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

Supporting Information

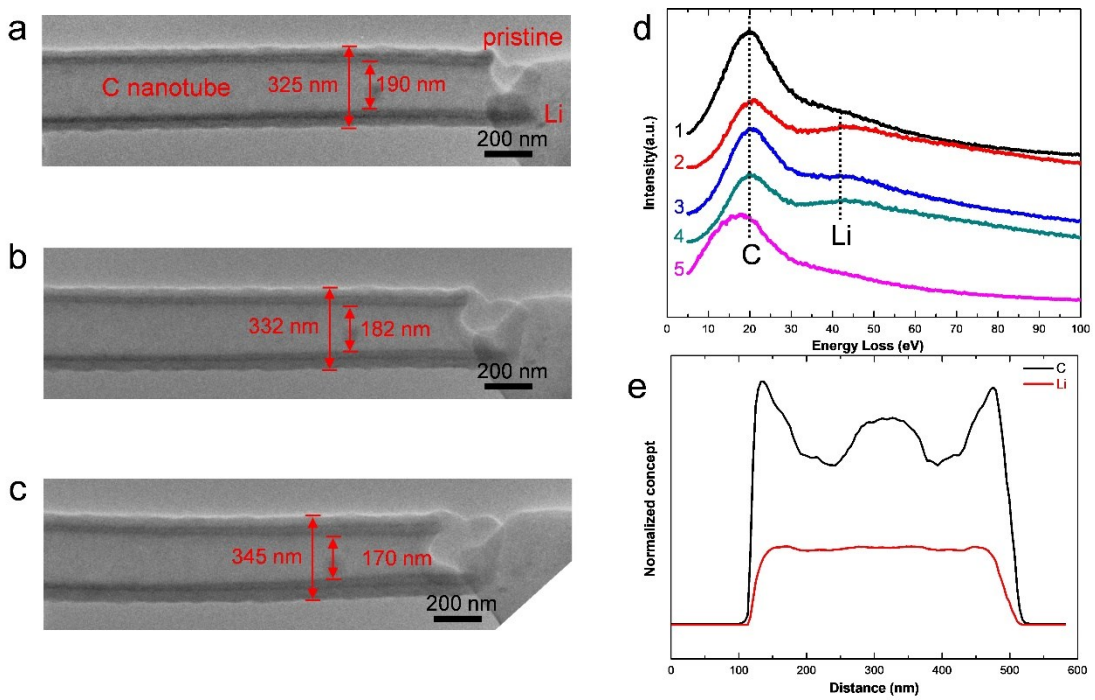
Concave Directing the Deposition of Lithium Metal on Inner Surface of Graphitic Carbon Tubes to Enable Lithim-Metal batteries

Weilin Chen[a‡], Youran Hong[b‡], Zehua Zhao[a], Yuting Zhang[a], Linhai Pan[a], Jia Wan[a], Jiangwei Wang[b]*, Haiyong He[a]*

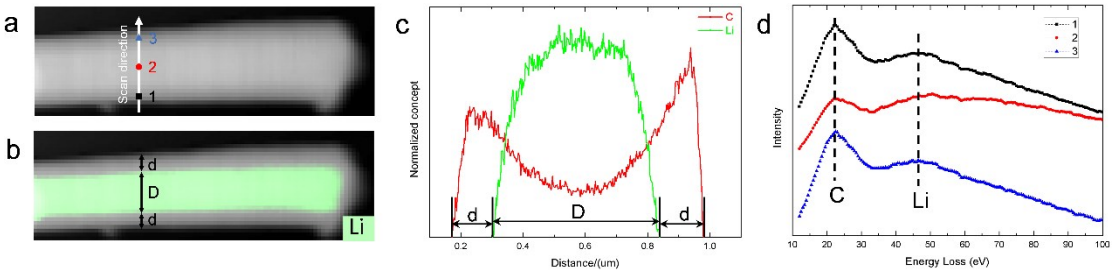
a: Ningbo Institute of Materials Technology and Engineering Chinese Academy of Sciences Ningbo 315201, China;

b: Center of Electron Microscopy and State Key Laboratory of Silicon Materials, School of Materials Science and Engineering, Zhejiang University, Hangzhou 310027, China.

‡Authors with equal contribution.



Supplementary figure 1. (a-c) A typical lithiation process of GCT. (d) EELS spectra of carbon edge and Li edge at five different positions figure1b. (e) The EELS line scan along the red line in figure 1b.

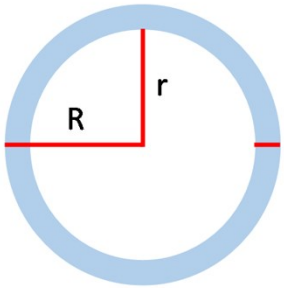


Supplementary figure 2. (a) ADF image of the lithiated nanotube. (b) Li EELS mapping. (c) The EELS line scan along the white line in S3 (a). (d) EELS spectra of C edge and Li edge at three different positions.

V= the volume of the whole tube, V_{inter}= inter volume of tube, ρ_{Li} (the density of Li) , ρ_c (the density of C), C_{Li} (theoretical specific capacity of Li)

$$\text{Specific capacity} = \frac{capacity}{mass}$$

$$= \frac{V_{inter} \cdot \rho_{Li} \cdot CL_i}{(V - Vinte_r) \cdot \rho_c}$$



R_0=22nm

$$= \frac{\pi r^2 \cdot h \cdot \rho_{Li} \cdot C_{Li}}{[\pi(R^2 - r^2)] \cdot h \cdot \rho_c} \qquad r=R-2R_0$$

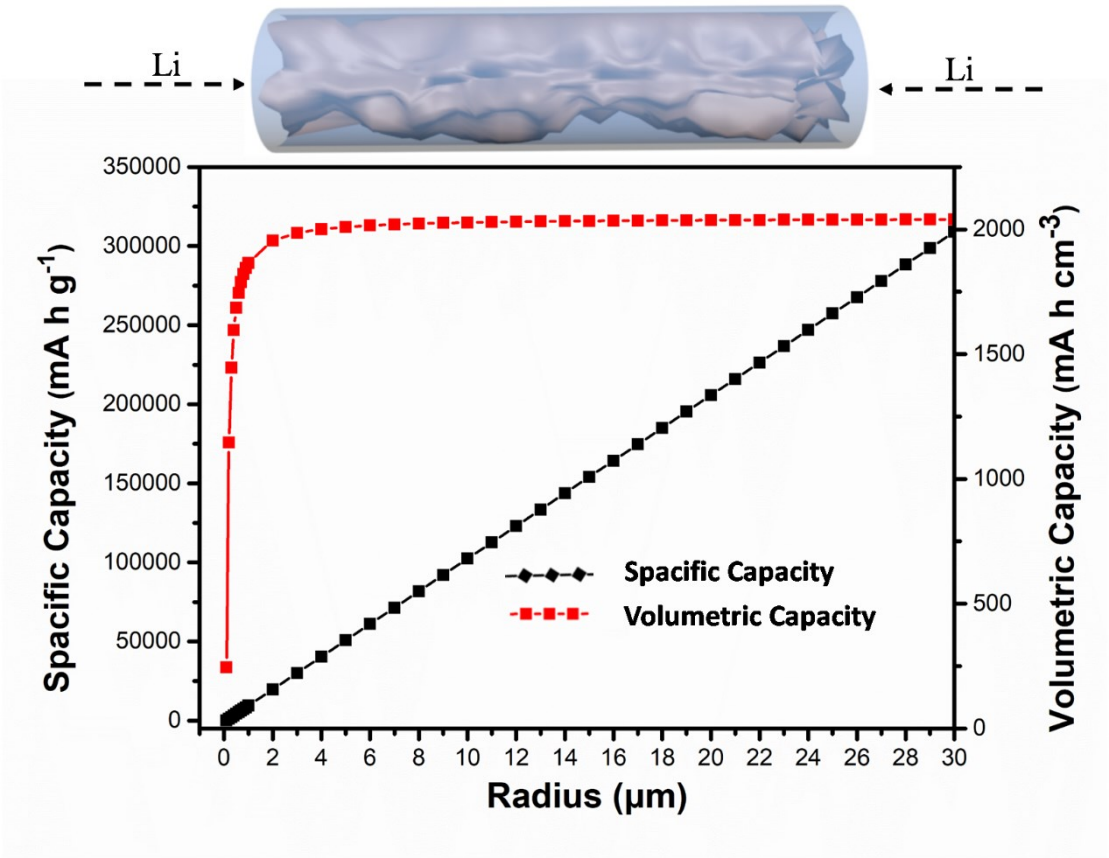
$$= \frac{\pi(R^2 - 4RR_0 + 4R_0^{\,2}) \cdot \rho_{Li} \cdot C_{Li}}{4\pi\rho_c(RR_0 - R_0^{\,2})}$$

$\rho_{Li} = 0.53$ g/cm^3 $\rho_c = 2.25$ g/cm^3 $C_{Li} = 3860$ mA h g^{-1} R_0=22 nm

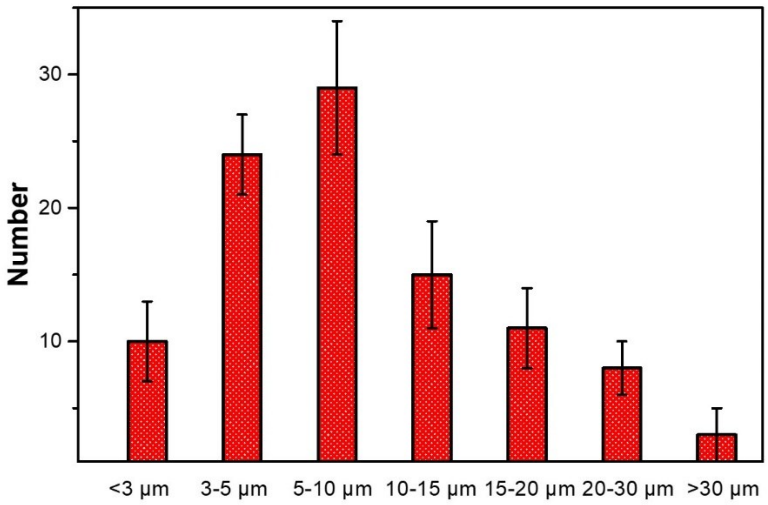
$$\text{Volumetric capacity} = \frac{\pi r^2 \cdot \rho_{Li} \cdot C_{Li}}{\pi R^2} \qquad r=R-2R_0$$

$$= \frac{\pi(R^2 - 4RR_0 + 4R_0^{\,2}) \cdot \rho_{Li} \cdot C_{Li}}{\pi R_0^{\,2}}$$

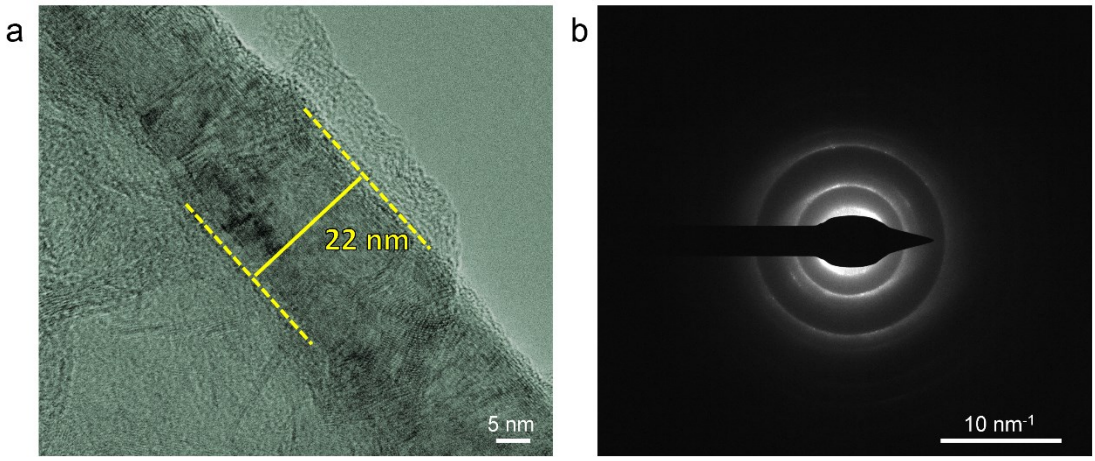
$\rho_{Li} = 0.53$ g/cm^3 $C_{Li} = 3860$ mA h g^{-1} R_0=22 nm



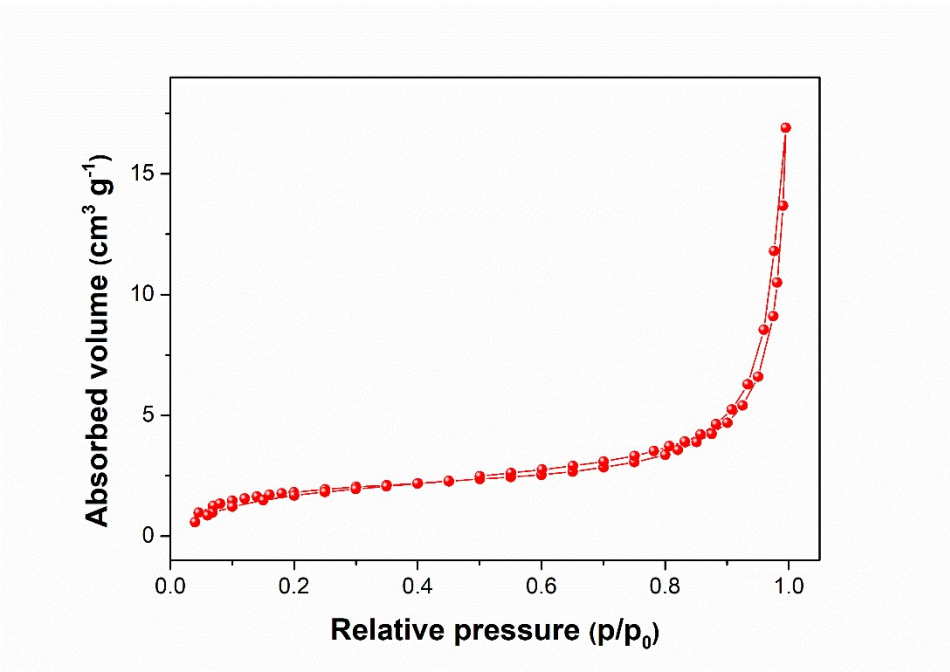
Supplementary figure 3. Ideal model for lithium deposition in tubes.



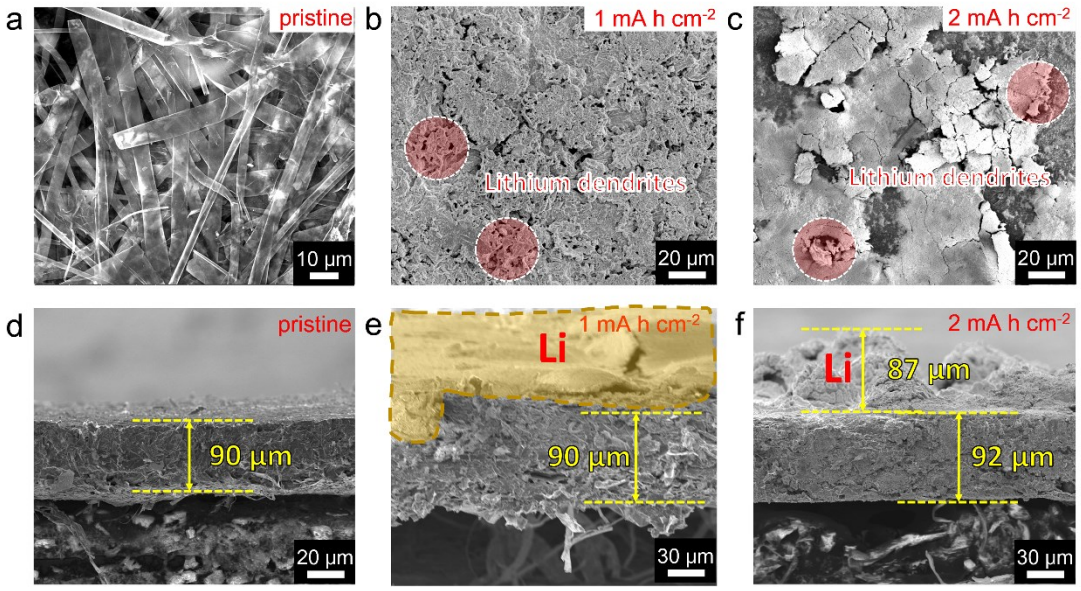
Supplementary figure 4. Diameter distributed of S-GCT.



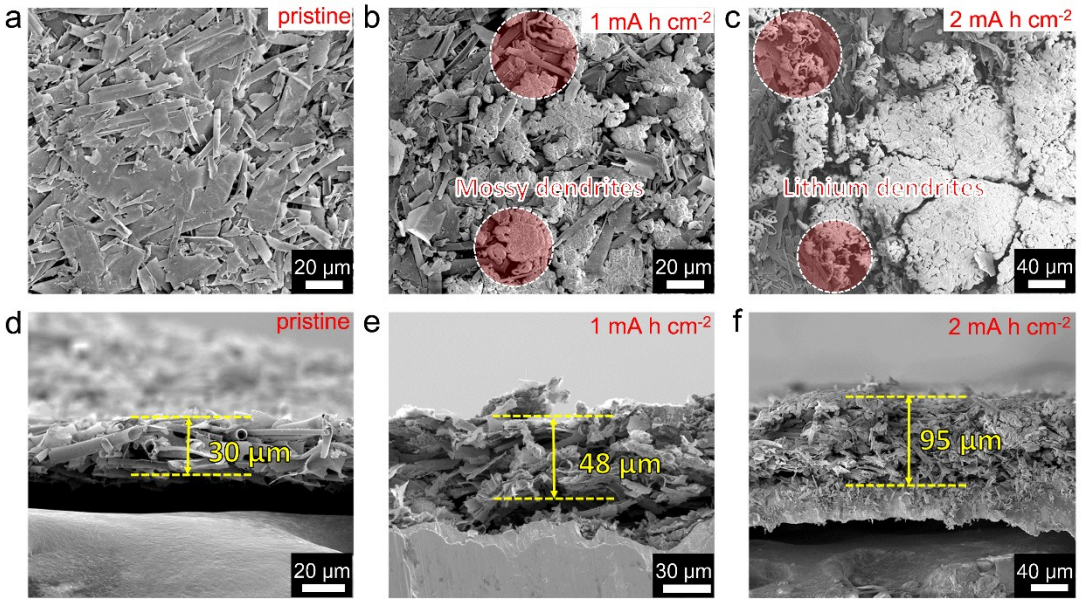
Supplementary figure 5. (a) Wall thickness of S-GCT. (b) Corresponding selected area diffraction (SAED) of the S-GCT.



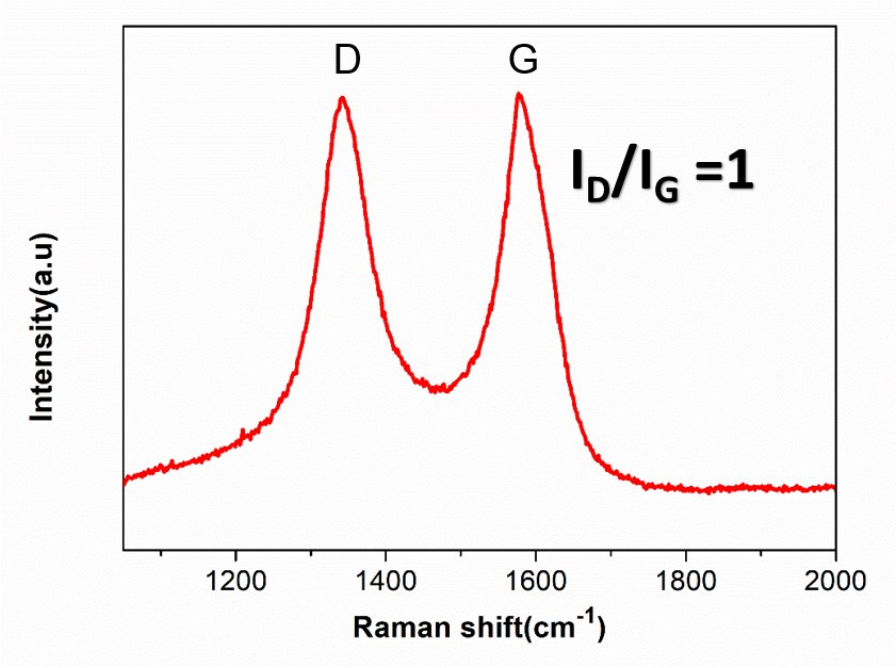
Supplementary figure 6. Nitrogen adsorption-desorption isotherm of S-GCT.



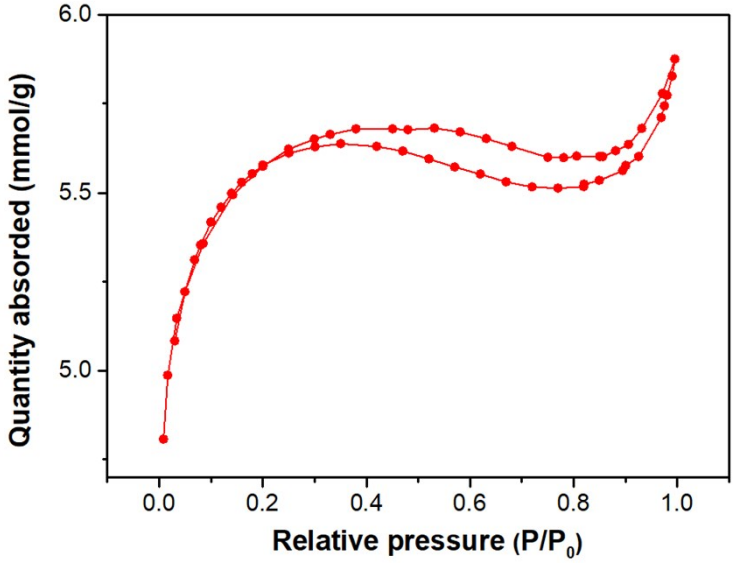
Supplementary figure 7. SEM image of Li metal deposition behavior of S-GCR. (a, d) pristine S-GCR electrode. (b, e) After plating 1 mA h cm^{-2}. (c, f) After plating 2 mA h cm^{-2}.



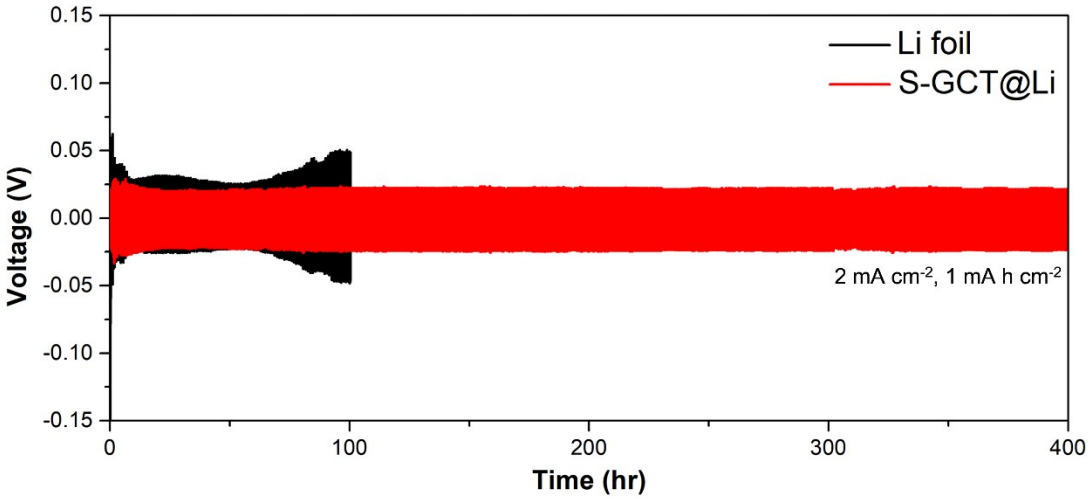
Supplementary figure 8. SEM image of Li metal deposition behavior of S-ACT. (a, d) pristine S-ACT electrode. (b, e) After plating 1 mA h cm^{-2}. (c, f) After plating 2 mA h cm^{-2}.



Supplementary figure 9. Raman spectra of the S-ACT.



Supplementary figure 10. Nitrogen adsorption-desorption isotherm of S-ACT.



Supplementary figure 11. Voltage profiles of metallic Li plating/stripping in two styles of symmetric cells (S-GCT@Li and Li foil) at 2 mA cm^{-2} for 1 mA h cm^{-2}

Supplementary Table 1. Comparison of the Li volumetric and specific capacity of S-GCT and other reported electrodes

Electrode	Thickness /μm	Current density /mA cm^{-2}	Capacity /mAh cm^{-2}	Cycle number	Coulombic Efficiency/%	Volumetric Capacity /mAh cm^{-3}	Specific Capacity / mAh g^{-1}	References
Graphitized Carbon Fiber (GCF)	1000	0.5	8	70	98	80	1254 (8 mAh cm^{-2})	Adv.Mater., 2017, 1700398
Hollow Corbon Fiber (HCF)	165	1	6	75	99	363	2400 (6 mAh cm^{-2})	Joule 2017,1,1
Unstacked Graphene (UG)	30	1	2	50	93	667	2666 (2 mAh cm^{-2})	Adv.Mater., 2016, 28, 2155
N-doped Graphene (NG)	150	2	5	100	96	333	2002 (5 mAh cm^{-2})	Angew. Chem., 2017,56,7764
N-enriched Graphitic Carbon Sponge(NGCS)	140	1	3	70	97	214	3175 (4 mAh cm^{-2})	J Power Sources, 2018, 386:77-84
Graphite Carbon Microtubes (GCM)	120	5	10	100	97.5	830	9130 (10 mAh cm^{-2})	Adv. Mater., 2017, 1700783
Crumpled Graphene Balls (CGB)	120	1	10		98	833	1428.6 (10 mAh cm^{-2})	Joule., 2018, 2, 184
S-GCT	**30**	**0.5**	**1**	**400**	**99.3**	**1333**	**7800 (4 mAh cm^{-2})**	**This work**
		1	**2**	**200**	**99.1**			
		2	**4**	**160**	**98.5**			

Cost calculation for preparation of S-GCT: the tube furnace can prepare up to 100 g of sample at a time, wherein the carbon content is about 4% of the total mass.

Item name	Dosage	specification	Unit price	Price
Aluminosilicate fiber	100 g	>99%	2000 RMB/t	0.2 RMB
Electric charge	10 kw h		0.5 RMB/kw	5 RMB
Gas (Ar)	200 L	>99.99%	20 RMB/m^3	4 RMB
Gas (CH_4)	2 L	>99.99%	200 RMB/m^3	0.4 RMB
Total price	9.9 RMB			

Supplementary Table 2. Cost calculation.

The price of the S-GCT is $Per_g \approx 2.5$ RMB, and the specific capacity of the S-GCT is 7.8 Ah g^{-1}, we can figure out Per_{Ah} = 0.32 RMB=0.0476 $ (2.5 RMB g-1/ 7.8 Ah g^{-1}).