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Review

Recent Developments of Biopolymer-Based Composite Membranes as Sustainable Separators for Lithium-Ion Batteries

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ABSTRACT. The increasing adoption of electric vehicles and large-scale energy storage systems has intensified demand for high-performance and sustainable lithium-ion battery separators. This review summarizes recent progress in biopolymer-based separators as environmentally friendly alternatives to conventional polypropylene/polyethylene membranes. Fourteen research articles published between 2020 and 2026 are analyzed, focusing on biopolymers such as bacterial cellulose, chitosan, lignin, alginate, silk fibroin, and poly(L-lactic acid). The discussion covers material classification, membrane fabrication techniques, and modification strategies, including composite formation and crosslinking. Overall, the reviewed studies indicate that biopolymer-based separators generally exhibit improved electrolyte wettability, enhanced ionic conductivity, and adequate thermal stability, leading to competitive electrochemical performance and good cycling stability. In addition to performance benefits, the use of biomass-derived materials and water-based fabrication processes offers sustainability and potential biodegradability advantages. This review highlights current research trends and outlines key challenges related to testing standardization and manufacturing scalability for future lithium-ion battery applications.

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INTRODUCTION

Energy needs continue to increase along with technological advances and human activities in various sectors, including transportation, industry, and household needs. This condition underscores the importance of energy storage devices, especially as renewable energy sources and electric vehicles become more widespread. Among various energy storage technologies, lithium-ion batteries (LIBs) remain the primary choice because they offer high energy efficiency and density, as well as a relatively long service life. According to an International Energy Agency (IEA) report, global demand for LIBs in 2030 is projected to exceed 3,500 GWh. This may occur alongside the increasing use of electric vehicles and large-scale energy storage devices (IEA, 2023).

The main structure of the LIB consists of a negative electrode (anode), a positive electrode (cathode), and a porous separator (separator). The separator prevents direct contact between the electrodes (anodes and cathodes) while allowing lithium ions to move during charging and discharging. The separator must have good physical and chemical properties to work optimally, including appropriate porosity, controlled pore size, uniform thickness, ease of electrolyte absorption, and adequate thermal and mechanical stability (Barbosa *et al.*, 2020). Polyolefin-

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based separator industries, such as polyethylene (PE) and polypropylene (PP), are still commonly used. However, this material has several limitations, including hydrophobicity that makes it difficult to absorb electrolytes, limited thermal resistance, and the potential for shrinkage at high temperatures, which can trigger battery malfunctions (Yang *et al.*, 2022). In addition, its nature, which does not decompose naturally, also causes environmental problems.

These limitations encourage the use of biopolymers as environmentally friendly alternative materials. Biopolymers such as chitosan, nanocrystalline cellulose (CNC), alginate, and starch are considered more environmentally friendly, easy to renew, and well compatible with liquid and gel electrolytes (Lizundia and Kundu, 2021). However, the use of pure biopolymers still faces obstacles, such as insufficient mechanical strength and relatively low ion permeability. To address these shortcomings, biopolymer composite membranes are a growing focus of research, combining biopolymer matrices with inorganic reinforcements such as SiO_2, Al_2O_3, graphene oxide, or nanoclay to enhance separator performance.

In addition to improving technical performance, the use of composite biopolymers aligns with global demands for more sustainable technology. The use of renewable materials supports the principles of the circular economy. It can reduce the environmental impact of synthetic polymer-based battery waste. In fact, some biopolymer composite membranes developed in recent research show performance that rivals commercial polyolefin separators in terms of ion conductivity, thermal stability, and service life (Liu and Chuan, 2021). The development of biopolymer composite membranes not only offers technical solutions but also represents a strategic step toward accelerating the transition to cleaner, safer energy storage technologies.

Based on this description, this article aims to review the latest developments in the design, fabrication, and characteristics of biopolymer composite membranes as LIB separators, by reviewing the different types of biopolymers and reinforcing materials used and analyzing the contribution of each to membrane structure, ion conductivity, thermal stability, and electrochemical performance. In contrast to the previous reviews that generally focused on material classification, this article adopts an integrated analytical approach that links the chemical structure of biopolymers, fabrication methods, and electrochemical performance in a single complete study, so this article is expected to provide scientific insights and strategic direction for the development of efficient, safe, and sustainable biopolymer-based separators for future LIB applications.

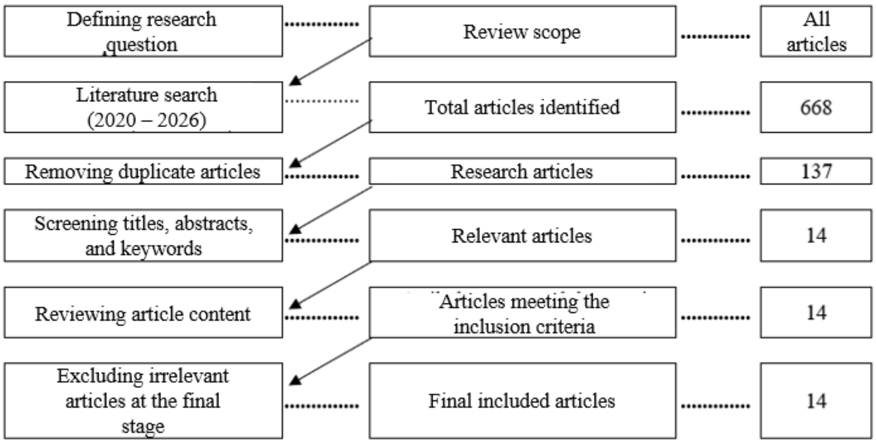

Figure 1. Systematic mapping study process.

The approach used in the preparation of this review article is a systematic mapping study (SMS) (Figure 1) to summarize the existing evidence on the topic being studied, identify gaps in the research, provide suggestions for future research, and provide background and position new research activities (Petersen *et al.*, 2015). This approach was used to map and analyze the latest developments in biopolymer-based LIB separators in a structured and comprehensive manner. The literature search for this review was conducted in the ScienceDirect database using the keywords "membrane separator", "lithium-ion batteries", "biopolymer", and "composite", limited to 2020 – 2026 to ensure relevance to the latest research developments. Only research articles are included for review material, so that the analysis is based on experimental findings that can be accounted for. Inclusion criteria include studies that discuss biopolymer-based separators or biopolymer composites, describe membrane fabrication methods, and report key performance parameters such as ionic conductivity, thermal stability, and/or electrochemical performance in batteries. The exclusion criteria include duplicate articles, review articles, non-English publications, and studies that do not present separator performance data in a relevant manner. The potential for literature bias is minimized through publication-year restrictions and gradual screening at the title, abstract, and article levels. The selected articles were then mapped and analyzed to identify research trends, linkages between chemical structures, fabrication methods, and electrochemical performance, as well as research gaps in the development of lithium-ion differential biopolymer separators.

BIOPOLYMER SEPARATOR IN LITHIUM ION BATTERY

A separator is a porous polymer membrane placed between the positive and negative electrodes in a cell that prevents direct physical and electrical contact between the electrodes while still allowing ion transfer (Figure 2). Although the separator is an inactive element in a battery, the characteristics of the separator, such as porosity, pore size, mechanical strength, and thermal stability, greatly affect ion transfer, cycle life, performance, and battery safety. Therefore, separators are one of the key components in LIB (Lingappan *et al*., 2023).

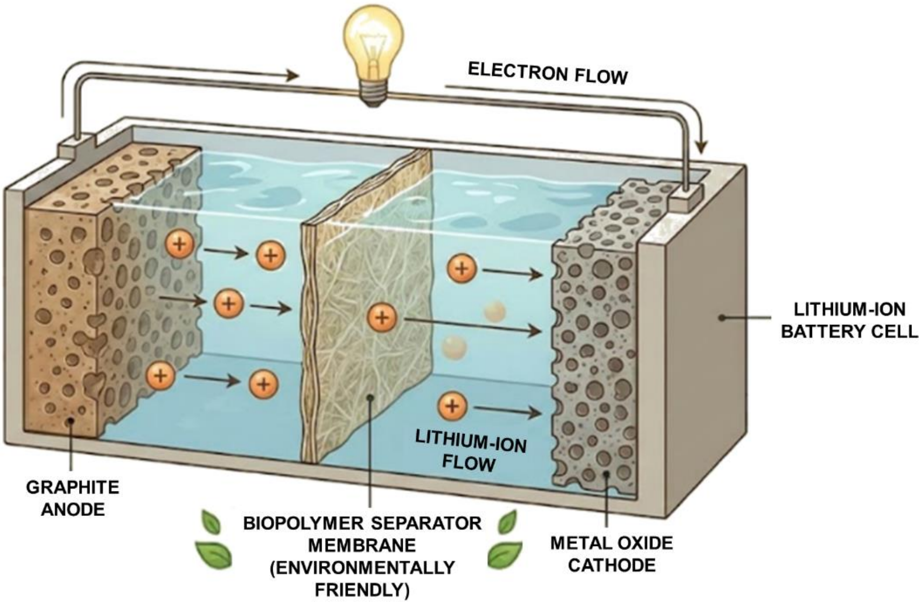

Figure 2. Biopolymer separator in a lithium-ion battery.

Modern battery separators are increasingly switching to biopolymers for environmental and performance benefits, as well as improved battery safety. Compared to conventional polyolefin-based separators that are difficult to decompose and contribute to long-term waste, biopolymers such as cellulose are *biodegradable* and therefore more environmentally friendly. In terms of safety, biopolymers generally exhibit better thermal stability, thereby reducing the risk of thermal shrinkage, internal short circuits, and *overheating* in high-capacity LIBs. In addition, the chemical and mechanical properties of biopolymers can be engineered to improve *wettability* with electrolytes, thereby enabling more efficient lithium-ion transfer and stabilizing the charging and discharging processes.

Biopolymer chains contain hydroxyl (–OH), amines (–NH2), or carboxylates (–COOH) groups (Figure 3) that are capable of forming porous membrane networks strong enough to separate electrodes and facilitate ion diffusion when modified with *plasticizers*, lithium salts, or electrolyte gel phases. Recent studies show that biopolymers as separators can balance performance both mechanically and electrochemically, thereby supporting the circular economy as an alternative to environmentally friendly separators (Beg *et al*., 2025).

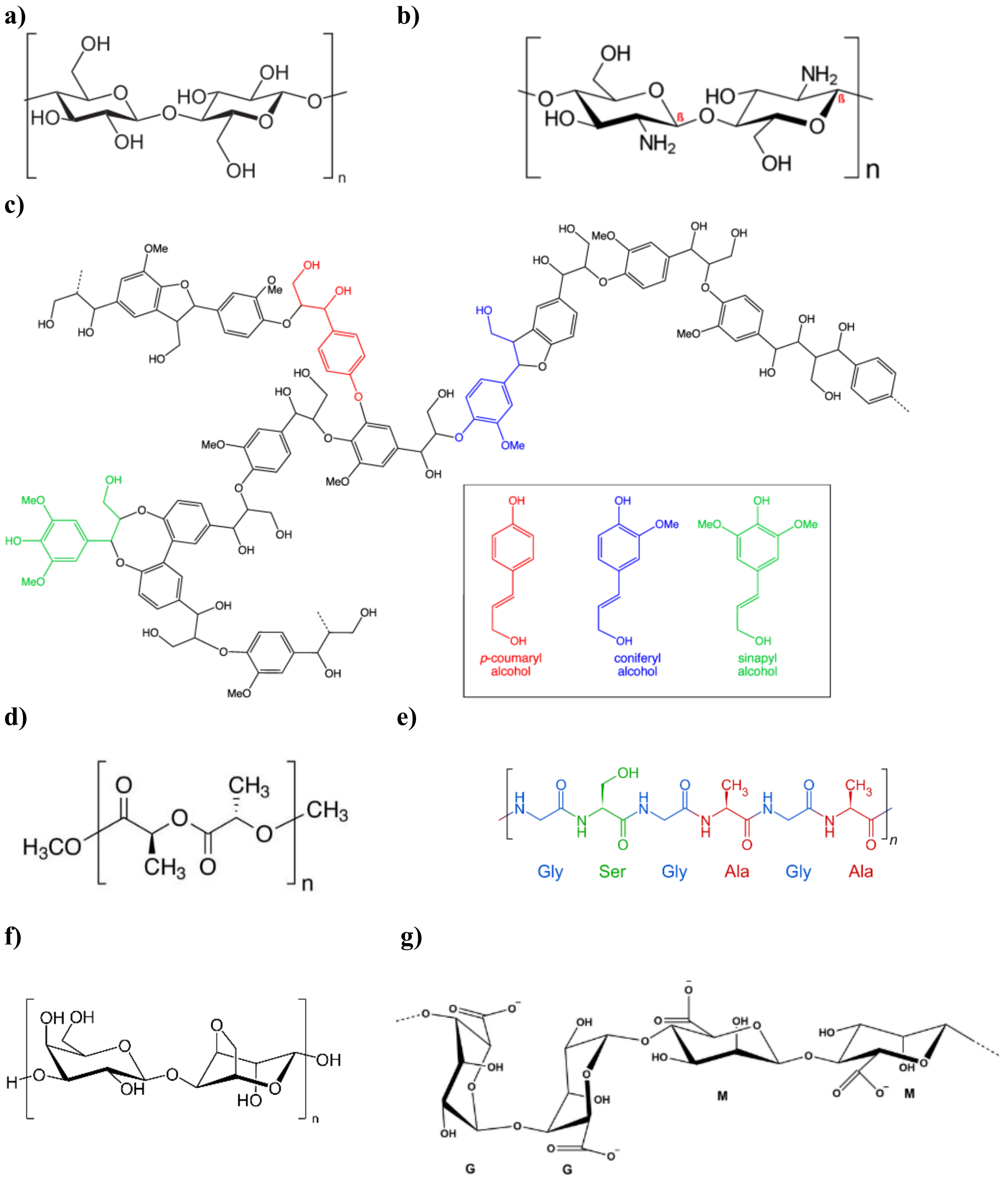

Figure 3. Biopolymer structure is (a) cellulose, (b) chitosan, (c) lignin, (d) poly(l-lactic acid), (e) silk fibroin, (f) agarose, and (g) alginate.

Biopolymer

Biopolymers are natural macromolecules composed of repeatable units and derived from biological sources (Figure 4). According to the European Bioplastics Association, biopolymers include bio-based, biodegradable, or both bio-based and biodegradable materials. Biopolymers are seen as important in sustainable materials development because they can reduce dependence on fossil fuels and support a circular economy. Its biocompatible, biodegradable, and versatile properties make biopolymers widely used across sectors, from packaging to advanced technology applications (Dutta and Sit, 2024).

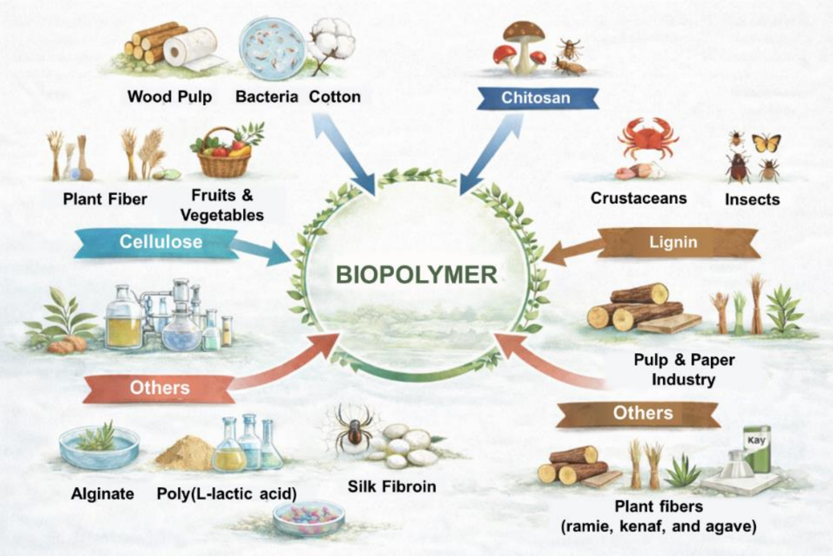

Figure 4. Source of biopolymers.

Cellulose

Cellulose is a linear polysaccharide composed of glucose units connected by β-acetal bonds, is chiral, hydrophilic, biodegradable, and acts as the main structural component of the plant cell wall (Dahman, 2017). These characteristics make cellulose-based biopolymers attractive as LIB separator matrices, owing to their engineeringable porosity, high thermal stability, and hydrophilic properties that support Li-ion safety and transport.

Cellulose, especially in the forms of bacterial cellulose (BC) and cellulose nanofiber (CNF), is increasingly being studied as an alternative to conventional hydrophobic polyolefin separators that are susceptible to thermal shrinkage. Hydroxyl group-rich three-dimensional nanofiber networks provide excellent electrolyte wettability and heat resistance up to 150 – 160 °C, thereby supporting improved LIB safety. Nevertheless, pure cellulose has the disadvantage of a dense and less uniform pore distribution, which can limit ionic conductivity and Li-ion transfer rates. To overcome these limitations, various structural and composite engineering strategies have been developed, such as CNF modification with polyethylene glycol (PEG) and polystyrene porosity, to increase porosity and ionic conductivity (Ko *et al*., 2025), as well as oxidation and grafting of chitosan in BC to increase the distance between fibers and uniformize pore distribution (Cheng *et al*., 2023).

In addition to pore engineering, chemical functionalization has been shown to be effective in improving the performance and safety of separators, for example, reinforcement with bacterial cellulose nanocrystals (BCNCs) of *nata de coco* waste, resulting in micropore membranes with high dimensional stability of up to 150 °C and better electrolyte absorption (Ajkidkarn and Manuspiya, 2020). Furthermore, the integration of cellulose with functional materials such as MOF UiO-66-NH$_2$ in BC results in a refractory *biodegradable separator* with a high ion transfer number (Jia *et al*., 2024), while polydopamine (PDA) coating plays a role in suppressing dendrite growth through physical and chemical trapping mechanisms that stabilize Li$^+$ ion flux (Bawanwal *et al*., 2024). Thus, while cellulose offers significant advantages, a composite and functionalization approach is key to overcoming weaknesses and achieving competitive separator performance for LIB applications.

Chitosan

Chitosan is a natural polysaccharide resulting from chitine deacetylation, composed of repeated units of glucosamine bonded β(1→4). These biopolymers are widely explored because they are commercially available, low-cost, *biodegradable*, and rich in functional groups (–OH and –NH$_2$), which are easily modified through simple chemical reactions (Reineke and Davis, 2012). As a cationic biopolymer, chitosan is readily protonated in organic electrolytes, thereby increasing the dissociation of lithium salts, increasing the fraction of free Li ions, and strengthening interactions with the liquid electrolyte. Although chitosan offers these advantages, its use as a separator membrane in its pure form still faces limitations, particularly regarding mechanical strength, structural stability, and suboptimal ionic conductivity.

Recent studies confirm that these limitations can be effectively overcome through physical structure engineering and chemical hybridization, which play an important role in maximizing chitosan's performance as a separator. For example, chitosan nanofibers (CSNFs) are capable of forming a more controlled porous structure with very high electrolyte affinity, demonstrated by an absorption capability of up to 218%, far surpassing the commercial separator of Celgard 2325, thereby accelerating the transfer of Li ions and improving the electrochemical performance of battery cells (Song *et al.*, 2024). In addition, the development of collagen-chitosan composite membranes demonstrates synergistic intermolecular interactions, in which chitosan increases thermal stability while decreasing the membrane's degree of crystallinity, thereby forming amorphous pathways that support ion mobility. Optimum composition of chitosan:collagen (30%:70%) results in high ionic conductivity (>0.49 mS/cm) and an electrochemical stability window of up to 5 V vs Li/Li$^+$ (Song *et al.*, 2024) that meets the safety and stability requirements for high-voltage battery applications (Andonegi *et al.*, 2025). Overall, chitosan is a promising biopolymer separator candidate for high-performance LIBs.

Lignin

Lignin is the aromatic macromolecule that makes up the main plant cell wall, composed of randomly connected phenylpropanoid units (C6–C3), forming complex polymer networks. Structurally, lignin functions as a cellulose–hemicellulose binder that provides rigidity and mechanical resistance to woody tissues (Beaumont and Zweben, 2018). Lignin in energy storage technology has evolved from a passive filler to an active functional component in battery separators. This development is evident in the formulation of methacrylated lignin (lignin methacrylate, LMA) combined with polyvinyl alcohol (PVA) and 2-acrylamido-2-methylpropanesulfonic acid (AMPS) via electrospinning and in situ UV curing, resulting in a crosslinked polymer network with high structural stability in liquid electrolytes. The rigid aromatic structure of lignin serves as the backbone of the hydrogel, increasing mechanical strength by up to 48.2 MPa and providing thermal stability without shrinkage up to 200 °C, far exceeding polyolefin separators, which deform above 130 °C (Dou *et al.*, 2024).

In terms of electrochemistry, the presence of sulfonate groups ($–SO_3^–$) of AMPS and the morphology of porous nanofibers significantly increase the transfer of Li ions. The lignin-based separator exhibits an ionic conductivity of 1.16 mS cm^{-1} with a lithium ion transport number (t^+) of 0.64, thereby suppressing concentration polarization and improving ion transfer efficiency. The battery cells using this separator maintain a specific capacity of 149.3 mAh g^{-1} at 5C, confirming superior cycle stability and fast charging capabilities. These findings confirm the potential of lignin as a high-performance and sustainable battery separator material (Dou *et al.*, 2024).

Nevertheless, despite its promising electrochemical and thermal performance, lignin still faces fundamental challenges for industrial applications, especially related to the variability of biomass sources and the heterogeneity of chemical structures, which are strongly influenced by plant types, growing conditions, and also extraction methods (kraft, organosolv, and soda). This variation certainly has a direct impact on the reproducibility of mechanical, thermal, and electrochemical properties, and it raises the problem of inter-batch consistency, which is crucial in manufacturing. Recent studies show that technical lignin generally requires advanced processes such as purification, fractionation, or chemical modification to achieve more uniform material properties, which increases process complexity and energy consumption (Zhang *et al.*, 2023; Yan *et al.*, 2025).

This condition confirms that while lignin offers advantages as a renewable, biodegradable, and potentially low-cost material, achieving high separator performance often requires additional chemical engineering that can reduce sustainability benefits (there is a trade-off between performance and sustainability). Therefore, the challenge ahead lies not only in improving performance but also in developing efficient strategies for source standardization, quality control, and modification processes so that lignin can be adopted consistently and economically in industrial-scale LIB applications (Zhang *et al.*, 2023; Dou *et al.*, 2024; Yan *et al.*, 2025).

Poly (L-lactic acid) (PLLA)

In addition to well-researched cellulose and chitosan, the exploration of biopolymers for battery separator applications is now expanding to include alternatives that offer better thermal stability and processing flexibility. PLLA is a biodegradable thermoplastic polymer derived from renewable resources, produced through the fermentation of lactic acid from biomass such as corn, sugarcane, and cassava, so it is seen as a prime candidate for replacing fossil-based plastics (Barbosa *et al.*, 2020; Ali *et al.*, 2023). With an intrinsic viscosity of about 5.0 dL g^{-1}, PLLA has good mechanical strength, thermal stability, and processing ability. As a semicrystalline polymer, the structural properties of PLLA can be engineered through the stereochemical regulation of the chain (PLLA/PDLA ratio and stereocomplex formation), the use of *nucleating agents*, as well as thermal treatments such as *controlled annealing* and stretching, so that the degree of crystallinity and crystal morphology can be adjusted to the needs of the application (Zhao *et al.*, 2023).

In energy storage applications, PLLA shows high potential as a porous separator membrane matrix, as its microstructure and porosity can be controlled through solvent-casting techniques and polymer concentration.

PLLA membranes are reported to exhibit very high thermal stability without significant shrinkage up to 250 °C and to achieve competitive porosity (72%) and ionic conductivity (1.6 mS cm^{-1}) compared to conventional microfiber glass-based polyolefin separators (Barbosa *et al.*, 2020). However, PLLA also has limitations, especially the relatively low wettability of the electrolyte and ionic conductivity, as well as the potential for long-term degradation due to ester groups. Therefore, the development of PLLA-based separators requires structural modification or a composite approach to achieve adequate performance and stability for LIB applications.

Silk Fibroin (SF)

Silk fibroin (SF) is a fibrous protein derived from the cocoons of silkworms and spiders, composed of about 20 amino acids. It is known to exhibit high biocompatibility and bioactivity, supporting tissue regeneration by regulating cytokine expression and cell proliferation (Zhou *et al.*, 2024). In addition to biomedical applications, SF also has the potential to serve as a functional material in energy storage technology, owing to its high biocompatibility and its ability to form porous structures that support ion transfer. However, SF still has drawbacks, including limited chemical and electrochemical stability in organic electrolytes, because its peptide backbone is susceptible to degradation under high-potential conditions and during long-term cycling. The mechanical properties and dimensional stability of SF are highly dependent on the β sheet phase content, which is sensitive to processing conditions, non-solvent types, and moisture levels, thereby posing reproducibility and consistency challenges across batches. In addition, SF is hygroscopic, readily absorbing water, which can disrupt electrolyte stability and increase the risk of side reactions in the LIB system (Pan *et al.*, 2022).

The weaknesses of SF as a separator membrane can be overcome through advanced engineering, such as composite reinforcement, crosslinking, or surface modification, to improve mechanical, thermal, and electrochemical stability, enabling its application as a high-form LIB separator. One effective approach is microstructure control via the phase-inversion method, in which non-solvent selection plays a key role in controlling SF membrane microstructures. The use of isopropanol results in more open, interconnected, and homogeneous pores compared to ethanol, thereby reducing the tortuosity of ion paths and increasing ionic conductivity (3.2 mS cm^{-1}). These findings confirm that protein-based biopolymers such as SF can maintain mechanical integrity while supporting rapid Li-ion transfer at high C rates, with electrochemical performance highly dependent on the control of membrane microstructures (Reizabal *et al.*, 2020).

Agarose

Agarose is a naturally occurring polymer extracted from red seaweed and is composed of repeated units of D-galactose and 3,6-anhydro-L-galactose (Švec *et al.*, 2003). In battery applications, agarose can be engineered into a glycerol-laminated non-pore solid gel layers such as Modified Agarose-Glycerol (MAGly). In this system, the ion transfer mechanism does not depend on the physical pore pathway but rather on diffusion in the gel tissue, effectively suppressing the growth of lithium dendrites. The elasticity of the agarose–glycerol tissue allows the membrane to adapt to changes in anode volume during the plating–stripping process, while its dense properties serve as an active mechanical barrier against dendrite penetration. In addition to improving the electrochemical stability of lithium-metal batteries, MAGly's water-based, hazardous-solvent-free fabrication process underscores its potential as an eco-friendly separator aligned with green manufacturing principles (Blin *et al.*, 2020).

However, agarose still has limitations, including decreased mechanical and thermal stability at high temperatures, relatively low ionic conductivity compared to porous separators due to diffusion within the gel, and hydrophilic properties that increase the risk of water absorption and side reactions. In addition, the dependence of mechanical and electrochemical properties on polymer concentrations, plasticizer types, and processing conditions poses reproducibility and inter-batch consistency challenges for industrial applications. Therefore, the development of agarose-based separators generally requires a hybridization or composite reinforcement approach to improve stability, ionic conductivity, and performance consistency.

Alginate

Alginate is an anionic polymer produced by brown algae and bacteria, composed of residues of α-L-guluronic acid (G) and β-D-mannuronic acid (M), which are connected by glycosidic bonds 1,4. These biopolymers are non-toxic, biodegradable, low-cost, and have good biocompatibility and non-immunogenic properties. However, the composition and sequence of G/M blocks are highly dependent on the algae source, environmental conditions, and extraction methods; thus, they greatly influence physicochemical properties such as mechanical strength, ionic stability, and response to ionic crosslinking (Paques *et al.*, 2014).

The development of a calcium-alginate (Ca-Alg) membrane in the LIB separator via the phase-inversion method, using an environmentally friendly water–ethanol solvent system and an ionic crosslinking mechanism with Ca^{2+} ions, has been shown to improve mechanical integrity, thermal stability, and electrolyte compatibility compared to sodium alginate. The electrochemical performance exhibits very high cycle stability, with capacitance retention >90% after 100,000 cycles and approximately 89% after 250,000 cycles, confirming the potential of Ca-Ag as a reliable separator (De Marco *et al*., 2025).

However, for large-scale industrial applications, such as lignin, alginate still faces significant challenges related to biomass sources and G/M composition heterogeneity, which impact the reproducibility of mechanical and electrochemical properties and the control of inter-batch consistency. Studies have shown that achieving consistent separator performance generally requires standardizing raw materials, controlling G/M ratios, or using hybridization and composite approaches to suppress variations in material properties (Lee and Mooney, 2012). Therefore, although alginate offers a significant sustainability advantage, developing alginate-based separators at an industrial scale requires balancing electrochemical performance optimization with control strategies to manage material variability and ensure consistent, cost-effective adoption.

Synthesis Methods and Fabrication Strategies

Solvent Casting

The solvent-casting method is one of the most commonly used synthesis approaches for fabricating biopolymer-based separator membranes, as it allows control over membrane structure, including thickness, porosity, and homogeneity. In PLLA-based membranes, solvent casting enables control of pore microstructure by regulating polymer concentration and solvent evaporation rate, thereby directly affecting electrolyte absorption and ionic conductivity. Studies show that PLLA membranes synthesized by solvent casting at 10 wt% exhibit a porosity of approximately 72% and an ionic conductivity of 1.6 mS cm^{-1}, as well as thermal stability without significant shrinkage up to 250 °C, providing electrochemical performance comparable to that of glass fiber-based commercial separators. In addition, solvent casting produces a more uniform pore distribution than mechanical or sintering methods, thereby reducing the tortuosity of ion paths and improving the efficiency of Li^+ displacement. The semicrystalline structure of PLLA formed during solvent removal also contributes to increased mechanical strength and resistance to dendrite penetration, as reflected in the high cycle stability and discharge capacity at medium to high C rates (Barbosa *et al*., 2020).

In comparison, in the collagen–chitosan system, although the main method used is freeze-drying, studies confirm that solution-based processing, including solvent casting, plays an important role in determining intermolecular interactions, hydrogen-bonded tissue formation, and phase homogeneity. These parameters are directly correlated with the high porosity (>90%), large electrolyte uptake, and ionic conductivity (>0.49 mS cm^{-1}) required for stable performance of LIB separators (Andonegi *et al*., 2025). Overall, the study shows that solvent casting not only serves as a simple, scalable fabrication method but also as an effective structural engineering tool to optimize the balance between porosity, thermal stability, mechanical strength, and electrochemical performance of biopolymer-based separator membranes.

Papermaking and *Filtration*

The papermaking method combined with vacuum filtration has proven effective for producing biopolymer-based separator membranes with a controlled, homogeneous, and easily scalable porous, fibrous structure. In BC separators, vacuum filtration enables the formation of tightly knit, interconnected three-dimensional nanofiber networks, allowing precise control of pore size and ion permeability through slurry concentration and filtration conditions. Studies show that BC pore size control directly improves the balance between electrolyte uptake, ionic conductivity, and resistance to dendrite penetration, reflected in improved cycle stability and rate performance in LIB cells (Cheng *et al.*, 2023).

A similar approach is applied to chitosan-based separators, where these biopolymers are processed as pulp through high-pressure beating and homogenization, then wet-laid into micro- or nanofiber paper. The porous structure, with low tortuosity and interconnected pores, enables more efficient Li^+ ion transfer than solid-film separators. Studies show that nanofiber chitosan separators with a fiber diameter of about 400 nm have optimal porosity, high electrolyte absorption (≈281%), ionic conductivity of up to 1.15 mS cm^{-1}, and non-shrinkage thermal stability of up to 160 °C, which contributes to improved rate performance and cycle stability compared to commercial polyolefin separators (Song *et al*., 2024).

The papermaking method on the CNF system results in uniform pore distribution, while vacuum filtration ensures adequate fiber densification to maintain mechanical integrity. The addition of PEG and PS particles as transient porogens, which are then removed, results in macrocavities that significantly increase porosity and membrane permeability while smoothing the Li-ion transfer pathway (Ko *et al*., 2025). Overall, the study confirms that the combination of papermaking, vacuum filtration, and pore engineering directly controls the structure–ion transfer–electrochemical stability relationship, making this approach a strategy for developing high-performance, environmentally friendly biopolymer separators that are feasible for industrial-scale LIBs.

Phase Inversion

Phase inversion is a controlled phase-separation process that transforms a homogeneous polymer solution from a liquid state into a solid. Phase inversion is becoming the most common method for fabricating porous polymer membranes. The structure, properties, and chemical interactions of the resulting membrane are greatly influenced by the type of polymer and the additives used in the casting solution (Purkait *et al*., 2018). In biopolymer-based battery separator applications, the immersion precipitation technique is widely used because it can control pore size and morphology via a solvent–non-solvent exchange mechanism.

SF membranes show that the non-solvent type directly determines pore morphology and ion-transfer properties. The use of ethanol triggers rapid phase separation, resulting in small pores with high tortuosity and low ionic conductivity, while isopropanol slows down the demixing process resulting in the formation of an open-porous (sponge-like) structure with more effective porosity, high electrolyte absorption (~350%), and ionic conductivity of ~3.2 mS cm^{-1}, which contributes to a decrease in internal resistance and an increase in battery rate performance (Reizabal *et al*., 2020).

The phase inversion approach is also effective in composite systems, such as PEBAX membranes reinforced with BCNCs, where the addition of BCNCs accelerates phase separation and promotes the formation of finger-like void-shaped macropores. This asymmetrical structure significantly improves the wettability of the electrolyte, electrolyte absorption, and mechanical and thermal stability, ultimately enhancing the stability of the battery cell cycle (Ajkidkarn and Manuspiya, 2020).

In addition, the development of alginate-based separators through a water–ethanol phase inversion system combined with Ca^{2+} ionic crosslinking confirms the advantages of an environmentally friendly approach. The polymer precipitation and ionic gelation processes occur simultaneously during dyeing, yielding a Ca-alginate network with controlled porosity, high mechanical strength, and stable electrochemical compatibility. The resulting separator demonstrates long-term cycle stability and consistent electrochemical performance, confirming that phase inversion serves not only as a membrane-forming method but also as a structural engineering tool for high-performance biopolymer separator stability (Ajkidkarn and Manuspiya, 2020; Reizabal *et al*., 2020; De Marco *et al*., 2025).

Electrospinning

The electrospinning technique is increasingly widely applied in the development of LIB separators because it can produce nonwoven nanofiber networks with high porosity, large surface area, and open ion-transfer paths, which are difficult to achieve with conventional stretching-based polyolefin separators. This three-dimensional random fiber structure forms interfiber pores with low tortuosity, thereby directly improving the electrolyte wettability and Li-ion mobility. The nanoparticle-reinforced PAN/polyimide system showed that the addition of SiO_2 improved the mechanical strength and thermal stability of the membrane and prevented the adhesion of excess fibers during electrospinning, thereby maintaining pore volume and higher total porosity compared to Celgard's commercial separators. This structure results in lower interface resistance and supports more stable speed and faster charging performance (Arifeen *et al*., 2021).

However, the fiber tissue produced by electrospinning is prone to swelling or shrinkage in the liquid electrolyte, which can degrade structural stability during long cycles. To overcome these limitations, a hybrid electrospinning–crosslinking approach through UV-curing with methacrylated lignin (LMA) as a macro-crosslinker was developed. Studies show that LMA can chemically lock fiber networks, resulting in cross-bonded membranes with high dimensional stability, thermal resistance up to ~200 °C, and increased ionic conductivity and Li^+ transport numbers. This chemical reinforcement significantly suppresses fiber structural degradation during battery operation and improves cycle stability at high current rates (Dou *et al*., 2024). Overall, the study confirms that electrospinning is a highly flexible fabrication technique, in which the control of fiber diameter, material composition, nanofiller addition, and post-fabrication treatments, such as crosslinking, can be synergistically optimized to produce separators with high porosity, good mechanical and thermal stability, and superior electrochemical performance.

Coating* and *Surface Modification

Surface modification and coating techniques have proven effective in improving the performance of battery separators by overcoming the limitations of base materials through control of pore structure, interfacial properties, and thermal stability. In PP–SiO_2 separators fabricated through the online construction method, SiO_2 nanoparticles are implanted simultaneously during the biaxial stretching process of β-crystal polypropylene, so that they are distributed not only on the surface but also on the internal pore wall without closing the ion path. This approach significantly smooths the coarse fibril, narrows the pore-size distribution, and lowers the Gurley value, thereby directly increasing ion permeability and ionic conductivity to ~0.80 mS cm^{-1}, while extending the electrochemical stability to ~4.8 V. In addition, the presence of hydrophilic SiO_2 increases electrolyte wettability and lowers interfacial resistance, thereby improving high-rate performance and long-term cycle stability compared to conventional PP separators (Ding *et al*., 2022).

Meanwhile, the PDA biomimetic coating approach for BC-based separators aims to control the battery's internal chemical reactions. The catechol and amine groups in the PDA serve as centers of effective chemical interactions that bind dissolved polysulfides in the Li–S battery system, thereby suppressing the shuttle effect and improving capacity stability during long cycles. Simultaneously, the PDA layer strengthens the BC nanocellulose network through hydrogen bonding and secondary covalent bonds, thereby improving the mechanical resistance and stability of the separator structure without compromising ion permeability. The increased tortuosity of the controlled ion path also helps balance the ion transfer and suppress the side reactions at the electrode interface (Baranwal *et al*., 2024). Overall, the study shows that surface coating and modification strategies are directly correlated with increased wettability, thermal stability, ion transfer, and electrochemical stability.

Composite* and *Crosslinking

Phase separation between the polymer and filler matrices is a major challenge in the manufacture of composite separators, as it can degrade structural stability and electrochemical performance. Studies show that chemical crosslinking with epichlorohydrin (ECH) effectively addresses this problem by covalently binding UiO-66-NH_2 particles to BC tissue. The chemical bond between the –OH cellulose and –NH_2 MOF groups results in a homogeneous filler distribution and a more uniform pore structure, thereby increasing electrolyte wettability, ionic conductivity (≈1.2 mS cm^{-1}), as well as lithium ion transport numbers (t^+ ≈0.5). This improvement in structure and ion transfer directly reduces the interface resistance and improves the stability of the battery cell cycle. In addition to its improved electrochemical performance, the BC/UiO-66-NH_2 separator exhibits high thermal stability without shrinkage and flame-retardant properties, confirming that the chemical crosslinking strategy is effective in producing a safe, stable, and high-performance composite separator for LIBs (Jia *et al*., 2024).

Overall, the selection of fabrication methods should be tailored to the material's purpose and characteristics. For the initial research stage or the development of simple biopolymers, the solvent-casting method is the right choice because it is relatively easy, low-cost, and flexible on a laboratory scale. In fibrous biopolymers such as cellulose, the papermaking method or vacuum filtration is superior because it can leverage the fibers' natural structure to produce a membrane with good mechanical integrity. However, if the main goal is to improve performance, especially porosity and surface area, electrospinning or phase inversion is highly recommended, as it can produce a controlled porous structure. Meanwhile, to improve safety and thermal resistance, applying a coating or making composites with ceramic materials is an effective strategy to strengthen the structural and thermal stability of the resulting membrane. Some synthesis methods and strategies for fabricating membrane separators are presented in Table 1.

Table 1. Synthesis methods and separation membrane fabrication strategies.

Synthesis Method	Mechanism of Stages	Pros	Cons
Solvent Casting	1. Dissolution of polymers in solvents until homogeneous 2. Pouring of the solution onto a flat substrate 3. Slow evaporation of solvents 4. Removal of dry film from the substrate	• The process is very simple and inexpensive; • Easy to do on a laboratory scale; • The resulting film is homogeneous.	• Difficult to control pore size; • The evaporation process takes a long time; • The potential for residual solvents is left behind.

Table 1. Synthesis methods and separation membrane fabrication strategies (continued).

Synthesis Method	Mechanism of Stages	Pros	Cons
Papermaking and Vacuum Filtration	1. Dispersion of fibers in water/solvent into suspensions (slurries) 2. Pouring of suspension over the strainer 3. Extraction of liquid by vacuum (filtration) until a wet sheet is formed 4. Pressing and drying	• It is suitable for fibrous biopolymers (cellulose); • Eco-friendly (usually water-based); • High scalability (similar to the paper industry).	• The pore size is too large (it needs to be tightly controlled); • The surface can be rough; • Requires binder optimization to be strong.
Phase Inversion	1. Polymer solution manufacturing 2. Printing of the solution on the substrate 3. Soaking in a non-solvent bath (coagulation) 4. The exchange of solvents with non-solvents triggers phase separation and pore formation 5. Washing and drying	• The control of porosity and pore structure is excellent; • Industry-standard method for porous membranes; • The pore structures are interconnected.	• Using organic solvents in large quantities; • The post-treatment process (washing) is quite long.
Electrospinning	1. High voltage application to polymer solution at needle tip 2. Taylor cone formation and nano fiber jet firing 3. Evaporation of solvents in the air as the fibers fly 4. Random accumulation of nanofibers on collectors forming nets	• Produces very high porosity (>80%); • The specific surface area is very large; • The nanofiber structure improves ionic performance.	• Low (weak) sheet mechanical strength; • Slow production rate (less efficient for large-scale production without modifications); • The cost of equipment is high.
Coating and Surface Modification	1. Preparation of slurry (mixture of ceramic particles and binder) 2. Slurry coating over the surface of the base separator (dip/spray) 3. Drying of the coating	• Drastically improves thermal stability (heat resistance); • Increase the wettability of electrolytes; • Adds battery safety.	• Increase the thickness and weight of the separator (decrease energy density); • It can clog pores if the particles are too large/thick.
Composite (Blending)	1. Mixing filler into the polymer solution before printing 2. Particle dispersion should be even 3. Printed using other methods (e.g., casting/ electrospinning)	• Combining material advantages (e.g., polymer flexibility + ceramic strength); • Improves ionic conductivity and tensile strength.	• Risk of agglomeration (clumping) of filler particles; • The interface between the polymer and the filler can be a weak point if they are incompatible.
Crosslinking	1. Formation of the initial membrane 2. Exposure to UV radiation or heat 3. A chemical reaction occurs to form a bridge between polymer chains	• Very high chemical and thermal stability (not easy to dissolve/melt); • Mechanical strength is significantly increased.	• The membrane can become brittle/stiff (less flexible); • The process is more complex and requires precise reaction control.

Performance and Feasibility of Separators

In LIB separators, performance is determined not only by their basic function as a separator between the anode and the cathode, but also by a number of key parameters that directly affect the efficiency, safety, and lifespan of the battery, such as thermal stability, ionic conductivity, *wettability,* and porosity (Table 2). Among these parameters, porosity plays a central role, as it determines the separator's ability to absorb and retain electrolytes, serve as a transfer path for Li^+ ions, and control internal resistance and the homogeneity of ion flux. The controlled, evenly distributed porosity enhances ionic conductivity and electrolyte wetness, whereas suboptimal porosity can limit ion diffusion or increase the risk of electrochemical instability.

Therefore, the evaluation of the separator's material feasibility cannot be based solely on testing its physical and structural properties; it must be validated through electrochemical testing directly in the battery cell. Performance parameters such as initial discharge capacity, rate capability, and cycle stability (Table 3) provide a more comprehensive picture of separator effectiveness under real operating conditions, as they reflect the synergetic effects between porosity, ionic conductivity, thermal stability, and separator–electrolyte interactions. Electrochemical testing is a crucial stage to ensure that the separator's pore characteristics and physical properties contribute to the overall improvement in the performance and reliability of the LIB.

Porosity

Porosity is an important structural parameter in LIB separator applications because it directly determines the electrolyte absorption ability, ionic conductivity, and electrochemical performance and stability of the cell. The porous structure allows the electrolyte to be absorbed and evenly distributed within the separator matrix, thereby providing a continuous path for Li^+ ion transfer during charging and discharging (Arora and Zhang, 2004). A number of studies have shown that porosity with nano–mesoporous pore sizes (tens to hundreds of nanometers) can reduce ion path tortuosity and internal resistance, which has a direct impact on improving ionic conductivity, *rate capability*, and battery cycle stability (Cheng *et al*., 2023; Zhang *et al*., 2023). In contrast, pores that are too small can limit ion diffusion, whereas micrometer-scale pores may increase the risk of lithium dendrite penetration and internal short circuits, thereby reducing the cell's safety (Reizabal *et al*., 2020). Therefore, pore size control and distribution are very important design aspects in the development of LIB separators.

The results of comparing various separator systems show that porosity not only serves as a morphological parameter but also acts as a major controlling factor influencing Li^+ ion transfer, separator–electrolyte interactions, and long-term electrochemical stability. BC-based separators and their derivatives consistently exhibit superior electrochemical performance when pore sizes are in the range of tens to hundreds of nanometers, as they can balance efficient ion diffusion with ion flux homogeneity as well as suppress the formation of local electric fields that trigger dendrite growth. In contrast, separators with macro pores, such as SF membranes or freeze-dried collagen-chitosans, show that high porosity and large electrolyte uptake are not always accompanied by improved electrochemical performance, due to increased ion distribution inhomogeneity and dendrite risk, especially under high-current conditions. An alternative approach is demonstrated by non-porous or near-non-porous separators, such as the agarose–glycerol system, which effectively compresses dendrites through mechanical resistance, but has limitations in ionic conductivity due to the conduction mechanism dominated by the movement of a portion of the polymer chain. Overall, these findings confirm that the optimal porosity zone resides in interconnected nano–mesoporous structures, making pore size and distribution control more crucial than simply increasing total porosity in sustainable biopolymer-based LIB separator designs (Jia *et al*., 2024; De Marco *et al*., 2025).

Thermal Stability

Thermal stability is a crucial factor directly related to LIB safety, especially under high load or in the event of system failures that can trigger a drastic temperature rise. Conventional polyolefin separators generally begin to shrink or melt at temperatures above 130°C, increasing the risk of internal short circuits and thermal runaway. To overcome these limitations, various innovations have been developed, including SiO_2-based inorganic coatings, heat-resistant polymers such as polyimide (PI), and biopolymers such as BC, which offer higher thermal resistance. This approach allows the separator to maintain structural integrity at 150 – 200 °C, thereby improving safety.

As summarized in Table 2, composite separators exhibit the highest thermal stability due to the synergistic roles among thermostable polymers, nanofiber networks, and nanoceramic fillers. Specifically, PI acts as a matrix with a rigid imide structure and a decomposition temperature, while electrospun PAN forms an interconnected nanofiber network that strengthens its mechanical integrity. The thermal pressing process improves fiber adhesion and suppresses dimensional shrinkage, while SiO_2 nanoparticles act as a physical barrier, limiting polymer chain mobility and preventing pore collapse. The PAN/PI–SiO_2 combination produces a separator with no significant shrinkage up to 150 °C and is chemically and physically stable >250 °C, far surpassing conventional polyolefin separators.

Ionic Conductivity

Ionic conductivity represents the ease of movement of lithium ions (Li^+) across the separator. In commercial separators, overly dense pore tissue often becomes an obstacle to ion flow, increasing internal resistance and degrading performance, especially at fast charging. Studies on PEG/nanocellulose systems show that high fiber density slows down ion migration. In contrast, biopolymer separators such as chitosan nanofibers and lignin hydrogels are designed with interconnected porosity to accelerate ion transfer, further supporting high-power applications.

High ionic conductivity of 2.75 mS cm^{-1}, with the polymer lignin-2 (PL-2) variant showing the highest value (Table 2), was achieved through synergistic structural engineering and chemical interactions. The addition of methacrylated lignin (LMA) as *a crosslinker* disrupts the hydrogen bonds of the PVA matrix, lowers crystallinity, and expands the amorphous phase that facilitates the movement of Li^+. The sulfonate group ($–SO_3^-$) of AMPS acts as an ion-hopping site that accelerates ion migration. The PL-2 variant has a fairly stable optimal crosslinking density without inhibiting free volume, so, together with the porous nanofiber structure resulting from electrospinning and its high electrolyte affinity, it produces highly efficient ion transfer.

Table 2. Porosity, thermal stability, ionic conductivity, and wettability of the LIB separator.

Main Materials	**Porosity (nm)**	**Thermal Stability**		**Ionic conductivity (σ) (mS/cm)**	**Wettability**		**References**
		°C	**% Depreciation**		**Contact Angle ()°**	**Uptake (%)**	
Poly(L-lactic acid) (PLLA)	~100 – 500	~150	<5	0.74	~30	140 – 360	(Barbosa *et al.*, 2020)
Alginate and PEO	~150	150	0	1.2	~0	>200	(De Marco *et al.*, 2025)
Bacterial Cellulose (BC) and Chitosan.	40 – 100	200	~1.2	1.28	18.5	295	(Cheng *et al.*, 2023)
Nanocellulose and PEG	50 – 100	160	~2.1	1.51	12.4	262	(Ko *et al.*, 2025)
SiO_2/PAN/Polyimid	200 – 400	>250	0	1.98	8.2	456	(Arifeen *et al.*, 2021)
Chitose Nanofiber (CSNF)	50 – 150	200	<2	1.15	22.5	248	(Song *et al.*, 2024)
Collagen/Chitosane	~100	150	~3	1.42	42	185	(Andonegi *et al.*, 2025)
PP/ Fumed SiO_2	~80	150	~8.5	0.82	52	165	(Ding *et al.*, 2022)
PDA and BC	~45	200	~1.8	1.35	~0	312	(Baranwal *et al.*, 2024)
BC Nanocrystals / PEBAX	~120	160	0 – 2	1.75	48	158	(Ajkidkarn and Manuspiya, 2020)
Bacterial Cellulose (BC) / UiO−66−NH_2	-	241	-	1.15	-	215.1	(Jia *et al.*, 2024)
PVA / AMPS / Livine (LMA)	230 – 660	170	0	2.75	-	661.4	(Dou *et al.*, 2024)
Agarose/Glycerol (MAGly)	Non-porous (Solid)	180	<5	-	-	-	(Blin *et al.*, 2020)
Silk Fibroin	22,000 – 38,000	300	-	-	-	350	(Reizabal *et al.*, 2020)

Wettability

Wettability is the separator's ability to absorb and interact with liquid electrolytes, which is important for ensuring an even distribution of Li^+ ions during battery operation. Conventional PP/PE-based polyolefin separators are hydrophobic, making it difficult for the electrolyte to penetrate the pores and leading to high interfacial resistance. In contrast, biopolymer separators such as alginate and PLLA exhibit hydrophilic properties with low contact angles, enabling rapid, homogeneous electrolyte absorption. Good electrolyte absorption not only improves the efficiency of ion transfer but also simplifies the cell assembly process, as wetting of the separator occurs more evenly.

The PDA/BC separator exhibits superhydrophilic properties with a contact angle of 0° (Table 2), far exceeding that of Celgard commercial polypropylene-based separators. This advantage stems from the polydopamine layer's modification of the surface, which is rich in polar groups (–OH and $–NH_2$), thereby increasing surface energy and affinity for polar electrolytes. The high surface energy, combined with the three-dimensional nanofiber structure, amplifies the capillary effect and allows the electrolyte to penetrate quickly throughout the pores. As a result, PDA/BC achieves very high electrolyte uptake (312%), well above Celgard (<150%), thereby significantly lowering internal resistance and improving the efficiency of Li^+ ion displacement.

Discharge Capacity

The initial discharge capacity at a low current rate is an important indicator of the battery's ability to deliver energy to its full capacity, with the separator playing a crucial role by absorbing electrolytes and lowering interfacial resistance. Biopolymer separators, such as PVA/lignin and collagen/chitosan, exhibit high wettability, so the electrolyte evenly distributes across the separator and electrode, preventing a dry region. This condition allows more Li^+ ions to participate in electrochemical reactions, resulting in a specific capacity close to the theoretical value of the $LiFePO_4$ cathode (163 – 167 mAh/g).

In half-cell $LiFePO_4$/Li testing at 0.1 C, the PVA/Lignin separator showed an initial discharge *capacity* of 167 mAh g^{-1}. Excellent hydrophilic properties and electrolyte retention support this performance due to the presence of –OH and $–SO_3^-$ groups, as well as high ionic conductivity (2.75 mS cm^{-1}) that effectively suppresses internal resistance and ohmic polarization.

Rate Capability (C Rate)

Rate capability is an important parameter for electric vehicle applications that demand fast charging and discharging. At high *C-rates*, the separator should facilitate the efficient transfer of Li^+ ions without significant resistance. Separators with high porosity and interconnected ion paths, such as chitosan nanofibers and nano-silica/PI composites, have been shown to maintain performance up to 5 C, while dense porous separators are prone to voltage degradation and capacity degradation.

In *the capability rate test*, the chitosan nanofiber separator (CSNF), specifically the optimal variant of CSNF15, showed much superior performance compared to the Celgard 2325. At a rate of ≥2 C, Celgard's capacity declined sharply, while the CSNF remained stable and almost fully recovered as the current was returned to 0.2 C. These advantages stem from the superhydrophilic properties of chitosan and the high porosity and low tortuosity of the 3D nanofiber networks, which result in high ionic conductivity, low resistance, and minimal polarization.

Cycle Stability

Cycle stability is a key determinant of battery life, as separators must maintain mechanical and chemical integrity over hundreds to thousands of charge–discharge cycles. A critical challenge in long-term operation is the growth of lithium dendrites, which can penetrate the separator and trigger an internal short circuit. Various mitigation strategies have been developed, including the use of agarose-based protective coatings and surface modification with polydopamine, which effectively suppresses dendrite growth and maintains capacity retention above 90% after hundreds of cycles.

Compared with conventional polyolefin separators, biopolymer separators exhibit superior cycle stability through different structural mechanisms. The methacrylated lignin-based PL-2 separator maintains high capacity retention (>93 – 95%) and near-100% Coulomb efficiency in half $LiFePO_4$/Li cells, thanks to a crosslinking network that suppresses swelling, maintains nanofiber integrity, and stabilizes the SEI. Meanwhile, the Ca-Alg separator exhibits exceptional cyclical resistance of up to hundreds of thousands of cycles, supported by Ca^{2+}–alginate (egg-box) ionic cross-bonding, high mechanical-thermal stability, and even distribution of ion flux. These findings confirm that high wettability and stable 3D structural integrity are key to the long service life of biopolymer separators.

Table 3. Initial capacity, high performance rate, and cycle stability of the LIB separator.

Material Separator	**Battery System (Cathode/Anode)**	**Initial Capacity (*Low Rate*)**		**High Rate Performance (*High Rate*)**		**Cycle Stability (Capacity Retention)**			**References**
		Specific Capacity (mAh/g)	*C-rate*	Specific Capacity (mAh/g)	*C-rate*	% Remaining Capacity	Number of Cycles	*C-Rate*	
PLLA	LFP/Li	~160	0.1	~130	2	94	50	0.5	(Barbosa *et al.*, 2020)
Alginate	AC/AC (Supercapacitor)	31,94	15.65	22.22	450	99	250,0000	-	(De Marco *et al.*, 2025)
OBCS (BC/Kitosan)	LFP/Li	158	0.2	134	5	98.2	100	0.5	(Cheng *et al.*, 2023)
PEG/CNF	LFP/Li	148	0.2	108	5	91.5	100	-	(Ko *et al.*, 2025)
Nano-silica/PI	LFP/Li	162	0.1	147	1	-	100	-	(Arifeen *et al.*, 2021)
Chitosane Nanofiber	LFP/Li	161	0.2	123	5	96.5	200	1	(Song *et al.*, 2024)
Collagen/Chitosane	LFP/Li	163	0.1	~130	2	98	50	-	(Andonegi *et al.*, 2025)
PP + *Fumed* SiO_2	LFP/Li	~150	0.2	120	5	95.2	200	0.5	(Ding *et al.*, 2022)
PDA/BC	Li-S	1086	0.2	586	3	67	300	-	(Baranwal *et al.*, 2024)
BCNC/PEBAX	LFP/Li	~150	0.2	120	2	95	100	-	(Ajkidkarn and Manuspiya, 2020)
BC/UiO-66-NH_2	LFP/Li	162.7	0.2	132	5	96.4	200	1	(Jia *et al.*, 2024)
PVA/Lignin	LFP/Li	167	0.2	135	8	94.7	500	-	(Dou *et al.*, 2024)
Agarose/Glycerol	Li/ Li (Symmetrical)	-		-	-	Withstands >500 hours *of plating/stripping* without a short circuit (Anti-dendrite)			(Blin *et al.*, 2020)
Silk Fibroin	LFP/Li	148	0.1	~100	2	83	100	-	(Reizabal *et al.*, 2020)

Material Life Cycle and Degradation Test of Biopolymer Separators

Material Life Cycle

There are three main aspects of the material cycle of battery separators: waste-to-wealth, green manufacturing, and end-of-life (Table 4). Biopolymer-based separators excel in sustainability because they adopt the waste-to-wealth concept, using biomass waste as a high-value functional material. Scientifically, biomass such as cellulose, lignin, and chitosan is rich in active groups (–OH, –COOH, $–NH_2$) that enable strong interactions with electrolytes, crosslinking, and pore engineering without complex synthetic additives. BCNC has high crystallinity and elastic modulus as a mechanical reinforcer (Ajkidkarn and Manuspiya, 2020), lignin with a rigid aromatic structure can be engineered into a macro-crosslinker to improve the stability of the separator (Dou *et al.*, 2024), while chitosan from fishery waste is cationic and hydrophilic to support lithium salt dissociation and ion transfer (Beaumont and Zweben, 2018; Andonegi *et al.*, 2025).

Table 4. Life cycle of separator materials.

Material Separator	Upstream Phase (Source of Raw Materials)	Process Phases (Green Manufacturing)	Downstream Phase (End of Life/Degradation)	References
PLLA	Fermented plant starch (Corn/Cassava).	Organic solvent, PLLA (biocompatible)	It degrades via hydrolysis and enzymatic action into CO_2 and water.	(Barbosa *et al.*, 2020)
Alginate	Brown Algae Extract (Seaweed).	Water-based solvents and ethanol coagulation.	Fully biodegradable. It is safe for marine ecosystems if it is discarded.	(De Marco *et al.*, 2025)
OBCS	Bacterial cellulose (*Acetobacter xylinum*) + Chitosan (Shell).	The suction filtration method is simple. Energy-saving.	The composition is 100% organic, decomposed by soil microbes.	(Cheng *et al.*, 2023)
Chitosane Nanofiber	Crustacean shell waste (Shrimp/Crab).	Papermaking Method: A modified traditional papermaking technique. Without high voltage electrospinning.	Naturally degraded. Chitosan is antimicrobial yet biodegradable.	(Song *et al.*, 2024)
BCNC/PEBAX	Nata de Coco Waste.	Nanocrystal extraction (BCNC) as a polymer matrix reinforcer.	The biopolymer matrix is strengthened with cellulose, which is easily degradable.	(Ajkidkarn and Manuspiya, 2020)
BC/MOF	Bacterial cellulose.	*In situ* synthesis of MOF particles on cellulose fibers.	Soil Burial: completely destroyed in 30 days—one of the fastest degradation results.	(Jia *et al.*, 2024)
PVA/Lignin	Paper/pulp industry waste.	Electrospinning water-based solution (water-soluble PVA).	Reduce industrial lignin waste. PVA and lignin are both biodegradable.	(Dou *et al.*, 2024)
Agarose	Seaweed (Agar). Glycerol (a by-product of biodiesel) as a plasticizer.	Water solvent, simple casting process.	Pure biopolymer film, edible grade, is very safe for the environment.	(Blin *et al.*, 2020)
Silk Fibroin	Cocoon of the Silkworm (Bombyx mori).	Methanol/water to regulate porosity (Beta-sheet formation).	Natural proteins. Degraded by proteolytic enzymes in nature.	(Reizabal *et al.*, 2020)

From a green manufacturing perspective, the dominant chemical properties of biopolymers enable water-based processing or the use of green solvents, such as water–ethanol phase inversion, ionic gelation, and papermaking. This approach is safer than the use of volatile organic solvents in PP/PE separators, and also reduces VOC emissions, *solvent recovery needs*, and energy consumption because it does not require intensive thermal stretching (Blin *et al*., 2020; De Marco *et al*., 2025).

At the end-of-life stage, biopolymers have a clear and non-persistent degradation pathway. Glycosidic bonds in cellulose and chitosan, and peptide bonds in SF, can be broken down by microorganisms, with almost total degradation achieved in a matter of weeks (Reizabal *et al*., 2020; Jia *et al*., 2024). Meanwhile, PLLA is degraded through hydrolysis of ester bonds into lactic acid, which is further mineralized (Barbosa *et al*., 2020). The combination of these three aspects makes biopolymer separators in line with the principles of circular economy and sustainable battery technology without sacrificing electrochemical performance.

Degradation Test

Degradation testing is a systematic evaluation procedure to assess the ability of separator materials to decompose back into natural components, such as biomass, water, and CO_2, after the end of their useful life. In the context of sustainable batteries, this test serves to validate the "green" claims of biopolymer materials and scientifically differentiate them from conventional synthetic polymers. Fundamental differences in their chemical structure determine the degradation advantage of biopolymer separators over PP/PE.

Biopolymers such as cellulose, SF, and PLLA contain glycosidic bonds, peptides, and esters that are biologically and chemically labile, so they are easily broken by microbial enzymes or hydrolyzed into small, stable, non-toxic molecules, such as simple sugars, amino acids, and lactic acid. In soil burial tests, degradation occurs through the activity of soil microbes that produce extracellular enzymes; in cellulose, the enzyme cellulase breaks the β-1,4-glycosidic bond so that rapid depolymerization and thorough fragmentation occur in about 30 days, reflecting bulk degradation due to its hydrophilic properties that are easily penetrated by water and enzymes (Jia *et al*., 2024). In in vitro assays, SF decomposed selectively via peptide bond cleavage by a protease enzyme (Reizabal *et al*., 2020), while PLLA degraded via hydrolysis of ester bonds into lactic acid, which was further mineralized in the natural carbon cycle (Barbosa *et al*., 2020). In contrast, PP/PE separators with highly stable nonpolar C–C bonds do not undergo chemical biodegradation but are only physically fragmented into microplastics by UV exposure or mechanical stress, which are persistent and ecologically risky (Ding *et al*., 2022). Thus, the degradation test confirms that the chemical design of the biopolymer, aligned with the biological system, enables the return of materials to the natural carbon cycle and is an essential solution to suppress the accumulation of *e-waste* and microplastics in the future.

CONCLUSION

Biopolymers (bacterial cellulose, chitosan, lignin, and silk) can be used as LIB separators, with more environmentally friendly fabrication methods, including water-based solvents, papermaking techniques, and simple material modifications, while enabling the use of biomass waste as value-added raw materials. In terms of performance, biopolymer separators exhibit superior wettability, electrolyte absorption, ionic conductivity, and thermal stability compared to conventional PP/PE separators. In addition to competitive electrochemical performance, biopolymers offer sustainability advantages through natural biodegradability after the battery life has ended. The combination of high performance, enhanced safety, and sustainable material lifecycles makes biopolymers prime candidates for next-generation battery separators.

Although biopolymers show great potential as LIB separators, their widespread application is still limited by a number of fundamental challenges. The lack of a standardized test method that accurately reflects the characteristics of biopolymer separators makes it difficult to compare performance across studies. Meanwhile, the issue of manufacturing scale-up remains important, as many fabrication techniques are effective at the laboratory scale but have not been validated for mass production with high batch-to-batch consistency. In addition, the integration of biopolymers into advanced-generation batteries, particularly solid-state systems and quasi-solid electrolytes, still requires further understanding of interface compatibility and long-term stability. The commercial adoption of biodegradable separators also depends on the availability of clear regulations and safety standards. Therefore, future research should focus on developing standardized test protocols, scalable, cost-effective manufacturing approaches, and cross-disciplinary synergies to accelerate the implementation of safe, high-performance, and sustainable biopolymer separators.

CONFLICT OF INTEREST

There are no conflicts of interest in this article.

AUTHOR CONTRIBUTION

TPS: Conceptualization, Methodology, Software, Data Analysis, Manuscript Draft Writing; FK: Planning, Manuscript Draft Writing, Supervision; ASK: Manuscript Planning, Review and Editing, Supervision;

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DECLARATION OF GENERATIVE AI

During the preparation of this work, the author used ChatGPT to edit the manuscript in terms of content and structure, and to generate ideas for the graphical abstract. After using this tool/service, the author reviews and edits the content as needed and is solely responsible for the content of the published article.

REFERENCES

Ajkidkarn, P., and Manuspiya, H., 2020. Novel Bacterial Cellulose Nanocrystals/Polyether Block Amide Microporous Membranes as Separators for Lithium-Ion Batteries. *International Journal of Biological Macromolecules*, 164, 3580–3588. https://doi.org/10.1016/j.ijbiomac.2020.08.234.

Ali, W., Ali, H., Gillani, S., Zinck, P., and Souissi, S., 2023. Polylactic Acid Synthesis, Biodegradability, Conversion to Microplastics and Toxicity: A Review. *Environmental Chemistry Letters*, 21, 1761–1786. https://doi.org/10.1007/s10311-023-01564-8.

Andonegi, M., Serra, J.P., Silva, M.M., Gonçalves, R., Costa, C.M., Lanceros-Mendez, S., de la Caba, K., and Guerrero, P., 2025. Lithium-Ion Battery Separators Based on Sustainable Collagen and Chitosan Hybrid Membranes. *Electrochimica Acta*, 526. https://doi.org/10.1016/j.electacta.2025.146087.

Arifeen, W.U., Choi, J., Yoo, K., Shim, J., and Ko, T.J., 2021. A Nano-Silica/Polyacrylonitrile/Polyimide Composite Separator for Advanced Fast Charging Lithium-Ion Batteries. *Chemical Engineering Journal*, 417, 128075. https://doi.org/10.1016/j.cej.2020.128075.

Arora, P., and Zhang, Z. (John), 2004. Battery Separators. *Chemical Reviews*, 104, 4419–4462. https://doi.org/10.1021/cr020738u.

Baranwal, R., Lin, X., Li, W., Pan, X., Wang, S., and Fan, Z., 2024. Biopolymer Separators from Polydopamine-Functionalized Bacterial Cellulose for Lithium-Sulfur Batteries. *Journal of Colloid* and *Interface Science*, 656, 556–565. https://doi.org/10.1016/j.jcis.2023.11.138.

Barbosa, J.C., Reizabal, A., Correia, D.M., Fidalgo-Marijuan, A., Gonçalves, R., Silva, M.M., Lanceros-Mendez, S., and Costa, C.M., 2020. Lithium-Ion Battery Separator Membranes Based on Poly(L-Lactic Acid) Biopolymer. *Materials Today Energy*, 18, 100494. https://doi.org/10.1016/j.mtener.2020.100494.

Beaumont, P.W.R., and Zweben, C.H., 2017. Comprehensive composite materials II, Comprehensive Composite Materials II. https://doi.org/10.1016/c2016-1-04658-8.

Beg, M., Saju, J., Alcock, K.M., Mavelil, A.T., Markapudi, P.R., Yu, H., and Manjakkal, L., 2025. Biodegradable Biopolymers for Electrochemical Energy Storage Devices in a Circular Economy. *RSC Sustainability*, 3, 37–63. https://doi.org/10.1039/D4SU00468J.

Blin, P., Boury, B., Taguet, A., Touja, J., Monconduit, L., and Patra, S., 2020. Glycerol-Plasticized Agarose Separator Suppressing Dendritic Growth in Li Metal Battery. *Carbohydrate Polymers*, 247, 116697. https://doi.org/10.1016/j.carbpol.2020.116697.

Cheng, C., Yang, R., Wang, Y., Fu, D., Sheng, J., and Guo, X., 2023. A Bacterial Cellulose-Based Separator with Tunable Pore Size for Lithium-Ion Batteries. *Carbohydrate Polymers*, 304, 120489. https://doi.org/10.1016/j.carbpol.2022.120489.

Dahman, Y., 2017. Nanotechnology and Functional Materials for Engineers, Nanotechnology and Functional Materials for Engineers.

De Marco, A., Lacarbonara, G., Rea, M., Tombolesi, S., Petruzzelli, R., Gualandi, C., Focarete, M.L., and Arbizzani, C., 2025. Alginate-Based Separators for Sustainable Energy Storage Devices. *Journal of Power Sources*, 660, 238505. https://doi.org/10.1016/j.jpowsour.2025.238505.

Ding, L., Yan, N., Zhang, S., Xu, R., Wu, T., Yang, F., Cao, Y., and Xiang, M., 2022. Facile Manufacture Technique for Lithium-Ion Batteries Composite Separator via Online Construction of Fumed SiO2 Coating. *Materials & Design*, 215, 110476. https://doi.org/10.1016/j.matdes.2022.110476.

Dou, Y., Li, S., Wang, S., Gibril, M.E., and Kong, F., 2024. Utilizing Methacrylated Lignin as a Sustainable Macro-Crosslinker for Synthesizing Innovative PVA/AMPS Composites Crosslinked Hydrogel Nanofibers: A Potential Application for Lithium-Ion Battery Separators. *Composites Part B: Engineering*, 281, 111537. https://doi.org/10.1016/j.compositesb.2024.111537.

Dutta, D., and Sit, N., 2024. A Comprehensive Review on Types and Properties of Biopolymers as Sustainable Bio-based Alternatives for Packaging. *Food Biomacromolecules*, 1, 58–87. https://doi.org/10.1002/fob2.12019.

International Energy Agency, 2023. *Global EV Outlook 2023*, IEA, Paris <https://www.iea.org/reports/global-ev-outlook-2023> (diakses pada 3 Januari 2026).

Jia, S., Chen, Z., Li, Y., Li, C., Duan, C., Lim, K.H., and Kawi, S., 2024. Construction of Greenly Biodegradable Bacterial Cellulose/UiO-66-NH2 Composite Separators for Efficient Enhancing Performance of Lithium-Ion Battery. *International Journal of Biological Macromolecules*, 269, 131988. https://doi.org/10.1016/j.ijbiomac.2024.131988.

Ko, Y., Jung, H.Y., Jung, H.W., Ko, S.M., Lee, J.T., and You, J., 2025. A New Strategy for Accelerating Redox Kinetics in Lithium-Ion Batteries: Highly Porous Poly(Ethylene Glycol)/Nanocellulose Separators. *Carbohydrate Polymers*, 356, 123372. https://doi.org/10.1016/j.carbpol.2025.123372.

Lee, K.Y., and Mooney, D.J., 2012. Alginate: Properties and Biomedical Applications. *Progress in Polymer Science*, 37, 106–126. https://doi.org/10.1016/j.progpolymsci.2011.06.003.

Lingappan, N., Lee, W., Passerini, S., and Pecht, M., 2023. A Comprehensive Review of Separator Membranes in Lithium-Ion Batteries. *Renewable* and *Sustainable Energy Reviews*, 187, 113726. https://doi.org/10.1016/j.rser.2023.113726.

Liu, F., and Chuan, X., 2021. Recent Developments in Natural Mineral-Based Separators for Lithium-Ion Batteries. *RSC Advances*, 11, 16633–16644. https://doi.org/10.1039/D1RA02845F.

Lizundia, E., and Kundu, D., 2021. Advances in Natural Biopolymer-Based Electrolytes and Separators for Battery Applications. *Advanced Functional Materials*, 31. https://doi.org/10.1002/adfm.202005646.

Pan, P., Zhang, M., Cheng, Z., Jiang, L., Mao, J., Ni, C., Chen, Q., Zeng, Y., Hu, Y., and Fu, K. (Kelvin), 2022. Garnet Ceramic Fabric-Reinforced Flexible Composite Solid Electrolyte Derived from Silk Template for Safe and Long-Term Stable All-Solid-State Lithium Metal Batteries. *Energy Storage Materials*, 47, 279–287. https://doi.org/10.1016/j.ensm.2022.02.018.

Paques, J.P., van der Linden, E., van Rijn, C.J.M., and Sagis, L.M.C., 2014. Preparation Methods of Alginate Nanoparticles. *Advances in Colloid* and *Interface Science*, 209, 163–171. https://doi.org/10.1016/j.cis.2014.03.009.

Petersen, K., Vakkalanka, S., and Kuzniarz, L., 2015. Guidelines for Conducting Systematic Mapping Studies in Software Engineering: An Update. *Information* and *Software Technology*, 64, 1–18. https://doi.org/10.1016/j.infsof.2015.03.007.

Purkait, M.K., Sinha, M.K., Mondal, P., and Singh, R., 2018. Introduction to Membranes. pp. 1–37. https://doi.org/10.1016/B978-0-12-813961-5.00001-2.

Reineke, T.M., and Davis, M.E., 2012. Nucleic Acid Delivery via Polymer Vehicles, in: Polymer Science: A Comprehensive Reference. Elsevier, pp. 497–527. https://doi.org/10.1016/B978-0-444-53349-4.00239-9.

Reizabal, A., Gonçalves, R., Fidalgo-Marijuan, A., Costa, C.M., Pérez, L., Vilas, J.-L., and Lanceros-Mendez, S., 2020. Tailoring Silk Fibroin Separator Membranes Pore Size for Improving Performance of Lithium Ion Batteries. *Journal of Membrane Science*, 598, 117678. https://doi.org/10.1016/j.memsci.2019.117678.

Song, Y., Zhao, G., Zhang, S., Xie, C., Yang, R., and Li, X., 2024. Chitosan Nanofiber Paper Used as Separator for High Performance and Sustainable Lithium-Ion Batteries. *Carbohydrate Polymers*, 329, 121530. https://doi.org/10.1016/j.carbpol.2023.121530.

Švec, F., Tennikova, T.B., and Deyl, Z., 2003. Monolithic materials : preparation, properties, and applications, 1st ed. ed, Journal of chromatography library, v. 67. Elsevier, Amsterdam.

Yan, L., Huertas-Alonso, A.J., Liu, H., Dai, L., Si, C., and Sipponen, M.H., 2025. Lignin Polymerization: Towards High-Performance Materials. *Chemical Society Reviews*, 54, 6634–6651. https://doi.org/10.1039/D4CS01044B.

Yang, Y., Wang, W., Meng, G., and Zhang, J., 2022. Function-Directed Design of Battery Separators Based on Microporous Polyolefin Membranes. *Journal of Materials Chemistry A*, 10, 14137–14170. https://doi.org/10.1039/D2TA03511A.

Zhang, M., Tian, R., Tang, S., Wu, K., Wang, B., Liu, Y., Zhu, Y., Lu, H., and Liang, B., 2023. The Structure and Properties of Lignin Isolated from Various Lignocellulosic Biomass by Different Treatment Processes. *International Journal of Biological Macromolecules*, 243, 125219. https://doi.org/10.1016/j.ijbiomac.2023.125219.

Zhao, X., Yu, J., Liang, X., Huang, Z., Li, J., and Peng, S., 2023. Crystallization Behaviors Regulations and Mechanical Performances Enhancement Approaches of Polylactic Acid (PLA) Biodegradable Materials Modified by Organic Nucleating Agents. *International Journal of Biological Macromolecules*, 233, 123581. https://doi.org/10.1016/j.ijbiomac.2023.123581.

Zhou, L., Zhang, Y., Yi, X., Chen, Y., and Li, Y., 2024. Advances in Proteins, Polysaccharides, and Composite Biomaterials for Enhanced Wound Healing via Microenvironment Management: A Review. *International Journal of Biological Macromolecules*, 282, 136788. https://doi.org/10.1016/j.ijbiomac.2024.136788.