

Advancing Green Batteries: The Role of Lignin-Derived Carbon in Alkali Metal Ion Energy Storage

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The global transition toward a sustainable energy future necessitates groundbreaking innovations in energy storage technologies that are both environmentally congruent and financially viable. In the face of escalating global energy demands, the depletion of fossil fuel reserves, and the exacerbating effects of environmental degradation, the urgency for pioneering, cost-efficient, and eco-friendly bio-based energy storage solutions has reached a critical juncture. Lignin is the second most abundant natural polymer that primarily emerges as a by-product in the paper pulping and bio-refining industries. Its inherent multiplicity of eco-friendly attributes positions it as an exceptionally promising candidate for integration into next-generation

rechargeable battery technologies. This review meticulously synthesizes the latest advancements concerning lignin-derived electrode materials in alkali metal-ion batteries, with particular emphasis on the cutting-edge application of lignin-based electrodes. Furthermore, it critically analyzes the prevailing challenges and offers forward-looking insights into the transformative potential and future trajectories of lignin-derived materials within the ever-evolving landscape of energy storage technologies. The exploration of lignin's untapped potential represents not only a step toward more sustainable energy solutions but also a pioneering approach to overcoming the inherent limitations of current electrode materials.

1. Introduction

As the global shift toward green energy accelerates, developing sustainable, affordable, and efficient energy storage technologies has become a top priority. Lithium-ion (LIBs), sodium-ion (SIBs), and potassium-ion (KIBs) batteries, which are known as alkali metal-ion batteries (AMIBs), have been preferred for their high energy efficiency, long-lasting performance, low maintenance, and flexible power-to-energy capabilities.^[1–5] To create high-performance, eco-friendly rechargeable batteries, intense efforts have focused on optimizing electrodes and key components. Bio-derived materials are gaining attraction for their renewability and versatility. Carbon-based materials are sought after in energy storage for their superior conductivity, stability, and vast surface area.^[6–10] Among bio-based materials, lignin is a biopolymer abundant in aromatic structures, which has been extensively studied in recent years as a potential component for energy storage devices. Lignin naturally decomposes while mitigating the environmental impact of batteries. Lignin's renewability and high carbon content make it a strong candidate for advancing sustainable and high-performance energy storage in AMIBs.^[11–14] Lignin does not inherently surpass traditional carbon materials in all

energy storage applications.^[15] However, it presents unique advantages while making it a promising alternative in certain applications.^[16,17]

The rising interest in lignin stems from its abundance, sustainability, cost-effectiveness, and biodegradability. It offers superior sustainability over traditional carbon sources while reducing the environmental impact of energy storage. Recent research highlights its potential to drive eco-friendly and sustainable energy storage technologies.^[18–20] Lignin-based electrode materials boast high surface area, customizable pore structures, and rich functional groups while making them highly effective for energy storage applications.^[21,22] Lignin-based electrode materials have demonstrated remarkable performance across a range of energy storage systems especially in AMIBs. Recent studies have highlighted successful commercial instances of lignin-derived electrode materials and have suggested their substantial potential for widespread industrial adoption.^[23–26] Lignin-derived carbon features greater graphite layer spacing due to its complex 3D structure while offering a strong potential for use as anodes in AMIBs.^[27,28] Lignin's electroactive redox properties and rich functional groups offer key active sites for rechargeable battery development. Its exceptional thermal stability, oxidation resistance and hardness provide significant structural advantages for battery components.^[29–31] While many studies suggest lignin as a promising precursor for high-performance battery electrodes, few explore the complex links between its chemical and physical structures and the resulting properties of lignin-based electrodes.^[32–34] It is noteworthy that the pulp and paper industry is the primary source of lignin while producing large amounts of waste black liquor during pulping. While rich in lignin, black liquor is typically seen as a problematic waste, often used for low-value purposes like heat and electricity generation.^[35,36]

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Despite its abundance, the vast potential of lignin extracted from black liquor remains largely untapped.^[37–40] By reimagining its utilization, this abundant and underexploited resource could be harnessed as a critical component in the advancement of cutting-edge energy storage technologies. Such an approach not only alleviates the waste disposal challenges associated with black liquor but also fosters the integration of more sustainable practices within the pulp and paper sector while heralding a new paradigm for industrial symbiosis and resource valorization.^[41–43] Converting waste black liquor lignin into carbon offers a unique route for advancing energy storage systems. By leveraging lignin's unique properties, researchers can create electrode materials that are efficient, cost-effective and environmentally sustainable.^[44–46] Lignin is mostly obtained from the lignocellulose biomass of plants and trees. It provides mechanical strength and resistance to degradation because it is a major component of the plant cell walls.^[47] Due to its natural abundance and renewability, lignin is a material of great interest for a range of applications including energy storage technologies. As a result of the paper industry's pulping processes, lignin has always been considered as a by-product of wood and plant material. In the past, low-value products like fuel or inexpensive chemicals were made from lignin.^[48] Its potential as a high-performance material and especially in energy storage applications has remained largely unexplored until recently.^[49]

In the context of AMIBs, the increasing demand for more sustainable, cost-effective, and eco-friendly energy storage solutions has catalyzed the exploration of lignin as a candidate for

electrode materials.^[50] Researchers began recognizing the potential of lignin to serve as a high-carbon and electroactive material capable of enhancing the performance of rechargeable batteries, particularly as alternatives to traditional carbon-based materials such as graphite.^[51] The development of lignin-based electrodes has gained momentum with advances in material processing techniques aimed at improving lignin's electrochemical properties, such as conductivity, porosity, and ion diffusivity. These efforts have sparked significant research interest in leveraging lignin's unique characteristics to improve battery performance, reduce environmental impact, and promote the use of renewable resources in energy storage technologies.^[52] Lignin-based materials have emerged as an attractive option for AMIBs, particularly in the development of electrodes for LIBs, SIBs, and KIBs. One of the key advantages of lignin in AMIBs lies in its high carbon content, which is essential for enhancing the energy density and electrochemical performance of batteries.^[53] The intrinsic aromatic structure of lignin also provides a significant surface area while contributing to an increased number of electroactive sites. These sites are crucial for the reversible intercalation and deintercalation of ions during battery cycling while enhancing charge capacity and cycle stability. Several studies have demonstrated that lignin-based electrodes exhibit comparable performance as compared to traditional carbon-based materials in terms of charge capacity, cycling stability, and structural integrity.^[54]

Moreover, lignin-based carbon materials can be easily tailored to suit specific battery applications. Researchers have been experimenting with different chemical and physical modifications, such



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as carbonization and activation, to further enhance the porosity and conductivity of lignin-derived materials.^[55] Additionally, lignin's renewable nature and biodegradability make it an environmentally friendly alternative to conventional materials while aligning with the global push toward sustainable energy solutions. As the demand for more eco-conscious energy storage technologies grows, lignin's potential for use in AMIBs is being increasingly recognized, particularly in the context of potassium-ion and sodium-ion batteries, which have been identified as promising alternatives to lithium-based systems.^[56]

In addition to lignin's direct application in energy storage, researchers have also begun exploring hybrid materials that combine lignin with other nanomaterials, such as graphene, carbon nanotubes (CNTs), and metal oxides. These hybrid materials aim to further improve the conductivity, electrochemical performance, and ion diffusion of lignin-based electrodes while overcoming some of its inherent limitations.^[57] As a result, lignin-based materials are emerging as a viable candidate for high-performance and sustainable energy storage systems. Despite the promising potential of lignin-based materials for AMIBs, several challenges remain in fully realizing their widespread application. One of the main hurdles is the inherent heterogeneity of lignin, which can lead to inconsistencies in the material's performance. Lignin is a complex polymer with varying molecular structures while depending on the source and extraction process. This variability can result in batch-to-batch differences in performance while making it difficult to ensure consistent quality and reliability across large-scale production.^[58] Furthermore, the natural properties of lignin pose significant challenges in optimizing its performance as an electrode material. To overcome this issue, lignin often requires chemical modifications, such as activation or the incorporation of conductive additives (e.g., CNTs or graphene) to improve its conductivity and overall electrochemical performance.^[59] Another challenge lies in the scalability and cost-effectiveness of processing lignin into high-performance electrode materials. The techniques used to convert lignin into carbon or other conductive forms, such as pyrolysis or chemical activation, can be energy intensive and expensive.^[60] Additionally, the extraction and purification of lignin from biomass can present logistical and economic barriers for large-scale production. These factors can limit the widespread adoption of lignin-based electrodes unless more efficient and cost-effective processing methods are developed.^[61] Furthermore, while there have been significant advancements in understanding the electrochemical behavior of lignin-based materials, much remains to be learned about the detailed mechanisms that govern ion interactions within lignin during battery cycling.^[62,63] A deeper understanding of these mechanisms is critical for optimizing lignin's structural design to maximize ion diffusion, cycling stability, and overall performance. Researchers must also develop standardized testing methods to compare lignin-based materials across different battery chemistries, as performance can vary widely depending on the specific type of AMIB being used.^[64]

Due to its structural properties, lignin has the potential to be used as a new, renewable, and cheap material for AMIBs. As compared to other carbon-based materials, lignin is a better option due to its high carbon content, electroactive structure, and

renewability. While significant progress has been made with electrodes based on lignin, there is still work to be done in dealing with problems pertaining to heterogeneity, low conductivity, and scalability for processing. Optimization of lignin's structure using new processing methods will have to be researched further in order to improve performance, reduce production costs, and enhance overall yield. There is an increased global demand for energy storage solutions, and lignin will likely serve greater purpose in AMIBs. Researchers can help shift the world toward an eco-friendly future by optimizing lignin for high performance energy storage technologies, which will help AMIBs become widely used environmentally safe devices. Merging lignin with other advanced materials will help overcome current limitations and encourage commercial adaptation. With further collaborative efforts and innovation, refined lignin-based devices will revolutionize energy systems while combating the increased need for renewable energy storage.

This paradigm shift not only mitigates the environmental impact typically associated with conventional energy storage technologies but also augments the overall performance and sustainability of energy systems. Furthermore, this approach is inextricably linked to the principles of a circular economy, wherein waste is ingeniously repurposed into high-value resources while fostering sustainability within the pulp and paper industry. By leveraging the abundant lignin from waste black liquor, the sector has the opportunity to contribute to a more sustainable and environmentally-conscious future while addressing both economic and ecological imperatives.^[65–67] Key properties of lignin-derived electrodes in AMIBs are illustrated in (Figure 1) while providing a visual summary of their exceptional performance characteristics. In this review, we present a comprehensive analysis of research on lignin-derived electrode materials for AMIBs over the past five years. This review meticulously explores lignin-based anodes for AMIBs while providing an in-depth analysis of fabrication techniques and their applications in energy storage. It emphasizes the numerous challenges in the development of these materials while concerning their economic viability and environmental footprint. The review also evaluates the technological maturity and public acceptance of lignin-derived electrodes while identifying critical research opportunities and offering strategic recommendations to propel the field forward. Hence, it highlights the disruptive potential of lignin-based electrodes in transforming energy storage paradigms while championing sustainable and environmentally conscious innovations.

2. Working Mechanism of AMIBs

The paramount importance of utilizing lignin-derived electrodes in AMIBs resides in their potential to instigate a profound transformation in energy storage technologies while steering them toward a more sustainable trajectory.^[68–70] This development not only advances energy storage capabilities but also fortifies the progress of renewable energy technologies while positioning lignin-derived anodes as a compelling and forward-looking alternative for next-generation AMIBs.^[71,72] The operation of a rechargeable AMIB is predicated on the cyclical migration of

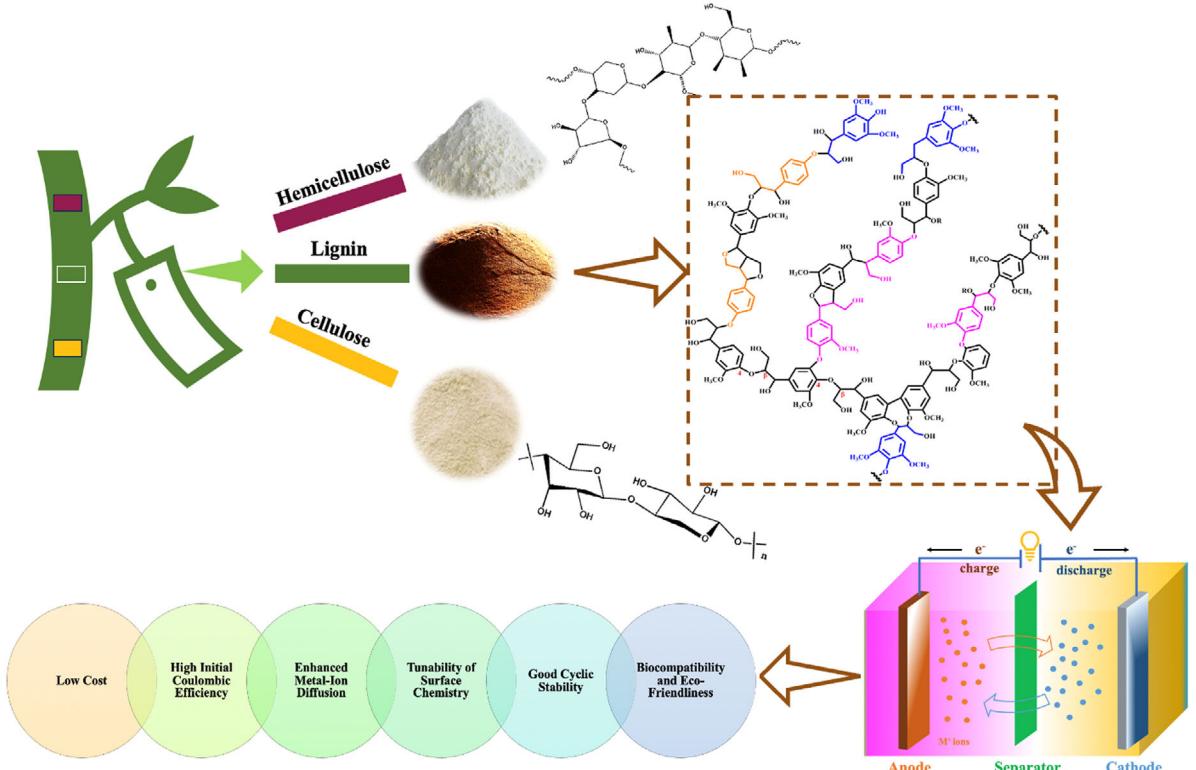


Figure 1. Some of the major properties of lignin-derived electrodes in AMIBs.

metal ions ($M^+ = Li^+, Na^+, K^+$) between the anode and cathode in a process which is enabled by the electrolyte. This dynamic mechanism of ion movement underpins both energy storage and discharge functions within AMIBs. During discharge, M^+ ions traverse the electrolyte while migrating toward the positively charged cathode while facilitating the energy release. This intricate ion transport mechanism is central to the battery's ability to continuously cycle between energy storage and discharge while ensuring sustained performance throughout its operational life.^[73] During the charging phase, lithium ions (M^+) migrate back to the anode through the same electrolyte, as depicted in (Figure 2). This process represents a meticulous inversion of

the discharge cycle, where the directional flow of M^+ ions is reversed.^[74] In the realm of AMIBs, specific parameters are meticulously measured to assess and quantify battery performance, which includes coulombic efficiency, rate capacity, and both reversible and irreversible capacity. The remarkable rechargeability of AMIBs is predominantly attributed to the reversible intercalation and de-intercalation of M^+ ions. However, empirical investigations have elucidated that, during these ions cycling processes, inevitable losses occur over successive cycles.^[75] As research advances, lignin-derived electrodes present a promising way to enhance AMIB performance and reduce environmental impact.^[76–78] As research and innovation progress, lignin-derived electrodes have the capability to greatly improve the efficiency, sustainability, and longevity of AMIBs.^[79]

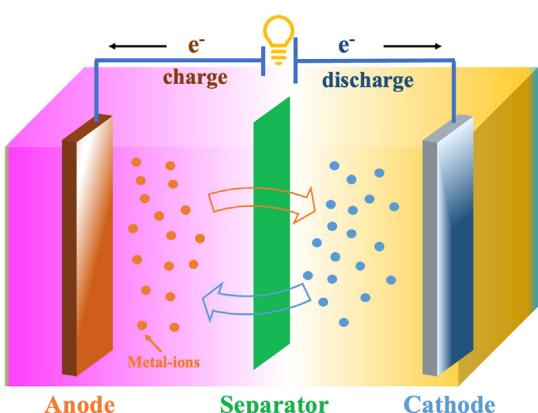


Figure 2. A schematic representation of the ion and charge transport mechanism within AMIBs.

3. Lignin

Lignin is a multifaceted and complex biopolymer that is a pivotal constituent of cellular architecture in a vast array of plants, particularly within the lignified tissues of wood and bark. Functioning as a critical structural element, it imparts mechanical rigidity and confers resistance to biotic degradation. Unlike cellulose, lignin exhibits an intricate and irregular molecular architecture, which is characterized by a network of highly branched and cross-linked phenolic compounds.^[80] This structural uniqueness is integral to the robustness and durability of plant materials while rendering lignin indispensable to the growth, structural integrity, and

ecological stability of vascular plants. Furthermore, lignin plays a quintessential role in the terrestrial carbon cycle while serving as a substantial reservoir of carbon within plant biomass, thus contributing to the sequestration of atmospheric CO₂. The singular molecular nature of lignin has piqued significant industrial interest while positioning it as a promising feedstock for a spectrum of sustainable applications like biofuels, bioplastics, and as a potential substitute for conventional fossil-derived materials. Lignin stands as the sole naturally occurring and renewable source capable of yielding aromatic polymers which has spurred research into its utilization as a feedstock for high-performance materials. Embedded within the plant cell wall, lignin collaborates synergistically with cellulose and hemicellulose while forming a complex and three-dimensional network that acts as a reinforcing scaffold. This molecular scaffolding plays an instrumental role in fortifying plant cell walls while enhancing their structural rigidity and enabling plants to withstand external mechanical stresses in order to maintain an upright growth.^[81–83] In addition to its biological functions, lignin holds considerable promise in the field of energy storage technologies. As a widespread by-product of the pulp and paper industries, it presents a promising foundation for bio-based storage systems. By utilizing its ability to form stable organic structures, researchers are creating advanced, eco-efficient energy devices that compete with traditional materials. As the demand for sustainable energy solutions grows, lignin is set to bridge the gap between intermittent renewable energy generation and long-term storage while addressing critical challenges like cycle stability, efficiency, and scalability to promote widespread adoption.

In future energy storage technologies, it is important to investigate the possibilities of lignin based electrode materials as a sustainable and comparatively economical alternative to conventional graphite and lithium-based materials.^[84] To fully understand the relevance of these materials, it would be beneficial to incorporate predictions on the potential market share and application prospects of lignin-based electrodes in the energy storage market. This exploration can provide insights into how these innovative materials could impact various industries, including electric vehicles (EVs), renewable energy storage, and portable electronics, which all rely heavily on advancements in energy storage technologies.^[5] The integration of lignin into energy storage systems is driven by its potential to enhance the overall performance of batteries. As the world continues its transition to renewable energy sources, the demand for energy storage systems that can efficiently store and release energy will only increase.^[85] The incorporation of lignin-based materials into energy storage devices can help meet this demand while offering an environmentally friendly and cost-effective solution. Specifically, lignin could be used as a component in the electrodes of batteries.^[86] These devices are essential for various applications, including EVs, grid energy storage for renewable energy systems, and portable consumer electronics. Lignin's potential to reduce production costs and reliance on non-renewable resources makes it an attractive option for the future of energy storage technology.^[87]

The global market for energy storage technologies is expected to grow rapidly in the coming years. According to market analysts, the energy storage market is projected to experience

substantial growth, which is driven by the advancements in renewable energy integration, the increasing popularity of EVs and the expansion of portable electronics.^[88] In addition, the EV market is expected to be a major driver for energy storage solutions, with significant investments being made in battery technology research and development. The need for energy storage systems that are both cost-effective and environmentally sustainable is fueling the exploration of alternative materials.^[89,90] The potential market share of lignin-based electrode materials will depend on several factors, including technological advancements in lignin processing, the scalability of production methods, and the overall performance of lignin-based energy storage devices.^[91] Early-stage research has shown promising results in terms of lignin's electrochemical properties, but there is still a need for further optimization to improve its efficiency and lifespan in commercial applications. Hence, lignin-based electrodes could secure a significant portion of the energy storage market, particularly in sectors that prioritize sustainability and cost-effectiveness.^[92]

In the EV industry, the demand for energy-dense, lightweight, and efficient batteries is essential for increasing the driving range and affordability of EVs. Lignin-based electrodes could play a role in reducing the cost of battery production while contributing to the overall performance. Additionally, lignin's renewable nature aligns with the goals of reducing the carbon footprint associated with the automotive industry.^[5] Lignin-based materials could help accelerate the adoption of EVs by providing a more sustainable and affordable alternative to traditional battery technologies. Furthermore, in renewable energy storage, lignin-based electrodes could be a game changer. Lignin-based materials could be used to develop more efficient energy storage devices while helping to address the intermittent nature of renewable energy sources. The potential to use lignin could make large-scale energy storage more accessible and affordable, especially in regions where the cost of renewable energy storage is currently a significant barrier.^[93]

Hence, the integration of lignin-based electrode materials in future energy storage systems represents an exciting opportunity for both researchers and industries focused on sustainability and innovation. By offering a renewable, cost-effective, and high-performance alternative to conventional electrode materials, lignin has the potential to disrupt the energy storage market. As the demand for environmentally friendly, efficient, and affordable energy storage solutions grows, lignin-based materials could secure a significant market share, particularly in sectors such as EVs, renewable energy storage, and portable electronics. Understanding the potential applications and market dynamics of lignin-based electrode materials will be crucial in shaping the future of energy storage technology. Further research and investment will be key to unlock the full potential of lignin-based materials and advancing their adoption in the global energy storage market.

3.1. Structure of Lignin

Lignin is a complex and high molecular weight biopolymer that plays a crucial role as a structural component in plant cell walls

while offering mechanical strength, rigidity, and resistance to degradation. It is mainly composed of three phenylpropanoid units (syringyl, guaiacyl, and p-hydroxyphenyl), which are connected through ether and carbon–carbon bonds to form a dense network of aromatic structures.^[94] This molecular architecture manifests as a hydrophobic and highly aromatic three-dimensional framework while conferring not only structural stability but also facilitating the efficient transport of water and nutrients within vascular plants. The inherent structural heterogeneity and complexity of lignin present both challenges and opportunities for its exploitation. The molecular framework of lignin is not amenable to a singular chemical formula due to its considerable structural diversity, which arises from variations in precursor composition and the diversity of methods employed for its extraction and purification. Lignin biosynthesis occurs through enzymatic dehydrogenative polymerization and free-radical coupling of aromatic precursors while yielding an extremely complex and variable polymer.^[95] Lignin's elemental composition is dominated by carbon (~60%), with hydrogen and oxygen in smaller proportions, and its molecular configuration remains a subject of ongoing debate.^[96] This discrepancy underscores the considerable structural variance inherent in lignin types while complicating the efforts to fully characterize its molecular architecture. Despite these complexities, lignin's unique structural attributes render it a material of immense potential.^[97] These same features also present significant challenges for processing and large-scale utilization. Lignin's distinctive molecular structure offers exceptional promise for advanced applications in the realm of sustainable energy storage technologies.^[98] The potential to harness lignin's high carbon content and aromatic complexity for the development of bio-based energy storage devices is increasingly being recognized. Moreover, modifications such as carbonization or chemical derivatization are actively being explored to enhance its functionality for a diverse range of applications across the

renewable energy, materials science, and biotechnological sectors. Hence, lignin's intricate, multifaceted molecular architecture positions it as a compelling and an albeit challenging candidate for diverse industrial applications.^[99] Ongoing research aimed at unraveling its structural nuances and refining processing methodologies which is critical to realize the lignin's vast untapped potential in the development of sustainable and bio-based alternatives to conventional fossil-derived materials and energy storage solutions. The structure of lignin along with the basic structural units of lignin is shown in Figure 3a,b.

3.2. Classification of Lignin

Lignin is a complex and a high-molecular-weight biopolymer that resides in the cell walls of plants, where it plays a pivotal role in conferring structural integrity and enhancing plant defense mechanisms. As one of the most ubiquitous natural polymers on Earth, lignin ranks second only to cellulose in its prevalence within terrestrial biomass. It is primarily composed of phenylpropanoid units, which are intricately linked by a diverse array of ether and carbon–carbon bond while rendering it highly resistant to microbial and chemical degradation.^[100] Lignin's classification is often based on its structural and chemical attributes, as well as its biosynthetic lineage. It is conventionally subdivided into three types which are known as guaiacyl (G), syringyl (S), and hydroxyphenyl (H) lignin. Each type is distinguished by the specific monolignols, which are polymerized to form the macromolecular structure.^[101] This tripartite division encapsulates the fundamental variations in lignin's composition as outlined in the following section.

3.2.1. Guaiacyl (G) Lignin

Guaiacyl lignin (G-lignin) is a major type of lignin found in plants, particularly in gymnosperms like conifers and some dicots. It is

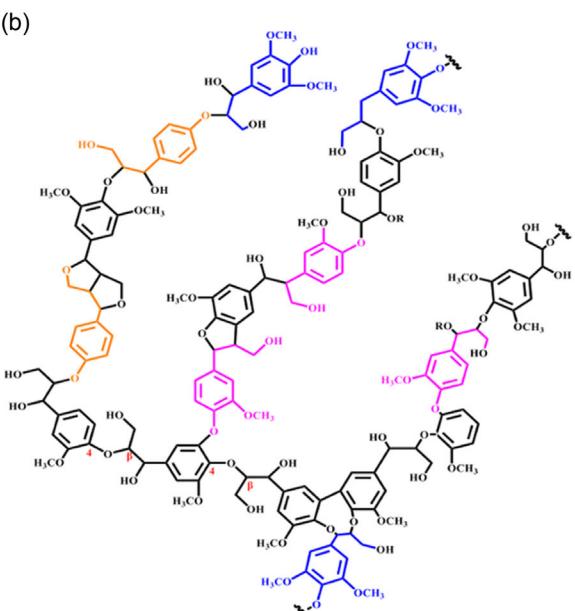
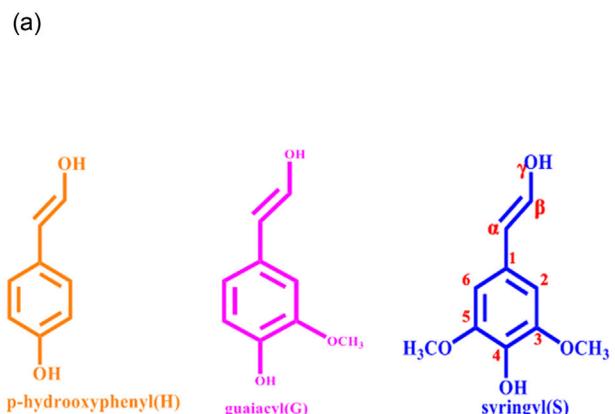


Figure 3. a) Basic structural units of lignin. b) Structure of lignin.

mainly formed from coniferyl alcohol and recognized by its aromatic structure, which includes methoxy groups (-OCH₃) attached to the phenylpropanoid units.^[102] These structural features give G-lignin its strength and rigidity while playing a crucial role in reinforcing plant cell walls. The main function of G-lignin is to provide mechanical support and protection to plant cells while helping plants to resist environmental stress such as drying out, physical damage, and attacks from microbes. Its durability and resistance to breakdown by enzyme make G-lignin important for various industrial applications especially in the paper industry. In addition to its structural role, the chemical characteristics of guaiacyl lignin position it as a highly promising candidate for avant-garde applications in energy storage technologies. Consequently, guaiacyl lignin assumes a pivotal function in fortifying and safeguarding plant architecture, especially within gymnosperms and certain dicotyledons. Its molecular composition, enriched with methoxy substituents, not only enhances its stability but also underscores its utility in both the pulp and paper industries and in the development of sustainable energy storage solutions.^[103] As interest grows in renewable materials, G-lignin's abundance in plant biomass positions it as a valuable resource for environmentally friendly innovations in energy storage and green energy solutions.^[104]

3.2.2. Syringyl (S) Lignin

Syringyl lignin (S-lignin) represents a critical subclass of lignin, which is synthesized from sinapyl alcohol. S-lignin is mainly found in the vascular tissues of flowering plants and helps maintain the strength of cell walls. Compared to the more rigid G-lignin, S-lignin has more methoxy groups (-OCH₃) attached to its aromatic rings, which gives it different properties and greater flexibility.^[105] This augmentation bestows an exceptional degree of flexibility upon its molecular architecture while rendering it more prone to microbial degradation and oxidative disintegration. The heightened methoxy substitution within S-lignin profoundly modulates its physicochemical attributes while amplifying its reactivity and diminishing its resistance to oxidative decomposition. Such enhanced reactivity renders S-lignin an exceptionally advantageous material for industrial processes in order to necessitate the efficient depolymerization of lignin. The expedited degradability of S-lignin aligns seamlessly with contemporary imperatives for advanced bioconversion technologies tailored to renewable energy solutions. Furthermore, the distinctive chemical signature of S-lignin positions it as an invaluable candidate for pioneering applications in bioengineering and energy innovation. Its heightened susceptibility to catalytic fragmentation underscores its potential as a cornerstone in the evolution of sustainable bioenergy systems. In particular, recent investigations into the electrochemical properties of S-lignin suggest its potential as a material in energy storage devices while offering a pathway to novel and eco-efficient energy storage solutions. As a renewable and abundant biopolymer, syringyl lignin holds substantial promise not only as a key player in plant biomass architecture but also as an agent in the advancement of green technologies.^[106] Its unique combination of flexibility, reactivity,

and biodegradability underscores its importance while reducing reliance on fossil fuels and fostering environmentally sustainable energy alternatives. The ongoing exploration of its properties and applications will likely lead to transformative innovations in bio-energy and materials science, while solidifying S-lignin's potential as a cornerstone of the bioeconomy.^[107]

3.2.3. Hydroxyphenyl (H) Lignin

Hydroxyphenyl (H) lignin is the least prevalent lignin isoform within the plant kingdom, which exists in minuscule quantities relative to the more ubiquitous G-lignin and S-lignin variants. Predominantly synthesized from *p*-coumaryl alcohol, it is most notably encountered in grasses, select monocots, and certain lower plant taxa. This lignin subtype is distinguished by the presence of a hydroxyl group (-OH) on the aromatic ring.^[108] The relative structural simplicity of H-lignin confers both heightened reactivity and reduced molecular complexity while rendering it more susceptible to depolymerization via chemical or microbial means. Its diminished degree of polymerization which is coupled with lower cross-linking results in a reduced molecular weight. This property is highly advantageous for processes requiring lignin breakdown.^[109] Although H-lignin constitutes a comparatively minor fraction of the lignin pool in most plant species (grasses and monocots), where its intrinsic structural simplicity generates both unique opportunities and constraints. Its reactivity and relative instability position it as a compelling target for biotechnological exploitation where efficient lignin deconstruction is critical. However, its propensity for degradation limits its applicability in industries that demand long-term structural integrity.

Ongoing advancements in the processing and valorization of H-lignin hold promise for unlocking novel avenues for its utilization in the bioeconomy while catalyzing the development of sustainable technologies and enhancing environmental stewardship. The triadic classification of lignin not only deepens our comprehension of plant physiology but also carries profound ramifications for the advancement of energy storage technologies. Owing to its intricate aromatic framework and phenolic moieties, lignin has garnered substantial interest for its prospective application in bio-electrochemical energy storage systems. The material's expansive surface area facilitates charge retention, while the distinctive chemical variations among lignin subtypes (guaiacyl, syringyl, and hydroxyphenyl) further modulate their electrochemical characteristics.^[110] Consequently, certain lignin isoforms may exhibit superior suitability for specific energy storage applications. Lignin's intrinsic conductivity and propensity for redox activity render it as a promising electrode candidate for batteries and capacitors. Ongoing research seeks to elucidate how the unique chemical profiles of lignin can optimize the performance of electrochemical devices while positioning lignin-based electrode materials as formidable alternatives to conventional electrode substances. Its abundant availability, cost-effectiveness, and renewable origins further elevate lignin as an attractive prospect for the development of sustainable energy storage solutions while offering a potential avenue for diminishing reliance on fossil-based technologies.^[111] The integration of lignin in energy

storage systems not only aligns with the global shift toward sustainability and renewable energy but also encapsulates the principles of a circular economy.^[112]

Lignin has gained attention as an alternative material for AMIBs due to its renewable, eco-friendly, and abundant nature. The types of lignin used in these batteries differ primarily in their source, chemical structure, and physical properties, which influence their electrochemical performance. Native lignin is directly extracted from plants. It is highly complex and irregular, which makes it difficult to use in high-performance applications without modification.^[113] Kraft lignin is a by-product of paper production. It is more uniform and thermally stable while making it suitable for use in composite materials and energy storage after additional processing. Wood-derived lignin varies between softwood and hardwood while influencing its reactivity and potential uses in biofuels and batteries.^[114] Agricultural lignin, extracted from residues like straw and husks, has higher porosity while making them suitable for energy storage materials.^[115] Carbonized lignin created through pyrolysis increases conductivity in battery anodes. In addition, modified lignin possesses ideal energy storage properties due to their enhanced solubility and ionic conductivity. Some modifications to chemical structure will be necessary for meeting specific requirements of different types of lignin, which directly impacts their practical use. As a derivative of biomass, lignin is a renewable resource while paving the way for sustainable electrode materials. The processing route and source of lignin dictate the ecological concern associated with lignin-based batteries. Agricultural waste or industrial by-product-derived lignin is often more environmentally friendly than wood-derived lignin. Hence, lignin-based materials not only help in the fabrication of eco-friendly and recyclable energy storage technologies but also promote biodegradability. The goal of ongoing research is to tailor these types of lignin for use in AMIBs while enabling high-performance energy storage solutions.^[116]

3.3. Synthesis Methods for Lignin Extraction

Lignin is a highly intricate and aromatic biopolymer, which constitutes a substantial proportion of plant cell walls in wood and other lignocellulosic biomasses. Owing to its remarkable abundance and distinctive structural characteristics, lignin has increasingly attracted attention as a renewable and versatile resource for a broad array of applications.^[117–126] The efficient extraction of lignin from biomass remains a pivotal step in unlocking its vast potential across these diverse sectors.^[127,128] This section aims to critically examine these various extraction techniques, offering a comparative analysis of their efficiency, environmental ramifications, and scalability.^[129] Additionally, it will address cutting-edge innovations that seek to optimize lignin recovery while simultaneously enhancing its applicability in downstream processes.

3.3.1. Organic Solvent Method

The organic solvent-based approach for lignin extraction has garnered significant attention as an efficacious technique for isolating lignin from lignocellulosic biomass. This methodology

capitalizes on the differential solubility of lignin relative to other constituent cell wall polymers by utilizing organic solvents to dissolve and extract lignin from plant matter. Organic solvents (alcohols, phenols, ketones, and esters) serve to disrupt lignin's intricate molecular architecture while enhancing the efficiency of its extraction.^[130] This methodology is esteemed as a more temperate and eco-friendlier alternative to harsher chemical processes while offering an effective means of lignin isolation. The organic solvent extraction relies on selective solubility while allowing lignin to be selectively dissolved. The distinctive hydrophobicity and pronounced aromaticity of lignin render it more readily soluble in organic solvents compared to the polysaccharidic constituents of plant cell walls.^[131] A range of organic solvents (alcohols, phenols, ketones, and esters) can be used to disrupt the forces that keep lignin's structure intact while enabling its extraction from biomass. The process can be enhanced with heat, pressure, and additives to improve solvent penetration and lignin solubilization.

This method's key advantage lies in its milder conditions compared to harsher chemical treatments, which can degrade lignin and result in the loss of valuable biomass.^[132] This method preserves lignin and biomass integrity while offering a sustainable and nondestructive approach for lignin extraction.^[133] A salient merit of this technique is its comparatively benign environmental impact, especially when juxtaposed with conventional chemical pulping methods, which often entail aggressive reagents that can degrade both lignin and other valuable biomass constituents. In contrast, organic solvent-based extraction operates under milder and more controlled conditions while safeguarding the structural fidelity of both lignin and the biomass in order to promote an eco-friendlier lignin recovery. Moreover, this approach yields lignin of superior purity while rendering it highly amenable to subsequent valorization into biofuels or biochemicals. The solvents in this method are generally less toxic than those in traditional pulping and can be reused to reduce environmental impact. However, challenges such as solvent volatility, flammability, and toxicity require strict safety protocols. Additionally, the energy and cost of solvent recovery pose barriers to large-scale adoption. Ongoing research aims to refine solvent selection, improve process efficiency, and enhance sustainability for broader application.^[134] The organic solvent method for lignin recovery shows promises with a lower environmental impact than traditional pulping and excels in high-purity extraction. However, challenges such as solvent toxicity, recycling, and energy use remain. Ongoing advancements in solvent choice and process optimization are set to enhance its feasibility for both lab and industrial applications.

Organic solvent extraction demonstrates various distinctive benefits during the process. The extraction process under relatively mild operating conditions protects the both lignin structure and the biomass components while maintaining extracted material quality. The selection of organic solvents consisting of alcohols, phenols, ketones, and esters creates both solvent-partner affinity and separation efficiency for obtaining pure lignin isolated from plant cell wall composition.^[135] Organic solvent extraction stands as a better environmental choice compared to tough chemical methods that frequently damage both lignin and other

plant substances during treatment. The product obtained from organic solvent extraction matches the requirements for biofuels along with biochemicals because it reaches high purity levels. The recovery of solvents together with their recycling capability helps decrease both environmental damage and operational expenses. The procedure necessitates strict safety procedures because it presents several issues including toxic solvents along with volatile and flammable conditions.^[136] The cost along with the energy requirements to recover solvents act as barriers to large-scale industrial implementation and reduce prospects for massive lignin extraction processes. Scientists currently investigate three main areas to enhance solvent choice selection and optimize procedures and reduce environmental impacts toward large-scale economic sustainability of this technique.^[137]

3.3.2. Enzyme Hydrolysis

Lignin is a highly complex polyphenolic biopolymer in plant cell walls, which has garnered increasing attention for its potential in energy storage due to its dense aromatic framework and unique electrochemical characteristics. To fully exploit lignin's capabilities, adopting advanced extraction techniques is essential. Enzymatic hydrolysis has emerged as a sustainable and progressively viable alternative to traditional chemical methods. This process employs specific enzymes to degrade lignin into smaller and more reactive fragments while enhancing its properties for energy storage applications. Enzyme hydrolysis offers a more selective, energy-efficient, and environmentally benign approach while positioning it as a key strategy for producing high-performance lignin-based electrodes for sustainable energy storage solutions.^[138] This process employs specific enzymes to degrade lignin into smaller, more reactive fragments, enhancing its properties for energy storage applications. Enzyme hydrolysis offers a more selective, energy-efficient, and environmentally benign approach, positioning it as a key strategy for producing high-performance lignin-based electrodes for sustainable energy storage solutions. While the cost is higher than conventional methods, it holds significant promise for bioethanol production and biorefining, yielding high-purity lignin suitable for conversion into high-value bio-based materials.^[139] The eco-friendly nature of enzyme hydrolysis aligns seamlessly with the increasing global demand for sustainable energy storage solutions. By selectively degrading lignin into smaller and functional fragments, this method enhances lignin's accessibility for use in cutting-edge energy storage materials. Unlike chemical methods that typically involve harsh reagents and high energy inputs, enzyme hydrolysis minimizes the reliance on detrimental chemicals, reduces energy consumption, and preserves the integrity of lignin's electrochemical properties. Despite its promise, enzyme hydrolysis does face certain challenges which includes the high cost of enzymes and the difficulties associated with scaling up the process for industrial applications.^[140] However, ongoing advancements in biotechnology, enzyme engineering, and process optimization are poised to address these obstacles while rendering enzyme hydrolysis as a commercially viable option for large-scale lignin extraction. As such, enzyme hydrolysis holds

significant potential to play a pivotal role in the development of sustainable and high-performance eco-friendly energy storage systems.

Enzyme hydrolysis represents a promising extraction method for lignin, which shows benefits through selection and sustainability components. The degradation process through enzymes transforms lignin into reactive fragments that improve its properties suited for energy storage applications.^[141] Through enzyme hydrolysis processes, researchers achieve both sustainable and energy-efficient lignin extraction while decreasing their need for harsh chemicals. Preserving electrochemical lignin properties becomes possible through this approach because it maintains vital properties needed for battery storage devices. Enzyme hydrolysis operates as a process subjected to various operational obstacles. Cost-effective enzyme technology faces hurdles due to production expenses along with processing complexities which stop commercial-scale operations.^[142] The limitations of enzyme hydrolysis become more manageable as nanotechnology and energy storage advances continue to develop. With the advancement of these technologies, enzyme hydrolysis demonstrates promise to become commercially accessible for sustainable high-performance lignin material production.^[143]

3.3.3. Ionic Liquid Method

Ionic liquids (ILs) have attracted considerable attention as solvents for lignin extraction due to their exceptional properties, which includes remarkable thermal stability, negligible vapor pressure, and highly tunable solvation capabilities. These distinctive characteristics render ILs particularly well-suited for lignin extraction from plant biomass, a process that is often complex and technically demanding. Lignin has emerged as a promising candidate due to its rich aromatic content, which holds significant potential for the development of sustainable, high-performance materials for batteries and other advanced energy storage devices.^[144] The ionic liquid-based method for lignin extraction presents several notable advantages over traditional extraction techniques. These include superior selectivity, milder reaction conditions, and the added benefit of ionic liquid recyclability across multiple extraction cycles. By effectively dissolving lignin, ILs facilitate the breakdown of the lignin structure without substantially compromising its functional properties, while enhancing the extracted lignin's suitability for integration into energy storage applications.^[145]

Consequently, the ionic liquid extraction technique emerges as a promising avenue for the development of cost-effective, environmentally benign energy storage materials derived from lignin. However, it is worth noting that many of the methods mentioned above have been primarily designed to optimize the yield and quality of cellulose, with less emphasis on the separation of lignin from its natural polymeric matrix. This has resulted in challenges in obtaining lignin with consistent structural and molecular weight characteristics. Therefore, despite the considerable promise of ionic liquid-based extraction, further refinement of the technique is necessary to ensure the production of lignin with well-defined properties while facilitating its

more widespread application in high-performance energy storage materials. The ionic liquid method for lignin extraction harbors immense promise in advancing the utilization of lignin-derived materials in cutting-edge energy storage technologies. ILs' exceptional ability to efficiently dissolve lignin under mild conditions facilitates the extraction of lignin with minimal degradation of its intrinsic functional properties. In comparison to traditional methodologies, the ionic liquid approach offers enhanced selectivity and sustainability while presenting an ecologically superior alternative that demands lower energy input and relies on fewer deleterious chemicals. However, challenges persist, particularly with regard to the high cost of ILs and their recyclability.^[146] Nonetheless, ongoing research dedicated to refining this process and develop more economically viable ILs holds the potential to increase the scalability of this technique. As technological advancements unfold, ionic liquid-based lignin extraction could emerge as a cornerstone for the production of high-performance and environmentally benign materials tailored for next-generation energy storage systems. Presently, the predominant fate of lignin is its incineration for electricity generation, with a mere 1–2% of the extracted lignin being repurposed as monomers for the synthesis of resins, adhesives, polymeric additives, and adsorbents. A multitude of initiatives has been undertaken to enhance the economic value of lignin, including its conversion into high-carbon lignin-based carbon nanofibers. Although lignin represents a vital renewable resource with expansive potential across diverse industries, its intricate and heterogeneous molecular structure presents formidable challenges in the extraction process. The choice of extraction technique hinges on several variables, such as the nature of the biomass, the required purity of the lignin, and the associated economic considerations. Future advancements in lignin extraction will likely concentrate on optimizing process efficiencies, minimizing environmental impact, and unlocking the potential of lignin for the production of high-value materials. As the global demand for sustainable alternatives to fossil-based products intensifies, the development of refined lignin extraction techniques will be indispensable in realizing the full potential of this versatile biopolymer.^[147]

The ionic liquid (IL) extraction approach for lignin recovery has become prominent due to its multiple exceptional characteristics including superior thermal stability, minuscule volatility, and adjustable solvent characteristics. The extraction of lignin with ILs occurs under gentle temperature conditions that preserves functional properties and enables high-performance material production suitable for energy storage applications.^[148] Researchers value the ionic liquid extraction approach because of its excellent selectivity and repeated recycling capability of ILs, which generates both economic and sustainable advantages. The ionic liquid extraction method offers a sustainable solution through low-energy requirements and minimal chemicals that harm the environment as compared to conventional techniques. The ionic liquid method encounters various obstacles for its implementation.^[149] The major obstacles include the expensive nature and the difficulties in recycling these solutions. The successful extraction outcomes demand further research to optimize the technique for achieving consistent lignin quality as well as yield

consistency. Additional research should be conducted to discover affordable ILs alongside optimization of extraction processes in order to achieve broader application and scalability in lignin-based energy storage technologies.

Hence, multiple synthesis techniques provide a comparative analysis for lignin extraction while highlighting their strengths and limitations. The organic solvent method is praised for its milder conditions, lower environmental impact, and ability to yield high-purity lignin while making it an effective and eco-friendly choice, though challenges such as solvent toxicity, volatility and the cost of solvent recovery remain. Enzyme hydrolysis, while environmentally benign and energy-efficient, is hindered by the high cost of enzymes and difficulties in scaling the process for industrial use.^[150] The ionic liquid method offers excellent selectivity, recyclability, and sustainability with minimal lignin degradation, but its high cost and the need for process optimization limit its practical application. Each method presents unique advantages for specific applications, yet all face barriers that require further research and technological improvements to enhance scalability, cost-effectiveness, and overall viability in lignin extraction.

Lignin offers several distinct cost advantages over traditional carbon-based materials while making it an attractive alternative for energy storage applications. One of the most significant cost benefits of lignin lies in its abundant availability as a by-product of the pulp and paper industry, which means it can be sourced at a relatively low cost.^[151] Unlike other high-performance carbon-based materials which require complex and expensive extraction or manufacturing processes, lignin is extracted through relatively simple and well-established methods during the pulping process. This gives lignin a clear economic edge over other materials, which can be costly due to its mining, refining and purification stages. In terms of raw material costs, lignin is typically far less expensive than synthetic carbon materials like graphene and CNTs.^[152] Graphene requires energy-intensive processes such as chemical vapor deposition (CVD) or liquid-phase exfoliation while contributing to high production costs. CNTs also require sophisticated synthesis methods, such as CVD or arc discharge while leading to higher prices. In contrast, lignin can be sourced directly from renewable biomass and processed in a more cost-effective manner.^[153] Additionally, lignin's potential for large-scale production is enhanced by the established infrastructure in the paper and forestry industries, which already handle massive quantities of biomass annually.

Additionally, the carbon content in lignin and its potential for conversion into advanced carbon products enhances its economic value. Through carbonization or activation, lignin can be a low-cost precursor to carbon electrodes utilized in batteries. This differs from other materials like activated carbon and graphene which have further processing steps that raise the cost value. Activated carbon is frequently produced from pricier materials such as coal or coconut shells and undergoes chemical activation at high temperatures using expensive materials. These cost comparisons also accentuate the benefits of lignin in terms of its cost and environmental impact. Other carbon materials, especially synthetic ones, undergo harsh extraction and refinement processes including toxic chemicals, substantial energy inputs,

and elevated costs while leading to greater environmental concerns. Unlike these, lignin is a renewable material that can be sustainably harvested without the potential damage environmentally harmful practices. This strengthens its associated external costs case. Additionally, these benefits can readily appeal to markets that help promote materials with sustainability certifications while supporting green energy initiatives.^[153]

4. Lignin-Derived Electrode Materials for LIBs

The escalating global demand for sustainable and highly efficient energy storage solutions has catalyzed the exploration of alternative materials for LIBs. Among the diverse range of candidates, lignin has garnered considerable attention as a promising source for electrode materials. Lignin is an intricate and three-dimensional polyphenolic compound primarily derived from the pulp industries and various biomass processing technologies. Its high carbon content, coupled with its multifaceted chemical structure, positions it as an ideal precursor for the synthesis of carbon-based materials, particularly for use in energy storage systems.^[154] In recent years, there has been a burgeoning scholarly focus on leveraging the distinctive attributes of lignin to engineer cutting-edge electrode materials for LIBs. These lignin-derived materials present numerous advantages, such as unmatched environmental sustainability, cost-efficiency, and the promise of superior electrochemical performance. Lignin-based electrodes are endowed with an array of desirable properties, which include tunable porosity, abundant functional groups, and the potential for structural customization.^[155] Transforming lignin into high-performance electrodes involves the application of various advanced preparation techniques, which include pyrolysis, hydrothermal carbonization, and chemical activation. These approaches not only optimize the structural and electrochemical properties of lignin-derived materials but also facilitate surface functionalization while enhancing charge storage capacity and cycling stability. This section delves into the considerable potential of lignin as a foundational material for LIB electrode fabrication while reviewing key advancements in its preparation and modification. It further explores the material's viability for large-scale energy storage.

4.1. Pristine-Lignin Electrodes

The pursuit of eco-friendly and high-efficiency materials for LIBs has catalyzed the investigation of unconventional electrode substances while surpassing the conventional graphite and metal oxide paradigms. Lignin is a multifaceted and copious biopolymer, which has attracted considerable scholarly interest owing to its renewability and its promising electrochemical prowess. Pristine lignin offers a unique combination of environmental benefits, structural versatility, and electrochemical activity. As a naturally occurring, carbon-rich material, lignin-derived electrodes can provide a cost-effective, sustainable alternative for the anodes and cathodes in LIBs. This section discusses the potential of pristine lignin as a promising electrode material while focusing on

its intrinsic properties, challenges in processing and performance optimization and recent advancements in its application for next-generation LIBs. By harnessing the full potential of pristine lignin, it is possible to create eco-friendly and high-capacity energy storage solutions that could contribute to the development of more sustainable energy systems.^[156] Lignin is extracted from biomass and pyrolyzed into a carbon-rich material which is then processed into electrode materials for LIBs.^[157] Lignin-based materials boast a high specific surface area, superior conductivity, and rapid lithium-ion diffusion while enabling exceptional capacity and cycling stability for LIB anodes.^[158]

Lignin is widely recognized as a potential precursor for hard carbon (HC) as the straightforward carbonization of unmodified lignin yields a product with inadequate surface area and porosity. In response to this challenge, Li et al. pioneered a sophisticated method for synthesizing lignin-derived porous carbon while employing a self-template approach coupled with surface modification in order to enhance the material's structural attributes and performance (Figure 4a).^[159] Oxygen functionalized groups are strategically incorporated to fine-tune the surface chemistry of lignin, while metal ions are deliberately selected to coordinate with these oxygenated sites while promoting the formation of carbonates that function as self-templates to meticulously regulate the pore structure. Moreover, the aromatic framework of lignin serves to spatially disperse the metal ions while facilitating the creation of homogeneously distributed pore-forming sites, as confirmed by scanning electron microscopy (SEM) imaging (Figure 4b). As an anode, lignin-derived porous carbon (CCL) exhibited a notable specific capacity of 500 mAh g⁻¹ at a current density of 50 mA g⁻¹. It remarkably preserved 99% of its capacity even after 1000 cycles at a rigorous current density of 1000 mA g⁻¹, while showcasing an exceptional rate performance across a range of current densities (Figure 4c,d). This research introduces a cost-effective anode for LIBs and proposes a regulatory framework for biomass-derived carbon while reducing lignin waste and mitigating environmental pollution. It demonstrates an advanced technique for synthesizing lignin-based porous carbon using a self-template method. Through the introduction of oxygen-functionalized groups and the coordination of metal ions, the pore structure of the carbon material is meticulously controlled. This strategy offers a low-cost, sustainable solution for LIB anodes while reducing lignin waste and its environmental impact. Without performance data, the focus is on the novel method for creating bio-based carbon with tailored pore structures for energy storage applications.

Lignin-based HC presents substantial potential as an energy storage material. However, directly carbonizing lignin often fails to yield the desired electrochemical performance. Du et al. introduced an innovative pre-oxidation strategy which effectively augments the reversible capacity of HC (Figure 5a).^[160] The alkyl groups are converted into peroxides and alcohols while facilitating intermolecular cross-linking within the lignin framework. This transformation leads to the formation of a material with a highly disordered nanotexture while manifesting a densely porous structure, as revealed by SEM imaging (Figure 5b). Owing to these structural enhancements, the optimized HC exhibited a superior Li⁺ storage performance while delivering a reversible capacity of

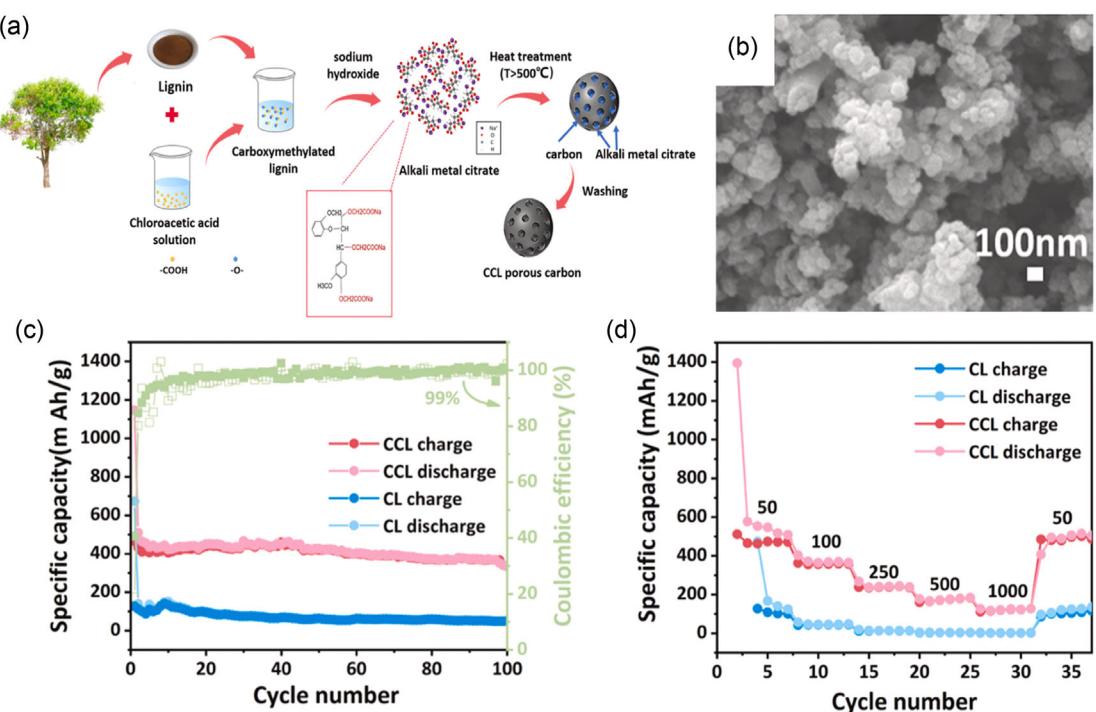


Figure 4. a) Schematic diagram depicting the preparation process of CCL. b) SEM image showing the surface morphology of the CCL electrode. c) Cycling performance of the synthesized electrodes. d) Rate performance of CCL and CL measured at different current densities. Adapted with permission.^[159] Copyright 2022, Elsevier.

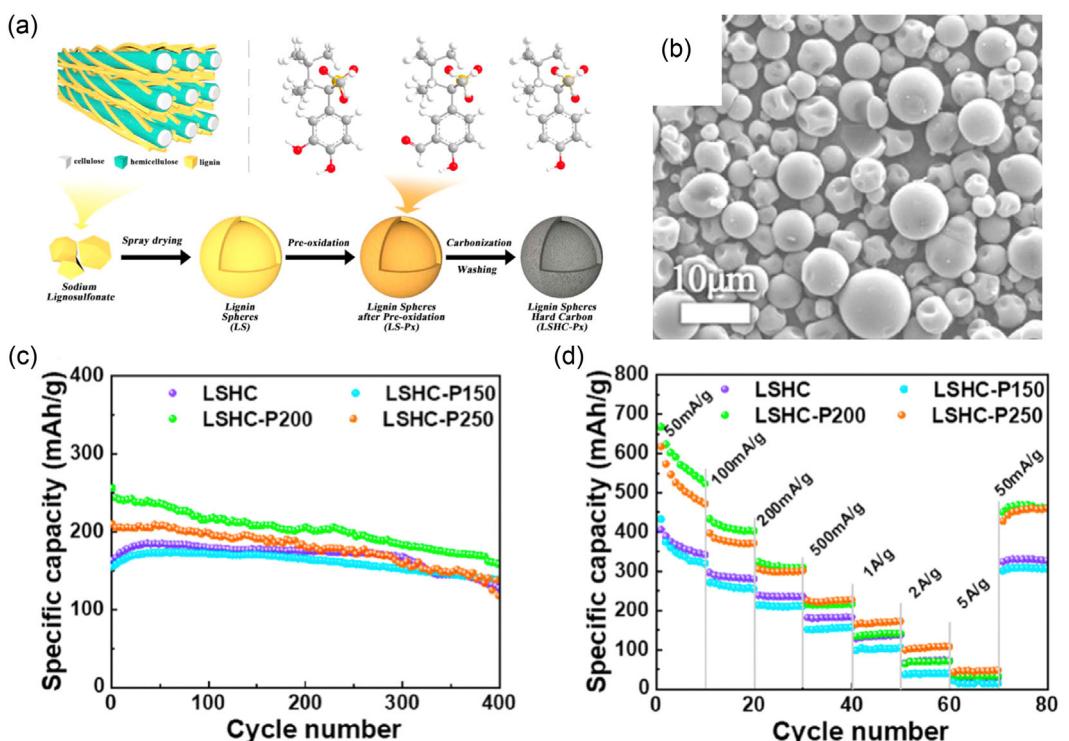


Figure 5. a) Schematic representation of the LSHC-P200 preparation process. b) SEM image showing the surface structure of the LSHC-P200 electrode. c) Cycling stability of the synthesized electrodes. d) Rate capability of the synthesized electrodes tested at different current densities. Adapted with permission.^[160] Copyright 2021, Elsevier.

$\approx 190 \text{ mAh g}^{-1}$ after 400 cycles, coupled with outstanding rate capabilities (Figure 5c,d). This research furnishes profound insights into the deliberate engineering of high-performance anodes for LIBs. It elucidates a pre-oxidation approach that substantially enhances the electrochemical properties of HC while elevating its potential for a wide array of energy storage uses. By transforming alkyl groups into peroxides or alcohols and promoting intermolecular cross-linking, the process leads to a material with a highly random nano-texture and abundant porosity. These structural improvements are key to enhance the capacity and stability of carbon anodes. The study provides valuable insights into designing high-performance HC anodes for LIBs and other energy storage systems.

Lignin-based porous carbon (LPC) is recognized for its sustainability and eco-friendliness, which presents a considerable promise as an anode material for LIBs. Nevertheless, its potential is constrained by the inherent heterogeneity of the lignin precursor. Hence, a comprehensive understanding of the interconnections between lignin properties, the architecture of the porous carbon, and the kinetics of Li^+ storage is of paramount importance. In this regard, Wu et al. fabricated LPCs from fractionated lignin through the use of an environmentally benign and recyclable activating agent while proposing a novel strategy to enhance their performance in energy storage applications (Figure 6a).^[161]

The architectural framework of the LPCs was meticulously engineered through the strategic modulation of the lignin macromolecules' molecular weight, linkage density, and glass transition

temperature while facilitating the emergence of a complex and three-dimensional flower-like structure. This nuanced manipulation of the lignin's intrinsic molecular properties served to direct the formation of an extraordinarily sophisticated porous morphology while optimizing both the structural integrity and electrochemical performance of the material (Figure 6b). As an anode, the meticulously crafted LPCE-70 exhibited a remarkable reversible capacity of 528 mAh g^{-1} at a current density of 0.2 A g^{-1} after 200 cycles while showcasing outstanding rate capabilities. This extraordinary combination of high-capacity retention and rapid charge-discharge performance underscores the material's exceptional electrochemical stability while highlighting its potential for sustained and high-efficiency operation in advanced energy storage systems (Figure 6c,d). The lignin-based LPCE-70 electrode enhances Li^+ storage via diffusion and capacitive mechanisms. This work presents a novel approach to synthesize LPC with a 3D flower-like structure by optimizing lignin's molecular properties in order to maximize the lithium-ion capacity. It emphasizes the importance of molecular tailoring for improving LPC as an anode material in LIBs.

Pristine-lignin derived electrodes present a promising and sustainable alternative for LIBs while offering significant potential to address the growing demand for eco-friendly, cost-effective energy storage solutions. While challenges remain in optimizing the electrochemical performance, recent advancements in material design, functionalization, and structural modification have demonstrated the feasibility of lignin as a viable candidate for

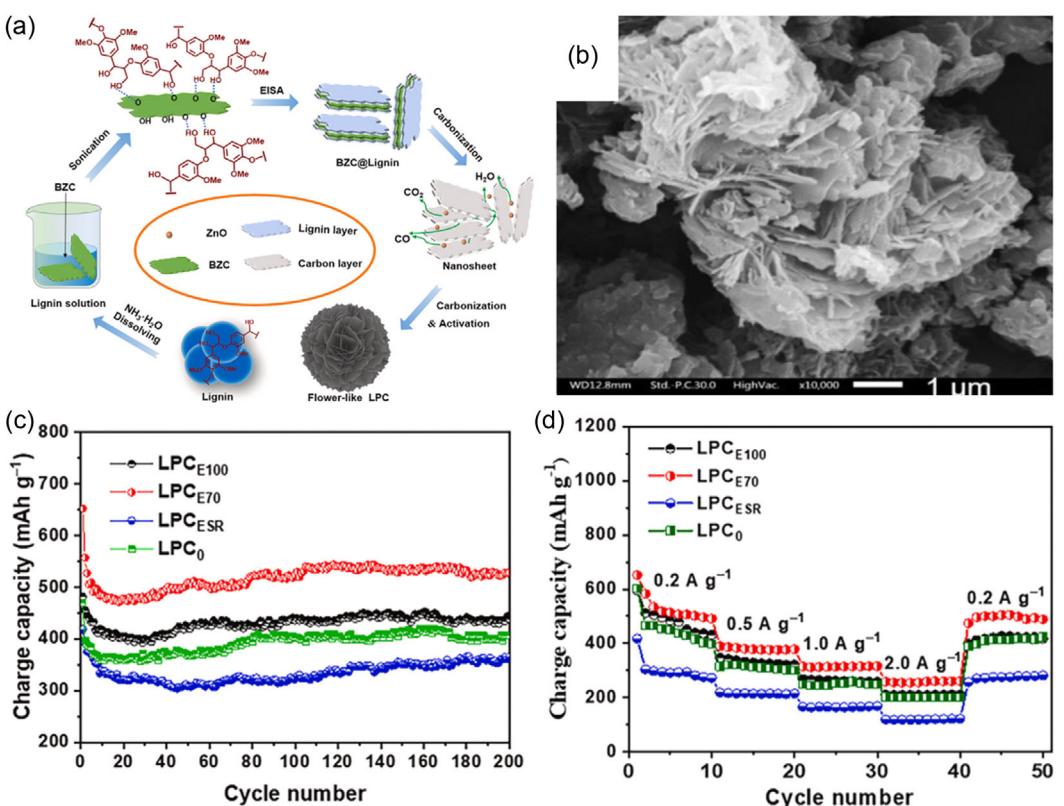


Figure 6. a) Schematic diagram outlining the preparation process of the LPC composite. b) SEM image illustrating the surface morphology of the $\text{LPC}_{\text{E}70}$ electrode. c) Cycling performance of the synthesized electrodes. d) Rate performances comparison of the synthesized electrodes at different current densities. Adapted with permission.^[161] Copyright 2023, Elsevier.

high-performance LIBs. The unique properties of lignin position it as an attractive material for next-generation battery technologies. Continued research into the development of pristine-lignin derived electrodes while improving their conductivity, stability, and energy density will be crucial to unlock their full potential.

This section explores the potential of lignin-derived materials for lithium-ion battery (LIB) electrodes while highlighting various strategies to enhance their electrochemical performance. While lignin is an attractive precursor for energy storage materials, its raw form lacks the necessary structural properties for effective use in LIBs. To address this, three distinct approaches are used to improve lignin-based materials, which include self-template carbonization, pre-oxidation, and molecular fractionation. Li et al. employed a self-template method with surface modification, resulting in lignin-derived porous carbon (CCL), which showed a remarkable specific capacity of 500 mAh g^{-1} at 50 mA g^{-1} and 99% capacity retention after 1000 cycles at 1000 mA g^{-1} , offering an excellent balance of high capacity and long-term stability. Du et al. used pre-oxidation to modify lignin's molecular structure, promoting cross-linking and enhancing porosity, resulting in lignin-derived HC with a reversible capacity of 190 mAh g^{-1} after 400 cycles, though its capacity and rate performance were lower than CCL. Lastly, Wu et al. fractionated lignin to optimize its molecular structure before creating a 3D flower-like porous carbon (LPC), which achieved a reversible capacity of 528 mAh g^{-1} at 0.2 A g^{-1} , surpassing both the pre-oxidized HC and CCL in terms of capacity. However, the cycling stability and rate performance of LPC were slightly less impressive than that of CCL. These findings suggest that while each method offers distinct advantages, the self-template carbonization approach appears to be the most promising for high-performance LIB anodes. Through a detailed comparison of these experimental data, this section concludes that each modification method brings its own set of strengths and weaknesses. The overall analysis suggests that surface modification, molecular fractionation, and pre-oxidation are all viable strategies for enhancing the electrochemical properties of lignin-derived materials. However, further refinement and optimization of these techniques are necessary to fully harness the potential of lignin for sustainable and high-performance energy storage applications.

Research on pristine lignin-derived materials for LIBs ought to look into retention of electrochemical activity while improving material synthesis for better efficiency and environmental impact. One promising direction would be the development of hybrid modification strategies that self-template carbonization with molecular fractionation or pre-oxidation techniques, maximizing each method's strengths. Self-doping frameworks may also assist in framework templating for enhancing functionality of structural morphology to deliver superior conductivity, ion diffusion, and capacity retention. Moreover, examining the electrocatalytic capabilities of various metal ion dopants within self-template methods may offer other tailored properties for specific LIB functionalities. Another focus of interest would include scaling of these pristine lignin-based materials while aiming for greener synthesis routes as an area for further exploration. Sophisticated LPC coatings may also be responsible for enhancement of cycling stability and rate performance alongside their hybrids such as

graphene or CNTs to bolster wrought preferential alignment and stratified structural integrity. Expanding the scope of lignin exploitation from various sources of plant waste may also aid in capturing economics of sustainable high performance anode feedstocks. These endeavors will be most helpful to promote lignin based materials as functional substitutes for sustainable energy storage systems.

4.2. Lignin-Silicon Oxide/Zinc Oxide Electrodes

The development of advanced electrode materials for LIBs is essential to meet the growing demand for high-energy-density, long cycle life, and environmentally sustainable energy storage solutions. A promising approach involves using composite materials to improve the performance of LIBs. Lignin offers strong sustainability benefits, while silicon oxide and zinc oxide are known for their high capacity and stability in energy storage. Combining lignin with these oxides creates a synergistic effect while enhancing conductivity, structural strength, and overall battery performance. This section explores the potential of lignin-based composites with silicon oxide and zinc oxide as innovative electrode materials for LIBs while highlighting their unique properties, challenges in design, and progress in optimizing their performance for future energy storage technologies. Through these innovations, lignin-silicon oxide/zinc oxide composites may contribute to more efficient, sustainable, and high-capacity LIBs. Lignin-extracted carbon is widely acknowledged as one of the most promising materials for electrochemical energy storage owing to its sustainability, cost-effectiveness, and excellent conductivity. However, its performance in LIBs is severely limited by the irregular microporosity that impedes its Li^+ storage capabilities. In an innovative approach, Huang et al. synthesized a porous carbon, encapsulated with SiO_2 (denoted as LHC/ SiO_2 -21), which possesses a highly intricate three-dimensional honeycomb mesoporous structure. This was achieved via a dual-template-assisted self-assembly strategy while representing a cutting-edge method to overcome the structural limitations of biomass-derived carbon and significantly enhance its electrochemical performance for LIBs (Figure 7a).^[162] The intricately ordered mesoporous framework synergistically facilitated expedited lithium-ion diffusion kinetics while simultaneously providing a significantly enhanced density of available lithium-ion storage sites as shown by the SEM imaging (Figure 7b). When utilized as an anode, LHC/ SiO_2 -21 manifested an extraordinary reversible capacity of 1109 mAh g^{-1} , coupled with exceptional rate performance and long-lasting cycle stability while underscoring its formidable potential for sustained energy storage (Figure 7c,d). This work demonstrates a dual-template-assisted self-assembly strategy to synthesize biomass-based electrodes. The resulting material features a well-ordered and three-dimensional mesoporous structure that promotes fast Li^+ diffusion. The study emphasizes the importance of structural design in enhancing the electrochemical performance of lignin-derived electrode materials as anodes in LIBs while showcasing how carefully engineered mesoporous architecture can improve lithium-ion storage capabilities.

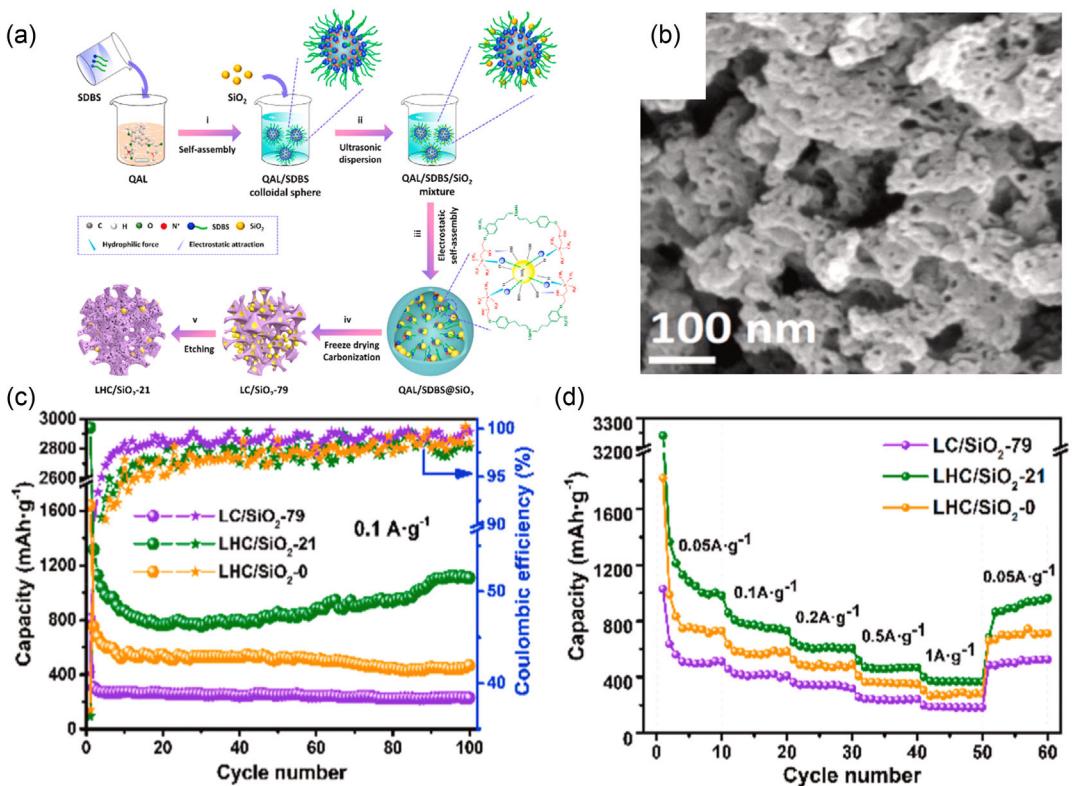


Figure 7. a) Schematic illustration depicting the fabrication process of the LHC/ SiO_2 -21. b) SEM image showcasing the surface morphology and microstructure of the LHC/ SiO_2 -21. c) Electrochemical cycling performance of the synthesized electrodes. d) Comparative rate performance of electrodes at varying current densities. Adapted with permission.^[162] Copyright 2021, Elsevier.

Lignin has been extensively explored as a high-quality carbon source in the synthesis of carbon electrodes. However, due to the inherently disordered, three-dimensional network structure of lignin molecules, conventional carbonization techniques are ill-suited for the production of high-performance porous carbon electrodes with well-ordered architectures. To address this limitation, Xi et al. proposed a novel lignin-based porous carbon (HCSF) to significantly enhance the electrochemical performance of LIBs. Utilizing electrostatic forces, lignin/silica nanocomposites were synthesized through an *in situ* self-assembly and co-precipitation method while employing quaternary ammonium-functionalized lignin and sodium silicate as precursors. Following carbonization and activation, HCSFs were obtained while offering a promising solution to the structural and electrochemical challenges faced by traditional lignin-derived carbon materials (Figure 8a).^[163]

The special hollow shell structure of HCSFs was also shown using SEM (Figure 8b). The fabricated HCSFs exhibited an exceptional electrochemical durability in order to preserve a reversible specific capacity of 480 mAh g⁻¹ after 200 cycles at a current density of 200 mA g⁻¹ while simultaneously demonstrating remarkable rate performance as an anode material in LIBs (Figure 8c,d). The enhanced energy storage capabilities of these materials can be chiefly attributed to the unique structure of the HCSFs, which provides optimal channels for rapid ionic diffusion. Additionally, the nitrogen doping effectively amplifies the number of electrochemical active sites while augmenting their performance.

This work epitomizes the pioneering advancement of lignin-based porous carbon frameworks (HCSFs) while showcasing their potential to markedly elevate the electrochemical efficiency of LIBs. An *in situ* self-assembly and coprecipitation method is used to generate lignin/silica nanocomposites, which were then carbonized and activated to form HCSFs with hollow porous structures enriched with nitrogen and SiO_2 . The unique hollow shell structure facilitates rapid ion transport while enhancing the material's electrochemical performance as an anode in LIBs. This study highlights the potential of engineered porous structures and elemental doping to improve energy storage materials. ZnO has garnered attention as a potential anode material owing to its impressive theoretical capacity, low operating potential, abundant natural availability, and nontoxic nature. However, its considerable volumetric expansion during cycling leads to pulverization and agglomeration while undermining its performance. In response to this challenge, Yu et al. innovatively synthesized a porous carbon using rice husk (RH) lignin, upon which ZnO nanoparticles were uniformly anchored while reinforcing the material's structural resilience and optimizing its electrochemical functionality (Figure 9a).^[164]

The singular porous morphology imparted remarkable stability and superior electrical conductivity while substantially elevating the material's structural integrity and electrochemical efficiency (Figure 9b). The integration of RH as a carbon source markedly augmented the biomass materials' efficacy in the refining process. The resulting samples exhibited highly promising Li⁺

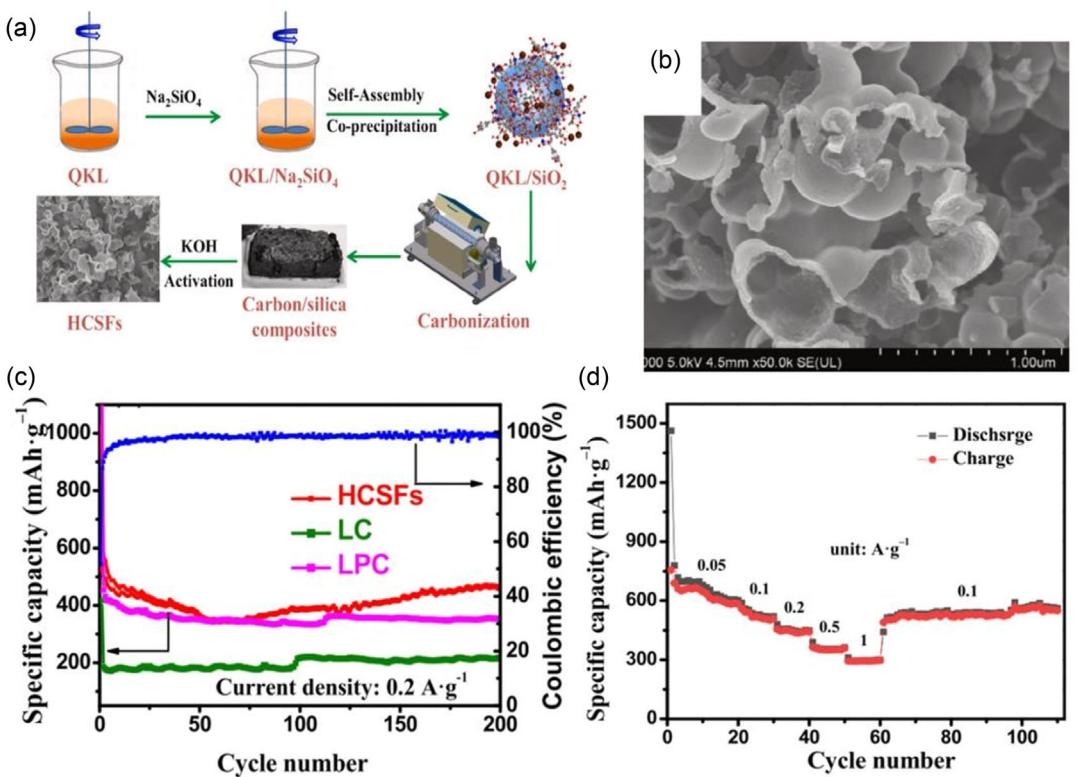


Figure 8. a) Schematic diagram of the HCSFs preparation process. b) SEM image showing the surface of the HCSFs electrode. c) Cycling performance of the synthesized electrodes. d) Rate performance of HCSFs at different current densities. Adapted with permission.^[163] Copyright 2021, Elsevier.

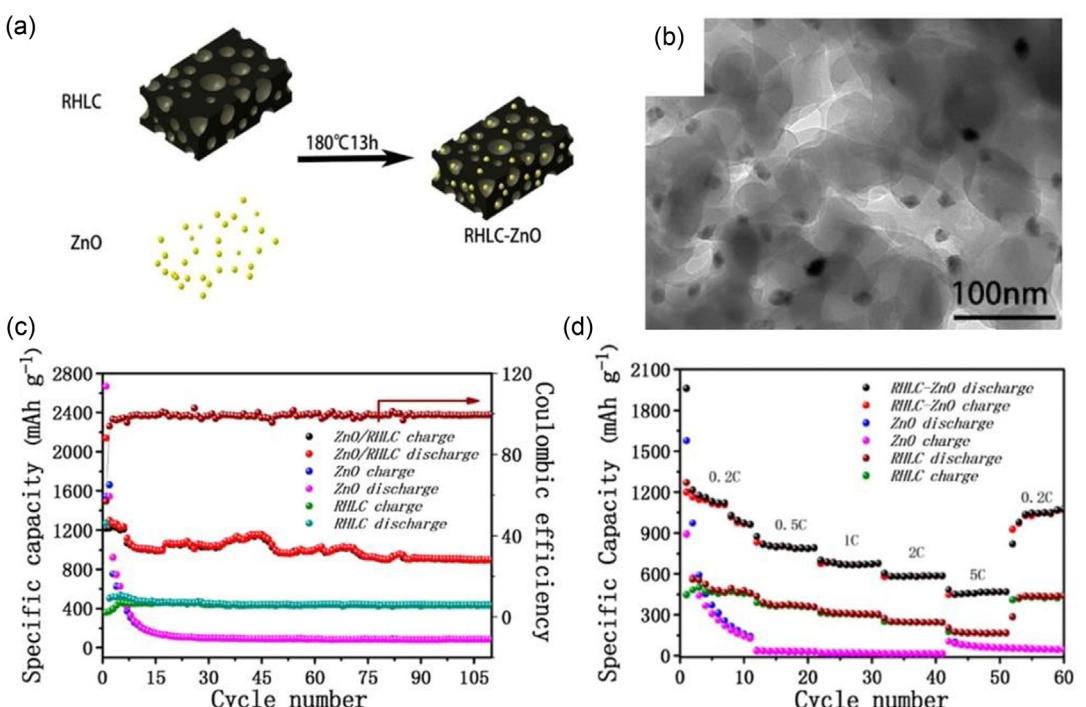


Figure 9. a) Schematic diagram illustrating the preparation steps of RHLC-ZnO. b) SEM image revealing the surface morphology of RHLC-ZnO electrode. c) Cycling performance of the synthesized electrodes. d) Rate performance comparison of the synthesized electrodes at various current densities. Adapted with permission.^[164] Copyright 2020, Springer.

storage properties and stellar electrochemical performance while achieving a discharge capacity of 898 mAh g^{-1} at a 0.2C after 110 cycles, coupled with exceptional rate retention (Figure 9c,d). This investigation showcases the fabrication of a porous carbon framework synthesized from RH lignin, which acts as a resilient matrix for the uniform embedding of ZnO nanoparticles. The resultant composite material harmoniously combines the inherent advantages of the carbon skeleton's intricate porous architecture with the functional properties of the ZnO nanoparticles while offering superior stability and enhanced electrical conductivity. The strategic use of rice husk as a carbon source not only maximizes the efficient utilization of biomass feedstocks but also aligns with sustainability principles while rendering the process more ecologically viable. The research accentuates the formidable potential of this composite material as an anode for LIBs, with a particular focus on its intricately devised structural framework aimed at bolstering both stability and electrochemical performance during prolonged cycling. In an innovative advancement, Xi et al. engineered a large-scale and hierarchically porous carbon material through a cost-efficient and eco-friendly activation methodology. This approach harnesses lignin in conjunction with ZnCO_3 while facilitating the creation of a high-performance and sustainable material (Figure 10a).^[165]

The three-dimensional innovative hierarchical carbon (HLPC) boasted a highly desirable microstructural architecture with an abundance of mesopores that substantially augmented its intrinsic properties and performance as shown through SEM (Figure 10b). This architecture is meticulously realized through the activation

process alongside the in situ formation of ZnO nanoparticles while templating an influence on the structure. As a result, the resultant hierarchical HLPC achieved a remarkable specific capacity of 550 mAh g^{-1} after 200 cycles at a current density of 0.2 A g^{-1} while demonstrating the exceptional rate performance (Figure 10c,d). The volumetric capacitances realized in this investigation stand as the most elevated among all reported lignin based carbon materials while setting a new benchmark in the field. In contrast to conventional activation agents, ZnCO_3 emerges as a highly sustainable, noncorrosive, cost-effective, and recyclable alternative while offering expansive potential for the fabrication of biomass-based carbon materials. This study exemplifies the synthesis of a hierarchical porous carbon while utilizing ZnCO_3 within an environmentally conscious activation process. The resultant HLPC showcased an intricate hierarchical framework while brimming with a dense network of mesopores. The research highlights the remarkable potential of ZnCO_3 as a green and economically viable activator for producing biomass-derived carbon materials while presenting an innovative and effective strategy for designing materials with precisely tailored structural characteristics for advanced energy storage applications.

Despite being one of the most promising anode materials for LIBs, the commercialization of silicon (Si) is fraught with numerous formidable challenges. In this regard, He et al. pioneered a remarkably simple yet highly effective approach that markedly ameliorates the cycling stability of Si-based anodes while offering a promising solution to some of the material's most pressing issues (Figure 11a).^[166] A distinctively porous yet densely compact

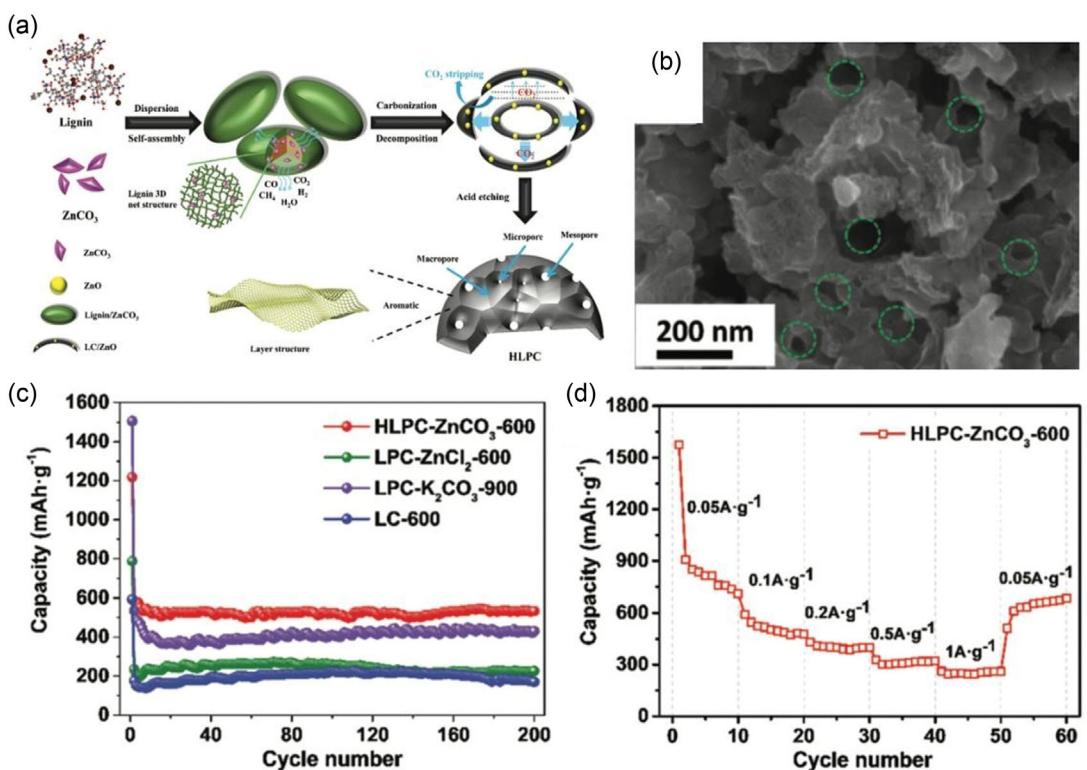


Figure 10. a) Schematic representation of the fabrication process for HLPC-ZnCO₃. b) SEM image highlighting the surface morphology and structural features of HLPC-ZnCO₃ electrode. c) Long-term cycling stability and performance of the synthesized electrodes. d) Rate performance of HLPC-ZnCO₃ at various current densities. Adapted with permission.^[165] Copyright 2020, Royal Society of Chemistry.

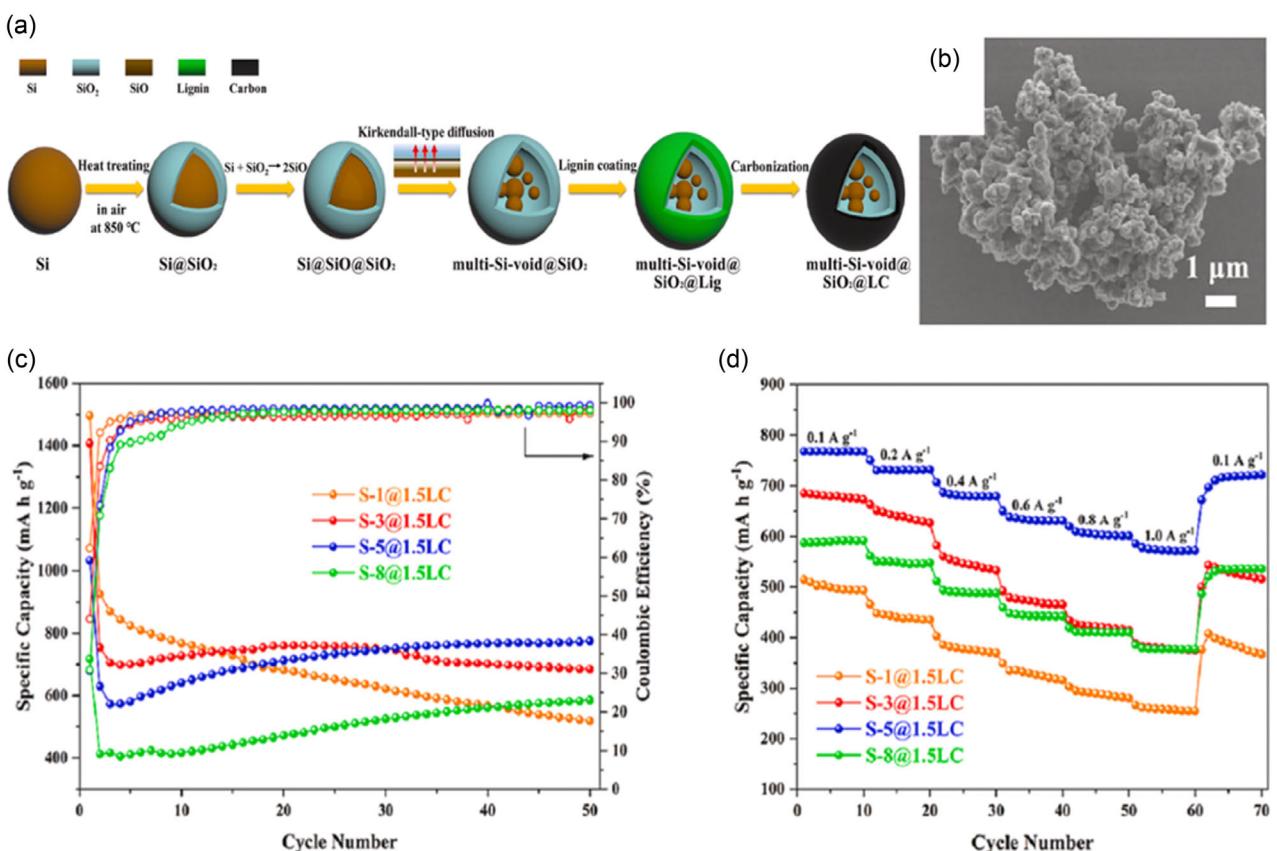


Figure 11. a) Schematic diagram depicting the stepwise preparation process of Si-5@1.5LC. b) SEM image illustrating the detailed surface morphology of Si-5@1.5LC electrode. c) Electrochemical cycling stability of the synthesized electrodes. d) Comparative rate capabilities of the synthesized electrodes under different current densities. Adapted with permission.^[166] Copyright 2023, Elsevier.

morphology was unveiled through SEM while showcasing an elaborate structural configuration characterized by a high degree of porosity alongside remarkable compactness (Figure 11b). The ideal electrode material exhibited remarkable durability and sustained a specific capacity of 759 mAh g⁻¹ after 1300 cycles at a current density of 1.0 A g⁻¹ while showcasing exceptional rate performance (Figure 11c,d).

The exceptional electrochemical performance arises from the synergistic interaction of four components (voids, silicon, silicon dioxide, and lignin-based carbon), which enhance cycle stability. Si nanoparticles were first transformed into a multi-Si-void@SiO₂ structure, then lignin was coated onto this structure through acid precipitation and hydrogen bonding. In situ carbonization resulted in the multi-Si-void@SiO₂@lignin-based carbon composite. This lignin-silicon oxide and lignin-zinc oxide composite hold promise for sustainable LIB materials. The integration of lignin with silicon oxide and zinc oxide enhances both the environmental sustainability and electrochemical performance of electrode materials. This synergy combines lignin's carbon structure with the mechanical reinforcement from silicon and zinc oxides. While challenges in synthesis and processing remain, ongoing research aims to improve conductivity, stability, and mitigate volume expansion. With further advancements, lignin-based composites could significantly advance LIB performance and contribute to more sustainable energy storage solutions.

The development of advanced electrode materials for LIBs is crucial to meet the increasing demand for energy storage solutions with higher energy density, longer cycle life, and sustainability. Composite materials, particularly those combining lignin with oxides like silicon oxide (SiO₂) and zinc oxide (ZnO), offer a promising route to enhance LIB performance by improving conductivity, structural integrity, and overall battery performance. Lignin faces limitations due to its irregular microporosity, which restricts lithium-ion storage. Various approaches have been explored to address these challenges, such as Huang et al.'s dual-template-assisted self-assembly of LHC/SiO₂-21, which featured a mesoporous structure that improved lithium-ion diffusion and achieved a high reversible capacity of 1109 mAh g⁻¹ after 200 cycles. Similarly, Xi et al.'s synthesis of HCSF using electrostatic forces and in situ self-assembly produced a hollow shell structure with enhanced ionic diffusion while yielding 480 mAh g⁻¹ after 200 cycles and benefiting from nitrogen doping to improve electrochemical performance.

Incorporating ZnO nanoparticles into lignin-based materials has also shown promise, with Yu et al.'s RHLC-ZnO composite exhibiting excellent structural stability and a discharge capacity of 898 mAh g⁻¹ after 110 cycles, thanks to the synergy between the porous structure of rice husk lignin and ZnO. Furthermore, the Si-5@1.5LC composites developed by He et al. integrating voids, silicon, SiO₂, and lignin-based carbon, demonstrated enhanced

cycling stability and a specific capacity of 759 mAh g^{-1} after 1300 cycles while emphasizing the importance of structural design for improving performance. These studies underscore the critical role of structural and chemical modifications, such as mesoporous architecture, hollow structures, and the integration of ZnO and SiO_2 , in enhancing the electrochemical performance of lignin-based composites while offering promising solutions for the next generation of LIBs.

Research goals should aim toward the improvement of the integration of lignin and advanced nanostructured materials to maximize performance of LIBs. In particular, creating new composite materials with lignin while using transition metal oxides or phosphates could greatly improve energy density and capacity retention. Furthermore, modification of the fabrication methods can result in greater structural uniformity and stability while leading to improved cycling performance. Another way of augmenting the charge transfer rate and discharging rate of lithium ions is by employing templating agents or selective etching methods to improve the porosity and surface area of lignin based composites. Further studies on the nitrogen, sulfur, and phosphorus doping of the lignin are needed to enhance the conductivity and electrochemical stability. Also, the addition of conductive carbon materials such as graphene or CNTs into electrodes could increase the overall conductivity and cycling stability of the electrodes. Lastly, significant efforts must be directed toward scale and scope while incorporating waste lignin from different industries, which would reduce production costs and offset the negative environmental impacts of LIBs. Hence, comprehensive studies on the long-term performance, rate capability, and real-world applicability of these composites in full-scale batteries will be essential to validate their commercial potential.

4.3. Lignin-Si Electrodes

Lignin-derived electrodes for LIBs have attracted substantial interest as a viable strategy to bolster the sustainability of energy storage systems. Lignin serves as a carbonaceous precursor that can be converted into a highly conductive and stable material suitable for LIB applications. When amalgamated with silicon, lignin mitigates inherent challenges. The synergy between lignin and silicon in composite electrodes capitalizes on the individual strengths of both while fostering materials that are not only ecologically advantageous but also exhibit enhanced performance. This discourse explores the promise of lignin-silicon composite electrodes while emphasizing their combined advantages and recent strides toward high-performance energy storage solutions. With ongoing research, lignin-silicon electrodes could emerge as a cornerstone for sustainable LIBs. Despite silicon's remarkable theoretical electrochemical capacity, its commercialization is impeded by significant challenges which include volumetric expansion during cycling, poor cycling stability, and low initial Coulombic efficiency. To overcome these challenges, carbon materials provide an effective solution by serving as a mechanical buffer to mitigate the volumetric expansion of silicon while simultaneously enhancing the overall electrical conductivity of the anode. This dual role enables these materials to support an

exceptionally high specific capacity and substantially improve cycling stability. In this regard, Li et al. innovatively utilized a lignin-derived resin as a carbon source and coupled with silicon to form silicon-based composite. The resulting composite not only addresses the critical issues of silicon anodes but also offers a pathway toward more efficient, stable, and sustainable LIBs (Figure 12a).^[167]

The resin derived from lignin was methodically deposited onto the surface of silicon nanoparticles and the carbonaceous layer was synthesized through the pyrolysis of the resin. The morphological and compositional characteristics of the carbon-silicon architecture were rigorously investigated using SEM. The SEM analysis unequivocally demonstrated that the carbon layer was seamlessly and uniformly deposited over the silicon nanoparticles while forming an even and coherent coating (Figure 12b). The electrochemical characteristics of the samples were thoroughly examined. When contrasted with pure phenolic resin-coated silicon, the cycling performance of Si/C-LPR composite showed a profound improvement. Specifically, the Si/C-LPR composite exhibited an outstanding electrochemical performance while achieving an initial specific capacity of 782 mAh g^{-1} , which remarkably sustained 605 mAh g^{-1} after 100 cycles, alongside exceptional rate capabilities (Figure 12c,d). This work demonstrates the development of a lignin-based phenolic resin composite material for enhancing the electrochemical performance of silicon anodes in LIBs. The study highlights how the lignin-based phenolic resin improves the cycling stability and electrochemical properties of silicon anodes through enhanced cycling performance and rate capability compared to conventional phenolic resin coatings. Yang et al. meticulously engineered a silicon/carbon hollow microsphere through a refined spray-drying process while harnessing recycled silicon and lignin feedstocks (Figure 13a).^[168]

Morphological and structural characterizations elucidate that the micron-sized silicon particles provide ample nucleation sites for lignin while ensuring a homogeneous carbon distribution on the silicon surface as shown through SEM (Figure 13b). The synthesized silicon/carbon composite (containing 50% lignin) manifested superior electrochemical properties as an anode material for LIBs. It exhibited an outstanding reversible capacity of 1147 mAh g^{-1} after 100 cycles at 0.2 A g^{-1} , alongside an extraordinary rate performance (Figure 13c,d). The use of recycled silicon and renewable lignin offers a cost-effective and scalable approach for producing high-capacity and long cycle life anodes. This study demonstrates the synthesis of a silicon/carbon composite via spray drying, where silicon facilitates uniform carbon coating and self-assembly. The composite shows excellent electrochemical performance while highlighting its potential as a promising anode material for LIBs. The comprehensive utilization of Si-rich biomass is restrained by macromolecular lignin and a large amount of ash. Li et al. treated rice husks (RHs) by alkali extraction and acid precipitation, and then obtained lignin- SiO_2 composite which is modified by carbonization, ball milling, magnesiothermic reduction, and additives in order to form Si/C composite (Figure 14a).^[169] The highly porous morphology was seen through SEM, which helped in attaining a significant electrochemical performance (Figure 14b). Through these processes,

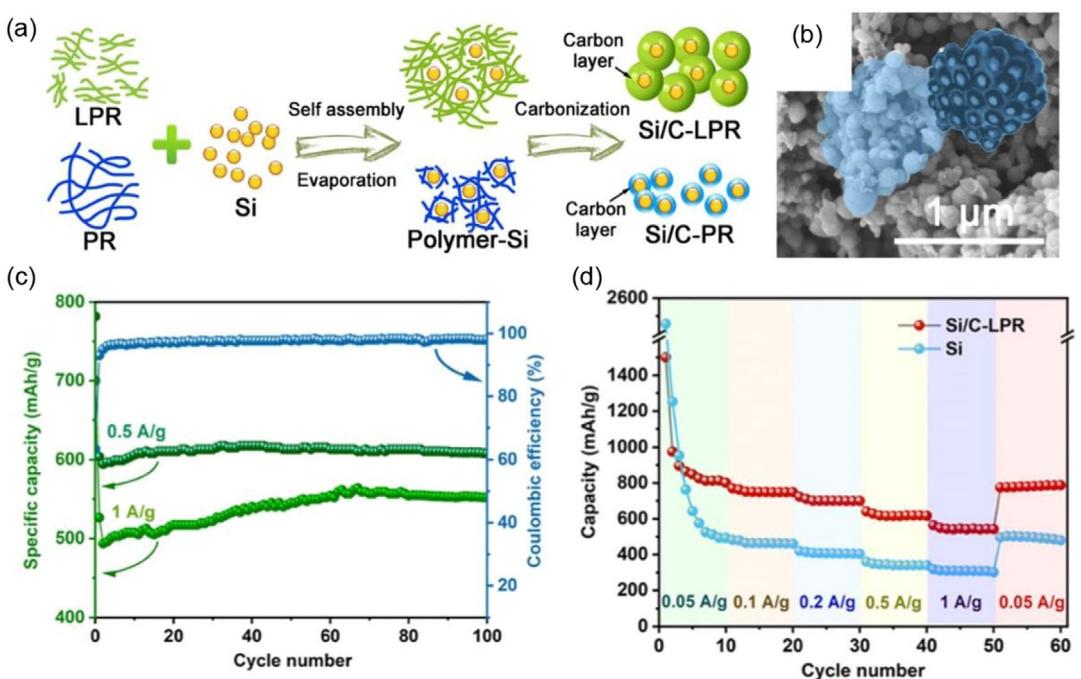


Figure 12. a) Schematic diagram depicting the stepwise preparation process of Si/C-LPR. b) SEM image illustrating the detailed surface morphology of Si/C-LPR electrode. c) Electrochemical cycling stability of the Si/C-LPR and Si electrodes. d) Comparative rate capability of Si/C-LPR and Si electrodes under various current densities. Adapted with permission.^[167] Copyright 2023, Royal Society of Chemistry.

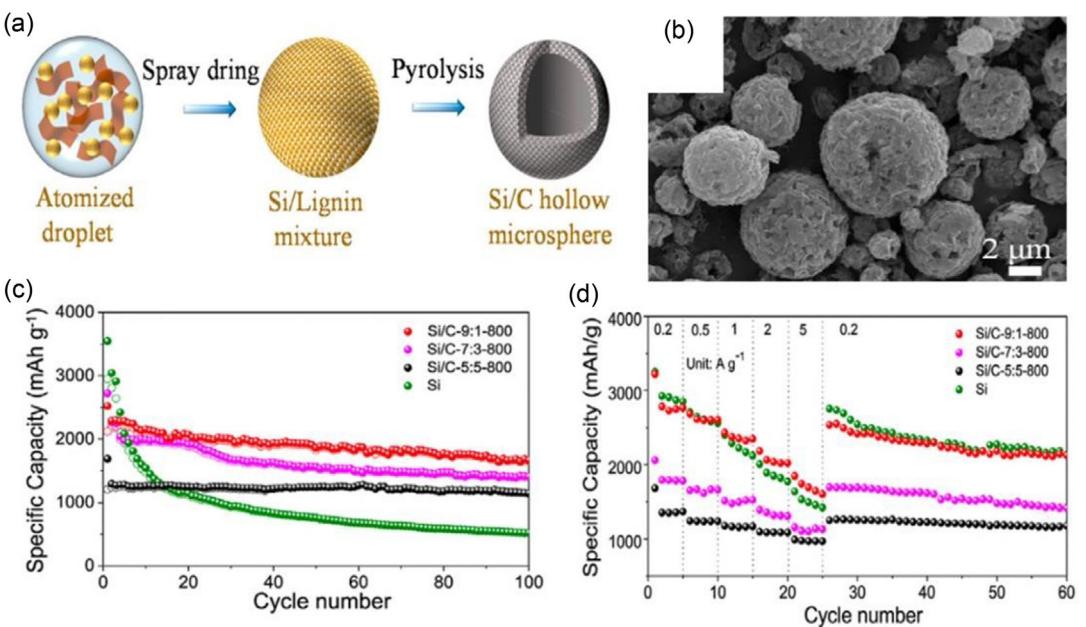


Figure 13. a) Diagram of the Si/C preparation process. b) SEM image of the Si/C-9:1-800 electrode. c) Cycling performance of the synthesized electrodes. d) Rate performance of the synthesized electrodes at different current densities. Adapted with permission.^[168] Copyright 2021, Elsevier.

an Si/C composite with excellent electrochemical properties is obtained and performs stable cycling performance with high specific capacity retention of 572 mAh g⁻¹ at 1 A g⁻¹ after 1000 cycles along with excellent rates (Figure 14c,d). This work demonstrates a method for the comprehensive utilization of Si-rich biomass by addressing the challenges posed by macromolecular lignin and ash content. The highly porous morphology

contributes to improved electrochemical performance, particularly in the form of enhanced cycling stability and capacity retention. The study highlights the potential of using Si-rich biomass as a sustainable source for high-performance silicon-based anode materials in LIBs. The burgeoning silicon photovoltaics industry produces an immense quantity of silicon waste each year, while lignin not only results in considerable resource depletion but also

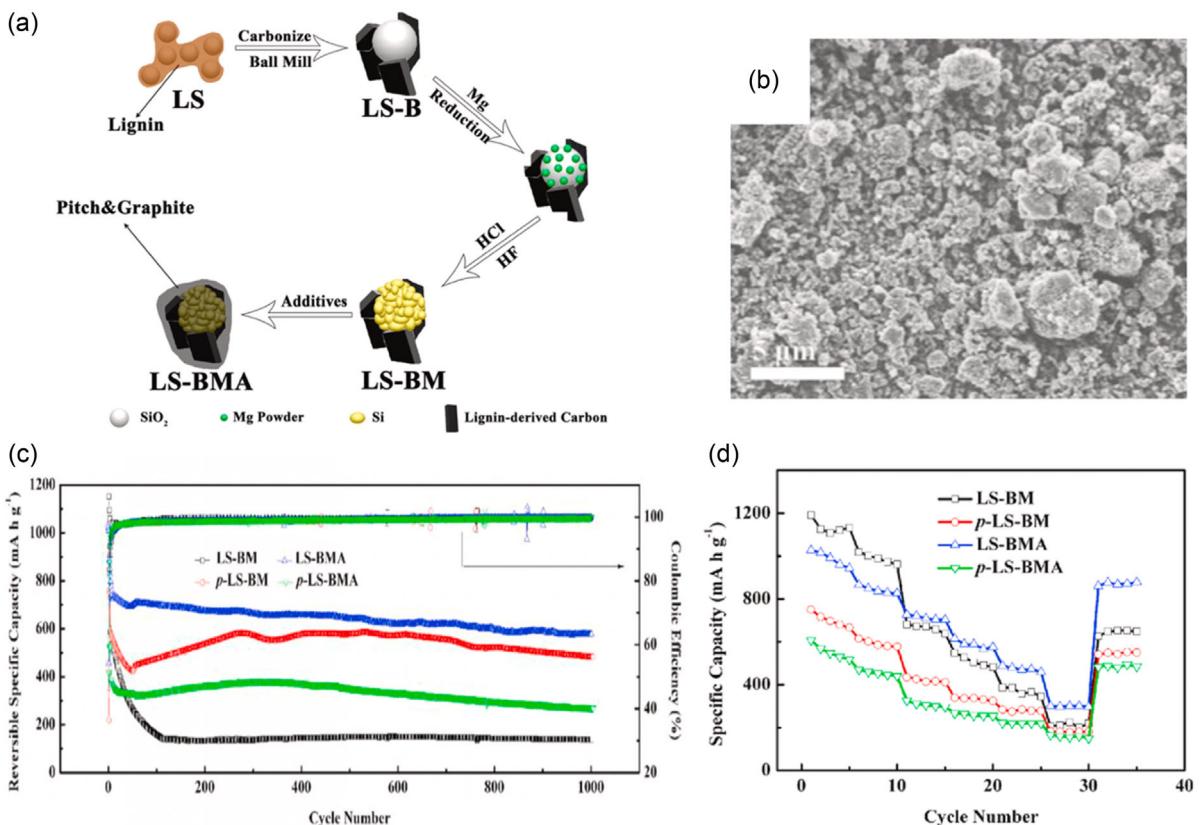


Figure 14. a) Schematic diagram illustrating the detailed fabrication process of the LS-BMA. b) SEM image providing insight into the surface morphology and characteristics of LS-BMA electrode. c) Electrochemical cycling stability and performance of the synthesized electrodes. d) Comparative analysis of the rate performances of the synthesized electrodes at varying current densities. Adapted with permission.^[169] Copyright 2021, Elsevier.

exacerbates environmental pollution on a large scale. LIBs are in search of materials that offer superior performance. In this regard, silicon Si-based anodes emerge as the most promising candidates for next-generation LIBs owing to their vastly higher capacity relative to conventional graphite anodes.

Liu et al. put forth an innovative strategy wherein the silicon and lignin waste materials are repurposed to synthesize silicon/carbon (Si/C) anode composites for LIBs. This approach employed a sophisticated coprecipitation technique, driven by electrostatic interactions, followed by a meticulously controlled thermal annealing process to optimize the material's structural integrity and electrochemical performance (Figure 15a).^[170] The resulting Si/C composite manifested an intricately advanced material architecture within a resilient carbon matrix. This sophisticated configuration adeptly mitigates the fundamental limitations of silicon materials while conferring an enhanced electrochemical stability and performance (Figure 15b). The resultant Si/C composite exhibited a remarkable initial charge capacity of 1016 mAh g⁻¹ in LIBs. Additionally, it demonstrated an impressive capacity retention of 74.5% at a current density of 0.2 A g⁻¹ following 100 charge-discharge cycles while underscoring its superior electrochemical stability (Figure 15c). Furthermore, the Si/C composite demonstrated an exceptional rate capability while achieving a remarkable charge capacity of 575 mAh g⁻¹ at a relatively high current density of 2 A g⁻¹, which represents 63.4% of the capacity maintained at 0.2 A g⁻¹ which emphasizes its formidable

electrochemical prowess and resilience under high-rate cycling conditions. (Figure 15d). The use of industrial silicon and lignin waste offers a sustainable pathway for developing high-capacity anode materials for LIBs with significant economic and environmental benefits. This study demonstrates how silicon and lignin by-products can be utilized to create silicon/carbon composites while featuring micron-sized particles with silicon nanoparticles embedded in a carbon matrix. The Si/C composites show great potential for high capacity, stability, and sustainability in LIBs while presenting a viable alternative to conventional anode materials.

As material processing and design techniques improve, lignin-Si composites hold the potential to play a crucial role in advancing the next generation of LIBs while offering a more sustainable and high-capacity energy storage solution for a wide range of applications. These materials not only improve the charge-discharge efficiency and cycle stability of LIBs but also offer a more environmentally friendly and cost-effective solution by utilizing renewable biomass resources. While challenges such as scalability and consistency remain, ongoing research into the energy storage applications of lignin-based carbon materials holds great promise for advancing next-generation LIBs.

Lignin-derived electrodes have gained considerable attention as a sustainable approach to improve the performance of LIBs, particularly when combined with silicon to address challenges such as silicon's volumetric expansion, poor cycling stability, and low initial

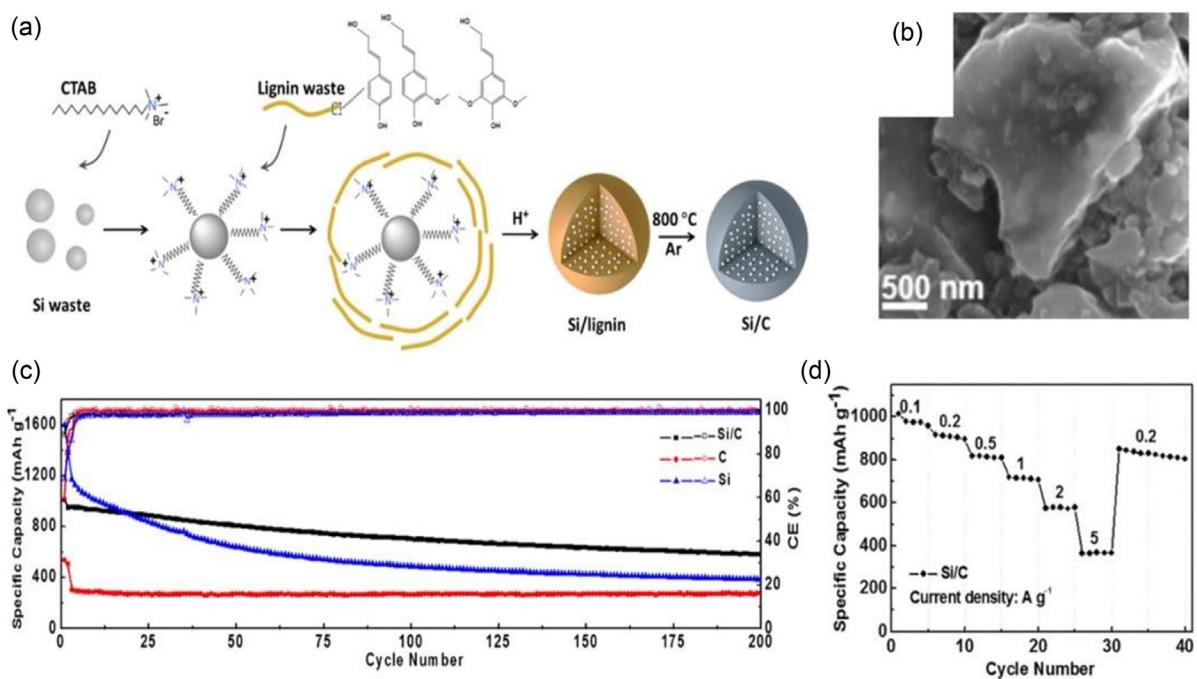


Figure 15. a) Schematic representation of the multistep synthesis process for Si/C. b) SEM micrograph capturing the intricate surface morphology and structural attributes of Si/C electrode. c) Long-term cycling stability and electrochemical performance of the synthesized electrodes. d) Rate capability of Si/C across a range of current densities. Adapted with permission.^[170] Copyright 2020, American Chemical Society.

Coulombic efficiency. Lignin serves as a precursor for creating conductive and stable composites that enhance the electrochemical performance of silicon anodes. Li et al. demonstrated the use of a lignin-derived resin to form a carbon layer around silicon nanoparticles while significantly improving the cycling performance and rate capability of the resulting Si/C-LPR composite, with a notable initial capacity of 782 mAh g⁻¹ and a stable 605 mAh g⁻¹ after 100 cycles. In a similar vein, Yang et al. engineered silicon/carbon hollow microspheres using recycled silicon and lignin while achieving a reversible capacity of 1147 mAh g⁻¹ after 100 cycles and exceptional rate performance. Furthermore, Li et al. also explored rice husk-derived lignin to form a SiO₂ composite that achieved a high specific capacity retention of 572 mAh g⁻¹ after 1000 cycles after carbonization and ball-milling. Additionally, Liu et al. used a coprecipitation technique to synthesize Si/C composites from silicon and lignin waste while resulting in an initial charge capacity of 1016 mAh g⁻¹ with 74.5% retention after 100 cycles. These advancements underscore how lignin-based composites can enhance the cycling stability, rate performance, and sustainability of silicon anodes while offering a promising path toward high-performance and environmentally friendly LIBs by utilizing renewable and waste materials.

Considerable efforts in the research of lignin-derived electrodes for LIBs need to be directed toward increasing the economic viability and scalability of lignin-based composite materials manufacture. One of such promising avenues is developing more efficient lignin extraction and processing methods directed toward improving the uniformity and consistency of the carbon structures with better consolidation to silicon and superior electrochemical characteristics. Further studies could examine hybrid composites with other bio-based or nanomaterial additives, such

as graphene or conductive polymers, which aim at increasing the electrical conductivity and the mechanical strength of the composites. In addition, more advanced surface modification methods for lignin-derived carbon may help in enhancing the long-term cycling stability of silicon's volumetric expansion reduction. The use of lignin from different sources of biomass could also be tested to investigate how using different feedstocks would modify the sustainability and environmental benefits in large-scale implementations. Moreover, new methods of synthesis could manipulate the battery morphology/porosity and electrode performance in an unprecedented way while resulting in excellent batteries. Hence, a deeper understanding of the fundamental mechanisms governing the interaction between lignin-derived materials and silicon is crucial to further improve the initial coulombic efficiency (ICE) and overall lifespan. Such advancements may lead to the development of renewable LIBs that are more efficient, environmentally sustainable and commercially feasible.

5. Lignin-Derived Electrode Materials for Sodium-Ion Batteries (SIBs)

Lignin-derived electrode materials for SIBs epitomize a transformative confluence of nature's ingenuity and cutting-edge electrochemical engineering while heralding a new epoch in the quest for sustainable and high-performance energy storage solutions. Lignin is an intricate polyphenolic biopolymer, which is ubiquitously embedded in lignocellulosic biomass. It has been emerged as an unparalleled contender for the next generation

of electrode materials due to its exceptional structural diversity, renewability, and the inherent electrochemical properties.^[171] This exceptional biopolymer harbors an immense and underexplored potential when subjected to advanced depolymerization and activation techniques while yielding carbonaceous matrices that demonstrate unparalleled capacity, superior cycling performance, and remarkable electrochemical resilience. In the realm of SIBs, lignin's exploitation transcends mere academic curiosity while representing a crucial advancement in energy storage solutions. Its valorization not only disrupts the conventional paradigm of traditional electrode materials but also substantially diminishes the environmental impact of battery production and disposal. As lignin-derived electrode research advances, it is primed to instigate a transformative shift in energy storage while harmonizing the complexities of biological polymers with the exigencies of contemporary technological imperatives.

5.1. Pristine–Lignin Electrodes

The burgeoning demand for sustainable and high-performance energy storage systems has catalyzed an unprecedented interest in alternative battery technologies. Lignin has garnered significant attention for its exceptional potential in the development of SIBs. As a carbon-rich macromolecule derived from plant biomass, pristine lignin offers a unique advantage in the synthesis of novel electrode materials while exhibiting remarkable electrochemical properties, structural integrity, and environmental compatibility. This discourse delves into the transformative role of pristine lignin-derived electrodes which not only address the pressing demands for efficiency and sustainability in sodium-ion

technology but also herald a new paradigm in the integration of bio-derived materials into next-generation energy storage devices. HC has surfaced as an exceptionally promising anode material for sodium-ion batteries owing to its affordability, relatively low operational voltage, and notable specific capacity. However, a significant challenge remains in the synthesis of exceptional HC anodes from cost-efficient carbon precursors. Moreover, the continual fracture of the SEI during repeated cycling accelerates capacity degradation while complicating the long-term stability of battery performance. To mitigate these persisting issues, Zheng et al. have advanced a lignin-derived carbon anode that not only inherently cultivates a resilient and stable SEI but also concurrently addresses the challenges of material synthesis and performance deterioration. (Figure 16a).^[172] The SEM and TEM micrographs elucidated the intricate, compact, and porous morphology embedded within the HC matrix as vividly illustrated in (Figure 16b,c). This study introduces a HC anode (H-1400BW), which exhibited a robust and organic-rich SEI. This SEI enhances the Na^+ transfer, improves the structural stability, and prevents rupture. The anode demonstrated an outstanding performance while achieving 289 mAh g^{-1} after 400 cycles with excellent cycling stability and capacity retention (Figure 16d,e). This superior SEI performance culminates in an overall enhancement of the sodium storage capabilities while providing a significant advancement in the development of high-performance and sustainable sodium-ion battery (SIB) anodes. This study exemplifies a lignin-derived HC anode for sodium-ion batteries while focusing on optimizing the SEI to prevent capacity loss from SEI rupture and reformation. The anode's porous structure promotes a stable and organic-rich SEI while enhancing Na^+ ion

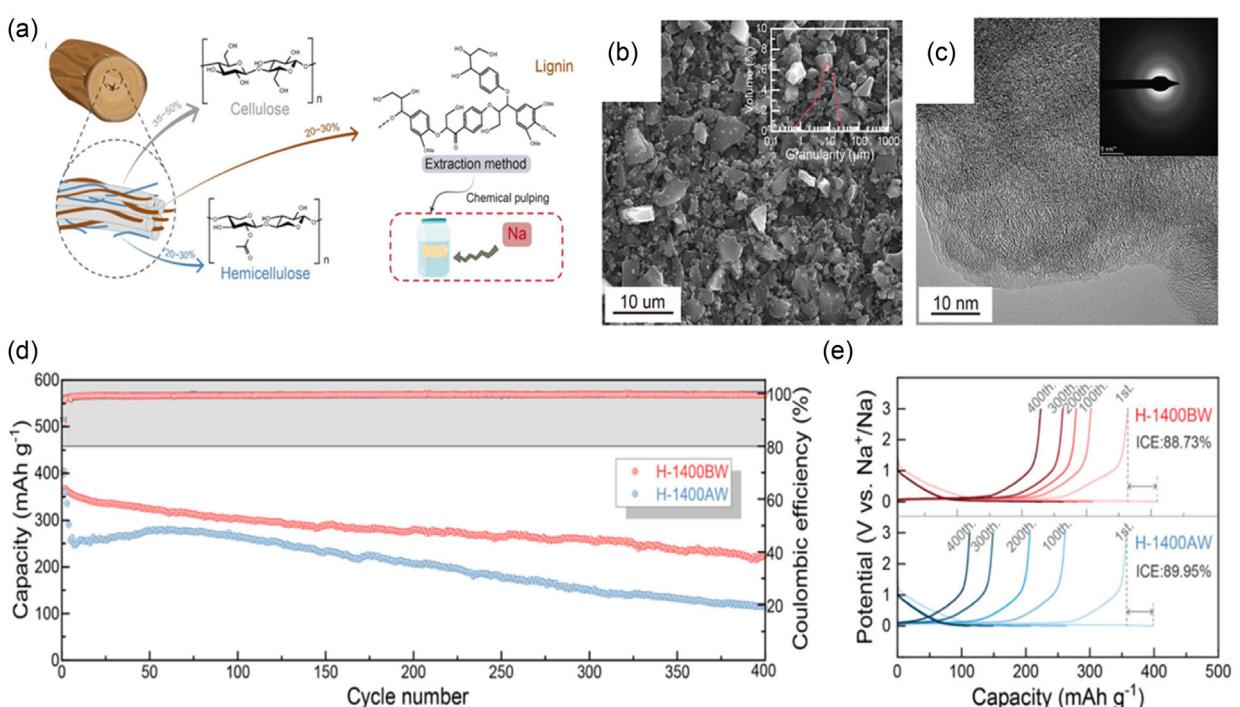


Figure 16. a) Schematic representation of the multistep synthesis process for H-1400BW. b,c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of H-1400BW electrode. d) Long-term cycling stability and electrochemical performance of the synthesized electrodes. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[172] Copyright 2023, Wiley.

transfer and cycle stability. Additionally, a novel gas-phase-assisted washing technique is used to selectively remove excess sodium while enabling the upcycling of industrial lignin into high-value HC. This work also demonstrates an effective strategy for improving the performance and stability of SIB anodes by enhancing the SEI and utilizing industrially available lignin as a precursor material. This advancement could lead to the development of high-performance, cost-effective, and sustainable anode materials for SIBs.

Rechargeable sodium-ion batteries often experience accelerated degradation of electrodes under elevated temperatures, coupled with the prohibitively high costs associated with the synthesis of electrode materials. Consequently, there is a pressing need to investigate novel organic electrodes that offer both cost-effectiveness and remarkable adaptability to volumetric fluctuations. She et al. explored the pristine lignin as a potential anode material for sodium-ion batteries, with its sodium storage performance assessed at ambient temperature and 60 °C (Figure 17a).^[51] The macromolecular structure of lignin is depicted through SEM (Figure 17b). The lignin-based anodes demonstrated exceptional capacities at various current densities and voltages along with the exceptional performances of 250, 199 mAh g⁻¹ at 2,001,000 mA g⁻¹ alternatively (Figure 17c–e). The 60 °C electrode also exhibited exceptional results at various current densities (Figure 17f). This can be attributed to the generation of abundant reactive functional groups, particularly carbonyl (C=O) moieties while resulting from the homogeneous cleavage of the bonds present within the lignin macromolecular structure. This research not only underscores the viability of lignin as a high-performance anode material but also paves the way for

the exploration of a broader range of natural organic materials for energy storage applications. This study could stimulate the development of environmentally conscious, economical, and durable electrode materials from biomass while leveraging renewable organic resources to advance more efficient and broadly accessible energy storage technologies.

HC is universally acknowledged as the most commercially promising anode material for sodium-ion batteries. Additionally, lignin emerges as a highly attractive precursor for the production of low-cost HCs. Yang et al. exquisitely modulated the HCs by variations in carbonization temperature to ensure the unique structural architecture and electrochemical characteristics of lignin (Figure 18a).^[173] The microstructure of lignin-derived HC (LDHC-1200) anode exhibited a compact morphology as shown through SEM (Figure 18b). All lignin-derived HCs at various temperatures showed crystallinity as depicted through XRD characterization (Figure 18c).

An augmentation in the carbonization temperature induces a diminution in the specific surface area while enhancing the initial Coulombic efficiency. Additionally, the slope and plateau capacities of the LDHC anode are subjected to precise modulation through the regulation of the carbonization temperature. The LDHC (subjected to carbonization at 1200 °C) demonstrated unparalleled sodium-ion storage performance while manifesting significant capacity and an exceptional rate performance along with high reversible sodium-ion capacity of 284.7 mAh g⁻¹ (Figure 18d–f). This approach signifies the development and optimization of lignin-derived HCs (LDHCS) as an anode material for sodium-ion batteries. The study highlights the significant influence of carbonization temperature on the microstructure,

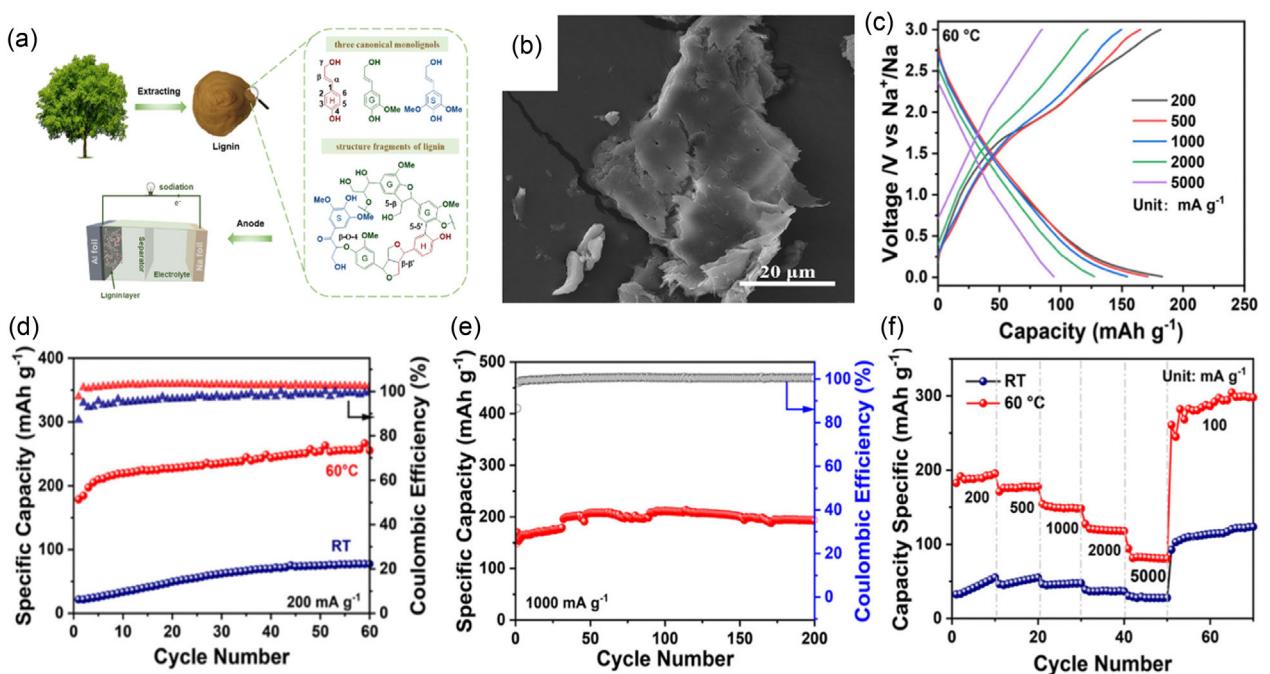


Figure 17. a) Schematic representation of the multistep synthesis process for 60 °C electrode. b) SEM micrograph capturing the intricate surface morphology and structural attributes of 60 °C electrode. c) Capacity vs voltage profiles of 60 °C electrode. d,e) Long-term cycling stability and electrochemical performance of the 60 °C electrode at 200 and 1000 mA g⁻¹. f) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[51] Copyright 2024, Wiley.

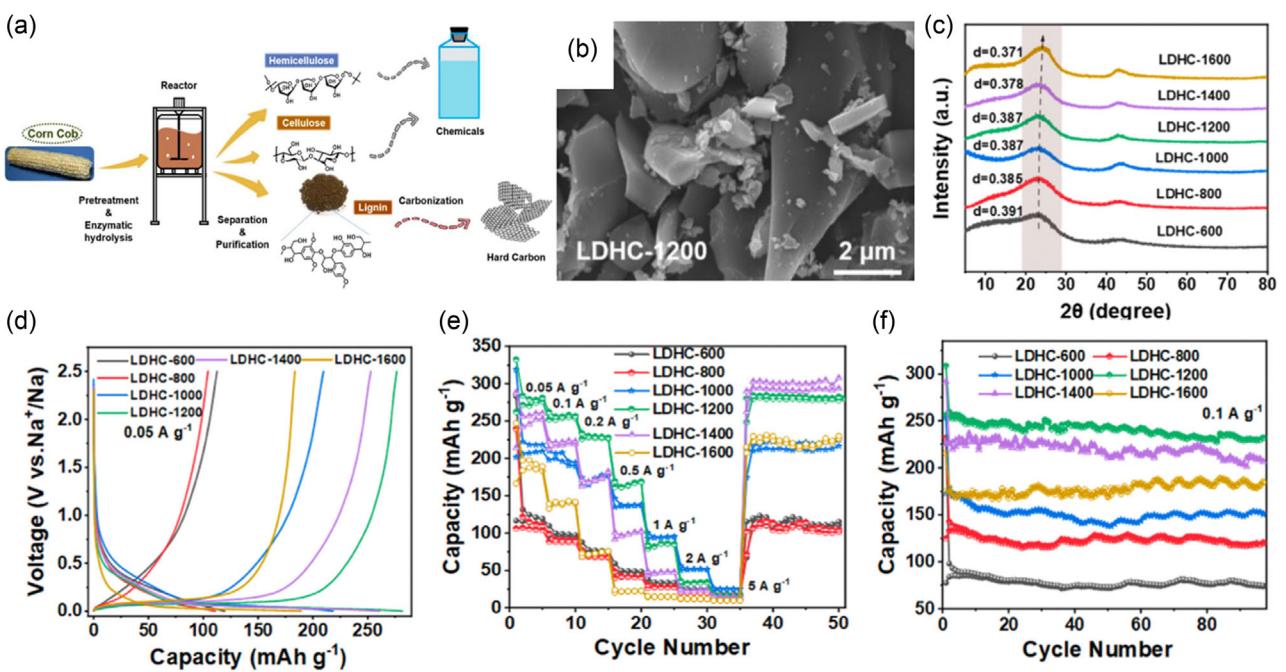


Figure 18. a) Schematic representation of the multistep synthesis process for LDHC-1200. b) SEM micrograph capturing the intricate surface morphology and structural attributes of LDHC-1200 electrode. c) XRD spectra of the synthesized electrodes. d) Capacity vs voltage profiles of the synthesized electrodes. e) Long-term cycling stability and electrochemical performance of the synthesized electrodes. f) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[173] Copyright 2023, Elsevier.

crystallinity, and electrochemical performance of these HCs. Specifically, by varying the carbonization temperature, the lignin-derived HCs exhibited distinct structural and electrochemical characteristics while leading to improved sodium-ion storage capacity, Coulombic efficiency, and rate performance. HC plays a pivotal role in the advancement of SIBs. Nevertheless, challenges such as suboptimal ICE and limited specific capacity continue to impede the progress of HC-based materials. Chen et al. synthesized HC materials (LDHC) from lignin through precise control of carbonization temperature (Figure 19a).^[174] The electrochemical storage mechanisms within SIBs were explored in depth, and the influence of carbonization temperature on the structural and chemical properties of the HC was thoroughly investigated through SEM and TEM analyses (Figure 19b,c). Owing to its optimized microstructure, the lignin-derived HC carbonized at 1600 °C (LDHC-1600) exhibited an impressive ICE of 81.2%, along with a reversible discharge capacity of 303 mAh g⁻¹ (Figure 19d). Furthermore, the anode material contributed a low-potential plateau (74.75%) of the total capacity while equating to 226.5 mAh g⁻¹ along with excellent rates (Figure 19e,f).

The mechanistic insights revealed that sodium ions are predominantly adsorbed within the sloped region, while intercalation and filling processes dominate within the plateau region. This work demonstrates the potential of lignin-derived HC as a promising anode material for SIBs. By carefully controlling the carbonization temperature, this work explores how the structural and chemical properties of HC can be optimized for better electrochemical performance. The study reveals complex sodium-ion storage behaviors while highlighting lignin-derived electrodes as a promising alternative for SIBs. These electrodes offer

advantages like low cost, sustainability, and scalability. However, challenges such as low initial efficiency, capacity, and cycle stability persist due to lignin's structural changes during carbonization. Despite these hurdles, ongoing research suggests strong potential for the future development of lignin-based electrodes in energy storage.

The growing demand for sustainable and high-performance energy storage systems has driven the exploration of lignin as a promising precursor for SIB anodes. Lignin offers exceptional electrochemical properties, structural integrity and environmental compatibility while making it ideal for novel electrode materials. Transforming lignin into HC has been extensively studied to address challenges such as material synthesis, stability and performance degradation in SIBs. Lignin's high-carbon content and renewable nature make it an attractive and low-cost option for high-capacity anodes, which are crucial for SIBs due to their low operational voltage and notable specific capacity. However, a key challenge lies in synthesizing high-performance HC anodes while ensuring long-term stability, especially with the breakdown of the solid electrolyte interphase (SEI) during cycling. Zheng et al. addressed this by developing a lignin-derived carbon anode with a resilient and stable SEI while promoting Na⁺ transfer and enhancing structural stability in order to achieve 289 mAh g⁻¹ after 400 cycles.

The unique porous structure of the anode (H-1400BW) enhanced the formation of an organic-rich SEI while contributing to long-term cycling stability. Additionally, Zheng's gas-phase-assisted washing technique facilitated the upcycling of industrial lignin into high-value HC while improving anode performance and mitigating SEI rupture. Further studies by She et al.

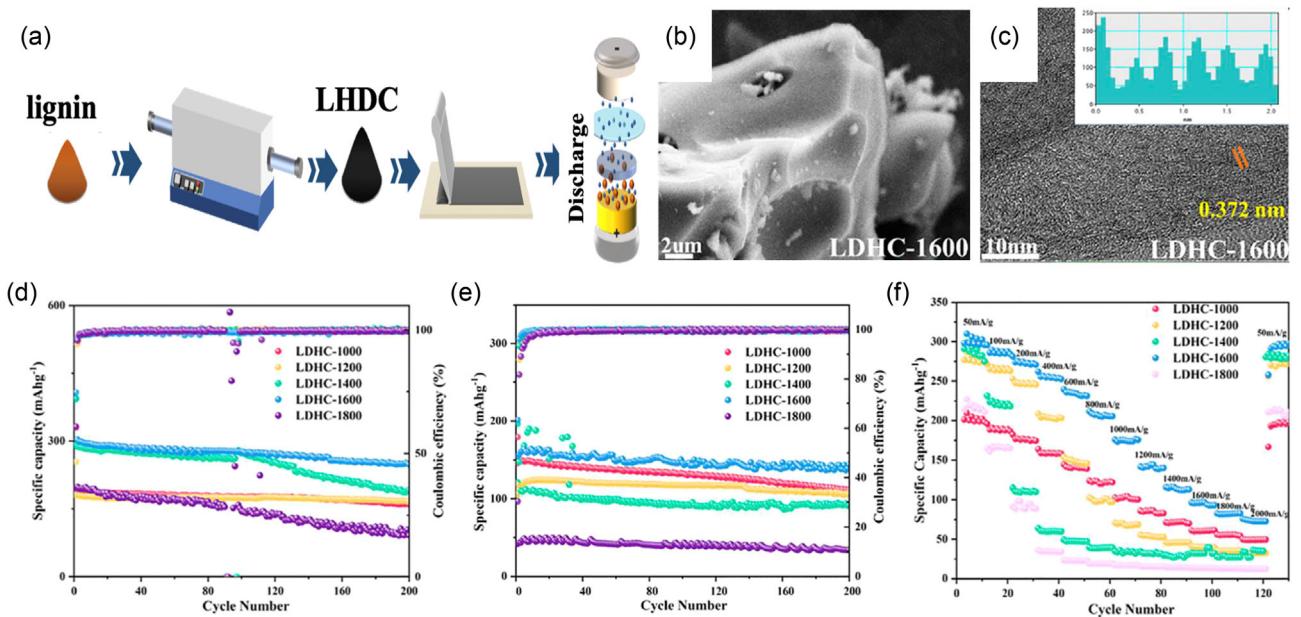


Figure 19. a) Schematic representation of the multistep synthesis process for LDHC-1600. b,c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of LDHC-1600 electrode. d,e) Long-term cycling stability and electrochemical performance of the synthesized electrodes. f) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[174] Copyright 2022, Elsevier.

demonstrated that pristine lignin, with its ability to generate reactive carbonyl groups, showed excellent sodium storage performance while achieving 250 mAh g^{-1} at 200 mA g^{-1} and 199 mAh g^{-1} at 1000 mA g^{-1} . Yang et al. and Chen et al. explored the impact of carbonization temperature on the electrochemical properties of lignin-derived HCs while revealing that increasing the temperature optimizes the Coulombic efficiency and electrochemical performance. Lignin-derived HCs carbonized at 1200°C and 1600°C achieved impressive reversible capacities (284.7 and 303 mAh g^{-1} , respectively), with excellent cycling stability and rate capability. These findings underscore the significance of precise carbonization conditions for improving the performance and stability of lignin-based anodes. These studies highlight the potential of lignin as a sustainable and high-performance material for next-generation sodium-ion batteries while offering a pathway for cost-effective, environmentally friendly, and high-capacity electrode materials.

Future studies regarding the application of lignin on SIB anodes should aim at improving the synthesis procedure to optimize their stability and electrochemical performance. One such approach is the combination of lignin with other bio-derived compounds like cellulose or hemicellulose to enhance the structural and cycling stability of the anodes. Moreover, there is a need to further develop gas-phase-assisted washing for better impurity removal and for better homogeneity of the final HC structure. The effect of different carbonization parameters (pressure, atmosphere, and heating rate) should also be comprehensively examined to improve the production of high-performance HC with adequate sodium storage capability. Researchers could look into new materials or methods that help improve the toughness of the organic-rich SEI so that better cyclic durability is achieved after multiple charge and discharge processes. Moreover, the use of lignin anodes should be competitively analyzed at scale

to determine its feasibility while keeping in mind that lignin is relatively cheap and sustainable for the industry. Hence, analyzing the environmental effects and life cycle assessment of lignin-based anodes relative to conventional materials will be essential to confirm their prospective effectiveness as a sustainable substitute in energy storage technologies.

5.2. Nitrogen-Doped Lignin Electrodes

The surging demand for sustainable and efficient energy storage solutions has driven the investigation of alternative battery technologies that address the shortcomings of conventional lithium-ion systems. Among the emergent alternatives, SIBs have attracted considerable attention due to the ubiquity and cost-effectiveness of sodium. However, significant hurdles persist, particularly in the development of high-performance electrodes capable of efficiently intercalating and de-intercalating sodium ions. In this regard, lignin has surfaced as a promising electrode material owing to its advantageous structural framework, substantial carbon content, and inherent sustainability. Recent advancements indicate that nitrogen doping (N-doping) can enhance the electrochemical characteristics of lignin while augmenting its conductivity, mechanical stability, and reactivity. Nitrogen-doped lignin electrodes have demonstrated remarkable performance while exhibiting high capacity, rapid charge-discharge rates, and superb cycling stability within SIBs. This section examines the rationale behind employing nitrogen-doped lignin as an innovative electrode material for SIBs while outlining its advantages, challenges, and its prospective role in revolutionizing future energy storage technologies. Chen et al. synthesized nitrogen-doped porous ultrathin layered carbon (LC) through the pyrolytic treatment of alkaline lignin in the presence of melamine

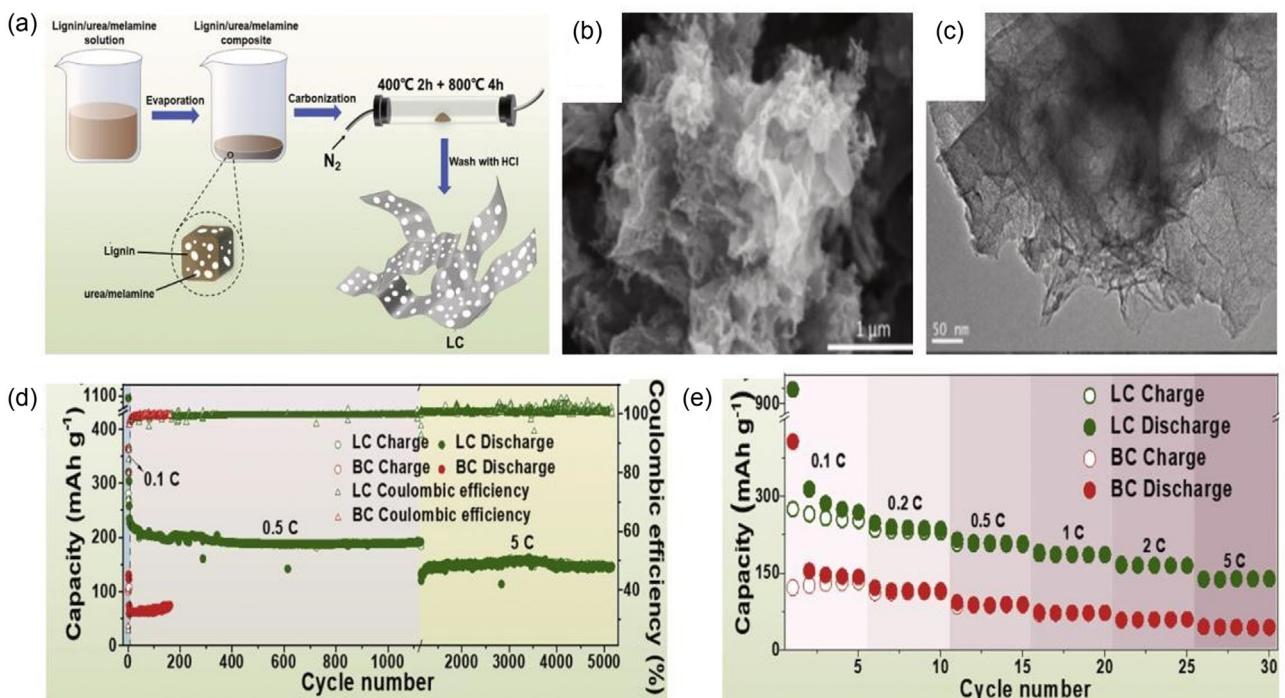


Figure 20. a) Schematic representation of the multistep synthesis process for LC b,c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of LC electrode. d) Long-term cycling stability and electrochemical performance of the synthesized electrodes. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[175] Copyright 2020, Elsevier.

and urea (Figure 20a).^[175] The resulting LC displayed an ultrathin graphitic carbon architecture, which is marked by a vast surface area and considerable pore volume as revealed through SEM and TEM (Figure 20b,c). The incorporation of nitrogen further augments the material's capacity while enhancing its electrochemical reactivity. The contributions from capacitive and diffusive mechanisms underscore the improved sodium ion storage kinetics while resulting in extraordinary rate capabilities. LC maintained a remarkable cycling durability with negligible capacity degradation after 5000 cycles at a high current rate of 5C along with extraordinary rate capabilities (Figure 20d,e). This work demonstrates the successful synthesis of nitrogen-doped porous ultrathin LC from alkaline lignin through pyrolytic treatment in the presence of melamine and urea while offering a novel approach for SIB anodes. The study highlights the unique structural features of the resulting LC which are crucial for enhancing electrochemical performance. The incorporation of nitrogen into the material improves its electrochemical reactivity and sodium ion storage kinetics while contributing to both capacitive and diffusive charge storage mechanisms. Additionally, the work emphasizes the material's excellent cycling stability while showcasing the potential of low-cost and sustainable feedstocks for next-generation energy storage technologies.

Fan et al. meticulously synthesized nitrogen-doped lignin-based carbon microspheres via a hydrothermal process while employing 3-aminophenol as the nitrogen dopant.^[176] An exhaustive investigation of the structural evolution, coupled with an in-depth analysis of its profound impact on electrochemical performance, is presented. The resulting nitrogen-doped carbon microspheres (NLC) exhibited a highly refined, spherical

morphology, enriched with a plethora of active sites, an extraordinarily ultra-microporous architecture (<0.7 nm) and a significantly enlarged interlayer spacing (Figure 21a–c). These structural intricacies are intricately linked to the remarkable electrochemical properties demonstrated by the material. The incorporation of defect-induced heterogeneity and the abundance of ultra-microporosity served to significantly augment the sloping capacity, while the pronounced interlayer spacing helped to enhance the plateau capacity (Figure 21d). The microspheres exhibited a rapid sodium ion adsorption and intercalation kinetics while delivering exceptional electrochemical performance. They achieved a capacity retention of 90% after 300 cycles at 0.4C while maintaining a stable rate capability across a range of current densities (Figure 21e,f).

This work demonstrates the successful synthesis of nitrogen-doped lignin-based carbon microspheres (NLC) through a hydrothermal process while using 3-aminophenol as the nitrogen dopant. The study provides a thorough analysis of the material's structural evolution while emphasizing its spherical morphology, ultra-microporous architecture and enlarged interlayer spacing. These structural features are shown to contribute to enhanced electrochemical performance by increasing the number of active sites and improving sodium ion adsorption and intercalation kinetics. The incorporation of defects and heterogeneity helps in enhancing material's charge storage capacities. Additionally, the work highlights the promising electrochemical properties of NLC while suggesting its potential as a high-performance anode material for sodium-ion batteries. Zhang et al. introduced a highly refined emulsion-solvent evaporation strategy that effortlessly converts lignin into nitrogen co-doped HC microspheres

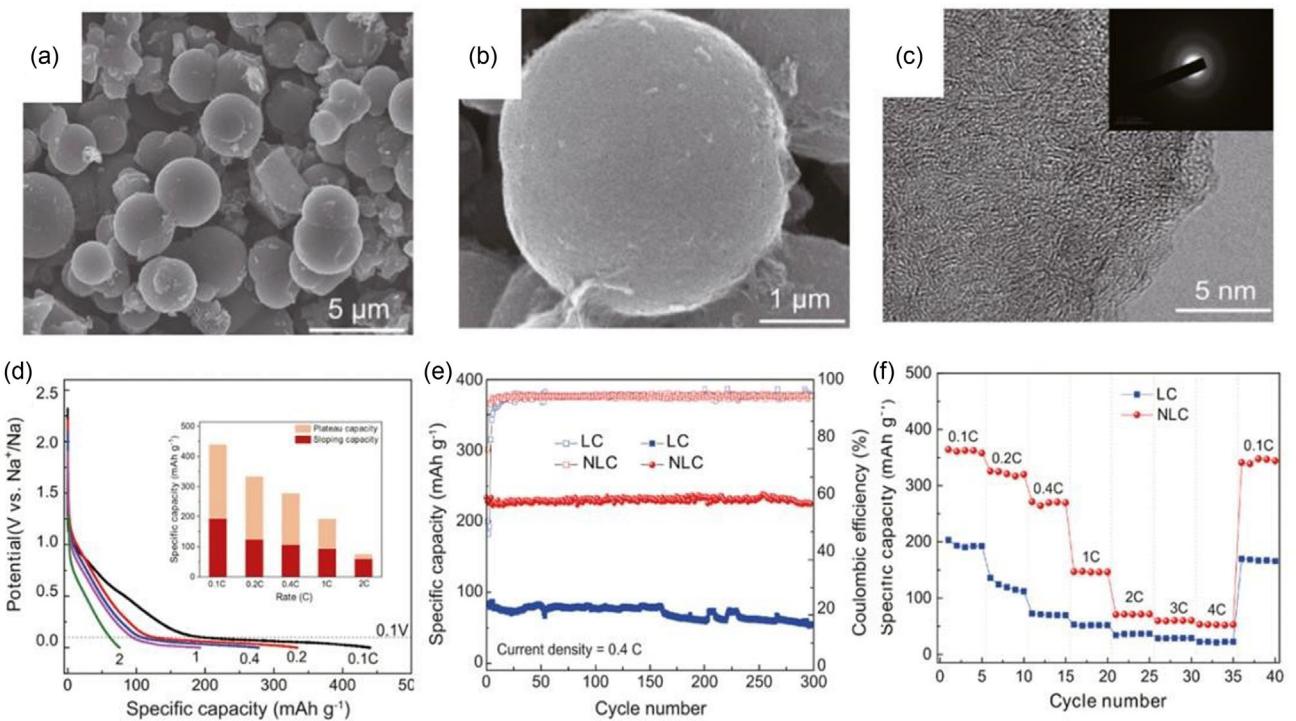


Figure 21. a–c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of NLC electrode. d) The specific capacity vs potential graph. e) Long-term cycling stability and electrochemical performance of the synthesized electrodes. f) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[176] Copyright 2020, Elsevier.

(NPL-1300) (Figure 22a).^[177] Through an exhaustive suite of physicochemical characterizations, it is established that the electrode material possesses an intricately engineered microstructure with porous spheres (Figure 22b,c). Electrochemical assessments in SIBs unambiguously reveal that these lignin-derived carbon microspheres operate in full alignment with the universally accepted adsorption-intercalation mechanism for Na⁺ storage. These microspheres deliver strikingly high and exceptionally well-balanced sodium storage performance while showcasing a substantial reversible capacity. Additionally, the anodes demonstrated an exceptionally high initial Coulombic efficiency, unrivaled rate capability at various current densities and extraordinary cycling stability while maintaining the capacity of 248 mAh g⁻¹ with a remarkable retention of 92.2% after 200 cycles at 0.1 A g⁻¹ (Figure 22d–f).

This work not only heralds a transformative and low-cost methodology for the valorization of lignin but also significantly advances the development of eco-friendly energy storage systems. This work demonstrates the development of a novel, low-cost approach for converting lignin into nitrogen co-doped HC microspheres (NPL-1300) while using an emulsion-solvent evaporation strategy. The study showcases the intricate microstructure of the resulting material containing porous spheres that are well-suited for sodium ion storage. Through comprehensive physicochemical characterizations, the material is found to operate according to the widely accepted adsorption-intercalation mechanism for Na⁺ storage. The work highlights the significant potential of this lignin-derived material for energy storage applications while emphasizing its remarkable electrochemical

performance. Furthermore, this approach advances the sustainable and eco-friendly utilization of lignin while offering a promising route for advanced SIBs.

Nitrogen-doped lignin electrodes represent a promising advancement in the field of SIBs while offering a sustainable and cost-effective alternative to traditional anode materials. The incorporation of nitrogen not only enhances the conductivity and electrochemical stability of the lignin-derived carbon structure but also significantly improves its sodium-ion storage capacity, rate performance and cycling stability. The unique porous and ultrathin morphology of the nitrogen-doped lignin electrodes provides abundant active sites for sodium storage while facilitating rapid ion diffusion and superior charge/discharge kinetics. Moreover, the exceptional cycling performance underscores the potential of these materials for long-term energy storage applications. The exceptional structural versatility, sustainability and inherent electrochemical properties of lignin endow these materials with the potential to rival or even surpass traditional battery electrodes in performance, efficiency and environmental compatibility. As a renewable resource of unparalleled abundance, lignin offers an exquisite avenue for addressing the pressing challenges of energy storage sustainability while mitigating the deleterious impacts of conventional materials and fostering the development of more eco-conscious and cost-effective solutions. The ongoing refinement of lignin-based electrode architectures underscore their pivotal role in the evolution of next-generation SIBs while unlocking new realms of performance and application. Hence, the strategic valorization of lignin in energy storage systems affirms lignin's place as a cornerstone

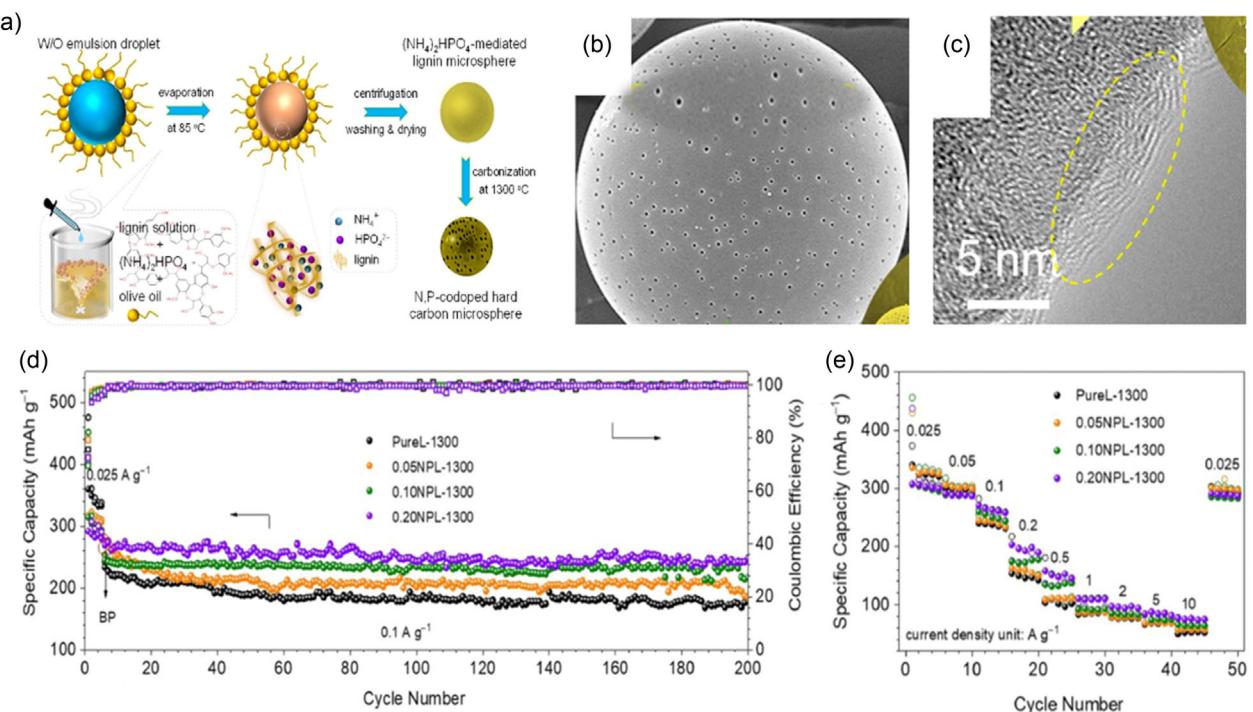


Figure 22. a) Schematic representation of the multistep synthesis process for 0.20NPL-1300. b,c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of 0.20NPL-1300 electrode. d) Long-term cycling stability and electrochemical performance of the synthesized electrodes. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[177] Copyright 2021, Wiley-VCH GmbH.

material in the pursuit of a more energy-secure and environmentally resilient future.

The investigation of nitrogen-doped lignin electrodes has shown great promise for enhancing the performance of sodium-ion batteries (SIBs) while offering a sustainable and cost-effective alternative to traditional anode materials. Recent studies, such as those by Chen et al. and Fan et al. have demonstrated that nitrogen doping significantly improves the electrochemical properties of lignin-derived carbon materials while enhancing their conductivity, mechanical stability and reactivity. Chen et al. synthesized nitrogen-doped porous ultrathin LC via pyrolytic treatment of alkaline lignin with melamine and urea while resulting in a material with a high surface area, significant pore volume, and improved sodium-ion storage kinetics in order to show excellent cycling stability after 5000 cycles at a high current rate. Fan et al. synthesized nitrogen-doped lignin-based carbon microspheres (NLC) using a hydrothermal process with 3-aminophenol while creating microspheres with an ultra-microporous structure and enlarged interlayer spacing in order to enhance the material's sodium-ion storage and intercalation performance. These studies highlight the importance of nitrogen doping in improving electrochemical reactivity and ion storage capacity, with both LC and NLC showing impressive rate capabilities and long-term cycling stability. Additionally, Zhang et al. employed an emulsion-solvent evaporation strategy to create nitrogen co-doped HC microspheres (NPL-1300) from lignin while demonstrating excellent sodium-ion storage, high Coulombic efficiency, and remarkable capacity retention. Hence, nitrogen-doped lignin electrodes offer enhanced performance due to their unique porous structures, abundant active sites, and improved electrochemical stability

while making them a promising material for next-generation and sustainable energy storage technologies.

Further analysis on the nitrogen-doped lignin electrodes should seek to improve the methods of synthesis to optimize the electrodes' electrochemical performance and commercial scalability. One such promising direction includes devising novel nitrogen-doping precursors and pyrolysis processes to relieve diffusion limitations and improve porosity along with surface chemistry of the material as well as ion diffusion and capacity retention over long cycling periods. Moreover, the addition of other heteroatoms (sulfur or phosphorus) could additionally improve the electronic conductivity and the electrochemical stability of the lignin-based carbon materials. Researchers ought to consider new lignin sources from agricultural wastes or wood by-products while ensuring the economically sustainable practices to improve the final material properties. To solve the problem with scalability, the development of low-cost environmentally benign processing methods could facilitate the mass production of nitrogen-doped lignin electrodes with appropriate performance. In addition, further detailed investigations on the practical use and durability issues of these materials in full-cell setups would make contributions to understand the potential of these materials in energy storage systems.

6. Lignin-Derived Electrode Materials for Potassium-Ion Batteries (KIBs)

Lignin-derived electrode materials for KIBs herald a revolutionary synthesis of biomolecular ingenuity and next-generation

electrochemical innovation, which is poised to transcend conventional paradigms in energy storage technology. Lignin presents an extraordinary repository of structural complexity and functional versatility while positioning it as an eminently viable candidate for the development of high-performance electrodes in KIBs. With potassium, the integration of lignin into KIBs not only reflects the convergence of sustainability with cutting-edge materials science but also offers an unparalleled opportunity to craft electrodes that exhibit exceptional electrochemical properties.^[178] When subjected to advanced de-polymerization, activation, and carbonization processes, lignin-based materials unveil remarkable capabilities which include superior charge storage capacity, long-cycle stability, and exceptional rate performance. In this context, lignin represents not merely a sustainable feedstock, but a transformative material that can rival or even outperform traditional electrode materials.^[179] The exploration of lignin in KIBs is emblematic of a future where biomaterials form the cornerstone of scalable, environmentally harmonious, and high-efficiency energy storage solutions.

6.1. Pristine/Carbon-Coated Lignin Electrodes

Pristine- and carbon-coated lignin electrodes have emerged as a highly promising and innovative class of materials for KIBs while offering a sustainable and eco-friendly alternative to traditional energy storage systems. Lignin is known as a naturally abundant and renewable biopolymer, which possesses a unique and versatile chemical structure that can be engineered to enhance electrochemical performance. By strategically coating lignin with carbon, its intrinsic limitations (poor conductivity and insufficient structural integrity) are effectively mitigated. The carbon coating not only amplifies electrical conductivity but also stabilizes the material's architecture while enabling superior potassium-ion storage and faster ion diffusion. This synergistic combination of renewable lignin and conductive carbon delivers remarkable improvements in rate capability and cycling stability while making it a formidable contender for next-generation KIBs. The dual-modification approach offers a compelling strategy for harnessing the untapped potential of lignin as a low-cost and high-performance electrode material while positioning carbon-coated lignin as an exciting candidate for scalable, high-efficiency energy storage applications. Porous HC anodes present a significantly enhanced cycle stability and superior discharge capacity for KIBs when compared to conventional graphite anodes. However, the majority of commercially available HCs are derived from relatively costly high-molecular-weight polymers. Zhu et al. ingeniously repurposed industrial by-products from the pulping and papermaking to synthesize lignin as HC.^[180] Black liquor solids and sludge were meticulously blended via ball milling and subjected to co-pyrolysis while resulting in the formation of highly porous HC anodes (BLDSC) (Figure 23a). The direct pyrolysis of black liquor yields HCs with remarkable efficiency while obviating the need for lignin extraction. Furthermore, the pore-expanding agent derived from deinking sludge significantly augments the pore structure of the resulting HCs while leading to the development of an abundant network of

mesopores as depicted through SEM and TEM (Figure 23b,c). The synthesized anodes exhibited an exceptionally low average capacity decay of just 0.06% per cycle at 100 mA g⁻¹ after 500 cycles along with exceptional rates at various current densities (Figure 23d,e).

This work demonstrates an innovative approach to utilize industrial by-products, specifically black liquor solids and deinking sludge, for synthesizing HC anodes. By blending these by-products via ball milling and subjecting them to co-pyrolysis, this study produces highly porous HCs (BLDSC) without the need for lignin extraction. The incorporation of the pore-expanding agent from sludge significantly enhances the pore structure while creating a network of mesopores that is beneficial for sodium-ion storage. The work showcases a sustainable and cost-effective method for converting waste materials into valuable energy storage components while emphasizing the potential for creating high-performance anode materials for sodium-ion batteries. This approach not only improves material efficiency but also advances the recycling and valorization of industrial waste in the context of energy storage technologies. Hard carbons have garnered significant attention as promising alternatives to graphite for anodes in KIBs. The integration of graphitic nanodomains into HCs can facilitate stable potassium-ion storage with diminished potential plateaus and reduced hysteresis. Zhong et al. tailored the crystalline structure of HC anodes through the incorporation of graphene oxide within renewable lignin-derived precursors (Figure 24a).^[181] The resulting modified HC (termed QLGC) exhibits well-defined graphitic nanodomains embedded in the carbon matrix while promoting stable potassium-ion de-intercalation dynamics (Figure 24b,c). As a result, the QLGC anode demonstrated a robust and stabilized capacity of 273 mAh g⁻¹ at 100 mA g⁻¹ while showcasing significant rates and sustained cycling stability (Figure 24d,e).

This approach shows a successful design and synthesis of a modified HC material (QLGC) with well-defined graphitic nanodomains and expanded interlayer spacing, which enhances potassium-ion de-intercalation. The modification leads to a more stable and efficient charge storage mechanism while suggesting that QLGC anodes are well-suited for use in KIBs. By employing electrokinetic analyses and comprehensive physicochemical characterizations, this study explores the charge storage mechanisms and crystallographic features of the anode material. This work also emphasizes the importance of engineering the crystalline structure of HCs to improve the electrochemical performance and long-term cycling stability of KIBs while contributing to the development of next-generation energy storage technologies. Lignin is exceptionally abundant in aromatic units. While precursors laden with aromatic structures generally yield soft carbon upon carbonization while forming a rigid framework that results in HC, Jiang et al. employed a graphene oxide-catalyzed decomposition and reassembly process to intricately modify lignin's side chains (Figure 25a).^[182] Compared to directly carbonized lignin, the carbonized reconstructed lignin exhibited significantly fewer defects and micropores (Figure 25b,c).

These characteristics can be tuned by adjusting the amount of graphene oxide catalyst. When tested as an anode for K-ion batteries, the carbonized reconstructed lignin delivered a higher

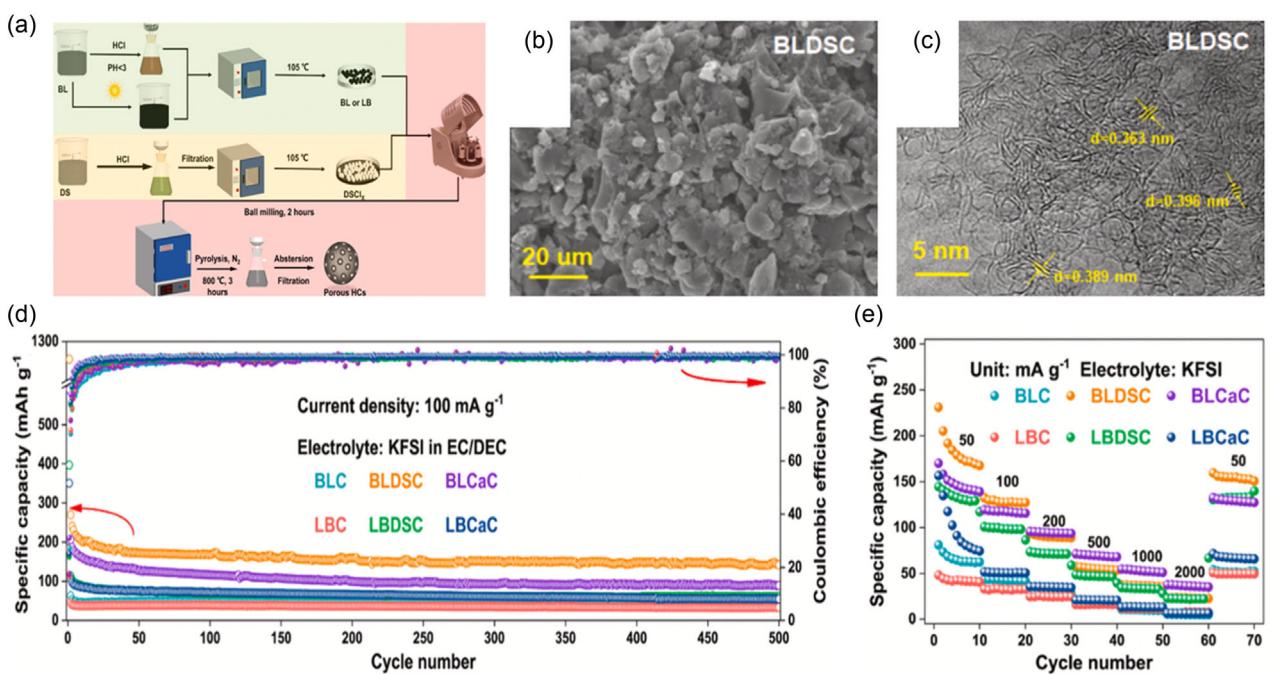


Figure 23. a) Schematic representation of the multistep synthesis process for BLDSC. b,c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of BLDSC electrode. d) Long-term cycling stability and electrochemical performance of the synthesized electrodes. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[180] Copyright 2024, Elsevier.

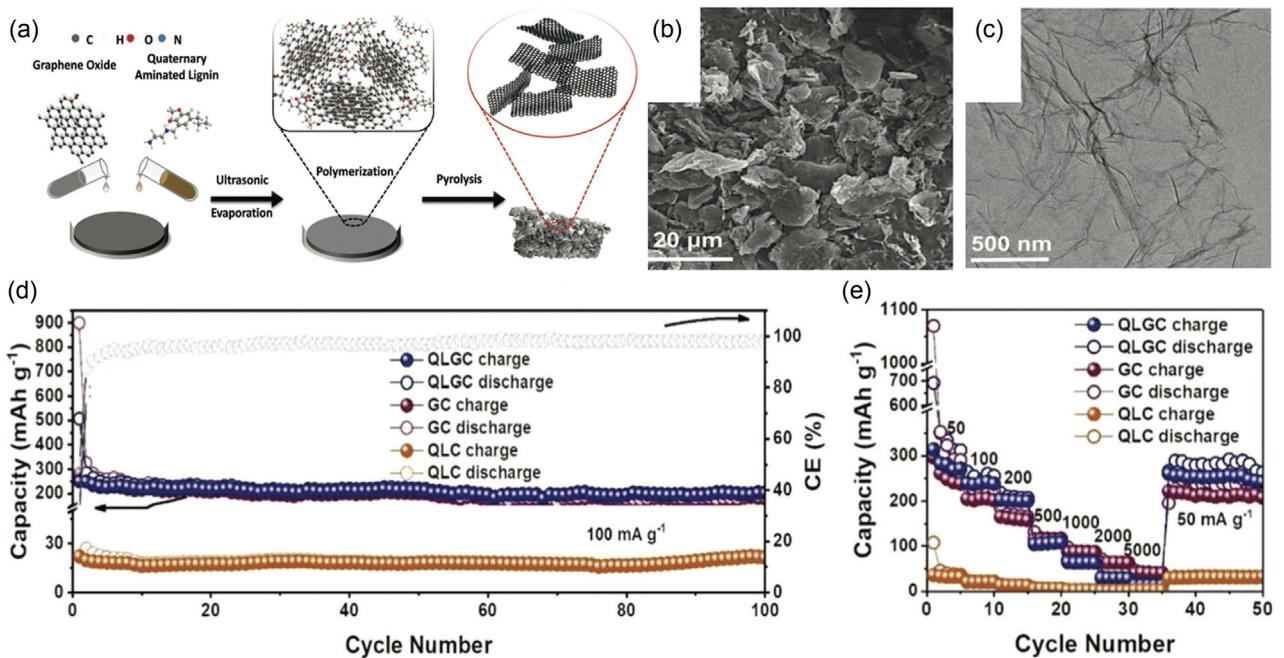


Figure 24. a) Schematic representation of the multistep synthesis process for QLGC. b,c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of QLGC electrode. d) Long-term cycling stability and electrochemical performance of the synthesized electrodes. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[181] Copyright 2022, Wiley-VCH GmbH.

capacity of 211 mAh g⁻¹ at 0.3 A g⁻¹ at low-potential ranges along with exceptional rates (Figure 25d,e). This work highlights that voltage hysteresis in K-storage is a significant issue and should be prioritized in the design of future electrode materials. The reconstruction strategy presented in this work offers a

promising and low-cost solution for improving K-ion battery performance. This study unveils a novel strategy for enhancing the electrochemical characteristics of lignin-based HCs in KIBs. Through a graphene oxide-catalyzed decomposition and repolymerization process, lignin is selectively modified to diminish

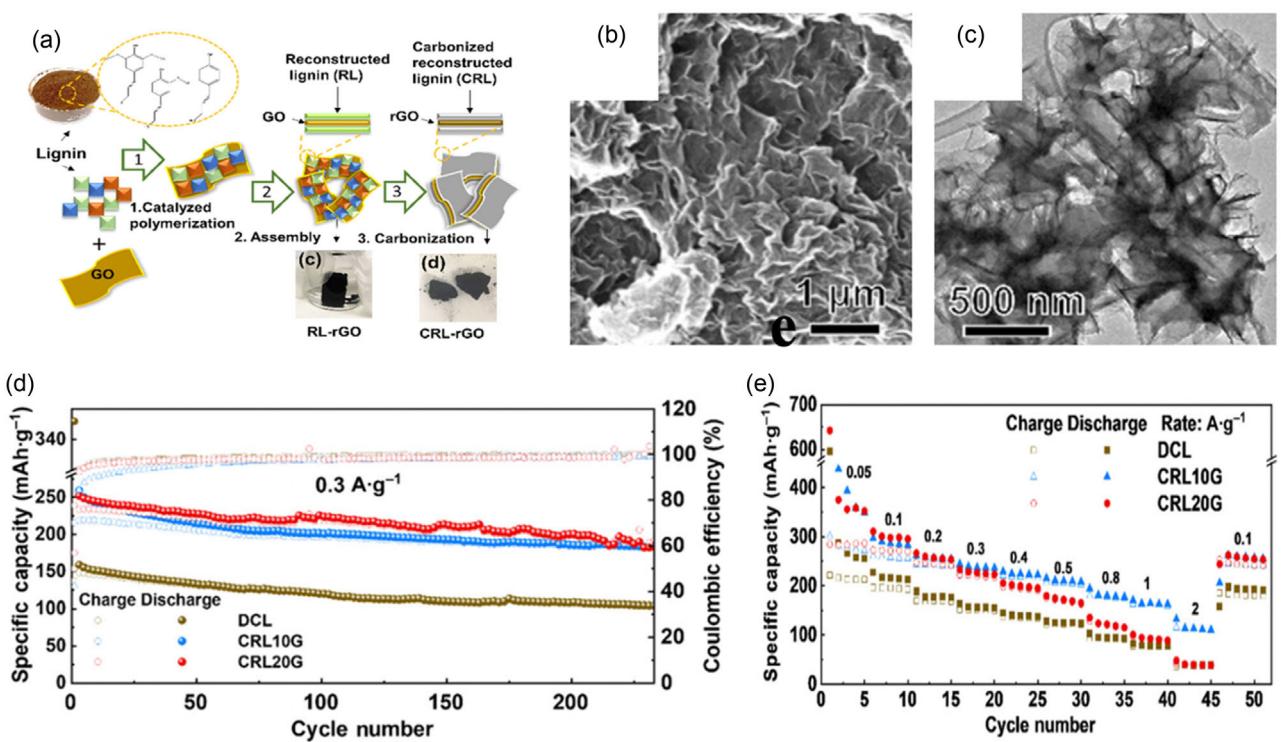


Figure 25. a) Schematic representation of the multistep synthesis process for CRL-RGO. b,c) SEM and TEM micrographs capturing the intricate surface morphology and structural attributes of CRL-rGO electrode. d) Long-term cycling stability and electrochemical performance of the synthesized electrodes. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[182] Copyright 2021, Springer.

defects, oxygenated functional groups, microporosity, and inter-layer spacing while augmenting K-ion storage stability and efficiency. The research highlights the critical role of minimizing voltage hysteresis to optimize energy efficiency and presents this structural tuning as a cost-effective method for improving KIB anodes. The carbon-coated lignin electrodes exhibit superior electrochemical performance, reduced voltage hysteresis, and enhanced cycling stability while maintaining reversible potassium-ion storage over extended cycles. This work offers a sustainable pathway for the development of high-performance lignin-derived electrodes for commercial energy storage systems.

Pristine- and carbon-coated lignin electrodes are emerging as sustainable and high-performance materials for KIBs, with modifications to enhance their conductivity and stability. Lignin benefits from carbon coatings that improve electrical conductivity and potassium-ion storage while leading to better cycling stability and rate capability. Researchers have also utilized industrial by-products like black liquor solids and deinking sludge to produce porous HC anodes while offering improved cycling stability and reduced costs. Notable advancements include Zhu et al.'s work with waste-derived materials while achieving 0.06% capacity decay per cycle over 500 cycles. Further improvements have been made by integrating graphene oxide, which enhances the crystalline structure and potassium-ion de-intercalation, as observed in Zhong et al.'s development of modified HC (QLGC) with a stable capacity of 273 mAh g⁻¹ at 100 mA g⁻¹. Similarly, Jiang et al. modified lignin's side chains with graphene oxide while reducing defects and enhancing ion storage stability which lead to a higher capacity of 211 mAh g⁻¹

at 0.3 A g⁻¹. Among these methods, Zhong et al.'s integration of graphene oxide into the HC structure stands out with the highest stable capacity of 273 mAh g⁻¹ at 100 mA g⁻¹ while demonstrating superior electrochemical performance compared to other approaches. These collective innovations have significantly improved lignin-based electrodes' electrochemical performance while positioning them as a sustainable alternative for next-generation energy storage technologies. Hence, these advancements underscore the significant potential of lignin-based electrodes as a sustainable and cost-effective solution for energy storage while demonstrating how leveraging waste materials and enhancing the properties of lignin can pave the way for more environmentally friendly alternatives to traditional battery technologies.

The modification of electrodes made from lignin for KIBs demands attention in a couple of aspects to improve their effectiveness and ease of use. First, more advanced methods of carbon coatings should be studied to enhance uniformity and conductivity. Moreover, optimizing the integration of graphene oxide with lignin may also involve reduction of imperfections at the molecular level to enhance ion storage capacity. Combination of lignin with other natural polymers or inorganic materials like transition metal oxides could also be studied to form hybrid materials with high energy densities and cycling stability. The creation of economical and scalable methods for the production of porous carbon anodes from waste materials like black liquor solids and deinking sludge also seems promising. If the processes used to enhance the energy density of the electrodes with reduced environmental impact are aligned with environmental

sustainability, then materials based on lignin could be proposed widely as electrodes for next-generation energy storage systems.

6.2. Other Lignin-Based Electrodes

Lignin-based materials are emerging as promising alternatives for advanced potassium-ion battery (KIB) electrodes while offering sustainability and cost-effectiveness. Recent studies highlight zinc-coated lignin composites and carbon nanobelt aerogels (CNBs), which show significant improvements in capacity and stability. Zinc ions enhance charge storage and strengthen electrode structure while boosting overall performance. Additionally, the incorporation of lignin-derived CNBs onto VSe₂ nanosheets (VSe₂@CNB) provide a high surface area and conductive framework that enhances the potassium-ion storage capacity of the electrodes. These innovative lignin-based electrode designs not only optimize the electrochemical performance but also contribute to the sustainability of KIBs by utilizing renewable biomass resources. This section reviews the latest advancements in lignin-based electrodes while focusing on the role of zinc coatings and CNBs in improving the performance of KIBs. KIBs are gaining attention as a promising option for grid-scale energy storage due to their potential for low-cost operation. Although carbon is a leading candidate for KIB anodes, but carbon anodes still face significant challenges related to limited cycle life and rate capability due to the small interlayer spacing of sp²-hybridized carbon. In this context, Wang et al. synthesized a biomass-derived carbon with a significantly expanded interlayer spacing of 0.44 nm through a zinc-assisted pyrolysis process (Figure 26a).^[183]

This unique structure confers the carbon with remarkable electrochemical properties, including superior capacity, rate

capability, and cycling stability. The increased interlayer spacing facilitates faster potassium ion diffusion and mitigates volume expansion during the potassiation process while enhancing rate capability and cycle durability. Moreover, the carbon's interconnected network structure and the porous morphology shortens the electron and ion transport paths while optimizing performance (Figure 26b,c). This study demonstrated that the carbon's large capacity of 184 mAh g⁻¹ after 10,000 cycles at 5 A g⁻¹ was maintained along with excellent rate capabilities at different current densities (Figure 26d,e). These impressive characteristics are attributed to the large interlayer spacing which promotes efficient potassium ion intercalation and reduces volume expansion. Additionally, doping enhances the K⁺ storage capacity, while the modified graphitic structure improves rate capability and cycle stability.

This work presents a biomass-derived carbon anode for KIBs with expanded interlayer spacing which was achieved through a zinc-assisted pyrolysis process. The increased spacing improves potassium ion diffusion, reduces volume expansion, and enhances electrochemical performance. The unique carbon structure also promotes efficient electron and ion transport. This study also illustrates that expanding the interlayer spacing of carbon anodes can address key challenges in KIBs while making it a promising solution for high-power and low-cost energy storage for grid-scale applications. KIBs have attained considerable attention recently owing to their low cost, low redox potential, and the abundance of naturally available potassium. However, their performance has been hindered by the larger size of K⁺ ions and the substantial volumetric changes during charge and discharge cycles, which result in poor electrochemical kinetics and cycling stability. Feng et al. fabricated the material through

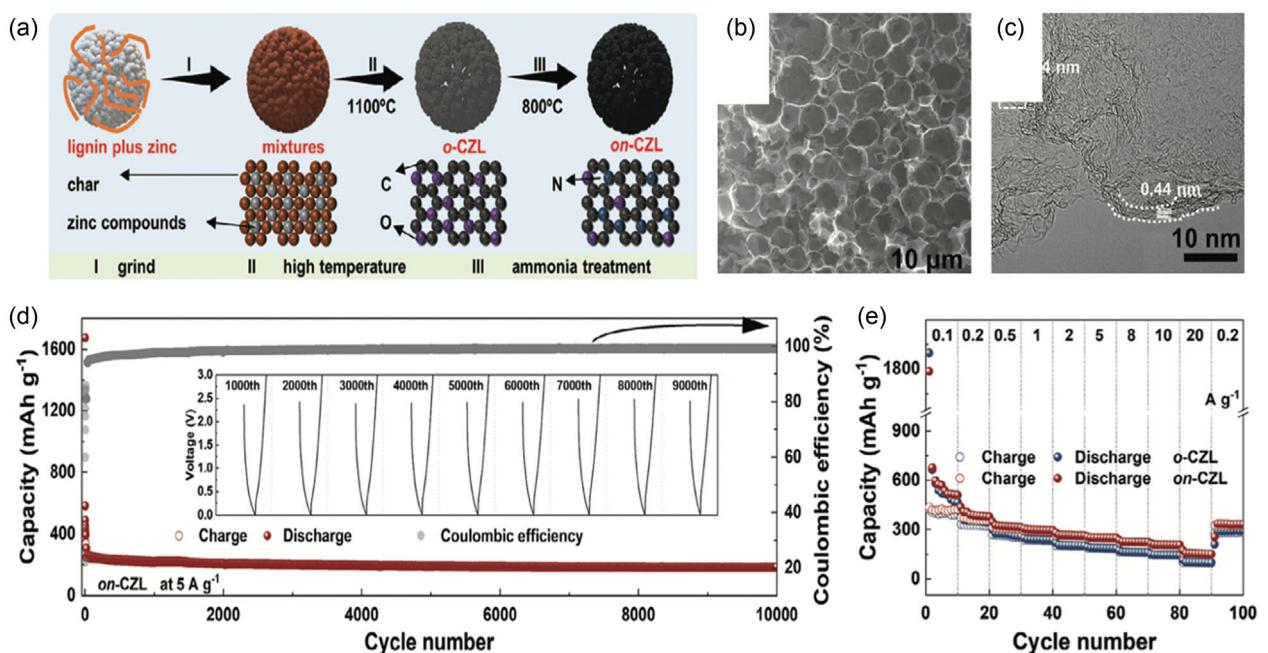


Figure 26. a) Schematic representation of the multistep synthesis process for on-ZCL electrode. b,c) SEM and TEM micrograph capturing the intricate surface morphology and structural attributes of on-ZCL electrode. d) Long-term cycling stability and electrochemical performance of on-ZCL. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[183] Copyright 2024, Wiley-VCH GmbH.

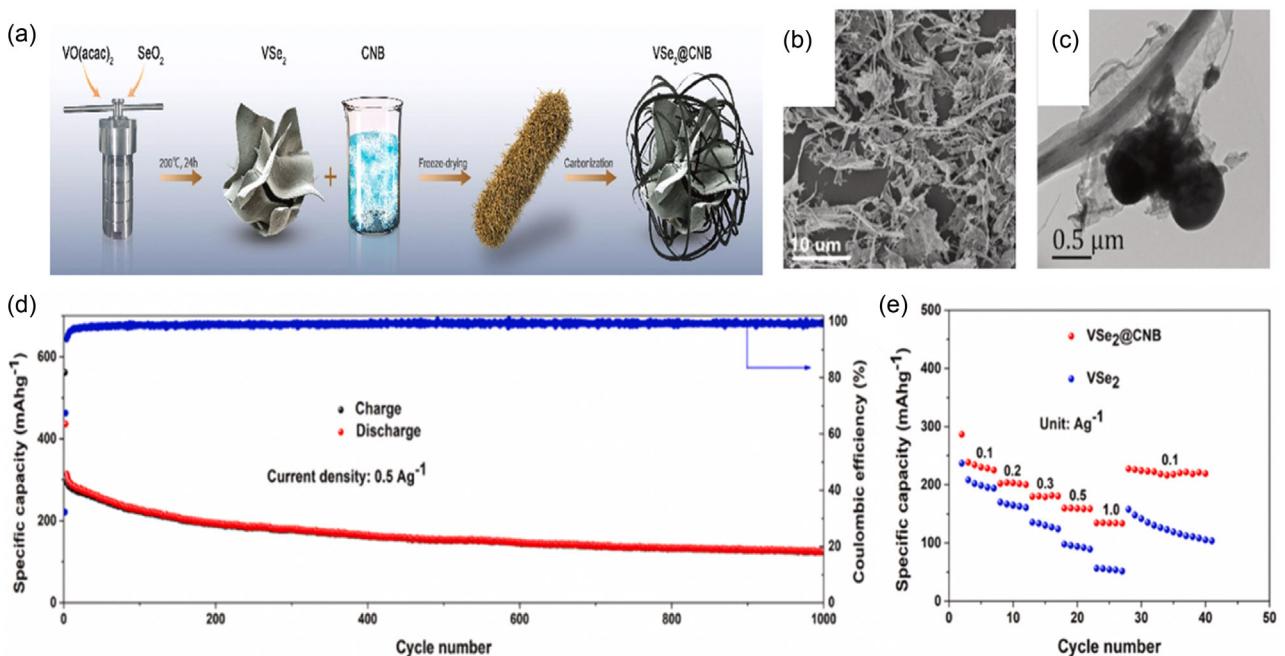


Figure 27. a) Schematic representation of the multistep synthesis process for VSe₂@CNB. b,c) SEM and TEM micrograph capturing the intricate surface morphology and structural attributes of VSe₂@CNB electrode. d) Long-term cycling stability and electrochemical performance of VSe₂@CNB electrode. e) Rate capability of the synthesized electrodes across a range of current densities. Adapted with permission.^[184] Copyright 2022, Elsevier.

the selenization of VO(acac)₂, followed by polymerization and coating with CNBs (Figure 27a).^[184] The resulting VSe₂@CNB composite offers enhanced conductivity and structural protection, owing to the interlinked and highly porous structure of the nanobelt aerogels (Figure 27b,c). The VSe₂@CNB demonstrated an impressive potassium-storage performance while achieving a high reversible capacity of 125 mAh g⁻¹ at 500 mA g⁻¹ after 1000 cycles, with a high Coulombic efficiency of 99% along with significant rates (Figure 27d,e). This work demonstrates the development of a novel anode material for KIBs while combining cellulose/lignin-based CNBs with VSe₂ nanosheets (VSe₂@CNB). The composite material is fabricated through a two-step process involving selenization and polymerization, followed by the coating of CNBs onto VSe₂ nanosheets. The resulting VSe₂@CNB composite offers enhanced conductivity, structural protection, and high surface area due to the porous and interlinked nature of the CNBs. The study shows that this composite material provides a promising solution for addressing challenges in KIBs, which includes poor electrochemical kinetics and cycling stability during charge and discharge cycles. The potassium storage mechanism is characterized by reversible phase evolution, where K⁺ ions intercalate into the VSe₂ nanosheets while inducing a conversion reaction that transforms VSe₂ into K₂Se. The work demonstrates that the VSe₂@CNB composite anode material offers improved electrochemical performance while making it a strong candidate for use in KIBs for energy storage applications.

Lignin-based materials have proven to be a promising avenue for the development of advanced electrodes in KIBs. The inclusion of zinc coatings and lignin-derived carbon has significantly elevated both the capacity and stability of KIBs. Zinc-coated lignin composites benefit from the synergistic effects of zinc ions,

enhancing charge storage and maintaining electrode integrity, while lignin-based CNBs provide a high-surface-area, conductive network that boosts potassium-ion storage, and cycling performance. Lignin-based materials are emerging as a promising solution for sustainable and cost-effective KIB electrodes, with recent studies focusing on zinc-coated lignin composites and CNBs/CNBs that significantly improve capacity, stability, and overall performance. Zinc ions in these composites enhance charge storage, strengthen the electrode structure, and boost KIB performance, while incorporating lignin-derived CNBs onto VSe₂ nanosheets (VSe₂@CNB) provides a high-surface area and conductive framework that further enhances potassium-ion storage capacity. KIBs are gaining attention for their low-cost operation, low redox potential, and the abundance of potassium, but carbon anodes still face challenges related to limited cycle life and rate capability due to small interlayer spacing. Wang et al. addressed this by developing a biomass-derived carbon with expanded interlayer spacing of 0.44 nm through a zinc-assisted pyrolysis process while improving potassium-ion diffusion in order to reduce volume expansion and enhancing electrochemical performance. The resulting carbon exhibited remarkable capacity, rate capability, and cycling stability with 184 mAh g⁻¹ after 10,000 cycles at 5 A g⁻¹. Meanwhile, Feng et al. developed a composite material by combining cellulose/lignin-based CNBs with VSe₂ nanosheets, which showed enhanced conductivity, structural protection, and high surface area. This composite achieved a reversible capacity of 125 mAh g⁻¹ at 500 mA g⁻¹ after 1,000 cycles with excellent Coulombic efficiency (99%) while demonstrating the effectiveness of combining lignin-derived carbon materials with advanced nanomaterials to improve electrochemical performance. Wang et al.'s method uses a zinc-assisted pyrolysis process to expand

interlayer spacing in biomass-derived carbon while improving potassium-ion diffusion and cycling stability, while Feng et al.'s approach combines cellulose/lignin-based CNBs with VSe₂ nanosheets to enhance conductivity and structural protection while offering a more stable electrochemical environment. Both methods improve KIB performance and focus on multiple aspects which include ion diffusion, stability, conductivity, and surface area. Hence, lignin-based materials, particularly through zinc coatings and CNBs, optimize KIB performance while contributing to the sustainability of the technology by utilizing renewable biomass resources. These innovations address challenges like poor electrochemical kinetics and cycling stability while making lignin-based electrodes a promising option for next-generation, high-power, and low-cost energy storage systems for grid-scale applications.

The future work on lignin-based materials should target improving the interfaces with other nanomaterials by optimizing the synthesis methods. One of such advanced directions is the modification of the surfaces of ion conductive lignin-based composites by attaching certain functional groups to enhance their binding with potassium ions. In addition, the enhancement in specific surface area, electrical conductivity, and stability of KIB electrodes may be achieved upon studying the hybridization of lignin-based carbons with other 2D materials like graphene or MXenes. With regard to the electrochemical performance, it

is important to focus further on the influence of the zinc coating thickness on the lignin composites in order to optimize the charge/discharge cycles and rate capabilities of the electrode. In addition, there are concerns about the ability to maintain consistent performance across scalable commercial applications of these materials. Therefore, there is a need to design reasonable and efficient approaches for the production of these composite electrodes. Hence, understanding the long-term cycling behavior and failure mechanisms of lignin-based KIB electrodes under real-world conditions will be essential for their practical implementation in grid-scale energy storage systems.

These innovations not only enhance the electrochemical properties of KIBs but also promote the sustainability of the technology by utilizing renewable biomass resources. As research continues, lignin-based electrodes are poised to play a critical role in advancing the performance and sustainability of KIBs, contributing to the development of next-generation energy storage systems. The incorporation of lignin-derived electrode materials into KIBs represents a transformative leap in the nexus between sustainable materials science and cutting-edge electrochemical engineering. The inherent complexity and exceptional electrochemical attributes of lignin, when strategically exploited through advanced material processing techniques, yield electrodes that challenge and potentially surpass the performance of conventional alternatives in terms of capacity, cyclability, and

Table 1. Summary of electrochemical performance of lignin-based anodes for AMIBs.

Electrode	Batteries	Electrochemical performance [mAh g^{-1}]	Current density [mA g^{-1}]	Cycles	Surface area [$\text{m}^2 \text{g}^{-1}$]	References
CCL porous carbon	LIB	500	50	100	233	[159]
Hard carbon	LIB	584	50	100	707	[160]
Lignin-derived porous carbon	LIB	528	200	200	596	[161]
Lignin-derived porous carbon encapsulated SiO ₂	LIB	1109	100	100	1107	[162]
Lignin-based porous hollow carbon shell frameworks	LIB	480	200	200	980	[163]
Rice husk lignin-based porous carbon	LIB	898.	100	110	539	[164]
3D hierarchical Lignin-derived porous carbon	LIB	550	200	200	1300	[165]
Si-Lignin	LIB	759	100	1300	1670	[166]
Si/C-LPR	LIB	605	500	100	1543	[167]
Si/C microsphere	LIB	1147	200	100	943	[168]
Rice husk lignin/SiO ₂	LIB	572	1000	1000	466	[169]
Si/C Composite	LIB	757	200	200	974	[170]
Sn/Lignin	SIB	374	20	100	353	[172]
Pristine lignin	SIB	262	200	60	6	[51]
Lignin-derived hard carbon	SIB	250	100	100	536	[173]
hard carbon	SIB	303	100	200	228	[174]
Lignin-derived nitrogen-doped porous ultrathin	SIB	138	150	5000	–	[175]
Nitrogen-doped lignin	SIB	233	50	300	727	[146]
N, P doped hard carbon	SIB	248	100	200	1148	[177]
Hard carbon	KIB	303	100	500	582	[180]
crystalline Lattice of hard carbon	KIB	283	50	100	163	[181]
Lignin/Carbon nanosheets	KIB	220	300	250	320	[182]
Lignin	KIB	320	1000	600	–	[183]
Cellulose/Lignin	KIB	227	100	100	237	[184]

rate capability. The use of potassium ions as charge carriers, coupled with the renewable nature of lignin, offers an unprecedented opportunity to mitigate the ecological and economic challenges associated with traditional energy storage systems. This synthesis of biological material with high-tech innovation not only promises to diversify the material landscape of KIBs but also aligns seamlessly with the global imperative for greener, more efficient, and cost-effective energy solutions. The ongoing refinement of lignin-based electrodes stands to revolutionize the energy storage paradigm, setting the stage for a new era of scalable, sustainable, and high-performance battery technologies. As such, the valorization of lignin in KIBs is poised to play an integral role in shaping the future trajectory of energy storage, offering a compelling vision of a more resource-efficient and environmentally resilient technological ecosystem. A brief summary of electrochemical performance of lignin based anodes for AMIBs has been depicted in **Table 1**.

7. Challenges of Lignin-Derived Electrode Materials in AMIBs

Lignin-derived electrode materials for AMIBs are rapidly garnering unprecedented attention, fueled by the urgent demand for eco-friendly solutions in next-generation energy storage devices. Lignin emerges as an exceptionally compelling candidate for electrode fabrication, owing to its renewable, bio-based nature and its rich carbon content, which can be meticulously transformed into highly conductive carbonaceous frameworks. However, despite the tremendous promise, the development of lignin-based electrodes is beset by a multitude of significant and intricate challenges that hinder their full realization and widespread application.^[185] One of the primary challenges is the low conductivity of pure lignin, which can limit the electrochemical performance of the electrodes. Lignin typically requires further processing which includes carbonization and incorporation with conductive additives in order to improve its electrical conductivity. An additional impediment lies in the comparatively suboptimal cycling stability and rate performance of electrodes derived from lignin. These materials frequently undergo deleterious structural transformations or progressive deterioration throughout successive charge–discharge cycles while culminating in a marked diminution of their capacity retention as the cycling regimen advances.^[186]

Lignin-derived electrodes exhibit lower energy densities compared to traditional materials, which are mostly used in commercial AMIBs. Hence, developing a cost-effective, efficient, and environmentally sustainable manufacturing process remains a key obstacle. Additionally, lignin's inherent heterogeneity complicates ensuring consistent quality and performance. However, future strategies such as combining lignin with conductive polymers or metal oxides and refining carbonization techniques could help address these issues and enhance large-scale viability.^[187] Advancements in material design could significantly enhance its conductivity and structural integrity. Additionally, refining the carbonization process may improve cycling stability and

increase energy densities while paving the way for more efficient energy storage solutions.^[188] Moreover, the incorporation of lignin-derived constituents into hybrid or composite matrices could further amplify the overall electrochemical efficacy of the electrodes. The long-term outlook for lignin-based anode materials in AMIBs appears exceptionally auspicious, especially with regard to their sustainability and economic viability.

As ongoing research progressively refines the understanding of lignin's intrinsic electrochemical properties, lignin-derived electrodes may emerge as a plausible and competitive substitute for conventional materials while fostering more ecologically sound and cost-effective energy storage solutions. However, a holistic and multidimensional strategy will be crucial in overcoming the myriad technical and material challenges that currently hinder their widespread commercial deployment.^[189] Foremost among these challenges is the enhancement of electrical conductivity, the stabilization of structural integrity during repetitive cycling, and the scalability of production techniques. To overcome these limitations, it is imperative that advanced production must be prioritized. Such research will facilitate a more precise control over the structural design, porosity, and overall material properties while ensuring greater stability and durability under operational conditions.^[190] Surface modification strategies, the incorporation of functional additives, and novel processing techniques present promising avenues for bolstering the performance characteristics of lignin-derived electrodes.

Lignin is primarily generated as a by-product during the pulping procedure of paper and bioethanol industries. Despite its potential, the widespread adoption of lignin as an electrode material faces several challenges, particularly due to the low purity and inconsistent quality of lignin produced by current extraction methods. These methods (kraft, sulfite, and organosolv processes) are optimized for other applications but not for producing high-quality lignin suitable for energy storage devices like AMIBs.^[191] The relatively low yield of high-purity lignin, combined with the complex and energy-intensive extraction processes, increases production costs while limiting its commercial viability as a direct electrode material. One of the main hurdles in using lignin for AMIB electrodes is its structural complexity and inherent heterogeneity. Lignin macromolecules vary widely in composition, which negatively impacts their electrochemical performance and makes it difficult to utilize lignin directly in battery systems without further refinement.^[192]

To improve lignin's performance, significant modifications such as chemical treatments, carbonization, and hybridization with conductive materials are often required. These modifications, however, come with additional costs and complexities, especially when compared to conventional electrode materials like graphite or commercial LIB materials. Nevertheless, lignin remains an attractive alternative due to its sustainability. It is a renewable and widely available resource that could reduce reliance on scarce and expensive raw materials like graphite, cobalt and lithium, which are commonly used in lithium-ion and sodium-ion batteries. Lignin can be sourced from agricultural residues, forestry by-products, and even municipal waste while making it an environmentally friendly feedstock with the potential for low-cost production. As research progresses in optimizing the

extraction, purification, and modification processes, the cost of lignin-derived electrodes could decrease while positioning them as competitive alternatives to traditional materials in terms of both performance and cost efficiency.^[93] For lignin-based electrodes to become commercially viable, cost-effective methods for lignin extraction, purification and conversion into functional electrode materials need to be developed. Innovations in biorefinery technologies and process optimization could help in lowering production costs while improving the electrochemical performance of lignin-derived materials. Additionally, strategies like doping with metal ions, hybridization with conductive polymers and carbonization could enhance lignin's energy density, cycle stability and conductivity while addressing key limitations of lignin-based electrodes. While these approaches show promise, they may introduce further processing steps and costs. However, if these challenges are addressed, lignin-derived electrodes could offer a sustainable, cost-effective and high-performance alternative to conventional materials while playing a significant role in the future of energy storage. The future of lignin-derived electrodes for AMIBs hinges on the development of scalable, efficient production processes and ongoing improvements in electrochemical performance. With continued research and technological advancements, lignin could become a key player in the next generation of energy storage technologies while offering a renewable and cost-effective alternative to current battery materials.^[193] Moreover, ensuring compatibility with existing energy storage technologies necessitates the development of innovative components and manufacturing approaches that can seamlessly integrate lignin-derived electrode materials with conventional systems. Furthermore, to catalyze progress in this area, standardized testing protocols and regulatory frameworks must be established to expedite the evaluation and commercialization of lignin-based electrode technologies.

Lignin-derived electrode materials for AMIBs show great potential, but their development faces key challenges that must be addressed for commercial use and better performance. One major issue is the large initial capacity loss and low first-cycle Coulombic efficiency of lignin-based electrodes.^[194] This inefficiency principally arises from the inherent fragility and electrochemical instability of untreated lignin compounded by the genesis of a solid-electrolyte interphase (SEI) that irrevocably consumes alkali-metal ions during the initial charge-discharge cycles. The pronounced early-stage capacity attenuation represents one of the most formidable impediments to the utility of lignin-based electrodes. This phenomenon is attributed to the propensity of raw lignin to undergo irreversible chemical alterations or morphological reconfigurations during the preliminary cycling stages. These alterations frequently facilitate the development of an SEI layer or other irreversible processes while inducing substantial capacity degradation. Although such behavior is ubiquitous among nascent electrode materials, it is particularly exacerbated in lignin due to the heterogeneity of its macromolecular architecture and the presence of diverse functional groups. Enhancing the Coulombic efficiency during the initial cycle is indispensable for optimizing the overall performance while positioning lignin-based electrodes as a plausible alternative to conventional materials.

Lignin's intricate and polymorphic architecture is characterized by disparate proportions of diverse phenolic constituents while giving rise to erratic electrochemical performance. The inherent heterogeneity within lignin-derived electrodes induces significant variability while undermining their cycling stability and capacity retention over successive charge-discharge cycles.^[195] The structural intricacy of lignin can impede the uniform dispersion of both ions and electrons while detrimentally influencing its rate capability and overarching electrochemical performance. Furthermore, lignin's intrinsic insulating properties present a formidable obstacle in its application for energy storage systems. Despite lignin-based electrodes demonstrating promising specific capacity, their inherently low electrical conductivity severely constrains their performance at elevated charge-discharge velocities. To address this issue, lignin often needs to be modified or hybridized with conductive materials like CNTs, graphene, or conductive polymers. However, these modifications can increase the complexity and cost of production while limiting the scalability of lignin-based electrodes. One of the most attractive features of lignin is its renewable and abundant nature.^[196] It is widely available as a by-product in industries such as paper manufacturing and bioethanol production and can also be sourced from agricultural residues and municipal waste.

The article examines the barriers that stand in the way of practical lignin-based electrode usage by focusing mainly on electrical conductivity limitations and structural instability challenges. Two aspects remain vital for achieving proper functioning and reliability of energy storage system electrodes. These limitations appear within lignin based electrodes due to their complex molecular structure. Their electrical conductivity remains poor since the polymeric framework of lignin contains no free-moving electrons that would enable efficient electricity transmission.^[197] Lignin-based materials degrade structurally during cyclic charge-discharge operations due to an effect on their mechanical stability.^[198] The degradation process decreases the long-term effectiveness of lignin-based electrodes than traditional metals and carbon conductive materials. The successful integration of lignin into energy storage technologies demands the resolution of these operational constraints.^[199]

The first strategy suggested to tackle these challenges is novel material design. One potential solution is to modify the chemical structure of lignin to enhance its conductivity. Researchers could explore the incorporation of functional groups into the lignin structure that could facilitate electron transfer, or even design lignin derivatives with conjugated systems that improve charge mobility.^[17] For example, introducing aromatic groups or incorporating conductive heteroatoms such as nitrogen or sulfur could potentially increase the conductivity of lignin.^[200] Additionally, modifying the molecular weight or branching structure of lignin could lead to improved mechanical properties while making the material more stable and durable under repeated charging cycles. Such structural modifications could enhance both the electrical performance and the longevity of lignin-based electrodes while addressing the key issues of low conductivity and poor structural stability.^[201]

Another avenue for improving lignin-based electrodes is the development of composite materials. By combining lignin with

other conductive materials, such as CNTs, graphene, and conductive polymers, researchers could create hybrid materials that leverage the advantages of both components.^[123] Carbon-based materials, for instance, are known for their excellent electrical conductivity, while lignin provides a renewable and biodegradable base. The synergistic combination of lignin and carbon materials could significantly improve the conductivity of the electrodes without sacrificing their sustainability. Similarly, the addition of conductive polymers could further enhance the electrochemical properties of the composite materials.^[202] Composite materials could also help mitigate the structural instability of pure lignin electrodes, as the stronger and more stable components in the composite would improve the overall mechanical strength and resistance to degradation.^[203] The development of such composite systems is a critical step toward making lignin-based electrodes competitive with traditional electrode materials in energy storage applications. Furthermore, the incorporation of nanoscale materials into lignin-based electrodes could be another promising approach. Nanoscale materials (nanoparticles or nanowires) possess unique properties due to their high surface area and excellent electronic characteristics. When integrated into lignin, these nanoscale materials could enhance the conductivity and electrochemical performance of the electrode materials.^[204] For instance, incorporating metallic nanoparticles like silver or gold could improve electron flow, while nanowires made from conductive polymers could provide better electron transport pathways.^[205] This solution could address both the conductivity and structural stability issues by increasing the material's surface area and providing additional support to the lignin matrix while preventing structural breakdown during cycling.^[206]

Finally, the article concludes the importance of the preparation of advanced process in order to surmount the gaps in lignin-based electrodes. The traditional methods for the synthesis of lignin-based materials are generally not able to produce the microstructural properties required for high-performance electrodes.^[207] Nevertheless, electrodes with microstructures can be fabricated by means of innovative processing techniques such as electrospinning, hydrothermal synthesis, or sol-gel method while resulting in better microstructures and improved mechanical integrity. With the aid of these advanced preparation techniques, the morphology of the materials can be much better controlled while resulting in better conductivity and stability. For instance, electrospinning can therefore be employed to fabricate nanofibrous lignin-based materials with high surface area, better charge storage, and electron transport.^[208] However, hydrothermal synthesis could allow the formation of lignin composites with desired properties, particularly increased conductivity as well as strengthened structural stability. Hence, these advanced techniques, new material designs, and composite strategies have shown great promise in introducing lignin as a viably used electrode material for large-scale energy storage applications. Hence, while lignin-based electrode materials face significant challenges, this section presents several viable strategies to address these issues. The modification of lignin's chemical structure, the development of composite materials, the incorporation of nanoscale materials, and the utilization of advanced preparation processes are all key areas that hold promise for improving the performance

of lignin-based electrodes.^[209] As research continues to explore these directions, lignin could emerge as a sustainable, high-performance alternative to traditional electrode materials while offering both environmental benefits and enhanced functionality in energy storage technologies.

8. Future Prospects of Lignin-Derived Electrode Materials in AMIBs

Lignin-derived electrode materials for AMIBs are emerging as a promising solution in the pursuit of sustainable and eco-friendly energy storage technologies. Lignin offers a unique advantage as an electrode material due to its bio-based origin and carbon-rich content. As the demand for more environmentally responsible and cost-effective energy storage solutions intensifies, lignin has gained significant attention for its potential to replace traditional materials in AMIBs. However, despite its promising characteristics, lignin faces challenges such as low electrical conductivity, suboptimal cycling stability and lower energy densities compared to conventional electrode materials. Nevertheless, ongoing advancements in material design, carbonization techniques and hybrid composites are opening new pathways to enhance the performance and scalability of lignin-based electrodes. With further research and innovation, lignin-derived materials could offer a sustainable, efficient, and competitive alternative to conventional electrodes while paving the way for the next generation of energy storage devices.

Lignin is an environmentally friendly material that could help reduce dependence on scarce and expensive raw materials. This sustainability aspect aligns well with the growing demand for green technologies and circular economies while positioning lignin as a promising candidate for future electrode materials. With advances in biorefinery technologies and improved extraction methods, the cost of lignin-derived electrodes could be reduced. Moreover, lignin's biogenic origins make it a promising material for the development of low-cost and scalable AMIBs, especially if improvements in processing technologies can overcome current challenges related to purity and consistency.^[210] As research progresses, efficient purification and modification methods could allow for the high-yield, low-cost production of lignin-based electrodes that compete with traditional materials. Lignin's chemical structure offers the potential for customization through various modification strategies. For example, lignin can be carbonized to enhance its conductivity or hybridized with conductive polymers and metal ions to improve electrochemical performance. These strategies allow for the fine-tuning of lignin's properties to meet the specific demands of AMIBs, which includes enhancing rate capability, cycle stability and energy density. Continued innovation in material design and processing techniques could lead to optimized lignin-based electrodes with superior performance characteristics while helping them to compete with more established materials like graphite. Beyond cost and performance, lignin-based electrodes offer significant environmental benefits. Lignin is biodegradable and using it in energy storage systems could help mitigate the environmental impact

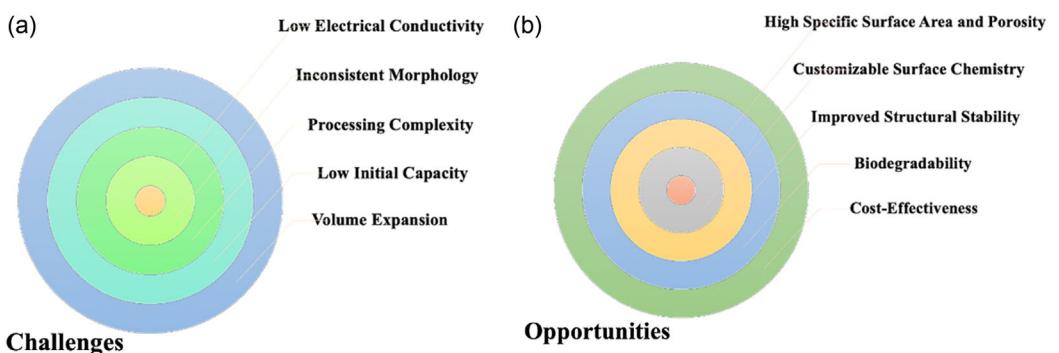


Figure 28. a) Certain challenges associated with lignin-derived electrodes. b) Key opportunities linked to lignin-derived electrodes.

of conventional battery materials, which often involve toxic or non-renewable substances.^[211] By incorporating lignin into battery technology, we can create more sustainable energy storage solutions that contribute to reducing the environmental footprint of modern energy systems.

Although lignin-based electrodes for AMIBs are promising and sustainable in terms of costs and the environment, but there are still gaps that need to be filled in order for them to be fully utilized. The primary challenges that remain obstacles along with structural and conductivity issues are high initial capacity loss, low first-cycle Coulombic efficiency, and low heterogeneity. Focusing on the extraction, purification, and modification of lignin can make electrodes more competitive and transform them into a sustainable alternative to traditional battery materials.^[212] Lignin-based electrodes have the potential to revolutionize affordable and sustainable alkali metal-ion batteries, change the future of energy storage, and help transform it into something more sustainable. The compatibility of these materials suggests that active tests in actual conditions are needed in order to determine versatility, durability and longevity within various energy storage systems. In order to fully utilize lignin-derived electrodes in energy storage applications, a multifaceted approach from both industrial and academic circles is required to maximize their innovation potential.^[213] The advancement of environmentally friendly and cheaper AMIBs that incorporate lignin-based electrodes for lignin energy systems is critical to the future of lignin electrodes. Additionally, the potential for scaling up recovery and processing of lignin from readily available biomass makes it an attractive material for large industrial applications. By increasing the performance, longevity, and economic factors of bio-based electrodes, lignin has the potential to revolutionize energy storage systems. Some of the major challenges and opportunities associated with lignin derived electrodes are shown in Figure 28a,b.

9. Conclusions

Lignin-derived anodes for AMIBs represent a promising avenue for developing sustainable and cost-effective energy storage solutions. As a renewable biopolymer, lignin offers numerous advantages which includes its abundance, low cost, and biodegradability. Recent investigations have elucidated the potential of

lignin as a highly effective constituent for anode materials to augment electrochemical performance. Lignin-based anodes exhibit commendable capacity, exceptional cycling stability, and notable aptitude for high-rate charge/discharge processes while positioning them as promising alternatives to traditional anode materials. However, several obstacles persist in the comprehensive optimization of lignin for applications in AMIB. Its relatively diminished electrical conductivity and inadequate structural robustness during repetitive cycling can precipitate a gradual decline in capacity over extended use. To mitigate these limitations, a range of sophisticated modifications, including the integration of conductive additives and the employment of carbonization techniques, have been rigorously investigated to elevate the material's overall efficacy.

In this review, we offer an in-depth examination of lignin-based electrode materials of diverse forms in AMIBs while focusing on the link between lignin's chemical/physical structure and the characteristics of its derived materials. Numerous studies confirm the viability of lignin and its derivatives as electrodes for rechargeable batteries. Yet, several challenges remain in advancing lignin-based AMIBs. The relationship between the chemical structure of lignin and the crystalline structure of lignin-based carbons remains unclear while posing challenges in understanding how lignin's complex macromolecular structure can be effectively transformed into carbon materials with varying graphite crystalline structures and functional groups crucial for the performance of AMIBs. Lignin offers significant potential as a sustainable material for rechargeable batteries, but its stabilization remains costly and inefficient due to its complex chemical composition. By applying chemical or physical modifications and exploring new techniques, these challenges can be addressed. Lignin-derived anodes could provide an eco-friendly alternative to conventional materials in next-generation AMIBs, but continued advances in conductivity, durability and scalability are crucial for realizing their full potential in energy storage applications.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: alkali metal-ion batteries • anode • energy storage • lignin-derived electrode • sustainability

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