

## Employment of Nanolignin in Energy Storage Devices

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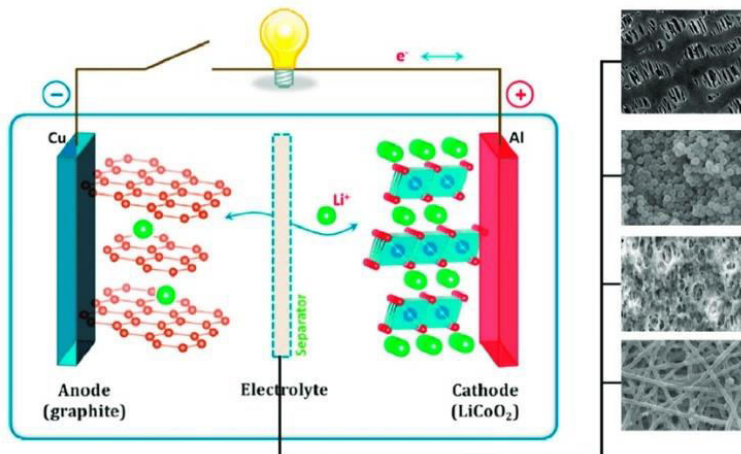
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### ABSTRACT:

In recent years, there has been a lot of focus on renewable energy storage devices as scientists attempt to optimize their performance. This paper delves into how research into nanolignin materials over the past few decades has aided in advancing energy storage devices because of nanolignin's good qualities (cheap cost, high availability, and lack of toxicity). This paper discusses the advantages and disadvantages of using nanolignin as a battery material. This paper begins with a review of state of the art in energy storage, lithium-ion battery fundamentals (including anodes, cathodes, and electrolytes), and nanolignin materials. A modification to increase the stability of energy storage and a case study of the Kraft nanolignin-carbon composite as a green cathode material is offered to illustrate the concept. In the following section, we discussed how oxidizing Kraft nanolignin yields cathode materials. Lastly, the study provides a high-level summary of the current state of the field, an analysis of the most urgent difficulties, and recommendations for future research into nanolignin-based energy storage devices.

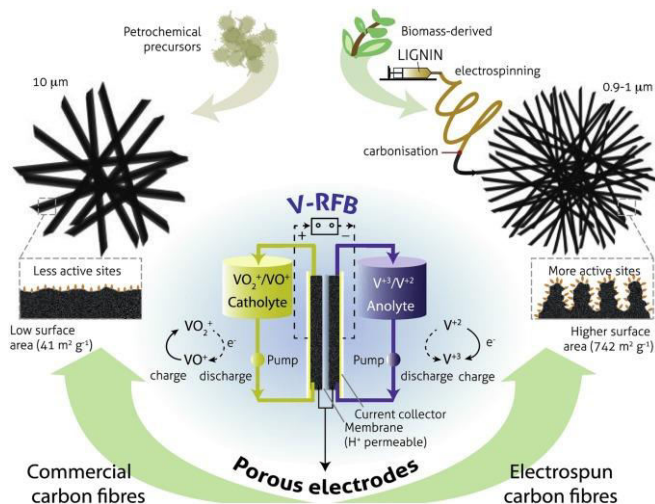
### 1. Energy Storage Technology

Energy is essential to modern life but poses significant economic, security, and environmental challenges. The outcome is a projected 60% increase in electricity demand between 2016 and 2040, alongside a 25% rise in global energy consumption. Electrochemical energy storage systems, such as electromechanical capacitors (ECs), fuel cells, and rechargeable batteries (including Li-ion batteries (LIBs), Na-ion batteries (NIBs), and K-ion batteries (KIBs)), are state-of-the-art in terms of storing and converting energy (Liu, Wan, et al. 2021). During the electrochemical energy storage, the electrodes and electrolytes undergo electronic and faradic procedures to store the fuel. To address the problems with current energy systems, scientists are investigating innovative sustainable, rechargeable energy solutions (Cao, Kitamoto, et al. 2021). Carbohydrates with porous microstructures, high mechanical strength, and excellent electrical conductivity are frequently used for this purpose. Since they are made from porous and high-surface activated carbons, graphite anodes used as conductive substrates in Li-S batteries are among the best alternatives to LIBs (Ma et al. 2021). Figure:1 is a schematic representation of the various electrochemical charge storage systems.



**Figure:1 Various Electrochemical Charge Storage Mechanisms. Reproduced from (Asghar, Anwar, and Naveed 2019) under the Creative Attribution Commons License 4.0 (CC-By 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

Therefore, it is necessary to research and create low-cost, sustainable precursors to design efficient, safe, and environmentally friendly methods of constructing carbon compounds. Due to its low cost, wide availability, quick renewal, and common environmental effect, biomass has been increasingly exploited during the past decade to develop carbon-based energy sources. In addition to being the most vital natural aromatic compound, it also contains a significant amount of paper and bio-refinery waste (Sun, Liu, and Li 2021b). Due to its peculiar polymeric structure, nanolignin is a valuable precursor for high-performance carbon compounds like graphene. Almost all available nanolignin was wasted since it was used as a cheap fuel to generate heat and power. Reusing nanolignin waste products as fuel for inexpensive energy generation is a significant cost-saver (Sun, Liu, and Li 2021a). Figure 2 depicts nanolignin-based electrodes for energy storage.



**Figure: 2 Nanolignin-based Electrodes for Energy Storage Application. Reproduced from (Liu et al. 2021) with permission from Elsevier.**

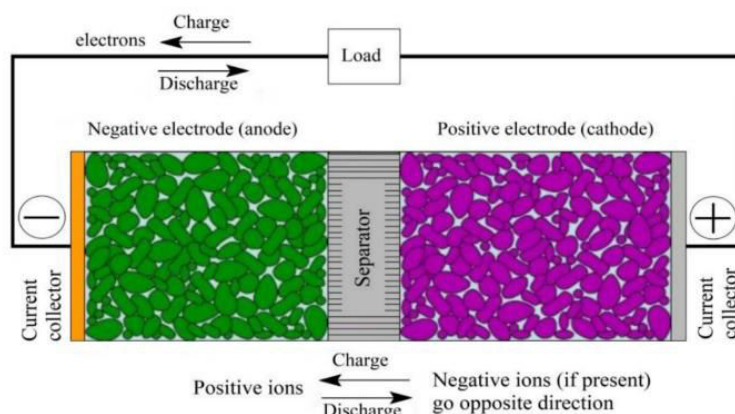
### **1.1.1 Related Work/Background**

None of the applications explored by José Luis and his team showed significant promise for nanolignin. The biopolymer's potential as a medium for storing energy has piqued the curiosity of scientists and researchers. Two-collector electrodes, electrochemical pseudo-condensers, fuel cells, lead-acid batteries, rechargeable batteries, and similar devices also use it. When needed, it can serve as a fuel-cell expander. Nanolignin decreases the cost and toxicity of energy storage devices while increasing their performance (Espinoza-Acosta et al., 2018). Reduced graphite oxide (RGO)/low-dimensional carbon (LDC) is used to create aerogels, which are extremely lightweight and elastic. In aerogels, the pores and cell walls are all neatly aligned. If the thickness and electrical conductivity of the cell walls are kept the same, but the interface polarization effect is increased, the cells become better at absorbing waves. Cell walls in aligned cell structures are larger and more integrated than in RGO aerogels, which boosts their multiple reflection properties. By contrasting the effects of various on-site compression operations in the parallel and normal directions, we can see the significance of microstructures in EMI shielding performance (Zeng et al. 2018). Li, Wenqi, and colleagues reported that nanolignin-derived carbon compounds have recently received attention as potential energy storage materials. The endocarps of walnuts and peaches were processed with a deep eutectic solvent (DES) to extract nanolignin. Sugar yields of over 90% were achieved with DES enzyme hydrolysis, which also allowed for the efficient removal of nanolignins. Endocarp biomass's molecular weight drops dramatically after treatment with DES nanolignin (Li 2019). Wu, Xiaoyu, and their team developed electrogene to create a high-performance electrical energy product and component. Nanolignin has demonstrated potential as a raw material; it is the second-largest natural polymer. Technological and economic assessments indicate that engineered nanolignin carbon structures may provide a cheap supply of carbon products. These findings demonstrate the benefits to both existing electrical power grids and nanolignin consumption/regeneration initiatives from increasing the usage of high-quality energy components created from nanolignin. Nanolignin-derived materials with great performance have specific requirements, and the author lays them out here (Wu et al. 2020). Kwang Ho Kim and colleagues detail recent developments in using nanolignin to produce polymers, dyes, glues, and fertilizers. Its value has shifted from a commodity to a potential source of precious metals and other resources. Recent studies have focused on nanolignin because of its potential medical, electrochemical, and multimedia applications. Nanolignin's physiological consistency, low cost, and availability make it a desirable material, but the difficulty of recovering and depolymerizing it is a significant scientific problem. Nanolignins have several potential applications in the medical industry, from pharmaceuticals to medical equipment to topical treatments. Moreover, they can be shaped into three-dimensional nanolignin-plastic structures. This research looks at the pharmacological, electrochemical, and additive manufacturing (AM) potential of nanolignin (Yu and Kim 2020). Jonatan and colleagues initially mentioned low-temperature thermal energy storage (TES) using the lignin-based polymer. Significant barriers exist to expanding the heat-storage capacity of lignin-based polymers (LBPs). Accordingly, the highest latent heat values were in LBPs rich in tetrahydrofuran (THF) and had low oxirane/THF mass ratios. It was shown that the melting temperatures of the manufactured LBPs were strongly inversely related to the oxirane/THF mass ratio. According to our best knowledge, low-temperature TES has been investigated and considered for nanolignin-based PCM-derived biomass. The energy required for the liquid-to-solid phase transformation in LBPs with bio-PCM potential (Perez-Arce et al., 2021). Liu claims that due to its high carbon content, low cost, and a massive number of functional groups,

nanolignin is the second-largest organic polymer in nature. There has been a rise in the use of nanolignin and its derivatives in recent years for these purposes, specifically as electrodes and energy storage media. Then, state-of-the-art nanolignin-based batteries and supercapacitors can be created with the highest quality electrons. Finally, an improved approach to energy storage based on nanolignin electrodes is described (Liu, Xu, Liu, Zhang, Liu, Li, Du, Si, et al. 2021). According to Diego and his colleagues, the Kappa value and the effectiveness of nanolignin are affected by the removal procedure, temperature, pH, resident duration, and pressure. The effectiveness and storage of activated carbon-based nanolignin highly depend on the amount of potassium hydroxide used. Metal doping reduces carbon and hydrogen storage capacity, creates fullerene-like cage structures, and makes the surface less accessible (Lobato-Peralta et al. 2021). According to Dayton and his colleagues, the modern energy economy is built on a foundation of fossil fuels. Large enough energy storage facilities are required to successfully store and transmit renewable energy even in the dead of winter. However, widespread use of energy storage will not happen unless significant progress is made in the field. Large amounts of energy can be stored in sodium-ion batteries with hard carbon anodes. Unlike lithium, sodium is both more affordable and better for the environment but requires a different anode design. To create the nanolignin-generated carbon composite, the nanoscale crystallites' amorphous graph-matrix structure is subject to intensive engineering (Kizzire 2021). According to Feijun, the proliferation of pollution, reliance on fossil fuels, and overall energy consumption drive the research and development of alternative energy sources. Supercapacitors utilize a variety of electrodes and polymer electrolytes. Biomass carbons and electrodes are now practical options for supercapacitors due to their attractive look, low cost, renewability, and environmental friendliness (Lin, Wang, and Shao 2021). The research group led by Zheng claims it is challenging to produce extremely bendable nanocarbon compounds from biomass. The production of high-quality green nano-green fibrous materials has been simplified and made more practicable. The chemical groups, branch structure, molecular weight, and spatial molecular architectures of nanolignin are only some of the many ways it is fractionated. Phosphatized nanolignin and cellular acetate are combined to create green carbon fibers (CF). Full, homogeneous biomass yields carbon molecules with irregular shapes and sizes (Zheng et al. 2021). According to Guo and coworkers, the electrochemical performance of a porous commercial carbon supercapacitor electrode (Guo et al. 2021).

### **1.1.2 Lithium-ion batteries (LIBs)**

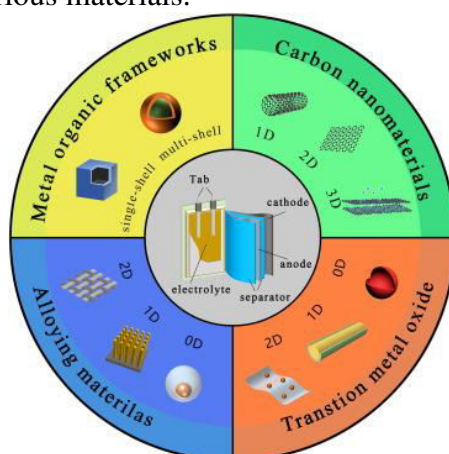
M.S. Whittingham sold the LIB chemical in 1976, and in 1991, SONY began commercial sales. LIBs have been widely adopted due to the portability of Li metal. In terms of electro positivity and electroactivity, lithium is the most prominent element on the planet (Chen, Wu, Ren, et al. 2021). A Lithium-Ion Battery Overview is shown in Figure: 3.



**Figure:3 Lithium-Ion Battery Overview. Reproduced from (Al-Gabalawy, Hosny, and Hussien 2020) under the Creative Attribution Commons License 4.0 (CC-BY 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

### 1.1.3 LIBs Anode materials

The low voltages required by LIBs make the anode materials useful in various chemical processes. As a result of its high electrochemical efficiency, metallic Li is frequently used as an anode material in primary Li cells. Rechargeable LIBs have a serious safety problem with dendrite development, but Li can solve this problem. Carbonate materials and tin-silver alloys received much attention (Shen and Gele 2021). As seen in Figure:4, anodes in lithium-ion batteries can be made from various materials.

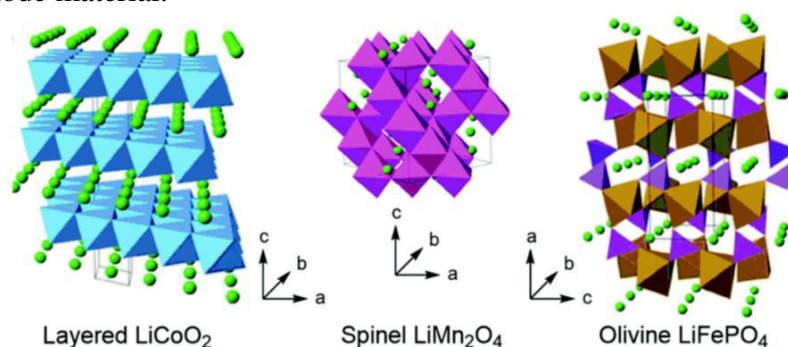


**Figure:4 Anode Materials of Lithium-ion Batteries. Reproduced from (Cheng et al. 2021) with permission from Elsevier.**

### 1.1.4 LIBs Cathode Materials

Due to the cathode materials, the intercalation method limits LIB's capability. Rechargeable LIBs have found commercial success thanks partly to using cathodes made of metal chalcogenides (such as  $\text{TiS}_2$  and  $\text{MoS}_2$ ), manganese or vanadium oxides, metallic lithium, metallic lithium alloys, and graphite. Over the past decade, multiple layered compounds, including  $\text{LiCoO}_2$ ,  $\text{LiNiO}_2$ , and the 3-dimensional  $\text{LiMn}_2\text{O}_4$  spinel phase, have been investigated as potential cathode materials (Wang, Wu et al. 2021).  $\text{LiMPO}_4$  ( $\text{M}=\text{Fe}, \text{Mn}, \text{Co}$ ), a novel polyanion-based chemical, has garnered much attention due to its low cost and lack of human health risks. Li-air and Li-S batteries, which are at the forefront of battery technology,

have the potential to outperform typical LIBs that rely on inserts vastly. The synthesis of renewable organic materials is an intriguing and "green" alternative to more conventional inorganic systems (Zhou et al. 2021). Figure: 5 depicts the structural and functional features of the  $\text{LiFePO}_4$  cathode material.

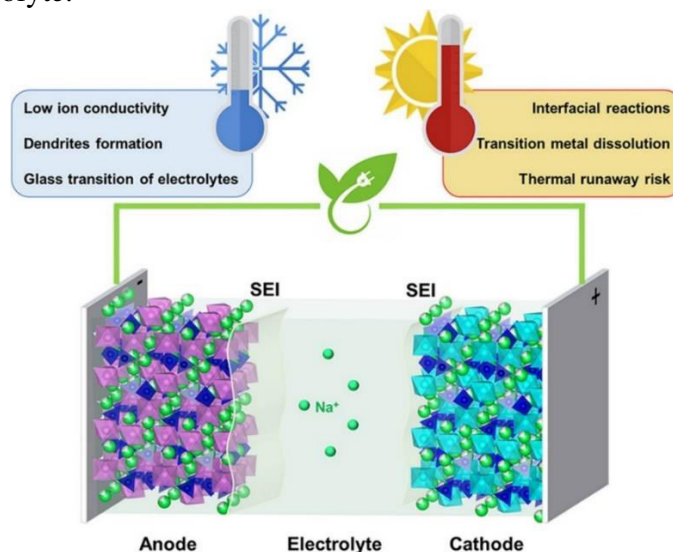


**Figure:5 structure and performance of the  $\text{LiFePO}_4$  cathode material. Reproduced from (Somo et al. 2021) under the terms of the Creative Commons License 4.0 (CC-By 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

### 1.1.5 Organic Electrodes for Batteries

#### 1.1.5.1 Introduction and Principle

The main ecological danger of inorganic electrodes is the infrastructure needed to recover, treat, and recycle hazardous or heavy metal waste. As a result of the high production energy requirements, greenhouse gases are frequently released to a meteoric rise in the popularity of organic electrode materials (Wu et al., 2022). The raw material cost is the most crucial variable in any marketing effort. As a result, the cathode might be responsible for up to 30% of the LIB cost of the device. It is crucial to cater to the needs of applications that don't call for outstanding performance but require materials. EESS electrodes began using dichloroisocyanuric acid as a cathode material in 1969. In a study by Xue et al. 2021, Figure:6 shows a sodium-ion battery with an organic electrolyte.



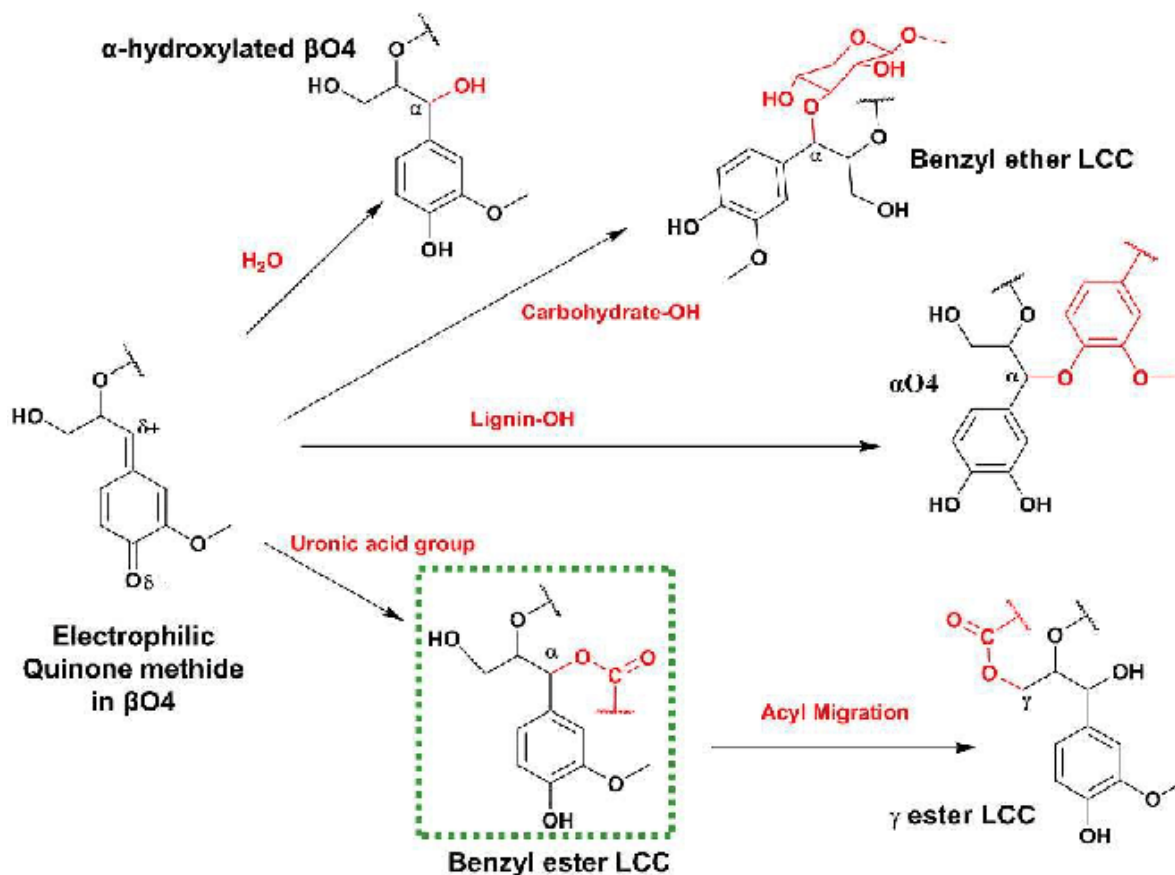
**Figure: 6 Sodium-ion Batteries in Organic Electrolytes. Reproduced from (Zhu and Wang 2020) under the terms of the Creative Attribution Commons License 4.0 (CC-By 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

Producing organic electrodes is difficult because of the solubility requirements of an electrolyte. The cycle's reliability decreases when the battery is discharged simultaneously as the active component. Incorporating a redox-active unit into a polymeric backbone is one approach to solving this issue. Until the turn of the millennium, carbonyl conjugates and radical nitroxyl polymers were the dominant players in this space (Liu, Du, et al. 2021). Some organic cathode materials rival the performance of their more conventional inorganic counterparts in terms of energy density, power density, and cycle stability. Several radicals, carbonyl compounds, conductive polymers, organosulfur compounds, and extremists were researched to construct organic Li batteries. The vast array of properties inherent to biomaterials makes them more adaptable than their more conventional counterparts. Layer structure and metal value are altered due to the intricate intercalation processes required for inorganic redox reactions (Anthony et al. 2021, McHugh et al. 2021).

#### 1.1.5.2 Quinones

Carbonyl compounds were the subject of intensive organic research in the '70s and '80s. Carbonyl groups introduced directly into multivalent anions are more effective in blocking electron processes. Because of their high electroactivity and capacity to absorb two electrons, LIB compounds are particularly interested in studying Quines and their derivatives (Cao, Wang, et al. 2021). Quinone functions as an electron carrier in photosynthesis and aerobic respiration. When dealing with small organic compounds in electrolytes, voltage, conductivity, and solubility issues arise. Unfortunately, polymeric electrodes may have difficulty isolating molecules with low electrical conductivity. Figure:7, Another option is to combine the active substance with conductive carbon and polymerize it (Xu et al. 2021). Nanolignin quinone derivatives are synthesized chemically.

Well-fabricated conjugated quinone-based polymers are capable of excellent efficiency as electrodes; this is attributed to the polymers' rapid kinetic response. These organic salts exemplify this new class of conjugated carbonyl compounds. At each stage of the cycle's progression, energy is depleted by the shifting coordination structure. Polymer-coordinating electrodes aren't the optimal solution (Taskin et al. 2021). Organic electrodes can be effective, but their manufacture often necessitates using hazardous and expensive chemicals. From biodegradable organic electrode materials like quinone, dopamine, purpurin, flavin, tannin, and nanolignin, phenolic compounds or their derivatives were transported and transformed into usable energy. Most of the structural makeup of phenolic or phenolate molds depends on redox activity (Wang, Lee et al., 2021).



**Figure:7 Quinones from Nanolignin and Nanolignin-Related Compounds. Reproduced from (Giummarella, Gioia, and Lawoko 2018) under the Creative Attribution Commons License 4.0 (CC-By 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

### 1.1.5.3 Relation between organic electrodes used in lithium-ion batteries and quinones

The cost of producing energy-intensive items like smartphones and laptops is high. Thus, there is a lot of money going into the study and development of battery materials. Although switching to renewable energy sources is challenging, it is ultimately the responsibility of the people (Zhu and Jing, 2022). The current energy storage techniques are inefficient and unreliable and must be replaced immediately with more modern alternatives.

Since their introduction, inorganic compounds have been widely used in numerous battery applications across industry and academia. Electrode materials with high capacity and controllable redox potentials can also be made using organic chemicals (Gaudin and Aubry, 2022, Shi et al., 2022). There is growing interest in using all-organic batteries, and these materials are a promising option because they can provide their capabilities without requiring transition metals. The category of carbonyl compounds called quinones has been the subject of a great deal of research. A wealth of materials is available online about quinone batteries' chemistry. Many applications can benefit from its high gravimetric capacity and solubility in battery electrolytes.

Consequences, such as decreased fat synthesis, can result from quinone molecules' insolubility in water. The larger molecules that result from these procedures make it harder to synthesize them,



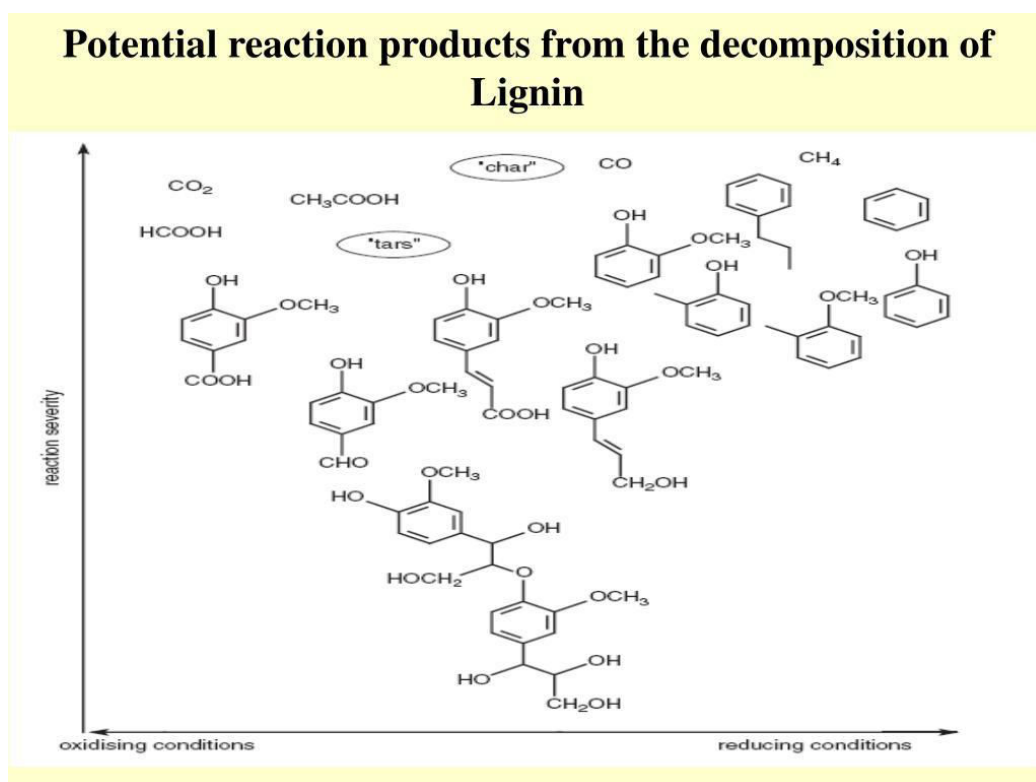
raise production costs, and prevent significant weight loss during processing (Bamgbopa et al., 2022). Novel approaches to selecting, developing, and manufacturing organic battery materials are required. Hydrogen bonding, a powerful intermolecular attraction, can make materials and electrodes more stable and less prone to breaking. H-connections (hydrogen bonds), which are ubiquitous, can be employed to build stable linkages between DNA's base pairs.

Similarly, to how organic battery components work, it appears to do the same thing. The very hygroscopic di-sodium rhodizonate was bound together with NaOH. The H-bonding between the electrode and the molecule was more critical than the molecule's solubility in keeping the electrode in its original form. Hydrogen bonding in intermolecular interactions and the behavior of neutral, sub-atomic-sized molecules (Kosgei, Kirimi, and Growth 2022).

### 1.1.6 Introduction to the nanolignin

A significant resource for the long-term production of sustainable human fuels, chemicals, and Minerals can be extracted from lignocellulosic biomass. Biomass, and wood, have long been used for these purposes (Du et al. 2021b).

Using biomass polysaccharides as a feedstock to produce fuels and chemicals has become increasingly widespread in recent decades (Wan et al. 2021). Greenhouse gas emissions might be reduced worldwide if petroleum were replaced with a sustainable, renewable feedstock. Biorefineries have mainly studied the sugars pentose and hexose sucres, fermented into ethanol using natural or synthetic yeast or bacterial strains (Du et al. 2021a). Figure 8 depicts potential reaction products resulting from the breakdown of nanolignin.

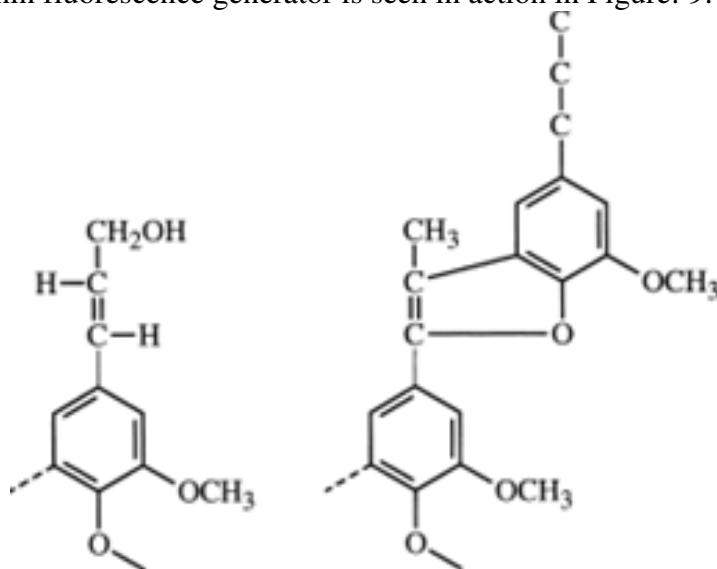


**Figure: 8 Potential Reaction Products from Nanolignin Decomposition. Reproduced from (Cherubini and Strømman 2011) with permission from Elsevier.**

There is an active investigation into using acids, bases, hot water, vapor, organic solvents, and ionic fluids as thermochemical deconstruction and pre-treatment techniques in tandem with this biomass deconstruction paradigm (Li et al., 2021). Anaerobic rumen bacteria and cellulolytic fungus have been successfully applied to commercial applications of carbohydrate enzyme active systems. However, thanks to (meta)genomics, we have been able to hone in on specific polysaccharide breakdown enzymes and suggest new lines of research into this area of study. Due to its durability and diversity, nanolignin is increasingly used to generate electrical power. Because nanolignin is so crucial to integrated biorefining, it has been the subject of extensive research and development as of late. Increased financing from governments and corporations for worldwide research institutes and consortia has led to a rise in ground-breaking discoveries and breakthroughs during the past decade. Since chemical and biological nanolignin valuation catalysis is still in its infancy, recent breakthroughs in this area are presented (Zhang et al. 2021).

**1.1.6.1 origin and nature of nanolignin**

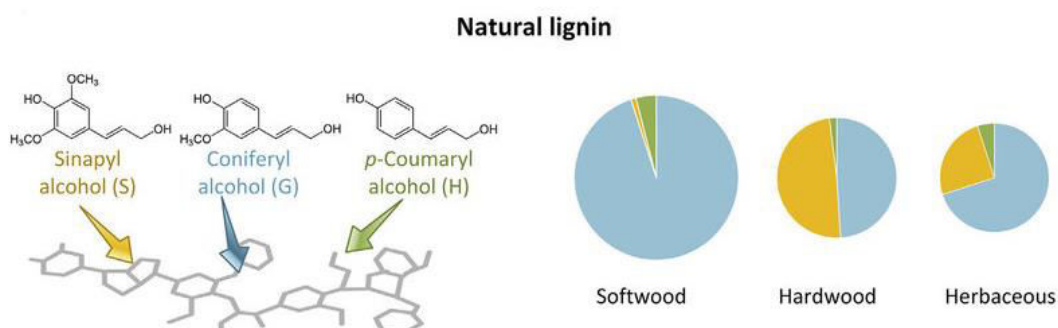
**Origin of Nanolignin** - Hydroxycinnamoyl alcohols make up the bulk of the chemical composition of this plant, which is a phenolic polymer termed nanolignin. Earth's terrestrial plant life can only succeed if the technology to produce nanolignin is refined (Makuza et al. 2021). Nanolignin in the cell walls of tracheophytes allows the water-conducting tracheal components to survive the negative pressure induced by transpiration. The development of land plants has resulted in several modifications to the biosynthesis monomer, probable precursors of nanolignin polymers, polymerization, and the regulatory mechanism for nanolignin biosynthesis (Lyu et al. 2021). The nanolignin fluorescence generator is seen in action in Figure: 9.



**Figure:9 Origin of Nanolignin Fluorescence. Reproduced from (Albinsson et al. 1999) with permission from Elsevier.**

**Nature of Nanolignin** - All vascular plants, including algae and fungi, have nanolignin in their intercellular gaps, cell walls, and other cellular components. The metabolic process of

photosynthesis increases competition for sunlight by regulating water flow in living organisms (primarily by fortifying cell walls to prevent collapse, but also, to a lesser extent, through liquid transfer) (Cai et al. 2021). It appears to be highly stable against breakdown due to a large concentration of H bonds and powerful chemical connections. Several links between wood sugars and this (hemicelluloses). Groundwood paper contains nanolignin, which is explosive and problematic in printed materials, contradicting the notion of durability and usefulness. During the pulping and bleaching operations, nanolignin is removed from the fibers (Huang et al. 2021). The chemical composition and monetary value of nanolignin are displayed in Figure 10.



**Figure:10 Composition and Valorization of Natural Nanolignin: Natural occurrence and composition of nanolignin. Reproduced from (Becker and Wittmann 2019) with permission from Elsevier.**

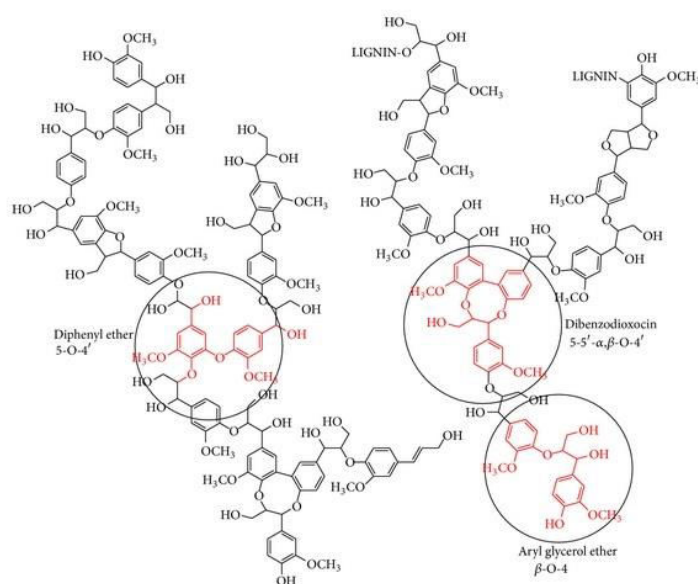
Remember that nanolignin is a composite substance with many different parts. These polymers contain phenylpropane, a ring of three carbons in the benzene family that makes them amorphous and complicated in three dimensions. Scientists haven't settled on a single definition because naturally occurring ones have molecular weights of 15,000 or higher (Slattery, Dunn, and Kendall 2021). Wood, paper, and nanolignin-containing board all contain carboxylic acids, and their degradation continues to yield acids. As the papermaking process progresses, nanolignin and other delicate cellulose fiber components are eliminated. Natural nanolignins (protonanolignins) have been found in all types of plants (Ryu et al. 2021, Masias, Marcicki, and Paxton 2021, Xie and Lu 2020). There are specialized pulping processes where substantial quantities of nanolignin can be removed, and then other methods can be used to separate the fibers from the band. When nanolignin is removed, the paper's binding ability and strength improve (Wu, Maier, and Yu, 2020). The removal of nanolignin can be accomplished mechanically, semi-chemically, or chemically, and all three methods can be used to make high-yield pulp. Preventing fiber separation and nanolignin buildup by heating the paper pulp is a desirable goal. nanolignin's higher resistance to the pulping chemicals than celluloses cause frequent process disruptions (Li, Sun, et al. 2020).

There has been a shift from using dioxin-based bleaches to chlorine-based replacements at many bleaching operations (Li, Liu, et al., 2020). Some pulp mills choose expanded alignment over bleaching when nanolignin removal falls short. Smook postulates that reducing kappa values and increasing total washing effects can be accomplished by applying a two-stage, partially counter-current cooking sequence to Kraft pulping in a continuous digester. Sulfite pulping generates black kraft liquor and liginosulfonates, which are highly sought. Dust control on roadways, fabric

and brick coloring, animal feed, paper and cardboard production, and concrete admixture are various uses for this versatile ingredient (Shan, Li, and Pang 2020b).

### 1.1.6.3 Chemical structures of nanolignin

Freudenberg and his colleagues investigated the origins of nanolignin between 1940 and 1970. Some compounds that must be created before the final product may be obtained are coniferyl, sinapyl alcohol, and P-coumaryl (Shan, Li, and Pang 2020a). When monomers combine in nature, a non-linear chain reaction occurs, giving rise to nanolignin (p-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol) (Abraham 2020). As far as natural polymers go, nanolignin has some of the most complex chemical properties.



**Figure 11 Chemical Structure of Nanolignin. Reproduced from (Wang, Tucker, and Ji 2013) under the terms of the Creative Attribution Commons License 4.0 (CC-By 4.0).**

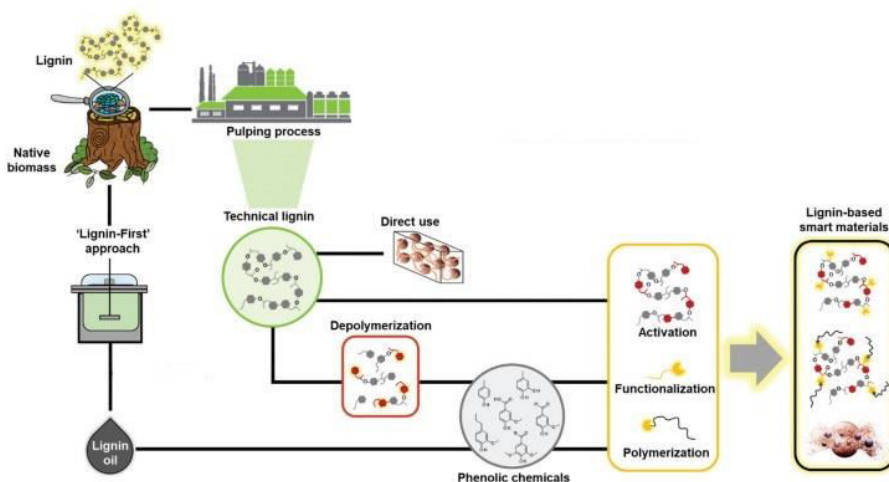
<http://creativecommons.org/licenses/by/4.0/>.

Compared to cellulose and protein, nanolignin and its degradation products exhibit lower optical activity because they include several asymmetric centers instead of a single one (Feng et al., 2020). Standard disassembly methods are ineffective because of the dense network of carbon-carbon bonds present in each structural unit. Little is known about the chemistry of plant-made nanolignin. Since Björkman-processed milled wood nanolignin (MWL) is structurally comparable to the original nanolignin and continues to dominate the nanolignin market, it is used by most wood-chemical specialists (Leskinen et al. 2017). Due to its complex regulation mechanism, guaiacum nanolignin is the dominant form of lignin in many wood species. Syringa nanolignin significantly affects the development of numerous plant species, and this is true even though there are many distinct wood quality stresses. Nanolignin's atomic structure is depicted in Figure: 11.

### 1.1.6.4 Nanolignin-based smart materials

With the ability to be synthesized as polymers, oligomers, dimers, and monomers, nanolignin has several potential applications ranging from plant biomass to intelligent materials. The chemical

structure of nanolignin is critically essential in plant evolutionary and differentiative processes, and recent advancements in analytical tools have made examining chemical bonds a lot less scary (Grey and Hall, 2020). Depolymerized and soluble in solution, nanolignin results from the thermochemical treatment of cellulosic fibers. This process involves the alteration of condensation bonds at reactive locations (Hong et al. 2021). Although nanolignin-based polymers and composites are readily available, there is still a need to explore very pure or split nanolignins for application in demanding domains like biomedical engineering. Increasing polymer performance with technological nanolignins is a fast and efficient method (Chen, Wu, Su, et al. 2021). Nanolignin is an antioxidant that makes plastics more durable by reflecting the sun's rays, and flame retardants are helpful since they prevent fires from starting in the first place. Nanolignin's limited utility is because it is incompatible with most polymers and has a low melting point (Jiao et al. 2021). Nanolignin can be modified chemically to improve polymer properties. Due to a chemical alteration that enables the grafting of ordinary monomers or polymers onto nanolignin macromonomers, producing nanolignin-based functional polymers is a relatively straightforward process. Many monomers (hydroxyl groups, alkyl halide esters, etc.) that react with the nanolignin core result in polymer chains that run perpendicular to the core (Tian et al. 2021). Shown in Figure: 12 is a cutting-edge material that makes use of nanolignin.



**Figure 12 Nanolignin-based intelligent materials. Reproduced from (Moreno and Sipponen 2020) with permission from Elsevier.**

The grafted chains' lengths can be adjusted to suit each user's requirements, and in-depth characterization can be carried out in advance using this technology (Zhao et al. 2021). Precursor-setting is essential in synthesizing many different materials, as seen by the enormous diversity of technical nanolignins. The biggest obstacle to making aromatic compounds is the defiant nanolignin created during the pulping process. Aromatic monomers and dimers enhance Nanolignin's oil fraction with lower molecular weights (Ding et al. 2021, Cao, Yang, et al. 2021).

#### **1.1.6.5 Isolation Techniques**

Nanolignin's physicochemical properties and effects on plants are significantly modified by the insulation techniques (also known as pre-treatment operations) used to create it (e.g., molecular weight distribution, solubility, and thermal properties). The production of technical nanolignin, which is chemically separate from soluble nanolignin, requires the use of a solution developed

for the latter (Kang et al. 2021, Liu, Han, et al. 2021). New functional groups are introduced into the lignin structure, and lines, especially ether linkages, are partially disrupted, all steps necessary to create nanolignin. After being isolated, nanolignin's system undergoes a series of additional steps that further complicate its structure (such as intramolecular condensation).

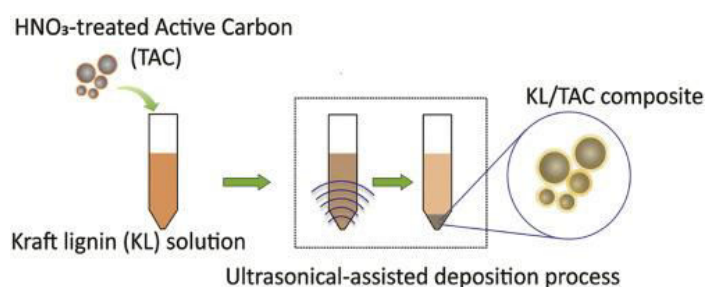
### **1.3 Kraft nanolignin-carbon composite for sustainable cathode materials**

#### **1.3.1 Objective and motivation**

It has been demonstrated that phenolic groups are essential to forming nanolignin. Demethylation of aromatic methoxy groups generates quinone/hydroquinone electroactive pairs that can be used to manufacture electrochemical nanolignin. Organic redox systems containing quinone/hydroquinone pairs are often present in naturally occurring phenolic compounds (e.g., humic acids, flavonoids, tannic acid, and dopamine). Two-step proton redox processes ( $2e^-/2H^+$ ), dependent on pH and the Nernstian reduction potential, are set in motion by quinone/hydroquinone pairs in acidic aqueous solutions (Xiao et al. 2021).

However, there are limitations to employing conductive polymer additives. Polymer degradation occurs, reducing cycling stability (Yang et al. 2021a). Doping with small and mobile counter-anions can boost the conductivity of several polymers, including PPy; however, this is usually accompanied by a drop in rate capability, an increase in self-discharge, and a decrease in storage density. Polymerization could negatively impact the environment if toxic organic solvents or strong acids are used. As an alternative, the electrosynthesis method allows nanolignin to be seamlessly incorporated into a surface polymer film (Yuan et al. 2021).

Finally, studying composites is complex, and figuring out the mass ratio of the components required to thwart storage growth is even more so. For nanolignin electrodes, the addition of carbon compounds improves their electronic conductivity. With the use of carbon composites incorporating nanolignin, the ratio of the individual components in the final hybrid material can be altered (Liang et al. 2021). Figure:13 depicts the incorporation of Kraft nanolignin onto Active Carbon.



**Figure:13 Kraft Nanolignin modified Active Carbon. Reproduced from (Zhou et al. 2019) with permission from Elsevier.**

Carbon-based composite electrodes have been demonstrated to have significant chemical stabilities, allowing them to function in high electrolyte concentrations while mitigating the effect of ion transport parameters. The difference between conductive and nonconductive polymers best illustrates this point. Nanolignin has been developed for battery-like charge storage, and several different conductor carbon materials have been employed (Tan et al. 2021).

Multiple redox peaks corresponding to this faradic process have been identified through cyclic voltammetry; however, some have been mislabelled as pseudo-capacity. Utilizing energy storage technology like CNTs and RGOs allows for long-term, reliable, and environmentally friendly solutions to health and safety issues (Wu et al. 2021). By incorporating renewable and sustainable materials, the cost of conductive carbon was lowered. Although lignosulfonate is in short supply, it has been studied by several groups as a potential electrode material (Incer-Valverde et al., 2021).

### **1.3.2 Electrochemistry of Kraft nanolignin-carbon composites**

The ratio of carbon to nanolignin could be adjusted, and both 1:1 and lower ratios were available to the researchers. Prototype battery ratios are identical to mass-produced redox composite batteries (Gao et al. 2021). A clear signature of each material was present in each porous composite sample. Because nanolignin and carbon particles were incorporated extensively into the electrode fabrication process, they were impossible to detect. Put another way; the capacity is independent of the materials used to make it. People with CH have a wide range of skill levels. Adding even a bit of nanolignin to a system can yield considerable gains in efficiency (Feng et al. 2021, Alirahmi, Razmi, and Arabkoohsar 2021). The load transmitted to the front carbon atoms would be minimal with a nanolignin layer of this thickness. Electrochemically active nanolignin and carbon surfaces are used in the storage device, as shown by cyclic voltammograms, which also reveal a high concentration of faradaic and non-faradaic charges (Razmi et al. 2021).

Higher electrolyte concentrations increase the total capacity of the composite, demonstrating the proton's significance. Electrolyte concentration variations often have little effect on non-faradaic charge storage (Balali and Stegen, 2021). These data show that the EDL loading capacity of conductive carbon coated with nanolignin is unaffected. Despite incorporating more nanolignin into the mix, the CL-based composite samples showed the same capacity contribution to faradaic loading storage. Because of its propensity to produce a thick coating that inhibits the flow of electrolyte ions, nanolignin is likely present in both solutions (Beck et al. 2021).

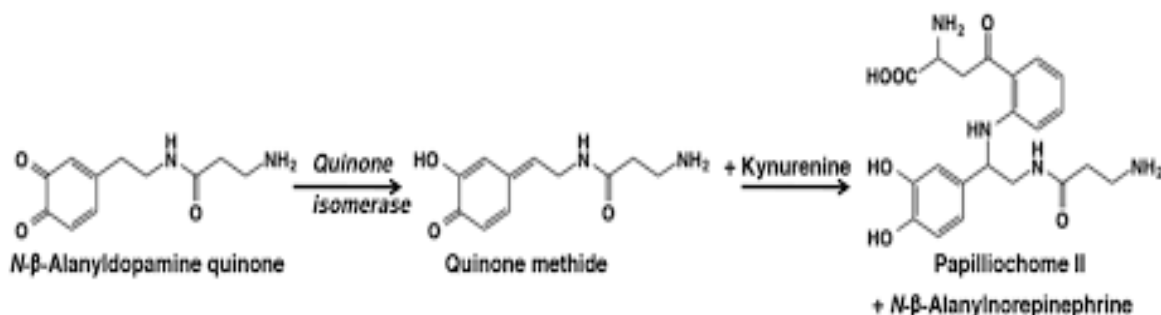
The compost electrode outperforms the standard graphite collector in terms of electrical conductivity and electron transport efficiency due to its higher carbon content (Lin et al. 2021). Increased surface area and ionic conductivity result from nanolignin's ability to form a thin layer of conductive carbon on a large surface. The redox-active moisture in the nanolignin reacts with the lignin's electrolyte ions, causing this effect (Rahbari, Arabkoohsar, and Alrobaian 2021). The composite paves the way for efficient ion transport at the electrode-electrolyte interface and a load-transfer mechanism with low resistance by forming a thicker layer of nanolignin on the outer surface of the conduction carbon. Warburg resistance represents a material's inability to allow charge to flow at high or medium frequencies, whereas the periphery of the diagram demonstrates its low-frequency diffusion properties (Giarola et al. 2021).

### **1.4 Modification of Kraft nanolignin with dialdehyde crosslinkers for cathode materials**

Nanolignin has been studied extensively due to its adhesive qualities, but the material also has numerous other possible industrial applications (Ye et al. 2021). In plants, it is a glue that prevents the cellulose fibers from unravelling. Scientists investigated whether or not using nanolignin in the production of polyurethane, phenol-formaldehyde (PF), and epoxy resins

would reduce their respective health risks. During battery electrode production, an electrode binder is employed to glue the active materials and carbon conductive additive to the current collector (Wei et al. 2021). Binders hold current collectors in place while the electrode is being assembled. What keeps these things together significantly affects their mechanical quality, cycle stability, safety, and environmental impact (Deshpande et al., 2021).

Electrochemical stability and binding capacity are barely affected by connecting electrode parts, even if they all have sufficient surface activities. Though they defluorinated well, they produce harmful waste products. They are difficult to recycle and expensive to dispose of (Meng et al., 2021). Hydrophobic electrodes have high internal resistance because they isolate and block ions, preventing the electrodes from interacting with the electrolyte. The CMC is preferable because it has no adverse effects on the environment and is inexpensive. Water-soluble CMC electrodes are equally effective as those manufactured with a fluorinated binder, but they can't be used in wet environments (Yang et al. 2020). Nanolignin, when used as an electrical component and with its critical features exploited, may improve the performance of composite electrodes. Nanolignin's storage capacity is diminished because of its low molecular weight, which is quickly hydrolyzed during charging and discharging (Lawoko, Berglund, and Johansson 2021). Theoretically, Figure 14 shows that the carbonyl groups in quinones are conjugated.



**Figure: 14 Conjugated carbonyl groups in quinones and their theory. Reproduced from (Ito, Sugumaran, and Wakamatsu 2020) under the Creative Attribution Commons License 4.0 (CC-By 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

In acid and alkaline circumstances, condensational reactions occur in the most reactive nanolignin structure, phenolic and noncyclic ether groups. The phenolic hydroxyl groups, another significant functional group, have also been interconnected with diacids, isocyanate, and epichlorohydrin (Hait et al. 2021). The cross-linking of electron-rich locations on the nanolignin aromatic rings also causes polymerization. The method of cross-linking nanolignin using cross-linking chemicals such as formaldehyde is Methylation, also known as hydroxymethylation. During the Methylation process, especially in the ortho positions of phenolic groups, reactive areas of nanolignin are joined to hydroxyl methyl groups. The composite electrodes were adsorbed by low molecular nanolignin across a broad carbon surface and heat cross-linking with glyoxal (Tang et al. 2020).

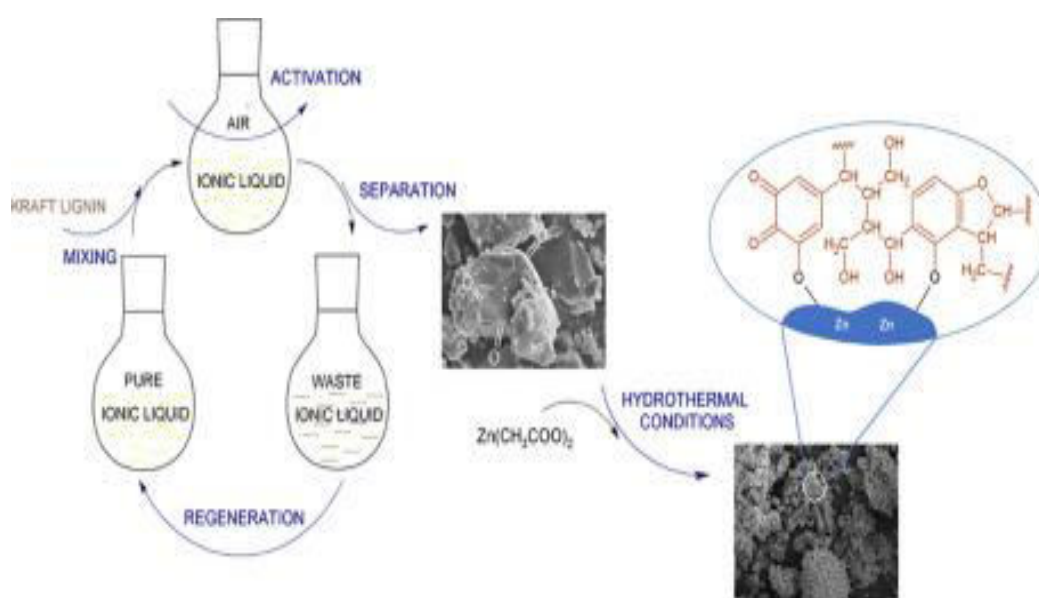
### **1.5 Oxidation of Kraft nanolignin for cathode materials**

Nanolignin mainly consists of non-electrical phenolic functional groups. Nanolignin's long-term stability can be attributed to the existence of hydroquinone and quinone and the dominance of methyl phenyl ethers and diphenyl ethers. The free methoxy or dimethoxy phenol group can be



oxidized electrochemically to produce hydroquinone, which, after being demethylated, can create quinone. However, the potential of nanolignin for load storage is limited by the availability of its methoxy groups to electrolytes (Tran et al. 2017).

It is not surprising that both exhibit electroactivity because they are a redox reaction pair. Increasing the nanolignin's quinone/hydroquinone ratio may be essential to increase its electrochemical activity for LIB uses (Du et al. 2019b). LIB employs a straightforward, non-chemical oxidation process. In recent years, improved methods of oxidizing chemical nanolignin have been discovered. It is standard practise to use oxidation to create compounds with strong odors (Garca Negrón et al., 2017). Vanillin, vanillic acid, syringic acid, and many more chemicals are excellent examples. Figure 15 shows an ionic liquid can be used to control the oxidation of kraft nanolignin at low temperatures. Different types of catalysts have been used in various technologies, such as organ metalized catalytic oxidation, biomimetic oxidation methods, and enzyme-based oxidation strategies. Many gentler oxidizers were used to protect the ring and restrict oxidation to specific groups. Fremy's salt and other oxidizing chemicals have essential drawbacks, despite their many benefits. By rupturing the circle, one can obtain free acid groups from oxidized nanolignin, for instance (Geng et al. 2017).



**Figure: 15 Controlled oxidations of kraft nanolignin in mild conditions using ionic liquid. Reproduced from (Klapiszewski et al. 2019) with permission from Elsevier. Copyright (2019).**

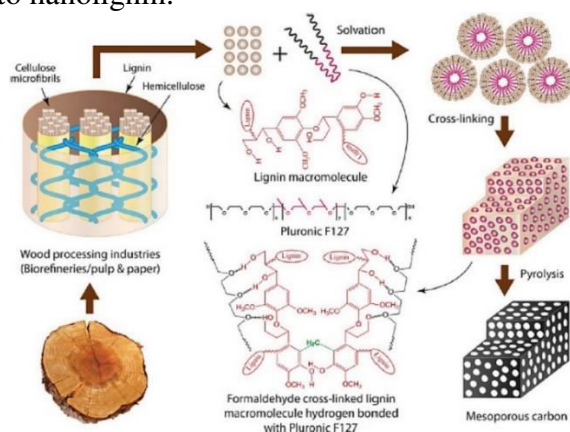
One of the more efficient approaches to producing quinone derivatives is the oxidative processing of hydroquinone dimethyl ethers, especially demethylation. Methyl ethers of phenol derivatives are highly stable over many reaction conditions and require an additional demethylated process. CAN be the most effective reagent in synthesizing quinones across the board for oxidizing processes, including the efficient oxidative demethylation of dimethoxybenzene (Mukhopadhyay et al., 2018). Information from the nanolignin model chemical was used to predict the oxidation products. The quality of oxidized nanolignin materials has been determined by various solid-state tests, such as FTIR and UV Vis. Both

modified and unmodified composite electrodes were developed for lithium-ion batteries (Chaleawert-upon and Liedel 2017).

## **1.6 Other advanced carbon materials from nanolignin for electrodes**

### **1.6.1 Carbon Fibres**

Thanks to their outstanding qualities, carbon fibers are becoming increasingly popular in the construction sector. Some examples of these qualities include exceptional tensile strength, low weight, excellent flexibility, and low thermal expansion. Carbon fibers are a vital component of modern composites, which have several uses in various industries, including aviation, automobile manufacturing, bridge construction, and wind power generation (Zeng et al. 2019). The beneficial characteristics of carbon fibers are often perpendicular to the fiber's long axis due to the orientation of the graphite layers. The basic materials used to create carbon fiber are the source of many beneficial qualities. Due to the high cost of oil, polyacrylonitrile (PAN) and mesophase fiber are the main precursors used to create modern carbon fiber. It is important to note that administrative expenses are included in the overall price (Chaleawertumpon et al. 2017). Nanolignin is already being processed commercially because of its high demand as a low-cost carbon fiber precursory material. For nearly fifty years, scientists have investigated nanolignin as a possible antecedent of carbon fiber. Figure 16 illustrates the transformation of carbon-based materials into nanolignin.



**Figure: 16 Nanolignin-Derived Advanced Carbon Materials. Reproduced from (Chatterjee and Saito 2015), with John Wiley and Son's permission.**

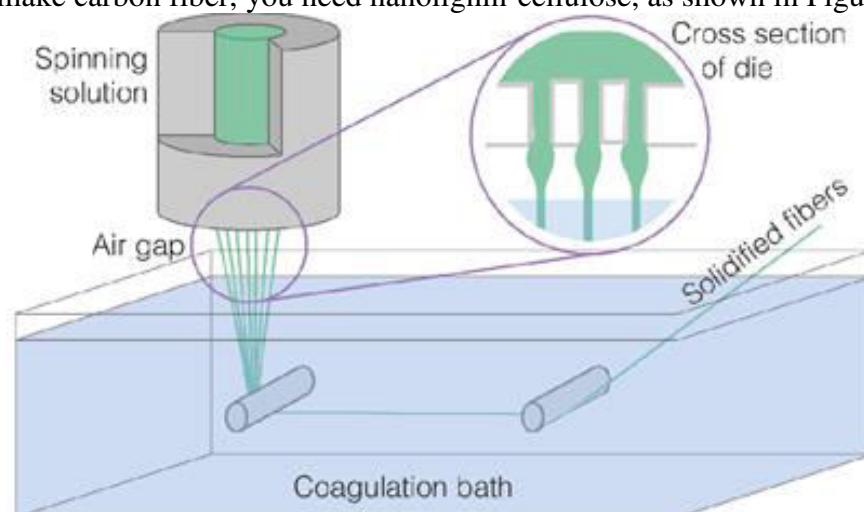
Sources of nanolignin carbon fibers include: It is worth noting that nanolignin carbon fibers can be created either by extruding nanolignin alone or by combining nanolignin with co-extruding chemicals. Pure nanolignin must go through three processes—carbonized spinning, therapeutic stabilization, and carbonization—before it can be spun into carbon fiber (Nirmale, Kale, and Varma 2017). Later, they go through graphitization to become structural fibers or activation for different applications. The procedure creates a strong and lightweight material using nanolignin and high-quality carbon fiber (both on the scale of atoms in thickness) (Kumar, Hietala, and Oksman 2019).

### **1.6.2 Spinning**

During the spinning process, the final carbon fiber's diameter, form, and strength are all determined. Fusion spinning, solvent-assisted spinning, and wet spinning are the most prevalent

techniques for transforming nanolignin into precursor fibers (Dou et al. 2017). Nano-nanolignin fiber-based products have recently been produced using electrospinning and melt-blown processes. If the material has a low melting viscosity, you can spin it at a low temperature. In either its cross-linked or dissolved forms, nanolignin's melting point should be lower than that of the corresponding lignin. After the nanolignin melt has been extruded into the dust, it is spun over a spinneret or wound onto a spinning winding machine. Sometimes a stream of rotating air is used to propel the nanolignin melt. The precursor fiber can be protected from oxidation by spinnable inert nanolignin (Saito et al. 2019).

Solvent-assisted spinning allows nanolignin to be dissolved in a suitable solvent and subsequently incorporated into fibers. The fiber is then dried in a hot or cold dish, depending on which method was used to remove the nanolignin solution (Xi, Yang, et al. 2019). When the solvent is completely gone, the fiber is dry. Voltage-driven solutions, such as a rotator, are commonly employed to alter nanolignin discharged from a spinneret on an earthed collection. To begin, we develop a solvent that is thin enough to be spun but strong enough to dissolve nanolignin (Aro and Fatehi 2017). Given the potential difference between the spinneret and the collector, the nanolignin solution will become electrically charged as it travels through the spinneret. To make carbon fiber, you need nanolignin-cellulose, as shown in Figure 17.



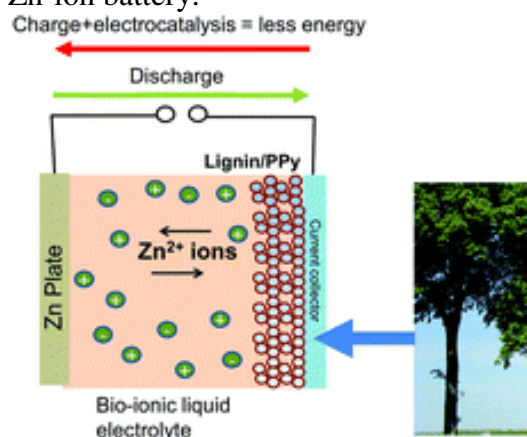
**Figure: 17 spinning of nanolignin-cellulose carbon fiber. Reproduced from (Wang et al. 2019) under the terms of the Creative Attribution Commons License 4.0 (CC-BY 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

When the surface load repulsions of the nanolignin solution surpass the surface voltage of the liquid drop, the answer will begin to flow out of the spinnerets. After the solvents quickly evaporate, the nanolignin mixture is spun into fibers, and the resulting fibers are collected in an earthed collector. Nanolignin fibers, which several different spinning techniques can make, are far too thin to be of any practical use. These filaments are transformed into carbon fibers following a carbonization and thermal stabilization process (Park et al. 2019).

## **1.7 Composites from nanolignin for electrodes**

### **1.7.1 Carbon/nanolignin composites**

Improvements in load storage capacity at the electrode can be attributed to the addition of the Q/QH<sub>2</sub> redox couple, which was synthesized from phenol nanolignin groups. Composites comprising nanolignin and carbon have promising properties (Hanuová 2021). The increased redox reactivity of the surface quinone molecules causes a pseudo-capacitance with a robust reversible component. This behavior is due to the incorporation of reduced graphene oxides (RGOs) into a conductive matrix containing nanolignin nanocrystals (Navarro-Suarez et al. 2018). Furthermore, electroactive nanolignin has been used to create lignosulfonate/single-wall carbon nanotube EC hydrogels with HNO<sub>3</sub> pressure sensitivity. High-performance, non-metallic, and flexible supercapacitors were developed utilizing lignosulfonate functional graphic composites, allowing them to compete with transition metal-based ECs in terms of energy and power density. With the help of lignosulfonate, single-wall carbon nanotubes, and electroactive nanolignin, pressure from HNO<sub>3</sub> can trigger a reaction in the hydrogel (Zhang and Shen 2019). Nanolignin composite electrode and bio-ionic liquid electrolyte are shown in Figure:18, illustrating a biodegradable Zn-ion battery.



**Figure:18 Biodegradable Zn-ion battery with a nanolignin composite electrode and bio-ionic liquid-based electrolyte. Reproduced from (Lahiri et al. 2021) under the terms of the Creative Attribution Commons License 4.0 (CC-By 4.0). <http://creativecommons.org/licenses/by/4.0/>.**

### **1.7.2 Nanolignin-derived carbon / active material composites**

Considering the advantages of active, highly theoretical materials paired with high conductivity carbon and excellent cycling stability, research on materials that could become feasible electrodes has essentially been exhausted. Extensive research has been done on carbon-active material composites (Beisl, Friedl, and Miltner 2017). As a result, research into composites containing carbon generated from nanolignin and other functional components has increased. Because of their high surface area and porous nature, nanolignin-based NiO-carbon composites are suited for usage as the electron storage medium in supercapacitors. Meanwhile, lamella graphene carbon/Fe<sub>3</sub>O<sub>4</sub> nanocomposite products with homogeneous microstructures have been developed by growing graphene sheets using nanolignin (Wang, Pu et al. 2019).

### **1.7.3 Nanolignin / active materials and nanolignin / polymer composites**

Since its redox moieties Q/QH<sub>2</sub> contribute to its electrochemical activity, nanolignin has attracted a lot of studies (Liu et al. 2018). Energy storage using nanolignin polymer composites has also shown promise. It has been demonstrated that a ternary composite system consisting of

polypyrrole, a nanolignin derivative, and phosphomolybdic acid has a greater load-storage capacity than polypyrrole alone (Demir, Tessema et al. 2018). The use of lignosulfonate-doped alkaline, polypyrrole electrodes, nanolignin/PEDOT polymer electrodes, polyaniline, etc., all verified these results. Although nanolignin composites may one day be employed in this capacity, research into electrochemical energy storage has continued for some time. Further investigation into structure-performance correlations in composites is needed before its supervised mass production is possible (Cui et al., 2019).

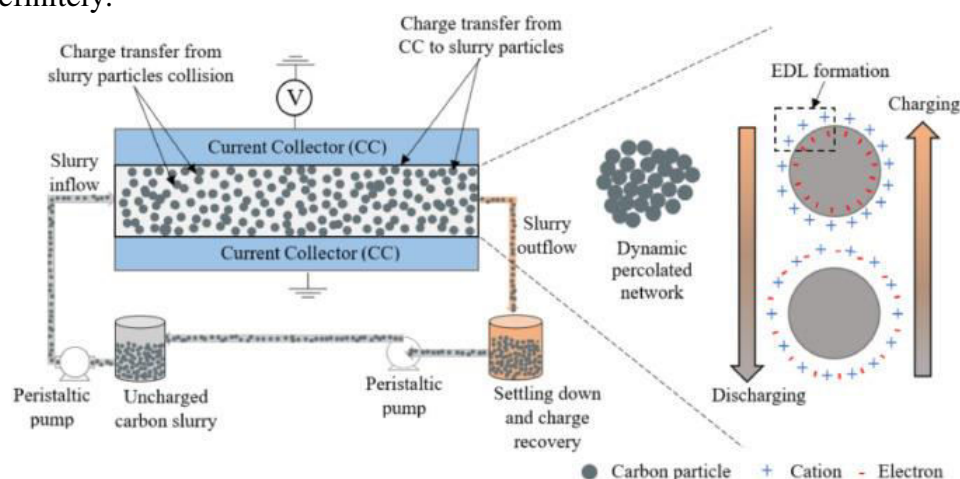
## 1.8 Nanolignin-based materials without carbonization as binders and separators

### 1.8.1 Nanolignin-based binder without carbonization

Polymers not affected by electrochemical reactions are commonly used to make binders in electrochemical energy systems (Du et al. 2019a). Binders are essential because they contribute to the cycle stability of the electrode, allowing for the development of lightweight, high-level binding devices that are mechanically flexible and electrochemically stable. Problems with conventional LIBs include high production costs, high fire risks, and the need for volatile processing (e.g., polyvinylidene difluoride (PVDF) as a binder). Compared to PVC binders, LINO binders are more environmentally friendly and cost-effective. To lessen the impact on volume, the Si anode was cycled with a water-soluble binder called nanolignin (Liao et al. 2020).

### 1.8.2 Nanolignin-based Separator without Carbonization

To minimize the possibility of electrical shorts and to maximize the availability of effective ion transfer channels, it is essential to physically separate the positive and negative electrodes in electrochemical energy storage devices. Commercial performance, production costs, and ionic transfer kinetics for polyolefin-based microporous separators remain problematic despite developments. Due to their increased thermal stability and ionic conductivity, sustainable green feedstock-based alternatives have gained renewed interest in recent years as separators (Peng et al. 2018). As shown in Figure: 19, nanolignin can be used to create electrodes that can store energy indefinitely.



**Figure:19 Nanolignin-based electrodes for energy storage. It was reproduced by (Mourshed et al. 2021) with permission from Elsevier.**

## 1.9 Conclusions

This paper reviews the topic of energy storage before delving into the existing literature on the subject, emphasizing the results of numerous investigations on using nanolignin in energy storage devices. The quest for and study of organic electrodes for batteries is currently a prominent focus of scientific inquiry. A kraft nanolignin carbon composite for sustainable cathode materials with objective and motivation follows this section, as does a discussion of the origin and nature of lignin, its chemical structure, nanolignin-based innovative materials, isolation methods (including kraft and sulfite), and a kraft nanolignin carbon composite for sustainable cathode materials with objective and motivation. Materials for the cathode, such as oxidized kraft nanolignin, are discussed in this section. Nanolignin films, carbon fibers, microstructured carbon fiber mats, activated carbons, templated carbons, activated carbon fibers, and carbon fibers made from nanolignin are all broken down and examined in detail. Nanolignin film, a kind of carbon fiber, can be made from this substance (such as carbon fibers, spinning, thermostabilization, and carbonization). This paper concentrates on nanolignin and its applications, including nanolignin composites, nanolignin-generated carbon/active material, and nanolignin/polymer composites. This paper explores the possibility of using nanolignin-based materials as carbon-free binders and separators. Carbon monoxide-free binders and separators are just one example.

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