

Electronic Supplementary Information

A Low-Self-Discharge High-Loading Polysulfide Cathode Design for Lithium–Sulfur Cells

Cheng-Che Wu^{‡a}, Yun-Chung Ho^{‡a} and Sheng-Heng Chung^{*ab}

^a Department of Materials Science and Engineering, National Cheng Kung University, No.1 University Road, Tainan City 70101, Taiwan

^b Hierarchical Green-Energy Materials Research Center, National Cheng Kung University, No.1 University Road, Tainan City 70101, Taiwan

[‡]Equal contribution

*Sheng-Heng Chung: SHChung@gs.ncku.edu.tw

Supporting Figures

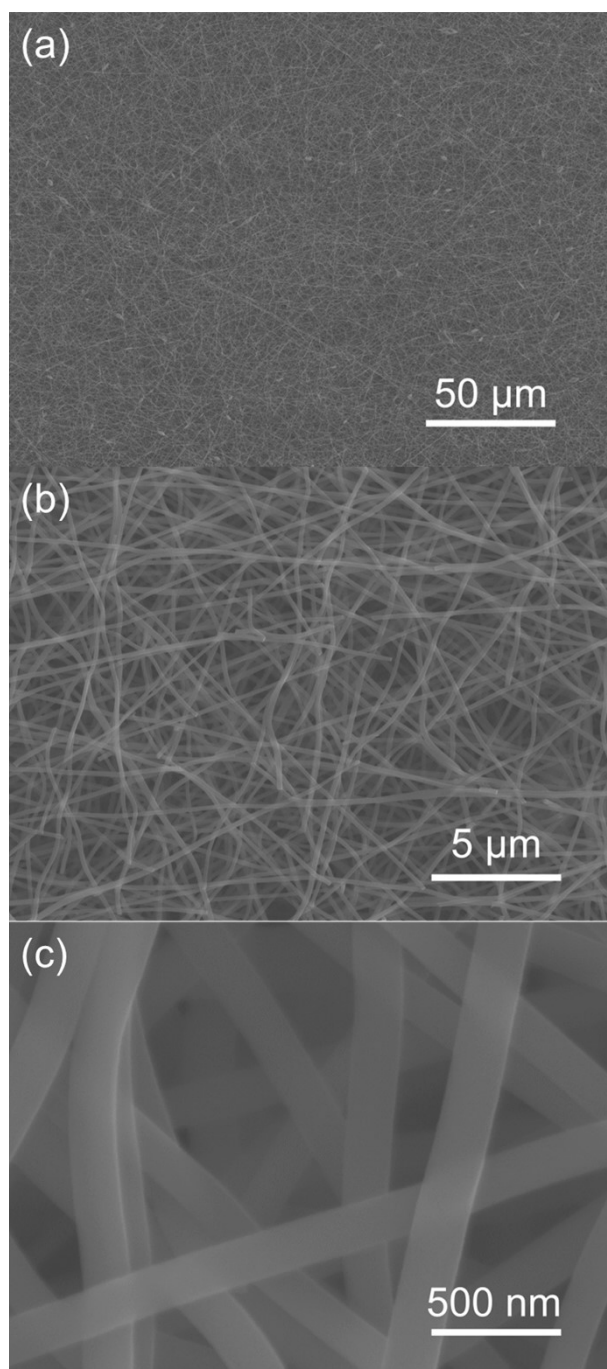


Fig. S1. SEM morphological observation of carbonized electrospun substrate (cPAN) at various magnifications.

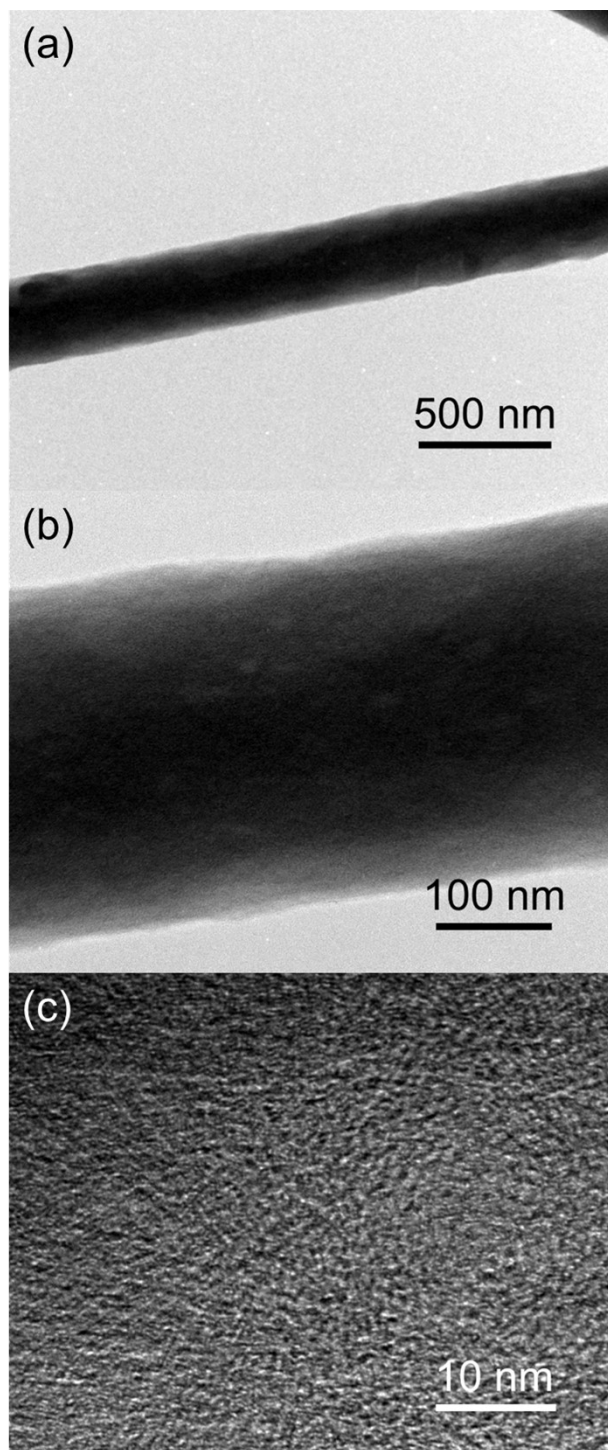


Fig. S2. TEM microstructural observation of carbonized electrospun substrate (cPAN) at various magnifications.

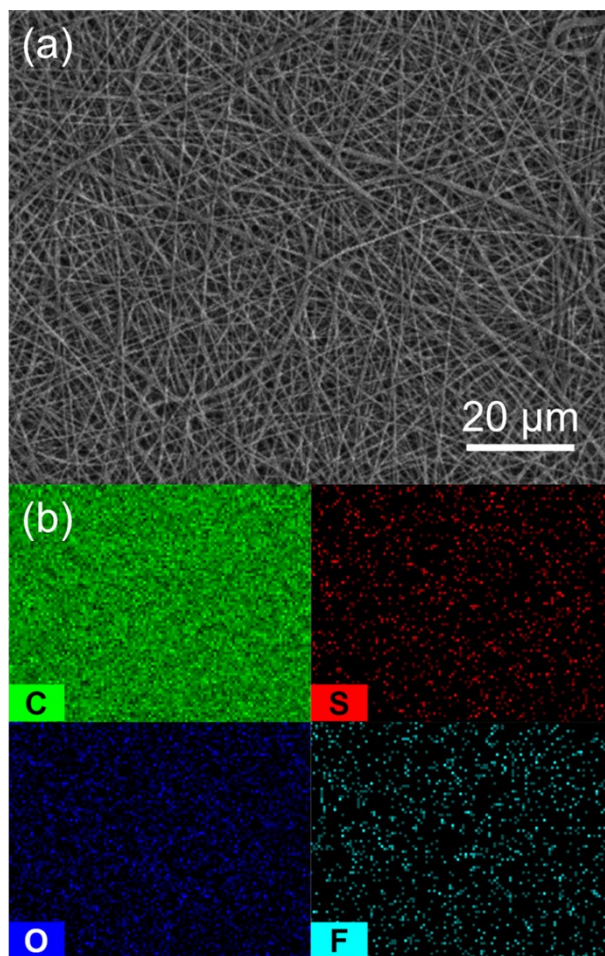


Fig. S3. SEM/EDS elemental mapping of carbonized electrospun substrate (cPAN) at low magnification.

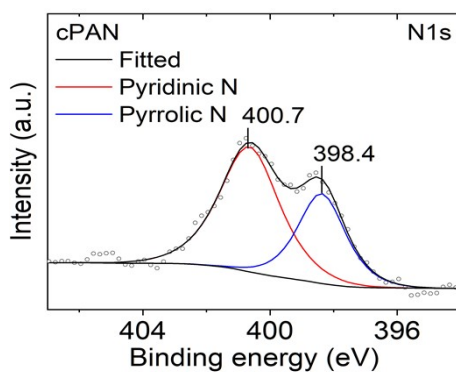


Fig. S4. Deconvolution of N1s spectra of the XPS analysis of carbonized electrospun substrate (cPAN).

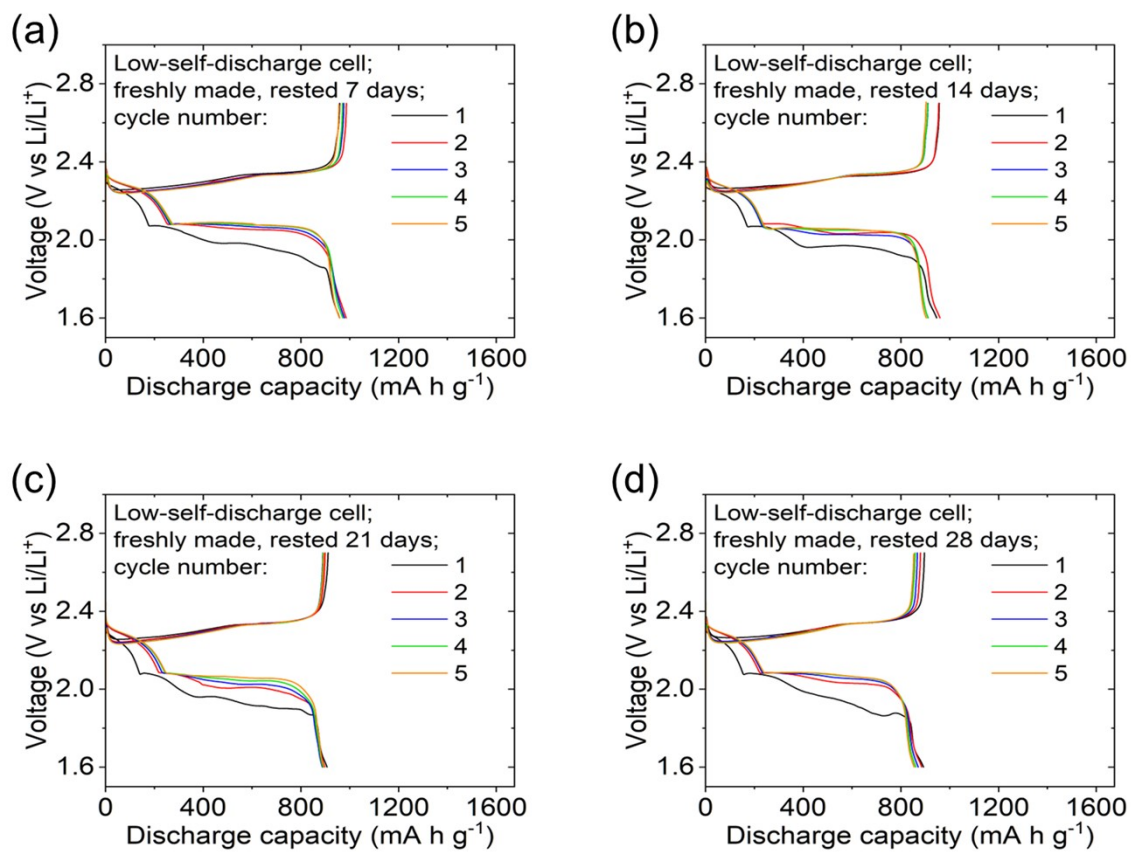


Fig. S5. Charge–discharge voltage profiles of the freshly made cells after resting for (a) 7, (b) 14, (c) 21, and (d) 28 days.

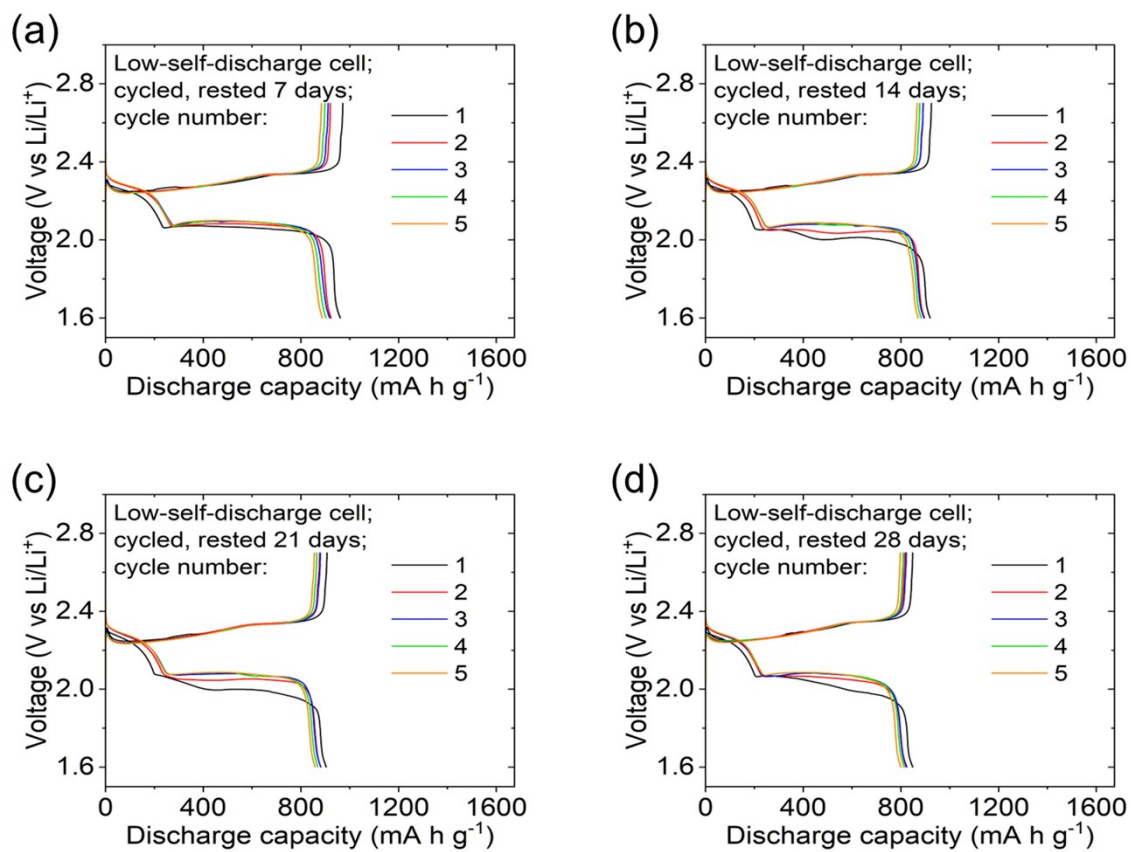


Fig. S6. Charge–discharge voltage profiles of the cycled cells after resting for (a) 7, (b) 14, (c) 21, and (d) 28 days.

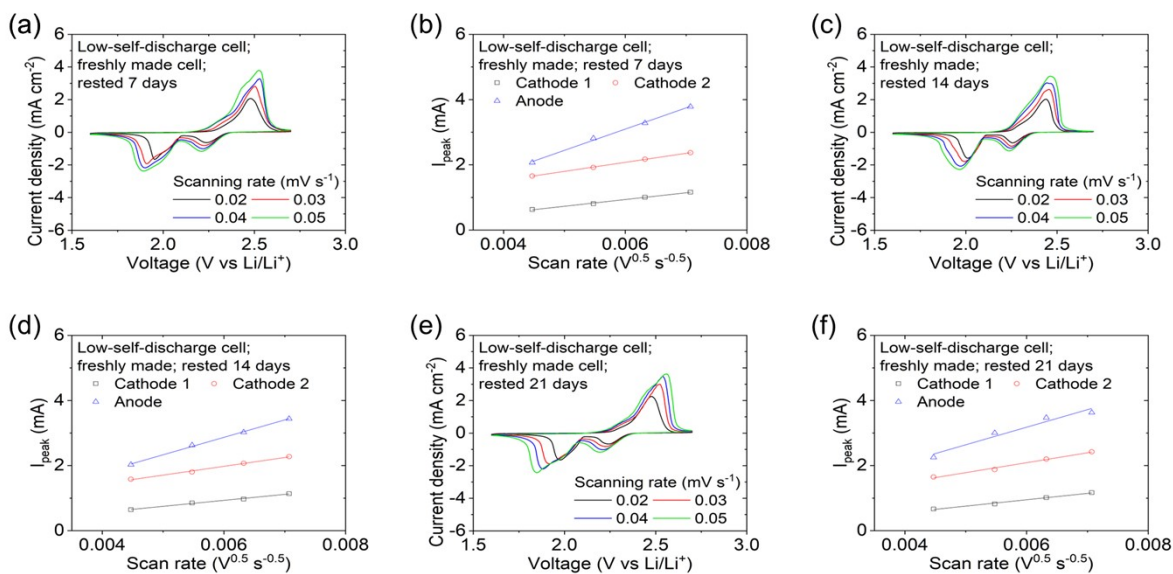


Fig. S7. Cyclic voltammetry (CV) curves and lithium-ion diffusion coefficients of the freshly made cells after resting for (a,b) 7, (c,d) 14, and (e,f) 21 days.

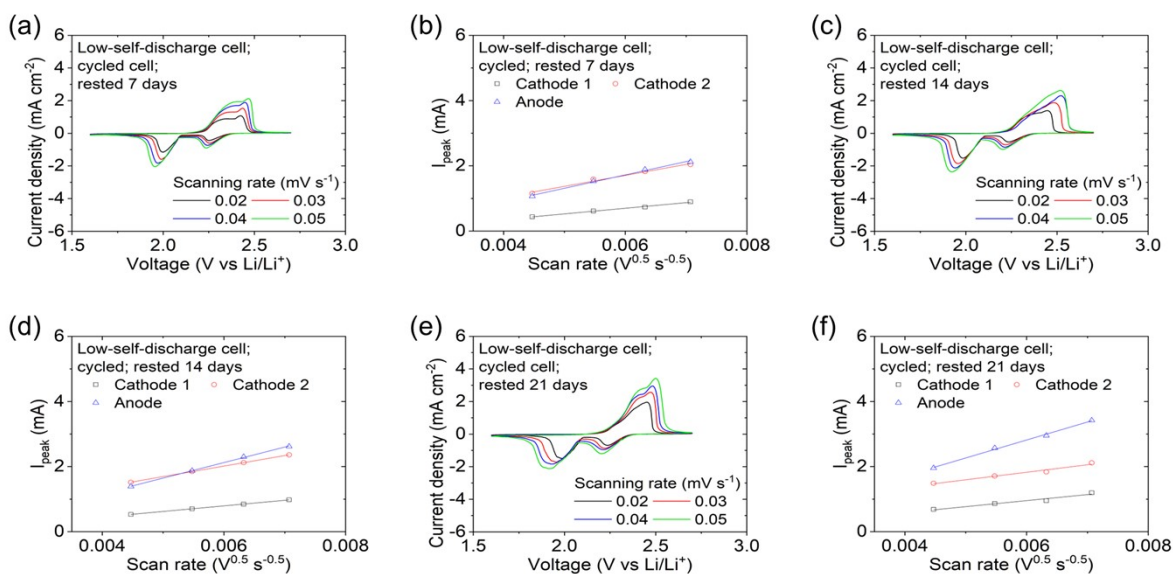


Fig. S8. Cyclic voltammetry (CV) curves and lithium-ion diffusion coefficients of the cycled cells after resting for (a,b) 7, (c,d) 14, and (e,f) 21 days.

Supporting Tables

Table S1. Lithium-ion diffusion coefficients of the low-self-discharge cells at various static and dynamic storage times for 7, 14, 21, and 28 days.

Cell condition	C_1 ($\text{cm}^2 \text{s}^{-1}$)	C_2 ($\text{cm}^2 \text{s}^{-1}$)	A ($\text{cm}^2 \text{s}^{-1}$)
freshly made reference cell	2.4×10^{-8}	6.4×10^{-8}	2.2×10^{-7}
cycled reference cell	1.3×10^{-8}	3.0×10^{-8}	2.4×10^{-7}
7 days static storage	2.2×10^{-8}	3.9×10^{-8}	2.1×10^{-7}
7 days dynamic storage	1.5×10^{-8}	5.7×10^{-8}	8.4×10^{-8}
14 days static storage	1.7×10^{-8}	3.7×10^{-8}	1.5×10^{-7}
14 days dynamic storage	1.5×10^{-8}	5.3×10^{-8}	1.2×10^{-7}
21 days static storage	1.9×10^{-8}	4.7×10^{-8}	1.5×10^{-7}
21 days dynamic storage	1.7×10^{-8}	2.7×10^{-8}	1.5×10^{-7}
28 days static storage	2.2×10^{-8}	8.2×10^{-8}	1.3×10^{-7}
28 days dynamic storage	1.5×10^{-8}	4.6×10^{-8}	1.1×10^{-7}

Table S2. Self-discharge performance of various lithium–sulfur cells.

Sulfur loading (mg cm^{-2})	Sulfur content (wt%)	Electrolyte-to-sulfur ratio ($\mu\text{L mg}^{-1}$)	Resting time (day)	Capacity-retention rate (%)	Time-dependent capacity-fade rate (% per day)	Cycle rate (C)	Battery status	year	Ref.
0.90	64.0	n/a	7	96.10	0.56	0.2	fresh, charged	2017	S1
1.00	56.0	30	10	70.00	3.00	0.1	cycled, charged	2017	S2
2.00	50.0	n/a	3	97.53	0.82	0.2	cycled, discharged	2017	S3
1.00	n/a	n/a	4	90.60	2.35	1	cycled, discharged	2017	S4
3.10	62.4	8	1	94.16	5.84	0.1	cycled, charged	2018	S5
5.20	62.4	8	1	93.89	6.11	0.1	cycled, charged	2018	S5
6.50	62.4	8	1	89.80	10.20	0.1	cycled, charged	2018	S5
1.50	56.0	gel	3	90.70	3.10	0.1	cycled, charged	2018	S6
1.20	70.0	22	3	91.20	2.93	0.5	cycled,	2018	S7

1.20	70.0	22	3	87.70	4.10	0.5	discharged cycled, discharged	2018	S7
1.00	57.8	n/a	7	91.00	1.29	0.2	cycled, charged	2018	S8
1.40	50.0	25	3	84.40	5.20	0.2	cycled, charged	2018	S9
0.50	50.4	500	1.67	98.50	0.90	0.1	fresh, charged	2018	S10
1.80	72.0	22	3	95.10	1.63	n/a	cycled, charged	2018	S11
1.00	60.0	40	3	90.90	3.03	2	cycled, discharged	2019	S12
1.20	56.0	25	4	92.80	1.80	0.2	cycled, charged	2019	S13
8.60	70.0	9	30	90.00	0.33	0.1	cycled, discharge	2019	S14
1.50	41.3	18	10	93.60	0.64	0.05	cycled, charged	2019	S15
1.40	64.0	32	2	86.40	6.80	0.1	cycled, charged	2019	S16
2.30	70.0	15	60	95.80	0.07	0.2	cycled, charged	2019	S17
3.70	70.0	15	60	98.20	0.03	0.2	cycled, charged	2019	S17
1.60	69.6	12.5	10	89.76	1.02	0.2	fresh, charged	2019	S18
1.60	69.6	12.5	30	64.85	1.17	0.2	fresh, charged	2019	S18
1.60	69.6	12.5	50	54.97	0.90	0.2	fresh, charged	2019	S18
1.50	60.0	n/a	30	92.70	0.24	0.2	cycled, charged	2019	S19
1.30	64.0	15	3	94.30	1.90	0.5	cycled, discharged	2019	S20
2.50	70.0	10	30	89.70	0.34	1	cycled, charged	2020	S21
1.50	75.0	70	0.17	94.72	31.06	0.5	cycled, charged	2020	S22
1.50	75.0	70	1	87.70	12.30	0.5	cycled, charged	2020	S22
4.74	n/a	n/a	20	98.31	0.08	0.5	cycled, charged	2020	S23
3.00	60.0	20	2	97.08	1.46	0.5	cycled, charged	2020	S24
3.00	70.0	15	20	85.70	0.72	1	cycled, charged	2020	S25
1.40	60.0	n/a	2	94.80	2.60	0.2	fresh, charged	2020	S26
2.00	51.0	30	60	88.60	0.19	n/a	fresh, charged	2020	S27
1.91	n/a	n/a	4	97.00	0.75	0.1	cycled, charged	2020	S28
4.00	75.0	10	4.17	88.40	2.78	0.1	cycled, charged	2021	S29
1.00	60.0	15	3	98.62	0.46	0.2	cycled, charged	2021	S30
1.50	56.0	15	5	91.80	1.64	n/a	cycled, charged	2021	S31
1.00	56.0	35	12	88.66	0.94	1	fresh, charged	2022	S32

n/a	72.3	n/a	2	90.85	4.58	0.2	cycled, charged	2022	S33
1.00	56.0	n/a	7	95.10	0.70	0.5	cycled, charged	2023	S34
1.00	60.0	20	2	96.04	1.98	0.5	cycled, discharged	2023	S35
1.50	56.0	16.7	5	96.63	0.67	n/a	fresh, charged	2023	S36
1.50	56.0	16.7	1	99.69	0.31	n/a	cycled, charged	2023	S36
4.03	66.8	10	7	93.01	1.00	0.1	fresh, charged	this paper	
4.03	66.8	10	14	91.84	0.58	0.1	fresh, charged	this paper	
4.03	66.8	10	21	88.06	0.57	0.1	fresh, charged	this paper	
4.03	66.8	10	28	86.60	0.48	0.1	fresh, charged	this paper	
4.03	66.8	10	60	82.91	0.28	0.1	fresh, charged	this paper	
4.03	66.8	10	90	76.80	0.26	0.1	fresh, charged	this paper	

Supporting References

- S01 Y. Pang, Y. Wen, W. Li, Y. Sun, T. Zhu, Y. Wang and Y. Xia, *J. Mater. Chem. A*, 2017, **5**, 17926-17932.
- S02 Z. Lin, X. Li, W. Huang, X. Zhu, Y. Wang and Z. Shan, *ChemElectroChem*, 2017, **4**, 2577-2582.
- S03 F. Liu, Q. Xiao, H. B. Wu, F. Sun, X. Liu, F. Li, Z. Le, L. Shen, G. Wang, M. Cai and Y. Lu, *ACS Nano*, 2017, **11**, 2697-2705.
- S04 X. Hong, J. Jin, T. Wu, Y. Lu, S. Zhang, C. Chen and Z. Wen, *J. Mater. Chem. A*, 2017, **5**, 14775-14782.
- S05 L. Ma, H. Lin, W. Zhang, P. Zhao, G. Zhu, Y. Hu, R. Chen, Z. Tie, J. Liu and Z. Jin, *Nano Lett.*, 2018, **18**, 7949-7954.
- S06 T. Chen, W. Kong, Z. Zhang, L. Wang, Y. Hu, G. Zhu, R. Chen, L. Ma, W. Yan, Y. Wang, J. Liu and Z. Jin, *Nano Energy*, 2018, **54**, 17-25.
- S07 K. Yang, L. Zhong, Y. Mo, R. Wen, M. Xiao, D. Han, S. Wang and Y. Meng, *ACS Appl. Energy Mater.*, 2018, **1**, 2555-2564.
- S08 L. Yan, N. Luo, W. Kong, S. Luo, H. Wu, K. Jiang, Q. Li, S. Fan, W. Duan and J. Wang, *J. Power Sources*, 2018, **389**, 169-177.
- S09 S. Suriyakumar, A. M. Stephan, N. Angulakshmi, M. H. Hassan and M. H. Alkordi, *J. Mater. Chem. A*, 2018, **6**, 14623-14632.
- S10 K. Yang, L. Zhong, R. Guan, M. Xiao, D. Han, S. Wang and Y. Meng, *Appl. Surf. Sci.*, 2018, **441**, 914-922.

- S11 Y. Ansari, S. Zhang, B. Wen, F. Fan and Y.-M. Chiang, *Adv. Energy Mater.*, 2019, **9**, 1802213.
- S12 B. Zheng, L. Yu, Y. Zhao and J. Xi, *Electrochim. Acta*, 2019, **295**, 910-917.
- S13 T. Chen, W. Kong, M. Fan, Z. Zhang, L. Wang, R. Chen, Y. Hu, J. Ma and Z. Jin, *J. Mater. Chem. A*, 2019, **7**, 20302-20309.
- S14 R. Yu, S.-H. Chung, C.-H. Chen and A. Manthiram, *Energy Storage Mater.*, 2019, **18**, 491-499.
- S15 S. Majumder, M. Shao, Y. Deng and G. Chen, *J. Power Sources*, 2019, **431**, 93-104.
- S16 L. Dong, J. Liu, D. Chen, Y. Han, Y. Liang, M. Yang, C. Yang and W. He, *ACS Nano*, 2019, **13**, 14172-14181.
- S17 Z. Ye, Y. Jiang, J. Qian, W. Li, T. Feng, L. Li, F. Wu and R. Chen, *Nano Energy*, 2019, **64**, 103965.
- S18 J.-X. Lin, Y.-X. Mo, P.-F. Zhang, Y.-Y. Li, Y.-J. Wu, S.-J. Zhang, Z.-G. Gao, J.-D. Chen, W.-F. Ren, J.-T. Li, Y. Zhou, L. Huang and S.-G. Sun, *Mater. Today Energy*, 2019, **13**, 267-276.
- S19 Z. Sun, X.-L. Wu, Z. Peng, J. Wang, S. Gan, Y. Zhang, D. Han and L. Niu, *Small*, 2019, **15**, 1902491.
- S20 Q. Zhao, K. Zhao, G. Ji, X. Guo, M. Han, J. Wen, Z. Ren, S. Zhao, Z. Gao, R. Wang, M. Li, K. Sun, N. Hu and C. Xu, *Chem. Eng. J.*, 2019, **361**, 1043-1052.
- S21 M. Waqas, Y. Han, D. Chen, S. Ali, C. Zhen, C. Feng, B. Yuan, J. Han and W. He, *Energy Storage Mater.*, 2020, **27**, 333-341.

- S22 C. Choi, D.-Y. Lee, J. B. Park and D.-W. Kim, *ACS Sustainable Chem. Eng.*, 2020, **8**, 15134-15148.
- S23 Z. Wu, S. Yao, R. Guo, Y. Li, C. Zhang, X. Shen, T. Li and S. Qin, *Int. J. Energy Res.*, 2020, **44**, 3110-3121.
- S24 Q. Hu, J. Lu, C. Yang, C. Zhang, J. Hu, S. Chang, H. Dong, C. Wu, Y. Hong and L. Zhang, *Small*, 2020, **16**, 2002046.
- S25 M. Waqas, A. Manzoor Soomro, S. Ali, S. Kumar, S. Chan, K. Hussain, F. Hussain Memon and S. Ahmed Shaikh, *ChemistrySelect*, 2020, **5**, 12009-12019.
- S26 D. Capková, M. Al máši, T. Kazda, O. Čech, N. Király, P. Čudek, A. S. Fedorková and V. Hornebecq, *Electrochim. Acta*, 2020, **354**, 136640.
- S27 C. Y. Zhang, G. W. Sun, Y. F. Bai, Z. Dai, Y. R. Zhao, X. P. Gao, G. Z. Sun, X. B. Pan, X. J. Pan and J. Y. Zhou, *J. Mater. Chem. A*, 2020, **8**, 18358-18366.
- S28 Y. Li, C. Wang, W. Wang, A. Y. S. Eng, M. Wan, L. Fu, E. Mao, G. Li, J. Tang, Z. W. Seh and Y. Sun, *ACS Nano*, 2020, **14**, 1148-1157.
- S29 J. Liu, X. Liu, Q. Zhang, X. Liang, J. Yan, H. H. Tan, Y. Yu and Y. Wu, *Electrochim. Acta*, 2021, **382**, 138267.
- S30 C. Zhang, R. Du, J. J. Biendicho, M. Yi, K. Xiao, D. Yang, T. Zhang, X. Wang, J. Arbiol, J. Llorca, Y. Zhou, J. R. Morante and A. Cabot, *Adv. Energy Mater.*, 2021, **11**, 2100432.
- S31 J. Liu, K. Li, Q. Zhang, X. Zhang, X. Liang, J. Yan, H. H. Tan, Y. Yu and Y. Wu, *ACS Appl. Mater. Interfaces*, 2021, **13**, 45547-45557.
- S32 S. Xiao, L. Huang, W. Lv and Y.-B. He, *ACS Appl. Mater. Interfaces*, 2022, **14**, 1783-1790.

- S33 S.-N. Xu, T. Zhao, L.-L. Wang, Y.-X. Huang, Y.-S. Ye, N.-X. Zhang, T. Feng, L. Li, F. Wu and R.-J. Chen, *J. Energy Chem.*, 2022, **67**, 423-431.
- S34 T. Nie, Y. Zhu, M. Fang, L. Ma, J. Xu, Y. Cao, S. Hu, X. Zhang and D. Niu, *J. Colloid Interface Sci.*, 2023, **640**, 908-916.
- S35 C. Dong, F. Liu, Y. Yang, H. Zhao, H. Huo, J. Zhou and L. Li, *ACS Appl. Energy Mater.*, 2023, **6**, 3894-3902.
- S36 Y.-H. Liu, L.-X. Li, A.-Y. Wen, F.-F. Cao and H. Ye, *Energy Storage Mater.*, 2023, **55**, 652-659.