Universal One-Step Graphitization of Silicon-Enriched Biomass-Derived Carbon in Molten Salts for Lithium-Ion Battery Anodes

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The equation for the diffusion coefficient of lithium ions is D_{GIT}[1–3]:

Equation (1)

$$D_{Li^+} = \frac{4}{\pi\tau}\left(\frac{m_B V_M}{M_B S}\right)^2\left(\frac{\triangle E_s}{\tau\left(dE_\tau \middle| d\sqrt{\tau}\right)}\right)^2 \left(\tau \ll L^2/D_{Li^+}\right) \quad (1)$$

If the relation between potential and $\tau^{1/2}$ follows a linear proportionality, equation (1) can be simplified even more to equation (2),

$$D^{GITT} = \frac{4}{\pi\tau}\left(\frac{m_B V_M}{M_B S}\right)^2\left(\frac{\triangle E_s}{\triangle E_\tau}\right)^2 \quad (2)$$

τ represents the duration of the constant current pulse, while m_B, V_M, S, and M_B denote the mass, molar volume, electrode-electrolyte interface area, and molar mass of the electrode material, respectively. ΔE_s is the voltage difference during the open circuit period, while $\Delta E\tau$ denotes the overall voltage change of the cell in response to a constant current pulse.

The equation for the diffusion of lithium ions is CV method:

$$D_{Li^+} = \frac{I_P^2\, R\, T}{0.1992\, F^3\, S^2\, c_0^2\, v}$$

$$I_P = \left(\frac{D_{Li^+}\ 0.1992\, F^3\, S^2\, c_0^2}{R\, T}\right)^{\frac{1}{2}} v^{\frac{1}{2}}$$

$$k = \left(\frac{D_{Li^+}\ 0.1992\, F^3\, S^2\, c_0^2}{R\, T}\right)^{\frac{1}{2}}$$

In the above equation, Ip represents the peak intensity (mA), R (J K^{-1} mol^{-1}) is the gas constant, T (K) is the temperature, F (C mol^{-1}) is the Faraday constant, S is the surface area of the electrode, which is 1.13 cm^2, Co is the molar concentration of lithium ions in the material (mol cm^{-3}), v represents the scan rate (mV s^{-1}), and k is the slope of the fitted curve.

The evaluation of lithium storage mechanisms is performed by analyzing the current (i) corresponding to the oxidation/reduction peaks in the cyclic voltammetry (CV) curves along with their corresponding scan rates (v). The equation is as follows:

$$i = av^b$$

$$\log(i) = \log(a) + b\log(v)$$

In the above equation, a and b are constants. When b approaches 1, the electrode is primarily controlled by capacitive adsorption behavior. When b approaches 0.5, the electrode is primarily controlled by ion diffusion behavior. Moreover, the capacitance contribution and diffusion contribution of the electrode can be further calculated using the equation $i = k_1v + k_2v^{1/2}$, where k_1 and k_2 are constants. The term k_1v represents the capacitance contribution of the electrode, while $k_2v^{1/2}$ represents the diffusion contribution of the electrode.

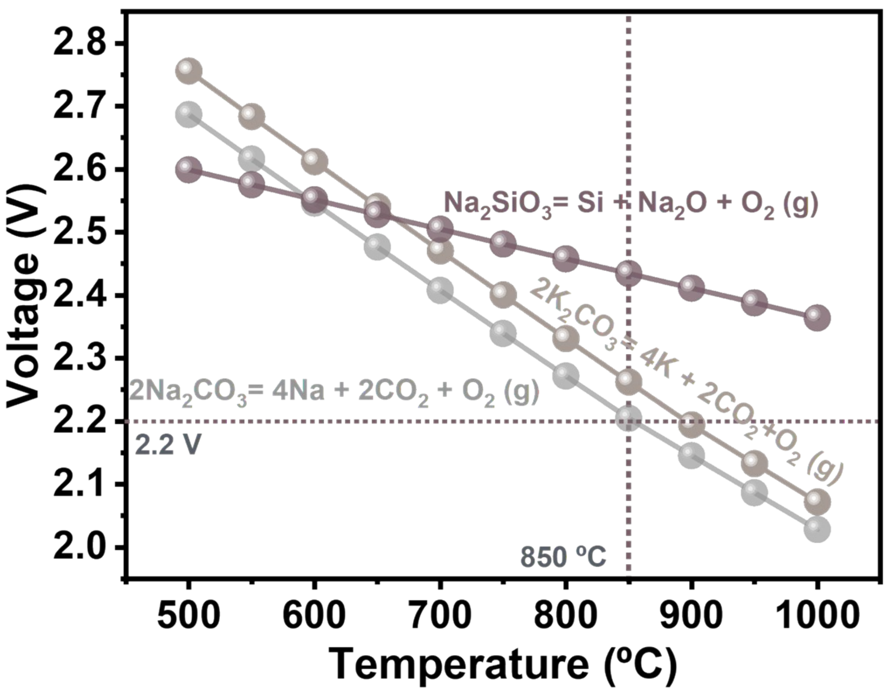

Figure S1. Profiles of the Gibbs free energy of different reactions as a function of temperature.

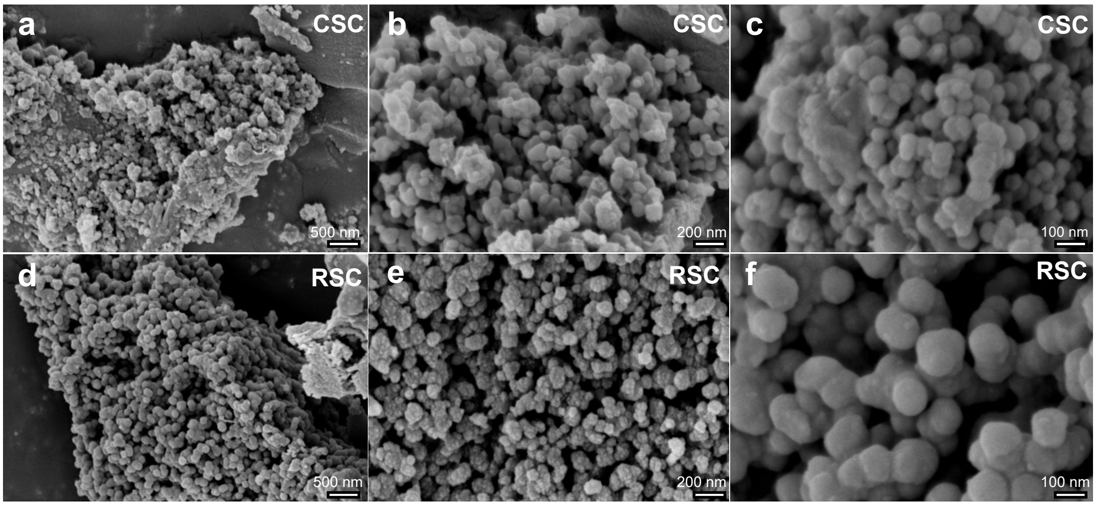

Figure S2. SEM images of different agricultural wastes.

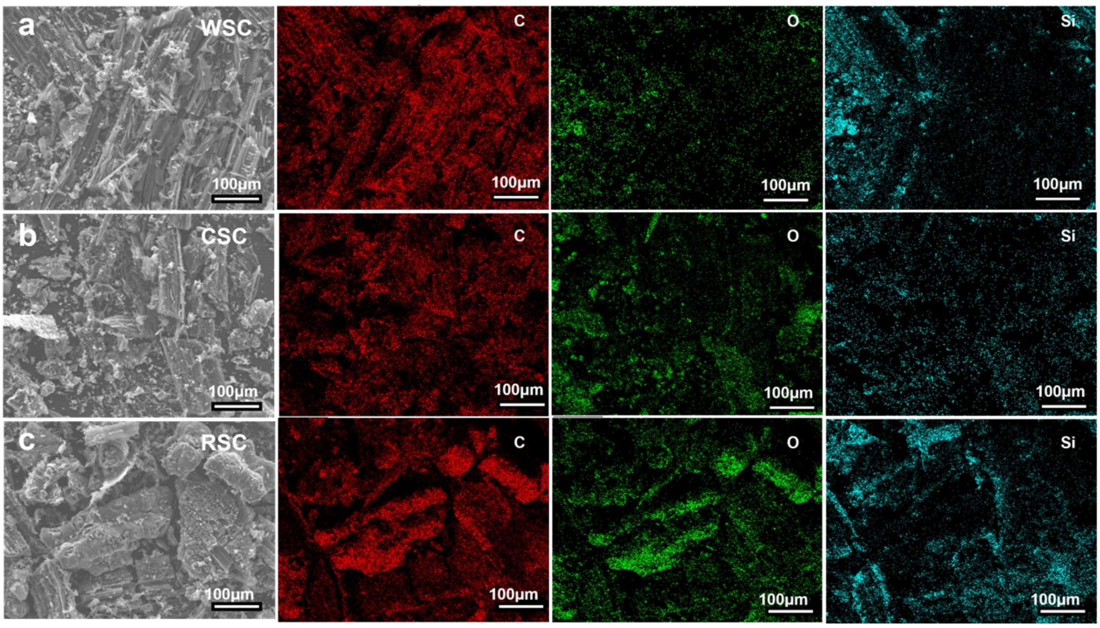

Figure S3. SEM-EDS mapping of the (a) WSC (b) CSC (c) RSC.

Table S1. Quantitative EDS analysis of major elements in WSC

Element	Weight	Atomic
C	79.06%	84.91
Si	5.18%	2.38
O	15.77%	12.71

Table S2. Quantitative EDS analysis of major elements in CSC

Element	Weight	Atomic
C	71.58%	78.85
Si	6.62%	3.12
O	21.8%	18.03

Table S3. Quantitative EDS analysis of major elements in RSC

Element	Weight	Atomic
C	54.28%	64.66
Si	14.42%	7.35
O	31.3%	27.99

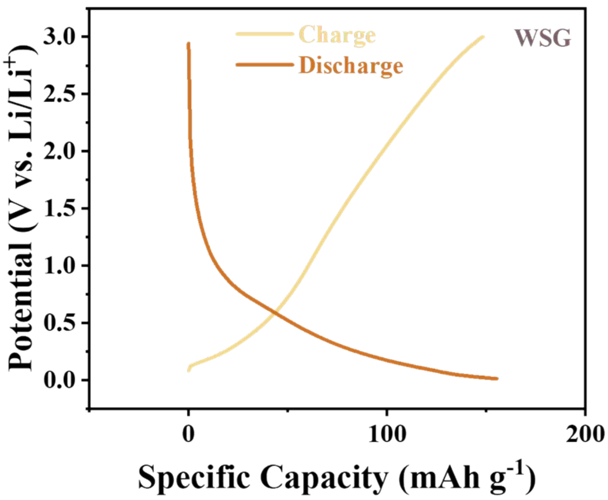

Figure S4. Charge-discharge curves after lithium intercalation.

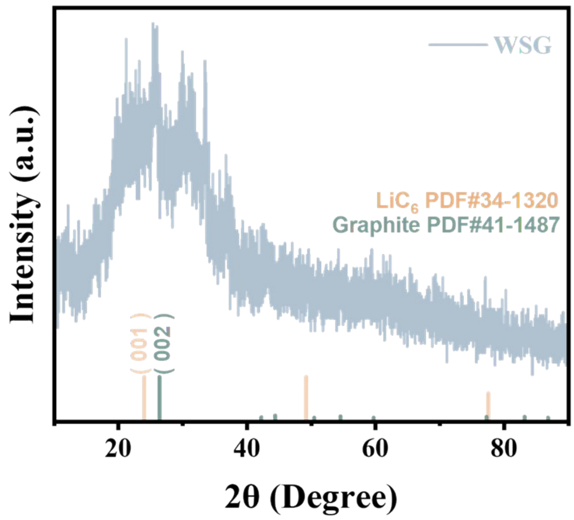

Figure S5. XRD pattern of the electrode sheet after lithium intercalation.

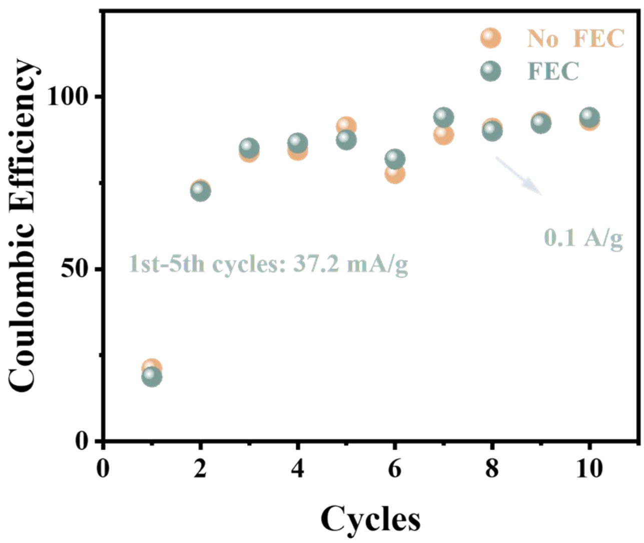

Figure S6. Coulombic efficiency of electrodes with and without FEC addition.

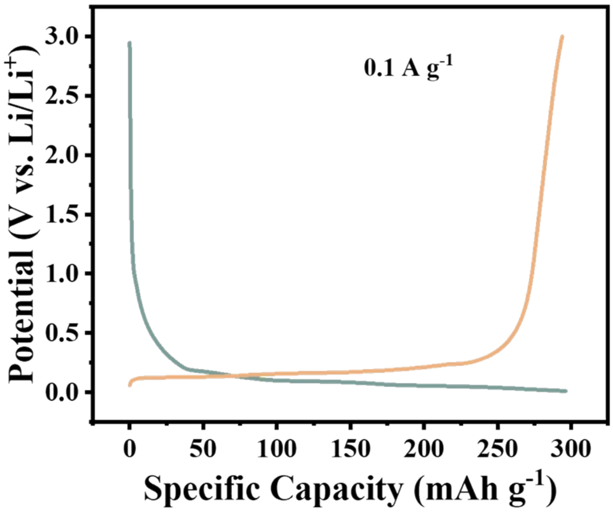

Figure S7. Charge-discharge curves of the commercial graphite electrode.

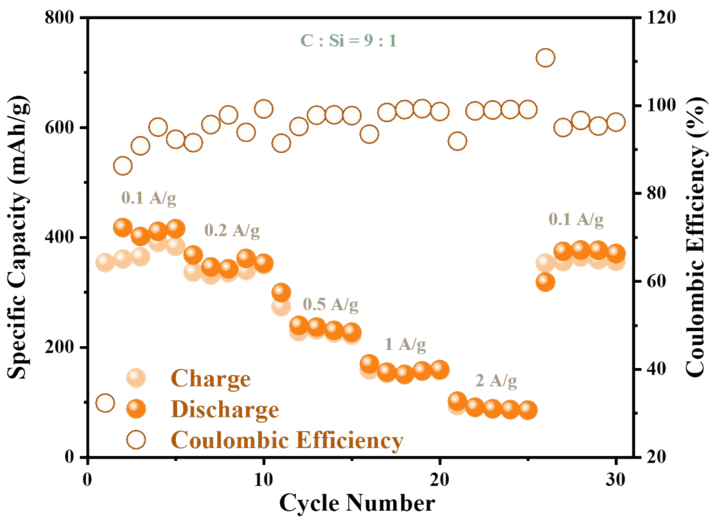

Figure S8. Rate performance of the Si/C electrodes (C: Si = 9: 1).

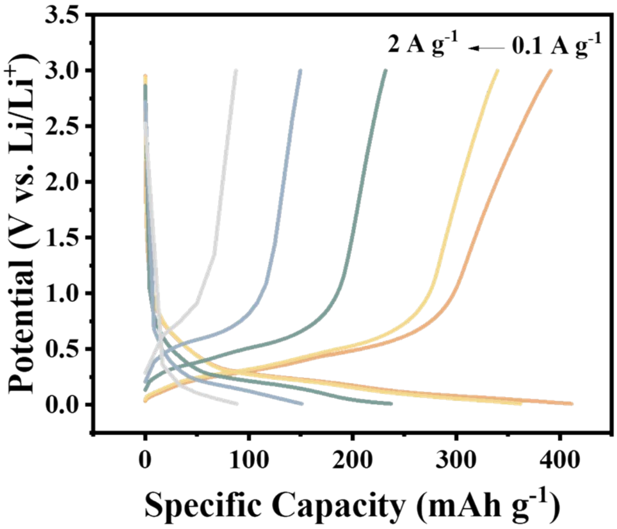

Figure S9. Charge-discharge curves of the Si/C electrodes (C: Si = 9: 1).

References

1 P. Li, J.-Y. Hwang and Y.-K. Sun, *ACS Nano*, 2019, **13**, 2624–2633.

2 M. Han, Z. Lin, X. Ji, Y. Mu, J. Li and J. Yu, *Mater. Today Energy*, 2020, **17**, 100445.

3 J.-I. Lee, Y. Ko, M. Shin, H.-K. Song, N.-S. Choi, M. G. Kim and S. Park, *Energy Environ. Sci.*, 2015, **8**, 2075–2084.