-CNTs: Graphene-CNT, 3D GA: 3D-printed graphene aerogel, MXene@EG: $Ti_3C_2T_x$@electrochemically exfoliated graphene, TP@EG: Thiophene nanosheets and electrochemically exfoliated graphene, LSG/Ni-CAT MOF: Ni-catecholate-based MOF on 3D laser-scribed graphene, rGO/Ag-NW: Reduced graphene oxide /silver nanowire hybrid electrodes.

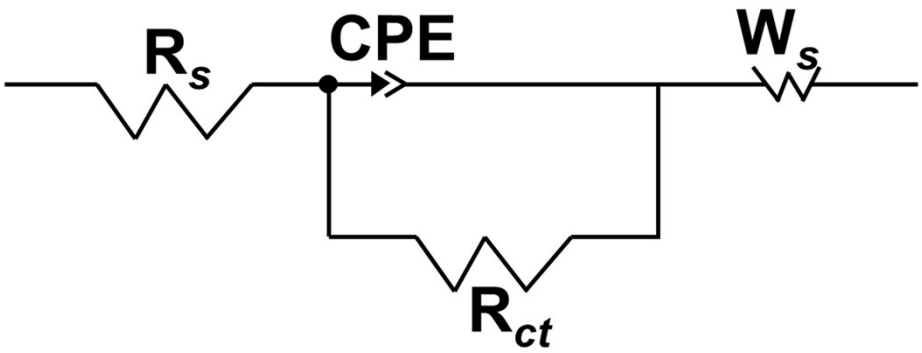


**Fig. S1** Equivalent circuit model for micro-supercapacitor testing.

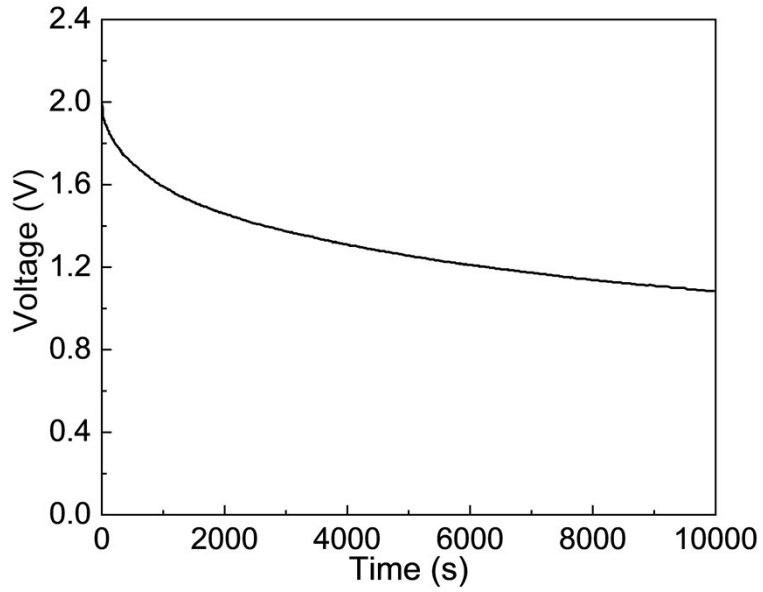


**Fig. S2** Self-discharge curve under a 2 V voltage window.

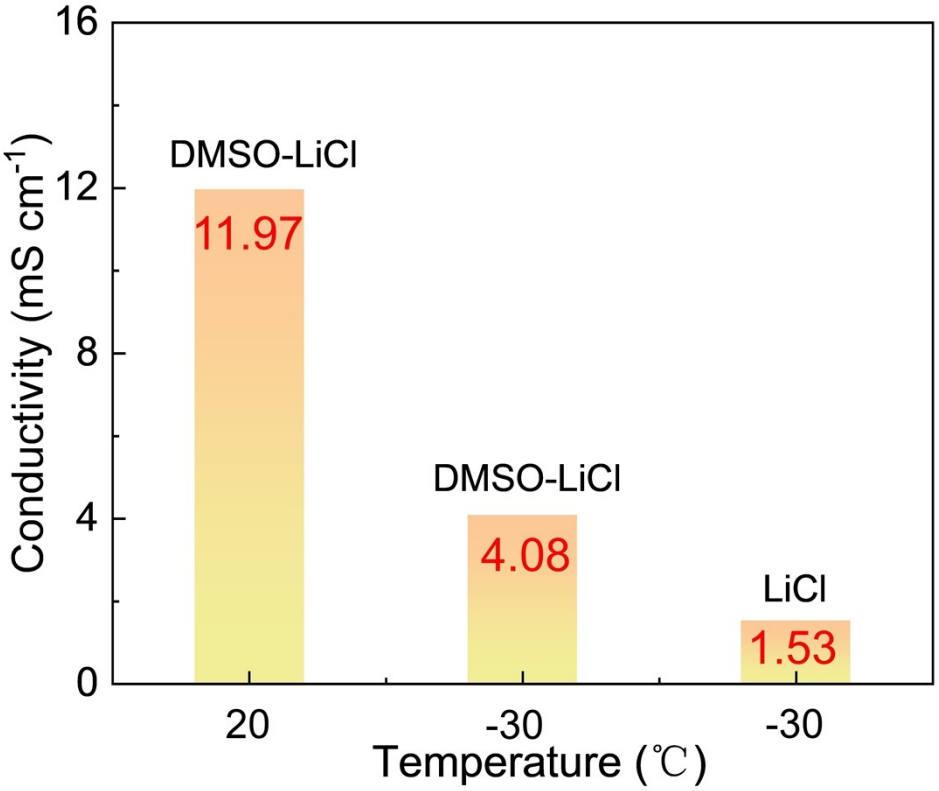


**Fig. S3** Ionic conductivities of different gel electrolytes at various temperatures.

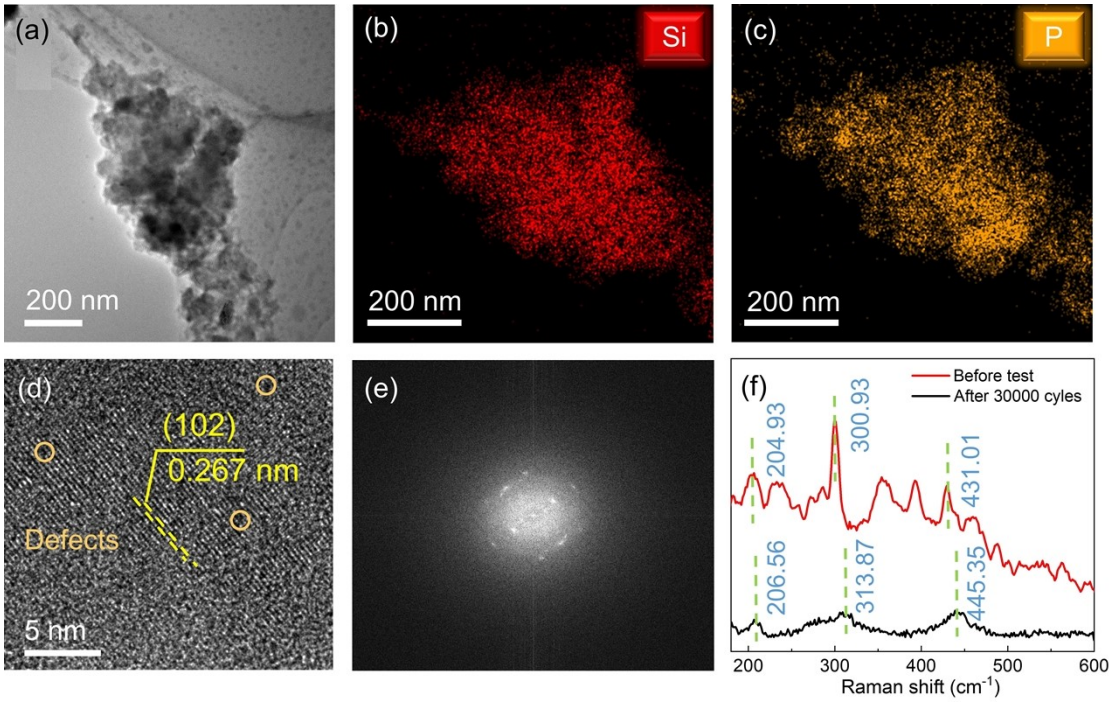


**Fig. S4** (a) TEM images of Pnma-$SiP_2$ after cycling, along with the corresponding elemental distribution maps of (b) Si and (c) P. (d) High-resolution TEM images of Pnma-$SiP_2$ after cycling. (e) Fourier transform pattern of Pnma-$SiP_2$. (f) Raman spectra of Pnma-$SiP_2$ before and after cycling.

**References**


1. Y. Chang, X. Sun, M. Ma, C. Mu, P. Li, L. Li, M. Li, A. Nie, J. Xiang and Z. Zhao, *Nano Energy*, 2020, **75**, 104947.
2. H. Xiao, Z. S. Wu, L. Chen, F. Zhou, S. Zheng, W. Ren, H. M. Cheng and X. Bao, *ACS Nano*, 2017, **11**, 7284-7292.
3. T. Liu, R. Ren, Z. Qi, J. Hu, Y. Chen, Y. Huang, Y. Guo, H. Cao, M. Liang and J. Sun, *Journal of Power Sources*, 2024, **602**, 234307.
4. L. Cao, S. Yang, W. Gao, Z. Liu, Y. Gong, L. Ma, G. Shi, S. Lei, Y. Zhang and S. Zhang, *small*, 2013, **9**, 2905-2910.
5. N. Kurra, M. K. Hota and H. N. Alshareef, *Nano Energy*, 2015, **13**, 500-508.
6. Y. Wang, Y. Zhang, G. Wang, X. Shi, Y. Qiao, J. Liu, H. Liu, A. Ganesh and L. Li, *Advanced Functional Materials*, 2020, **30**, 1907284.
7. B. Yao, S. Chandrasekaran, H. Zhang, A. Ma, J. Kang, L. Zhang, X. Lu, F. Qian, C. Zhu and E. B. Duoss, *Advanced Materials*, 2020, **32**, 1906652.
8. H. Li, Y. Hou, F. Wang, M. R. Lohe, X. Zhuang, L. Niu and X. Feng, *Advanced Energy Materials*, 2017, **7**, 1601847.
9. Z. Wu, Y. Zheng, S. Zheng, S. Wang, C. Sun, K. Parvez, T. Ikeda, X. Bao, K. Müllen and X. Feng, *Advanced Materials*, 2017, **29**, 1602960.
10. S. Bellani, E. Petroni, A. E. Del Rio Castillo, N. Curreli, B. Martín-García, R. Oropesa-Nuñez, M. Prato and F. Bonaccorso, *Advanced Functional Materials*, 2019, **29**, 1807659.
11. X. Li, B. Yang, X. Zhang, S. Duan, X. Jiang, M. Ma, W. Yi, H. Sun, Z. Wang

and Y. Chu, *Journal of Power Sources*, 2023, **568**, 232986.

12. H. Wu, W. Zhang, S. Kandambeth, O. Shekhah, M. Eddaoudi and H. N. Alshareef, *Advanced Energy Materials*, 2019, **9**, 1900482.

13. G. Huang, N. Li, Y. Du, Q. Feng, H. Xiao, X. Wu and S. Fu, *ACS Applied Materials & Interfaces*, 2018, **10**, 723-732.