Balancing interlayer spacing, pore structures and conductivity endows hard carbon with high capacity for rechargeable aluminum batteries

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Figure S1. Contact angle test of P-5 and P-M.



Figure S2. Galvanostatic charge/discharge curves of (a) P-M, (b) P-H, and (c) P-5 at 500 mA \cdot g⁻¹ after various cycles. Galvanostatic charge/discharge curves of (d) P-M, (e) P-H, and (f) P-5 at different current densities.



Figure S3. Coulombic Efficiency of P-5 and P-M at 500 mA $\cdot g^{\text{-1}}$.



Figure S4. Cycling stability of P-H and P-5 at $1 \text{ A} \cdot \text{g}^{-1}$ after 1000 cycles.



Figure S5. (a) CV curves of P-H and P-M at 1.0 mV/s. (b) b value of P-H and P-M



Figure S6. the Nyquist plot of P-5, P-M, and P-H 9 (a) before and (b) after cycling at 500 mA \cdot g⁻¹.

Site -	Electrical conductivity (s/cm)			
	P-5	P-H	P-M	
Site 1	0.724	1.288	1.292	
Site 2	0.721	1.289	1.294	
Site 3	0.721	1.290	1.293	
Average value	0.722	1.289	1.292	

Table S1. Electrical conductivity tests of P-5, P-H, and P-M.

Matariala	Elemental analysis (<i>at</i> .%)			
wrateriais	С	0	Ν	
P-5	89.6	10.4	-	
P-H	94.6	3.36	2.04	
P-M	90.23	4.26	5.51	

Table S2. Elemental analysis of P-5, P-H, and P-M.

Materials	Specific capacity/mAh·g ⁻¹ (I/mA·g ⁻¹)	Cycling performance		
		Current density (mA·g ⁻¹)	Cycle number	Capacity (mA h·g ⁻¹)
P-M	323(500)	1000	1000	109
FNG[1]	300(60)	60	40	225
Natural graphite[2]	110(99)	660	6000	60
EG[3] (expanded graphite)	110(1000)	5000	27500	100
SG[3] (synthetic graphite)	152.5(500)	5000	500	75.5
Nanosheet-bricked PG[4] (porous graphite)	104(1000)	100000	3000	90
CG[5] (commercial graphite)	101(1000)	5000	30000	60
3D GF[6] (graphitic foam)	90(1000)	1200	4000	60
3D GMN[7] (graphene network)	56(3000)	2400	200	57
Defect-free GA[8]	97(50000)	5000	25000	100 ± 3
FLG[9] (few-layer graphene)	173(1000)	10000	5000	78
TLG[10] (three-layer graphene)	197(200)	5000	1000	147
GF@CFC[11] (carbon fiber cloth)	140(100)	3000	300	60
CMK-3[12] (commercial ordered mesoporous carbon)	32(270)	980	36000	28
CMK-8[13]	100.5(300)	2000	30000	46.4
Coconut PAC[14] (porous activated carbon)	150(100)	1000	1500	80
CNF[15] (commercial carbon nanofiber)	95(50000)	10000	20000	105
MoS ₂ / MNC[16] (N-doped carbon)	191.2(500)	1000	1700	127.5
NC@ZnSe[17]	172(300)	500	250	60

Table S3. Electrical performances of various carbon materials

Reference

- J.V. Rani, V. Kanakaiah, T. Dadmal, et al., Fluorinated Natural Graphite Cathode for Rechargeable Ionic Liquid Based Aluminum–Ion Battery, Journal of The Electrochemical Society. 2013, 160: A1781–A1784.
- [2] M.C. Lin, M. Gong, B. Lu, et al., An ultrafast rechargeable aluminium-ion battery, Nature. 2015, 520: 325–328.
- [3] D. Muñoz-Torrero, A. Molina, J. Palma, et al., Widely commercial carbonaceous materials as cathode for Al-ion batteries, Carbon. 2020, 167: 475–484.
- [4] C. Zhang, R. He, J. Zhang, et al., Amorphous Carbon-Derived Nanosheet-Bricked Porous Graphite as High-Performance Cathode for Aluminum-Ion Batteries, ACS Applied Materials and Interfaces. 2018, 10: 26510–26516.
- [5] X. Dong, H. Xu, H. Chen, et al., Commercial expanded graphite as high-performance cathode for low-cost aluminum-ion battery, Carbon. 2019, 148: 134–140.
- [6] Y. Wu, M. Gong, M.C. Lin, et al., 3D Graphitic Foams Derived from Chloroaluminate Anion Intercalation for Ultrafast Aluminum-Ion Battery, Advanced Materials. 2016, 28: 9218–9222.
- [7] G.Y. Yang, L. Chen, P. Jiang, et al., Fabrication of tunable 3D graphene mesh network with enhanced electrical and thermal properties for high-rate aluminum-ion battery application, RSC Advances. 2016, 6: 47655–47660.
- [8] H. Chen, F. Guo, Y. Liu, et al., A Defect-Free Principle for Advanced Graphene Cathode of Aluminum-Ion Battery, Advanced Materials. 2017, 29:.
- [9] H. Huang, F. Zhou, P. Lu, et al., Design and construction of few-layer graphene cathode for ultrafast and high-capacity aluminum-ion batteries, Energy Storage Materials. 2020, 27: 396– 404.
- [10] Y. Kong, C. Tang, X. Huang, et al., Thermal Reductive Perforation of Graphene Cathode for High-Performance Aluminum-Ion Batteries, Advanced Functional Materials. 2021, 31: 1–9.
- [11] C. Liu, Z. Liu, Q. Li, et al., Binder-free ultrasonicated graphite flakes@carbon fiber cloth cathode for rechargeable aluminum-ion battery, Journal of Power Sources. 2019, 438: 226950.
- [12] Z.A. Zafar, S. Imtiaz, R. Li, et al., A super-long life rechargeable aluminum battery, Solid State Ionics. 2018, 320: 70–75.
- [13] C. Li, P.C. Rath, S.X. Lu, et al., Ordered nano-structured mesoporous CMK-8 and other carbonaceous positive electrodes for rechargeable aluminum batteries, Chemical Engineering Journal. 2021, 417: 129131.
- P. Thanwisai, N. Chaiyapo, P. Phuenhinlad, et al., Mesoporous and defective activated carbon cathode for AlCl₄⁻ anion storage in non-aqueous aluminium-ion batteries, Carbon. 2022, 191: 195–204.
- [15] Y. Hu, S. Debnath, H. Hu, et al., Unlocking the potential of commercial carbon nanofibers as free-standing positive electrodes for flexible aluminum ion batteries, Journal of Materials

Chemistry A. 2019, 7: 15123–15130.

- [16] S. Guo, H. Yang, M. Liu, et al., Interlayer-Expanded MoS₂/N-Doped Carbon with Three-Dimensional Hierarchical Architecture as a Cathode Material for High-Performance Aluminum-Ion Batteries, ACS Applied Energy Materials. 2021, 4: 7064–7072.
- [17] J. Li, W. Liu, Z. Yu, et al., N-doped C@ZnSe as a low cost positive electrode for aluminum-ion batteries: Better electrochemical performance with high voltage platform of ~1.8 V and new reaction mechanism, Electrochimica Acta. 2021, 370: 137790.