Electronic Supplementary Information (ESI)

Interconnected highly graphitic carbon nanosheets derived from wheat stalk as high performance anode materials for lithium ion battery

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Experimental Section

Material Preparation

Highly graphitic carbon nanosheets (HGCNS) were prepared by carbonization and graphitization of the hydrothermal product of wheat stalk. Detailed procedures are as follows: The raw material was first cut into small pieces (about 3 cm in length) and washed with deionized water, and then dried at 60 °C for 12 h before use. Within the underlying hydrothermal treatment, 4 g of the cleaned wheat stalk and 280 mL of homogeneous 3 M KOH solution were placed in a 400 mL Teflon-lined stainless steel autoclave. The autoclave was sealed and heated at a temperature of 150 °C for 6 h and then allowed to cool down to room temperature naturally. The resulting samples were fetched out, gathered by vacuum filtration with filter paper, washed by ultrasonication in 120 mL distilled water for 30 min and dried at 80 °C in an electric oven. Next, the as-prepared products were calcined at 800 $^\circ C$ for 3 h in N_2 atmosphere at a heating rate of 5 °C min⁻¹ and a N₂ flow rate of 150 cm³ min⁻¹. The evolved carbon was washed with 0.5 M HCl solution to remove the residual KOH and dried in a vacuum oven at 80 °C for 12 h. Finally, the product was thermally treated at 2600 °C for 5 min using a graphite furnace in an argon atmosphere. The synthetic processes and production of each step are shown in Figure 1. For comparison, the cleaned wheat stalk carbonized at 800 °C directly without hydrothermal

and graphitization is called WS, the graphitic WS is denominated as GWS. The non-graphitization carbon nanosheets (CNS) were also prepared, according to similar procedures.

Material Characterization

The morphologies and structures of the samples were investigated by SEM (Nova NanoSEM230, Japan), TEM (JEM-2100F, Japan), HRTEM (JEM-2100F, Japan; Titan G2 60-300, Netherlands), SAED measurements and atomic force microscopy (AFM, NanoScope (R) III, Bruker, USA). The crystalline structures of the products were performed by X-ray diffraction (XRD, Rigaku-TTRIII, Japan). X-ray photoelectron spectra (XPS) were recorded by using an X-ray photoelectron spectrometer (K-Alpha 1063) with a monochromatic Al Ka X-ray source. Raman spectra were obtained using a LabRAM Hr800 from HORIBA Jobin Yvon. Surface area, pore volume and pore size were recorded from nitrogen adsorption and desorption isotherms performed on a Surface Area and Porosity Analyzer (ASAP 2020 HD88). The total pore volume of the samples was calculated at a relative pressure of 0.999 (P/P_o). The specific surface area and pore size distribution were determined according to the Brunauer-Emmett-Teller (BET) theory and the nonlocal density functional theory (NLDFT) model from the adsorption branch of the isotherm. Elemental analysis was done using a EuroEA3000 (Leeman, USA) Analyzer. Digital photographs were taken with a Nikon COOLPIX

s4300 Camera. Following determination of d_{002} , the degree of graphitization was calculated according to

$$D_{g} = \frac{3.44 - d_{002}}{3.44 - 3.354} \times 100\%$$
(1)

In this equation, 3.44 represents the interlayer spacing of turbostratic graphite (in Å) and 3.354 is the interlayer spacing for single crystal graphite (in Å).

Electrochemical Measurement

For electrochemical tests, the composite electrodes were fabricated by mixing 80% active materials with 10% conductive carbon black, and 10% polyvinylidene fluoride (PVDF) binder and dispersing the mixture in N-methyl-2-pyrrolidinone (NMP) to prepare slurry. The slurry was then coated on copper foil using a doctor blade, followed by evaporating the NMP at 120 °C overnight in a vacuum oven, resulting in electrodes with a mass loading of ~1 mg cm⁻². The electrodes were cut into disks of 10 mm in diameter and then were assembled into 2025 coin-type cells in a glove box (Super 1220/750, Shanghai Mikrouna Co. Ltd.) filled with Ar gas. Lithium foil was used as the counter electrode, and a polyethylene film was used as the separator. The electrolyte was 1 M LiPF₆ in ethylene carbonate/dimethyl carbonate (1:1), the related amount of electrolyte was 150 µL for a coil cell. Galvanostatic charging-discharging was carried out on LAND CT-2001A in the potential range of 0-3.0 V versus Li/Li⁺ at a certain current density value. The cyclic voltammogram (CV) measurements were conducted on a PARSTAT 4000 electrochemical workstation in the same range at a scanning rate of 0.2 mV S⁻¹. The first cycle began with the discharge process, corresponding to the insertion of lithium into the electrodes. The electrochemical impedance spectroscopy (EIS) was performed by a PARSTAT 4000 electrochemical workstation in the 100 kHz to 10 mHz frequency range in automatic sweep mode from high to low frequency.



Figure S1. SEM (a-b) and TEM images (c-d) of WS.



Figure S2. SEM (a-b) and TEM images (c-d) of GWS.



Figure S3. SEM (a-b) and TEM images (c-d) of CNS.



Figure S4. HRTEM images of HGCNS.

samples	C [wt %]	H [wt %]	O [wt %]
HGCNS	93.20	0.16	0.20

Table S1. The combustion elemental analysis for HGCNS.



Figure S5. Cyclic voltammograms of WS (a), CNS (c), GWS (e) electrodes between 0 and 3 V at a scan rate of 0.2 mV s⁻¹, galvanostatic discharge/charge curves of WS (b), CNS (d), GWS (f) electrodes at 0.1 C.



Figure S6. SEM (a-b) and TEM images (c-d) of flake graphite.



Figure S7. (a) Galvanostatic discharge/charge curves of flake graphite electrode at 0.1 C, (b) extended cycling performance of flake graphite electrode at 0.1C.



Figure S8. Galvanostatic discharge/charge curves of WS (a), CNS (b), GWS (c), and flake graphite (d) electrodes at different rates.



Figure S9. (a) Galvanostatic discharge/charge curves of CNS-1600, CNS-2100, and HGCNS electrodes at 0.1 C, (b) extended cycling performance of CNS-1600, CNS-2100, and HGCNS electrodes at 1 C (The electrode was firstly discharged-charged at 0.1 C for two cycles to activate the battery).



Figure S10. Typical Nyquist plots of HGCNS electrode after 2000 cycles at 5 C.

 Table S2. Kinetic parameters of HGCNS electrode after 2000 cycles at 5

 C.

Samples	$R_{e}\left[\Omega ight]$	$R_{\rm f}\left[\Omega\right]$	$R_{ct}\left[\Omega\right]$	$i_0 [mA cm^{-2}]$
HGCNS	10.52	25.58	41.53	0.618

Note: The exchange current density i_0 was calculated according to the equation of $i_0 = RT/nFR_{ct}$.