

Supplementary Information for

Plasma-coupled electrothermal ultrahigh-temperature graphitization of non-graphitizable hard carbons via heterogeneous edge-seeded growth

Pei-Yu Cao^{1,5}, Zhiwei Wang^{2,5}, Ming-Ze Yang^{1,5}, Su-Fan Hu^{1,5}, Junjie Fang², Huang
Huang³, Long Zhang¹, Tianze Zhang⁴, Yu Gu¹, Jing-Hua Tian¹, Wanlin Guo², Hu Qiu^{2*},
Ye-Chuang Han^{1*}, Zhong-Qun Tian¹

¹Innovation Laboratory for Sciences and Technologies of Energy Materials of Fujian
Province, State Key Laboratory of Physical Chemistry of Solid Surfaces, College of
Chemistry and Chemical Engineering, Xiamen University, Xiamen 361005, China.

²State Key Laboratory of Mechanics and Control for Aerospace Structures, Key
Laboratory for Intelligent Nano Materials and Devices of the Ministry of Education,
Institute of Nano Science, Nanjing University of Aeronautics and Astronautics, Nanjing
210016, China.

³State Key Laboratory of Photovoltaic Science and Technology, College of Smart
Materials and Future Energy, Fudan University, Shanghai 200433, China.

⁴State Key Laboratory of Electronic Thin Film and Integrated Devices, School of
Physics, University of Electronic Science and Technology of China, Chengdu 611731,
China.

⁵These authors contributed equally to this work.

*Corresponding author: giuhu@nuaa.edu.cn (H.Q.); ychan93@xmu.edu.cn (Y.-C.H.).

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1. Supplementary Text

1.1 Molecular Dynamics Simulations

In all MD simulations, the C/H/O-2016 force field developed by Ashraf and van Duin¹ was adopted, owing to its validated accuracy in modeling high-temperature pyrolysis of organic compounds and reliability in describing carbon-condensed phases and C/H/O chemistry^{2,3}. A time step of 0.1 fs was used, and temperature was controlled with the Nosé–Hoover thermostat^{4,5}. Periodic boundary conditions were applied in all directions. All simulations were conducted using the LAMMPS⁶. All visualization and analysis were performed using VMD⁷.

Since cellulose is the primary component of biomass straw, it was chosen as a representative model for hard carbon precursors. The model construction process is illustrated in Supplementary Fig. 1a. Starting from a 1,4- β -D-glucopyranose monomer, twenty monomers were connected to form a long-chain polymer. Twenty such polymer chains were then randomly placed in a cubic simulation cell to build a three-dimensional hard carbon model, using an initial low density of 0.5 g/cm³ to prevent atomic overlaps. The system then underwent energy minimization, followed by NVT equilibration simulation at 300 K for 100 ps. It was then gradually compressed in the NPT ensemble at 300 K and 0.1 GPa for another 100 ps. After compression, the structure was re-equilibrated in the NVT ensemble to reach the experimental cellulose density (~1.03 g/cm³)⁸. Subsequently, the system was heated to 2500 K at a rate of 10 K/ps and maintained under the NVT ensemble for carbonization (Supplementary Fig. 1b). This process continued until about 80% of hydrogen atoms were removed, releasing gaseous species such as H₂O, CO₂, CO, and H₂. As a result, the oxygen mass percentage decreased from 49.6% to 8.2%, while the carbon mass percentage increased from 44.4% to 87.5% (Supplementary Fig. 1c). Following carbonization, the gaseous byproducts were removed, and the system's density was preserved. Finally, graphitization simulations were performed across a temperature range of 1500–4000 K.

1.2 Kinetic analysis of sp²-carbon evolution using the Johnson–Mehl–Avrami–Kolmogorov (JMAK) model

The temporal evolution of the sp²-hybridized carbon fraction was analyzed using the classical JMAK model^{9–11}, which was originally developed to describe nucleation-and-growth-controlled phase transformations and has also been applied to sp² ordering and graphitization kinetics in carbonaceous materials^{12,13}. The normalized transformation fraction, $X(t)$, was fitted as:

$$X(t) = 1 - \exp[-(kt)^n] \quad (1)$$

k is the apparent ordering rate constant, and n is the Avrami exponent. In this work, $X(t)$ was defined from the sp²-hybridized carbon fraction as:

$$X(t) = \frac{f_{sp^2(t)} - f_0}{(f_\infty - f_0)} \quad (2)$$

where $f_{sp^2}(t)$ is the sp^2 -hybridized carbon fraction at time t , and f_0 and f_∞ denote the initial and apparent limiting sp^2 fractions, respectively. Accordingly, the fitted function for the sp^2 fraction can be written as:

$$f_{sp^2}(t) = f_0 + (f_\infty - f_0)\{1 - \exp[-(kt)^n]\} \quad (3)$$

During fitting, f_0 was taken as the initial sp^2 fraction, whereas f_∞ , k , and n were obtained as fitting parameters.

1.3 Finite element simulation

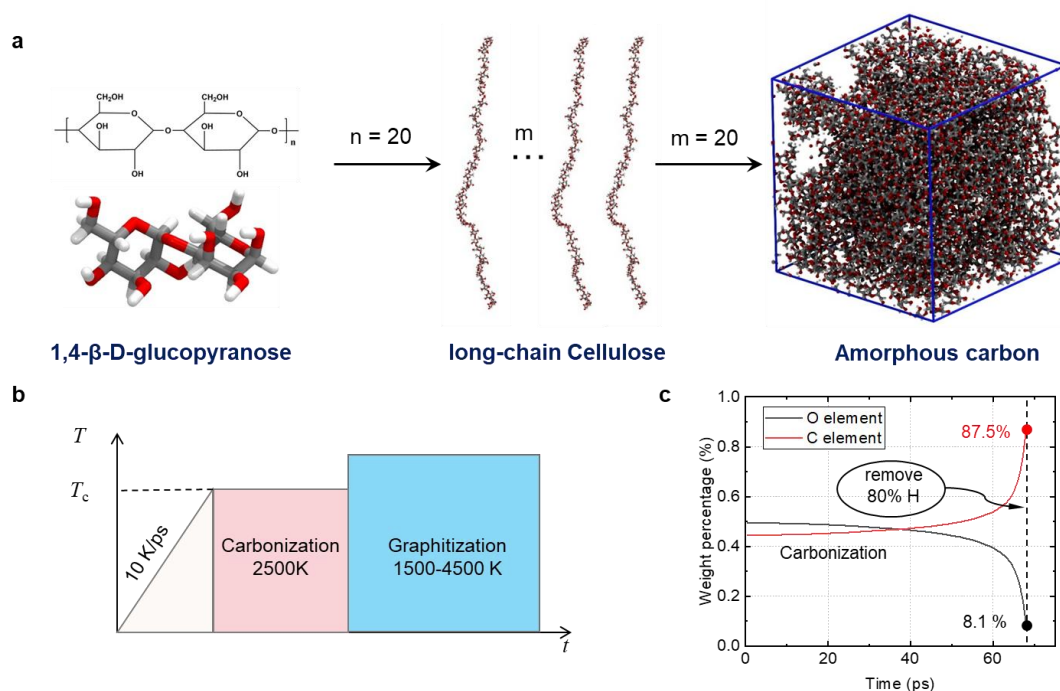
We conducted coupled electro-thermal simulations using the electric current and solid heat transfer modules in COMSOL Multiphysics. The model geometry consists of carbon felt in different shapes, with a cylindrical sample area embedded in the center. We set a constant temperature of 3800 K at the upper and lower boundaries of the sample area to simulate high-temperature plasma. The simulation considers Joule heating as a secondary heat source, and the corresponding heat generation is expressed as:

$$Q = J \cdot E = \sigma \cdot |E|^2 \quad (4)$$

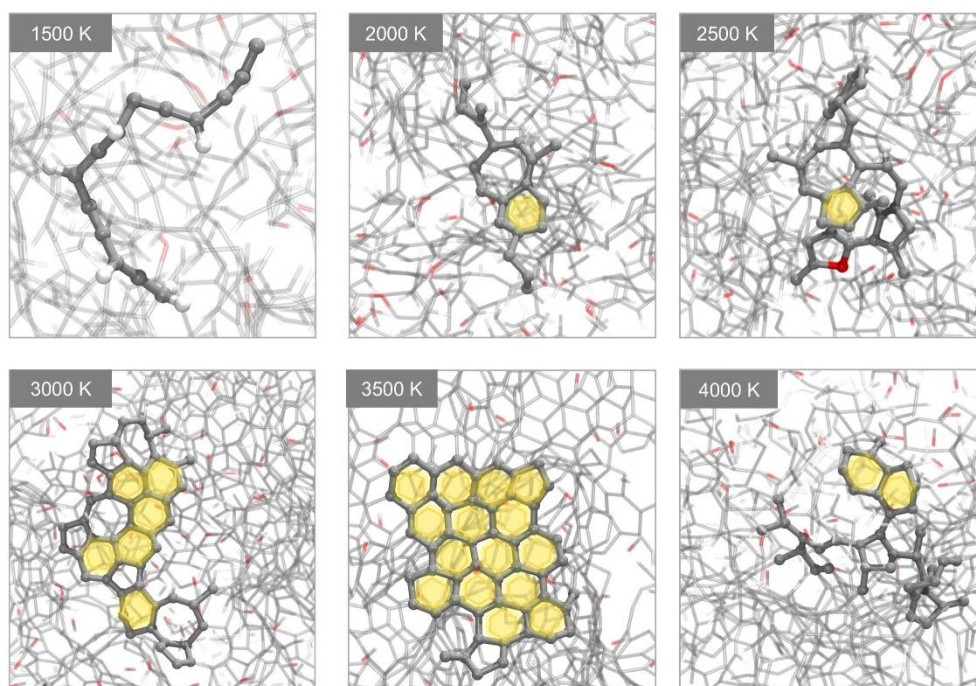
where J is the current density, E is the electric field, and σ is the electrical conductivity. A voltage was applied to the top boundary of the model while the bottom boundary was grounded, establishing a current flow. The current distribution was first solved using the electric currents module; the resultant Joule heating was then coupled as a heat source in the Heat Transfer in Solids module to compute the transient temperature field.

To achieve a target core sample temperature of 3500 K, we repeatedly adjusted the applied voltage to determine the steady-state power required to keep this temperature stable. Next, we optimized the voltage sequence to rapidly heat the sample close to 3500 K, then reduced the voltage to sustain the set temperature. We computed energy consumption by integrating volumetric power values for each second during the heating process. For material properties, including electrical and thermal conductivity, we adopted either standard reference parameters or actual experimental data.

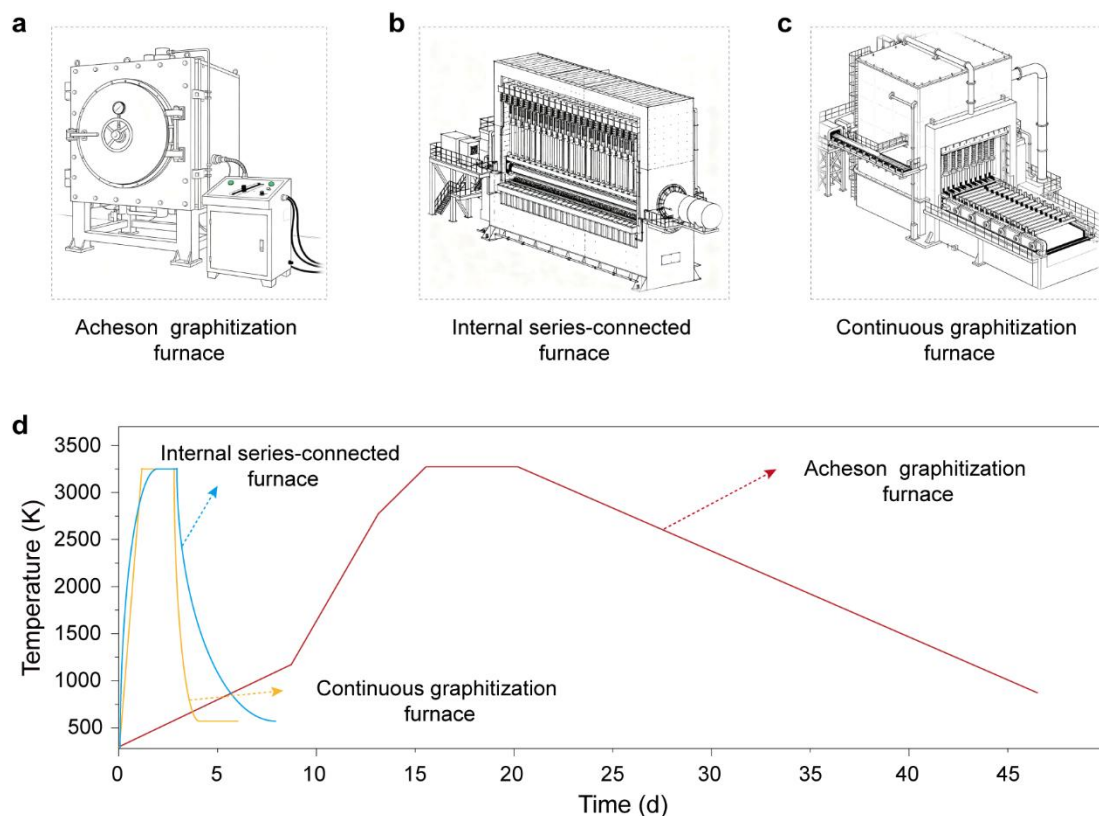
To further clarify the granular-scale mechanism of localized Joule heating, we established a microscopic sub-model. This model consists of two spherical carbon particles with a diameter of 5 μm each. We set their center-to-center spacing at 4.98 μm to form an overlap of 0.02 μm . Such overlap reproduces the partial contact interfaces commonly found in packed powder beds during graphitization. We imposed a potential difference between the two particles. Afterwards, we adopted the electric currents module to calculate the distributions of the electric field and current density. We further derived the volumetric Joule heating value based on Eq. 4 and imported this data into the Heat Transfer in Solids module. This process helps obtain the transient temperature distribution within and near the particle contact region. The established sub-model demonstrates that contact resistance at particle connections leads to temperature gradients and the emergence of potential hot spots.



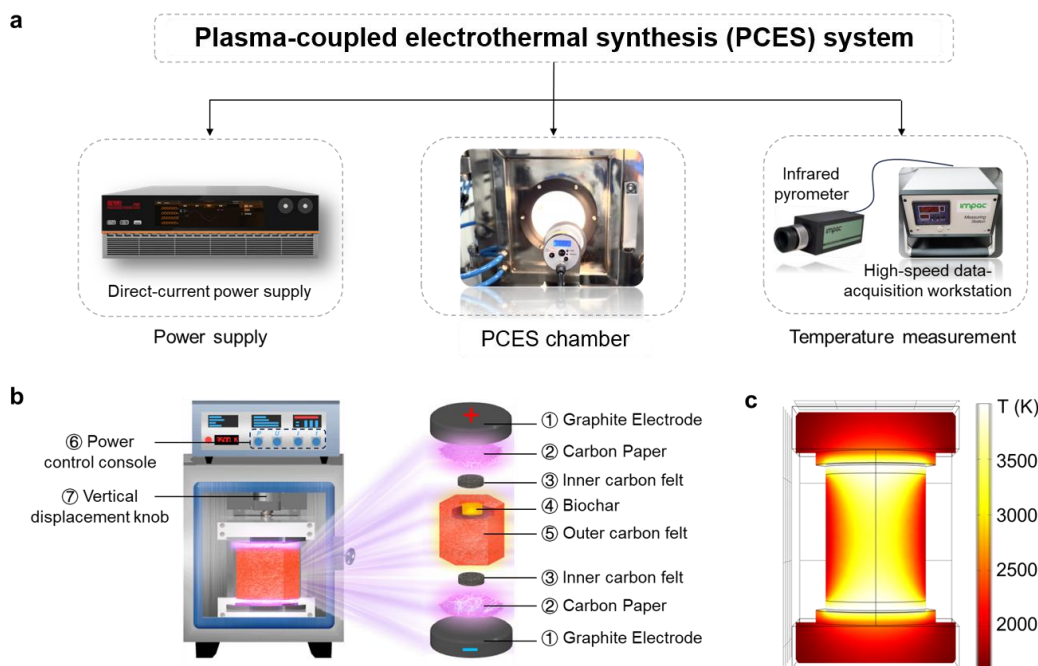
Supplementary Fig. 1 | Construction of the cellulose-derived hard carbon model and ReaxFF MD simulation workflow. (a) Schematic of the hierarchical assembly of the cellulose precursor model. Individual 1,4-β-D-glucopyranose units ($n = 20$) form long cellulose chains ($m = 20$), which are then packed into an amorphous carbon simulation box. (b) Temperature-time profile of the multi-stage simulation procedure, including initial heating (10 K ps^{-1} ramp), carbonization (1300 K isothermal hold), and graphitization (1500–4000 K isothermal treatments). (c) Elemental weight percentage evolution of carbon (red) and oxygen (black) during carbonization, showing that removal of ~80% of hydrogen species drives the carbon content to 87.5% and oxygen to 8.1%.



Supplementary Fig. 2 | Final structures in 1000 ps long ReaxFF MD simulations of hard carbon graphitization at different temperatures. Six-membered carbon rings, the basic structural units of graphite, are highlighted in yellow. Oxygen atoms are marked in red. At 1500 K, low thermal energy traps carbon atoms in initial chain-like defective structures. At 2000–2500 K, partial bond breakage facilitates ring formation while generating metastable topological defects. These defects include twisted rings, oxygen-containing heterocycles, and 5/7-membered carbon rings. At 3000–3500 K, sufficient thermal energy removes all defects and forms continuous sp^2 graphitic domains. At 4000 K, severe thermal vibration breaks C=C bonds, causing structural degradation and reducing the overall degree of graphitization.



Supplementary Fig. 3 | Schematic configurations and heating/cooling profiles of industrial graphitization furnaces. (a–c) Schematic diagrams of (a) Acheson graphitization furnace, (b) internal series-connected furnace, and (c) continuous graphitization furnace. (d) Typical temperature profiles of the Acheson furnace¹⁴, internal series-connected furnace¹⁵, and continuous graphitization furnace (<https://www.vacfurnace.com/continuous-high-temperature-graphite-furnace>) during a typical graphitization process. Traditional industrial graphitization processes typically require prolonged heating and cooling cycles extending over several days to weeks. In stark contrast, the PCES process achieves superior graphitization degrees within seconds, representing a transformative leap in energy efficiency and temporal throughput.



Supplementary Fig. 4 | Plasma-coupled electrothermal ultrahigh-temperature synthesis (PCES) system. (a) Experimental setups of PCES. (b) Left, experimental setup of the PCES system, featuring a plasma-coupled heating chamber with an integrated sample holder. Right, schematic diagram of the PCES device, in which precursor powders were compressed into a pellet and then placed in the middle of the cylindrical carbon felt, with a carbon paper sandwiched between the carbon felt and the graphite electrode. When current is applied, ultra-high-temperature plasma is initially ignited at the microscopic contact points between the short carbon fibers in the felt and the carbon paper surface. This localized plasma, coupled with intense Joule heating, rapidly propagates through the highly porous, three-dimensional network of the carbon felt via thermal conduction, heating the entire carbon felt volume to 3500 K. (c) Simulation of temperature distribution during PCES, showing uniform heat penetration due to localized plasma energy deposition.

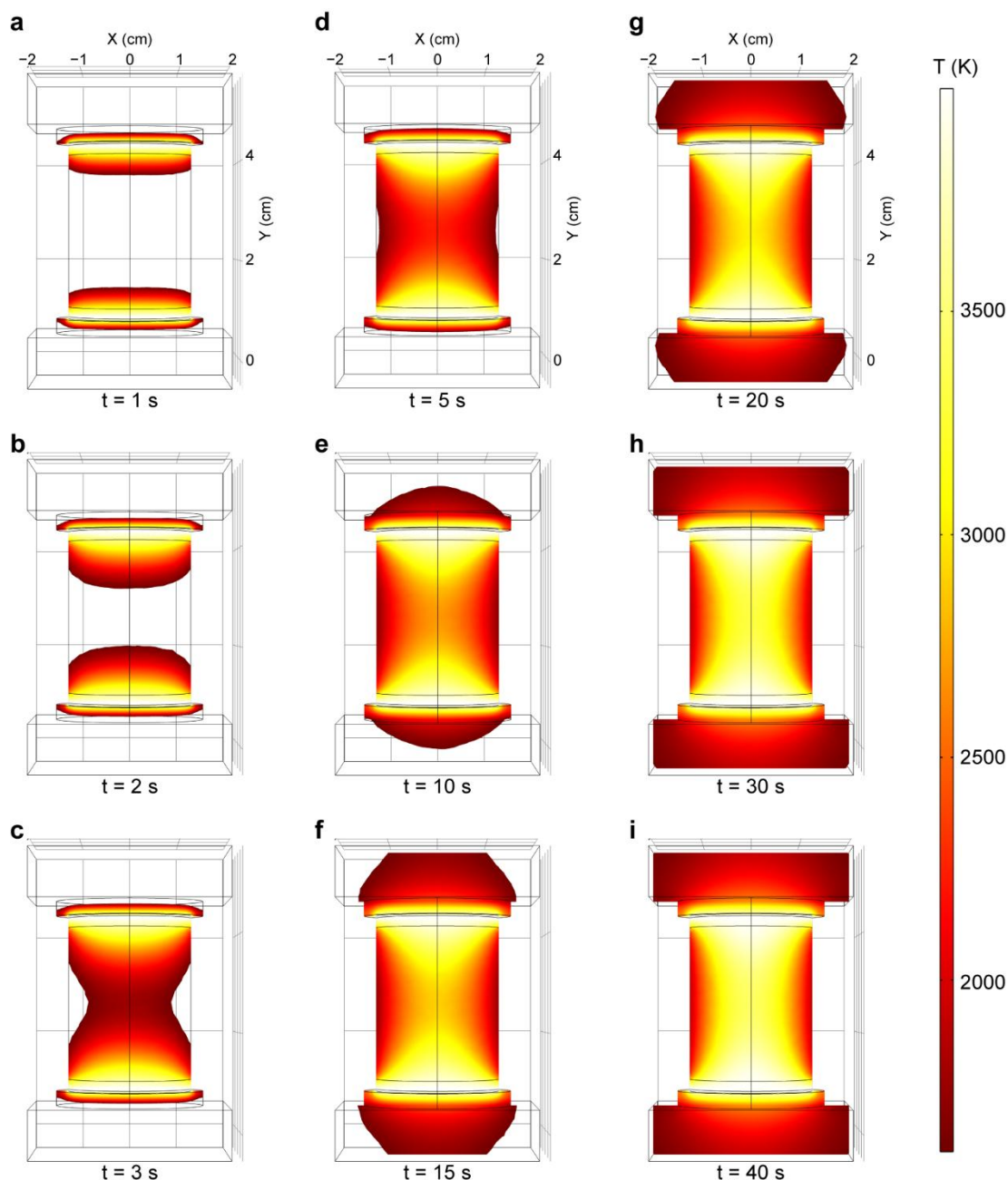
Note: Detailed introduction to the PCES apparatus and operating principle:

The PCES system comprises three core modules: a programmable direct-current (DC) power supply, a sealed reaction chamber, and a high-speed infrared pyrometer for real-time temperature monitoring.

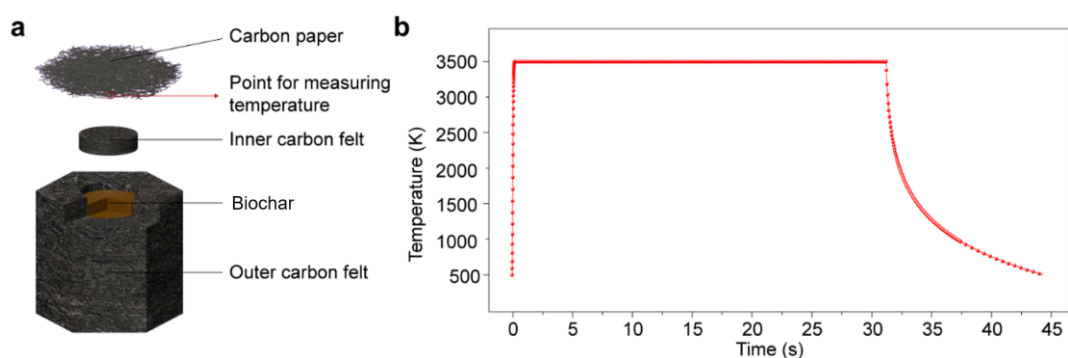
The reaction chamber adopts a coaxial layered design. High-density DC current is applied through graphite electrodes (①) to conductive carbon papers (②), which serve as uniform electrical contact interfaces. The biochar precursor pellet (④) is sandwiched between two inner carbon felts (③), which isolate the sample from carbon papers to prevent contamination while enabling efficient axial heat transfer. An outer carbon felt sleeve (⑤) encloses the entire reaction zone, acting as an effective thermal insulator to minimize radial heat loss. The system is controlled by a power console (⑥) that precisely regulates input current, voltage, and heating duration. A vertical

displacement knob (⑦) adjusts the inter-electrode gap to accommodate different sample sizes and ensure optimal mechanical compression, which minimizes interfacial contact resistance required for reliable plasma generation.

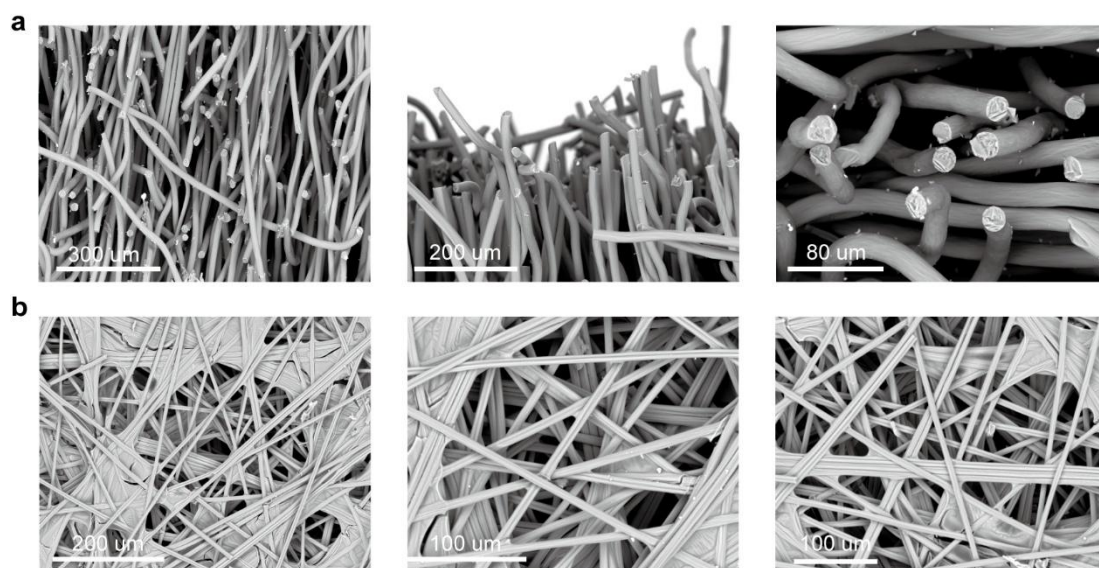
Upon electrification, resistive Joule heating first raises the sample temperature to ~2800 K within 14 seconds. Meanwhile, localized ultrahigh-temperature plasmas form rapidly at the microscopic contact interfaces between carbon papers (②) and carbon felts (③⑤). The synergistic combination of volumetric Joule heating and interfacial plasma heating delivers spatially uniform temperatures exceeding 3500 K throughout the sample within 30 seconds. For operational safety, the chamber is equipped with an integrated water-cooling jacket to protect external components. Customizable gas ports allow processing under controlled inert, reducing, or vacuum atmospheres.



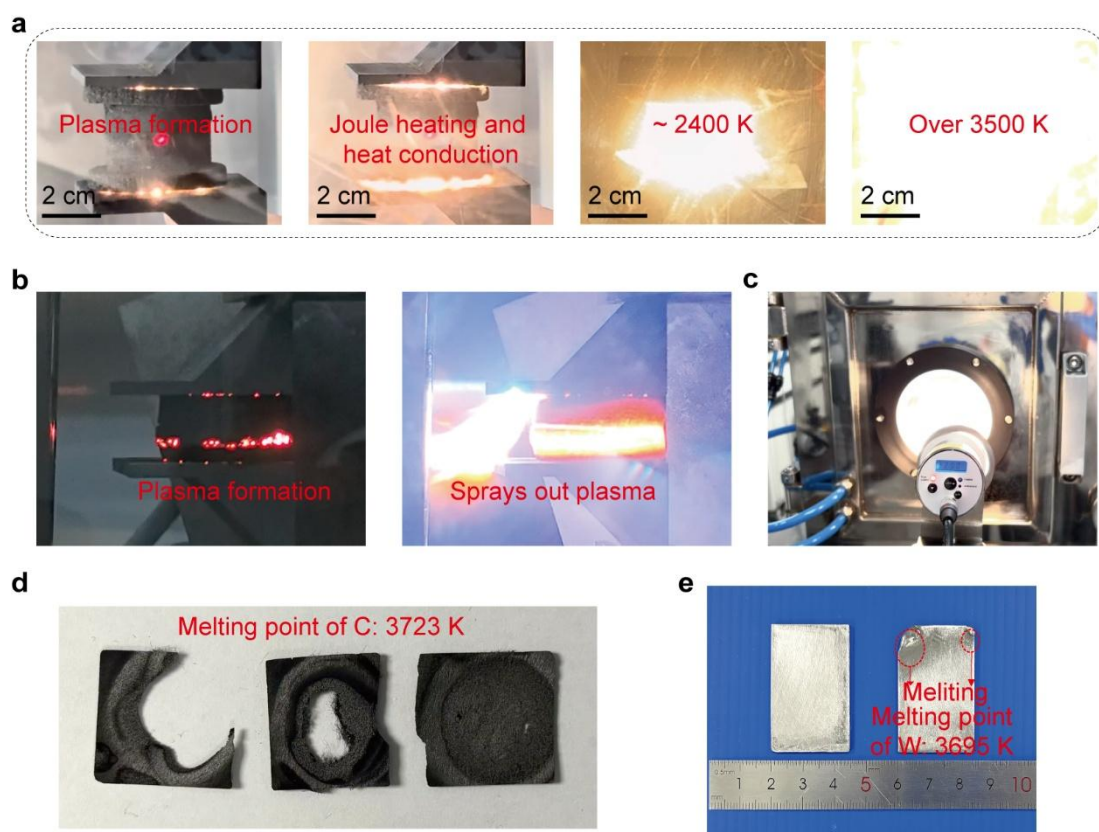
Supplementary Fig. 5 | Finite element temperature field simulations of the PCES device. (a–i) Temporal evolution of the simulated spatial temperature field, demonstrating the characteristic Joule heating ($t = 1\text{--}14\text{ s}$) and plasma-coupled heating ($t > 14\text{ s}$) mechanisms. (a–e) illustrate the rapid initial heating phase dominated by Joule heating, which raises the sample temperature to $\sim 2400\text{ K}$. (f–i) demonstrate the transition following the formation of contact-induced interfacial ultrahigh-temperature plasma, achieving a spatially uniform ultrahigh-temperature environment ($> 3500\text{ K}$). This thermodynamically extreme yet kinetically controlled field minimizes thermal gradients and provides the energy flux necessary to bypass the solid-state carbon diffusion barrier ($\sim 682\text{ kJ mol}^{-1}$), enabling direct, non-equilibrium phase transitions to stable graphite.



Supplementary Fig. 6 | Characterization of the interfacial plasma temperature during PCES. (a) Schematic of the experimental setup, identifying the specific measurement point at the interface between the carbon paper electrode and the carbon fiber felt. (b) Measured temperature-time profile of the plasma region. The results demonstrate the ultrafast thermal response of the PCES system. When electrified, contact-induced plasma causes an immediate temperature spike. It hits the pyrometer's 3500 K measurement limit in just seconds. This localized ultrahigh-temperature state stays stable during electrification. It sustains a steady energy flux far higher than what pure Joule heating can produce.

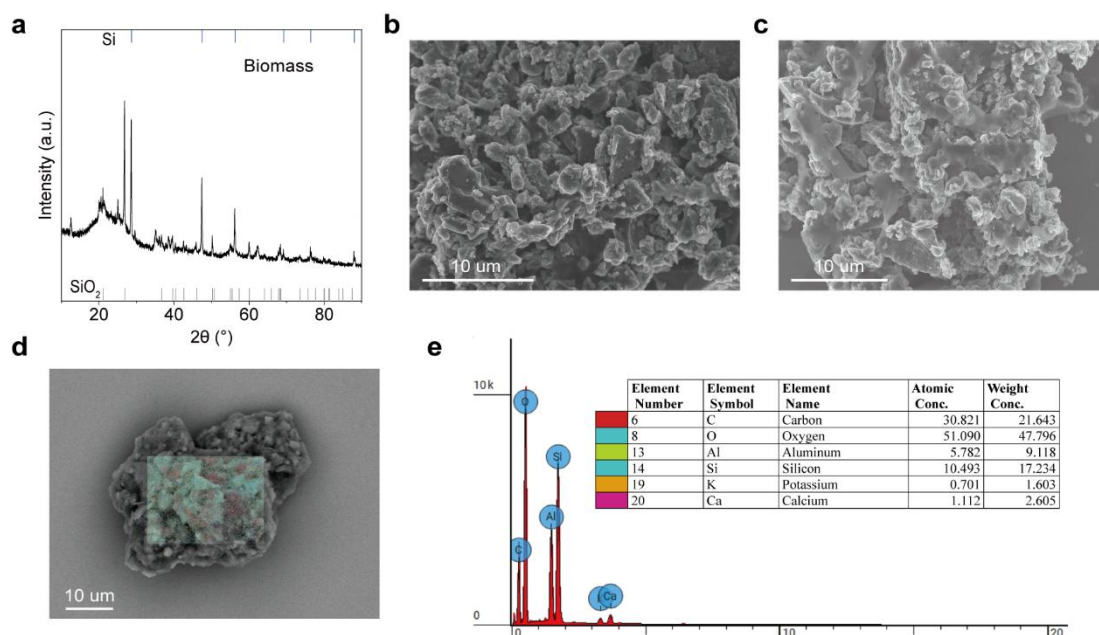


Supplementary Fig. 7 | Microstructural characterization of the PCES heating elements. (a) SEM images of the carbon fiber felt, showing a random, high-porosity network of short carbon fibers. (b) SEM images of the carbon paper surface reveal an integrated fibrous structure with a relatively planar morphology. Randomly arranged fibers inside felt create dense, tiny point contacts when pressed against the carbon paper electrode. These limited contact areas generate high initial contact resistance, which supports localized Joule heating. As current density rises, fiber intersections and contact points between fibers and carbon paper become primary sites of localized electrical breakdown, allowing the generation of interfacial ultrahigh-temperature plasma above 3500 K.

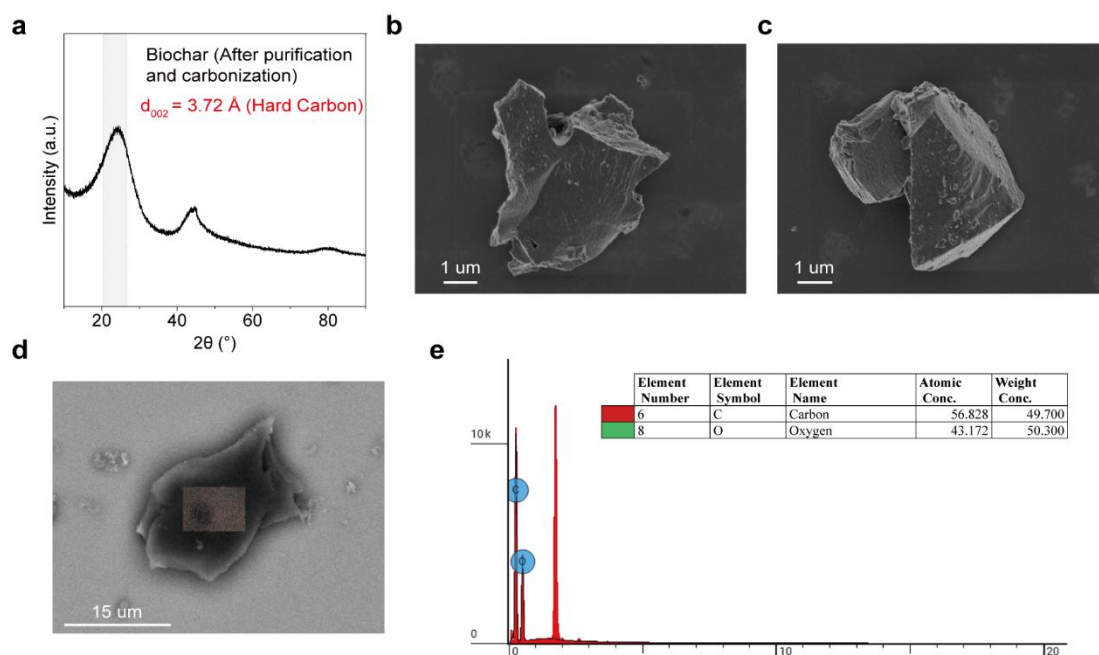


Supplementary Fig. 8 | Experimental visualization and temperature verification of the PCES process. (a) Sequential digital images showing the evolution of the PCES process: from initial plasma formation at contact interfaces to bulk Joule heating and heat conduction, ultimately establishing a stable ultrahigh-temperature state exceeding 3500 K. (b) Photographs reveal that high contact resistance at the fiber/paper interfaces triggers intense arc discharges and plasma spraying. (c) Digital image of the real-time temperature monitoring setup using an infrared pyrometer, confirming peak temperatures up to 3500 K. (d) Photographs of the carbon felt and carbon paper after PCES treatment. The extreme temperatures localized within the plasma region induced significant sublimation of the carbon precursors. (e) Comparison of a pristine tungsten sheet and one after *in-situ* PCES exposure.

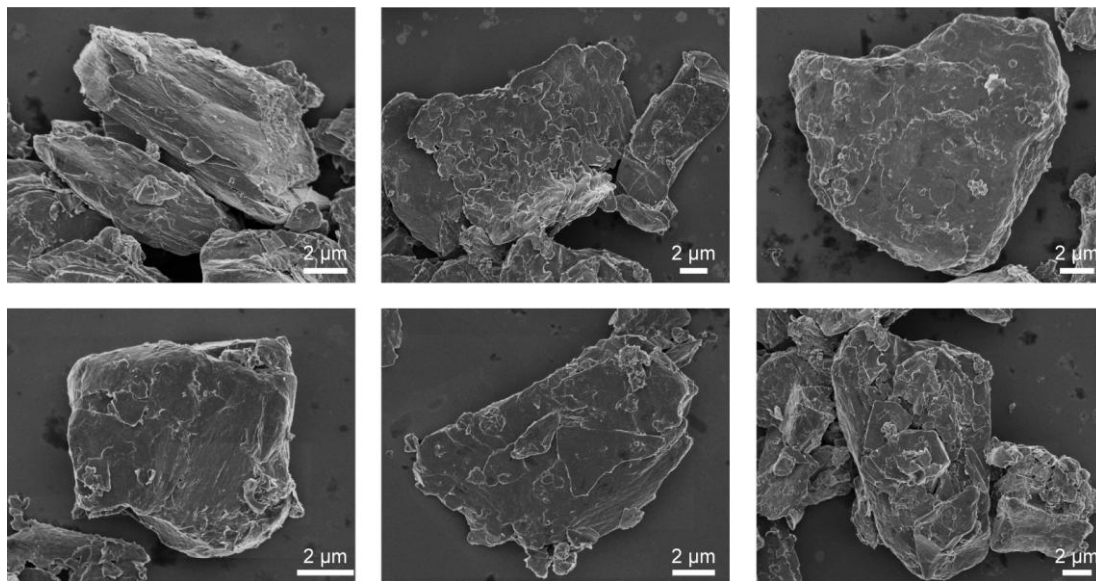
Note: Peak local temperatures exceed 3500 K during PCES was independently validated by the melting of tungsten foil (melting point, 3695 K). The localized melting at the edges of the tungsten sheet serves as a definitive physical benchmark, indicating that local temperatures exceed the melting point of tungsten (3695 K) during PCES.



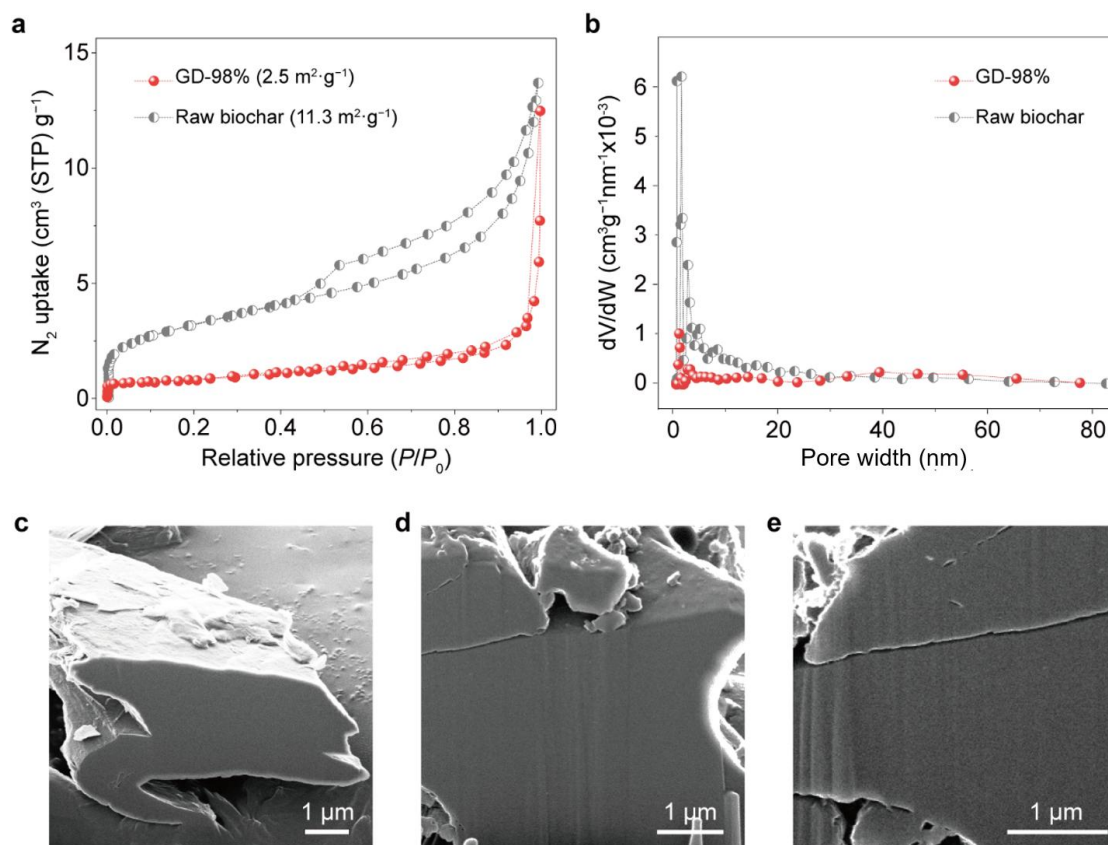
Supplementary Fig. 9 | Characterization of straw biochar. (a) XRD patterns of straw indicate that the straw contains a large amount of silicon and silica impurities. (b-c) SEM images reveal the inherent porous and heterogeneous microstructure of the untreated straw biochar. (d-e) SEM-EDS elemental mapping and the corresponding energy-dispersive X-ray spectrum, confirming the high concentration of Si, O, Al, K, and Ca within the straw framework.



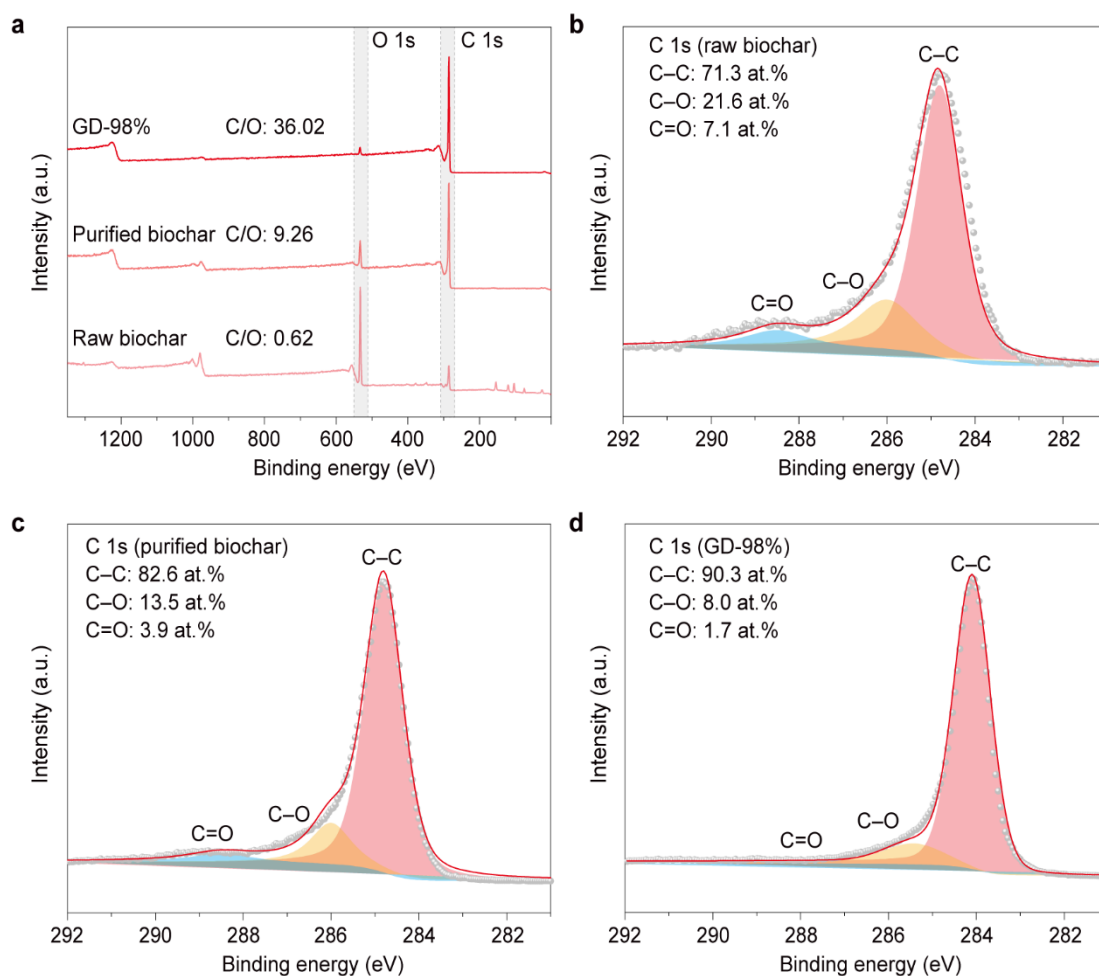
Supplementary Fig. 10 | Characterization of purified biochar. (a) XRD patterns of straw after HF purification and carbonization, showing the complete disappearance of silicon and silica signals. The interlayer distance of straw is greater than 0.37 nm, which proves that the precursor belongs to hard carbon materials. (b-c) SEM images illustrating the morphology of the purified biochar particles. (d-e) SEM-EDS elemental mapping and the corresponding spectrum, showing a high-purity framework composed exclusively of carbon and oxygen.



Supplementary Fig. 11 | SEM imaging of the PCES-synthesized highly graphitized GD-98% powder.

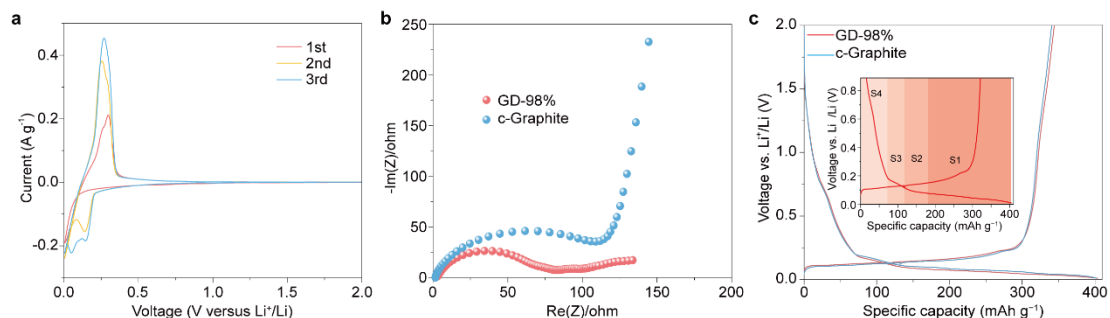


Supplementary Fig. 12 | Pore structure characterization and cross-sectional morphology of raw biochar and graphitized samples. (a) N_2 adsorption and desorption data of raw biochar and GD-98% sample. **(b)** Pore size distribution curves of raw biochar and GD-98% sample. **(c-e)** Cross-sectional SEM imaging of the PCES-derived graphite via FIB-SEM serial sectioning.



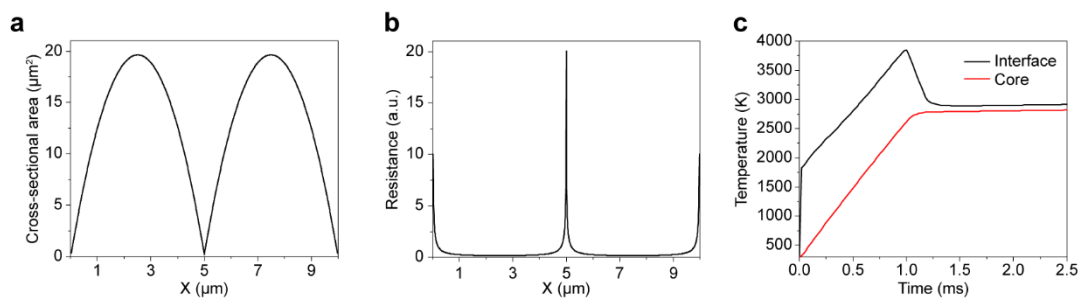
Supplementary Fig. 13 | Evolution of surface chemical composition and carbon bonding states. (a) XPS survey spectra of raw biochar, purified biochar, and GD-98% sample, illustrating the progressive increase in C/O atomic ratios from 0.62 to 36.02. (b-d) High-resolution C 1s spectra with peak fitting for (b) raw biochar, (c) purified biochar, and (d) GD-98% graphite. The deconvoluted peaks at ~284.8 eV, ~286.0 eV, and ~288.5 eV correspond to C–C, C–O, and C=O bonds, respectively.

Note: XPS analysis offers quantitative evidence for gradual carbon enrichment and thorough deoxidation throughout the PCES process. The results demonstrate that the PCES process effectively eliminates oxygen-containing functional groups. The dramatic increase in the C/O ratio and the C–C component (90.3 at.%) provides solid chemical evidence for the near-complete graphitization of the biomass precursor.



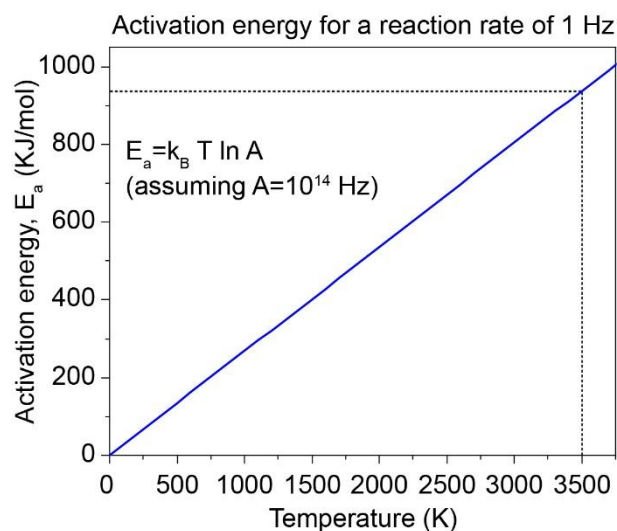
Supplementary Fig. 14 | Electrochemical performance of GD-98% and commercial graphite (c-Graphite) in a lithium-ion battery. (a) CV curves of the GD-98% electrode for the initial three cycles. **(b)** Nyquist plots obtained from EIS, highlighting the reduced charge-transfer resistance of GD-98% (red) compared to c-Graphite (blue). **(c)** Initial charging and discharging profiles of GD-98% (red) and c-Graphite (blue) at 0.05 C from 0.01V to 2V. Inset: Enlarged view of the low-voltage region. GD-98% electrodes show similar lithiation/delithiation plateaus (0.05 V and 0.15 V).

Note: It is worth noting that these evaluations were carried out using a standard additive-free electrolyte, which is 1 M LiPF₆ dissolved in EC/DMC/EMC. This test configuration enables a more accurate comparison of the intrinsic material properties.



Supplementary Fig. 15 | Localized contact resistance and ultrafast interfacial heating mechanism via PCES. (a) Geometric profile showing the cross-sectional area along the X-axis of two spherical particles in contact. (b) Corresponding resistance distribution calculated based on the geometric profile. (c) Simulated temperature evolution during the initial 2.5 ms of the PCES process.

Note: The simulation results elucidate the micro-scale origin of the ultrahigh-temperature field. Due to the constriction resistance effect, the current density is extremely concentrated at the infinitesimal contact interfaces between the carbon fibers. This localized high resistance triggers a nearly instantaneous temperature surge (up to 3800 K within 1 ms) at the interface, far outpacing the heating rate of the bulk core.

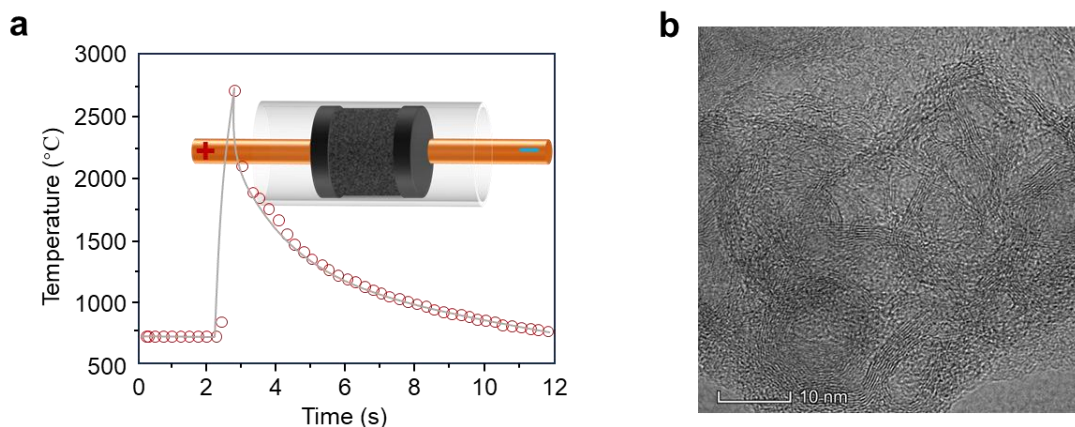


Supplementary Fig. 16 | Activation energy analysis for PCES graphitization.

Note: From the rearranged Arrhenius relation:

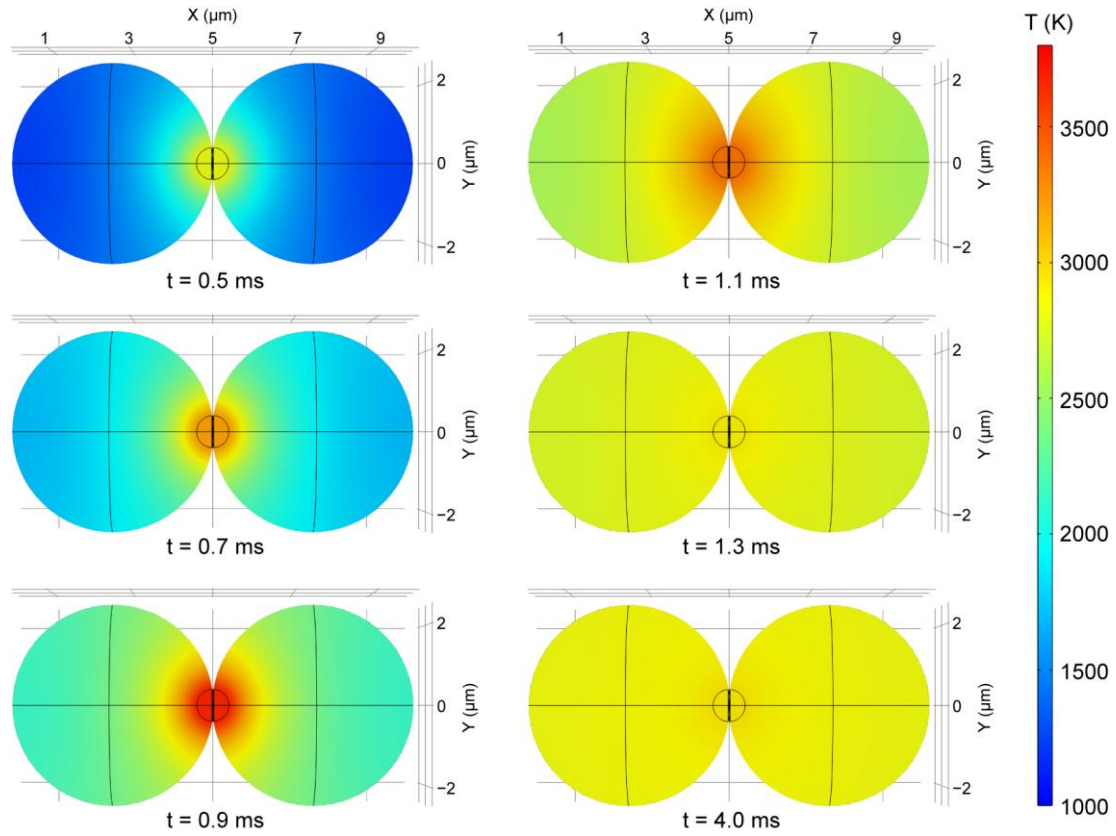
$$E_a = k_B T \ln[A/f] \quad (5)$$

At 3500 K, assuming $A \sim 10^{14}$ Hz and a reaction time of ~ 1 s¹⁶, we estimate an apparent activation energy of $E_a \approx 938$ kJ mol⁻¹. This estimate illustrates how the ultrahigh-temperature environment in PCES breaks down diffusion barriers, thereby allowing various carbon precursors to rapidly convert into highly graphitic graphite.

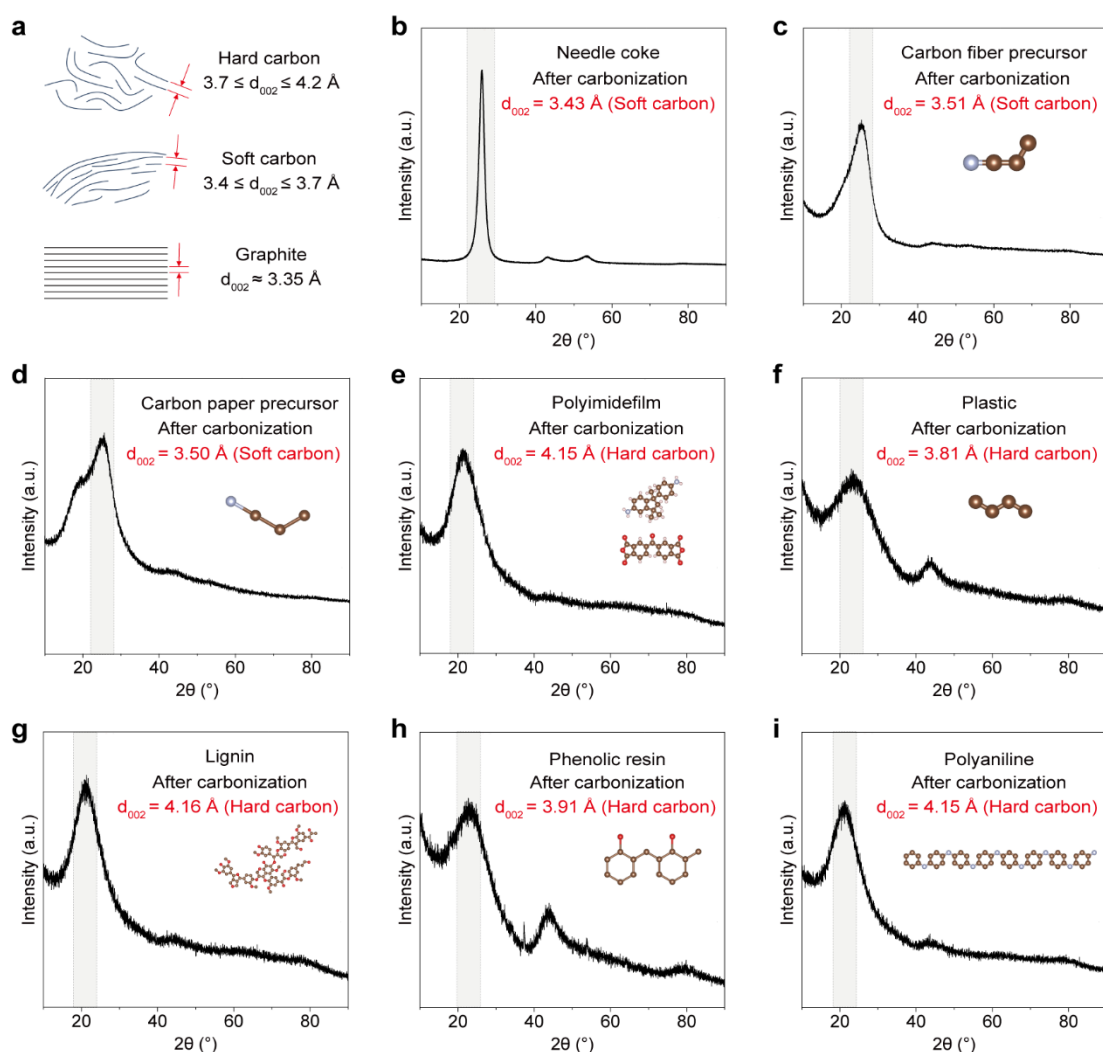


Supplementary Fig. 17 | Synthesis and characterization of turbostratic graphene via flash Joule heating (FJH). (a) Schematic of the FJH reaction cell and corresponding temperature-time profile, showing ultrafast heating to ~2700 °C (~3000 K) within 1 s followed by rapid quenching to room temperature. (b) High-resolution transmission electron microscopy image of FJH-synthesized turbostratic graphene, showing the characteristic turbostratic stacking structure with randomly rotated graphene layers.

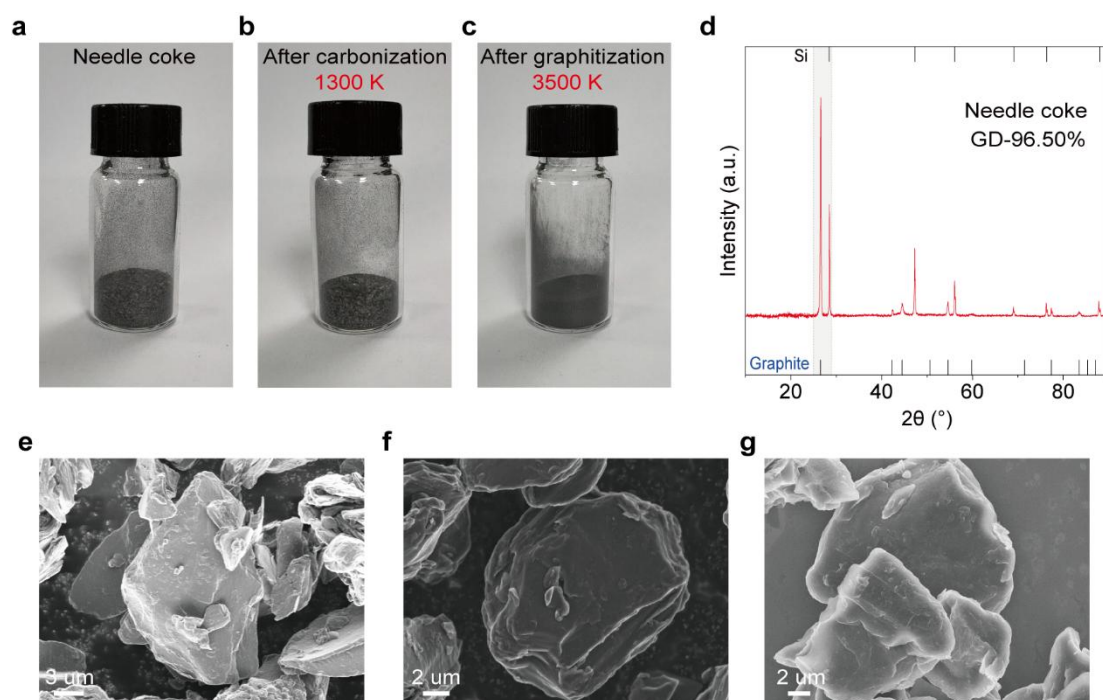
Note: Turbostratic graphene formation in FJH arises from extreme non-equilibrium thermal conditions, specifically ultrafast heating and quenching rates ($\sim 10^3 \text{ K s}^{-1}$), which are orders of magnitude higher than those of conventional furnace heating (typically $< 1 \text{ K s}^{-1}$). As demonstrated in previous studies^{17,18}, rapid vaporization-condensation of carbon species at ultrahigh temperatures generates isolated graphene nuclei, and subsequent fast cooling prevents these nuclei from rearranging into thermodynamically stable Bernal (AB) stacked graphite. Instead, the graphene layers retain random rotational orientations relative to one another, forming a turbostratic structure.



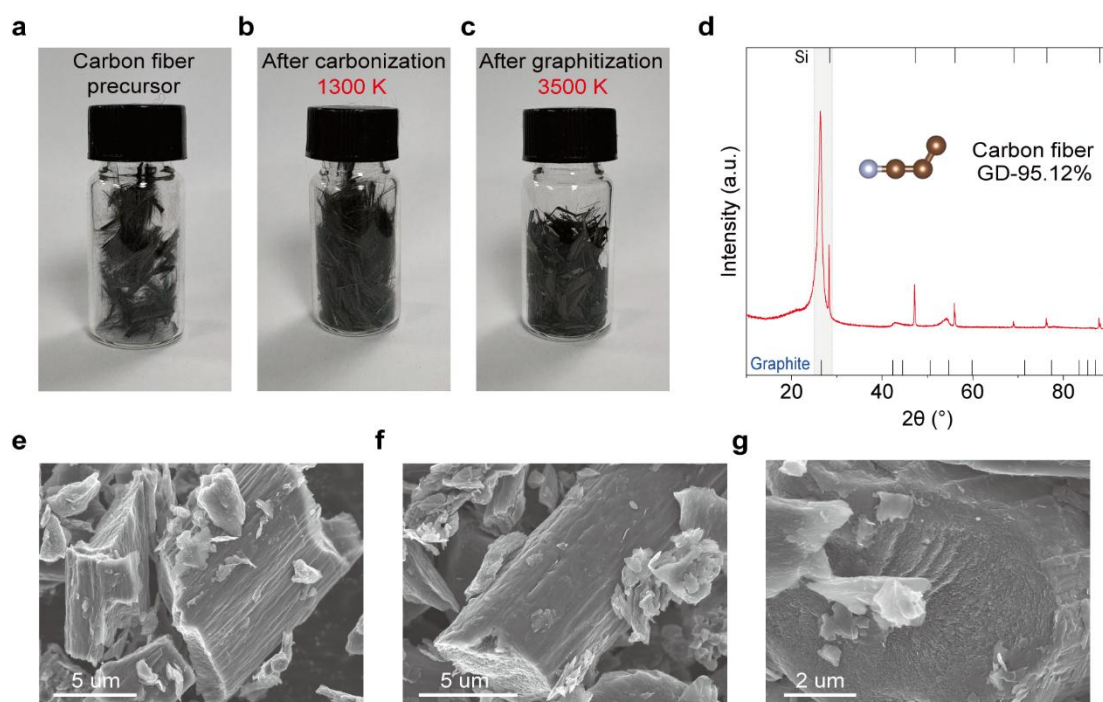
Supplementary Fig. 18 | Spatiotemporal evolution of the temperature field during the initial stage of PCES. COMSOL Multiphysics simulation snapshots show the temperature distribution across two particles in contact from $t = 0.5$ ms to 4.0 ms. At $t = 0.9$ ms, the interfacial temperature rises sharply to nearly 3500 K in the red zone. Meanwhile, bulk regions stay much cooler below 2500 K in the blue zones. By $t = 4.0$ ms, the temperature field grows more uniform. The system gradually approaches a quasi-equilibrium state with a stable temperature.



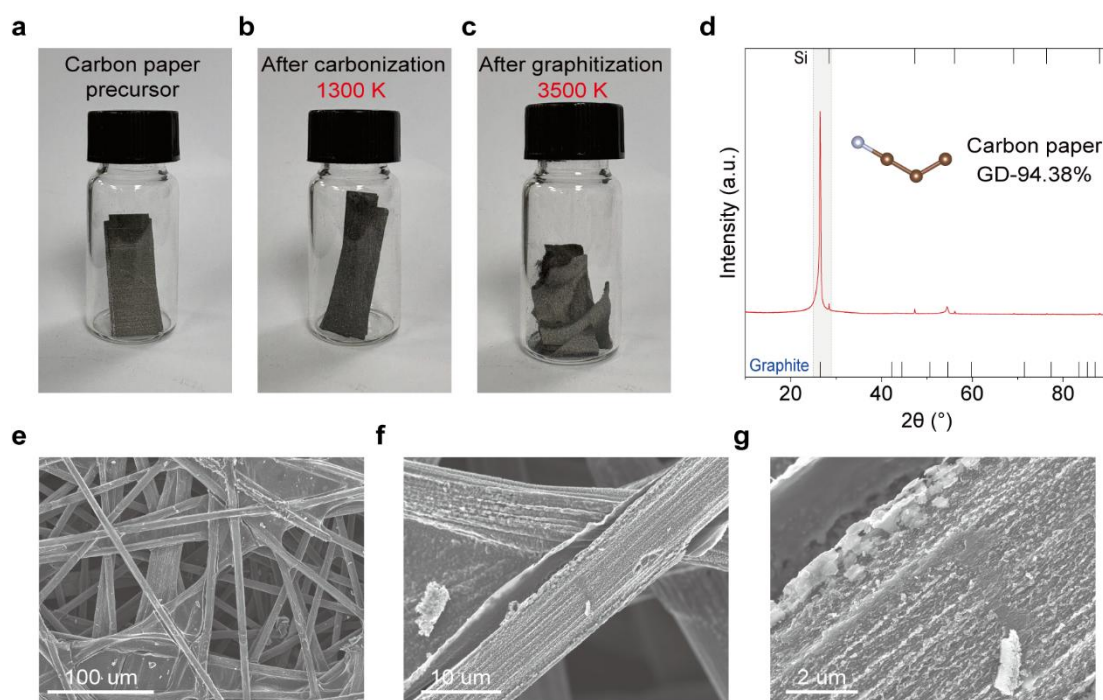
Supplementary Fig. 19 | Universality of the PCES method. (a) Schematic illustrations of hard carbon, soft carbon, and graphite. Specifically, hard carbons feature short-range graphitized domains, expanded interlayer distance ($0.37\text{--}0.42 \text{ nm}$)¹⁹. (b-i) XRD patterns of needle coke, carbon fiber, carbon paper, polyimide film, plastic, lignin, phenolic resin, and polyaniline after being carbonized at 1300 K. The broad diffraction peaks around $22\text{--}26^\circ$ are assigned to lattice planes of (002). The interlayer distance of needle coke, carbon fiber, and carbon paper is between 0.34 and 0.37 nm ($3.4 \text{ \AA} \leq d_{002} \leq 3.7 \text{ \AA}$), which proves that the precursors belong to soft carbon materials. The interlayer distance of polyimide film, plastic, lignin, phenolic resin, and polyaniline is all greater than 0.37 nm ($d_{002} > 3.7 \text{ \AA}$), which proves the precursors belong to hard carbon materials.



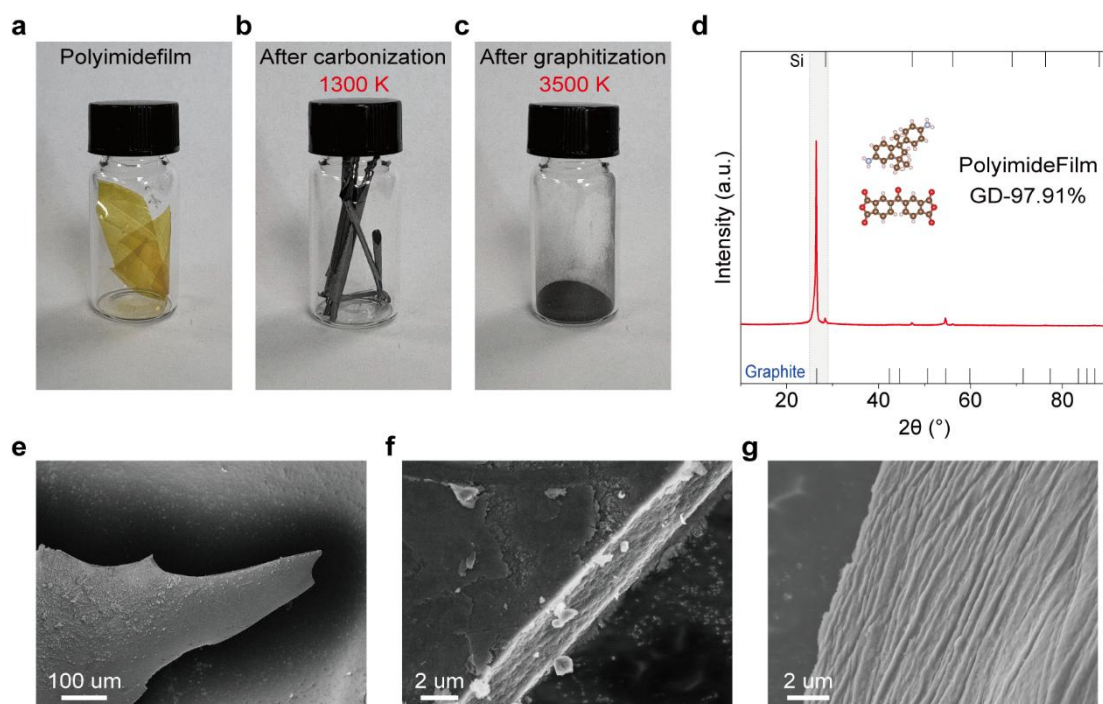
Supplementary Fig. 20 | Graphitization of needle coke. Photographs of (a) needle coke, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of needle coke after being graphitized at 3500 K. (e-g) SEM images of needle after being graphitized at 3500 K. The sharp and intense (002) diffraction peak confirms the successful transition to a highly ordered graphitic structure. The calculated degree of graphitization reaches a high value of 96.50%. Silicon was used as an internal standard for peak calibration.



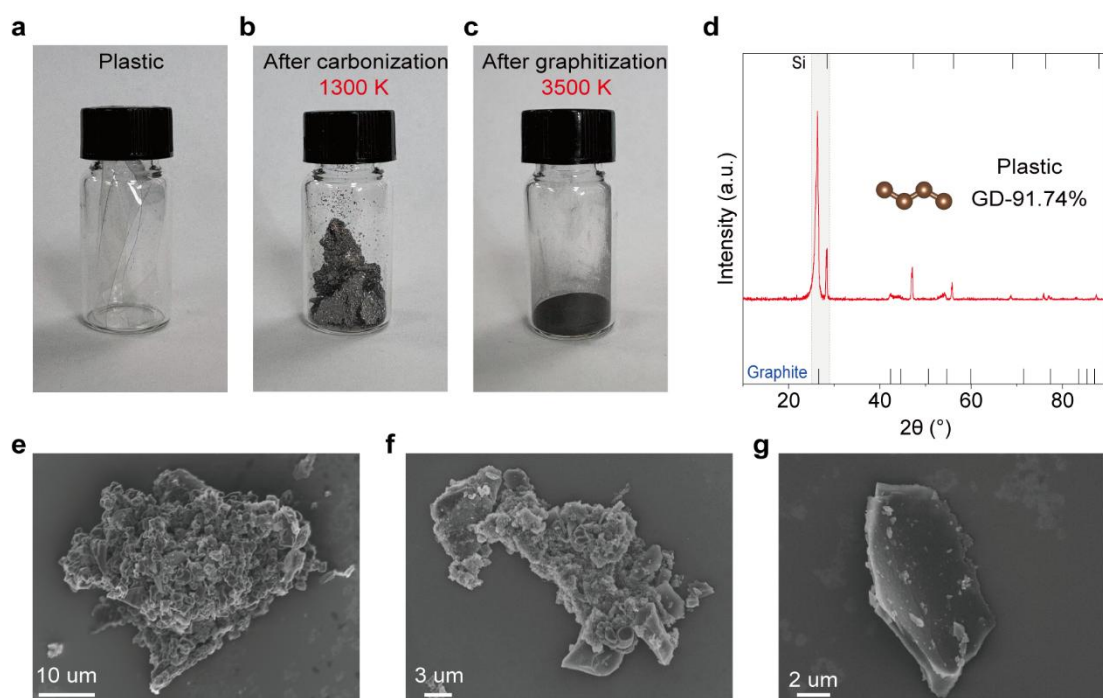
Supplementary Fig. 21 | Graphitization of carbon fiber precursor. Photographs of (a) carbon fiber precursor, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of carbon fiber precursor after being graphitized at 3500 K. (e-g) SEM images of carbon fiber precursor after being graphitized at 3500 K. The sharp and intense (002) diffraction peak confirms the successful transition to a highly ordered graphitic structure. The calculated degree of graphitization reaches a high value of 95.12%. Silicon was used as an internal standard for peak calibration.



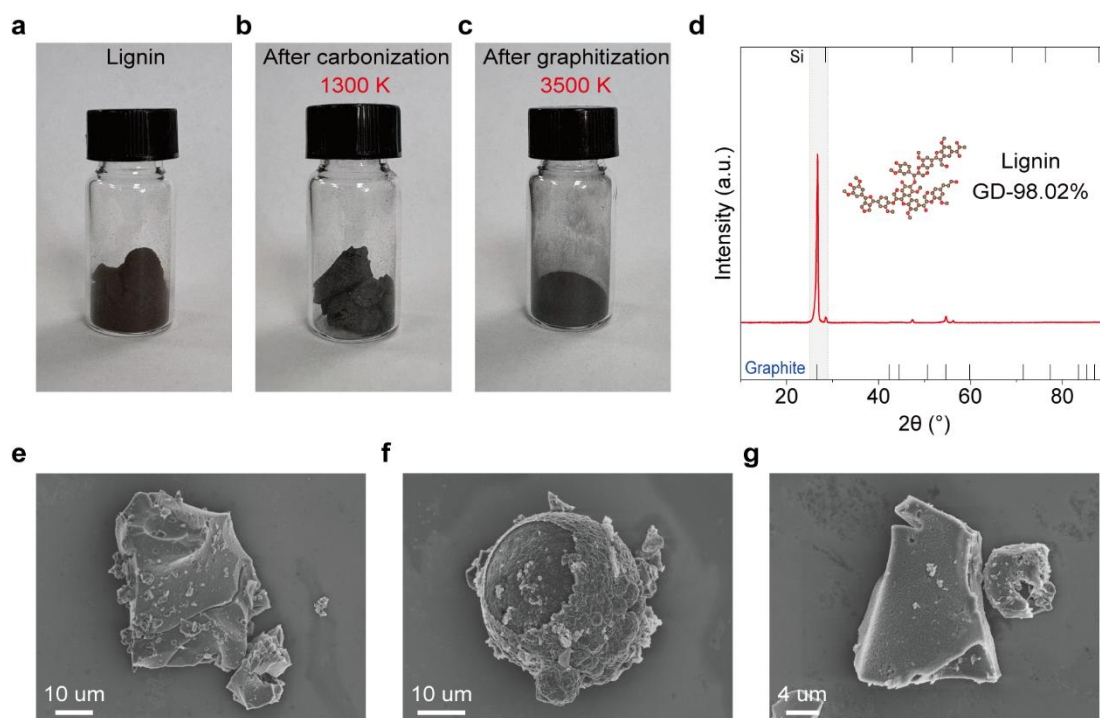
Supplementary Fig. 22 | Graphitization of carbon paper precursor. Photographs of (a) carbon paper precursor, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of carbon paper precursor after being graphitized at 3500 K. (e-g) SEM images of carbon paper precursor after being graphitized at 3500 K. The sharp and intense (002) diffraction peak confirms the successful transition to a highly ordered graphitic structure. The calculated degree of graphitization reaches a high value of 94.38%. Silicon was used as an internal standard for peak calibration.



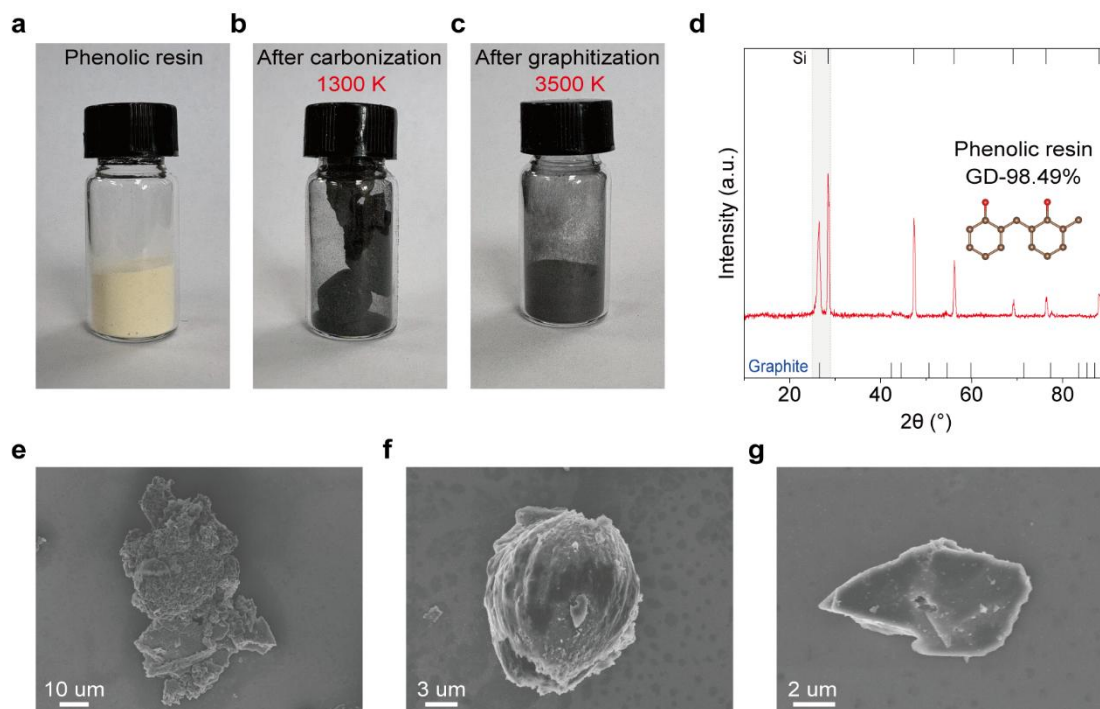
Supplementary Fig. 23 | Graphitization of polyimide film. Photographs of (a) polyimide film, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of polyimide film after being graphitized at 3500 K. (e-g) SEM images of polyimide film after being graphitized at 3500 K. The distinct and strong (002) diffraction peak verifies the successful formation of a highly ordered graphitic structure. The calculated graphitization degree is as high as 97.91%. Silicon serves as the internal standard for peak calibration.



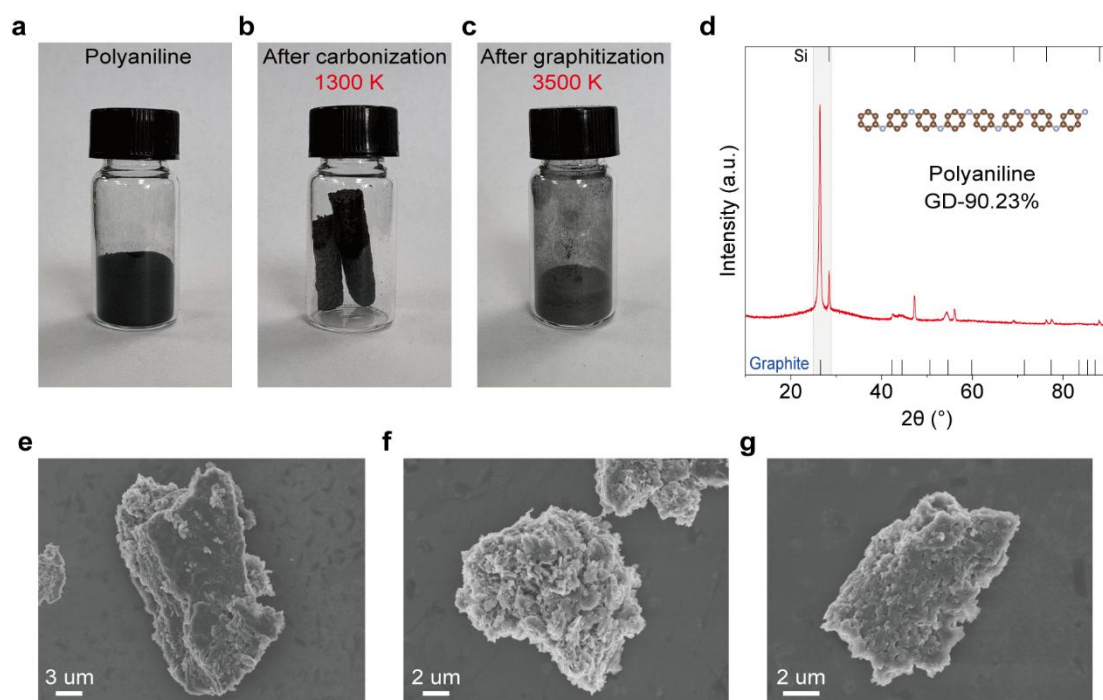
Supplementary Fig. 24 | Graphitization of plastic. Photographs of (a) plastic, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of plastic after being graphitized at 3500 K. (e-g) SEM images of plastic after being graphitized at 3500 K. The sharp and intense (002) diffraction peak confirms the successful transition to a highly ordered graphitic structure. The calculated degree of graphitization reaches a high value of 91.74%. Silicon was used as an internal standard for peak calibration.



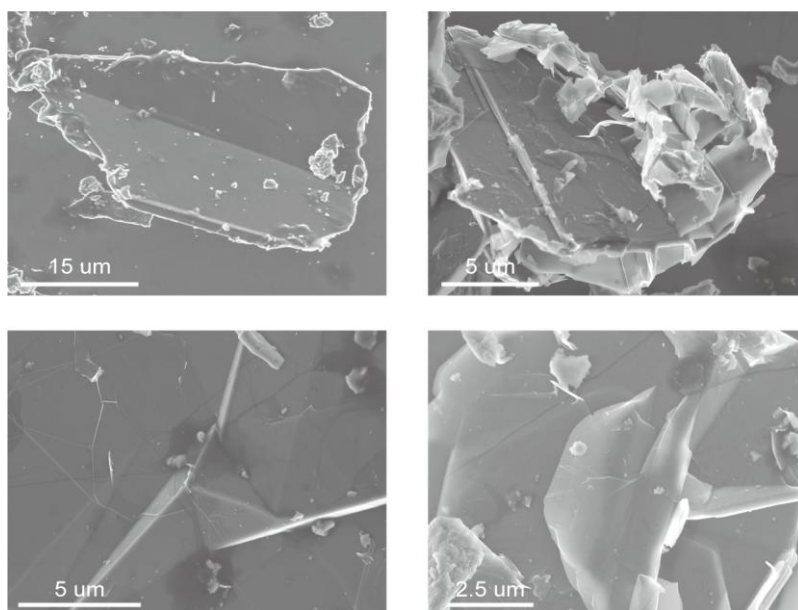
Supplementary Fig. 25 | Graphitization of lignin. Photographs of (a) lignin, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of lignin after being graphitized at 3500 K. (e-g) SEM images of lignin after being graphitized at 3500 K. The sharp and intense (002) diffraction peak confirms the successful transition to a highly ordered graphitic structure. The calculated degree of graphitization reaches a high value of 98.02%. Silicon was used as an internal standard for peak calibration.



Supplementary Fig. 26 | Graphitization of phenolic resin. Photographs of (a) phenolic resin, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of phenolic resin after being graphitized at 3500 K. (e-g) SEM images of phenolic resin after being graphitized at 3500 K. The sharp and intense (002) diffraction peak confirms the successful transition to a highly ordered graphitic structure. The calculated degree of graphitization reaches a high value of 98.49%. Silicon was used as an internal standard for peak calibration.

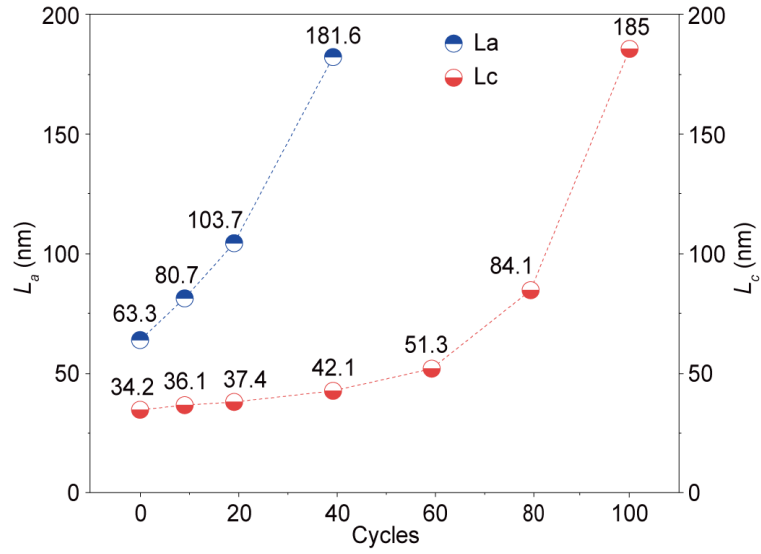


Supplementary Fig. 27 | Graphitization of polyaniline. Photographs of (a) polyaniline, (b) precursor after carbonization at 1300 K, and (c) product after graphitization via PCES at 3500 K. (d) XRD pattern of polyaniline after being graphitized at 3500 K. (e-g) SEM images of polyaniline after being graphitized at 3500 K. The sharp and intense (002) diffraction peak confirms the successful transition to a highly ordered graphitic structure. The calculated degree of graphitization reaches a high value of 90.23%. Silicon was used as an internal standard for peak calibration.



Supplementary Fig. 28 | Microstructural characterization of HOPG synthesized via cycling PCES treatment.

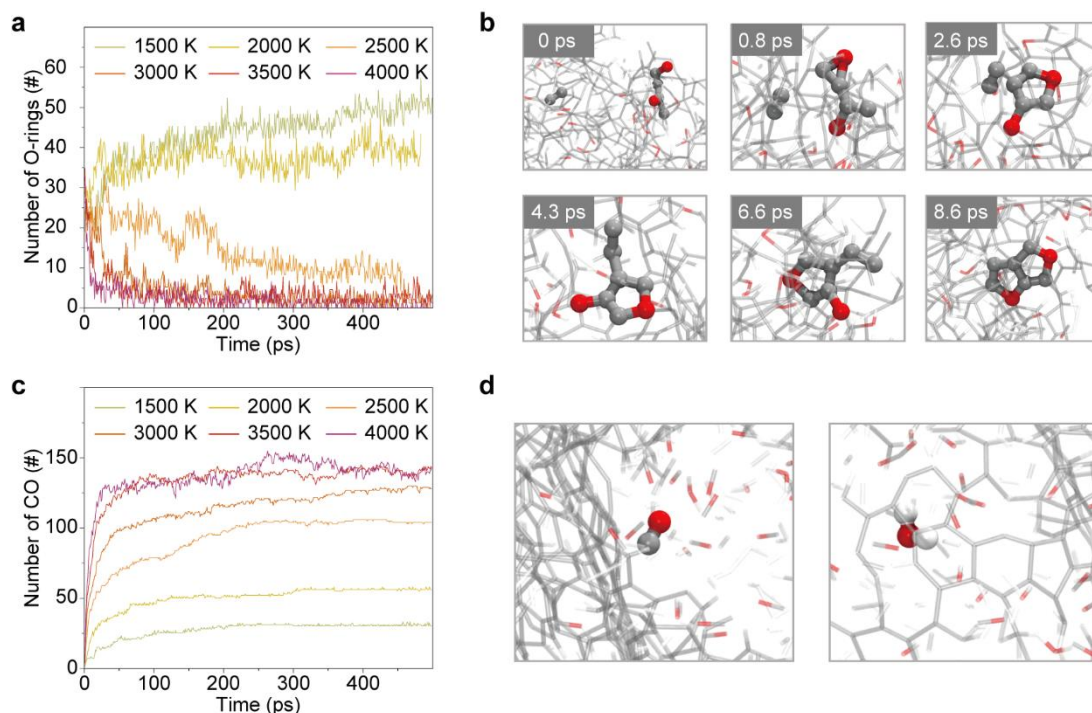
Note: The synthesis of HOPG via PCES represents a substantial improvement in manufacturing efficiency compared to traditional chemical vapor deposition or high-pressure annealing methods. Conventional HOPG synthesis is notoriously energy-intensive and slow, typically requiring sustained temperatures (2500-3000 °C) and high pressures (30 MPa) for over 24 hours with a low deposition rate of $\sim 0.1 \mu\text{m}/\text{h}^{20}$. In stark contrast, our PCES-based cyclic strategy utilizes contact-induced interfacial plasma to reach a peak temperature of 3500 K within mere 30 seconds. By repeating this ultrafast heating/cooling cycle, there is a complete transition from graphite to a highly oriented state after 60 cycles. SEM images of the HOPG powder at various magnifications, revealing a signature highly ordered, layered architecture with significant grain growth and sharp, well-defined terrace edges²¹.



Supplementary Fig. 29 | Evolution of in-plane grain size (L_a) and out-of-plane stacking height (L_c) as a function of thermal cycles. The specific calculation formula of L_a and L_c was calculated using the Scherrer equation:

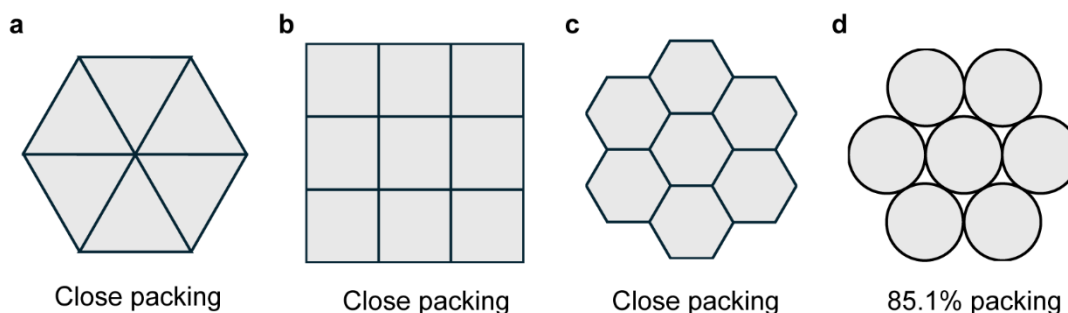
$$L \text{ (nm)} = \frac{K\lambda}{\beta \cos \theta} \quad (6)$$

Note: λ represents the wavelength of the incident X-ray, β represents the half-peak width of the diffraction peak, and θ represents the diffraction angle. For L_a calculations, the (100) reflection at $2\theta \approx 42^\circ$ was utilized. For L_c calculations, the primary (002) reflection at $2\theta \approx 26^\circ$ was utilized. $K = 1.84$ was applied to the (100) peak to reflect the lateral extent of the graphene layers. $K = 0.89$ was applied to the (002) peak to represent the mean thickness of the graphitic crystal stacks. Both L_a and L_c exhibit a monotonic upward trend, signaling the progressive transformation of graphite into HOPG under cycling PCES treatment²². After 60 thermal cycles, the (100) peak is significantly weaker than the dominant (002) peak, as the material becomes highly oriented, these in-plane peaks can become extremely difficult to resolve or fit accurately once they narrow beyond the instrumental threshold.



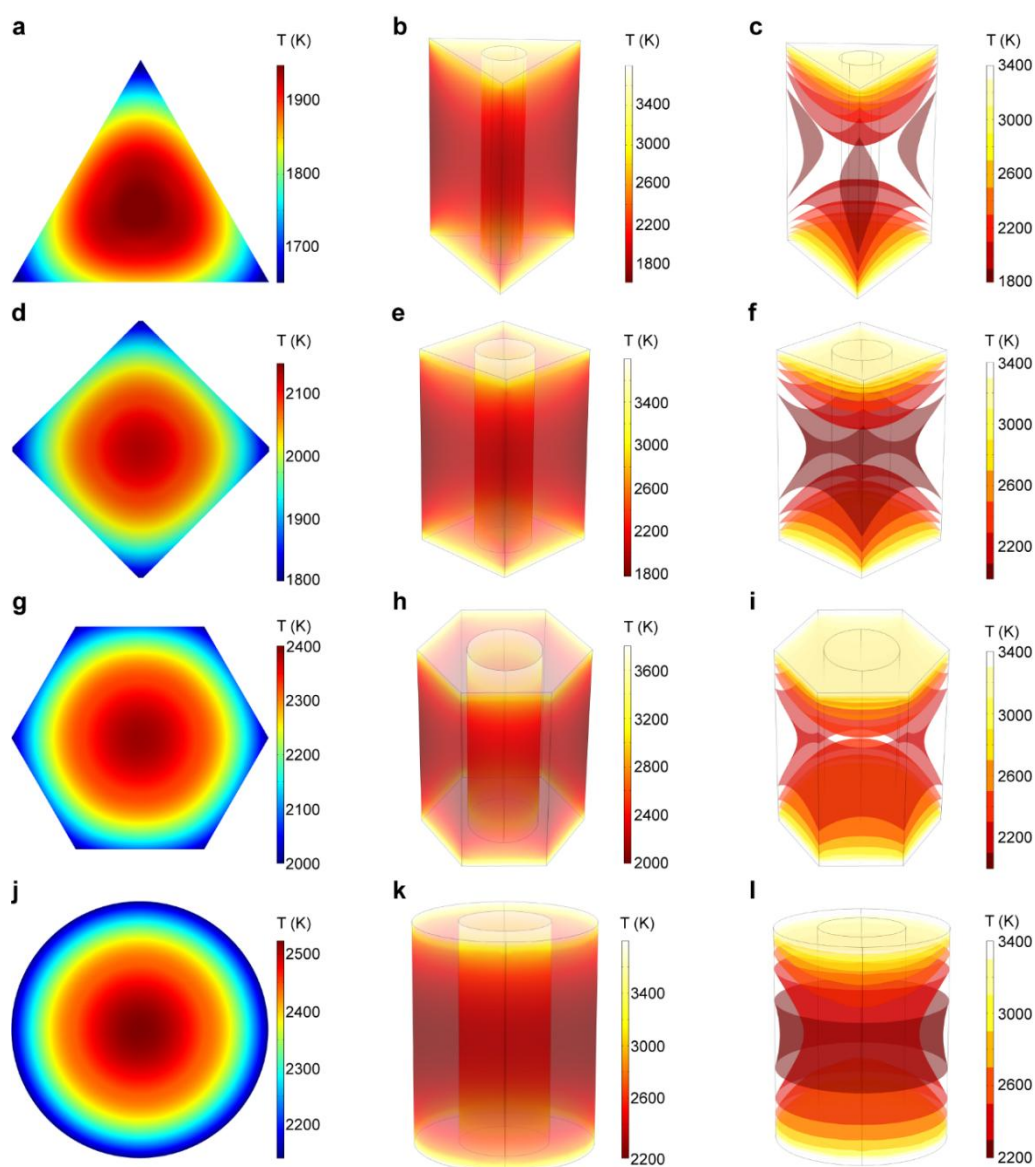
Supplementary Fig. 30 | Role of oxygen heteroatoms in hard carbon graphitization (ReaxFF MD simulations). (a) Temporal evolution of oxygen-containing ring structures at different temperatures, showing a sharp decrease in ring count above 2500 K. (b) Representative MD snapshots of the formation process of an oxygen-containing five-membered ring at 2000 K (0–8.6 ps). (c) Temperature-dependent evolution of CO molecule formation, showing a strong positive correlation between temperature and CO yield. (d) Representative snapshots of volatile CO and H₂O species generated during graphitization at 3500 K.

Note: Oxygen heteroatoms exhibit a dual role in hard carbon graphitization. At temperatures below 2500 K, oxygen atoms incorporate into the carbon network to form metastable oxygen-containing five-membered rings, which act as topological defects that block long-range graphitic ordering. Above 3000 K, these metastable heterocycles become thermodynamically unstable and decompose, releasing oxygen as volatile CO and H₂O species. This defect elimination process frees carbon atoms to reorganize into stable six-membered rings, significantly accelerating graphitization kinetics. Notably, the ultrahigh temperatures achieved by PCES are essential for driving this complete oxygen removal and defect healing, which cannot be efficiently accomplished by conventional furnace heating.



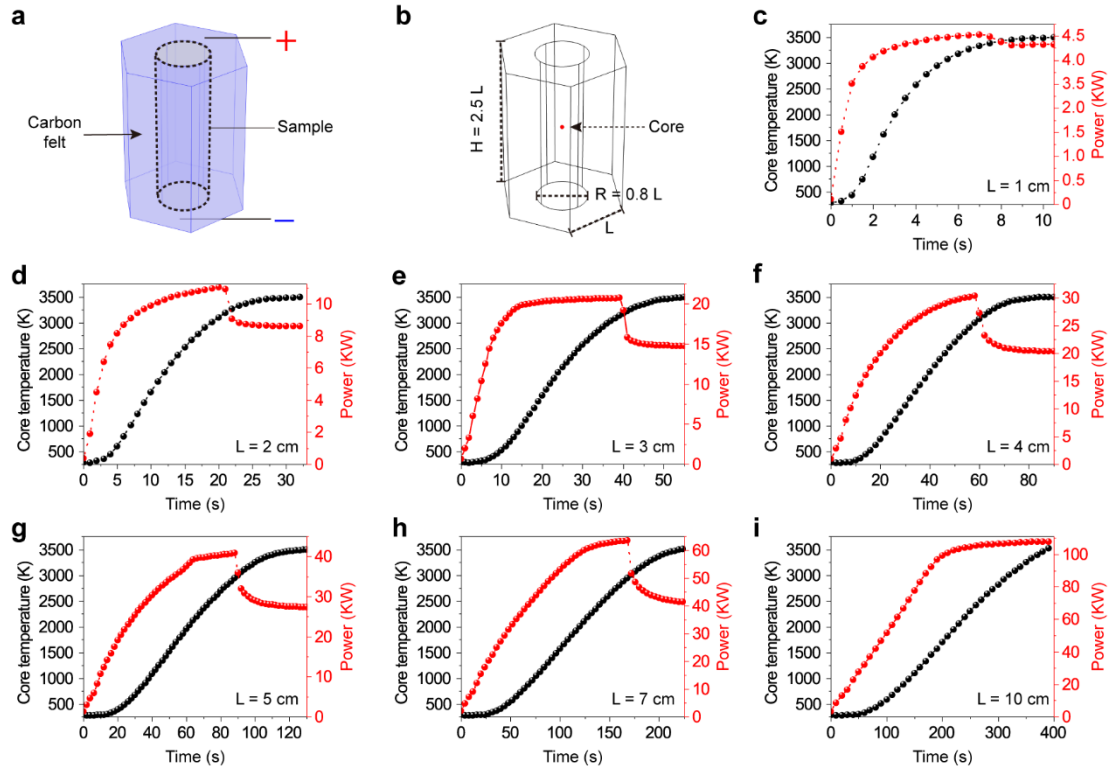
Supplementary Fig. 31 | Comparison of the packing efficiency of different geometric shapes. (a) Triangular prism, close packing. (b) Quadrangular prism, close packing. (c) Hexagonal prism, close packing. (d) Cylinder, 85.1% packing efficiency.

Note: The geometric design of heating elements (triangular prism, quadrangular prism, hexagonal prism, and cylinder) is evaluated through two key metrics: the l_a/l_b ratio, which represents the ratio between the longest and shortest edges; V_1/V_2 , which denotes the ratio of the volume between filler and the entire heater reactor (**Table S7**). A lower l_a/l_b ratio minimizes temperature gradients during operation, which is vital for achieving uniform graphitization. This aligns with classic heat transfer principles. Geometric symmetry reduces thermal anisotropy and ensures more uniform energy distribution.

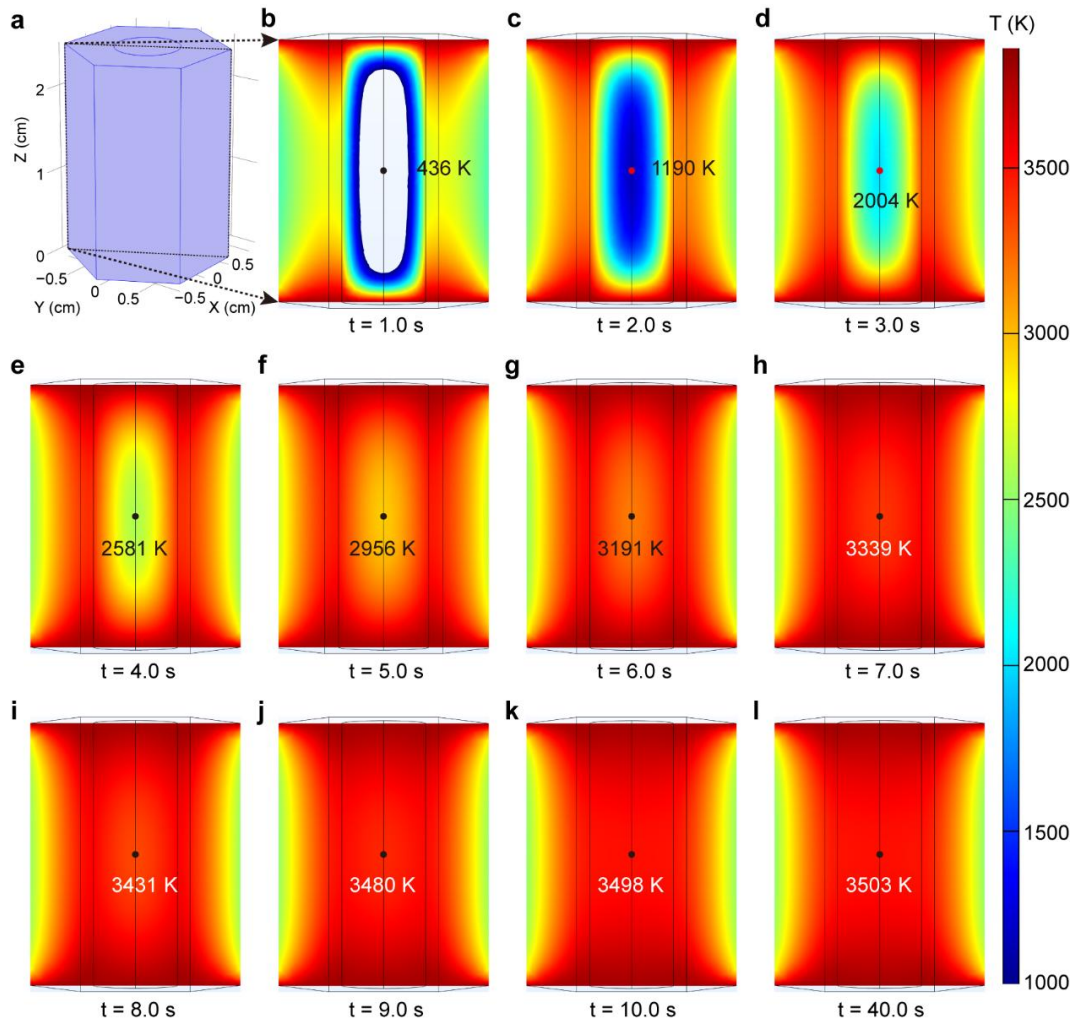


Supplementary Fig. 32 | Finite element simulations comparing the thermal field distributions of carbon felts with varying macroscopic geometries. (a, d, g, j) 2D cross-sectional temperature profiles. (b, e, h, k) Perspective views of the 3D thermal field. (c, f, i, l) Isothermal contour plots mapping the temperature gradients within the sample region. The simulated geometric models correspond to (a-c) a triangular prism, (d-f) a quadrangular prism, (g-i) a hexagonal prism, and (j-l) a cylinder.

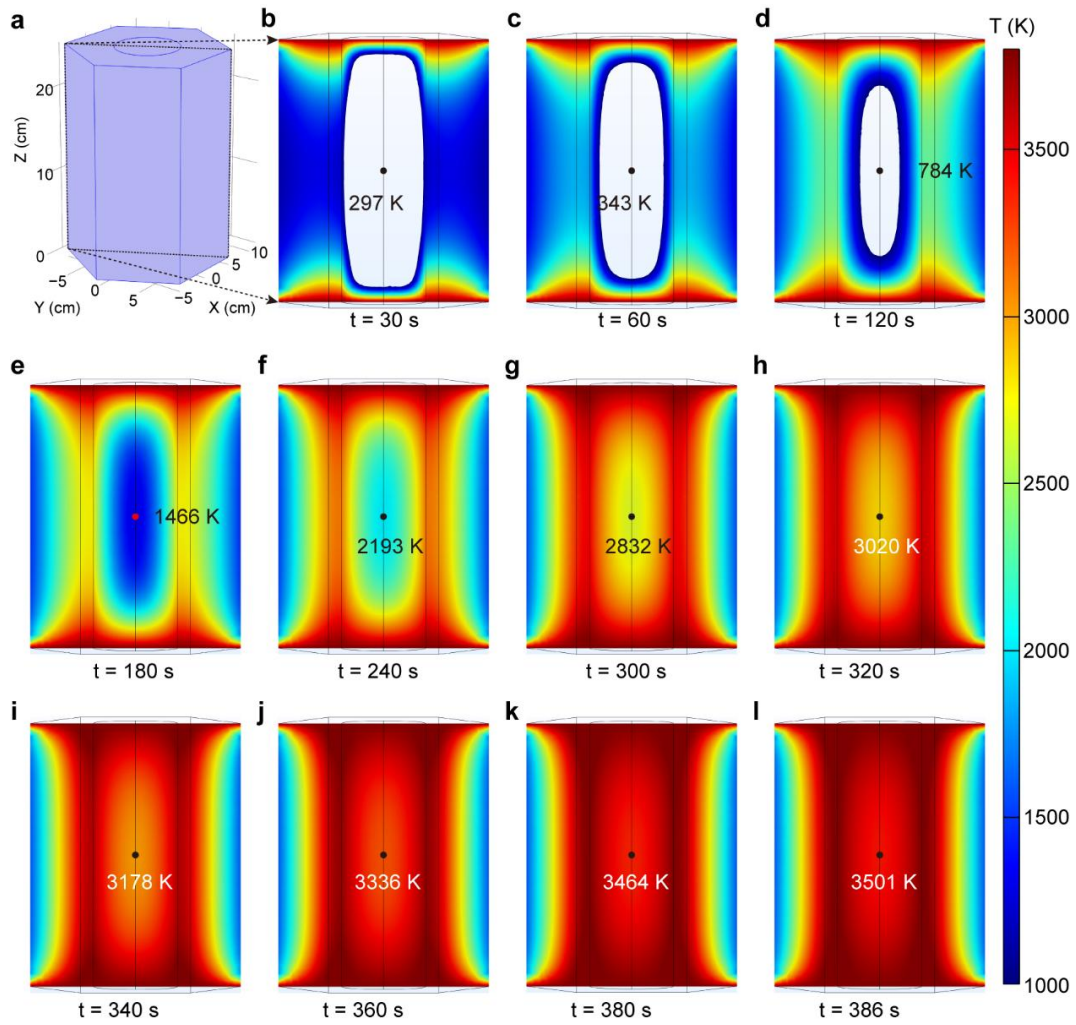
Note: The simulations reveal the profound impact of macroscopic shape on heating efficiency and spatial thermal uniformity. Geometries with sharp corners, such as the triangular and quadrangular prisms, exhibit severe thermal gradients and localized cold zones due to significant radiative heat dissipation at the acute edges. Furthermore, these acute-angled geometries are prone to severe powder accumulation. The precursor material easily gets trapped in the sharp corners, hindering complete material recovery and leading to unavoidable product loss during processing.



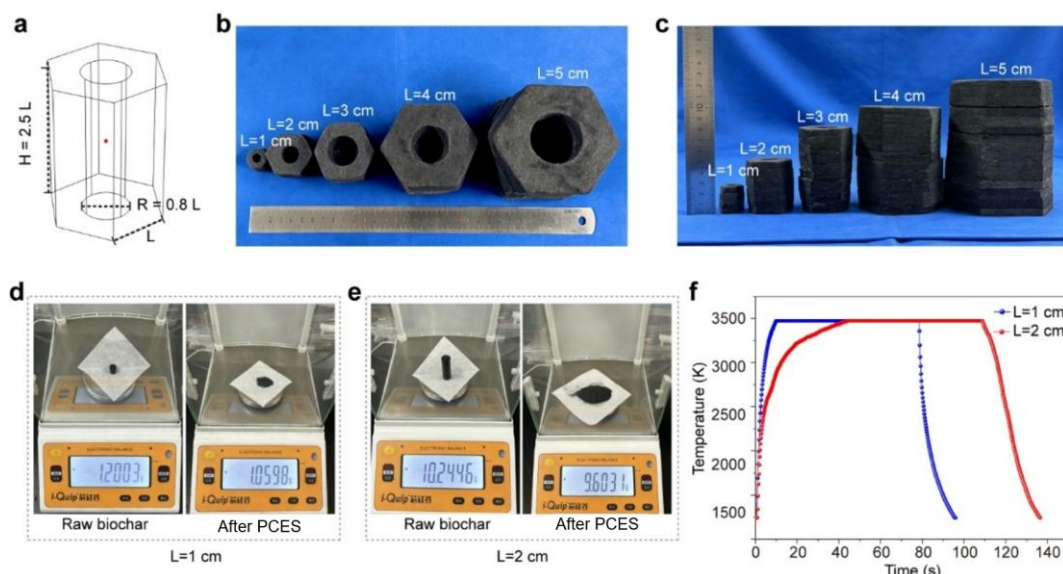
Supplementary Fig. 33 | Simulation results for electrothermal heating performance as a function of system scale. (a) Material configuration: a hollow hexagonal-prism carbon felt enclosing a central cylindrical sample, with plasma simulated by fixed 3800 K boundary conditions at the top and bottom interfaces. **(b)** Geometric parameters: the hexagonal prism has side length L and height $2.5 L$, the embedded cylindrical sample has a diameter of $0.8 L$ **(c–i)** Time evolution of core temperature and applied power for systems with $L = 1$ cm, 2 cm, 3 cm, 4 cm, 5 cm, 7 cm, and 10 cm, respectively.



Supplementary Fig. 34 | Cross-sectional temperature distribution over time for the scaled-down system with $L = 1$ cm. (a) Schematic of the model geometry and selected cross-section. (b–k) Temperature fields at $t = 1.0$ s to 10.0 s in 1-s increments; (l) Steady-state profile at $t = 40.0$ s. The core reaches ~ 3500 K by 10 s, demonstrating rapid heating capability.

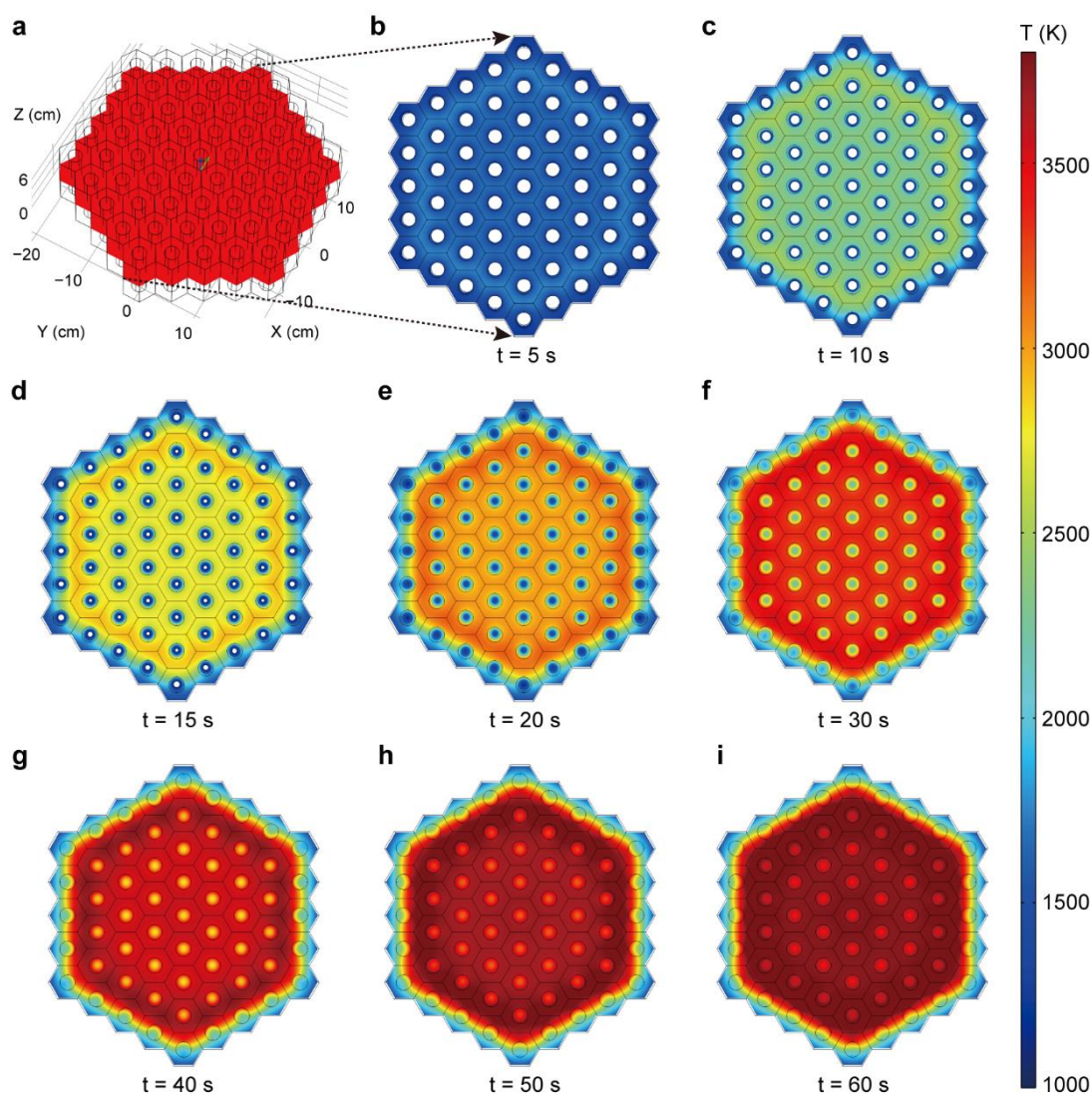


Supplementary Fig. 35 | Cross-sectional temperature evolution for the enlarged system with $L = 10$ cm. (a) Model schematic. (b–l) Temperature distributions at selected time points from 30 s to 386 s. The core temperature attains 3500 K only after 386 s, indicating significantly reduced heating efficiency at larger scales—highlighting the practical limitation on reactor size for ultrafast graphitization.

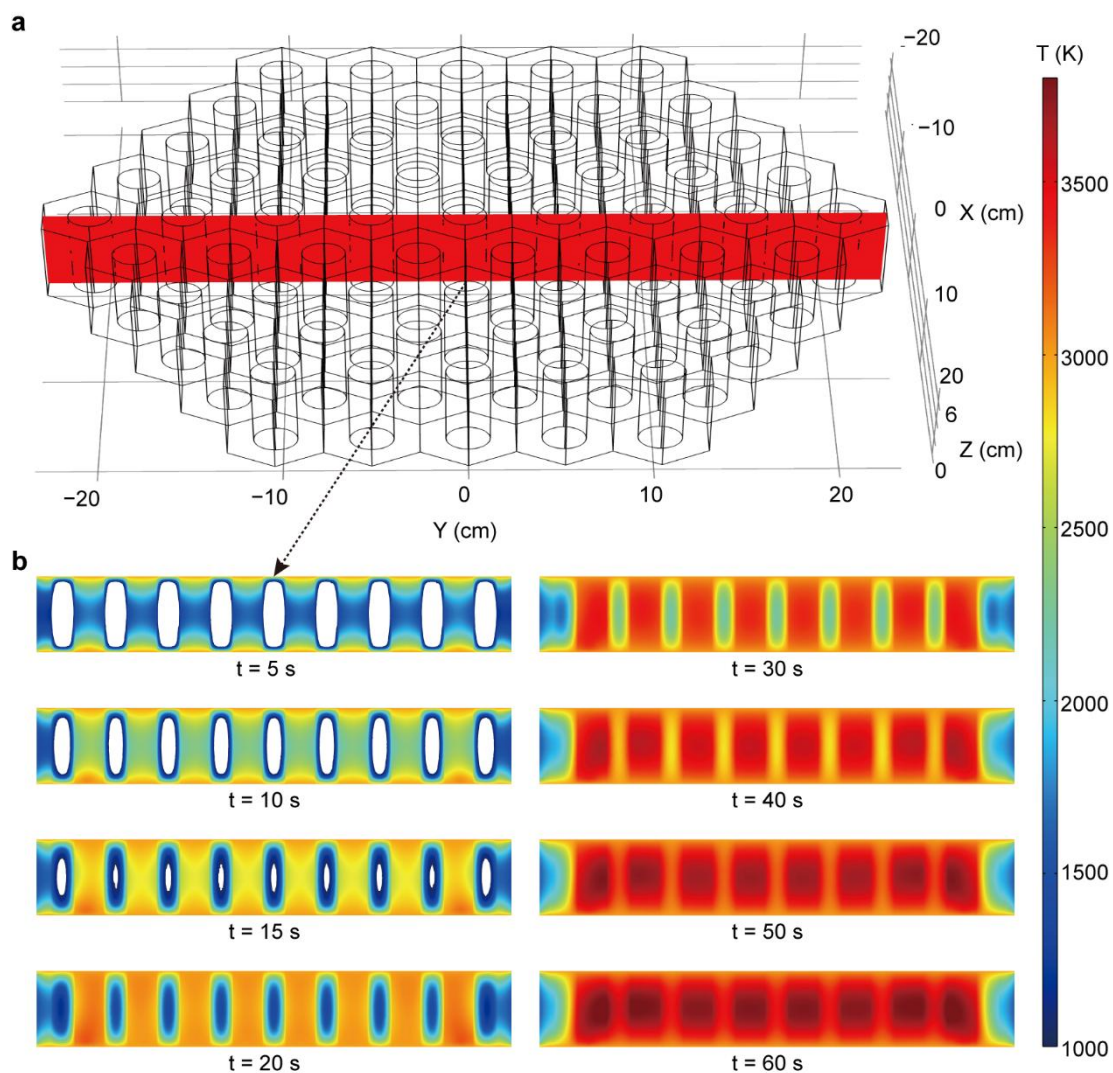


Supplementary Fig. 36 | Experimental validation of the PCES scaling kinetics. (a) Geometric schematic of the hexagonal prism heating module. (b-c) Top and side-view photographs of the carbon felt modules with side lengths ranging from $L = 1$ cm to $L = 5$ cm. (d-e) Digital photographs showing the mass and physical appearance of the compacted raw biochar precursor (left) and the resulting graphite product after the PCES process (right) for $L = 1$ cm and $L = 2$ cm modules, respectively. (f) Experimentally measured transient temperature profiles for the $L = 1$ cm and $L = 2$ cm modules. The $L = 1$ cm and $L = 2$ cm modules successfully rapidly ramped to the target equilibrium temperature of 3500 K in approximately 10 s and 30 s, respectively, highly consistent with the theoretical simulations.

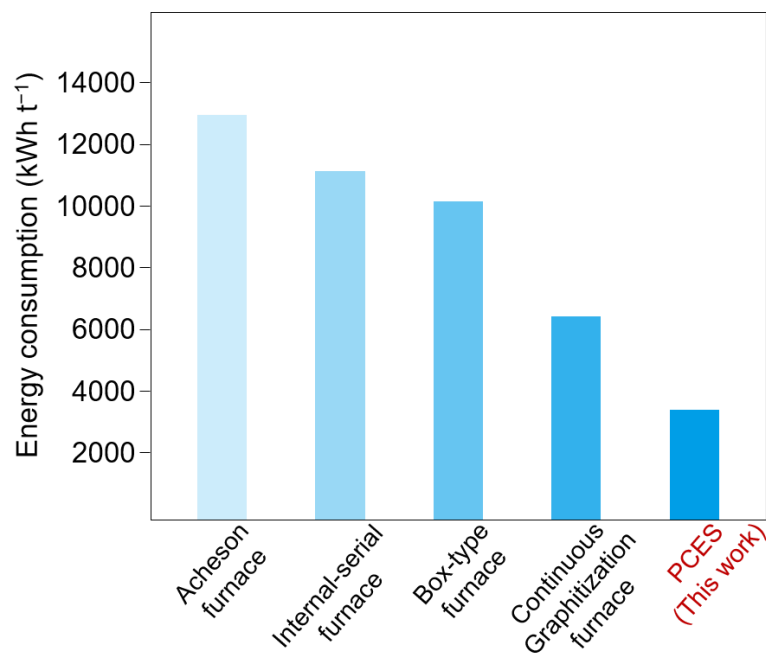
Note: To optimize the electrical resistance and ensure a successful Joule heating process, the raw biochar precursor was mechanically compacted into solid cylindrical pellets. The measured bulk density of the compacted precursor is approximately 1 g cm^{-3} . Furthermore, constrained by the 15 kW power supply, we experimentally evaluated the energy consumption specifically for the $L = 1$ cm and $L = 2$ cm modules. The actual measured energy consumptions were 62499 kWh t^{-1} and 21628 kWh t^{-1} , respectively. Both the transient heating times and the empirical energy consumption data demonstrate excellent agreement with our theoretical calculations. This comprehensive consistency supports the reliability of our scaling model and provides a basis for extrapolation to larger industrial scales and justifies the subsequent extrapolation to larger industrial dimensions.



Supplementary Fig. 37 | Thermal simulation of an integrated multi-unit reactor design. (a) Perspective view of the assembly, the red plane denotes the investigated cross-section. (b–i) Temperature distribution within this cross-section at $t = 5$ s, 10 s, 15 s, 20 s, 30 s, 40 s, 50 s, and 60 s. This integrated approach allows uniform heating up to 3500 K in under 60 seconds, while substantially improving batch throughput.



Supplementary Fig. 38 | Thermal simulation of an integrated multi-unit reactor design. (a) Perspective view of the assembly, the red plane denotes the investigated longitudinal section. (b) Temperature distribution within this longitudinal section at $t = 5$ s, 10 s, 15 s, 20 s, 30 s, 40 s, 50 s, and 60 s. This integrated approach allows uniform heating up to 3500 K in under 60 s, while substantially improving batch throughput.



Supplementary Fig. 39 | Comparative energy consumption analysis between traditional industrial furnaces and the PCES method.

Note: Traditional industrial graphitization relies on energy-intensive equipment such as the Acheson furnace (13000 kWh/t), internal-serial furnaces (11250 kWh/t), and box-type furnaces (10250 kWh/t), all of which suffer from prolonged heating cycles and significant radiative heat loss. Even advanced continuous graphitization furnaces consume approximately 6500 kWh/t. In contrast, using the PCES method confirms an average energy consumption of only 3594 kWh/t, demonstrating the exceptional energy efficiency and commercial viability of scaled PCES production. The source data were taken from reported literature²³.

Supplementary Table 1 | Temperature range, rated power, and fastest heating rate of the commercial graphitization furnace.

Manufacturer	Graphitization furnace	Temperature range (K)	Rated power (kW)	Fastest heating rate (K/min)
Chenxin Technology Co., Ltd.	CX-GF10/15VT	RT~3273	50	100
	CX-GF20/30VT	RT~3473	80	100
	CX-GF30/50VT	RT~3473	120	100
Thermal Technology LLC	1000-3560-FP-20	RT~2923	60	100
	1000-6580-FP-40	RT~2573	100	100
	HTG 9020-FP23	RT~2573	150	100
MTI Guangzhou Carbotech Co., Ltd.	GRF3000	RT~3273	70	100
	CB-Lab3000	RT~3373	60	100
	CB-UHT3000	RT~3273	60	100
	CB-3000	RT~3123	60	100
Omni R&D	ORDVGF1010	RT~3273	160	100
Hunan Xinrui Automation Equipment Co., Ltd.	JR-SMH-Φ10X15	RT~3373	30	100
	XR-LXM-10X20X50	RT~3073	80	100
	XR-SMH-Φ120X200	RT~3073	1200	100
Centorr Vacuum Industries	LF-2000	RT~2273	16	20
	LF-3000	RT~3273	18	20
MRF	Graphite Tube Furnace	RT ~ 3273	30	100
	LHTG 100-200/22-1G	RT ~ 2473	22	10
	LHTG 100-200/30-1G	RT ~ 3273	40	20
	LHTG 200-300/30-1G	RT ~ 3273	85	20
Hunan Fengpu Technology Co., Ltd.	Laboratory Graphitization Furnace	RT~3473	120	100
Hunan Jingtian Automation Equipment Co., Ltd.	JT-SML-10-C	RT ~ 3273	80	80
	JT-SML-100-C	RT ~ 3273	400	80
	JT-SML-340-C	RT ~ 3273	1200	80
Zhuzhou Nuotian Electric Heating Technology Co., Ltd.	NTI-SML-54W	RT ~ 3273	150	100
	NTI-SML-182W	RT ~ 3273	250	100
	NTI-SML-400W	RT ~ 3123	400	100

RT: room temperature.

Rated power: denotes the maximum sustained power output or power handling capacity that a device is designed to support continuously under its nominal operating conditions, including specified voltage, temperature, and cooling requirements. It represents the upper operational limit for safe and reliable long-term performance, as defined by the manufacturer.

Supplementary Table 2 | Comparison of different graphitization furnaces.

Indicator (unit)	Acheson furnace	Internal- serial furnace	Box-type furnace	Continuous graphitization furnace
Average furnace charge (t/furnace)	50-200	5-60	100-250	/
Maximum core temperature (K)	3373	3273	3323	3273
Energy consumption (kWh/t)	8800-13000	6500-8000	7000-10000	6500
Initial cycle efficiency (%)	96	98.6	93.4	93.1
Yield (%)	92.9	93	91	91
Production auxiliary materials	A large	No resistance material	A small	No resistance material
	amount of		amount of	
	resistance material		resistance material	
Comprehensive energy utilization rate (%)	30-40	40-50	40-60	50-60
Market share	50-55	5	35-40	/

The source of data was based on reported work by Gao, et al²³.

658 **Supplementary Table 3** | Comparison of different graphitization methods.

Raw material	Graphitization method	Temperature (K)	Heating up time	GD (%)	Ref.
Metallurgical coke	Joule heating	2273	~1.5 s	70	24
Lignin-derived carbon	Joule heating	2500	1 h	36.9	25
Asphalt, petroleum coke, and CO ₂ -derived carbon	Joule heating	3073	2 h	91.93	26
Activated carbon	Microwave heating	1773	5 min	85.8	27
Pitch coke	Microwave heating	1423	2 h	11.6	28
Anthracite	Induction heating	2873 3073	0.5 h	80.6 85.8	29
Pitch coke	Modified astro furnace heating	2573 2273 1523	25 h	69.7 29.1 11.6	30
Oil coke	Electrical furnace heating	2273 3273	1 h	26.7 91.86	31
Sucrose-derived carbon	Acheson furnace heating	3273	172 h	93	32
Lignin-derived carbon	Concentrated solar heating	2273	1 h	62.8	33
switchgrass				42.09	
cotton				53.95	
hemp	Electrical furnace heating	1437	1 h	63.37	34
miscanthus				75	
Corn stover				76.63	
Anthracite	Electrical furnace heating	1373	2 h	32.55	35
Petroleum coke	Electrical furnace heating	1573	3 h	77.32	36
Glucose-derived hard carbon	Electrical furnace heating	2273	3 h	93.02 46.51	37

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660 **Supplementary Table 4** | Comparison of different Joule heating equipment.

Commercial Joule heating equipment	Temperature range (K)	Peak rated power (kW)	Fastest rate of heating (K/s)
Hefei In-situ Technology	RT~3273	10/20	10000
UHS-3000Plus (Tianjin Zhonghuan Electric Furnace Co., Ltd.)	RT~3273	20	500
Zhongke Jingyan	RT~3273	18	20000
JDF-400 (Beijing Jingke Zhichuang Technology Development Co., Ltd.)	RT~3473	160	1000000
ESHOCK (Jilin Padike Precision Instrument Co., Ltd.)	RT~3273	8	100000
Flash Joule Heating Device LMJHD-A103	RT~3673	20	60000
Flash Joule Heating Device LMJHD-A100	RT~3273	20	100000
FJH-2026Aplus (Taiyuan Saiyin Materials Co., Ltd.)	RT~3873	3.5	3600
SSC-JHM (Shandong Shinsco Instrument Co., Ltd.)	RT~3073	15	1000
HTS (Shenzhen Joule Intelligent Control Technology Co., Ltd.)	RT~3273	6	10000

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662 **Supplementary Table 5** | Initial CE and reversible capacity of different methods.

Method	Initial CE (%)	Reversible capacity (mAh g ⁻¹)	Ref.
Citric acid leaching	~65.0	~330	38
Water leaching	~75.9	~339	39
Electrolysis	~81.9	~350	40
Furnace sintering	~62.9	~361	41
Flash Joule Heating	~78.1	~351	42
Water washing and Flash Joule Heating	~91.7	~343	43
Boric acid leaching and furnace sintering	~82.0	~332	44
Sulfuric acid leaching and furnace sintering	~94.0	~358	45
Directly and rapidly high-temperature upcycling	~90.0	~350	46
Molten salt electrochemical upcycling	~77.0	~300	47
Si metalloid catalysts and furnace sintering	~85	~330	32
PCES	~85.2	~351	This work

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Supplementary Table 6 | Thermal and electrical conductivities of graphite and amorphous carbon.

Materials	Thermal	Electrical
	Conductivity (W/(m·K))	Conductivity (S/m)
Graphite	~2000	$10^4 \sim 10^5$
Amorphous carbon	~0.1	$10^3 \sim 10^4$

Note: As shown in the table, the thermal conductivity of graphite (~2000 W/(m·K)) is four orders of magnitude higher than that of amorphous carbon (~0.1 W/(m·K))⁴⁸, while its electrical conductivity ($10^4 \sim 10^5$ S/m) also significantly surpasses that of its disordered counterpart ($10^3 \sim 10^4$ S/m)^{49,50}.

672 **Supplementary Table 7** | Comparison of parameters for different geometric shapes.

Parameter	Triangular prism	Quadrangular prism	Hexagonal prism	Cylinder
l_a	R	R	R	R
l_b	$\frac{R}{2}$	$\frac{R}{\sqrt{2}}$	$\frac{\sqrt{3}R}{2}$	R
$\frac{l_a}{l_b}$	2.000	1.414	1.155	1.000
V_1	$\frac{\pi}{16}R^2h$	$\frac{\pi}{8}R^2h$	$\frac{3\pi}{16}R^2h$	$\frac{7\pi}{4}R^2h$
V_2	$\frac{3\sqrt{3}}{4}R^2h$	$2R^2h$	$\frac{3\sqrt{3}}{2}R^2h$	$(8\sqrt{3} + 12)R^2h$
$\frac{V_1}{V_2}$	15.1%	19.6%	22.7%	21.3%

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674 **Supplementary Table 8** | Comparison of heating-up time, batch yields, and peak
675 temperatures across various conventional and emerging thermal technologies.

Method	heating-up time (s)	Yield per batch (g)	Peak temperature (°C)	Ref.
Laser-induced flash heating	~0.01	<0.01	~2500	51
Carbothermal shock	~0.05	~0.01	~2000	52
Flash Joule heating	~0.1	~0.03	~2727	17
Flash Joule heating	~0.05	~0.03	~2727	17
Flash Joule heating	~0.25	~1.2	~2727	17
Flash Joule heating	~0.5	~0.5	~2000	24
Flash Joule heating	~0.5	~1.0	~2627	24
Flash Joule heating	~0.25	~2.77	~2527	53
Flash Joule heating	~0.035	~0.04	~2727	54
Flash Joule heating	~249	~96.5	~3000	55
Flash Joule heating	~25	~15	~2500	56
In situ self-heating CVD	~1.0	~0.05	~1200	18
Spark Plasma Sintering (SPS)	~960	~3.0	~1600	57
Induction heating	~3900	~10.0	~1300	58
Hot pressing	~8400	~15	~1400	59
Conventional high- temp furnace	~22200	~20	~2500	60

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Supplementary Table 9 | Simulated and calculated parameters for individual reactors of various sizes.

<i>L</i> (cm)	Mass (g)	Heating up time (s)	Power (W)	Energy consumption (kWh t ⁻¹)
1	1.3	10	3337	58502
2	10.0	32	9814	23729
3	33.9	55	17473	14861
4	80.4	83	22736	11665
5	157.1	126	27236	9355
7	431.0	218	40343	7684
10	1256.6	386	62415	7127

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