

Article

High-Performance Cotton-Derived Carbon Fibers as Next Generation Anode Materials for Lithium-Ion Batteries

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Abstract

The increasing demand for lithium-ion batteries has intensified the search for sustainable alternatives to conventional graphite anodes. In this work, cotton-derived carbon fibers were prepared from commercial medical-grade cotton wool using a two-step pyrolysis process and investigated as hard-carbon anode materials for lithium-ion batteries. The structural and morphological properties of the prepared material were analyzed using X-ray diffraction, scanning electron microscopy, and transmission electron microscopy, revealing a predominantly amorphous carbon structure with a retained fibrous morphology after pyrolysis. Electrochemical performance was evaluated in CR2032 half-cells by cyclic voltammetry, electrochemical impedance spectroscopy, and galvanostatic charge–discharge measurements. The prepared hard carbon exhibited characteristic lithium-storage behaviour with irreversible processes during the initial cycle followed by stable reversible cycling. The material delivered a reversible capacity of approximately 780–800 mAh g^{−1} after 300 cycles at a current density of 100 mA g^{−1}, together with stable Coulombic efficiency and good rate capability. Post-mortem analysis confirmed that the electrode structure remained stable after repeated cycling. The obtained results demonstrate the potential of waste cotton as a renewable precursor for the preparation of high-performance hard-carbon materials and highlight the applicability of biomass-derived carbons for sustainable electrochemical energy-storage systems.

Keywords: cotton-derived carbon fibers; pyrolysis; anode material; lithium-ion batteries



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1. Introduction

Lithium-ion batteries (LIBs) are widely used as rechargeable power storage devices due to their high power capacity, high energy density, long cycle life, and minimal memory effect [1]. Their extensive deployment in portable electronics, electric vehicles, and grid-scale energy storage has led to a rapidly increasing demand for electrode materials, especially graphitic carbon used as the conventional anode [2,3].

This growing reliance on graphite presents significant economic, environmental, and geopolitical challenges. The extraction and processing of natural graphite are associated with high energy consumption, intensive water use, and significant environmental impacts associated with extraction and purification processes [4].

In general, the production of graphite anode materials is widely recognized as a major contributor to the overall life cycle footprint of LIBs, primarily due to their energy-intensive processing [5]. This is also reflected in the case of synthetic graphite. A recent life cycle assessment of synthetic graphite anode production in the United States reported greenhouse gas emissions of 29.7 kg CO₂ per kg of battery anode material (BAM), along with water consumption of 121 L kg⁻¹ and an energy demand of 580 MJ kg⁻¹. The process requires extremely high temperatures (typically above 2500 °C), and the graphitization step was identified as a major hotspot, contributing up to 74% of total environmental impacts [5].

From a supply chain perspective, graphite production is highly geographically concentrated, with China accounting for approximately 75–80% of global natural graphite mine production and an even larger share of downstream processing capacity [6,7]. This dominance becomes more pronounced at the processing stage, where China is responsible for 87% of graphite refining into battery-grade material production, including the manufacture of spherical graphite used in lithium-ion battery anodes [8]. As a result, the graphite value chain is significantly more concentrated than many other critical raw materials, with the top three producing countries accounting for nearly the entire global supply (94%) [7].

Recent analyses by the International Energy Agency highlight that demand for graphite is rapidly increasing due to electrification trends, particularly in electric vehicles and energy storage systems, with continued growth expected under all energy transition scenarios [8].

This rapid demand expansion, combined with highly localized production and processing, creates structural vulnerabilities in the supply chain, as widely recognized for critical materials systems [9]. Disruptions in a single dominant region—whether due to trade restrictions, policy changes, or market shifts—can propagate through global battery manufacturing systems, leading to supply insecurity and price volatility [8,9]. The strong asymmetry between globally distributed demand and regionally concentrated processing capacity reinforces strategic dependency on a limited number of suppliers, posing a significant challenge to the long-term resilience and sustainability of lithium-ion battery supply chains [7,9].

In addition to anode-related challenges, the broader reliance of LIB technology on inorganic and critical raw materials introduces further environmental and economic concerns, including the impacts associated with end-of-life battery waste and the rising cost and limited availability of cobalt-based cathode materials [10]. These combined factors highlight the urgent need for the development of sustainable, low-cost, and widely available alternatives to conventional electrode materials.

Fortunately, the recent push for active use of renewable resources prompted researchers to investigate bio-based materials, where plant-derived materials have been shown to be a promising alternative for graphite in the manufacturing of carbon-based anode materials [11]. Such bio-based precursors undergo heat treatment without or with restricted access to oxygen in a process called pyrolysis. Already investigated materials so far include, e.g., apple waste—285 mAh g⁻¹, rice husks—679.9 mAh g⁻¹, rice straw—200 mAh g⁻¹, banana fibers or peels—954 mAh g⁻¹, tea leaves—363 mAh g⁻¹, natural potato starch—531 mAh g⁻¹, and human hair—370 mAh g⁻¹ [12–18]. Using renewable resources also means investigation of material utilisation of waste that is rich in carbon, such as waste tire rubber, plastic bag waste, or unburned carbon concentrates [19]. Unlike crystalline graphite, where lithium storage mainly proceeds through staged intercalation to form LiC₆ with a theoretical capacity of 372 mAh g⁻¹, hard carbon generally stores lithium through a combination of processes related to its disordered structure. Depending on precursor chemistry and synthesis conditions, biomass-derived hard carbons may contain turbostratically stacked graphene-like domains, enlarged interlayer spacing, edge sites,

surface heteroatoms, and porous regions. These structural features are commonly associated with lithium storage through a probable combination of adsorption, insertion into disordered carbon layers, and pore-related storage. Consequently, precursor chemistry and pyrolysis conditions play an important role in tailoring the structure and electrochemical behaviour of biomass-derived hard carbons [20–22].

During high-temperature pyrolysis, carbon-rich materials obtained from these precursors undergo graphitization, leading to the formation of artificial graphite structures [23]. However, such high-temperature pyrolysis is energy intensive, which is why the focus shifted to lower-temperature pyrolysis (<1500 °C). Materials created during that process can be segregated into two groups: hard carbon and soft carbon.

The difference between the two groups is that hard carbon is a non-graphitizable carbon material and cannot create an orderly graphitic structure even at temperatures reaching 3000 °C or even higher [21]. This non-graphitizable character is commonly associated with cross-linked carbon structures formed during pyrolysis, which can inhibit the rearrangement and growth of graphitic layers. Depending on precursor chemistry and heat-treatment conditions, such structural constraints may also contribute to the development of disordered domains, defects, and porosity [21,24].

Soft carbons, on the other hand, are typically produced through a liquid-phase carbonization process in which liquid crystal precursor molecules undergo rearrangement and polycondensation during pyrolysis. As a result, the resulting carbon structure contains fewer cross-linked nanostructures and can be more easily graphitized at high temperatures [25].

In this work, we investigate cotton-derived carbon fibers as a sustainable and low-cost precursor for the preparation of hard carbon anode materials via pyrolysis. Commercial medical-grade cotton wool, consisting of high-purity cellulose, was used without further treatment as a model biomass precursor. Owing to its availability, purity, and well-defined composition, this precursor is a promising candidate for developing carbon materials with tunable structure and electrochemical properties. This study, therefore, aims to evaluate the potential of cotton-derived carbon fibers as an alternative anode material for LIBs.

2. Results and Discussion

2.1. Structural and Morphological Characterization

The X-ray diffraction (XRD) pattern of the cotton-derived carbon fibers is presented as the black curve in Figure 1. The recorded diffraction pattern clearly confirms a highly disordered (amorphous) structure of the pyrolyzed carbon, characterized by a low-intensity, broad diffuse scattering profile rather than well-defined crystalline reflections. Such behaviour is typical for turbostratic or poorly ordered carbon materials, where the absence of long-range order suppresses the formation of sharp Bragg peaks [26,27].

To distinguish the intrinsic signal of the sample from the background contribution of the sample holder (plexiglass), a separate measurement of the holder was performed and is shown as the orange curve in Figure 1. The comparison indicates that most of the low-angle scattering is significantly influenced by the holder, emphasizing the necessity of background correction in the analysis of low-crystallinity carbon systems.

In the diffraction pattern of the cotton-derived carbon fibers sample, only a single weak Bragg reflection is observable at approximately $2\theta \approx 33\text{--}35^\circ$, marked in Figure 1. Based on its position, it can tentatively be assigned to a trigonal phase of CaCO_3 (COD 1010962). The presence of this phase is attributed to residual calcium-containing mineral species naturally present in the cotton precursor and retained during the carbonization process. No post-synthesis acid-leaching treatment was applied, as the objective of this work was to maintain a simple and scalable preparation route with minimal processing

steps. Although only a single diffraction peak attributable to CaCO_3 was observed and the overall material exhibits a predominantly amorphous character [28], this tentative phase assignment is further supported by the TEM/SAED and EDS analyses discussed below.

Overall, the XRD analysis demonstrates that the cotton-derived carbon fibers consist predominantly of amorphous carbon with only trace amounts of crystalline phases, which is consistent with the expected structural evolution of pyrolyzed carbon fibers at relatively moderate processing temperatures [26].

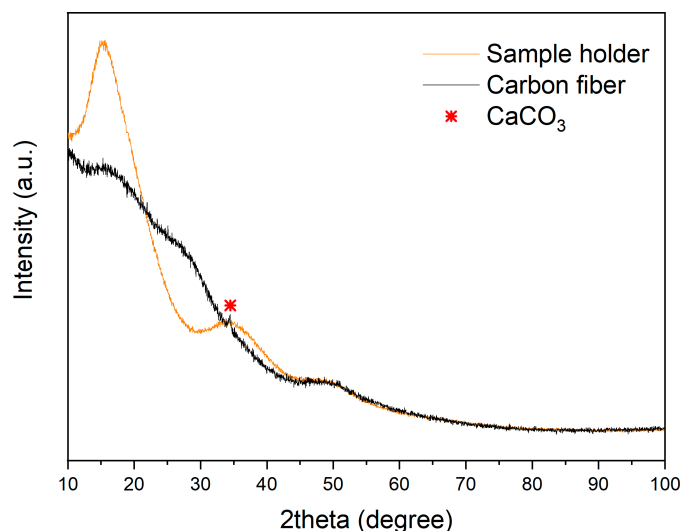


Figure 1. XRD patterns of cotton-derived carbon fibers and plexiglass sample holder, highlighting the predominantly amorphous structure and background contribution.

The morphology of the cotton-derived carbon fibers after pyrolysis was investigated by SEM at different magnifications, as shown in Figure 2a–c. The material retains a fibrous structure characteristic of the original cotton precursor, although partial shrinkage and surface roughening are observed as a result of thermal decomposition. The fibers exhibit a relatively disordered fibrous structure, which is typical for biomass-derived hard carbons and can facilitate electrolyte penetration and lithium storage [29,30].

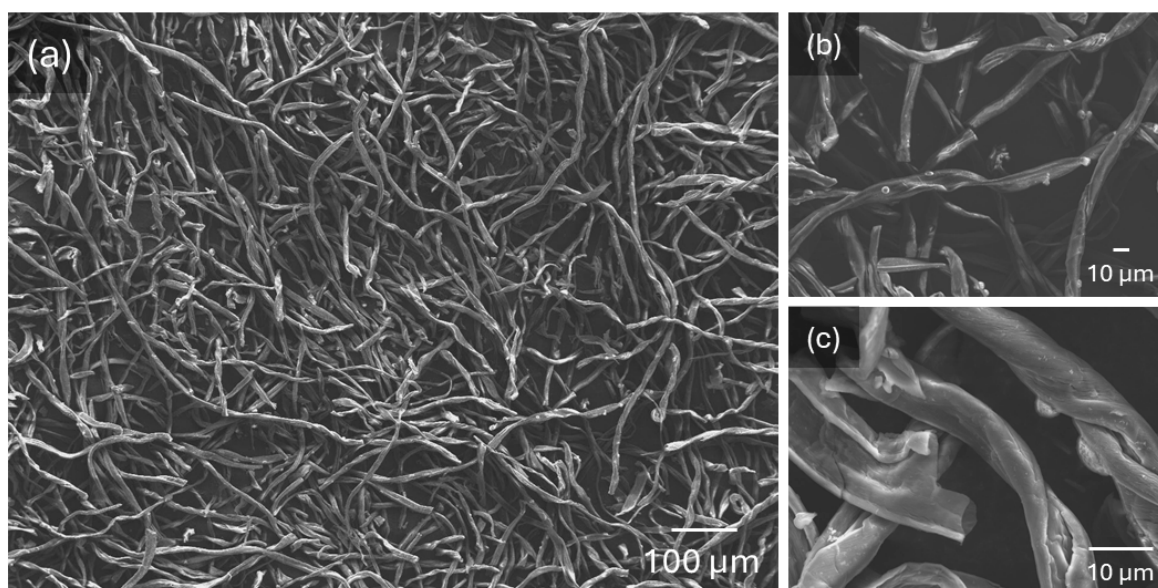


Figure 2. SEM images of cotton-derived carbon fibers after pyrolysis, demonstrating the retained fibrous structure at different magnifications (a–c).

Elemental analysis revealed that carbon is the major component of the material, with a content of 49.2 wt.%. A significant oxygen contribution (35.8 wt.%) was also detected, indicating the presence of oxygen-containing species within the carbonized material and/or inorganic oxygen-bearing phases formed or retained during pyrolysis. In addition, calcium was identified at 12.8 wt.%, while sodium and magnesium were present only in minor amounts of 1.8 wt.% and 0.4 wt.%, respectively. The presence of calcium is consistent with the XRD results, where a weak diffraction feature was tentatively assigned to a CaCO_3 phase. These inorganic species may originate from residual mineral impurities naturally present in the cotton precursor or from the thermal treatment process.

EDS elemental mapping in Figure 3a–d demonstrates a relatively homogeneous distribution of carbon and oxygen throughout the fibrous structure. The oxygen signal follows the morphology of the cotton-derived fibers, confirming the presence of oxygen-containing species within or on the carbonized material. However, EDS does not provide information on the bonding state of oxygen; therefore, the presence of specific oxygen-containing surface functional groups cannot be directly confirmed from these data. Calcium is present as finely dispersed domains distributed across the material, without the formation of large crystalline agglomerates, which is consistent with the low-intensity CaCO_3 related feature observed in the XRD pattern. These results indicate that the pyrolyzed cotton-derived material retains a chemically heterogeneous but structurally continuous fibrous network.

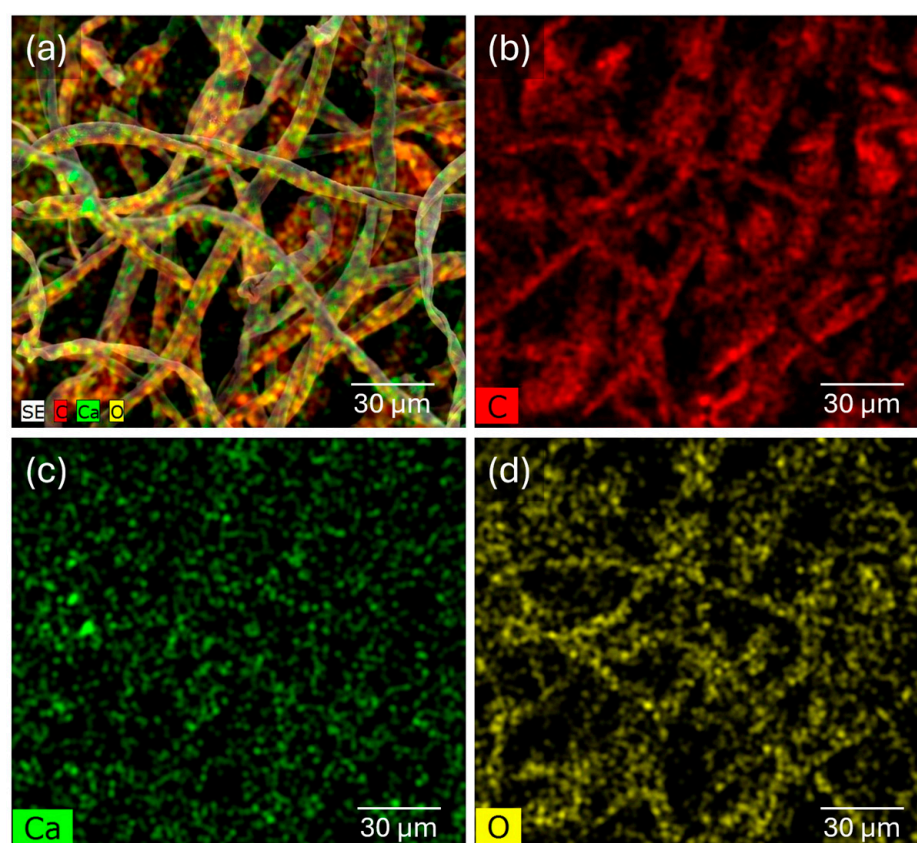


Figure 3. EDS elemental mapping of cotton-derived carbon fibers after pyrolysis (a–d).

The relatively high oxygen content (35.8 wt.%) is expected to influence the electrochemical behavior of the material during the initial lithiation process. Previous studies have shown that oxygen-containing species in carbonaceous anodes can participate in electrolyte decomposition and SEI formation, resulting in irreversible lithium consumption and reduced initial Coulombic efficiency [31]. In biomass-derived hard carbons, the initial irreversible capacity has also been associated with oxygen-containing surface groups and

lithium trapping within microporous domains, both of which contribute to capacity loss during the first cycle [32]. However, recent studies indicate that the effect depends not only on the total oxygen content but also on the nature of the oxygen species present. While reactive oxygen functionalities may enhance parasitic reactions and SEI growth, specific oxygen-related bonding environments can contribute to the formation of a thinner and more stable SEI layer, thereby improving the initial Coulombic efficiency [33].

Overall, the SEM observations with EDS elemental mapping results indicate that the pyrolyzed cotton-derived fibers maintain a disordered morphology that may be favorable for electrochemical applications [29].

The substructure of the cotton-derived carbon fibers and the fine particles present on their surface were investigated by transmission electron microscopy. Figure 4a shows a detail of a carbon fiber dispersed on an ultrathin carbon film, while Figure 4b presents the corresponding SAED pattern. The diffuse diffraction halo observed in the SAED pattern is consistent with the XRD results and confirms the predominantly amorphous structure of the carbon fibers. Figure 4c shows a particle located on the fiber surface. Based on its contrast and morphology, the particle appears to be polycrystalline or composed of an agglomerate of crystallites. This assumption is confirmed by the corresponding SAED pattern shown in Figure 4d, which exhibits characteristic diffraction rings indicative of a polycrystalline material. The measured diffraction rings were indexed to the CaCO_3 phase (COD 1010962), in agreement with the XRD results, while its chemical composition was further verified by EDS analysis.

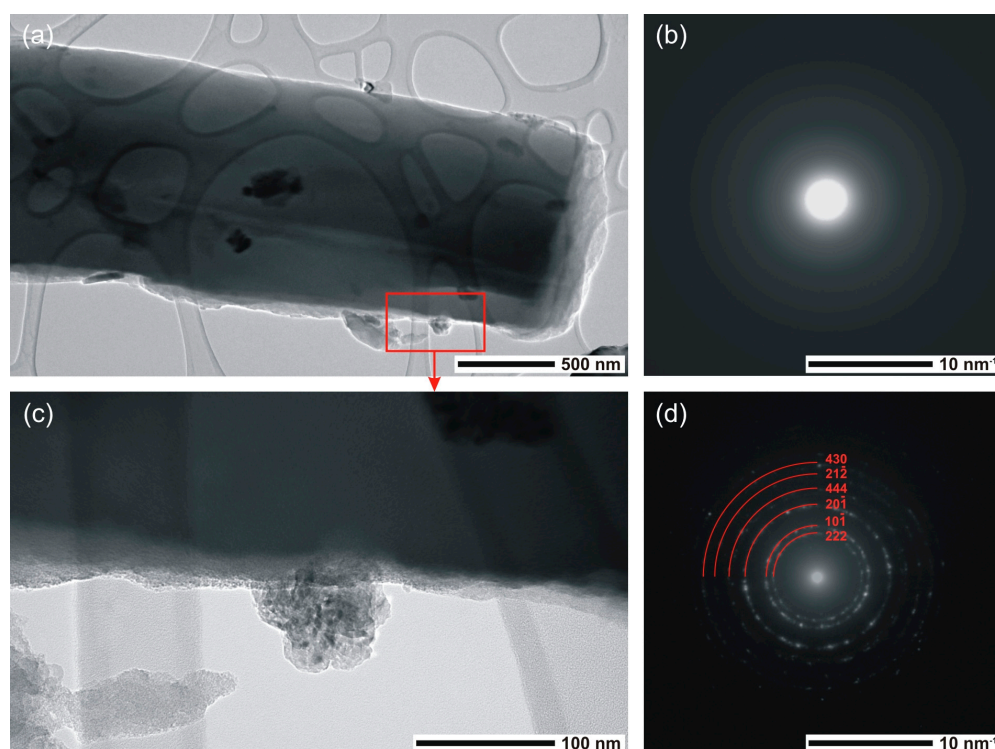


Figure 4. Bright-field TEM images and corresponding SAED patterns of a cotton-derived carbon fiber after pyrolysis (a,b) and a fine polycrystalline particle located on the fiber surface (c,d).

The structural features observed in the cotton-derived carbon fibers suggest that the material may be favourable for lithium storage even without additional physical or chemical activation. Cotton consists mainly of cellulose fibers with a naturally hierarchical fibrous morphology. During pyrolysis, dehydration, depolymerization, and aromatization of cellulose can generate a disordered carbon framework that may contain turbostratic graphene-

like domains, edge defects, residual oxygen-containing species, and micro-/nanoporous regions. Because the final treatment temperature was limited to 1000 °C and the holding time was short, excessive carbon-layer ordering and pore collapse were likely restricted, allowing the material to retain a predominantly disordered hard-carbon structure. These structural characteristics can be associated with additional lithium-storage sites and may contribute to lithium storage through a probable combination of adsorption at defect/edge sites, insertion into disordered carbon layers, and pore-related storage in nanoporous domains [21,24–26].

2.2. Electrochemical Performance

The electrochemical behaviour of the prepared cotton-derived carbon fibers half-cell was first evaluated by cyclic voltammetry (CV) in the potential range of 0.01–3.0 V vs. Li/Li⁺ at a scan rate of 0.1 mV s^{−1} (Figure 5a). During the first cathodic sweep, a pronounced irreversible reduction feature is observed at approximately 0.75 V, which disappears in subsequent cycles. This feature is attributed to electrolyte decomposition and the formation of the solid electrolyte interphase (SEI), along with other irreversible interfacial reactions occurring during the formation cycle. Such behaviour is characteristic of hard carbon materials and contributes to their low initial Coulombic efficiency [20,34,35]. This effect may be further enhanced by the relatively high oxygen content (35.8 wt.%) detected in the present material, since oxygen-containing species in carbonaceous anodes have been reported to promote electrolyte decomposition and SEI formation during the first lithiation cycle.

The first cycle also exhibits a significantly higher cathodic current in the low-potential region below 0.2 V, indicating additional irreversible lithium consumption that may be associated with SEI formation, irreversible interfacial reactions, and lithium trapping in structurally disordered regions of the carbon matrix. From the second cycle onward, the CV curves become much more similar, indicating improved reversibility and stabilization of the electrode/electrolyte interface after the formation cycle [20,34].

The broad and overlapping electrochemical features observed in the CV curves are characteristic of the disordered microcrystalline structure of hard carbon and suggest the presence of multiple lithium-storage environments rather than a single sharp phase transition [20,21,34–36]. Recent studies on pitch-derived hard carbon further suggest that the low-voltage region (0–0.2 V) is commonly associated with lithium storage within pores, whereas the intermediate-voltage region (0.2–0.9 V) is related to lithium insertion into interlayer spaces and the higher-voltage region (0.9–2.5 V) to interactions with edge-associated sites [37]. The broad and overlapping nature of the CV response suggests that the individual lithium-storage processes are not sharply separated, but rather occur as partially overlapping contributions. Therefore, the observed CV behaviour is consistent with the presence of multiple lithium-storage environments typically reported for disordered hard-carbon materials.

The galvanostatic charge–discharge profiles (Figure 5b) exhibit electrochemical behaviour typical of hard-carbon materials, including a sloping voltage region and a low-voltage region. The first cycle shows a significantly higher discharge capacity, which is attributed mainly to irreversible lithium consumption arising from SEI formation and other interfacial reactions. Additional irreversible lithium trapping in structurally disordered regions of the carbon matrix may also contribute to this behaviour. This response is consistent with the well-known low initial Coulombic efficiency of hard carbon materials. In subsequent cycles, the charge–discharge curves become progressively stabilized, indicating improved reversibility after the formation cycle. The sloping voltage profile and low-voltage region are generally consistent with the electrochemical response of disor-

dered hard-carbon materials. In the literature, these regions are often related to lithium adsorption at surface or edge sites, insertion into disordered carbon layers, and pore-related storage [20,35,36,38]. The disordered structure of hard carbon is generally associated with a distribution of lithium-storage environments and overlapping electrochemical processes rather than a single well-defined phase transition, which can contribute to voltage hysteresis [20,34,36]. Accordingly, the observed voltage profile is interpreted as a probable result of partially overlapping storage processes typical of hard-carbon materials, rather than as a set of individually resolved mechanisms. The cycling performance and Coulombic efficiency of the hard-carbon anode are shown in Figure 5c. The Coulombic efficiency increases rapidly during the initial cycles, rising from approximately ~85–90% in the first cycles to ~98–100% after ~5–10 cycles, and subsequently stabilizes at values close to unity. This behaviour indicates highly reversible lithium-storage processes after the formation stage and is characteristic of hard carbon systems with stabilized electrode/electrolyte interfaces and reduced parasitic reactions.

The evolution of the specific capacity (Figure 5c) reveals an initial discharge capacity of approximately ~500 mAh g⁻¹, followed by a slight decrease during the early cycles to ~350–380 mAh g⁻¹, and subsequently a continuous gradual increase during prolonged cycling. After 300 cycles at a current density of 100 mA g⁻¹, the reversible capacity reaches approximately ~780–800 mAh g⁻¹, indicating a remarkably high lithium-storage capability and progressive electrochemical activation of the hard-carbon structure. This behaviour is consistent with progressive electrode conditioning during cycling, which may involve improved electrolyte wetting, gradual stabilization of the electrode/electrolyte interface, and improved charge-transfer kinetics [39]. These processes can increase the electrochemically accessible fraction of the electrode during prolonged cycling. Therefore, the gradual capacity increase is interpreted as a probable consequence of progressive electrode conditioning and improved electrochemical accessibility, rather than being assigned to one specific structural process. The reversible capacity achieved after prolonged cycling exceeds the theoretical capacity of graphite (372 mAh g⁻¹), indicating that lithium storage in the cotton-derived carbon fibers cannot be explained solely by conventional graphite-like intercalation [23]. This behaviour can be discussed in the context of the fundamentally different lithium-storage mechanism generally reported for hard carbon materials compared with crystalline graphite [20,21,40]. In graphite, lithium storage is mainly limited by the formation of LiC₆, whereas hard carbons commonly exhibit a disordered structure that may include turbostratic carbon domains, enlarged interlayer spacing, surface or edge sites, and porous regions, depending on the precursor and synthesis conditions.

According to the adsorption-intercalation-pore-filling model commonly proposed for hard carbons, lithium storage may involve adsorption at surface or edge sites, insertion between disordered carbon layers, and pore-related storage within closed or partially accessible nanopores [20,21,35,39,40]. The electrochemical behaviour observed in the present work is consistent with such a multi-site lithium-storage concept. In the present cotton-derived carbon fibers, the amorphous XRD pattern, diffuse SAED response, retained fibrous morphology, and oxygen signal detected by EDS suggest a structurally disordered and chemically heterogeneous carbon material. These features may contribute to the observed progressive electrochemical activation during cycling. Accordingly, the capacity above graphite is discussed as a probable result of multiple, partially overlapping lithium-storage processes typical of hard carbon materials, rather than as direct evidence of one specific storage mechanism [20,21,35,39,40].

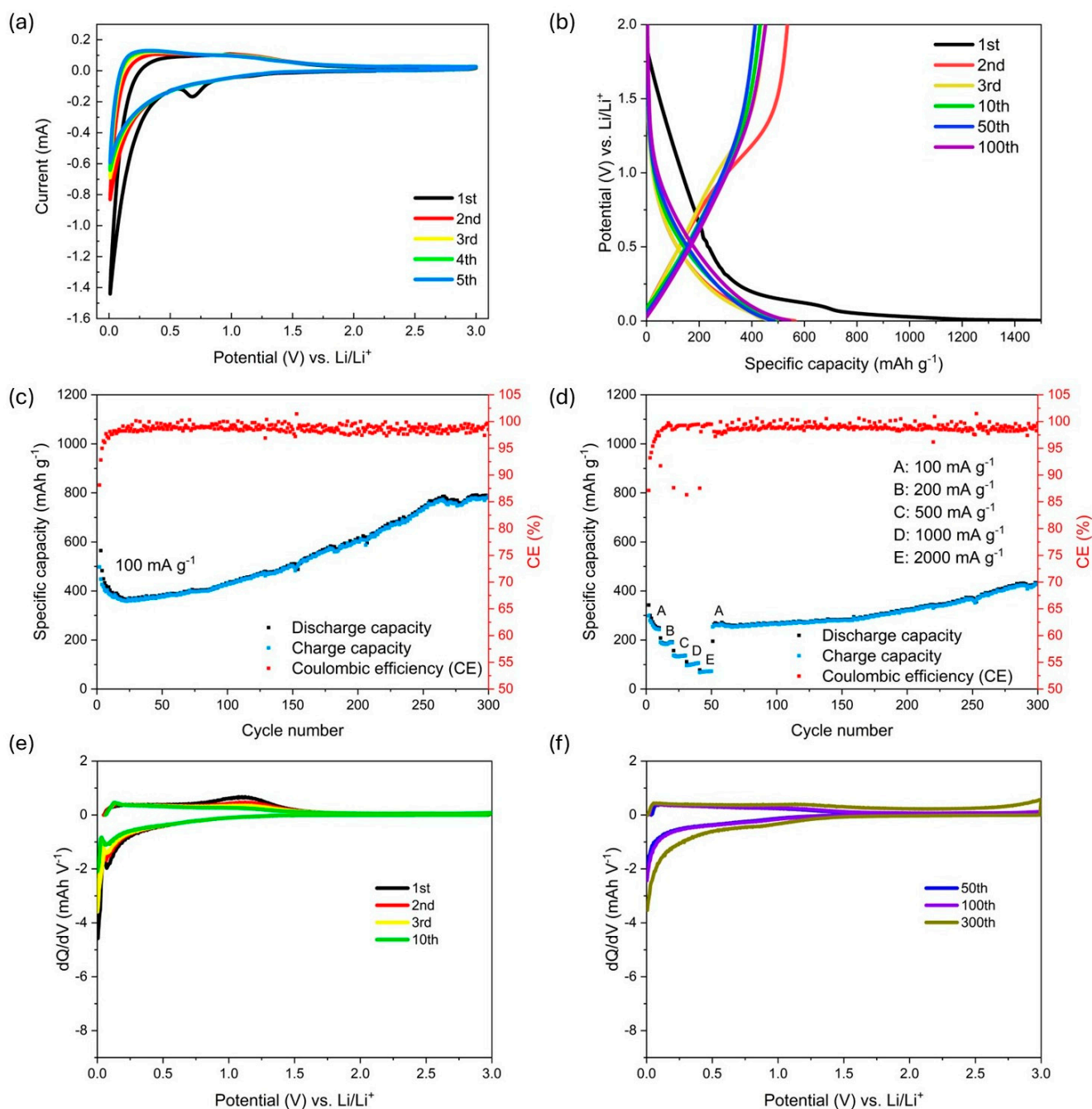


Figure 5. Cyclic voltammetry (CV) curves recorded at a scan rate of 0.1 mV s^{-1} in the potential window of $0.01\text{--}3.0 \text{ V}$ vs. Li/Li^+ (a), electrochemical performance of cotton-derived carbon fibers in lithium half-cells: galvanostatic charge-discharge profiles illustrating typical hard carbon behaviour (b), cycling stability at 100 mA g^{-1} (c), and long-term rate capability evaluated at different current densities ($100, 200, 500, 1000$, and 2000 mA g^{-1}), with 10 cycles at each current density followed by regeneration at 100 mA g^{-1} for 100 cycles (d), and differential capacity (dQ/dV) profiles during initial cycling (e) and prolonged cycling (f).

As summarized in Table 1, the present cotton-derived carbon fibers show competitive lithium-storage performance compared with biomass-derived carbon anodes tested at the same current density of 100 mA g^{-1} . Compared with waste makeup cotton-derived hollow carbon, the present material delivers a substantially higher reversible capacity after 300 cycles under identical current-density conditions. The present material also outperforms several non-doped biomass-derived carbons prepared from avocado seed, coffee oil

and coconut oil under comparable testing conditions. The main advantage of the present material is therefore the combination of a simple two-step pyrolysis route, untreated cotton precursor, absence of chemical activation, templating or doping steps, retained fibrous morphology and high long-term reversible capacity. It should also be noted that literature comparisons are influenced by electrode formulation, voltage window, mass loading, capacity normalization and cycling protocol; therefore, the table is intended as a qualitative comparison under similar current-density conditions rather than an absolute ranking.

Table 1. Comparison of biomass-derived carbon anodes tested at 100 mA g^{−1}.

Precursor	Treatment Route	Capacity	Advantages	Limitations	Ref.
Cotton waste	Two-step pyrolysis up to 1000 °C	780–800 mAh g ^{−1} after 300 cycles	Simple route, waste cotton precursor, retained fibrous morphology, high reversible capacity	Low initial Coulombic efficiency, high O/Ca content	This work
Waste makeup cotton	Direct pyrolysis/carbonization in N ₂ at 1000 °C	450 mAh g ^{−1} after 300 cycles	Cotton-derived waste precursor, hollow morphology, stable cycling	Lower reversible capacity than this work, low initial Coulombic efficiency	[41]
Avocado seed	Pyrolysis, no doping at 750 °C	315 mAh g ^{−1} after 100 cycles	Non-doped biomass carbon, simple waste precursor, stable capacity comparable to graphite	Lower capacity than this work; shorter cycling test	[42]
Coffee oil	Dry autoclaving at 700 °C	274 mAh g ^{−1} after 250 cycles	Non-doped biomass/waste precursor, stable cycling	Lower capacity than this work	[43]
Coconut oil	Incineration and piranha solution treatment	250 mAh g ^{−1} after 90 cycles	Biomass-derived carbon, same current density	Uses harsh post-treatment; lower capacity and shorter cycling	[44]

The rate capability of the hard-carbon anode was evaluated by varying the current density from 100 to 2000 mA g^{−1} (Figure 5d). The electrode exhibits a gradual decrease in capacity with increasing current density, followed by nearly complete recovery upon returning to lower current, indicating good rate performance and structural stability [45,46]. At elevated current densities, the specific capacity decreases from approximately ~300 mAh g^{−1} at 100 mA g^{−1} to ~70–100 mAh g^{−1} at 2000 mA g^{−1}, reflecting increasing kinetic limitations for lithium-ion diffusion and charge transfer within the disordered carbon framework. Nevertheless, after returning to 100 mA g^{−1}, the reversible capacity recovers to approximately ~270–290 mAh g^{−1}, confirming the structural robustness and reversibility of the electrode material.

The higher reversible capacity observed during long-term cycling (Figure 5c), compared with the capacity recovered after the rate-capability test (Figure 5d), may be attributed to differences in kinetic limitations under high current densities and progressive electrode conditioning during prolonged low-current cycling. Prolonged cycling at low current density may improve electrolyte wetting, stabilize the SEI layer, and gradually increase the electrochemically accessible fraction of the disordered carbon electrode. In contrast, the high-current conditions used during rate-capability testing impose kinetic limitations on lithium diffusion and charge transfer, which may suppress slower storage processes. However, without direct pore-structure analysis, the role of nanoporous storage sites cannot be confirmed [47].

The differential capacity (dQ/dV —Figure 5e,f) profiles provide further insight into the evolution of the lithium-storage behaviour during cycling. In the initial cycles, the dQ/dV response shows a weak low-potential feature around 0.03 V, which becomes less pronounced and is no longer clearly resolved after extended cycling. Apart from this minor feature, no sharp or well-defined peaks are observed, indicating the absence of distinct two-phase reactions or graphite-like staging transitions. Instead, the capacity is distributed over a broad potential range, which is consistent with the disordered nature of hard-carbon materials and the presence of multiple overlapping lithium-storage environments.

After prolonged cycling, the dQ/dV profiles become broader and flatter, suggesting a more distributed electrochemical response and gradual stabilization of the electrode/electrolyte interface. Together with the EIS results, this behaviour supports the interpretation that the gradual capacity increase is related to progressive electrode conditioning, improved charge-transfer kinetics, and increased electrochemical accessibility of the carbon electrode during cycling.

The combination of stable Coulombic efficiency, reversible lithium storage, and good rate capability confirms the good electrochemical stability and structural robustness of the hard-carbon anode during prolonged cycling [48].

EIS analysis shows significant but expected changes in the electrode/electrolyte interfaces during cycling. The high-frequency region initially exhibited two partially overlapping semicircles, which became progressively resolved upon cycling (Figure 6a,b). Additionally, immediately after the first cycle, another semicircle emerged at low frequencies (20–0.5 Hz) and was then present in all the remaining measurements. This semicircle warranted use of different circuits for fitting. The circuit before cycling has one less R–CPE pair (Figure 6c–e). Fitting was carried out in two steps. The initially higher frequency region was fitted. Then, the lower frequency region was fitted with the Warburg element with another CPE in parallel. Parameters and curves from the first fit were used as a baseline for the second fit. All fits had Efron R^2 above 99%. An augmented Warburg element was required because the low-frequency region was not at the typical 45° but was instead rising more sharply, indicating combined behaviour. From comparison of fitted impedances, we can see that R corresponding to the electrolyte is stable during cycling. Resistance R1 at high frequencies is slowly rising. We associate this semicircle with the SEI layer and conclude it is slowly increasing in size, therefore increasing resistance. Resistances R2 and R3 have decreasing trends, indicating a slow stabilization is still taking place (Figure 6f). This interpretation is further supported by the gradual increase in charge capacity during the initial cycles (Figure 5c).

2.3. Post-Mortem Analysis

SEM images of the prepared electrode surfaces before (Figure 7a–c) and after (Figure 7d–f) 10 cycles at a current density of 100 mA g^{-1} provide detailed insight into the morphological stability of the carbon-fiber-based electrodes. At lower magnifications (Figure 7a,b), the electrode surface appears relatively homogeneous and uniformly coated prior to cycling, with no visible cracks, delamination, or structural defects. After 10 cycles (Figure 7d,e), the overall morphology remains largely unchanged, indicating excellent mechanical integrity and electrochemical stability under repeated cycling conditions.

At higher magnification (Figure 7c), a more detailed microstructural arrangement becomes visible, revealing the composite nature of the electrode coating. The fibrous carbon network is clearly embedded within a matrix consisting of the binder (PVDF) and conductive additive (carbon black), which forms a continuous and well-interconnected structure. This homogeneous distribution of components contributes to the structural

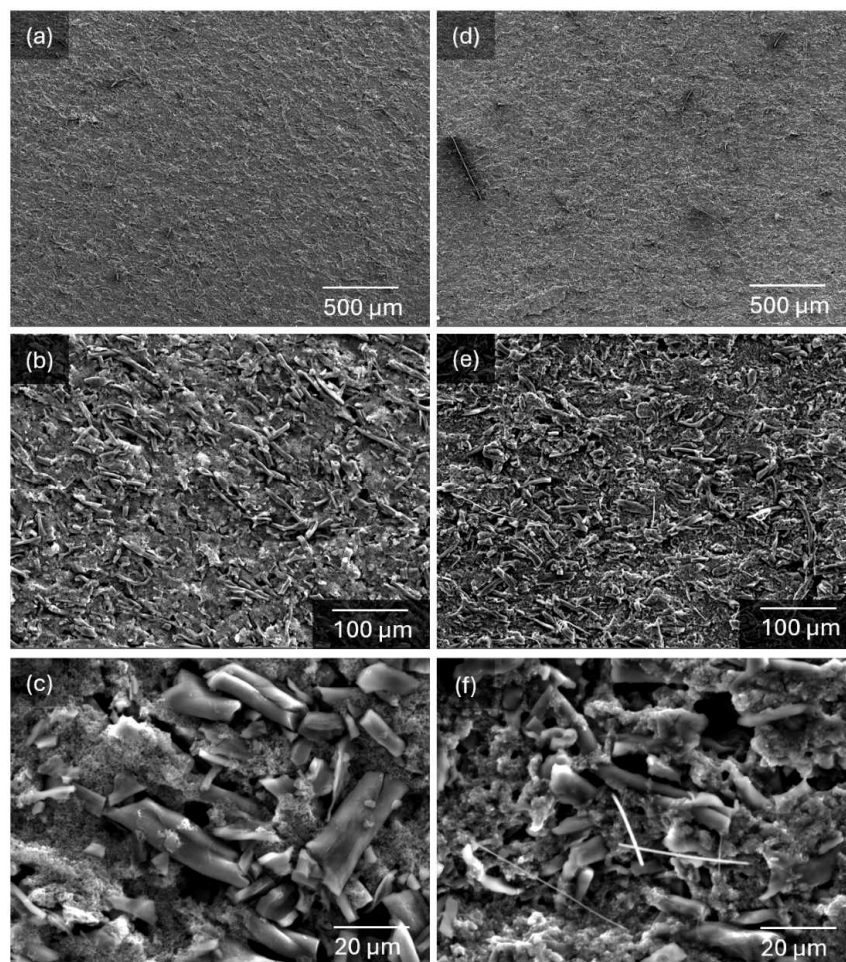


Figure 7. SEM images of the prepared electrode surfaces before (a–c) and after (d–f) 10 cycles at a current density of 100 mA g^{-1} .

After cycling (Figure 7f), the microstructure remains well preserved. In addition to the intact composite electrode architecture, fine needle-like features can be observed, which are attributed to glass fibers originating from the separator. These fibers are visible within the porous electrode surface but do not appear to induce mechanical damage or disruption of the electrode structure, suggesting good interfacial stability between the electrode and separator during cycling.

Overall, the SEM analysis confirms that the carbon-fiber-based electrodes maintain their structural integrity and composite architecture even after repeated electrochemical cycling, demonstrating high resistance to both mechanical and electrochemical degradation. Unlike silicon anodes, where significant volume expansion and continuous SEI growth are typically observed, such degradation phenomena are largely absent in this system [50].

The post-mortem XRD pattern of the cycled cotton-derived carbon fiber electrode after 10 cycles (Figure 8) exhibits reflections associated with the copper-based current collector, which are marked with red dots together with the corresponding crystallographic planes. The pattern is dominated by a broad low-angle diffraction band, indicating the presence of a largely amorphous carbon phase. An additional minor feature appears at $2\theta = 53.48^\circ$, marked with a green star (*). Due to the use of a Cobalt (Co) anode and the intense nature of the primary Cu (200) peak at $2\theta \approx 59.6^\circ$, this minor reflection is unambiguously assigned as an instrumental residual Co-K β radiation artifact originating from the current collector.

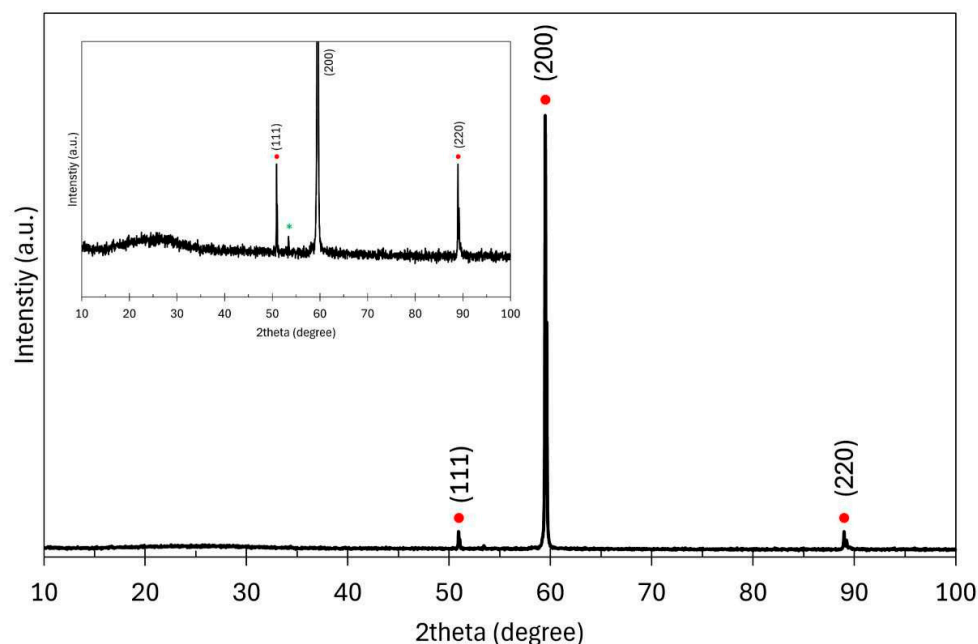


Figure 8. Post-mortem XRD pattern of the cycled cotton-derived carbon fiber electrode after electrochemical testing, showing a predominantly amorphous structure, reflections from the copper current collector (red dots). The minor feature at $2\theta = 53.48^\circ$ (green star, *) is assigned to a residual Co-K β radiation artifact associated with the Co anode and the intense Cu (200) reflection.

3. Materials and Methods

3.1. Material Synthesis

Cotton-derived carbon fibers were prepared from commercial medical-grade cotton wool (waste cotton) via a two-step pyrolysis process. The precursor material was placed into custom-made cylindrical capsules with lids, which were used to limit direct oxygen access during thermal treatment, and inserted into a furnace preheated to 240°C . The temperature was subsequently increased from 240°C to 600°C for 150 min, corresponding to a heating rate of $2.4^\circ\text{C min}^{-1}$, under a nitrogen atmosphere to induce carbonization of the biomass precursor. In the second step, the material was pyrolyzed at 1000°C for 30 min under a nitrogen atmosphere to promote further carbonization and structural stabilization of the resulting carbon material.

The reported temperature profile and atmosphere conditions were established and optimized by the industrial partner based on proprietary process know-how to ensure reproducibility and scalability of the carbonization procedure. The capsule-based configuration used during carbonization was likewise provided by the industrial partner. Certain engineering aspects of the process, including capsule design, gas-handling configuration, furnace setup, and related operational details, constitute proprietary industrial know-how and are therefore beyond the scope of the present work. The synthesis conditions reported above were maintained consistently throughout this study. The obtained carbon material was subsequently ground using a mortar and pestle to obtain a homogeneous powder.

3.2. Electrode Preparation and Cell Assembly

Electrochemical measurements were performed using CR2032-type coin cells. The electrode slurry was prepared by mixing pyrolyzed cotton-derived carbon fibers (70 wt.%), Super P carbon black (20 wt.%, Alfa Aesar, Karlsruhe, Germany), and polyvinylidene fluoride (PVDF, 10 wt.%, Alfa Aesar, Germany) dissolved in N-methyl-2-pyrrolidone (NMP, Sigma-Aldrich, Darmstadt, Germany) in a mass ratio of 7:2:1 (0.175 g, 0.050 g, and 0.3125 g and 0.025 g PVDF). The PVDF was added as 0.3125 g of PVDF/NMP solution, and an

additional 1.4 mL of NMP was added to form a homogeneous slurry. The mixture was homogenized using a planetary centrifugal mixer (Thinky ARE-250, Tokyo, Japan) at 2000 rpm for three cycles of 3 min until a uniform slurry was obtained.

The prepared slurry was coated onto a copper foil (current collector) using a Casting knife (BYK, Wesel, Germany). The coating thickness was set to 180 μm , and the slurry was applied in a single uniform pass to ensure a compact and homogeneous electrode layer. The coated electrodes were subsequently dried in a vacuum oven at 60 $^{\circ}\text{C}$ for approximately 24 h to remove residual solvent.

After drying, mechanically stable electrode films were obtained and circular electrodes with a diameter of 13 mm were punched using a precision electrode cutter (EL-CELL, Hamburg, Germany). Electrode mass loading was 1.36 mg/cm^2 for cyclic voltammetry, galvanostatic charge/discharge profiles, cycling stability and differential capacity (dQ/dV) profiles analysis. For rate capability, the electrode mass loading was 2.26 mg/cm^2 .

Coin cells-CR2032 were assembled in an Ar-filled glovebox (Jacomex, Dagneux, France) with controlled atmosphere ($\text{O}_2 < 1$ ppm, $\text{H}_2\text{O} < 1$ ppm) due to the high sensitivity of the materials to moisture and oxygen. Lithium metal foil (19 mm, Nanografi, Ankara, Turkey) served as the counter/reference electrode, while Whatman GF/A glass fiber (17 mm, Cytiva, Marlborough, MA, USA) was used as the separator. The electrolyte consisted of 1 M LiPF_6 (Sigma-Aldrich, Germany) dissolved in ethylene carbonate (EC) and diethyl carbonate (DEC) in a 1:1 volume ratio.

In the assembled cells, the prepared pyrolyzed cotton-derived carbon fibers electrode was used as the working electrode. The cell components were stacked in the following order: stainless steel casing, lithium metal foil, separator, electrolyte, working electrode, stainless steel spacer, spring, and bottom casing. A total of 120 μL of electrolyte was added to each cell prior to sealing. The cells were hermetically sealed using a hydraulic crimping device and subsequently used for electrochemical testing.

3.3. Electrochemical Measurements

The electrochemical performance of the assembled CR2032-type half-cells described above was evaluated. Cyclic voltammetry (CV) measurements were further performed to investigate the electrochemical behavior of the electrode material over a potential range of 0.01–3 V vs. Li/Li^+ at a scan rate of 0.1 mV s^{-1} , employing a BioLogic SP-150 potentiostat/galvanostat (BioLogic, Seyssinet-Pariset, France).

Galvanostatic charge–discharge cycling was carried out within a potential window of 0–3 V vs. Li/Li^+ using a battery testing system (Landt Instruments, New York, NY, USA). Specific capacities were calculated based on the mass of cotton-derived carbon fibers in the working electrode.

Electrochemical impedance spectroscopy (EIS) measurements were performed using a BioLogic SP-150 potentiostat/galvanostat (BioLogic, Seyssinet-Pariset, France) over a frequency range of 1 MHz–100 mHz with an AC current amplitude of 0.217 mA. Measurements were conducted before cycling and after 1, 2, 5, 10, and 50 galvanostatic charge–discharge cycles. Circuit fitting was performed using CIRFIT EIS Studio (HIOKI E.E. Corporation, Nagano, Japan; version 0.15.3).

3.4. Material Characterization and Post-Mortem Analysis

The X-ray diffraction (XRD) analysis of the pyrolyzed cotton-derived carbon fibers and post-mortem XRD of the electrodes were conducted using a Philips X'Pert Pro diffractometer (Malvern Panalytical, Almelo, The Netherlands) equipped with a cobalt anode ($\text{Co K}\alpha$ radiation, $\lambda = 1.7903$ Å) and operated at 40 kV and 40 mA. The measurements were

recorded over a 2θ range of 10° to 100° , with a step size of 0.033° and a dwell time of 60 s per step.

The morphology of the cotton-derived carbon fibers after pyrolysis was analyzed using a JSM-7000F scanning electron microscope (JEOL, Tokyo, Japan) equipped with an INCAx-sight energy-dispersive X-ray spectroscopy (EDX) detector (Oxford Instruments, Abingdon, UK).

TEM observations of the pyrolyzed cotton-derived carbon fibers were performed using a JEOL JEM-2100F UHR transmission electron microscope (JEOL, Tokyo, Japan) equipped with a Schottky field-emission gun and operated at 200 kV. Bright-field imaging was used for microstructural analysis, whereas phase characterization was performed by selected area electron diffraction (SAED). For TEM examination, the fibers were mechanically dispersed onto an ultrathin carbon film supported by a lacey carbon copper grid.

SEM images of the electrodes before and after cycling, as well as EDS mapping of the cotton-derived carbon fibers after pyrolysis, were acquired using a Tescan VEGA 3 LMU scanning electron microscope (Tescan, Brno, Czech Republic) equipped with an energy-dispersive X-ray spectroscopy (EDX) detector (Bruker, Billerica, MA, USA), which was used to determine the elemental composition of the samples.

4. Conclusions

In this work, cotton-derived carbon fibers were successfully prepared through a two-step pyrolysis process and evaluated as anode materials for lithium-ion batteries. The results demonstrate that waste cotton can serve as a low-cost and sustainable precursor for the preparation of hard carbon materials with promising electrochemical properties. Structural characterization confirmed the predominantly amorphous and disordered nature of the prepared carbon. XRD and SAED analyses revealed the absence of long-range crystalline ordering in the carbon fibers, while TEM further confirmed the amorphous character of the fibers and the presence of fine polycrystalline CaCO_3 particles on the fiber surface. SEM/EDS analysis showed that the fibrous morphology of the original cotton precursor was largely preserved after pyrolysis, forming a chemically heterogeneous and interconnected carbon structure. Electrochemical measurements revealed the characteristic behaviour of hard-carbon anodes, including irreversible processes during the first cycle followed by stable and reversible lithium storage in subsequent cycles. The material exhibited good cycling stability and a gradual increase in reversible capacity during prolonged cycling, reaching approximately $780\text{--}800\text{ mAh g}^{-1}$ after 300 cycles at 100 mA g^{-1} . This performance exceeds the theoretical capacity of graphite and suggests that lithium storage is not limited to conventional graphite-like intercalation.

The CV, galvanostatic charge–discharge, and dQ/dV results are consistent with the broad and distributed electrochemical response typically observed for disordered hard-carbon materials. The dQ/dV profiles showed no sharp, well-defined peaks or clear graphite-like staging transitions, but rather a broad response over a wide potential range, supporting the probable presence of overlapping lithium-storage processes. Together with the EIS results, this behaviour suggests gradual stabilization of the electrode/electrolyte interface and progressive electrochemical conditioning of the electrode during cycling. Based on the amorphous carbon structure observed by XRD and SAED, the retained fibrous morphology, and the electrochemical response, the high capacity is considered consistent with the multi-site lithium-storage behaviour generally reported for disordered hard carbons. The individual contributions of defect sites, oxygen-containing species, enlarged interlayer spacing, and micro-/nanoporous domains should therefore be regarded as probable, literature-supported contributions rather than individually resolved mechanisms in the present material. The electrode also showed stable Coulombic efficiency and good rate

capability, confirming the electrochemical stability of the prepared material. Post-mortem SEM analysis further confirmed that the electrode structure remained stable after repeated cycling without significant mechanical degradation. Overall, the obtained results indicate that cotton-derived carbon fibers are a promising sustainable alternative to conventional graphite anodes. Further optimization of the synthesis conditions and complementary structural characterization will be useful for improving the initial Coulombic efficiency and for clarifying the lithium-storage mechanisms in these biomass-derived carbon materials.

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