

Supporting Information

Selenium Infiltrated Hierarchical Hollow Carbon Spheres Display Rapid Kinetics and Extended Cycling as Lithium Metal Battery (LMB) Cathodes

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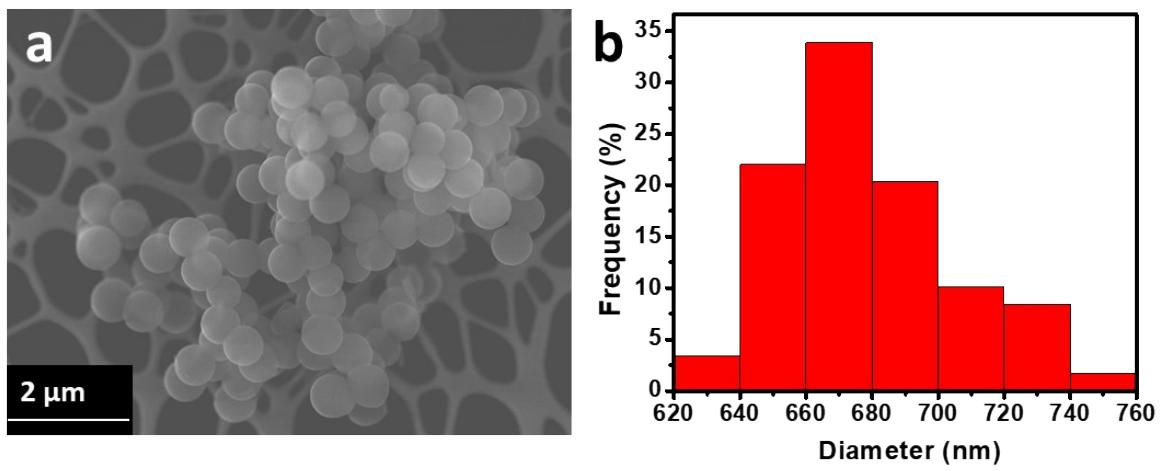


Figure S1. (a) SEM image and (b) size distribution of MCS.

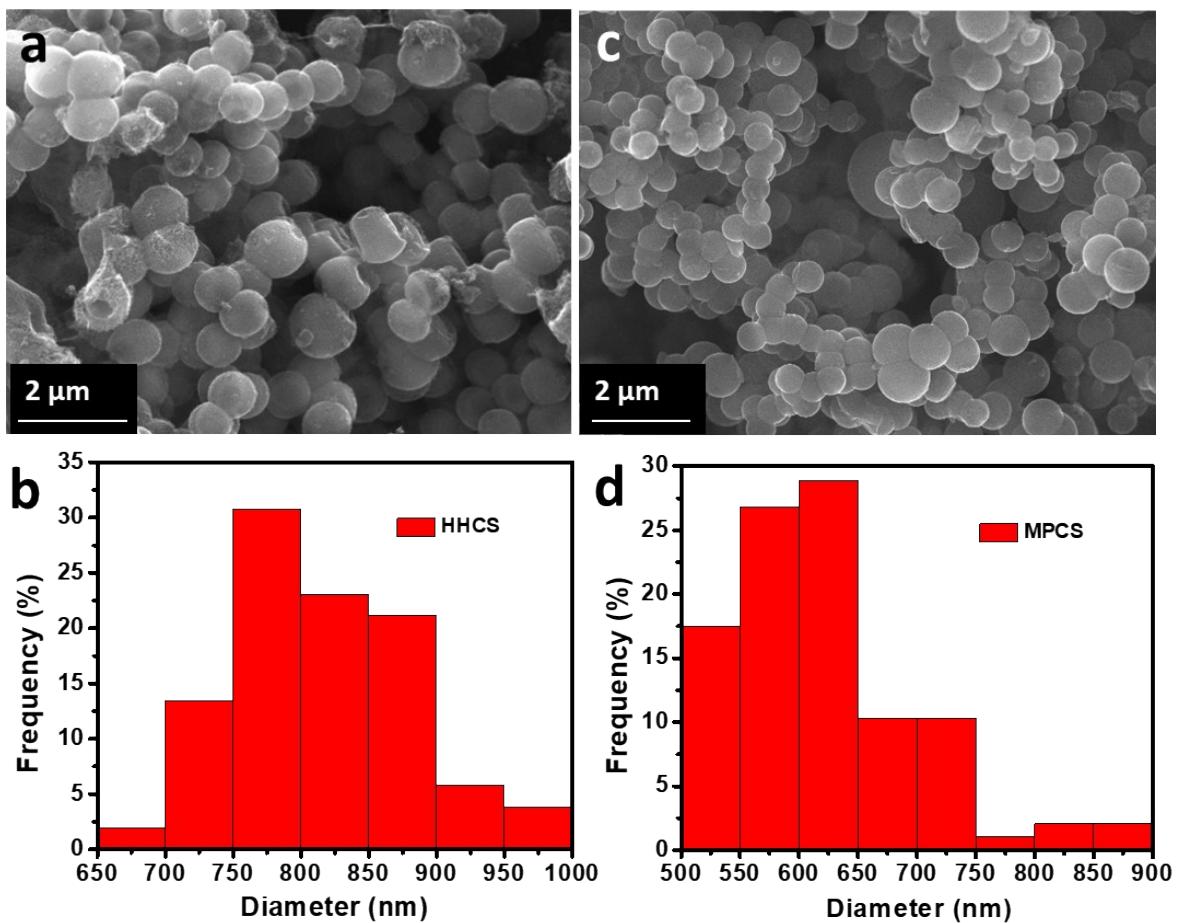


Figure S2. SEM images of **(a)** HHCS and **(b)** MPCS. Size distribution of **(c)** HHCS and **(d)** MPCS.

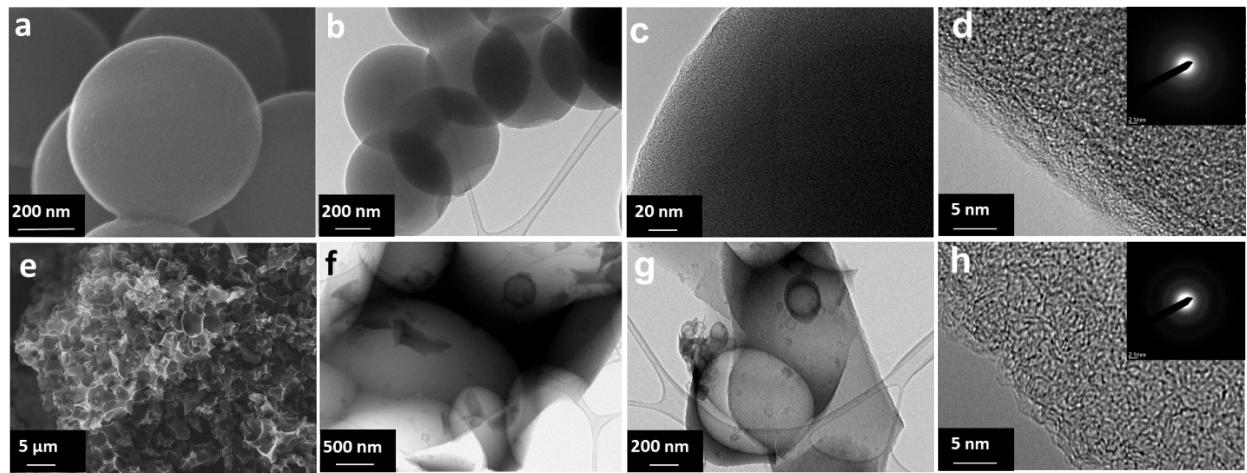


Figure S3. SEM images of (a) MPSCS and (c) MPCP. TEM images of (b, c) MPSCS and (f, g) MPCP. HRTEM images with SAED insets of (d) MPSCS and (i) MPCP.

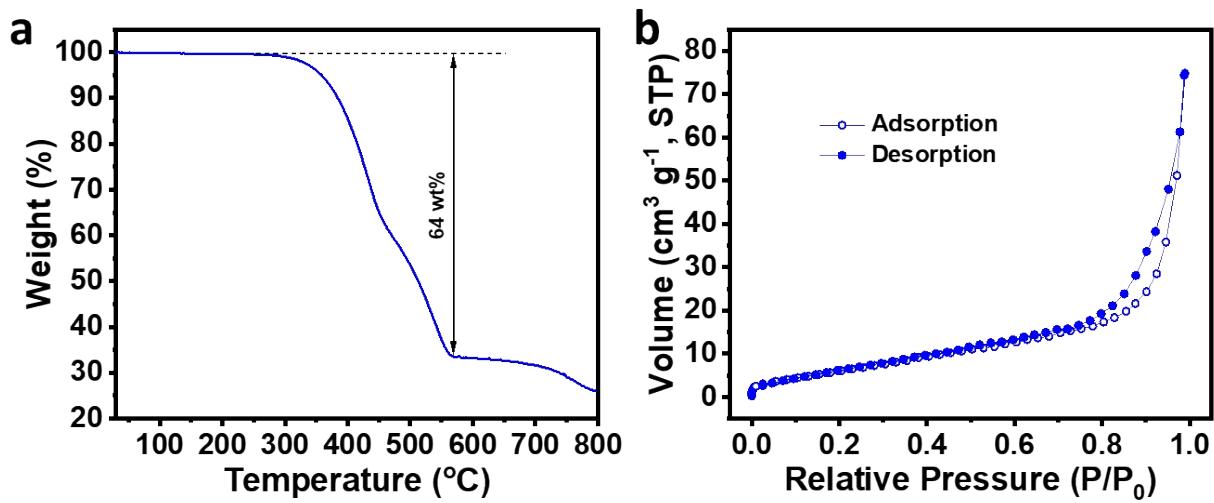


Figure S4. **(a)** TGA curve of $\text{Se}_{\text{HL}}@\text{HHCS}$ from room temperature to 800 °C, tested in Ar. **(b)** Nitrogen absorption-desorption isotherm of $\text{Se}_{\text{HL}}@\text{HHCS}$

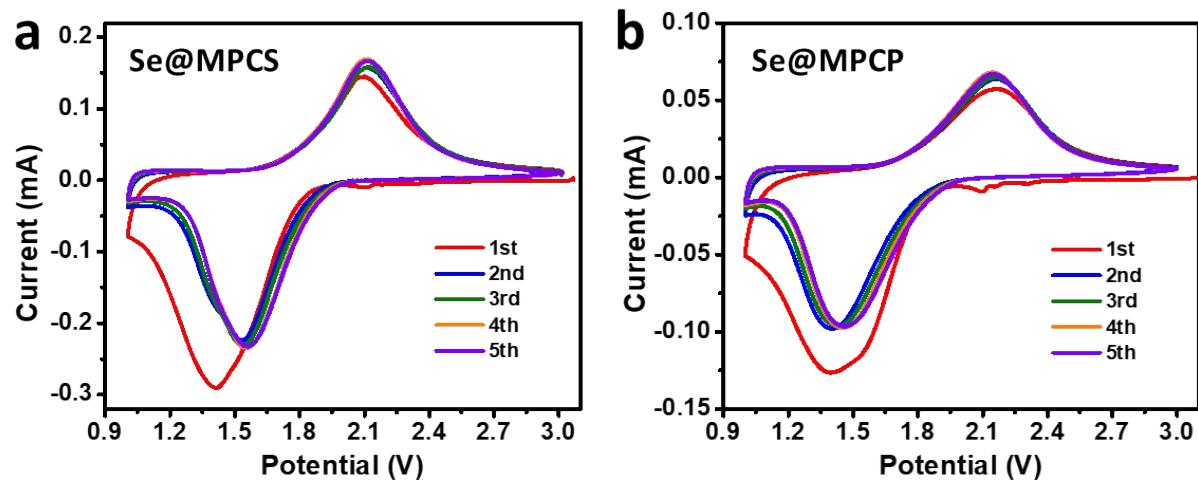


Figure S5. CV curves of **(a)** $\text{Se}@\text{MPCS}$ and **(b)** $\text{Se}@\text{MPCP}$ at a scan rate of 0.1 mV s⁻¹

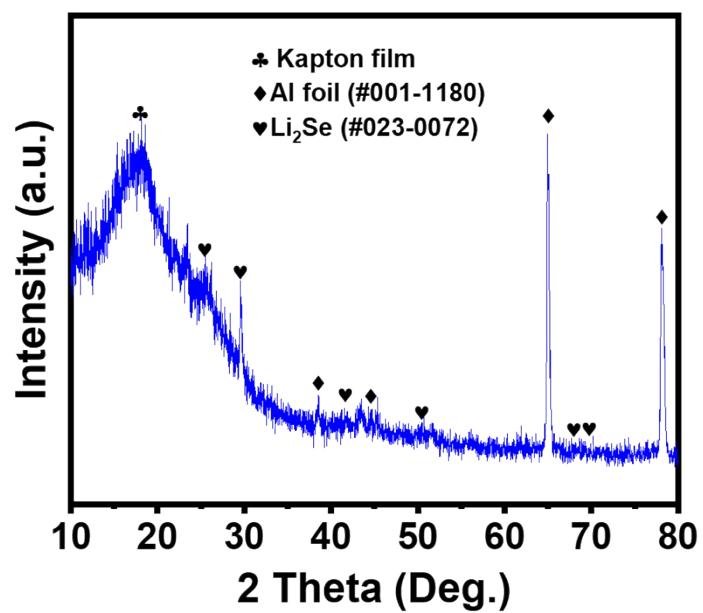


Figure S6. XRD profile of Se@HHCS discharged to 1 V at cycle 1.

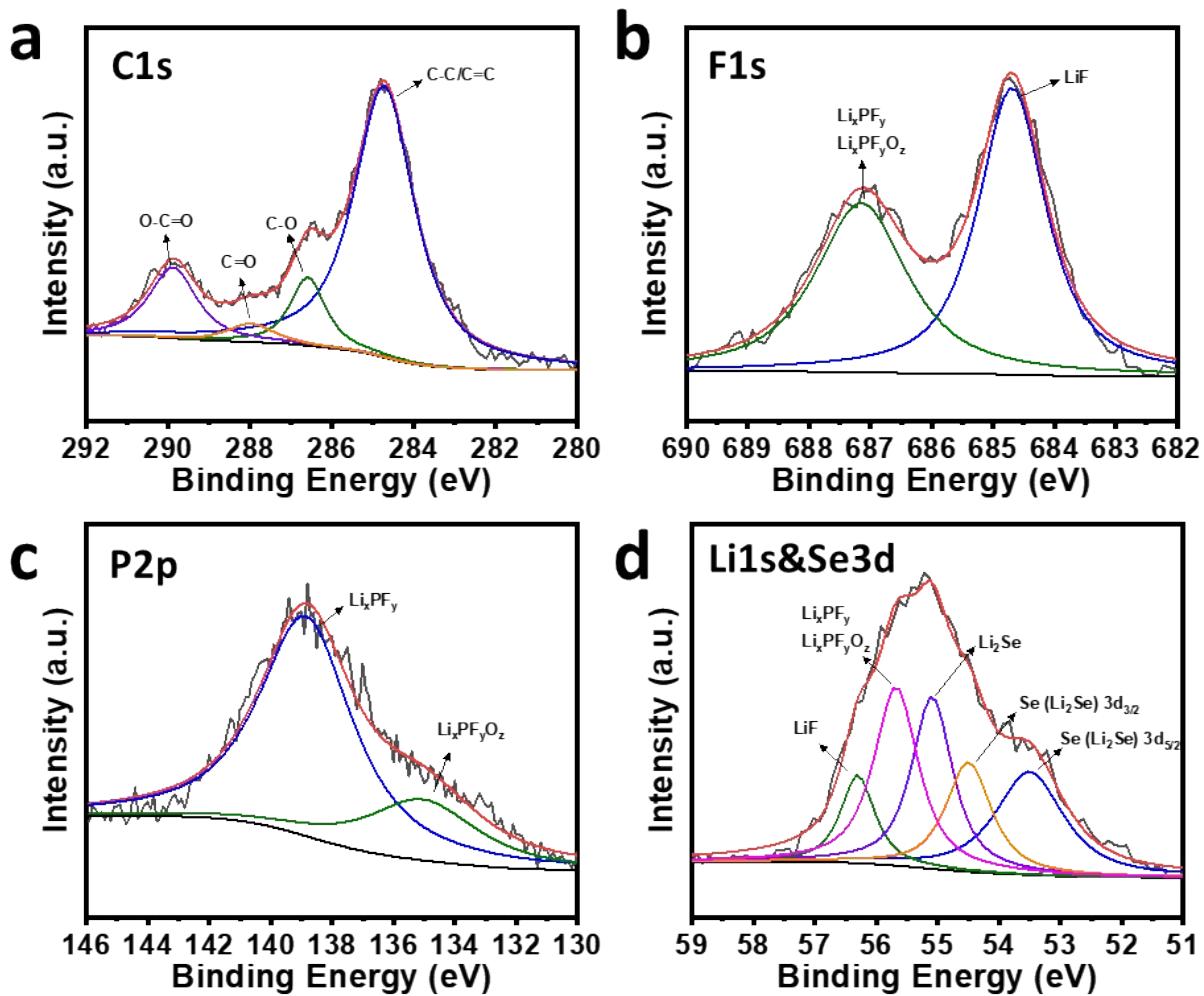


Figure S7. High-resolution XPS spectra of (a) C1s, (b) F1s, (c) P2p and (d) Li1s & Se3d of Se@HHCS discharged to 1 V at cycle 1.

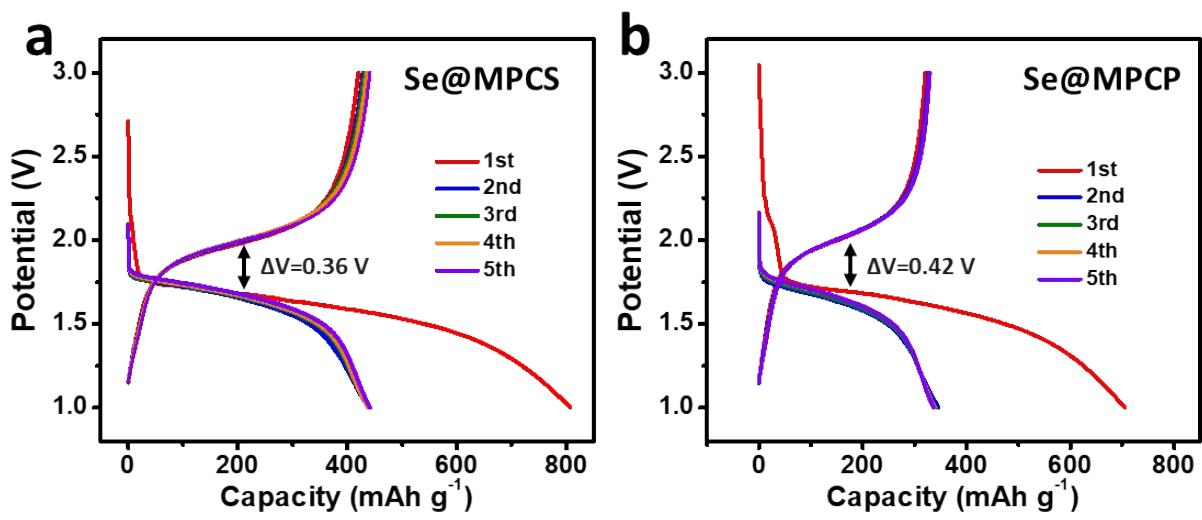


Figure S8. Galvanostatic profiles of (a) Se@MPCS and (b) Se@MPCP at 0.2C.

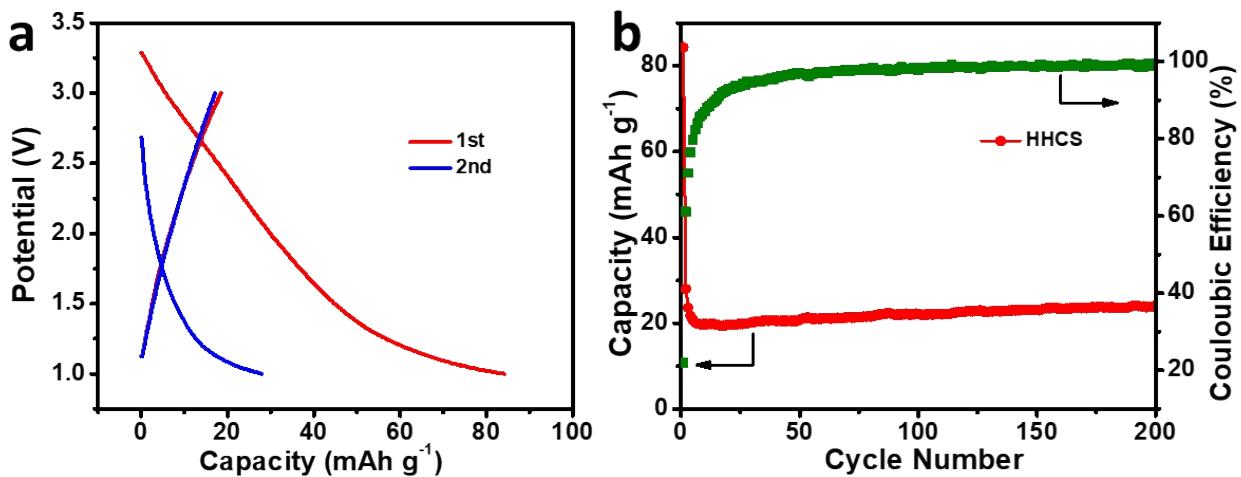


Figure S9. (a) Galvanostatic profiles and (b) cycling performance of HHCS at 0.2C.

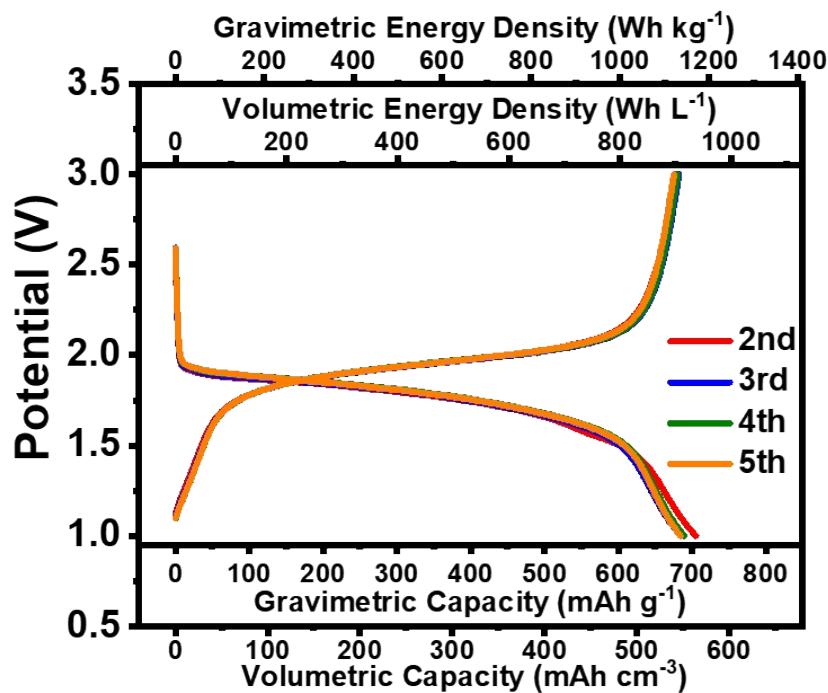


Figure S10. Galvanostatic profiles of $\text{Se}_{\text{HL}}@\text{HHCS}$ at 0.2C.

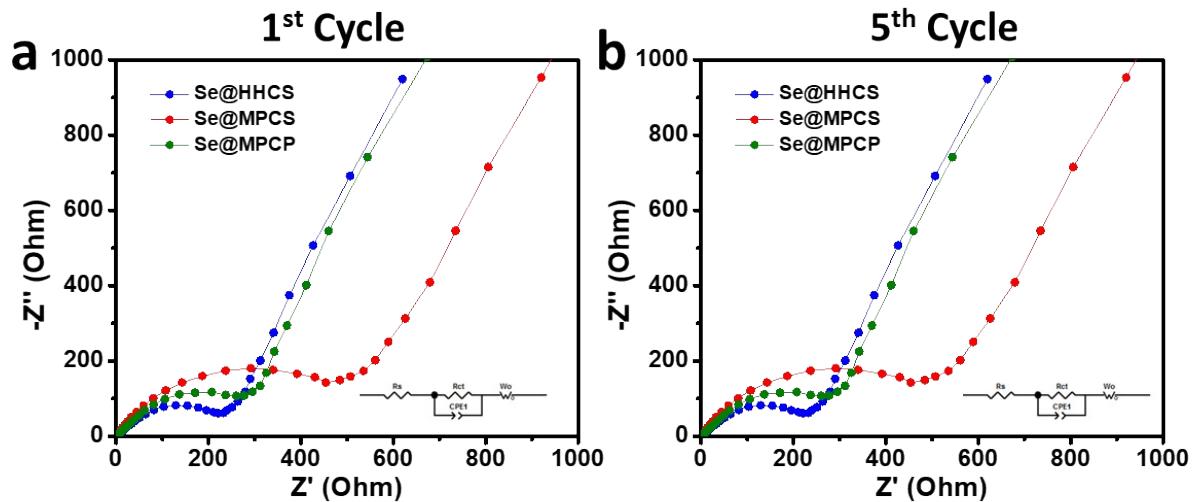


Figure S11. Nyquist plots of $\text{Se}@{\text{MPCS}}$ and $\text{Se}@{\text{MPCP}}$: (a) after 1st cycle and (b) after 5th cycle.

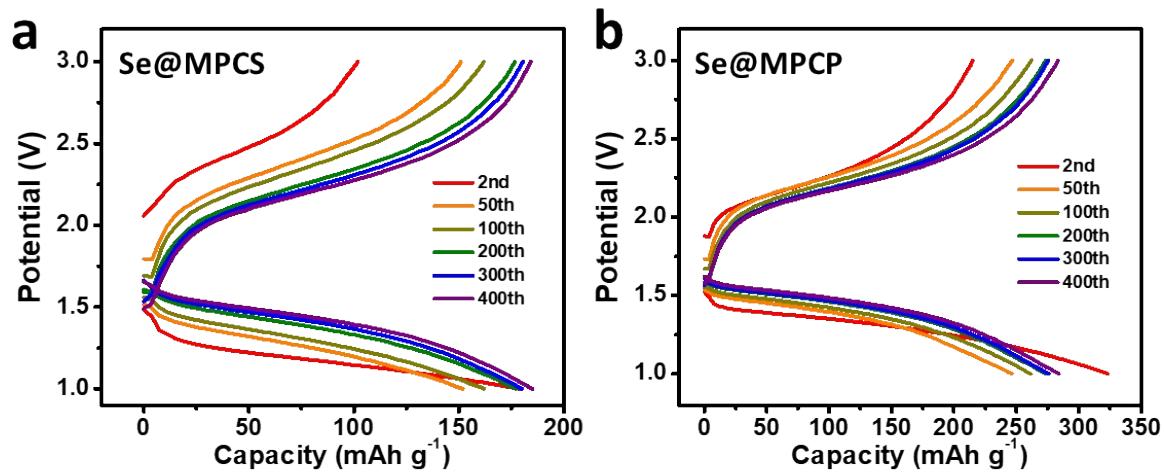


Figure S12. Galvanostatic profiles of (a) Se@MPCS and (b) Se@MPCP at 2C after 2nd, 50th, 100th, 200th, 300th and 400th cycles.

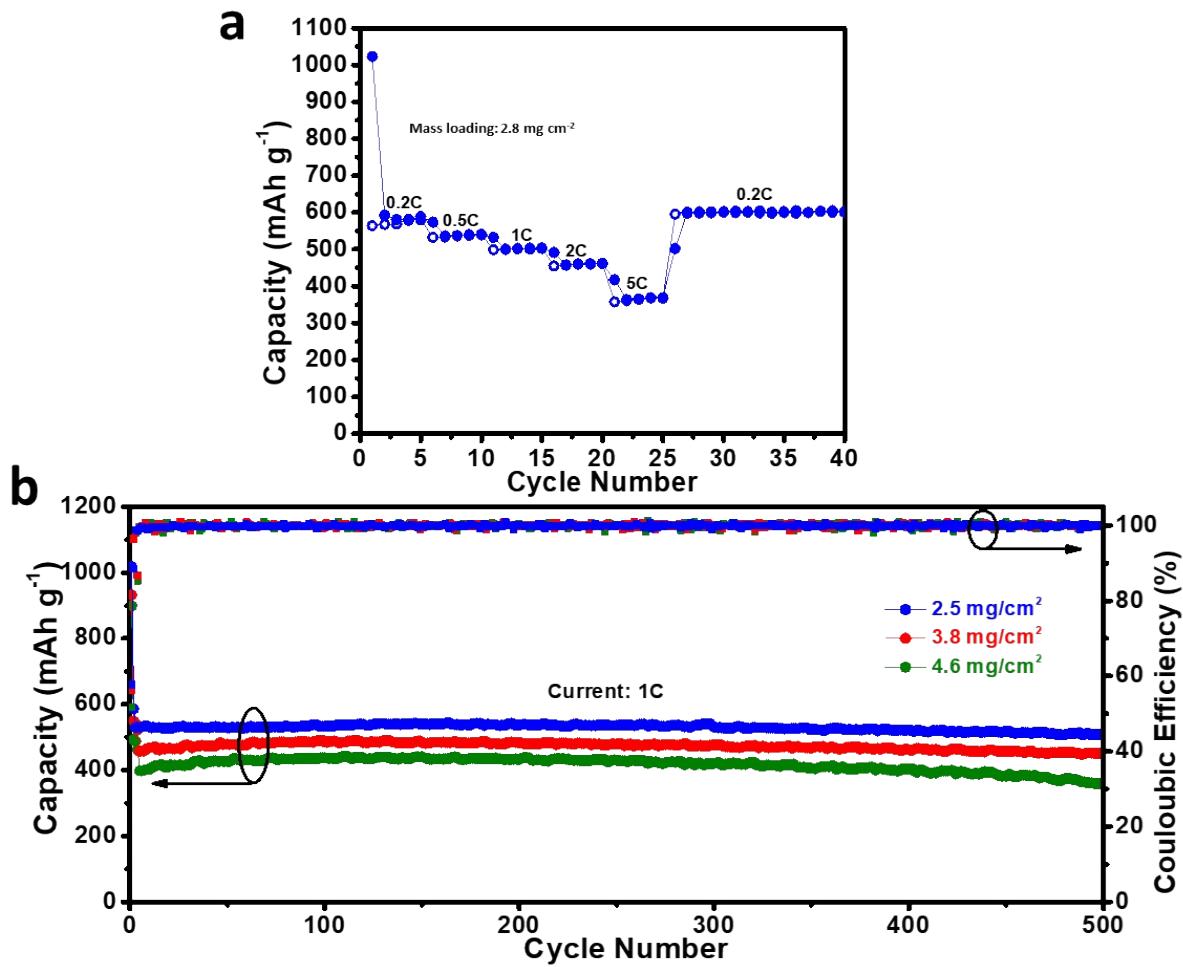


Figure S13. **(a)** Rate capability and **(b)** cycling performance of Se@HHCS at high electrode mass loadings.

Table S1. Comparison of cycling performance of Se@HHCS with Se-based cathodes from previously published reports.

Materials	Se loading (wt.%)	Current density (C)	Reversible capacity (mAh g ⁻¹) after (x) cycles	Reference
Se@HHCS		2	558 (500 th)	
	48	5	442 (1500 th)	
		10	357 (2000 th)	This work
	64	5	356 (600 th)	
		10	290 (800 th)	
Se/N-CSHPC	54	1	425 (200 th)	1
PCNS/Se	58	5	147 (750 th)	2
Se@HCS200	49	5	105 (100 th)	3
C/Se composite	55	2	363 (500 th)	4
Selenium				
nanowires/CNT composite	60	1	401 (500 th)	5
MPC/Se	50	0.5	354 (200 th)	6
Se@CoSe ₂ -PC	43	1	408 (100 th)	7
PANI@Se/C-G	52	5	403 (500 th)	8
NPC/CGB-Se	60	1	462 (1000 th)	9
Se/CMK-3	49	0.1	600 (50 th)	10
Se/CNSs	60	0.5	376 (1000 th)	11
Se@MICP	51	1	249 (3000 th)	12
Se-NCSs	56	1	301 (500 th)	13
Se/CMCs	50	2	166 (460 th)	14

HPTCs/Se	53	2	317 (900 th)	15
Se/(CNT@MPC)	50	1	352 (100 th)	16
MHPCS/Se	48	1	200 (100 th)	17
MiC/Se	44	0.5	400 (500 th)	18
Se/MCNF	50	1	403 (2000 th)	19
Se/MCN-RGO	62	1	400 (1300 th)	20
MWCNT/Se	60	4	231 (100 th)	21
Se-BPC	45	2	216 (80 th)	22
meso-C@Se composite	48	3	417 (100 th)	23
MCMs/Se	50	0.5	300 (100 th)	24
NCS/Se-50	50	5	287 (60 th)	25
HCPS/Se	41	0.5	299 (100 th)	26
PCM/Se	52	2	230 (510 th)	27
PCNFW/Se	33	1	324 (300 th)	28
Se-NCHPC	56	2	305 (60 th)	29
Graphene-Se@CNT	30	0.1	315 (100 th)	30
Se@3D MIL-68(Al)@MWCNTs	50	0.2	453 (200 th)	31
Se/SO-HPC	50	0.5	394 (400 th)	32
3DG-CNT@Se	51	0.2	504 (150 th)	33
Se@HPCNB	60	2	400 (1000 th)	34
Se-HPCF	50	5	202 (2000 th)	35
FNDPC@Se-1	40	~3	446 (500 th)	36

References

1. J. P. Song, L. Wu, W. D. Dong, C. F. Li, L. H. Chen, X. Dai, C. Li, H. Chen, W. Zou, W. B. Yu, Z. Y. Hu, J. Liu, H. E. Wang, Y. Li and B. L. Su, *Nanoscale*, 2019, **11**, 6970-6981.
2. W. Jin, H. Li, J. Zou, S. Inguva, Q. Zhang, S. Zeng, G. Xu and X. Zeng, *J. Alloys Compd.*, 2020, **820**, 153084.
3. R. Pongilat and K. Nallathamby, *J. Phys. Chem. C*, 2019, **123**, 5881-5889.
4. C. Zhao, Z. Hu and J. Luo, *Colloids Surf., A*, 2019, **560**, 69-77.
5. Y. Cui, X. Zhou, W. Guo, Y. Liu, T. Li, Y. Fu and L. Zhu, *Batter. Supercaps*, 2019, **2**, 784-791.
6. M. H. A. Shiraz, H. Zhu, Y. Liu, X. Sun and J. Liu, *J. Power Sources*, 2019, **438**, 227059.
7. J. Yang, H. Gao, D. Ma, J. Zou, Z. Lin, X. Kang and S. Chen, *Electrochim. Acta*, 2018, **264**, 341-349.
8. B. Wang, J. Zhang, Z. Xia, M. Fan, C. Lv, G. Tian and X. Li, *Nano Res.*, 2018, **11**, 2460-2469.
9. S. K. Park, J. S. Park and Y. C. Kang, *ACS Appl. Mater. Interfaces*, 2018, **10**, 16531-16540.
10. C. P. Yang, S. Xin, Y. X. Yin, H. Ye, J. Zhang and Y. G. Guo, *Angew. Chem. Int. Ed.*, 2013, **52**, 8363-8367.
11. S. F. Zhang, W. P. Wang, S. Xin, H. Ye, Y. X. Yin and Y. G. Guo, *ACS Appl. Mater. Interfaces*, 2017, **9**, 8759-8765.
12. Y. Liu, L. Si, X. Zhou, X. Liu, Y. Xu, J. Bao and Z. Dai, *J. Mater. Chem. A*, 2014, **2**, 17735-17739.
13. H. Lv, R. Chen, X. Wang, Y. Hu, Y. Wang, T. Chen, L. Ma, G. Zhu, J. Liang, Z. Tie, J. Liu and Z. Jin, *ACS Appl. Mater. Interfaces*, 2017, **9**, 25232-25238.
14. T. Liu, M. Jia, Y. Zhang, J. Han, Y. Li, S. Bao, D. Liu, J. Jiang and M. Xu, *J. Power Sources*, 2017, **341**, 53-59.
15. M. Jia, S. Lu, Y. Chen, T. Liu, J. Han, B. Shen, X. Wu, S.-J. Bao, J. Jiang and M. Xu, *J. Power Sources*, 2017, **367**, 17-23.
16. S. Xin, L. Yu, Y. You, H. P. Cong, Y. X. Yin, X. L. Du, Y. G. Guo, S. H. Yu, Y. Cui and J. B. Goodenough, *Nano Lett.*, 2016, **16**, 4560-4568.

17. T. Liu, C. Dai, M. Jia, D. Liu, S. Bao, J. Jiang, M. Xu and C. M. Li, *ACS Appl. Mater. Interfaces*, 2016, **8**, 16063-16070.
18. X. Wang, Y. Tan, Z. Liu, Y. Fan, M. Li, H. A. Younus, J. Duan, H. Deng and S. Zhang, *Small*, 2020, **16**, 2000266.
19. Y. Liu, L. Si, Y. Du, X. Zhou, Z. Dai and J. Bao, *J. Phys. Chem. C*, 2015, **119**, 27316-27321.
20. K. Han, Z. Liu, J. Shen, Y. Lin, F. Dai and H. Ye, *Adv. Funct. Mater.*, 2015, **25**, 455-463.
21. X. Wang, Z. Zhang, Y. Qu, G. Wang, Y. Lai and J. Li, *J. Power Sources*, 2015, **287**, 247-252.
22. Y. Qu, Z. Zhang, Y. Lai, Y. Liu and J. Li, *Solid State Ion.*, 2015, **274**, 71-76.
23. T. Liu, Y. Zhang, J. Hou, S. Lu, J. Jiang and M. Xu, *RSC Adv.*, 2015, **5**, 84038-84043.
24. L. Liu, Y. Wei, C. Zhang, C. Zhang, X. Li, J. Wang, L. Ling, W. Qiao and D. Long, *Electrochim. Acta*, 2015, **153**, 140-148.
25. Z. Li and L. Yin, *Nanoscale*, 2015, **7**, 9597-9606.
26. J. Li, X. Zhao, Z. Zhang and Y. Lai, *J. Alloys Compd.*, 2015, **619**, 794-799.
27. M. Jia, C. Mao, Y. Niu, J. Hou, S. Liu, S. Bao, J. Jiang, M. Xu and Z. Lu, *RSC Adv.*, 2015, **5**, 96146-96150.
28. J. Zhang, Z. Zhang, Q. Li, Y. Qu and S. Jiang, *J. Electrochem. Soc.*, 2014, **161**, A2093-A2098.
29. Y. Qu, Z. Zhang, S. Jiang, X. Wang, Y. Lai, Y. Liu and J. Li, *J. Mater. Chem. A*, 2014, **2**, 12255-12261.
30. K. Han, Z. Liu, H. Ye and F. Dai, *J. Power Sources*, 2014, **263**, 85-89.
31. C. Li, Y. Wang, H. Li, J. Liu, J. Song, L. Fusaro, Z.-Y. Hu, Y. Chen, Y. Li and B.-L. Su, *J. Energy Chem.*, 2021, **59**, 396-404.
32. X. Zhao, L. Yin, Z. Yang, G. Chen, H. Yue, D. Zhang, Z. Sun and F. Li, *J. Mater. Chem. A*, 2019, **7**, 21774-21782.
33. J. He, Y. Chen, W. Lv, K. Wen, P. Li, Z. Wang, W. Zhang, W. Qin and W. He, *ACS Energy Lett.*, 2016, **1**, 16-20.
34. W. D. Dong, W. B. Yu, F. J. Xia, L. D. Chen, Y. J. Zhang, H. G. Tan, L. Wu, Z. Y. Hu, H. S. H. Mohamed, J. Liu, Z. Deng, Y. Li, L. H. Chen and B. L. Su, *J. Colloid Interface Sci.*, 2021, **582**, 60-69.

35. X. Chen, L. Xu, L. Zeng, Y. Wang, S. Zeng, H. Li, X. Li, Q. Qian, M. Wei and Q. Chen, *Dalton Trans*, 2020, **49**, 14536-14542.
36. R. Qiu, R. Fei, T. Zhang, X. Liu, J. Jin, H. Fan, R. Wang, B. He, Y. Gong and H. Wang, *Electrochim. Acta*, 2020, **356**, 136832.