



Graphene-encased hard carbon as a high-performance anode for sodium-ion batteries

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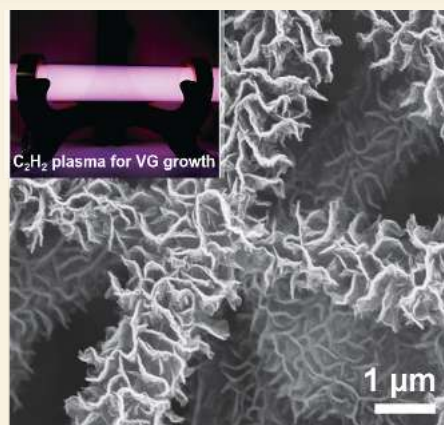
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Abstract: Hard carbon (HC) has emerged as the promising anode material for commercial sodium-ion batteries because of its low operating plateau (~ 0.1 V vs. Na^+/Na), cost-effectiveness, and the abundance of its precursors. While current research mainly focuses on pore engineering to optimize the plateau capacity, electron transport kinetics in the anodes remain largely overlooked. We address this critical aspect by uniformly grafting graphene (G) arrays onto cotton tissue-derived hard carbon (CTHC) by plasma-enhanced chemical vapor deposition. Benefiting from the more numerous and shorter electron transport channels provided by G, the electronic conductivity and ion transport capability of CTHC@G are significantly improved. The resulting CTHC@G anode has a superior sodium-storage capacity of 389.7 mAh g^{-1} , retains a capacity of 187.6 mAh g^{-1} at a high current density of 800 mA g^{-1} , and achieves an initial Coulombic efficiency of 92.17%.

Furthermore, when configured in a full cell with a $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ (NVP) cathode, the CTHC@VGNVP SIB has a high energy density of 145 Wh kg^{-1} at a power density of 1380 W kg^{-1} . This work highlights the great potential of graphene for enabling high-performance, cost-effective, and sustainable energy storage.

Key words: Sodium-ion battery; Hard carbon; Vertical graphene; Rate capability; Energy storage



1 Introduction

In recent years, sodium-ion batteries (SIBs) have gained attention as a promising alternative to lithium-ion batteries (LIBs)^[1–2]. This shift is largely fueled by growing concerns about the limited availability of lithium and its uneven distribution around the world. Lithium mining and recycling are often costly and environmentally taxing, presenting challenges for sustainable energy storage solutions. In contrast, sodium offers remarkable advantages in natural abundance, uniform distribution, and lower cost, making it particularly suitable for large-scale energy storage systems^[3–5]. Furthermore, SIBs exhibit inherent safety features, including the ability to be safely discharged to near-zero voltage, which simplifies storage and

transportation. Another significant economic benefit stems from the use of low-cost and lightweight aluminum as the anode current collector, replacing the heavier and more expensive copper foil required in LIBs^[6–7], thereby substantially reducing manufacturing costs.

Graphite, renowned for its excellent electronic conductivity and mechanical stability, is the dominant anode material in LIBs^[8]. However, it exhibits poor performance in SIBs due to a thermodynamic mis-

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match: the large ionic radius of Na^+ is incompatible with graphite's narrow interlayer spacing (0.34 nm), resulting in negligible sodium storage capacity (e.g., 20 mAh g^{-1} in ester-based electrolytes)^[9–10]. As a result, hard carbon (HC) has emerged as the most promising alternative anode, owing to its abundant precursors, low cost, and suitable operating potential^[11–13]. Hard carbon offers several advantages, including its low cost, diverse and abundant raw material sources and stable electrochemical performance. Despite these advantages, the commercialization of HC anodes is impeded by 2 major drawbacks: low initial Coulombic efficiency (ICE) and unsatisfactory rate capability. The latter directly limits the power density, restricting its application in scenarios requiring rapid charging or high-power delivery.

Scientific studies have established that the low-potential plateau capacity in hard carbons (HCs) stems from sodium cluster formation within closed nanopores. To engineer a high density of such pores, various activation agents—including MgO ^[14], MgCl_2 ^[15], KOH ^[16], CO_2 ^[17], ZnO ^[18], $\text{Zn}(\text{Ac})_2$ ^[19] and ethanol^[20], have been utilized to etch carbon precursors such as cellulose^[21–22], carbohydrates^[23–24], wood^[25–26], starch^[27], and resins^[28–29] during pyrolysis. While these modification strategies effectively enhance the sodium-storage capacity of HC, they further exacerbate its poor rate capability, which is inherently limited by insufficient electronic conductivity and sluggish ion transport. Consequently, developing electrode structures with simple fabrication feasibility and improving their electronic conductivity have become a core strategy to meet the demands of commercial applications.

Cotton tissue has recently emerged as a promising precursor for hard carbon anodes in sodium-ion batteries, owing to its inherent flexibility, self-supporting nature, low cost, and global availability^[30]. When subjected to direct carbonization, the resulting cotton tissue-derived hard carbon (CTHC) preserves the interconnected fibrous microstructure of the original material, which provides excellent mechanical integrity and eliminates the need for binders or conductive additives^[13,31]. This approach not only simpli-

fies electrode fabrication, but also represents an eco-friendly, low-waste pathway for sustainable material production. To further enhance its properties, here we integrated vertical graphene (VG) nanosheets onto the CTHC fiber surface by plasma-enhanced chemical vapor deposition (PECVD), forming a hierarchical CTHC@VG composite. The vertically aligned graphene architecture substantially increases the electroactive surface area and introduces abundant active sites, while simultaneously improving electronic conductivity across the electrode^[32–36]. This synergistic coupling facilitates rapid Na^+ adsorption and diffusion kinetics. As a result, the CTHC@VG anode exhibits a superior initial coulombic efficiency of 92.16% and a high reversible capacity of 389.7 mAh g^{-1} in ether-based electrolytes—significantly surpassing that of pristine CTHC (277.1 mAh g^{-1}). Moreover, it maintains a remarkable capacity of 187.6 mAh g^{-1} even at 800 mA g^{-1} , demonstrating superior rate capability.

2 Experiment

2.1 Material synthesis

Commercial cotton tissue was pre-oxidized in air at 200 °C (heating rate: 1 °C·min^{−1}) and subsequently carbonized in an Ar atmosphere at 1300 °C for 2 h (heating rate: 3 °C·min^{−1}) to obtain tissue-derived hard carbon (CTHC) paper.

For the CTHC@VG composite, pre-oxidized tissue was first carbonized at 800 °C for 2 h under Ar flow. Vertical graphene (VG) arrays were then grown in situ on the carbonized tissue via plasma-enhanced chemical vapor deposition (PECVD) using acetylene as the carbon source and Ar as the carrier gas. Specifically, the carbonized substrate was placed in a PECVD tube furnace, heated to 500 °C under vacuum (5 Pa), and then exposed to a $\text{C}_2\text{H}_2/\text{Ar}$ mixture (1 : 1 v/v, 20 mL·min^{−1}). The plasma was ignited at 500 W and maintained for 30 min. The resulting CTHC@VG sample was collected after natural cooling to room temperature.

2.2 Material characterization

Crystal structure was analyzed by X-ray diffrac-

tion (XRD, Bruker D8 Advance) with Cu $K\alpha$ radiation ($\lambda = 0.15418$ nm). Morphology was observed using scanning electron microscopy (SEM, SU 3500) at 15 kV. Raman spectra were acquired on a WITec alpha 300 R system with a 532 nm laser. Specific surface areas were determined from nitrogen adsorption–desorption isotherms using the Brunauer–Emmett–Teller (BET) method. Surface electronic states were examined by X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi). Electronic conductivity was measured with a four-point probe resistivity tester (ST2258C).

2.3 Electrochemical measurements

Freestanding CTHC and CTHC@VG were directly used as anodes with an active mass loading of $\sim 1.2 \text{ mg}\cdot\text{cm}^{-2}$. CR2032 coin cells were assembled with glass fiber (Whatman) as the separator and $1 \text{ mol}\cdot\text{L}^{-1}$ NaPF₆ in diglyme as the electrolyte. Galvanostatic charge–discharge (GCD) and galvanostatic intermittent titration technique (GITT) tests were conducted on a LAND battery test system between 0.01 and 2.5 V (vs. Na⁺/Na). For GITT, cells were discharged at $25 \text{ mA}\cdot\text{g}^{-1}$ for 20 min, followed by a 2 h relaxation period, repeated until the potential reached ≤ 0.01 V.

According to GITT results, D_{Na^+} is calculated using the following equation,

$$D_{\text{Na}^+} = \frac{4}{\pi\tau} \left(\frac{m_{\text{B}} V_{\text{m}}}{M_{\text{B}} S} \right)^2 \left(\frac{\Delta E_{\text{S}}}{\Delta E_{\tau}} \right)^2$$

where τ is the relaxation time, m_{B} is the mass of active material, M_{B} is the molar mass of active material, V_{m} is the molar volume of hard carbon, S is the surface area of active material, ΔE_{S} and ΔE_{τ} can be obtained from GITT curves. Electrochemical impedance spectroscopy (EIS) tests were performed on a Reference 3000 Gamry with 0.1 Hz–100 kHz and an amplitude of 0.5 mV. All tests were conducted at room temperature.

3 Results and discussion

The synthesis procedures for both CTHC and CTHC@VG are detailed in the Experimental section. Flexible self-supporting CTHC paper was fabricated

through pre-oxidation in air at 200 °C, followed by high-temperature annealing at 1300 °C under an argon atmosphere. In contrast, the preparation of CTHC@VG involved an additional step: prior to annealing, VG arrays were directly grown on the tissue-derived microfibers by PECVD (Fig. 1a). VG arrays constitute a distinctive graphene architecture characterized by sheets aligned perpendicularly to the substrate. This configuration offers several advantages over conventional planar graphene, including a significantly enlarged surface area, abundant exposed edges that serve as active sites, efficient vertical electron transport pathways, and improved mechanical flexibility and structural integrity^[32]. Notably, in this work, the aligned VG arrays were synthesized at a moderate temperature of 500 °C (Fig. 1b), which substantially lowers energy consumption compared to traditional CVD processes typically requiring temperatures exceeding 1000 °C^[37–38].

SEM characterization was employed to examine the morphology of CTHC and CTHC@VG (Fig. 1c–h). The CTHC sample maintains the self-interwoven mesh architecture of its precursor (Fig. S1), composed of numerous micron-scale fibers entangled into highly flexible, cross-linked microbands. This robust network underpins the material's excellent mechanical properties, allowing the papers to be repeatedly bent or folded without structural failure (Fig. S2). In the case of CTHC@VG (Fig. 1f–h), SEM images reveal dense and uniform VG coatings grown along individual hard carbon fibers. These VG structures, with their extended edges and interconnected open network, effectively enhance the electron transport kinetics within the hard carbon matrix.

Transmission electron microscopy (TEM) reveals distinct nanostructural differences between CTHC and CTHC@VG. As shown in Fig. 2a–b, CTHC exhibits a conventional hard carbon architecture characterized by localized short-range structural ordering, abundant defect sites, and a significant fraction of isolated closed pores. Clearly discernible are tightly stacked carbon layers with an interlayer spacing of 0.409 nm. In contrast, CTHC@VG exhibits a

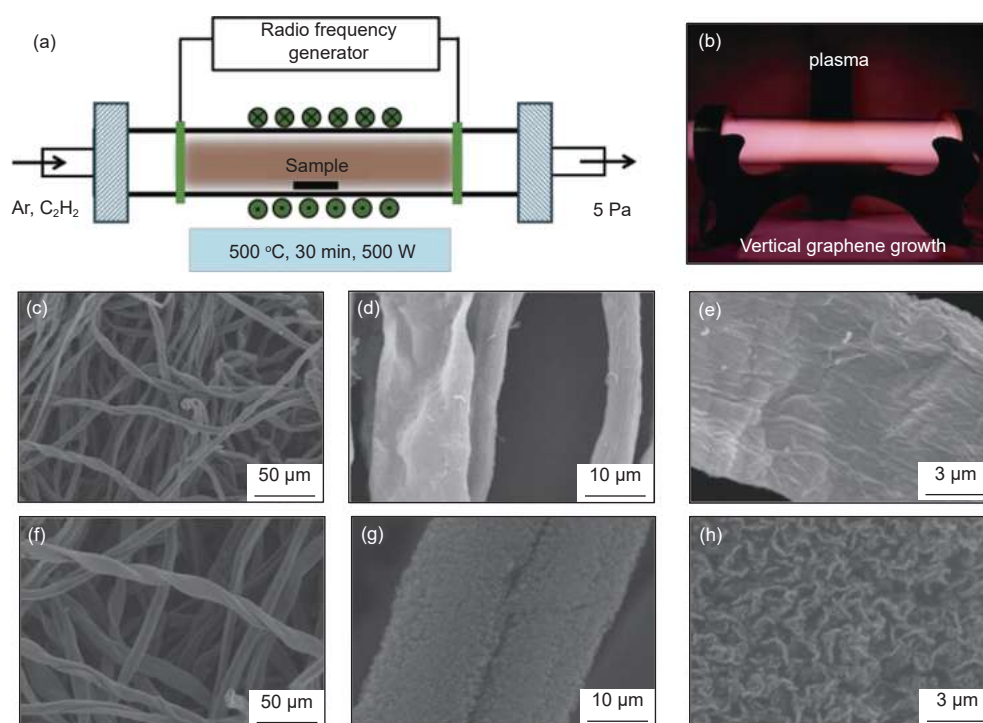


Fig. 1 Structural characterizations. (a) Illustration of the VG growth process on carbon fibers *via* PECVD. (b) Digital photograph of the C_2H_2/Ar plasma environment during VG synthesis. (c-e) SEM images of the CTHC paper. (f-h) SEM images revealing the dense coverage of VG nanosheets on the individual fibers of CTHC@VG

high proportion of short graphite-like domains with an interlayer spacing of only 0.349 nm (Fig. 2c-e). Compared to CTHC@VG, CTHC demonstrates a lower degree of graphitization, consisting primarily of disordered carbon layers with smaller crystallite sizes (L_a and L_c). Meanwhile, the VG component introduced via PECVD exhibits a highly graphitized layer structure, featuring larger L_a and L_c values and reduced interlayer spacing (Fig. 2f). The carbon microcrystalline structures were further probed by X-ray diffraction (XRD). As depicted in Fig. S3, the (002) diffraction peak of CTHC@VG shifts toward a lower angle relative to that of CTHC, indicating an increased average interlayer spacing (d-spacing) following VG integration. This expansion is likely attributable to the defect-rich nature of the VG framework. Moreover, the (002) peak intensity in CTHC@VG is lower than in CTHC, suggesting differences in stacking order and a reduced number of stacked layers in the graphene-enhanced composite. This tailored nanostructure in CTHC@VG is conducive to facilitating faster sodium-ion transport, a highly desirable trait for sodium-ion battery (SIB) applications^[39].

Raman spectroscopy was employed to probe the structural evolution from CTHC to CTHC@VG, as presented in Fig. 2g. Both samples exhibit the characteristic *D*-band ($\sim 1340\text{ cm}^{-1}$) and *G*-band ($\sim 1580\text{ cm}^{-1}$), which are attributed to the breathing mode of A_{1g} symmetry in disordered carbon structures and the in-plane stretching vibration of E_{2g} phonons in sp^2 -hybridized carbon domains, respectively. A notable decrease in the intensity ratio I_D/I_G was obtained from 1.189 for CTHC to 1.171 for CTHC@VG, indicating an enhanced degree of graphitization in the latter. This trend is consistent with the TEM observations, confirming that the introduction of VG effectively improves the structural ordering of the carbon matrix.

The surface chemical composition and bonding states of CTHC and CTHC@VG were further analysed by X-ray photoelectron spectroscopy (XPS). Survey spectra (Fig. S4a-b) confirm that both materials are predominantly composed of carbon and oxygen, with CTHC@VG exhibiting a marginally higher oxygen content. High-resolution deconvolution of the C 1s spectrum (Fig. S4c-d) reveals three component

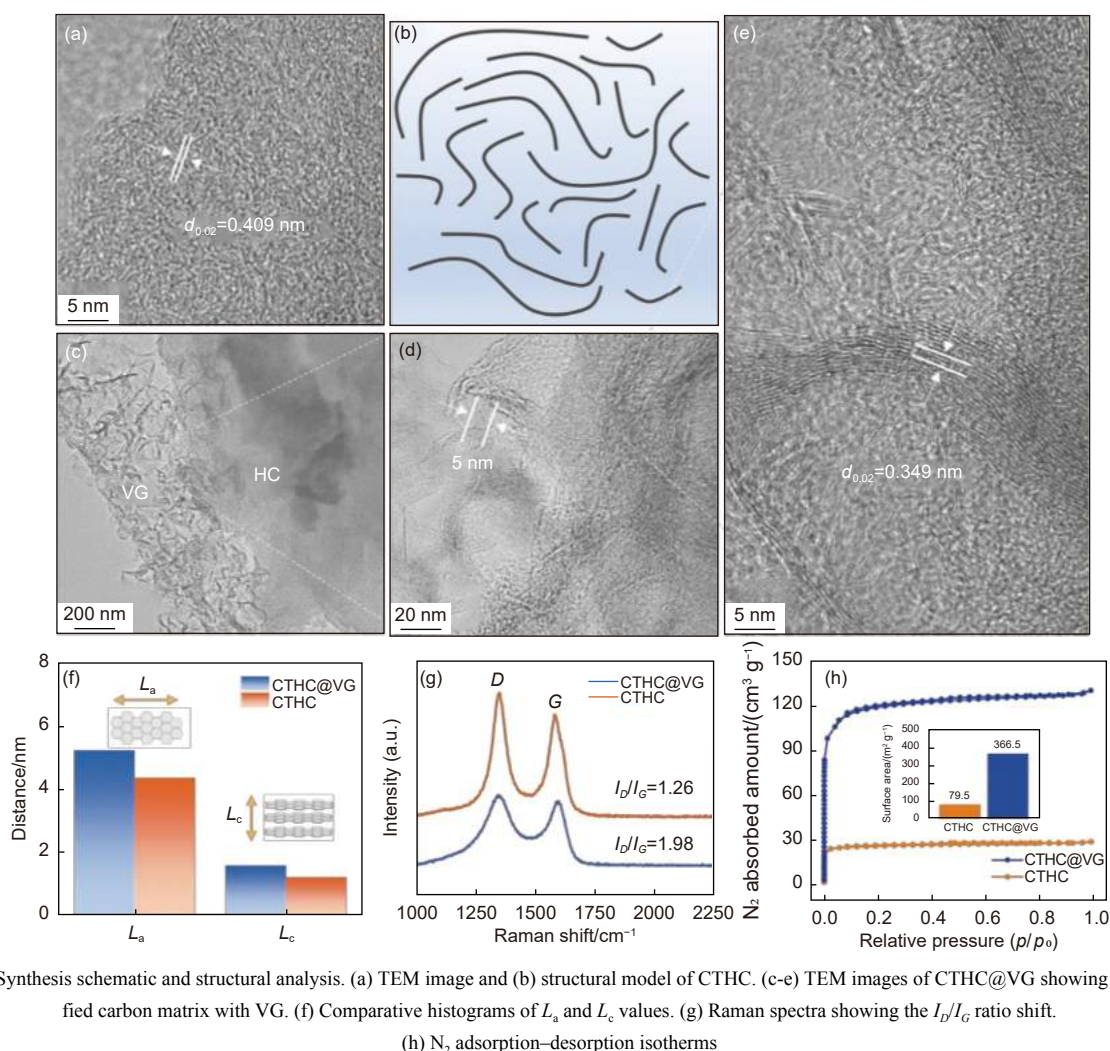


Fig. 2 Synthesis schematic and structural analysis. (a) TEM image and (b) structural model of CTHC. (c-e) TEM images of CTHC@VG showing the modified carbon matrix with VG. (f) Comparative histograms of L_a and L_c values. (g) Raman spectra showing the I_D/I_G ratio shift. (h) N_2 adsorption-desorption isotherms

peaks located at 284.8, 286.3 and 289.4 eV, corresponding to C—C/C=C, C—O and C=O bonds, respectively. Notably, the relative content of C—O groups in CTHC@VG is higher than that in CTHC, suggesting that the PECVD process facilitates the formation of oxygen-containing functional groups such as epoxy/ether bonds, which are known to contribute to enhanced sloping capacity in sodium-ion storage. This finding is corroborated by the O 1s spectra (Fig. S4e-f), where oxygen species are identified in the forms of C=O, C—O and —OH. Compared to CTHC, CTHC@VG displays increased proportions of C=O and —OH groups, which are expected to provide additional active sites for sodium ion adsorption, thereby further improving the sloping capacity.

To investigate the effect of VG on the surface

area of HC, N_2 adsorption-desorption measurements were performed. The N_2 adsorption-desorption isotherm of CTHC@VG exhibits a type-I isotherm (Fig. 2h), indicating the surface area of CTHC@VG is measured to be $366.5 m^2 g^{-1}$, which is larger than CTHC ($79.5 m^2 g^{-1}$), which confirms that VG significantly increases the contact area with the electrolyte. The abundant surface edges of vertically grown graphene provide fast transport pathways for sodium-ion migration to boost the plateau capacity. Meanwhile, the conformal VG coating acts as a barrier that blocks electrolyte solvent molecules from penetrating into the internal micropores of hard carbon. This dual effect suppresses irreversible side reactions and thus yields a high ICE.

To evaluate the effect of VG on sodium storage performance, half-cells were configured using CTHC

and CTHC@VG as working electrodes, sodium metal as the counter/reference electrode, and 1 mol L⁻¹ NaPF₆ in diglyme as the electrolyte. Galvanostatic charge-discharge (GCD) profiles measured at a low current density of 25 mA g⁻¹ (Fig. 3a) for both electrodes exhibit 2 characteristic regions: a sloping region above 0.1 V, attributed to Na⁺ adsorption on defect sites and functional groups, and a plateau region below 0.1 V, associated with Na⁺ intercalation into graphitic-like layers. Notably, CTHC@VG delivers a significantly higher reversible capacity of 389.7 mAh g⁻¹ compared to 277.1 mAh g⁻¹ for CTHC, along with an improved ICE of 92.17% versus 86.11% (Fig. 3b). The enhanced ICE of CTHC@VG is mainly attributed to the uniformly grown VG on its surface, which reduces the direct contact between the electrolyte and hard carbon matrix, thereby alleviating excessive electrolyte decomposition and the formation of solid-electrolyte interphase (SEI). This mechanism is corroborated by TEM analysis (Fig. S6), which shows that the SEI formed on CTHC@VG after three cycles is notably smoother and more uniform than that on pristine CTHC. More importantly, CTHC@VG exhibits increased capacity contributions in both the sloping and plateau regions relative to CTHC

(Fig. 3c). This comprehensive performance enhancement is attributed to the synergistic effects of the PECVD process. The in-situ plasma environment not only catalyzes the growth of VG on the hard carbon fibers but also induces controlled plasma etching, which generates abundant micropores and expands the internal surface area accessible to Na⁺. The resulting hierarchical structure—comprising a highly conductive VG network and a modified porous carbon matrix—collectively facilitates faster ion transport, provides additional active sites for Na⁺ adsorption, and improves the reversibility of intercalation, thereby boosting the overall sodium storage capacity.

Fig. 3d compares the rate capabilities of CTHC and CTHC@VG electrodes across increasing current densities. While both electrodes experienced a capacity decrease with escalating current density, CTHC@VG demonstrated a markedly more gradual decay, sustaining significantly higher capacities than CTHC at all tested rates from 25 to 800 mA g⁻¹. This superior kinetics is further highlighted at an ultra-high current density of 800 mA g⁻¹, where CTHC@VG delivered a capacity of 187.6 mAh g⁻¹, which is approximately three times that of CTHC (55.9 mAh g⁻¹). The exceptional reversibility and structural resilience of

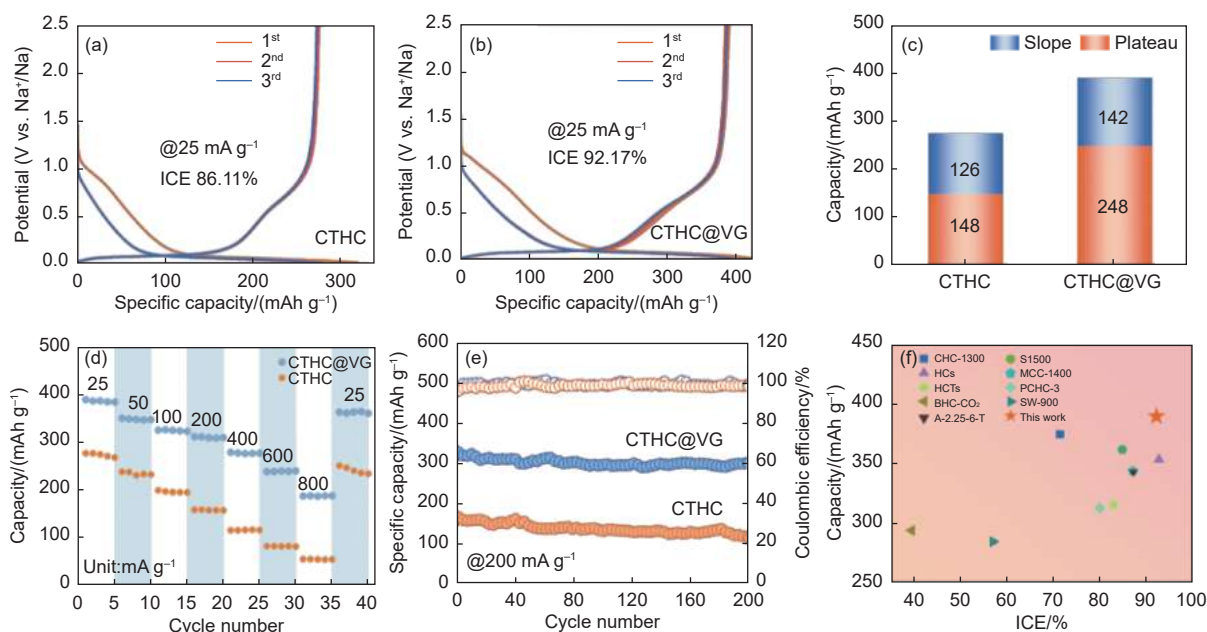


Fig. 3 Na⁺-storage performance of CTHC and CTHC@VG. (a, b) GCD curves at 25 mA g⁻¹. (c) Comparison of the plateau and sloping capacity based on the 2nd discharge/charge curves. (d) Comparison of rate capability. (e) Comparison on 200 cycles of CTHC@VG and CTHC at 200 mA g⁻¹. (f) The comparison of specific capacity and ICE of CTHC@VG with previous reported hard carbon anodes

CTHC@VG are confirmed as the specific current density was restored to 25 mA g^{-1} , with its capacity recovering nearly to the original value. This outstanding rate performance and recovery ability are attributed to the unique architecture of CTHC@VG: the vertically aligned graphene network provides a highly efficient electron transport pathway. To distinguish the distinct effects of argon plasma etching and VG growth on the materials, the electrochemical performances of electrospun hard carbon fibers (HCF) carbonized at identical temperature and Ar plasma-treated HCF (HCF-PAr) are added as reference controls in the Fig. S7. In contrast, HCF-PAr delivers a marginal improvement in sodium storage capacity relative to HCF ($302.5\text{--}351.2 \text{ mAh g}^{-1}$), its rate performance slightly deteriorates because the abundant newly formed porous structure on the surface impedes electronic conductivity and ion transport. Nevertheless, Ar plasma exerts a moderate auxiliary function during the PECVD growth of VG. Its mild etching generates abundant nucleation sites for VG deposition and reinforces interfacial bonding between VG and the carbon substrate. This result further verifies that the VG conductive network dominates the exceptional rate capability of CTHC@VG and synergistically boosts its sodium storage performance.

The long-term cycling stability of CTHC and CTHC@VG electrodes was further evaluated at a current density of 200 mA g^{-1} over 200 cycles. As illustrated in Fig. 3e, the CTHC@VG electrode demonstrates significantly enhanced cycling durability, retaining a reversible capacity of 304.1 mAh g^{-1} with a high-capacity retention of 87.3%. Throughout the cycling test, the charge-discharge plateaus remained stable, and the Coulombic efficiency was consistently maintained at nearly 100%, indicating highly reversible sodium storage behavior. In contrast, the CTHC electrode suffered from capacity degradation, declining from an initial 168.9 to 120.9 mAh g^{-1} after 200 cycles, corresponding to a retention rate of only 71.5%. The superior performance of CTHC@VG can be attributed to its robust hierarchical architecture, in which the vertically aligned graphene nanosheets not

only enhance electrical conductivity, but also effectively buffer volume variations during repeated Na^+ insertion/extraction. This tailored structure ensures mechanical integrity and continuous electron transport pathways, thereby mitigating active material degradation and enabling stable cycling.

To elucidate the kinetic and interfacial properties of the electrodes, a comprehensive analysis was conducted through electronic conductivity measurements, electrochemical impedance spectroscopy (EIS), and galvanostatic intermittent titration technique (GITT). The free-standing CTHC@VG film exhibited a notable enhancement in electronic conductivity, achieving 71.43 S m^{-1} —approximately 1.5 times higher than that of the pristine CTHC (28.57 S m^{-1}) (Fig. 4a). This significant improvement is attributed to the in situ growth of vertical graphene (VG) arrays on the carbon fiber surface by PECVD, which constructs an interconnected conductive network that facilitates efficient electron transport throughout the electrode.

Further interfacial kinetics were probed using EIS, with the Nyquist plots fitted using an appropriate equivalent circuit model (Fig. 4b). The spectra consist of a semicircle in the high-to-medium frequency region, corresponding to charge transfer resistance (R_{ct}), and a linear segment in the low-frequency region representing solid-state ion diffusion. The R_{ct} value of the CTHC@VG electrode was determined to be 9.52Ω , indicating favorable charge transfer kinetics. Moreover, the low-frequency region of the CTHC@VG electrode displays a steeper slope compared to that of CTHC, suggesting enhanced Na^+ diffusion behavior. The VG architecture not only provides continuous electron pathways but also guides rapid ion transport, contributing to reduced interfacial resistance and more reversible sodiation/desodiation processes. The minimal variation in the diffusion slope after cycling further confirms the structural robustness of CTHC@VG, which effectively maintains ion diffusion pathways during repeated cycling.

The sodium-ion transport kinetics of CTHC and CTHC@VG electrodes were quantitatively evaluated using GITT at a current density of 25 mA g^{-1} . The

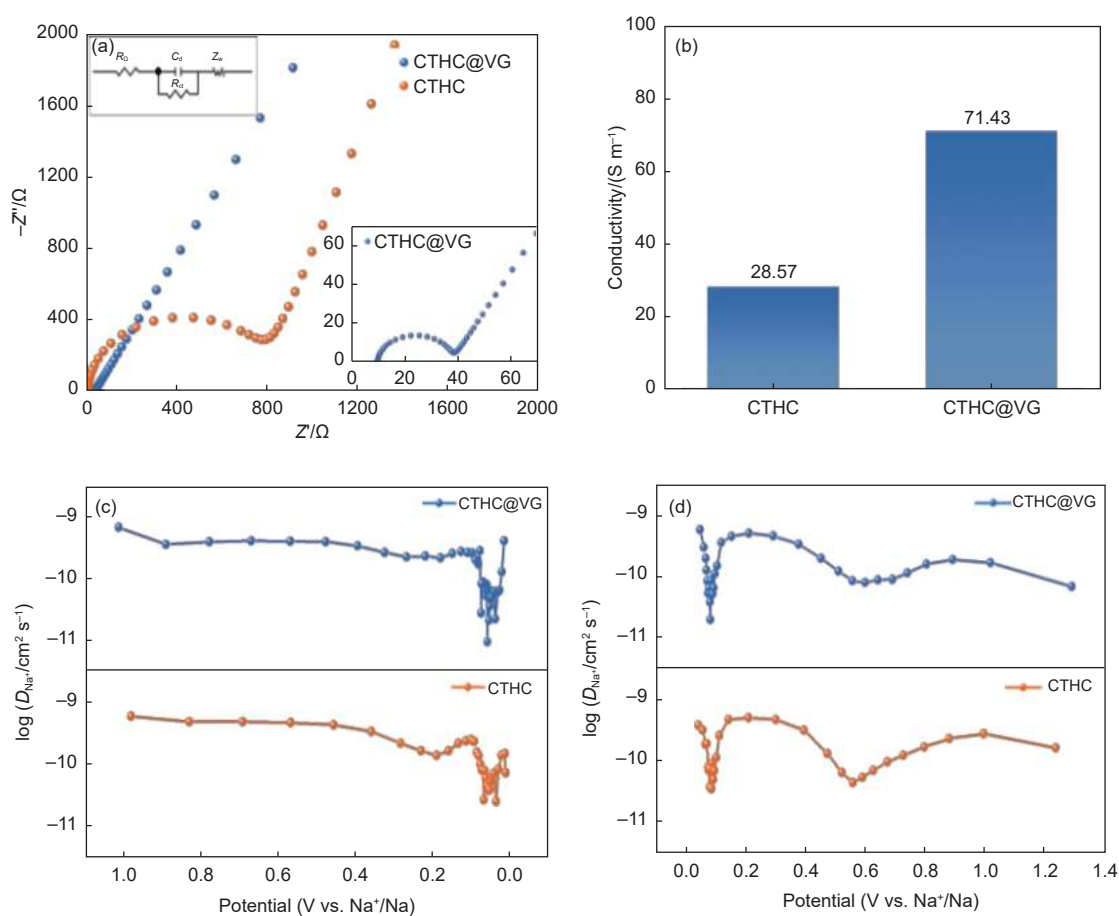


Fig. 4 Kinetic and interfacial analysis. (a) Electrical conductivity comparison. (b) EIS Nyquist plots with equivalent circuit (inset) and a magnified high-frequency region. (c, d) GITT-derived Na^+ diffusion coefficients during sodiation and de-sodiation

corresponding sodium-ion diffusion coefficients (D_{Na^+}) during sodiation and desodiation were derived from GITT profiles (Fig. S8) and are presented in Fig. 4c-d. Throughout the sloping region, both electrodes exhibit relatively stable and high D_{Na^+} values, indicative of rapid Na^+ adsorption on defect sites and carbon surfaces. In contrast, a pronounced decrease in D_{Na^+} is observed within the low-potential plateau region, which can be attributed to the slower solid-state diffusion process as Na^+ ions intercalate into the graphitic-like domains. Notably, this is followed by a distinct rebound in D_{Na^+} near 0.1 V, signaling the onset of pore-filling processes within the closed nanopores—a mechanism associated with the formation of quasi-metallic sodium clusters. Importantly, the CTHC@VG electrode demonstrates consistently higher D_{Na^+} values than CTHC across both the sloping and plateau regions, underscoring the role of the vertical graphene network in facilitating ion accessibility and

enhancing overall ionic diffusivity. This improved kinetic behavior aligns well with the superior rate performance observed in CTHC@VG and highlights its effectiveness in promoting fast and stable sodium storage.

To assess the practical viability of the CTHC@VG anode, a full sodium-ion battery (SIB) was constructed by coupling it with a $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ (NVP) cathode (Fig. 5a). The structural and electrochemical characteristics of the synthesized NVP are provided in Fig. S9. An optimal anode-to-cathode mass ratio ($m_{\text{CTHC@VG}} : m_{\text{NVP}} = 1 : 3$) was determined based on their respective specific capacities to maximize full-cell performance. Herein, the mass loading of NVP electrodes ranges from 3.6 to 4 mg, and full cells are tested within a voltage window of 1.5–3.8 V. The energy density is calculated based on the total active mass of both cathode and anode electrodes. During charging, Na^+ ions deintercalate from NVP and mi-

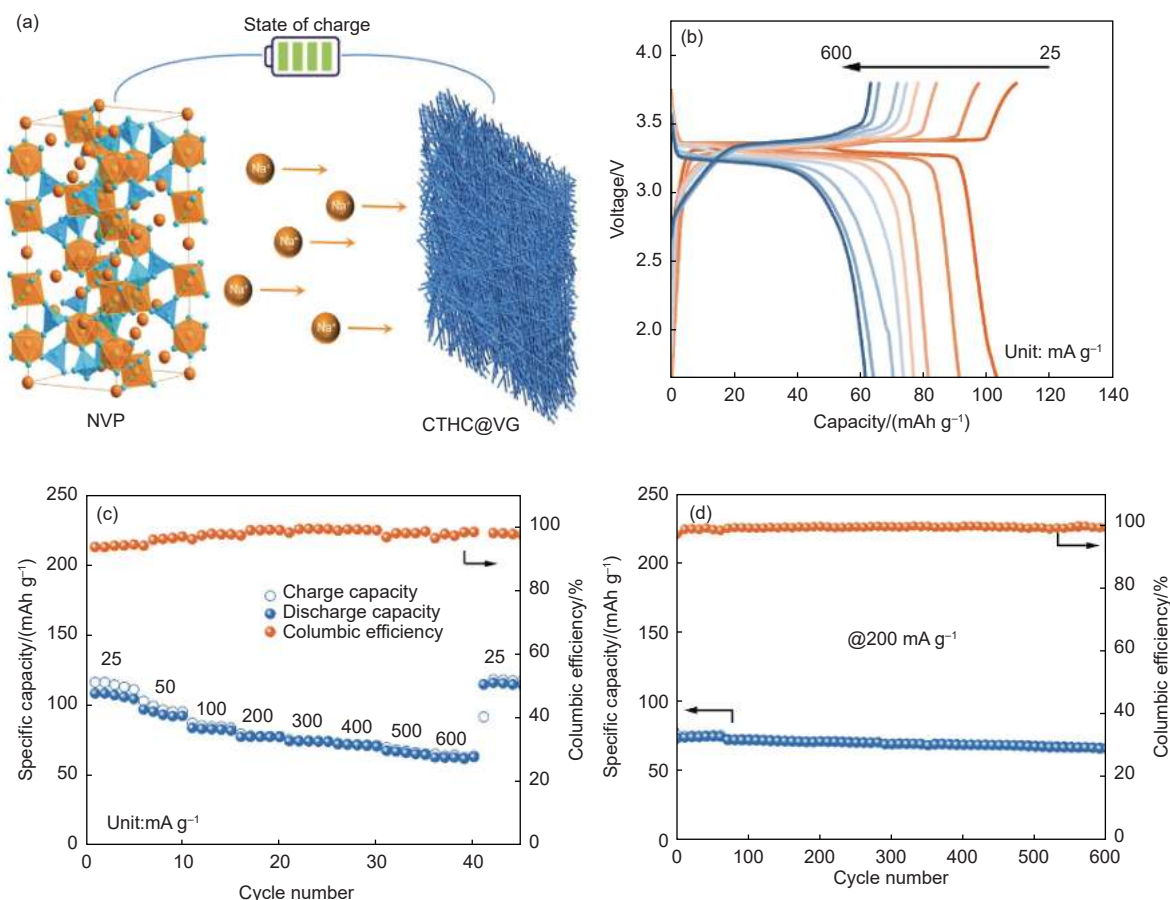


Fig. 5 Full SIB device performance: (a) Schematic of the full configuration, where the CTHC@VG anode is coupled with the $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ cathode during the charge process. (b) Charge/discharge curves of CTHC@VG||NVP SIBs at various current densities. (c) Rate performance of CTHC@VG||NVP SIBs at various current densities. (d) Cycling performance of CTHC@VG||NVP SIBs at 0.2 A g^{-1}

grate to the CTHC@VG anode, where they are stored, while PF_6^- anions adsorb onto the NVP surface. During discharge, this process reverses, converting chemical energy into electrical output. The full cell exhibits stable charge-discharge profiles with an average operating voltage of approximately 3.31 V (Fig. 5b). The rate performance of the CTHC@VG||NVP full cell was evaluated across current densities ranging from 25 to 600 mA g^{-1} (Fig. 5c). At 25 mA g^{-1} , the cell delivers a specific capacity of 116.7 mAh g^{-1} . Notably, even at a high current density of 600 mA g^{-1} , well-defined charge-discharge plateaus are retained, and a reversible capacity of 63 mAh g^{-1} is achieved, underscoring the excellent kinetics of the CTHC@VG anode. Furthermore, the full cell demonstrates outstanding cycling stability, retaining 90.5% of its initial capacity after 600 cycles (Fig. 5d). In terms of energy and power density, the device achieves 268.4 Wh kg^{-1} at a power density of 54.5 W kg^{-1} .

Even under demanding conditions (1380 W kg^{-1}), it maintains a high energy density of 145 Wh kg^{-1} , highlighting its potential for high-performance, practical sodium-ion storage systems.

4 Conclusion

In summary, this work demonstrates a significant advancement in sodium-ion battery technology through the rational design of a tissue-derived hard carbon anode integrated with vertical graphene nanosheets (CTHC@VG) by plasma-enhanced chemical vapor deposition. This unique architecture effectively addresses the persistent challenges of low ICE and limited rate capability in conventional hard carbon anodes. The optimized CTHC@VG anode achieves an exceptional ICE of 92.16% and superior high-rate performance (389.7 mAh g^{-1}), attributable to its robust interfacial stability, enhanced electronic

conductivity, and facilitated sodium-ion transport kinetics. When configured in a full cell with an $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ cathode, the CTHC@VG ||NVP system delivers a high energy density of 145 Wh kg^{-1} at a power density of 1380 W kg^{-1} , underscoring its practical potential. Our study provides a scalable and energy-efficient manufacturing strategy for developing high-performance SIBs, paving the way toward more economical and sustainable energy storage systems.

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