Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Ultra-fast and ultra-long-life Li ion batteries with 3D surface-porous graphene anodes synthesized from CO_2

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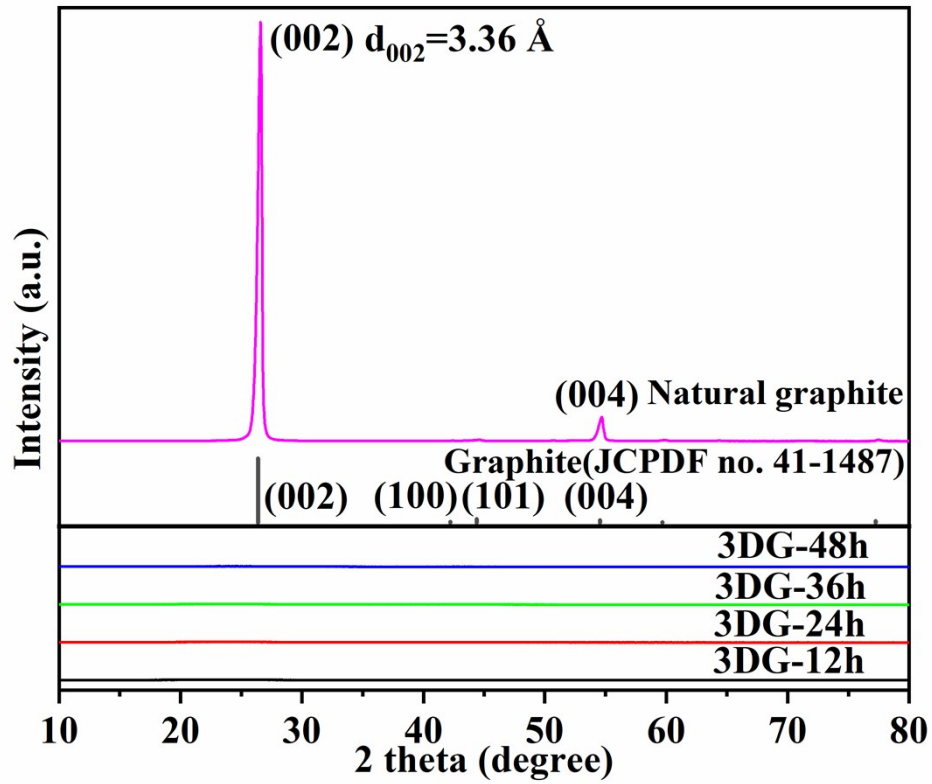

Figure S1. XRD patterns of as-synthesized 3DGs and commercial graphite.

Table SI. The structural parameters of 3DGs from XRD.

Samples	d_{002}	L_c	Sheet layers
3DG-12h	3.87	1.96	5.0
3DG-24h	3.85	1.96	5.0
3DG-36h	3.81	1.91	5.0
3DG-48h	3.79	1.91	5.0

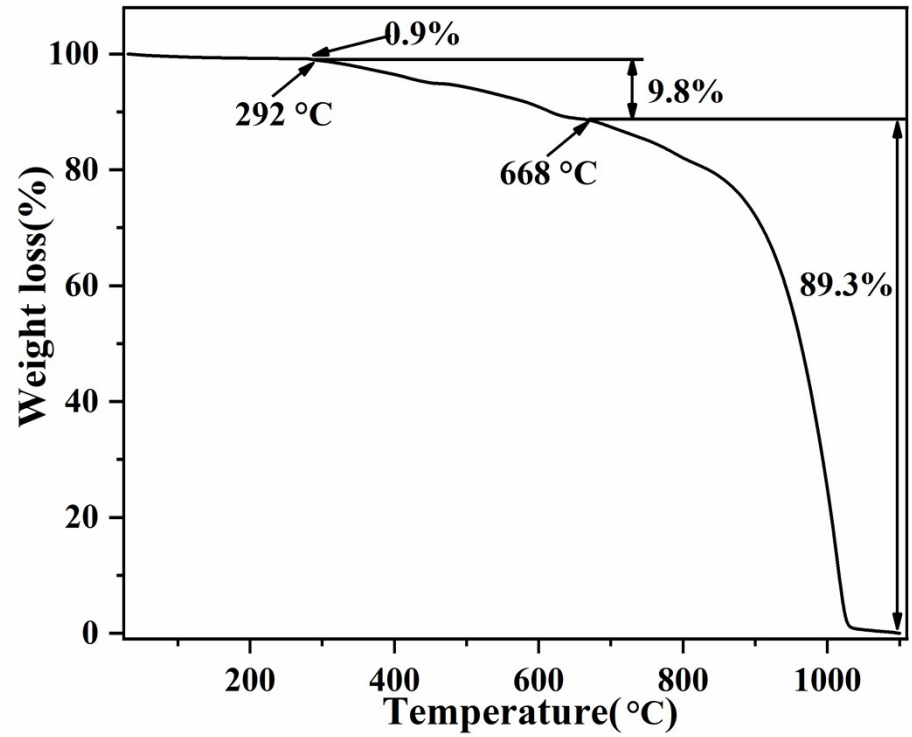

Figure S2. TG curve of 3DG-24h in carbon dioxide.

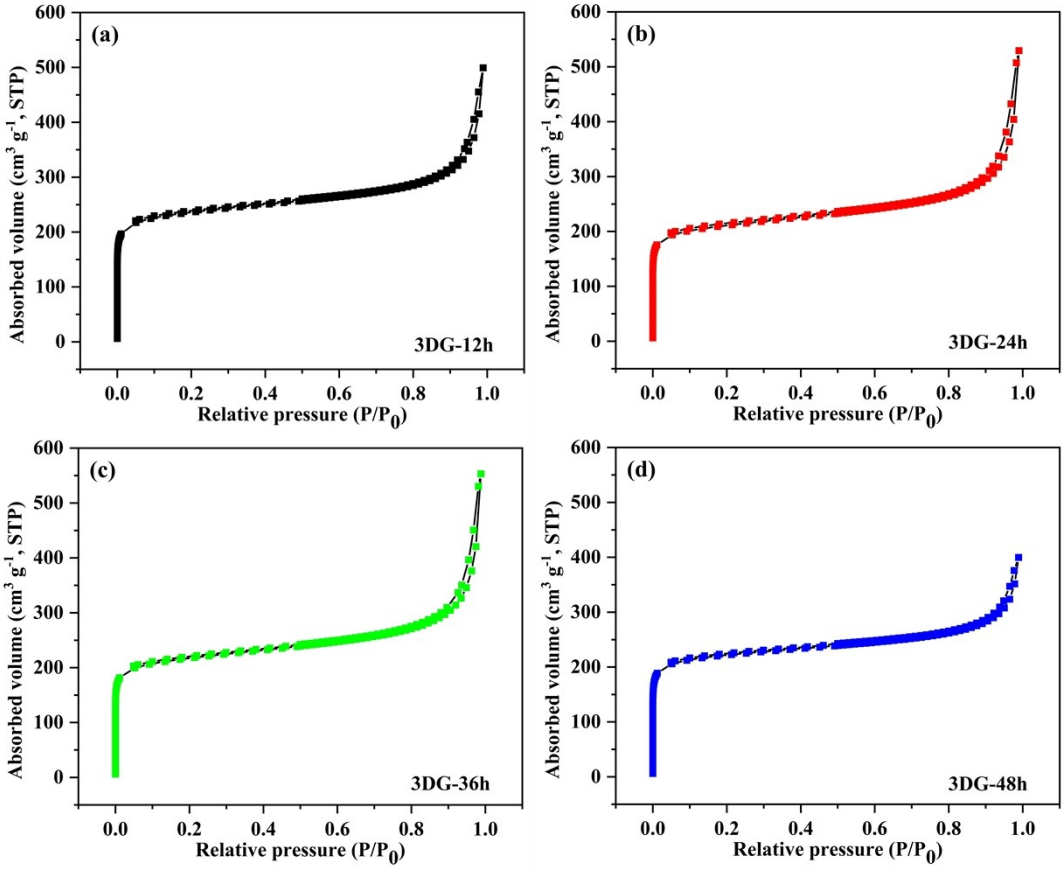

Figure S3. N_2 adsorption/desorption curves at 77 K of (a) 3DG-12h, (b) 3DG-24h, (c) 3DG-36h, and (d) 3DG-48h samples.

Table SII. The composition of as-synthesized 3DGs.

Samples	Raman	C1s (XPS)			Elemental (EDS)	
	I_D/I_G	sp^2-C (%)	sp^3-C (%)	C-O (%)	C (at.%)	O (at.%)
3DG-12h	1.02	73.1	14.4	12.4	93.57	6.43
3DG-24h	1.01	74.0	14.3	11.7	93.94	6.06
3DG-36h	1.00	74.5	14.5	11.0	94.62	5.38
3DG-48h	0.98	74.8	14.8	10.4	95.28	4.72

Table SIII. The initial capacity and coulombic efficiency of 3DGs at 0.2C.

Samples	charge capacity (mAh g^{-1})	discharge capacity (mAh g^{-1})	Coulombic efficiency (%)
3DG-12h	910	1842	49.4
3DG-24h	972	1965	49.5
3DG-36h	965	1961	49.2
3DG-48h	929	1896	49.0

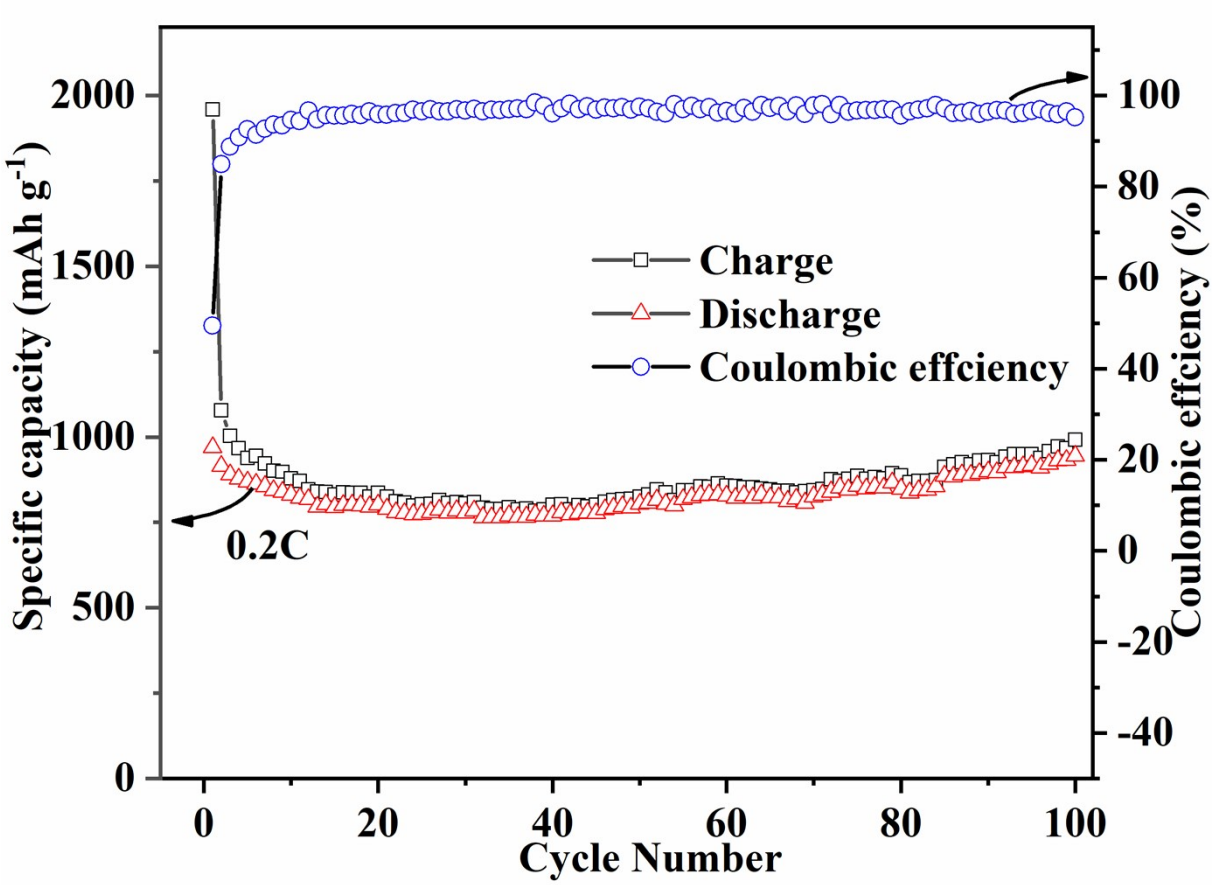

Figure S4. Cyclic performance and coulombic efficiency of 3DG-24h electrode at 0.2 C.

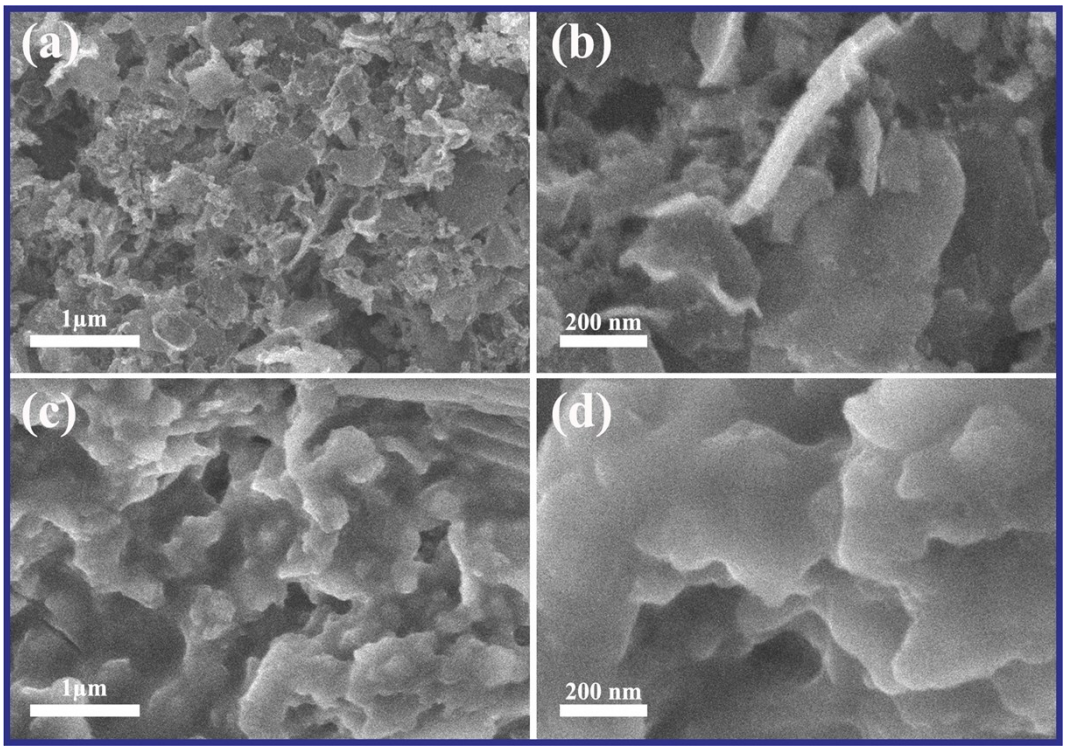

Figure S5. SEM images of 3DG-24h electrode: (a, b) for the pristine electrode and (c, d) for the electrode after 10000 cycles.

Table SIV. Comparison of electrochemical performance of 3DGs with previously reported carbon-based anode materials in LIBs.

Electrodes	Current density	Electrochemical properties	Cycle life (capacity retention)	References
3DG	1C = 372 mA g^{-1}	972-277 mAh g^{-1} at 0.2-150C	10000 cycles at 50 C (91.9%)	This work
Doped graphene Sheets	1C = 500 mA g^{-1}	1040-235 mAh g^{-1} at 0.1-50C	30 cycles at 0.1 C (83.6%)	S1
3D nitrogen-doped carbon frameworks	1C = 500 mA g^{-1}	675-279 mAh g^{-1} at 0.2-20C	10000 cycles at 20 C (54%)	S2
Carbon clusters @ hard carbon nanofibers	1C = 372 mA g^{-1}	549-129 mAh g^{-1} at 0.3-13.4C	1000 cycles at 2.7 C (99.7%)	S3
Carbon nanotube clusters@3DG	1C = 500 mA g^{-1}	1132-383 mAh g^{-1} at 0.2-10C	1000 cycles at 4 C (75%)	S4
Pomegranate-shaped Sn/SnO_x/nanocarbon composites	1C = 500 mA g^{-1}	526-289 mAh g^{-1} at 0.5-6C	5000 cycles at 15 C (89%)	S5
TiO_2/Micron carbon fibers	1C = 200 mA g^{-1}	690.4-100 mAh g^{-1} at 0.5-16C	5000 cycles at 10 C (92%)	S6
Co@ porous carbon nanosheets	1C = 500 mA g^{-1}	900-510mAh g^{-1} at 0.1-16C	1000 cycles at 16 C (90%)	S7
Graphene–silica (SiO_x)	1C = 700 mA g^{-1}	716.2-382.6 mAh g^{-1} at 0.1-5C	400 cycles at 5 C (88%)	S8
Nitrogen-containing carbon film	1C = 500 mA g^{-1}	908.4-325.9 mAh g^{-1} at 1-40C	800 cycles at 10 C (91.6%)	S9

Table SV. The R_s and R_{ct} values, obtained from fitting the equivalent circuit to the experimental data.

Samples	R_s/Ω	R_{ct}/Ω
3DG-12h	3.1	149.1
3DG-24h	2.6	170.2
3DG-36h	2.9	203.0
3DG-48h	2.5	235.5

Table SVI. Comparison of capacitive contributions of 3DGs with those of reported carbon-based anode materials in LIBs.

Electrodes	Capacitive contributions	Scan rate of CV	References
3DG	56.4%	1 mV s^{-1}	This work
High-defect mesopore-dominant porous carbon (HDMPC)	46.7%	10 mV s^{-1}	S10
Porous carbon (PC)	55.2%	0.75 mV s^{-1}	S11
Three-dimensional hollow carbon spheres/reduced graphene oxide nanocomposites (DHCSs/RGO)	52.6%	1 mV s^{-1}	S12
Boron-doped 3D hierarchical porous carbon network(B-CN)	36.0%	1 mV s−1	S13
Nitrogen-sulfur co-doped porous carbon (NSPC)	86.3%	0.75 mV s^{-1}	S11
Nitrogen-rich hierarchically porous carbon (NHPC)	64.0%	1 mV s^{-1}	S14
Nitrogen and phosphorus co-doped carbon nanosheets (NP-CNSs)	65.5%	0.8 mV s^{-1}	S15

References

S1. Z. S. Wu, W. C. Ren, L. Xu, F. Li and H. M. Cheng, *ACS Nano*, 2011, **5**, 5463-5471.

S2. J. Wang, H. Fan, Y. Shen, C. Li and G. Wang, *Chem. Eng. J.*, 2019, **357**, 376-383.

S3. C. Liu, N. Xiao, Y. Wang, H. Li, G. Wang, Q. Dong, J. Bai, J. Xiao and J. Qiu, *Fuel Process. Technol.*, 2018, **180**, 173-179.

S4. S. Z. Huang, J. Y. Wang, Z. Y. Pan, J. L. Zhu and P. K. Shen, *J. Mater. Chem. A*, 2017, **5**, 7595-7602

S5. M. D. Cao, M. Zhang, L. L. Xing, Q. Wang and X. Y. Xue, *J. Alloy Compd.* 2017, **694**, 30-39.

S6. D. D. Wang, G. H. Zhang, Z. Q. Shan, T. Y. Zhang and J. H. Tian, *Chem. Electro. Chem.*, 2018, **5**, 540-545.

S7. V. Etacheri, C. N. Hong, J. L. Tang and V. G. Pol, *ACS Appl. Mater. Inter.*, 2018, **10**, 4652-4661.

S8. I. H. Son, J. H. Park, S. Park, K. Park, S. Han, J. Shin, S. G. Doo, Y. Hwang, H. Chang and J. W. Choi, *Nat. Commun.*, 2017, **8**, 1-11.

S9. T. Yuan, Y. S. He, W. M. Zhang and Z. F. Ma, *Chem. Commun.*, 2016, 52, 112-115.

S10 J. Niu, R. Shao, M.Y. Liu, J.J. Liang, Z.P. Zhang, M.L. Dou, Y.Q. Huang and F. Wang, *Energy Storage Materials*, 2018, **12**, 145-152.

S11 M.Q. Du, Y.S. Meng, C. Wang, C.Y. Duan, F.L. Zhu and Y. Zhang, *Journal of Electroanalytical Chemistry*, 2019, **834**, 17-25.

S12 Y.Q. Zhang, Q, Ma, S.L. Wang, X. Liu and L. Li, *ACS Nano*, 2018, **12**, 4824-4834.

S13 D. Wang, Z.Y. Wang, Y. Li, K.Z. Dong, J.H. Shao, S.H. Luo, Y.G. Liu and X.W. Qi, *Applied Surface Science*, 2019, **464**, 422-428.

S14 C.M. Zheng, X.D. Hu, X.H. Sun, S. J. Yoo and X. Li, *Electrochimica Acta*, 2019, **306**, 339-349

S15 C. Gao, J.Z. Feng, J.G. Dai, Y.Y. Pan, Y.L. Zhu, W.H. Wang, Y.F. Dong, L.F. Cao, L. Guan, L. Pan, H. Hu and M.B. Wu, *Carbon*, 2019, **153**, 372-380.