

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

Ye-Chuang Han

ychan93@xmu.edu.cn

State Key Laboratory of Physical Chemistry of Solid Surfaces, Xiamen University

<https://orcid.org/0000-0001-8998-6061>

Pei-Yu Cao

State Key Laboratory of Physical Chemistry of Solid Surfaces, College of Chemistry and Chemical Engineering, IKKEM, Xiamen University

Zhiwei Wang

State Key Laboratory of Mechanics and Control for Aerospace Structures, Nanjing University of Aeronautics and Astronautics

Ming-Ze Yang

State Key Laboratory of Physical Chemistry of Solid Surfaces, Xiamen University

Su-Fan Hu

State Key Laboratory of Physical Chemistry of Solid Surfaces, College of Chemistry and Chemical Engineering, Innovation Laboratory for Sciences and Technologies of Energy Materials of Fujian Province

Junjie Fang

State Key Laboratory of Mechanics and Control for Aerospace Structures, Nanjing University of Aeronautics and Astronautics

Huang Huang

State Key Laboratory of Photovoltaic Science and Technology, Fudan University

Long Zhang

State Key Laboratory of Physical Chemistry of Solid Surfaces, College of Chemistry and Chemical Engineering, IKKEM, Xiamen University

Tianze Zhang

<https://orcid.org/0000-0001-8341-3510>

Yu Gu

State Key Laboratory of Physical Chemistry of Solid Surfaces, College of Chemistry and Chemical Engineering, IKKEM, Xiamen University <https://orcid.org/0000-0002-8099-4716>

Jing-Hua Tian

State Key Laboratory of Physical Chemistry of Solid Surfaces, College of Chemistry and Chemical Engineering, IKKEM, Xiamen University

Wanlin Guo

Nanjing University of Aeronautics and Astronautics <https://orcid.org/0000-0002-2302-8044>

Hu Qiu

State Key Laboratory of Mechanics and Control for Aerospace Structures, Nanjing University of Aeronautics and Astronautics

Zhong-Qun Tian

State Key Laboratory of Physical Chemistry of Solid Surfaces, College of Chemistry and Chemical Engineering, IKKEM, Xiamen University <https://orcid.org/0000-0002-9775-8189>

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**Plasma-coupled electrothermal ultrahigh-temperature
graphitization of non-graphitizable hard carbons via
heterogeneous edge-seeded layer 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.); yhan93@xmu.edu.cn (Y.-C.H.).

Abstract

Graphitization is inherently limited by solid-state carbon self-diffusion, whose activation energy (682 kJ mol^{-1}) ranks among the highest recorded for elemental solids, underpinning the 70-year dichotomy between graphitizable soft and non-graphitizable hard carbons. Industrial production currently relies on multi-day annealing of fossil-derived coke in Acheson furnaces. Here we report a plasma-coupled electrothermal ultrahigh-temperature synthesis (PCES) strategy that couples bulk Joule heating and localized interfacial plasma excitation in a two-stage sequence, achieving spatially uniform temperatures above 3500 K within seconds to directly surmount the carbon diffusion barrier. Ex-situ electron microscopy and ReaxFF simulations reveal a heterogeneous edge-seeded layer growth mechanism: reactive carbon species at particle interfaces condense epitaxially onto nascent sp^2 nuclei, bypassing sluggish bulk rearrangement and avoiding turbostratic disorder. We achieve 98 % graphitization of renewable hard carbon precursors, yielding Bernal-stacked graphite with performance matching commercial battery-grade graphite. A modular hexagonal reactor design decouples heating kinetics from scale, supporting projected ton-per-day output at about 60% lower energy than Acheson furnaces.

Main

Synthetic graphite is an indispensable functional material for modern energy storage, electronics, and advanced manufacturing, with lithium-ion battery anodes accounting for the largest share of global consumption^{1,2}. Driven by the rapid expansion of electric vehicle and grid-scale energy storage markets, global demand for battery-grade synthetic graphite is projected to grow 7.6-fold between 2022 and 2030, reaching approximately 13 million metric tons annually^{3–5}. For over a century, industrial graphite production has relied on the Acheson process and its derivatives, which treat fossil-derived feedstocks such as petroleum coke and coal tar pitch at 2700–3300 K for days to weeks^{6,7}. This long-standing manufacturing paradigm imposes a dual burden: near-

exclusive dependence on fossil feedstocks, and extremely high energy consumption and carbon emissions^{8,9}. Producing one ton of commercial synthetic graphite consumes approximately 11000 kWh of electricity and generates 9 tons of CO₂, with graphitization alone accounting for nearly half of total anode manufacturing costs^{10,11}. These challenges have created urgent demand for sustainable synthesis routes that reduce reliance on fossil resources and prolonged thermal equilibrium.

Carbon-negative biomass, with 146 billion metric tons generated annually worldwide, represents a compelling feedstock for sustainable graphite production^{12–14}. However, this goal has been blocked for more than 70 years by a fundamental dichotomy in carbon science: graphitizable “soft” carbons versus non-graphitizable “hard” carbons^{15–17}. Conventional production uses exclusively fossil-derived soft carbons, whose fusible sp²-hybridized aromatic domains rearrange into ordered graphite lattices above 2700 K¹⁸. In contrast, biomass- and plastic-derived hard carbons possess rigid cross-linked sp³ networks bridged by heteroatoms (e.g., O, N, S)¹⁹. This rigid structure prevents the layer sliding and coalescence required for graphitic ordering, making full graphitization extremely difficult even at 3300 K²⁰. The deeper, more universal barrier comes from reaction kinetics: graphitization is governed by solid-state carbon self-diffusion, which has an activation energy of approximately 682 kJ mol^{−1} and ranks among the highest recorded for elemental solids^{21,22}. This enormous barrier forces conventional processes to rely on days of isothermal holding to drive lattice reorganization, yet still often leaves residual defects and broad interlayer spacing distributions that degrade battery performance^{13,23,24}. Transition metal catalysis can reduce processing temperatures, but residual metallic impurities cause catastrophic battery failures such as internal short circuits, and no catalyst remains stable at the temperatures needed for complete hard carbon graphitization^{25,26}.

Ultrafast heating strategies, most notably flash Joule heating (FJH), have recently emerged as a transformative approach to high-temperature materials synthesis, achieving temperatures above 3000 K on sub-second timescales^{27,28}. These methods bypass the slow heating and cooling cycles of conventional furnaces and show promise

for carbon upcycling^{29–31}. Nevertheless, two critical limitations prevent their application to industrial production of high-quality crystalline graphite. First, the extreme non-equilibrium conditions of FJH typically produce turbostratic graphene with rotational stacking faults, rather than thermodynamically stable Bernal-stacked graphite, because rapid quenching kinetically traps growing carbon layers in disordered configurations^{29,32}. Converting turbostratic graphene into well-ordered graphite would require additional energy input to overcome an even larger thermodynamic barrier³³. Second, all rapid-heating technologies face an inherent scaling trade-off: increasing reactor volume reduces heating rates and temperature uniformity, degrading product consistency and limiting throughput^{27,28,34,35}. Resolving both the fundamental graphitization barrier and the engineering scaling challenge requires a new electrothermal design that combines ultrahigh temperature, spatial uniformity, and modular scalability.

Here we report a plasma-coupled electrothermal ultrahigh-temperature synthesis (PCES) strategy that synergistically couples bulk Joule heating with localized interfacial plasma excitation. This dual-zone design achieves spatially uniform temperatures above 3500 K within seconds, directly overcoming the carbon diffusion barrier. Using straw biochar as a model renewable hard carbon precursor, we demonstrate that PCES achieves near-complete graphitization with a 98 % graphitization degree, yielding crystalline graphite with electrochemical performance matching premium commercial battery-grade graphite. Ex-situ electron microscopy and ReaxFF molecular dynamics simulations reveal a heterogeneous edge-seeded layer growth mechanism: transient reactive carbon species generated at particle interfaces condense epitaxially onto nascent sp^2 nuclei, bypassing the sluggish bulk atomic rearrangement that limits conventional thermal graphitization. We further demonstrate a modular hexagonal reactor cluster design that decouples heating kinetics from production scale, supporting projected ton-per-day graphite manufacturing with about 60% lower energy consumption than industrial Acheson furnaces. This work overcomes the long-standing 70-year dichotomy between graphitizable soft carbons

and non-graphitizable hard carbons, provides a low-energy route to sustainable battery graphite, and establishes a promising generalizable platform for non-equilibrium extreme-temperature materials synthesis.

Temperature-dependent graphitization kinetics of hard carbon

The conversion of biomass-derived hard carbon into crystalline graphite proceeds via two core steps (Fig. 1a)¹³. The first is low-temperature carbonization at approximately 1300 K, which removes volatile components and forms a sp³-rich disordered biochar matrix. The second is ultrahigh-temperature annealing, which reconstructs the carbon framework into long-range ordered graphitic domains. The rate-limiting step of this process is solid-state carbon self-diffusion, which has an activation enthalpy of 682 kJ mol⁻¹ and ranks among the highest recorded for any elemental solid (Fig. 1b)^{27,28}. This formidable energy barrier explains why conventional graphitization routes, even with metal catalysis, fail to convert hard carbons into highly crystalline battery-grade graphite efficiently.

To define the temperature window for hard carbon graphitization at atomic resolution, we performed ReaxFF molecular dynamics simulations across 1500–4000 K, using a cellulose-derived amorphous carbon model representative of straw biochar (Supplementary Figs. 1,2)³⁶. At all tested temperatures, the sp² carbon fraction increases with time following a sigmoidal profile characteristic of nucleation-growth phase transformations (Fig. 1c). Fitting these data with the Johnson–Mehl–Avrami–Kolmogorov (JMAK) model shows that the apparent ordering rate constant k rises with temperature and peaks at 3500 K (inset of Fig. 1c). This trend indicates accelerated bond rearrangement and graphitic network formation at elevated temperatures^{37,38}. Further heating to 4000 K causes a sharp drop in k , because excessive thermal vibration breaks in-plane C=C bonds and degrades already formed graphitic domains.

The optimal graphitization temperature of 3500 K is corroborated by multiple structural descriptors. The C–C radial distribution function (RDF) at 3500 K exhibits the sharpest long-range periodicity, and matches the signature of crystalline graphite most closely

(Fig. 1d). The population of six-membered carbon rings, the fundamental building unit of graphite, also reaches its maximum at 3500 K (Fig. 1e). This trend is consistent with the emergence of large, continuous layered graphite domains in simulation snapshots (Fig. 1f). Collectively, these results establish that full graphitization of hard carbon requires temperatures approximately 800 K higher than for soft carbons⁷. This evidence arises from cross-linked sp³ domains and residual heteroatoms that impede layer rearrangement^{39,40}. As conventional industrial furnaces cannot reliably achieve and sustain such extreme temperatures uniformly (Supplementary Fig. 3, Supplementary Table 1)⁴¹, these findings motivated the design of our plasma-coupled electrothermal ultrahigh-temperature synthesis (PCES) strategy (Fig. 1g).

Graphitization of biochar via PCES in seconds

The PCES system adopts a dual-zone design. It includes a central bulk Joule heating zone for volumetric heating of carbon precursors, and two interfacial plasma zones that provide additional localized ultrahigh-temperature input (Fig. 2a). This synergistic integration of resistive Joule heating and contact-induced interfacial plasma heating achieves spatially uniform temperatures above 3500 K within seconds^{27,42}, and finite-element simulations verify the homogeneous temperature distribution (Supplementary Figs. 4,5). The superior heating efficiency of PCES arises from a two-stage mechanism. During the first 14 s, Joule heating dominates and rapidly raises the sample temperature to approximately 2800 K. Beyond this threshold, interfacial ultrahigh-temperature plasmas ignite spontaneously at carbon fiber junctions, delivering concentrated energy that drives the temperature to 3500 K within the next 30 s (Supplementary Figs. 6,7). At equivalent power input, PCES reaches a peak temperature 1100 K higher than pure Joule heating alone (Fig. 2b, Supplementary Fig. 8). This combined heating mode generates a uniform ultrahigh-temperature field on a second timescale, eliminating the steep thermal gradients that cause non-uniform and incomplete graphitization in traditional furnaces⁴¹.

Using purified straw biochar as a model hard carbon precursor (Supplementary Figs. 9,10), we synthesized graphite samples with systematically tunable graphitization

degrees. Quantified by X-ray diffraction (XRD) with a silicon internal standard to exclude baseline interference³¹, the sample treated at 3500 K exhibits a (002) interlayer spacing of 3.35 Å, corresponding to a graphitization degree of 98%, which we denote GD-98% (Fig. 2c). This value represents near-complete graphitization of a biomass-derived hard carbon. Raman spectroscopy confirms progressive defect healing. The I_D/I_G ratio decreases from 1.16 for raw biochar to 0.08 for GD-98% (Fig. 2d), indicating near-complete elimination of structural disorder³³.

Benchmarking against state-of-the-art graphitization technologies confirms the transformative performance of PCES (Fig. 2e, Supplementary Tables 2–4). Traditional furnace annealing achieves approximately 85% graphitization at 3000 K over 24 h, and microwave plasma methods reach approximately 70% at 3200 K over 10 min. By contrast, PCES attains 98% graphitization at 3500 K in seconds. Electron microscopy reveals the morphological and structural evolution from amorphous biochar particles to well-defined layered graphite, with clear lattice fringes verifying atomic-level ordering (Fig. 2f, Supplementary Fig. 11). The high-temperature treatment eliminates internal micropores and reduces specific surface area, consistent with cross-sectional SEM and pore size distribution analysis (Supplementary Fig. 12). X-ray photoelectron spectroscopy confirms progressive deoxygenation of the carbon framework (Supplementary Fig. 13). These microstructural improvements directly benefit ion transport and mechanical stability in battery applications.

When evaluated as a lithium-ion battery anode, the GD-98% electrode delivers an initial reversible capacity of 344 mAh g⁻¹ at 0.05 C (Supplementary Fig. 14) and a reversible capacity of 350 mAh g⁻¹ at 0.1 C (Fig. 2g). Galvanostatic cycling demonstrates excellent structural robustness, with negligible capacity degradation over 200 cycles and an average Coulombic efficiency above 99.5% during prolonged cycling (Fig. 2g). Under identical testing conditions, GD-98% exhibits a stabilized reversible capacity approximately 14 mAh g⁻¹ higher than that of premium commercial graphite (Supplementary Table 5). The enhanced electrochemical performance is directly attributed to the high crystalline order of PCES-derived graphite. A graphitization

degree of 98% ensures well-defined Bernal-stacked carbon lattices with minimal structural defects, which provide continuous, low-barrier pathways for reversible lithium intercalation and deintercalation^{6,43}. The low defect density contributes to structural integrity during repeated lithium insertion/extraction processes, enabling stable electrochemical cycling. These results verify that PCES produces battery-grade graphite from renewable hard carbon precursors on a second timescale, with electrochemical performance matching or surpassing conventional fossil-derived commercial products.

Heterogeneous edge-seeded layer growth mechanism

The graphitization mechanism of PCES differs fundamentally from conventional equilibrium thermal annealing. In PCES, structural reorganization initiates exclusively at particle–particle interfaces rather than uniformly throughout the bulk (Fig. 3a). This spatial selectivity originates from contact-resistance-induced localized Joule heating, which creates far higher energy densities at particle interfaces than in the particle interior (Supplementary Fig. 15). Interfacial temperatures exceeding 3500 K readily overcome the carbon diffusion barrier (Supplementary Fig. 16), generating nanoscale sp² graphitic domains that act as epitaxial seeds for subsequent growth (inset of Fig. 3a)^{44,45}. As these seeds expand, their high electrical and thermal conductivity gradually reduces interfacial resistance, creating a self-limiting heating effect (Supplementary Table 6)^{46–48}.

Flash Joule heating and other rapid thermal shock methods typically produce turbostratic graphene with rotational stacking faults instead of Bernal-stacked graphite^{27,29,32}. This occurs because extreme quenching rates kinetically trap growing carbon layers in rotationally disordered configurations^{29,32}. Converting turbostratic graphene to thermodynamically stable Bernal graphite requires overcoming an additional barrier as large as approximately 1640 kJ mol^{−1} (Supplementary Fig. 17)⁴⁶. In contrast, conventional furnaces establish uniform thermal equilibrium at temperatures below 3300 K over days (Fig. 3b), which is insufficient to activate hard carbon graphitization⁴⁰. PCES uniquely combines ultrahigh peak temperature,

controlled dwell time and interfacial nucleation, enabling ordered graphite growth rather than disordered carbon formation.

COMSOL Multiphysics simulations confirm that particle interfaces are the primary sites of energy deposition and atomic rearrangement. Both interface and bulk temperatures increase linearly with applied current, but interfaces exhibit much steeper temperature gradients. Transient profiles at microsecond resolution show that interfaces reach 3500 K within 1 ms, while the bulk remains below 2000 K at the same time point (Fig. 3c, Supplementary Fig. 18). This extreme spatiotemporal energy confinement is the origin of PCES's exceptional efficiency, as energy is concentrated precisely where sp^3 bond breaking and carbon rearrangement are needed.

Ex-situ transmission electron microscopy (TEM) and selected-area electron diffraction (SAED) directly visualize the progressive structural transformation driven by heterogeneous interfacial heating (Fig. 3d). At 1500 K, the particle core remains fully amorphous with diffuse SAED rings, while discontinuous few-layer graphite features appear at the surface. At 2500 K, isolated graphitic domains emerge in the core and the surface graphite layers thicken into continuous sheets. At 3500 K, uniform layered graphite extends throughout the entire particle, with sharp, well-defined SAED rings characteristic of crystalline graphite. This temperature-dependent evolution matches our simulation predictions and confirms that interfacial ultrahigh temperatures drive hexagonal ring formation and interlayer stacking alignment. Notably, the PCES strategy is generalizable to a wide range of renewable precursors (Supplementary Figs. 19–27). Repeated PCES cycling can further convert polycrystalline graphite into highly oriented pyrolytic graphite (HOPG, Fig. 3e, Supplementary Figs. 28,29), a capability that, to the best of our knowledge, has not been demonstrated by other rapid graphitization techniques⁴⁹.

Atomic-scale MD simulations reveal the detailed growth pathway under non-equilibrium PCES conditions (Fig. 3f). Time-resolved snapshots show that disordered carbon species first rearrange into locally ordered sp^2 graphitic fragments. Small graphitic clusters nucleate locally and then expand laterally, with new carbon structures

adding preferentially at the edges of existing graphitic domains. This observation supports a heterogeneous, edge-mediated growth mechanism rather than uniform bulk crystallization. During heating, the total number of carbon rings increases rapidly, with six-membered rings becoming the dominant population at 3500 K (Fig. 3g). This trend is consistent with the honeycomb topology of graphite and thermodynamic preference for graphitic configurations⁵⁰. Non-hexagonal rings persist throughout the simulation, indicating that the MD captures the onset of graphitization rather than full topological defect healing^{46,51}. Additionally, residual oxygen in the carbon matrix assists graphitization by oxidizing and removing disordered cross-linked structures at high temperatures (Supplementary Fig. 30).

Modular reactor design for scalable, low-energy industrial production

PCES achieves near-complete graphitization of sustainable biochar precursors in seconds, representing a breakthrough in time and energy efficiency. However, a fundamental challenge facing all emerging rapid-heating technologies is the inherent trade-off between production scale and temperature uniformity^{27,28}. Larger reactors suffer slower heating rates and wider temperature gradients, which degrade product consistency^{34,52}. To address this challenge, we used COMSOL simulations to guide the forward design of scalable PCES reactors, replacing empirical trial-and-error with simulation-driven optimization.

We first evaluated four geometric shapes, including triangular, square, hexagonal and cylindrical prisms, to minimize thermal anisotropy and maximize space utilization (Fig. 4a, Supplementary Fig. 31, Supplementary Table 7). The hexagonal prism design offers the optimal balance. It minimizes edge-length disparity to reduce thermal gradients while achieving high packing efficiency, making it well suited for batch processing (Supplementary Fig. 32). We then analyzed the trade-off between element size and performance for hexagonal prisms ranging from 1 to 10 cm in side length (Fig. 4b, Supplementary Figs. 33–35). Smaller elements (1 cm) reach 3500 K in approximately 10 s but have higher specific energy consumption ($\sim 60000 \text{ kWh ton}^{-1}$). Larger elements with 10 cm side length require approximately 400 s to reach target temperature but

achieve much lower average energy consumption approximately 7000 kWh ton⁻¹. This size–efficiency trade-off is consistent with fundamental Joule heating models, and we validated the simulation results experimentally with small-batch tests (Supplementary Fig. 36).

To resolve this trade-off, we propose a modular hexagonal prism cluster design that decouples heating kinetics from production scale. Compared with existing thermal technologies, PCES uniquely bridges the gap between ultrafast heating kinetics and macroscopic batch yields (Fig. 4c,d, Supplementary Tables 8,9). Each module retains the fast-heating kinetics of small elements, while parallel operation of multiple modules scales output linearly without compromising thermal uniformity. Thermal field simulations confirm that clustered hexagonal elements deliver uniform working temperatures above 3500 K across the entire reactor volume (Fig. 4e–g, Supplementary Fig. 37). All modules reach 3500 K within 60 s, and temperature fluctuations across the sample bed remain below 5% (Supplementary Fig. 38).

Technoeconomic analysis demonstrates the outstanding energy efficiency of scaled PCES production. Batch tests with 1.2 kg loads confirm an average energy consumption of ~4000 kWh ton⁻¹, corresponding to approximately 1.7 tons of graphite per day. This represents a 60% reduction compared with industrial Acheson furnaces (Supplementary Fig. 39). Peak power demand scales linearly with batch mass and responds on a second timescale, enabling seamless integration with renewable power grids such as solar and wind for flexible low-carbon production (Supplementary Table 9). Importantly, PCES eliminates dependence on fossil-derived precursors and metal catalysts, further reducing environmental footprint and production costs. This modular design provides a demonstrated, scalable pathway to battery-grade graphite manufacturing, establishing PCES as a viable and competitive platform for sustainable industrial graphite production.

312

313 **Conclusion**

314 In summary, we developed a PCES strategy that achieves seconds-scale graphitization
315 of traditionally non-graphitizable hard carbons at 3500 K, overcoming the 70-year-old
316 dichotomy between graphitizable soft carbons and traditionally non-graphitizable hard
317 carbons in carbon materials science. By synergizing bulk Joule heating with localized
318 interfacial plasma excitation, this approach directly overcomes the high kinetic barrier
319 of solid-state carbon self-diffusion. The process follows a heterogeneous edge-seeded
320 layer growth mechanism that bypasses the sluggish bulk atomic rearrangement limiting
321 conventional thermal graphitization, and avoids the turbostratic stacking disorder
322 typical of flash Joule heating approaches. Transient reactive carbon species formed at
323 particle interfaces condense epitaxially onto nascent sp^2 nuclei to drive ordered
324 crystalline growth. The resulting biomass-derived graphite exhibits a graphitization
325 degree up to 98% and electrochemical performance matching premium commercial
326 battery-grade graphite. Our modular hexagonal reactor cluster design decouples
327 ultrafast heating kinetics from production scale, overcoming the universal scaling trade-
328 off that plagues most rapid high-temperature synthesis technologies. Technoeconomic
329 analysis confirms an approximately 60% reduction in energy consumption relative to
330 industrial Acheson furnaces, supporting projected ton-per-day production of high-
331 quality graphite from renewable feedstocks. Beyond sustainable graphite
332 manufacturing, this work establishes a promising generalizable platform for non-
333 equilibrium extreme-temperature materials synthesis.

334

335 **Methods**

336 **Materials**

337 Raw straw (Foshan Perth Carbon Materials Technology Co., Ltd.), needle coke (KenYe
338 Technology Co., Ltd.), carbon fiber precursor (Toray Industries Inc.), carbon paper

precursor (Guangdong Hydrogen Development New Materials Technology Co., Ltd.), and 0.025 mm polyimide film (Foshan Huayong Technology Co., Ltd.) were used as received. Recycled plastic was sourced from post-consumer mineral water bottles. Dealkaline lignin (Beijing Huamaike Biotechnology Co., Ltd.), water-soluble phenolic resin with ~70 wt% solid content (Shanghai Macklin Biochemical Co., Ltd.), polyaniline (98%, Shanghai Macklin Biochemical Co., Ltd.), and silicon powder (99.99%, Shenzhen Kejing Zhida Technology Co., Ltd.) were used without further purification.

PCES system setup

The PCES system consists of a water-cooled reaction chamber, high-purity graphite electrodes, and a programmable DC power supply (PRD8V50, Xi'an Actionpower Electric Co., Ltd.). The power supply delivers an adjustable output of 0–80 V and 0–500 A, with $\pm 0.02\%$ full-scale accuracy. Sample temperature was recorded by a non-contact infrared pyrometer (IMGA 740, IMPAC) with a 500–3500 K measurement range and $<0.3\%$ uncertainty, coupled to a high-speed data acquisition system with 1 μs temporal resolution.

Graphitization of straw-derived hard carbon precursor

Raw straw was immersed in 40 wt% hydrofluoric acid for 48 h to remove inorganic impurities, then rinsed with deionized water to neutral pH and dried at 353 K. The purified straw was carbonized at 1300 K for 2 h in a horizontal tube furnace under nitrogen atmosphere, with a heating ramp rate of 10 K min^{-1} . The resulting biochar was ground into fine powder, pressed into pellets, and loaded into the core of a cylindrical carbon felt assembly. The assembly was sandwiched between two layers of conductive carbon paper and fixed with graphite electrodes.

The PCES process proceeded via two sequential heating stages. Joule heating dominated the first 14 s, raising the sample temperature to approximately 2800 K. Interfacial plasma then ignited spontaneously at fiber junctions, delivering additional localized energy to reach target peak temperatures within the next 30 s. Samples were

treated at peak temperatures of 2550 K, 2600 K, 2900 K and 3500 K to achieve graphitization degrees of 53%, 75%, 87% and 98%, respectively. After a 30 s isothermal hold at the peak temperature, the current was cut off and samples cooled naturally to room temperature. The as-prepared graphite was ground and sieved through 325-mesh and 500-mesh screens to control particle size, then homogenized for further characterization.

Graphitization of other precursors

Needle coke, carbon fiber precursor (polyacrylonitrile), carbon paper precursor, polyimide film, plastic, lignin, phenolic resin and polyaniline-derived carbon were processed following the same protocol. All precursors were first carbonized at 1300 K for 2 h under nitrogen, then subjected to PCES treatment at 3500 K with a 30 s isothermal hold.

Synthesis of highly oriented pyrolytic graphite

PCES-synthesized graphite was reground, pressed into pellets, and subjected to repeated PCES cycles at 3500 K with 30 s isothermal hold per cycle. Samples were treated for 10, 20, 40, 60, 80 and 100 cycles to tune the degree of structural ordering.

Material characterization

Scanning electron microscopy (Zeiss GeminiSEM 500) was used to examine sample morphology. X-ray diffraction measurements were performed on a Rigaku Ultima-IV system with Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) to quantify graphitization degree. Raman spectra were collected on a HORIBA XploRA PLUS system to evaluate structural defects and stacking order. Specific surface area and pore size distribution were measured with a TriStar II 3020 instrument. Atomic-scale structure was characterized by aberration-corrected transmission electron microscopy (JEOL ARM200F). X-ray photoelectron spectroscopy (PHI VersaProbe 5000) was used to analyze surface chemical states. Focused ion beam SEM (Crossbeam 350) was employed to prepare cross-sectional lamellae, with milling performed at accelerating voltages from 30 kV down to 2 kV and beam currents from 3 nA to 10 pA.

Electrochemical measurements

Working electrodes were prepared by mixing active material, Super P carbon, carboxymethyl cellulose, and styrene-butadiene rubber at a mass ratio of 95.5:1.5:1.2:1.8 in deionized water. The resulting slurry was cast onto copper foil with a doctor blade and dried under vacuum at 353 K for 12 h. CR2032 coin cells were assembled in an argon-filled glove box with O₂ and H₂O levels below 0.01 ppm, using lithium metal as the counter electrode, 1 M LiPF₆ in ethylene carbonate/dimethyl carbonate/ethyl methyl carbonate (1:1:1 by volume) as the electrolyte, and a Celgard 2500 polypropylene separator.

Galvanostatic cycling and rate performance tests were conducted on a Neware battery test system at room temperature. Cyclic voltammetry and electrochemical impedance spectroscopy measurements were performed on a CHI660D electrochemical workstation, with impedance tested over a frequency range of 200 kHz to 10 mHz at 5 mV AC amplitude.

Calculation of graphitization degree

The graphitization degree g was calculated from the (002) interlayer spacing d_{002} using the following equation:

$$GD = \frac{0.3440 - d_{002}}{0.3440 - 0.3354} \times 100\% \quad (1)$$

where 0.3440 nm corresponds to the interlayer spacing of fully non-graphitized carbon, and 0.3354 nm corresponds to ideal crystalline graphite. GD represents the graphitization degree, and d_{002} denotes the (002) interplanar spacing of the carbon material.

The interlayer spacing d_{002} was derived from Bragg's law:

$$d_{002} = \frac{n\lambda}{2\sin(\theta_{002})} \quad (2)$$

Where n represents the diffraction order ($n = 1$), λ denotes the X-ray wavelength ($\lambda = 0.1541$ nm), and θ_{002} indicates the diffraction angle of the (002) peak for the carbon

421 material.

422 A silicon internal standard was used to correct for instrumental angle deviation. The
423 corrected $2\theta_{111}^{Si}$ was obtained as:

$$424 \quad 2\theta_{002} = 2\theta'_{002} + \Delta \quad (3)$$

425 Where θ'_{002} represents the diffraction angle of the (002) peak measured from the carbon,
426 and Δ denotes the measurement error.

$$427 \quad \Delta = 28.443 - 2\theta_{111}^{Si} \quad (4)$$

428 Where θ_{111}^{Si} represents the diffraction angle of the (111) peak for Si powder.

429

430 **Data availability**

431 All data needed to evaluate the conclusions in the study are present in this Article or its
432 Supplementary Information. Source data are provided with this paper.

433

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Author contributions

Y.-C.H. conceived the idea and designed the experiments. Y.-C.H., H.Q., W.G. and Z.-Q.T. supervised the project. P.-Y.C., M.-Z.Y. and S.-F.H. carried out all the sample preparation and characterization experiments under the supervision of Y.-C.H. Z.W. and J.F. performed the MD simulation under the supervision of H.Q. M.-Z.Y. performed the COMSOL Multiphysics simulation and electrothermal mechanism analysis under the supervision of Y.-C.H. P.-Y.C., S.-F.H., L.Z. and T.Z. tested the electrochemical performance of synthetic graphite under the supervision of Y.G. and J.-H.T. H.H. assisted with TEM design and data analysis. Y.-C.H. and H.Q. wrote the manuscript with the contribution of all authors.

Conflict of Interest

The authors declare no competing interests.

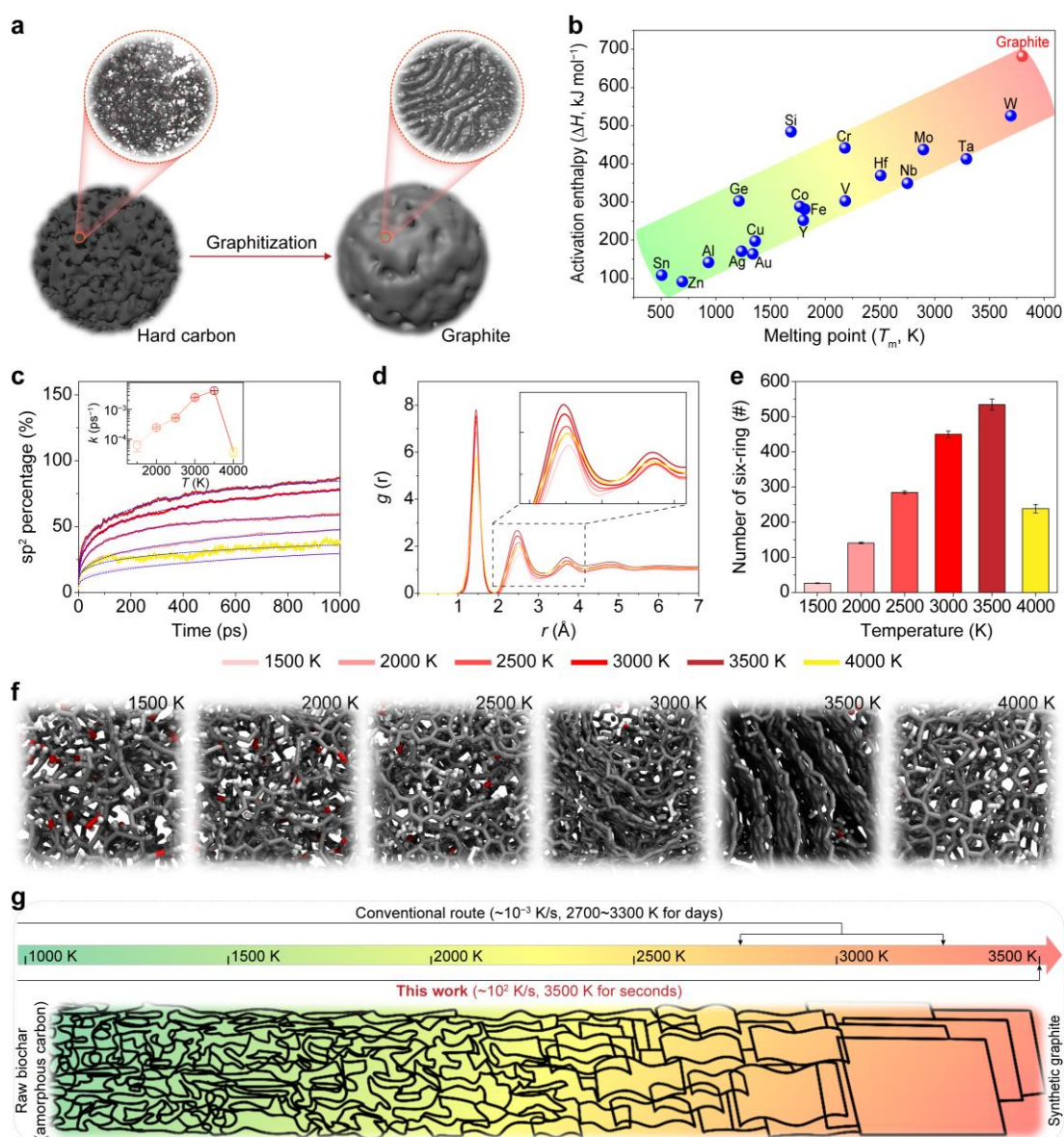


Fig. 1 | ReaxFF MD simulations of temperature-dependent graphitization kinetics in biomass-derived hard carbons. **a**, Schematic of the structural transformation pathway from disordered hard carbons to crystalline graphite, with insets showing typical atomic-scale morphologies obtained from our MD simulations. **b**, Correlation between the atomic diffusion activation enthalpy (ΔH) and the melting point (T_m) of different elements. The source data were taken from reported literature (22, 23). **c**, Temporal evolution of the fraction of sp^2 carbon at different temperatures. The blue dashed lines represent fittings based on the JMAK model, and the corresponding apparent rate constants k are shown in the inset. **d**, C–C RDF profiles at various temperatures. **e**, Number of six-membered carbon rings averaged over the final 100 ps of each MD trajectory. **f**, Final carbon network structures in simulations at various temperatures. **g**, Schematic comparison of conventional equilibrium furnace heating (top, 2700–3300 K for days) versus PCES (bottom, 3500 K for seconds) for graphitization of hard carbon precursors.

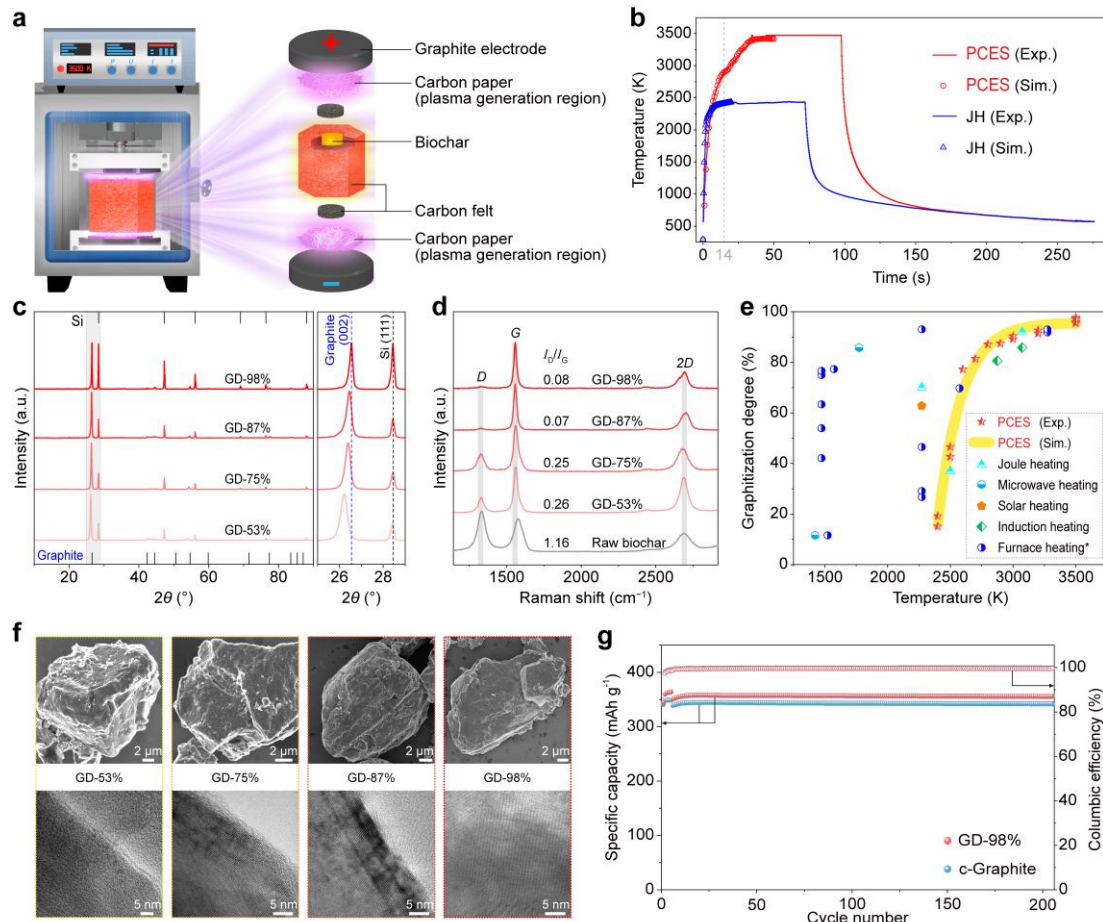


Fig. 2 | Rapid electrothermal graphitization of biochar via PCES. **a**, Schematic of contact-induced plasma generation at the intersection of vertical carbon felt fibers and horizontal carbon paper fibers. **b**, Comparative experimental and simulated temperature profiles of PCES (red) and pure Joule heating (blue) at identical power input. **c**, XRD patterns of graphitized samples (GD-53%, 75%, 87%, 98%) with Si internal standard, where the graphitization degree (GD) is quantified by the graphite (002) to Si (220) peak intensity ratio. **d**, Raman spectra of raw biochar and samples with different graphitization degrees. **e**, Benchmarking of PCES against state-of-the-art graphitization methods in terms of treatment temperature and achieved GD. **f**, SEM and corresponding high-resolution TEM images of PCES-synthesized samples at sequential graphitization stages. **g**, Long-term cycling performance and coulombic efficiency of GD-98% and commercial graphite (c-Graphite) for over 200 cycles at 0.1 C.

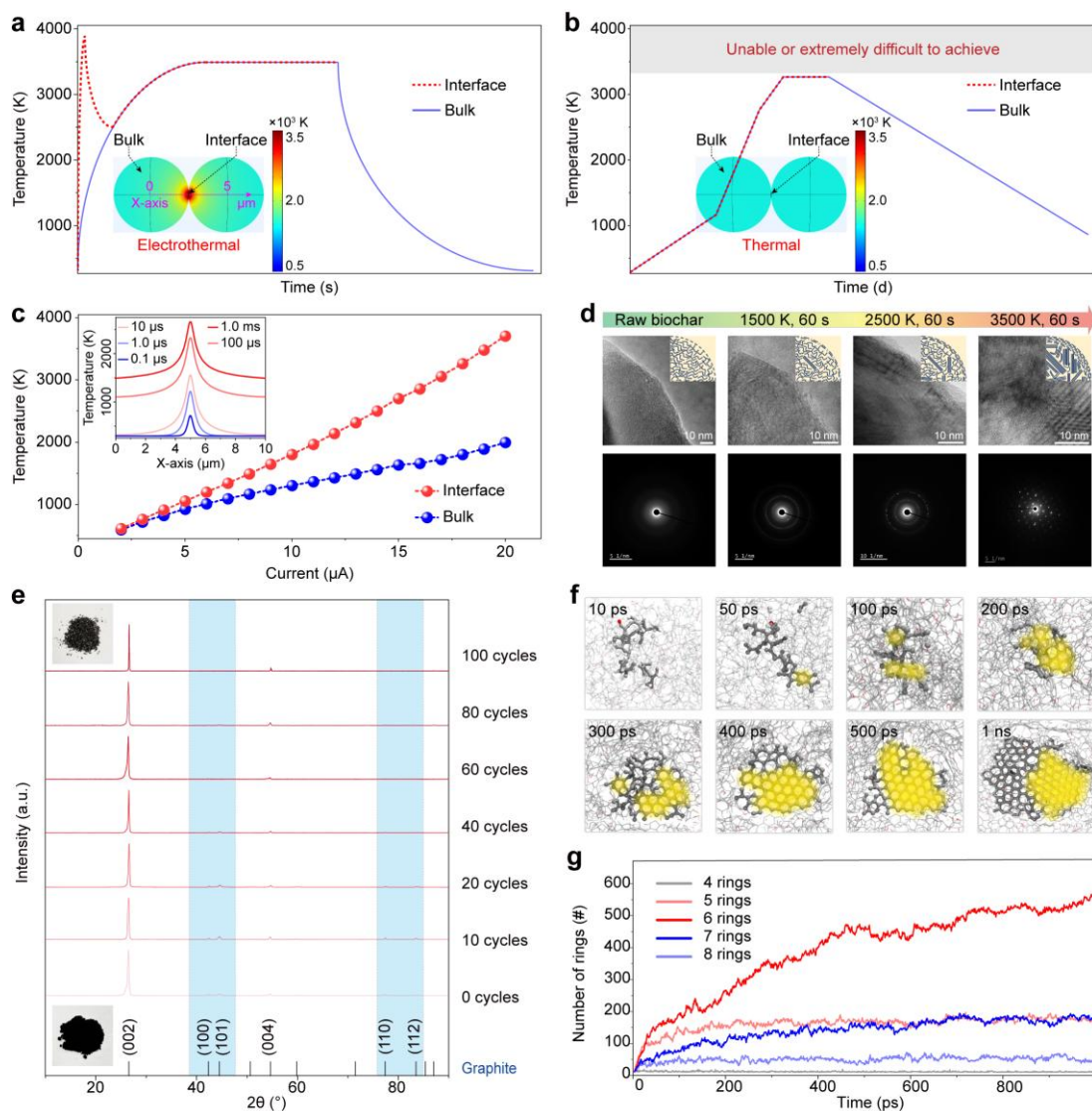


Fig. 3 | Heterogeneous edge-seeded layer growth mechanism during PCES. **a**, Temperature evolution of PCES, showing rapid heating to 3500 K in seconds, followed by quenching. Inset: Temperature distribution at the particle-particle interface, highlighting localized interfacial heating. **b**, Temperature profile of conventional equilibrium furnace heating over days. Inset: Uniform bulk temperature distribution with no interfacial enhancement. **c**, Current-dependent temperatures for interface and bulk regions. Inset: Transient temperature profiles at 0.1, 1.0, and 10 μ s. **d**, Ex-situ TEM images and corresponding SAED patterns of raw biochar and samples treated at 1500 K, 2500 K, and 3500 K for 60 s. **e**, The transformation of the material from ordinary graphite into HOPG by cycling PCES treatment. **f**, Schematic of the heterogeneous edge-seeded layer growth mechanism, illustrating epitaxial growth of graphite layers from interfacial sp^2 nuclei, with yellow highlighting the formation of hexagonal rings. **g**, Time dependent evolution of the number of different carbon ring structures in the MD simulation at 3500 K.

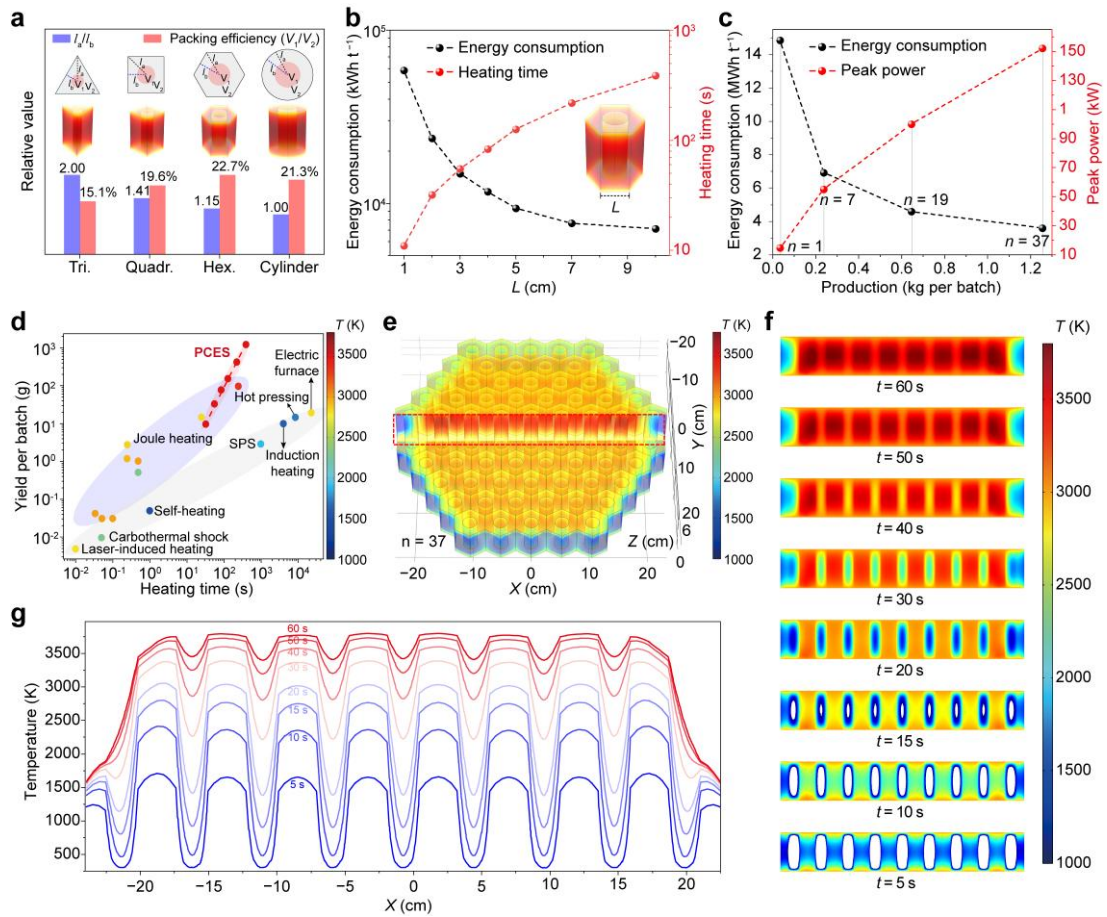


Fig. 4 | Industrial scalability and technoeconomic analysis of PCES for ton-scale graphite production. **a**, Comparison of edge length ratio (l_a/l_b) and packing efficiency (V_1/V_2) for triangular, square, hexagonal, and cylindrical heating elements. Lower l_a/l_b ratios minimize thermal gradients, while higher V_1/V_2 ratios improve space utilization. **b**, Trade-off between energy consumption and heating-up time for hexagonal prism elements of varying sizes (1–10 cm) at 3500 K. **c**, Energy consumption and peak power requirements for batch production (0.03–1.2 kg per batch) by using clustered hexagonal prism heating modules. **d**, Comprehensive comparison of batch yield, peak reaction temperature, and heating-up time between the PCES method and various heating technologies. **e**, Schematic of thermal field distribution in clustered hexagonal prism heating modules. **f**, Transient thermal profiles of a single module at 5–60 s intervals. **g**, Longitudinal cross-section of thermal field distribution in a multi-module reactor.

Supplementary Files

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