

## **Supporting Information**

### **Designed graphite with activated edge for fast-charging lithium-ion storage properties**

## **Experimental Methods**

### **Synthetic Process:**

Fabrication of GWAE precursor: The GWAE precursor was fabricated by using solution treating. First, 3.0 g pristine graphite (purchased from Laixi City Xin Carbon Source Commercial Department) was fully dispersed in the 200 mL distilled water by stirring and ultrasonic dispersion. Then, 100 mL 4.0 M sulfuric acid aqueous was slowly added to the graphite dispersed aqueous and stirring for 12 h at room temperature, subsequently 100 mL 1 M potassium hydroxide solution was poured into the mixing solution and stirring 12 h. The GWAE precursor was obtained by cleaning with distilled water and freeze dry. Besides, the mass of GWAE precursor is 2.802g and the production yield is 93.33%.

Fabrication of GWAE: The GWAE was synthesized by sintering at 800 °C for 10 h in the Ar<sub>2</sub>-H<sub>2</sub> atmosphere with a heating rate of 5 °C min<sup>-1</sup>. The obtained black powdered material was the prepared GWAE.

### **Material Characterizations:**

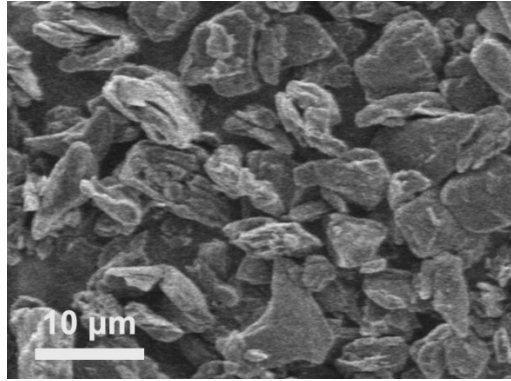
The X-ray diffraction (XRD) (Rigaku X-ray diffractometer, Cu K $\alpha$ ,  $\lambda = 1.5418 \text{ \AA}$ ) was employed to expounding crystal phases with scanning rate of 20° min<sup>-1</sup>. The morphological and microstructural were measured by the scanning electron microscope (SEM, Nova NanoSEM-23) and high-resolution transmission electron microscope (HRTEM), respectively. Raman spectra results were obtained on an HR-800 spectrometer (Jobin-Yvon LabRAM) at 514 nm laser excitation range from 200 to 3500 cm<sup>-1</sup>.

### **Electrochemical Properties Evaluation:**

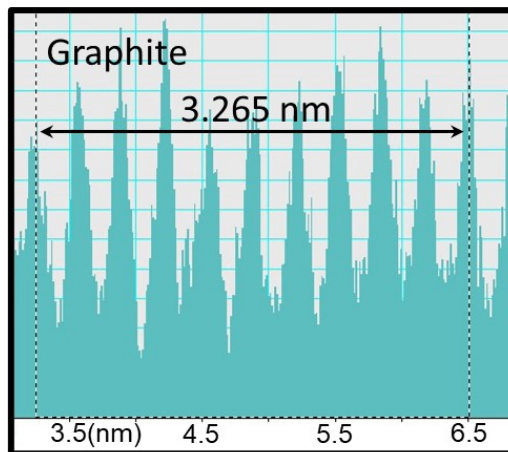
In half-cell, The GWAE and graphite electrode was made on the copper foil with the ratio 80:10:10 wt% of active composite, conductive carbon black and polyvinylidene fluoride (PVDF) in N-methyl pyrrolidone (NMP) solvent. The active materials loading in electrode is 3.0-5.0 mg cm<sup>-2</sup>. In a glovebox filled with argon, the CR2032 coin-cells were sealed with Celgard 2400 microporous polyethylene membrane, lithium foils and electrolyte. The electrolyte is 1M LiPF<sub>6</sub>, LiBF<sub>4</sub> and LiDFOB in carbonate acetate (EC): diethyl carbonate (DEC): ethyl methyl carbonate (EMC)=1:1:1 Vol%. The LAND2001CT is used for measuring the galvanostatic charge/discharge tests at 25 °C. The electrochemical workstation (Zahner Bio-Logic EC-LAB SP-300) measured the cyclic voltammetry (CV), electrochemical impedance spectra (EIS) and Tafel curves in the potential window between 0.01 and 3.0 V, an alternating current amplitude of 5 mV in the frequency range of 100000-0.01 Hz and voltage range from -0.04 V to 0.04 V, respectively. The full-cells were assembled with GWAE anode and LFP cathode in an Ar<sub>2</sub>-filled dry room. The cathode formulation was 90 wt.% active materials, 5 wt.% Super P carbon (SP) and 5 wt.% polyvinylidene fluoride (PVDF) in NMP onto a piece of aluminum foil. The anode electrode is similar with that in half-cells. 1M LiPF<sub>6</sub> in carbonate acetate (EC): diethyl carbonate (DEC): ethyl methyl carbonate (EMC)=1:1:1 Vol% was used as the electrolyte. The galvanostatic charge/discharge tests were measured in the voltage range of 1.0-3.6 V in full-cells.

### **Electrochemical tests for capacity contribution of intercalation:**

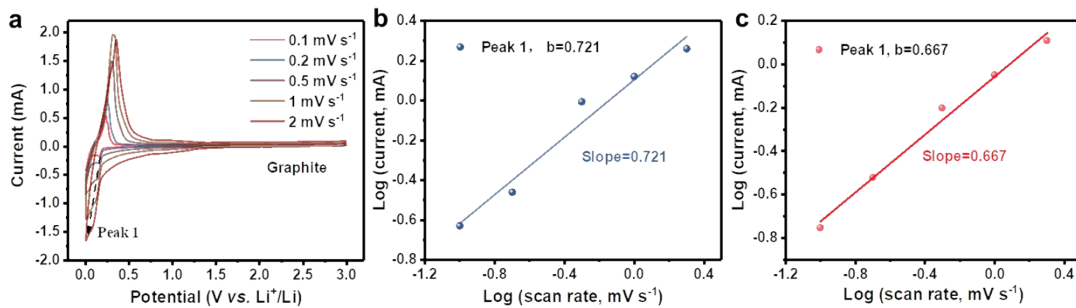
Graphite and GWAE were first mixed with PVDF and conductive carbon black in a weight ratio of 8/1/1 in NMP to cast onto 200  $\mu\text{m}$  copper foils. CR2032 cells were assembled with the working electrode, Li plate counter electrode and electrolyte of 1.0 M lithium hexafluorophosphate ( $\text{LiPF}_6$ ) in carbonate acetate (EC): diethyl carbonate (DEC): ethyl methyl carbonate (EMC)= 1:1:1 Vol% in argon atmosphere. As displayed in **Table S7**, all cells are firstly executed step 1.1 to 1.5 in the potential range of 0.01-3.0 V at rate of 0.1 C and the discharging capacity of the third cycle is defined as the standard capacity ( $Q_d$ ). Then cells are implemented by discharging to the  $Q_d$  at high rates with Li plating reaction occurrence, which is corresponding to the area of potential below 0 V in discharging curves. The simplified high rates statements with 1 C, 3 C, 5 C and 8 C are thereby implemented throughout step 2.1 to 2.4 to measure the capacity contribution of intercalation.



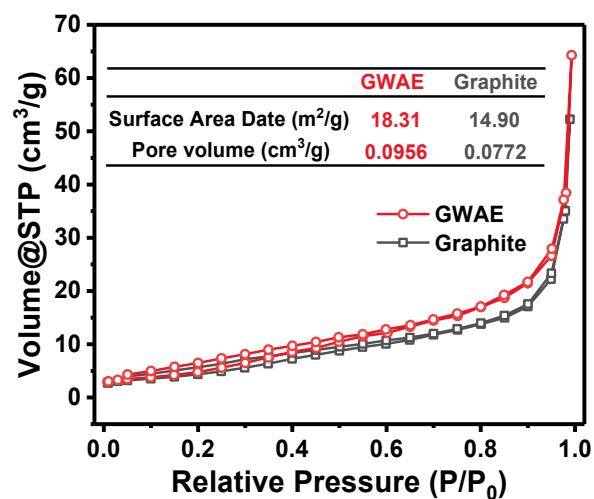
**Fig. S1.** The SEM images of graphite.



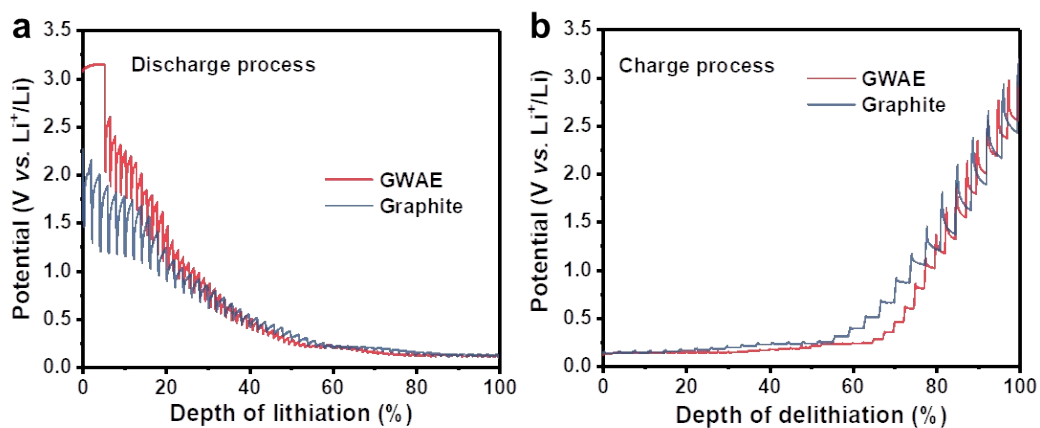
**Fig. S2.** Pristine calculated dates of layer spacing for graphite.



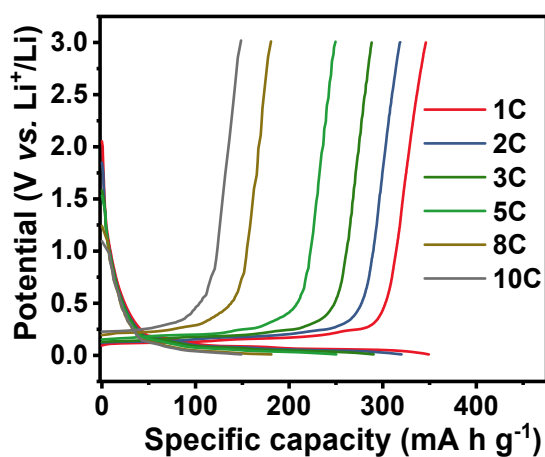
**Fig. S3.** (a) The CV curves at different sweep rates of graphite. The plots of  $\log(i)$  vs  $\log(v)$  calculated from CV curves of (b) graphite and (c) GWAE.



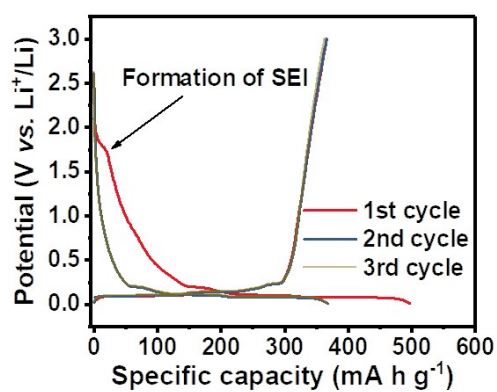
**Fig. S4.** The N<sub>2</sub> adsorption-desorption curves of the pristine graphite and GWAE.



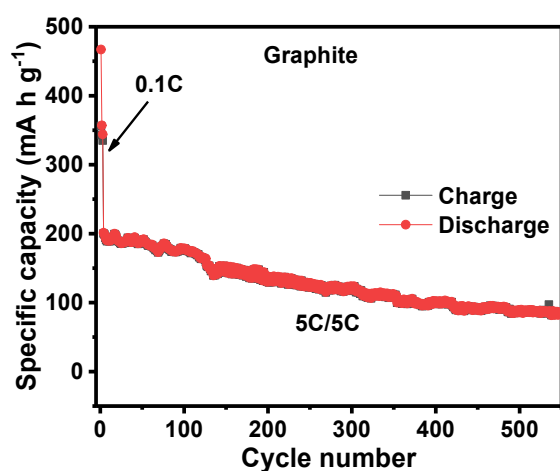
**Fig. S5.** GITT profiles (current pulse at 35 mA g<sup>-1</sup> for 10 min followed by 40 min relaxation) for GWAE and pristine graphite anode during (a) charge process and (b) discharge process.



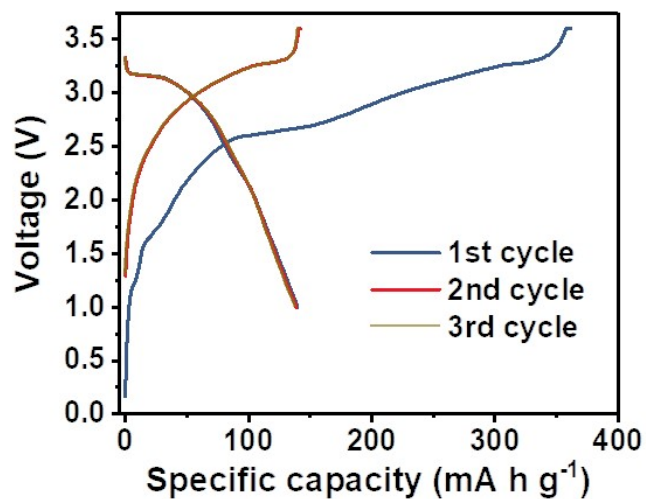
**Fig. S6.** Charge and discharge curves of GWAE at different rates range from 1 C to 10 C in half-cells.



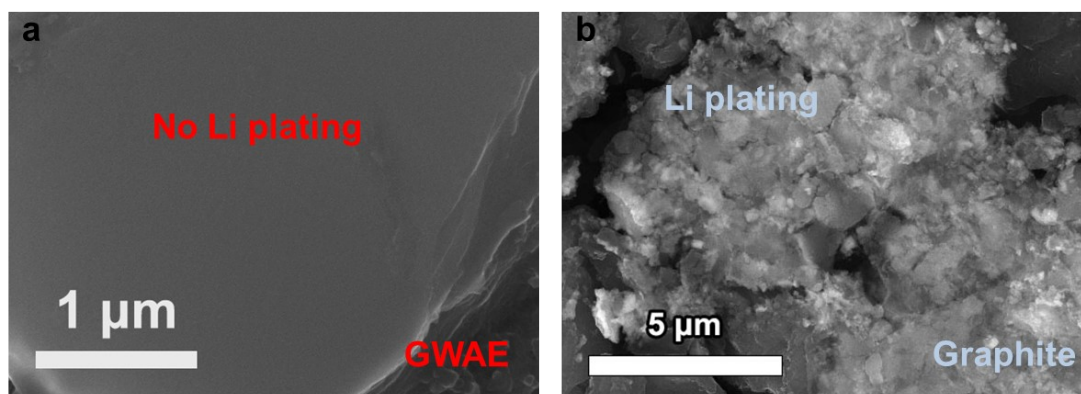
**Fig. S7.** Charge and discharge curves of GWAE at rate of 0.1 C in initial three cycles in half-cells.



**Fig. S8.** Cycle performance of graphite in half-cells at 5 C/5 C charge/discharge rate.



**Fig. S9.** Charge and discharge curves of GWAE at rate of 0.1 C in initial three cycles in full-cells with LFP cathode.



**Fig. S10.** SEM images of (a) graphite and (b) GWAE after 700 cycles.



**Table S1.** The  $R_{ct}$  values in various temperatures of HCRG and graphite.

Sample	$R_{ct}$ ( $\Omega$ )/ 0 °C	$R_{ct}$ ( $\Omega$ )/ 5 °C	$R_{ct}$ ( $\Omega$ )/ 10 °C	$R_{ct}$ ( $\Omega$ )/ 15 °C	$R_{ct}$ ( $\Omega$ )/ 20 °C	$R_{ct}$ ( $\Omega$ )/ 25 °C
HCRG	2120	1349	1020	580	400	210
Graphite	2550	1780	1200	790	520	340

**Table S2.** The  $R_{ct}$  values in charging and discharging process of HCRG and graphite.

Sample/Potential	-1.5 V	-0.6 V	-0.2 V	-0.1 V	0.1V	0.2 V	0.6 V	1.5V
HCRG	63.61	77.22	30.47	31.7	30.4	33.14	128.4	115.8
Graphite	123.8	126.7	39.16	47.06	41.19	44.34	132.1	125.7

**Table S3.** The calculated overpotential values at different current density for GWAE.

Current(mA )	0.2	0.5	1	2	5
	0.4257	0.9626	0.9487	0.9546	1.1461
	0.8405	0.6238	0.8358	0.6665	0.6427
	0.5841	0.7698	0.7003	0.6241	0.58
	0.5106	0.585	0.572	0.59	0.5404
	0.4799	0.5627	0.5255	0.5645	0.5122
	0.7583	0.5292	0.5137	0.5528	0.4731
	0.4204	0.4936	0.5782	0.5748	0.558
	0.412	0.4815	0.5494	0.5363	0.506
	0.7688	0.4731	0.47	0.5376	0.6017
	0.3869	0.4678	0.4632	0.4833	0.4839
	0.8637				

Average	0.57645	0.59491	0.61568	0.60845	0.60441
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**Table S4.** The calculated overpotential values at different current density for graphite.

Current/mA	0.2	0.5	1	2	5
	1.4655	1.2004	1.334	1.4661	2.1543
	1.716	0.9356	1.1691	1.3935	1.7308
	1.2122	0.9009	1.0863	1.2776	1.711
	1.0894	0.8712	1.0364	1.1775	1.5464
	1.0007	0.8457	0.8566	1.1474	1.4887
	0.6873	0.833	0.8126	1.1471	1.2894
	0.6331	0.8163	0.7989	1.1282	1.2122
	0.6176	0.7989	0.7822	1.1217	1.3105
	0.599	0.7781	0.7704	1.0841	1.1334
	0.5754	0.7515	0.8287	1.0981	1.0934
	0.5583				
Average	0.92314	0.87316	0.94752	1.20413	1.46701

**Table S5.** The calculated contribution of intercalation values of GWAE.

Rate/C	1	3	5	8
Sample 1	80.96	68.01	44.22	35.55
Sample 2	81.66	66.55	43.85	34.99
Sample 3	82.05	67.51	45.01	36.01
Sample 4	81.98	67.02	44.55	36.21
Sample 5	82.25	66.36	43.92	35.64
Average	81.78	67.09	44.31	35.68

**Table S6.** The calculated contribution of intercalation values of graphite.

<b>Rate/C</b>	<b>1</b>	<b>3</b>	<b>5</b>	<b>8</b>
Sample 1	77.20	50.55	37.43	23.92
Sample 2	78.12	50.43	36.88	24.86
Sample 3	76.89	50.98	38.22	24.44
Sample 4	77.35	51.02	37.43	23.98
Sample 5	76.89	51.27	37.94	24.30
Average	77.29	50.85	37.58	24.30

**Table S7.** The experimental steps involving pre-cycle and contribution of intercalation tests.

<b>Number</b>	<b>Step</b>	<b>Value</b>	<b>SOC/cycle number/cut-off volatge</b>
<b>1</b>	<b>Pre-cycle</b>		
1.1	Discharge	0.1 C	0.01 V
1.2	Relaxation	5 min	
1.3	Charge	0.1 C	3.00 V
1.4	Relaxation	5 min	
1.5	Cycle (1.1 to 1.4)	0.1 C	2 times
<b>2</b>	<b>Contribution of intercalation tests</b>		
2.1	Discharge	1 C/3 C/ 5 C/ 8 C	100%SOC
2.2	Charge	Corresponding discharge	3 V
2.3	Relaxation	5 min	
2.4	Cycle (2.1 to 2.3)		30 times

### Supplementary Note 1:

The  $\eta_{Li}^{Pl}$  is easy to decrease below 0 V vs. Li/Li<sup>+</sup> at high rate, which results in Li plating on the surface of graphite.

$$\eta_{Li}^{Pl} = \eta_{inter} + \eta_{diff} + \eta_e \quad (S1)$$

Where  $\eta_{inter}$ ,  $\eta_{diff}$  and  $\eta_e$  is corresponding to the polarization of lithium-ions intercalation at interface, lithium-ions diffusion in bulk and electrode diffusion, respectively.<sup>1</sup>

### Supplementary Note 2:

The value of b can be decided follows the equation:

$$i = av^b \quad (S2)$$

Where  $i$  is current and  $v$  is sweep rate. The value of b can be decided by calculating the oblique line of log(i)-log(v) curves, which is represented pseudocapacitive contribution (b = 1) or ionic diffusion-controlled reaction (b = 0.5).

The percentage of pseudocapacitive contribution can be further calculated by the following equation:

$$i/v^{1/2} = k_1v^{1/2} + k_2 \quad (S3)$$

The constants of  $k_1$  and  $k_2$  are obtained by this equation.<sup>2</sup>

### Supplementary Note 3:

The relative electrochemical active surface area was evaluated by the relationship between scan rate and peak current from the Randles-Sevcik equation.

$$I_p = (2.69 \times 10^5) n^{3/2} A D^{1/2} C_0 v^{1/2} \quad (\text{S5})$$

Where  $I_p$  is peak current.  $n$ , the number of transferred electrons.  $A$ , the active surface area.  $D$ , the diffusion coefficient of Li ion.  $C_0$ , the bulk concentration of Li ion.  $v$ , the scan rates.<sup>4</sup>

### Supplementary Note 4:

The value of  $D_{Li^+}$  can be calculated according to the simplified Fick's Second Law by following equation:

$$D_{Li^+} = \frac{4}{\pi} \left( \frac{V_m}{S F z_i} \right)^2 \left( \frac{dE/dx}{dE/dt^{1/2}} \right)^2 \quad (\text{S4})$$

where  $I_0$  is the current,  $V_m$ ,  $S$ ,  $F$ , and  $z_i$  denote the molar volume, interfacial area of electrode-electrolyte, Faraday constant, and the number of electrons per molecule during redox reaction,  $dE/dx$  is differential with final potential after rest to percentage of capacity,  $dE/dt^{1/2}$  is differential with potential to square root of time for each single-step.<sup>3</sup>

### Supplementary Note 5:

Hypothesizing that each transport step is a simple thermally activated process, that can raise the activation energies for  $\text{Li}^+$  transport during these reactions from their Arrhenius behaviors by drawing  $\log(R^{-1})$  versus reciprocal temperature  $1000/T$  plots:<sup>5</sup>

$$E_a = -19.144 \times \text{slope (kJ mol}^{-1}\text{)} \quad (\text{S6})$$

### Supplementary Note 6:

The corresponding contribution of intercalation of GWAE and graphite are calculated to further confirm the effect of activated edge. The contribution of intercalation ( $CE_{inter}$ ) and lithium plating ( $CE_{plat}$ ) are defined as the ratio of intercalation capacity ( $Q_{inter}$ , the capacity is keeping with potential above 0 V) to total discharge capacity ( $Q_d$ ) and the ratio of Li plating capacity ( $Q_{plat}$ , the capacity is keeping with potential below 0 V) to total discharge capacity ( $Q_d$ ):

$$CE_{inter} = \frac{Q_{inter}}{Q_d} \quad (\text{S7})$$

$$CE_{plat} = \frac{Q_{plat}}{Q_d} \quad (\text{S8})$$

$$Q_d = Q_{plat} + Q_{inter} \quad (\text{S9})$$

### Supplementary references

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4. X. Han, Q. Meng, X. Wan, B. Y. Sun, Y. Zhang, B. C. Shen, J. L. Gao, Y. L. Ma, P. J. Zuo, S. F. Lou and G. P. Yin, *Nano Energy*, 2021, **81**, 105635.
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