

Review

State of the Art and Recent Advances on Ester and Ether Derivatives of Polysaccharides from Lignocellulose: Production and Technological Applications

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Abstract

In an era defined by the imperative for sustainable, high-performance materials, this review examines the development and utility of key ester and ether derivatives from both cellulose and hemicellulose sourced from lignocellulosic biomass, with a special emphasis on waste feedstocks. Our findings indicate that these derivatives exhibit tunable physicochemical properties, enabling their broad use in established industrial sectors while also fueling the emergence of novel technological applications in nanotechnology, controlled delivery, tissue engineering, environmental remediation, electronics, and energy fields. This dual-polysaccharide platform demonstrates that underutilized biomass streams can be repurposed as valuable feedstocks, promoting a circular supply chain and supporting more sustainable solutions, thereby aligning with the goals of eco-friendly innovation in materials science. Future progress will likely depend on integrating green chemistry synthesis routes, optimizing waste-to-product conversion efficiency and scalability, and engineering derivatives for multifunctional performance, thus bridging the gap between commodity-scale use and high-tech material innovation.

Keywords: biorefinery; polysaccharides; cellulose; hemicellulose; etherification; esterification



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1. Introduction

In the search for sustainable and high-performance materials, the chemical modification of natural polysaccharides from lignocellulose, namely cellulose and hemicellulose, has emerged as a cornerstone of green materials science within the biorefinery concept for the most abundant biological material on Earth [1–3]. Cellulose is a primary structural component of plant cell walls and exists as a linear polysaccharide composed of repeating β -D-glucopyranose units linked through β -1,4-glycosidic bonds [4]. Each glucose unit contains three hydroxyl groups located at the C-2, C-3, and C-6 positions, which

enable extensive intra- and intermolecular hydrogen bonding that produces highly ordered crystalline regions interspersed with amorphous domains, contributing to its rigidity, insolubility in water, and resistance to most solvents [5]. Ester and ether derivatives of cellulose are well-established industrial products with wide global commercial applications due to their tunable solubility, film-forming and thickening ability, as well as biocompatibility and biodegradability, supporting a wide array of applications in pharmaceuticals, food, coatings, oil-well drilling, and textile fields [3]. The chemical structure of cellulose and hemicellulose is presented in Figure 1.

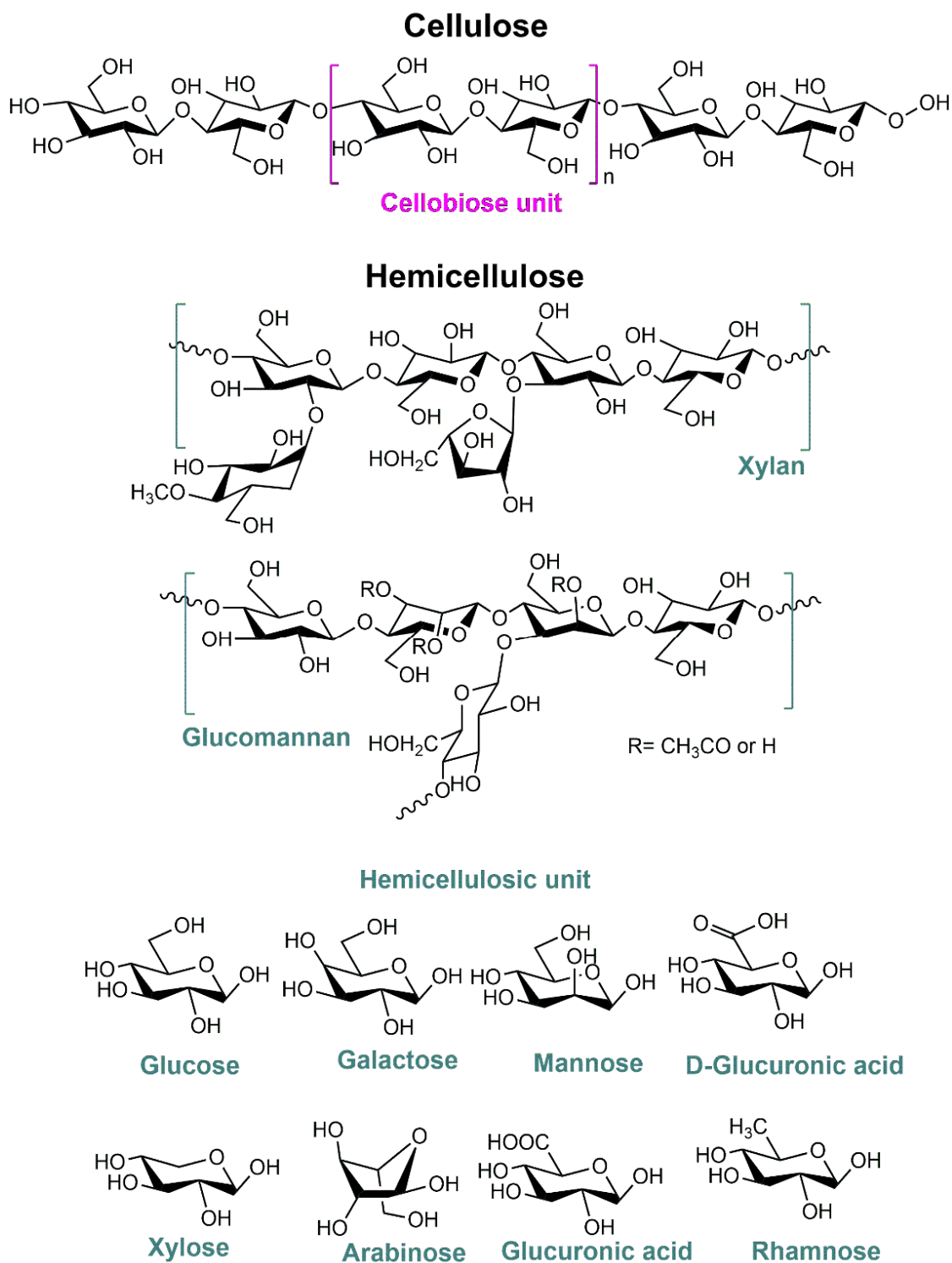


Figure 1. Chemical structure of cellulose and hemicellulose polysaccharides.

A transformative frontier in this field is the valorization of hemicellulose, a structurally amorphous, branched, heterogeneous polysaccharide composed of pentoses and hexoses (xylans, mannans, glucomannans with variable branching, acetylation, and uronic acids), containing hydroxyl groups at the C-2, C-3, and/or C-6 positions (Figure 1), often left unused in the pulp and biomass processing industry, through conversion into esters and ethers [6,7]. Recent studies have demonstrated the viability of producing xylan derivatives with excellent film-forming properties, moisture-barrier properties, and thermoplastic behavior. These advances support fully integrated biorefinery strategies, wherein cellulose, hemicellulose, and lignin are effectively converted into high-value functional materials such as packaging films, adhesives, hydrogels, and scaffolds [2,6]. Despite the industrial success of cellulose derivatives, hemicellulose derivatives remain underdeveloped due to their structural heterogeneity, variable composition, lower degree of polymerization, and difficulties in controlling dissolution, reactivity, and substitution patterns [8]. While cellulose derivatives benefit from well-established, scalable chemistries, hemicellulose derivatives have mostly been explored at the laboratory scale, with limited pilot- or industrial-scale applications [6].

The industrial relevance of cellulose and hemicellulose derivatives is underscored by robust market growth. The global market for cellulose esters and ethers reached approximately USD 7.84 billion in 2023 and is expected to nearly double to USD 15.40 billion by 2030, expanding at a CAGR of around 10.1% from 2024 to 2030 [9]. These figures reflect not only sustained demand in traditional sectors such as coatings, pharmaceuticals, and food, but also indicate rising interest in novel technological applications. Recent research has increasingly focused on valorizing waste lignocellulosic residues, such as agro-food byproducts, paper mill waste, and pulp processing streams, as cost-effective and sustainable feedstocks for ester and ether derivatization [10,11]. Lignocellulosic biomass is produced globally at a scale of approximately 182 billion tons per year, yet more than 90% of this material is currently underutilized by industry [12]. Furthermore, advanced delignification approaches enhance the accessibility of hydroxyl groups in the polysaccharide matrix, enabling more efficient and selective esterification and etherification of both cellulose and hemicellulose [13,14]. These innovations not only unlock the full potential of lignocellulosic biomass but also reinforce a paradigm shift toward resource-efficient feedstock utilization and circular bioeconomy frameworks in materials research.

The extraction and valorization of cellulose and hemicellulose generally require pre-treatment, given their intricate association with lignin and the strong hydrogen bonding within their structures. A wide range of strategies has been developed, often grouped into physical, chemical, biological, and hybrid routes. These processes aim to obtain polymers with high yield and purity, while ensuring conversion efficiency, recovery of all biomass components, and overall sustainability [15]. Physical approaches such as milling, centrifugation, hammering, extrusion, vibration, microwave, and ultrasound treatments reduce particle size and improve accessibility without generating liquid chemical waste, representing an environmentally sustainable option [16,17]. Chemical treatments—including alkaline or acidic hydrolysis, ozonolysis, oxidation, ionic liquids (ILs), deep eutectic solvents (DESs), renewable solvents such as ethyl lactate, and molten salt hydrates (MSHs)—stand out for their high effectiveness under relatively mild temperatures and pressures, while also offering potential environmental compatibility [6,18,19]. Acid hydrolysis is particularly effective for cleaving lignin–hemicellulose linkages in softwoods and agro-residues with high hemicellulose content, whereas alkaline treatments are advantageous for woody biomass, promoting matrix swelling, crystallinity reduction, and solubilization of both hemicellulose and portions of lignin. Post-treatment ethanol washing further enhances the purity of recovered hemicellulose [19]. Biological methods involving bacteria, fungi,

algae, or genetically engineered microorganisms represent highly sustainable alternatives, with low energy input and minimal hazardous waste generation, although often slower and less effective as stand-alone methods [20,21].

In parallel, hybrid physicochemical approaches such as steam explosion, ammonia fiber expansion (AFEX), liquid hot water hydrolysis (LHW), supercritical fluid extraction (SFE), autohydrolysis, and microwave-assisted extraction are widely used at pilot and industrial scales because of their relatively short processing times, moderate energy requirements, and lower capital costs [22–26]. For instance, hot-water autohydrolysis achieves selective hemicellulose solubilization at near-optimal pH $\approx$ 3.5 while preserving lignin, whereas steam explosion and microwave-assisted methods enhance the accessibility of both cellulose and hemicellulose. Each method presents trade-offs: chemical and thermal treatments ensure high efficiency but may generate byproducts requiring detoxification, while biological and physical approaches are greener yet often less efficient when applied alone.

Following extraction, the conversion of cellulose and hemicellulose into derivatives via esterification or etherification represents a critical step toward functional materials. Cellulose and hemicellulose esters and ethers are the two most widely explored classes of lignocellulosic derivatives, attracting increasing interest as functional and sustainable materials with improved physicochemical properties and broad industrial applicability [27–29], since modification provides tailored solubility, stability, and functional properties, supporting applications across materials, food, biomedical, packaging, and environmental sectors [6,8,30,31]. Cellulose esters such as cellulose acetate (CDA, CTA), cellulose acetate butyrate (CAB), cellulose acetate propionate (CAP), cellulose nitrate (CN), cellulose sulfate (CS), and cellulose benzoate (CB) have long been employed in biodegradable films, coatings, textiles, and photographic materials, as well as in pharmaceutical and food formulations, due to their excellent film-forming ability, strength, and chemical resistance [32]. Similarly, hemicellulose esters (e.g., acetates, propionates) enhance hydrophobicity, processability, and film-forming properties, enabling applications in bio-based packaging, coatings, hydrogels, and controlled-release biomedical systems [33–35].

Cellulose ethers such as methylcellulose (MC), ethylcellulose (EC), hydroxyethyl cellulose (HEC), hydroxypropyl cellulose (HPC), carboxymethylcellulose (CMC), and hydroxypropyl methylcellulose (HPMC) are produced through etherification of hydroxyl groups, with their solubility and strength governed by the degree of substitution (DS) and the type of substituent [3,36]. They find widespread use as stabilizers, thickeners, and film-formers in pharmaceuticals, cosmetics, and construction materials [6]. Analogously, hemicellulose ethers (e.g., carboxymethyl, methyl, hydroxyethyl, cyanoethyl) are produced via alkylation or hydroxyalkylation, improving solubility, flexibility, and functionality. For instance, carboxymethyl hemicellulose acts as a water-soluble polyelectrolyte suitable for drug delivery and thickening, while hydroxyalkyl derivatives are valued in adhesives, coatings, and personal care products [25,37].

These cellulose and hemicellulose derivatives are commonly characterized using a set of analytical techniques, with particular emphasis on determining the degree of substitution (DS), a key factor influencing their properties. These methods include NMR spectroscopy (^1H and ^{13}C) for detailed substitution patterns, FTIR for rapid functional group analysis, GC-MS for quantifying ester or ether groups after derivatization, and HPLC for analyzing complex substitution distributions. Size-exclusion chromatography (SEC), dynamic light scattering (DLS), and viscosity measurements are valuable for assessing molecular weight, solubility, particle size, surface charge, and polymer hydrodynamics affected by substitution. Combining these methods enables accurate DS evaluation and

comprehensive characterization, which is crucial for optimizing their performance in diverse applications [38–40].

Traditionally, the majority of these reactions are carried out under heterogeneous conditions, where cellulose remains in solid form and only its amorphous regions are accessible. Under alkaline activation, sodium hydroxide promotes fiber swelling, reduces crystallinity, and enhances hydroxyl reactivity, enabling nucleophilic substitution through an S_N2 mechanism [41]. While this approach is simple and widely used, it often leads to low degrees of substitution (DS), poor product uniformity, and limited solubility due to incomplete modification of crystalline regions [42]. To overcome these drawbacks, homogeneous modification has been developed, made possible by advanced solvents capable of dissolving cellulose at the molecular level. Ionic liquids (ILs), deep eutectic solvents (DESs), and inorganic molten salt hydrates (MSHs) are alternative solvent systems that have attracted significant interest for the dissolution and conversion of cellulose and hemicellulose [43].

ILs are salts composed of bulky organic cations and various anions with low lattice energy, which enables them to remain liquid at or near room temperature. Specifically, 1-ethyl-3-methylimidazolium acetate ([Emim][OAc]), 1-butyl-3-methylimidazolium chloride ([Bmim][Cl]), and 1-allyl-3-methylimidazolium chloride ([Amim][Cl]) can disrupt the strong hydrogen-bonding network within cellulose and hemicellulose, effectively dissolving these polysaccharides and facilitating further chemical transformations [44]. DESs are formed by mixing a hydrogen bond donor (e.g., urea, glycerol, or organic acids) and a hydrogen bond acceptor (e.g., choline chloride), resulting in a liquid with unique solvation properties at relatively low temperatures [45]. Common DESs include choline chloride–urea, choline chloride–glycerol, choline chloride–lactic acid, and choline chloride–oxalic acid, which destabilize hydrogen bonding within biomass, promoting solubilization and reactivity [44]. MSHs, such as molten zinc chloride or lithium chloride hydrates, are high-temperature ionic media that strongly interact with hydroxyl groups of polysaccharides, leading to their dissolution and depolymerization [46]. Collectively, these solvent systems provide tailored ionic environments that weaken hydrogen bonding and crystalline order, making cellulose and hemicellulose more accessible for chemical modification, hydrolysis, or conversion into value-added products [47].

These systems allow regioselective substitution of hydroxyl groups with significantly higher DS and uniformity, thus providing precise control over polymer properties, such as solubility, flexibility, and thermal stability, making the derivatives more suitable for industrial applications [48]. Among the solvent systems, ILs are particularly effective due to their ability to disrupt hydrogen bonding networks and dissolve even highly crystalline cellulose, although their high cost, potential toxicity, and limited recyclability remain significant barriers to large-scale deployment. DESs, in contrast, are low-cost, biodegradable, and more sustainable, but challenges remain regarding efficiency, reproducibility, and solvent recovery [49].

The advances in extraction and chemical modification illustrate a continuous evolution from conventional heterogeneous routes with limited reactivity to innovative homogeneous systems offering higher efficiency and tunability. Nevertheless, industrial implementation still faces challenges, including solvent recovery, cost reduction, and scale-up feasibility. Future progress will depend on integrating greener solvents and hybrid pretreatment strategies to enable efficient and sustainable valorization of polysaccharides into high-value-added products. In this context, this review aims to provide a comprehensive state-of-the-art overview of the feedstocks, production, and technological applications of ether and ester derivatives of cellulose and hemicellulose biopolymers. Particular attention is given to industrially relevant compounds and promising emergent derivatives that

combine renewable origin with performance properties competitive with or superior to their fossil-based counterparts. The main properties and applications of ethers and esters from cellulose are presented in Figure 2, and the main features of hemicellulose derivatives are shown in Figure 3. Anhydroglucose units are employed in the figures to simplify the visualization of the synthetic routes.

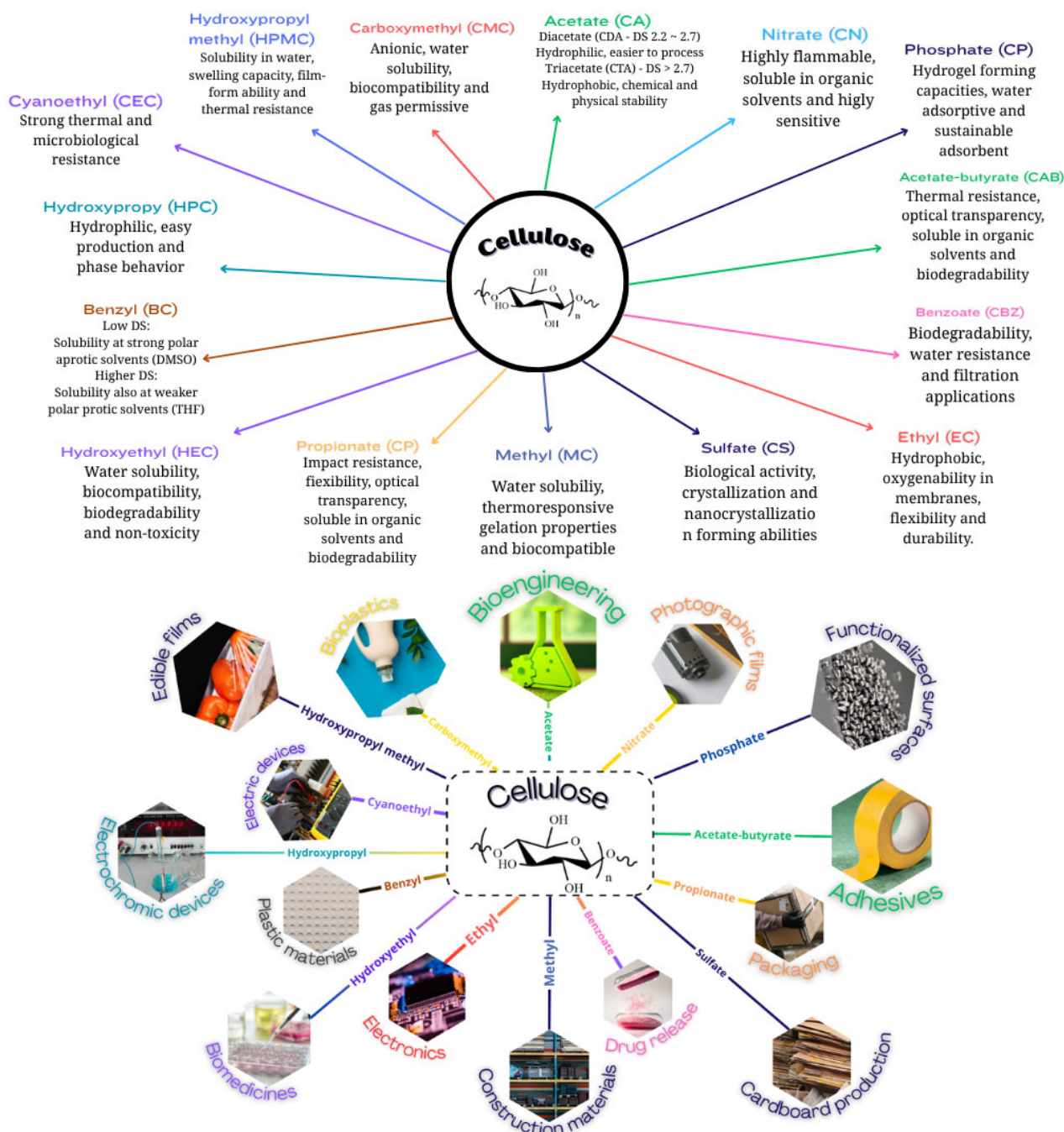


Figure 2. Properties and applications of the main cellulose ethers and esters.

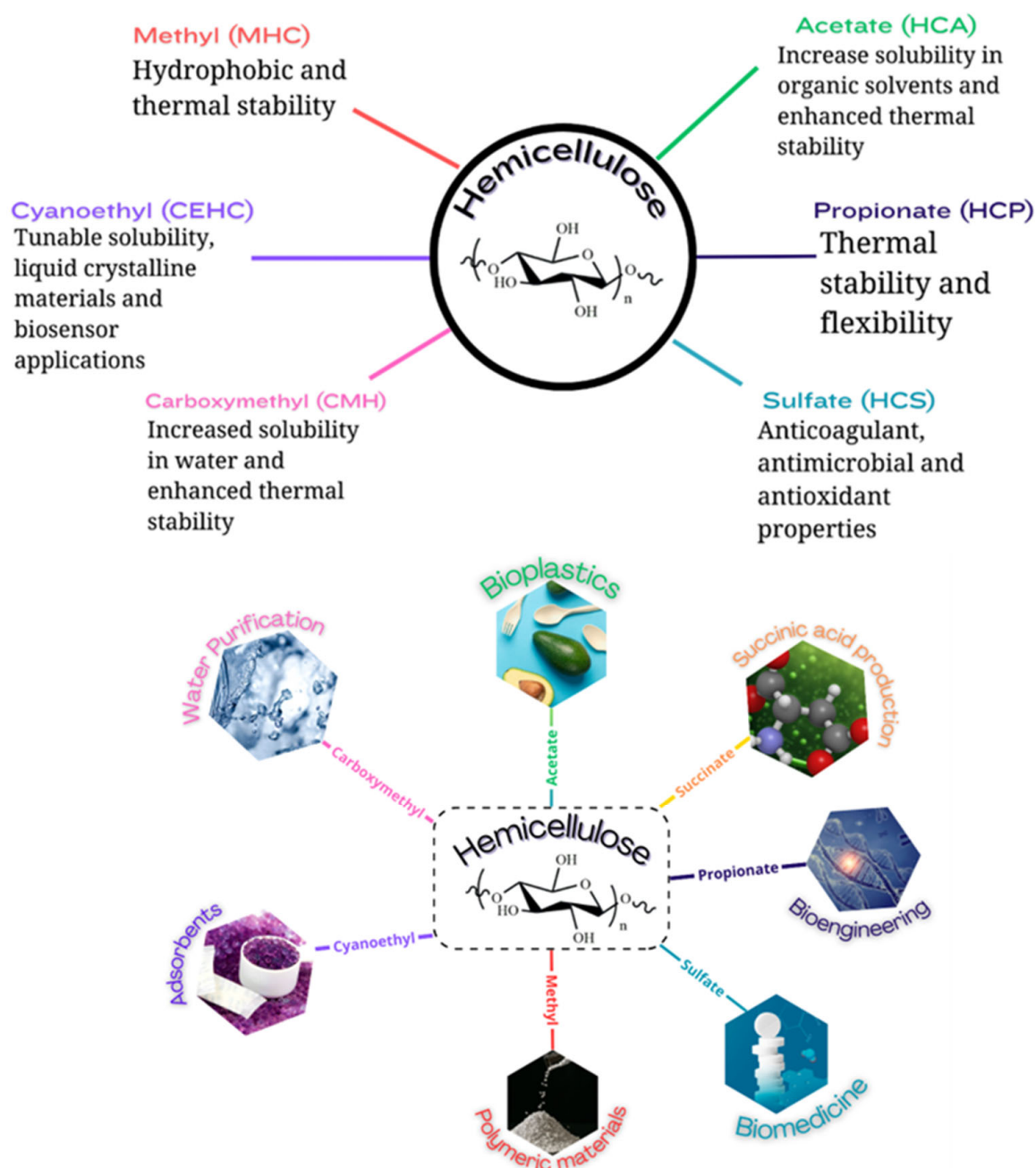


Figure 3. Properties and applications of the main hemicellulose ethers and esters.

2. Cellulose Esters

Cellulose esters are among the most important derivatives of cellulose, obtained through esterification of hydroxyl groups present in the polymer backbone. This modification improves solubility, thermal stability, and processability, expanding their applicability compared to native cellulose. Common examples include cellulose acetate, cellulose nitrate, and cellulose propionate, each with specific physicochemical properties tailored for different uses. These materials are widely employed in coatings, films, membranes, and drug delivery systems due to their biodegradability and mechanical strength. Additionally, cellulose esters can serve as renewable alternatives to petroleum-based polymers, contributing to sustainable material development. Their versatility makes them key materials in industries ranging from packaging and textiles to pharmaceuticals and biomedical engineering. The reaction pathways for the production of polysaccharide esters are illustrated in Figure 4.

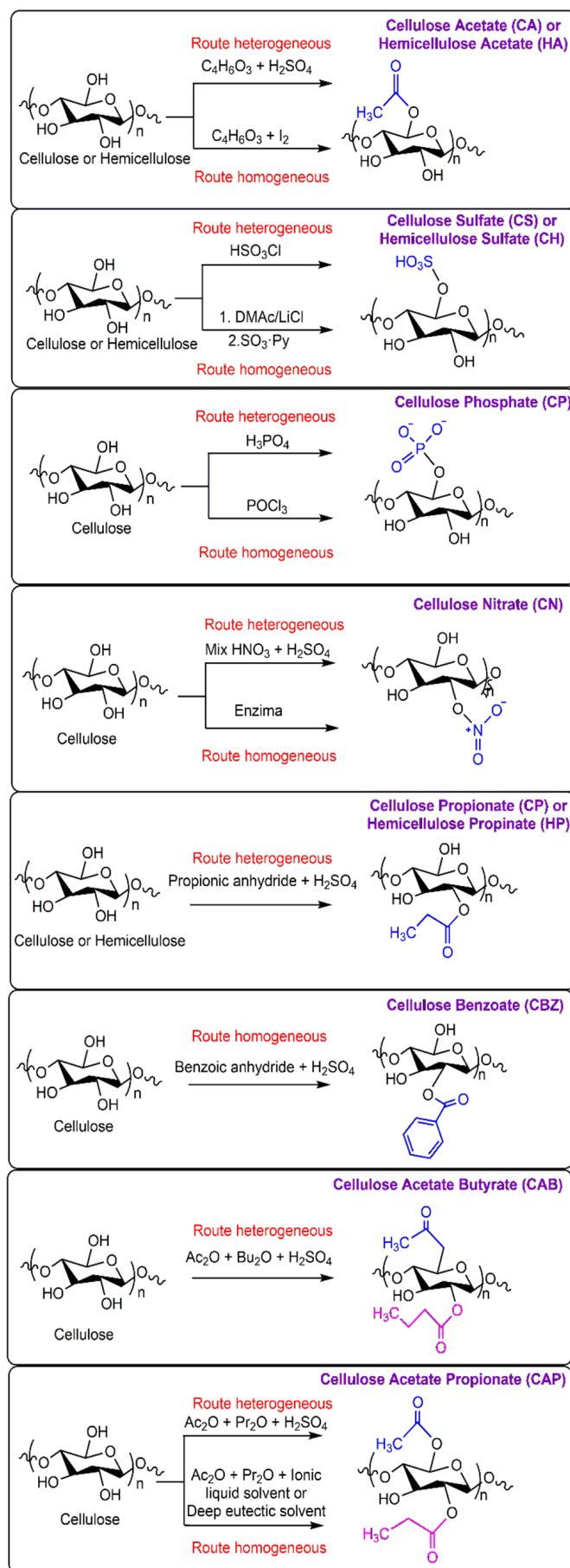


Figure 4. Reaction pathways for the production of the polysaccharide esters discussed in this review.

2.1. Cellulose Acetate Propionate (CAP)

Cellulose acetate propionate (CAP) is a cellulose-derived copolymer obtained by the esterification of hydroxyl groups in the cellulose backbone with both acetate ($-\text{COCH}_3$) and propionate ($-\text{COC}_2\text{H}_5$) substituents. This dual substitution imparts a combination of high impact resistance, flexibility, optical transparency, solubility in organic solvents, and biodegradability, which makes CAP an attractive alternative to conventional petroleum-based polymers in various industrial and technological applications [27,29]. The synthesis of CAP was first reported in the 1940s, initially aimed at producing coatings, films, and adhesives for the packaging and automotive sectors. Over the decades, advances in catalyst design, feedstock selection, and reaction optimization have improved the efficiency and sustainability of its production. The production process involves the esterification of cellulose with acetic anhydride and propionic anhydride, catalyzed by sulfuric acid, resulting in polymer formation and the release of acetic and propionic acids as byproducts. In the literature, this copolymer is widely applied when produced from commercial cellulose, as reported in studies by Gao et al. (2022), Martín-Alfonso et al. (2024), and Gao et al. (2023) [27–29]. From a structural perspective, CAP retains the β -1,4-linked D-glucopyranose units of cellulose but with varying degrees of substitution by acetyl and propionyl groups, influencing its thermal and mechanical performance.

The CAP market was valued at around USD 250 million in 2024 and is projected to reach USD 400 million by 2033, reflecting a 5.5% CAGR between 2026 and 2033 [50]. CAP represents an expanding niche in the global chemical sector, largely fueled by rising interest in biodegradable and eco-friendly alternatives. As a cellulose-based derivative, CAP is recognized for its optical clarity, thermal stability, and broad solvent compatibility. This thermoplastic material is utilized across diverse industries, including coatings, films, and photographic products, and also plays a role in the tobacco industry through cigarette filter manufacturing [50]. Traditionally, CAP is synthesized from high-purity commercial cellulose sources, such as wood pulp or cotton linters, due to their high crystallinity and low content of non-cellulosic impurities. Despite growing interest in circular bioeconomy approaches, the direct use of agricultural residues or other lignocellulosic wastes as feedstock for CAP synthesis remains underexplored, largely due to challenges in achieving sufficient cellulose purity and controlling the degree of substitution during functionalization [51].

The most common synthetic pathway for CAP involves acid-catalyzed esterification using a mixture of acetic anhydride and propionic anhydride in the presence of concentrated sulfuric acid, producing the copolymer along with acetic and propionic acids as byproducts. However, more sustainable and selective synthetic routes have emerged in recent years. Wen et al. (2021) reported the use of a core-shell heterogeneous catalyst ($\text{PS@PMA-ZrO}_2\text{-PW}_{12}$), which enabled high molecular weight CAP ($\approx 100,644$ Da) with a degree of substitution of 2.73, excellent transparency ($>90\%$), and flexibility (elongation at break $\sim 32.7\%$), all achieved without the need for plasticizers. Another promising strategy involves the immobilization of heteropolyacids on zirconia-polymethacrylate microspheres, offering a greener and more easily recoverable catalyst system while maintaining high product quality and minimizing acidic effluent generation [51].

In terms of applications, the unique balance of toughness, flexibility, and chemical resistance has led to its use in diverse sectors. In the coatings and inks industry, CAP serves as a binder and rheology modifier, enhancing adhesion, blocking resistance, and solvent release, particularly in flexographic inks and overprint varnishes. In food packaging, it is valued for its clarity and ability to form moisture-resistant and non-yellowing films [52]. Emerging applications include its integration into hydrogels for controlled-release systems, where CAP matrices can modulate the release rate of active compounds such as pesticides or pharmaceuticals, and composite formulations for electronics and biodegradable

structural components [53]. These features, combined with its partial biodegradability and compatibility with other biopolymers, position CAP as a key candidate for the development of high-performance, sustainable materials.

Although catalysis represents an effective and sustainable strategy for selectively obtaining this derivative, alternative routes have also gained prominence. Synthesis in ionic liquids, for example, allows the exploitation of their unique physicochemical properties, such as low vapor pressure, high thermal and chemical stability, and the ability to modulate catalytic activity and molecular selectivity, thus contributing to more efficient and reusable processes [47,54]. The derivatization of native cellulose is a challenging process, particularly due to the difficulty in solubilizing this polymer in a homogeneous medium. In this context, the main advantage of using ionic liquids as a reaction medium lies in their high capacity to disrupt the extensive network of hydrogen bonds present in cellulose, promoting its complete dissolution and, consequently, enabling more efficient derivatization. Highlighting this potential, Gao et al. (2025) synthesized cellulose acetate with a high degree of substitution ($DS = 2.82$) using the ionic liquid $[DBUC_8]Cl$. According to the authors, this ionic liquid has a dual function in the process, acting simultaneously as a reaction medium and a catalyst, which enhances the efficiency of cellulose transesterification and eliminates the need for additional catalysts [55].

Despite their promising properties, the widespread adoption of ionic liquids in cellulose derivatization processes still faces economic and methodological limitations due to their high cost and complexity in synthesis, as well as toxicity issues, a determining factor in their rejection on an industrial scale. These obstacles have driven the search for more affordable and environmentally safe alternatives, notably deep eutectic solvents (DESs), which combine low environmental impact, ease of production, and the potential to play a similar role in cellulose dissolution and modification [56]. Zhou et al. (2025) prepared cellulose acetate with degrees of substitution, DS , of $0.42 < DS < 1.81$, using a deep eutectic solvent composed of zinc chloride/phosphoric acid/water ($ZnCl_2/PA/H_2O$). As in the case of ionic liquids, the role of DESs in obtaining cellulose derivatives also focuses on the solubilization of the biopolymer. The authors point out that, if the raw material used is in the form of fibers derived from mixed fabrics, DESs can selectively solubilize it, preserving the other components [57]. Tian et al. (2025) introduced an innovative approach to the study of deep eutectic solvents (DESs), elucidating the bifunctionality of acidic active sites in metal-based systems. The high acidity of these systems significantly increased the interaction between bamboo fibers and the $ZnCl_2$ -lactic acid solvent, resulting in greater efficiency in the pulp purification step using the biomass adopted in the study. This route yielded cellulose acetate with a high degree of substitution ($DS = 2.84$) and a yield over 70%, demonstrating the system's potential for cellulose derivatization processes [58].

When comparing these studies, notable differences arise regarding feedstock origin and processing conditions. For instance, while Yahaya et al. (2025) employed purified cellulose from commercial sources, achieving high substitution degrees under relatively mild reaction conditions, other approaches using lignocellulosic waste feedstocks required more intensive pretreatment steps to remove residual lignin and hemicellulose before esterification. Such differences directly influence both reaction efficiency and the physicochemical profile of the final propionate derivatives, with waste-derived samples often exhibiting broader molecular weight distributions and slightly reduced thermal stability. This derivative is produced on an industrial scale for applications in films, automotive plastics and accessories, paints, and coatings [59]; however, reports on CAP production from waste biomass, as well as on the synthesis of an analogous polymer from hemicellulose, have not been reported in the literature and represent opportunities for innovation.

Studies point to CAP as an essential component for the development of advanced and eco-efficient polymer electrolytes. Gao et al. (2023) presented a novel gel polymer electrolyte (GPE) composed of polyimide (PI) and cellulose propionate acetate (CAP), manufactured by electrospinning. The combination of these materials resulted in a nanofibrous membrane with superior properties, such as high ionic conductivity and excellent thermal stability (above 200 °C), mechanical strength, and lithium-ion transport capacity. The PI–CAP-based GPE demonstrated superior performance in lithium batteries, with 95% capacity retention after 300 cycles and efficient inhibition of lithium dendrite growth. The incorporation of CAP into the PI-based GPE resolved critical issues such as poor wettability and mechanical strength, while improving the electrochemical performance and safety of lithium batteries [60].

In recent years, the production of CAP has advanced from conventional acid-catalyzed esterification to greener routes employing heterogeneous catalysts, ILs, and DESs. Traditional methods remain dominant for their scalability, but emerging strategies offer higher substitution degrees, better performance, and reduced environmental impact. ILs excel in cellulose dissolution and dual catalytic roles, though cost and recyclability limit industrial use, while DESs provide a safer and more economical option, albeit with challenges in selectivity. From purified cellulose to lignocellulosic residues, feedstock choice balances product uniformity with sustainability goals. Beyond established roles in coatings, films, and plastics, CAP shows growing promise in advanced sectors, particularly energy storage and biomedicine.

2.2. Cellulose Sulfate (CS)

Cellulose sulfate (CS) is produced by chemical substitution of hydroxyl groups (–OH) on cellulose with sulfate groups (–OSO₃H), releasing water as a byproduct. The properties of CS, such as solubility, charge density, and biological activity, depend strongly on the sulfation method used. The most common synthesis routes include heterogeneous, homogeneous, and acetosulfation (quasi-homogeneous) methods, each with advantages and challenges. Heterogeneous sulfation involves reacting solid cellulose with sulfating agents like chlorosulfonic acid or sulfur trioxide complexes in a non-solvent medium. Because cellulose remains mostly insoluble, sulfation mainly affects the surface hydroxyl groups, leading to lower degrees of substitution (DS) and less uniform substitution patterns. This method requires precise temperature control to avoid degradation and is preferred for simpler and cost-effective production of low-DS CS for applications where extreme purity or uniformity is not required [61–63]. Recent innovations also explore microwave-assisted sulfation, which accelerates the reaction and improves DS control by uniform heating and enhanced reagent diffusion within cellulose fibers [64,65]. Furthermore, enzymatic pretreatment of cellulose with cellulases prior to sulfation enhances reagent accessibility by selectively hydrolyzing amorphous regions, thus enabling more efficient and regioselective sulfation [66–68].

Homogeneous sulfation in a medium dissolves cellulose in solvents such as DMAc/LiCl or ILs, allowing the sulfating agents to uniformly react with all accessible hydroxyls on the cellulose chain (positions C2, C3, and C6). This technique yields higher DS values and more consistent substitution profiles, enabling fine control over CS properties such as solubility and charge density. The major drawback lies in the need for specialized solvents and complex purification steps, which increase production cost and complexity [69,70]. Acetosulfation is a regioselective, two-step process where cellulose is first acetylated at the C2 and C3 hydroxyls to protect these positions, followed by sulfation primarily at the C6 hydroxyl. This enables high DS values with defined substitution sites, making CS highly

suitable for biomedical applications requiring specific chemical functionalities. However, the multi-step procedure increases reaction time and complexity [71–73].

Recent studies, such as Johnson et al. (2024), investigated the synthesis of cellulose sulfate nanofibers (CSNFs), which were achieved from raw jute fibers via a heterogeneous sulfonation route using chlorosulfonic acid (CSA) in *N,N*-dimethylformamide (DMF). In this process, CSA simultaneously promotes partial pulping of the lignocellulosic matrix and introduces sulfate ester groups, which preferentially bind to the primary hydroxyls at the C6 position of the anhydroglucose units. The successful incorporation of sulfate functionalities was confirmed by FTIR (bands at 1204 and 813 cm^{-1}) and elemental sulfur analysis, which showed a progressive increase in sulfur content from 2.2 to 6.2% as the CSA dosage increased. Mass yields decreased from 54.1% (at the lowest CSA concentration) to 44.2% (at the highest), reflecting chain scission, glycosidic bond cleavage, and crystallinity reduction, as further supported by WAXD analysis. Although the degree of substitution (DS) was not explicitly reported, solid-state NMR spectroscopy indicated predominant functionalization at primary hydroxyl groups, consistent with typical cellulose sulfate derivatives. The resulting CSNFs exhibited remarkable performance in aqueous ammonium ion removal, achieving a maximum adsorption capacity of 41.1 mg/g according to the Langmuir isotherm model, a value superior to conventional sorbents such as zeolites, biochar, and carboxylated nanocellulose. Moreover, their adsorption efficiency remained stable over a broad acidic pH range (2.5–6.5), with zeta potential analysis confirming the strong anionic character conferred by sulfate groups. Beyond their demonstrated use in ammonium removal, cellulose sulfate nanofibers represent a versatile platform for ion-exchange systems, hydrogel formulations, and sustainable adsorbent technologies, positioning them as strategic materials for green chemistry and advanced environmental remediation [65].

Jokar et al. (2022) used cotton as a raw material for CS production via sulfonation. In their study, the biopolymer was impregnated with palladium to enable nanocatalysis aimed at reducing Cr^{4+} in contaminated water in Iran [74]. Given its broad applicability in industry, several other studies, including those by Li et al. (2021), Romanchenko et al. (2015), and Wu et al. (2019), have explored the use of this polymer derived from residual sources such as cotton, paper, wood, and other biomass to develop innovative materials with promising applications across multiple industrial sectors [75–77]. Among recent contributions in this area, Ma et al. (2023) investigated cellulose sulfation using a ternary deep eutectic solvent composed of sulfamic acid, urea, and choline chloride. This approach enabled simultaneous swelling and sulfation of cellulose, resulting in greater energy efficiency in the synthesis process. The method is innovative not only due to its operational simplicity but also because of its efficient use of resources, aligning with the principles of green chemistry by employing low-toxicity solvents that can be recovered and subsequently reused [78]. Years later, aiming to reuse cotton textile fibers, Zhao et al. (2025) synthesized sulfated cellulose nanocrystals (SCNCs) using a binary DES composed of sulfamic acid and urea. This system favored cellulose wetting and swelling, corroborating the observations regarding the efficiency of these solvents. Furthermore, changes in the intra- and intermolecular hydrogen bonding networks of the cellulose chains were identified, resulting in the formation of nanoscale particles [79].

Among its numerous applications, Su et al. (2023) describe the synthesis and application of lithium cellulose sulfate (CSL) as a conductive binder for the LiFePO_4 (LFP) cathode in lithium-ion batteries, highlighting its superior performance compared to the conventional polyvinylidene fluoride (PVDF)-based binder. The sulfate groups present in cellulose provide mobile Li^+ ions and facilitate the diffusion of these cations into the cathode, ensuring an ionic conductivity 2.5-fold greater than that of PVDF. CSL is stable enough for

battery application and offers a superior alternative to PVDF, with longer service life, better cleavage capacity, and less interfacial degradation. Lithium cellulose sulfate combines high ionic conductivity, excellent adhesion, and sustainability. This work paves the way for the use of polysaccharide derivatives in high-performance lithium batteries [64].

CS has been applied in industrial hydrogels, commercial pharmaceuticals, and scientific research in controlled drug release systems. It plays an important role in the encapsulation sector due to its mechanical strength, stability, and capacity to form semi-permeable membranes that enable nutrient exchange while preserving cell viability [80]. The use of CS is particularly significant in systems aimed at long-term cell survival, with increasing interest in drug delivery and regenerative medicine, both key areas driving the development and application of new CS-based materials [66,81,82]. In the energy field, this derivative is often transformed into cellulose sulfate lithium (CSL) by treating the acidic form of CS with a lithium-containing base, commonly lithium hydroxide (LiOH) or lithium carbonate (Li_2CO_3) [64,83]. This material acts as a multifunctional polyelectrolyte binder for Li-ion batteries and can improve fast charging capability. Eco-friendly separators for Zn-ion aqueous batteries have also been produced with CS, as reported by Yan et al. (2025) for the CS produced from bacterial cellulose. The 50 μm sulfonated separator shows high tensile strength (167 MPa) and ionic conductivity (13.1 mS cm^{-1}). Incorporation of sulfonate groups into the cellulose matrix suppresses sulfate ion migration, enhances zinc-ion transport, and prevents dendrite growth. A 1.2 Ah pouch cell with high cathode loading (21.6 mg cm^{-2}) was achieved, demonstrating its promise for scalable and durable aqueous batteries [84].

Cellulose sulfate (CS) stands out for the versatility of its sulfation routes, which allow fine control over substitution and structural features. This chemical flexibility has enabled the design of tailored materials that perform reliably in demanding contexts, from long-term cell encapsulation to ion-selective membranes in batteries. Beyond synthetic improvements, the critical challenge lies in linking structural precision to predictable performance across different application scales, a point that will determine how CS advances from promising laboratory results to mature technologies.

2.3. Cellulose Phosphate (CP)

Cellulose phosphate (CP) is a cellulose-derived polymer obtained through the substitution of hydroxyl groups ($-\text{OH}$) with phosphate groups ($-\text{OPO}_3\text{H}_2$), which imparts the material with unique properties such as water solubility, biodegradability, and biocompatibility [85,86]. These characteristics make cellulose phosphate a promising material for various applications, particularly in the biomedical field, including tissue engineering scaffolds, controlled drug-delivery systems, and functional hydrogels [85–90]. The reaction mechanism involves the phosphorylation of cellulose using phosphoric acid or phosphorylating agents such as phosphorus pentoxide and phosphoryl chloride, under controlled temperature and reaction time [85,89]. Phosphoric acid serves both as a reagent and as the reaction medium, promoting the efficient conversion of cellulose into its phosphorylated form [85,89]. Water is released as a byproduct during this process, and the degree of substitution can be tailored to meet the specific requirements of each application [86,88].

Recent studies have highlighted the versatility of cellulose phosphate in various technological fields, such as bone tissue engineering and bio-inks for regenerative medicine, thereby expanding its utility beyond traditional domains. In bone tissue engineering, phosphate-functionalized cellulose scaffolds support biomineralization and osteoconductivity; in bio-inks, phosphate-modified cellulose enables enhanced cell support and printability for bioprinting applications [85–87,90,91]. In addition, phosphorylated cel-

lulose nanofibers (P-CNF) have been reported as a promising eco-friendly alternative for organic fillers and ion-sieving barriers with flame-retardant properties in Li-ion batteries [92,93].

The reaction mechanism for producing CP involves the phosphorylation of cellulose using phosphoric acid or phosphorylating agents such as phosphorus pentoxide and phosphoryl chloride, under controlled temperature and reaction time, usually in an aqueous medium [85,89]. Phosphoric acid serves both as a reagent and as the reaction medium, promoting the efficient conversion of cellulose into its phosphorylated form [85,89]. Water is released as a byproduct during this process, and the degree of substitution can be tailored to meet the specific requirements of each application [86,88].

Rohaizu and Wanrosli (2015) investigated the synthesis of cellulose phosphate from microcrystalline cellulose extracted from empty fruit bunches of oil palm, a waste product of the palm oil industry. Derivatization was performed using orthophosphoric acid (H_3PO_4) and triethyl phosphate (Et_3PO_4) at different molar ratios, with reaction conditions adjusted to optimize phosphate group incorporation. The DS varied depending on the reagent ratio, directly influencing the polymer's physicochemical properties, such as water absorption capacity [94]. In the research developed by Abdulhameed et al. (2025), abundant rice husk waste was applied to produce crosslinked CP superabsorbent hydrogels (SAH) using an innovative rapid derivatization method under microwave irradiation. The aqueous medium containing the extracted cellulose and H_3PO_4 was heated in a microwave oven for 3 min (420 W), resulting in a SAH with high swelling capacity. The product was tested as a soil treatment for maize, with irrigation limited to the first 21 days and growth monitored over 12 weeks to simulate arid and semi-arid conditions. Rice husk SAH improved crop growth and yield, with optimal results at 5 g per pot. Treated plants continued to thrive under water stress, unlike controls, confirming its effectiveness in greenhouse cultivation [95].

Flame-retardant phosphorylated cellulose nanofibers (P-CNF) were produced from the derivatization of softwood Kraft pulp in a homogeneous medium using choline chloride/urea/ H_3PO_4 as a green reactive DES, followed by ultrasonication, in the investigation performed by Zhang et al. (2025). Reaction parameters significantly affected charge density and yield, with optimal conditions (150 °C, 2 h) achieving up to $2.91 \text{ mmol} \cdot \text{g}^{-1}$ and ~90% yield. The resulting P-CNF exhibited nanoscale dimensions (~5.1 nm width), excellent flame retardancy with an 89.1% reduction in peak heat release rate, a limiting oxygen index of 44.8%, and a 43% residual mass at 700 °C. Additionally, transparent films (>80% light transmittance) with robust mechanical strength ($86.4 \pm 13 \text{ MPa}$) and self-extinguishing behavior were obtained. This method provides a sustainable and scalable route for producing intrinsically flame-retardant nanocellulose, with promising potential in thermal insulation, electronics, and other advanced material applications [96].

In a similar approach, Gao et al. (2025) reported a one-step green synthesis of flame-retardant cellulose fabrics using a recyclable ternary deep eutectic solvent (TDES) prepared from phytic acid, sodium acetate, and urea (1:1:5 molar ratio). Cellulose fabrics ($12 \times 12 \text{ cm}^2$) were immersed in TDES at 120 °C for 30 min, washed to neutral pH, and oven-dried, yielding surface ammonium phosphate-modified cellulose fabrics (SACFs). The treated fabrics displayed excellent flame resistance, with a limiting oxygen index (LOI) of 48.0% maintained at 33.5% after 50 laundering cycles, along with sharp reductions in peak heat release rate (93.6%) and total heat release (54.0%). SACFs also showed strong antibacterial activity (99.8% inhibition of *E. coli*), enhanced dyeability with cationic dyes, and retained fire resistance post-dyeing. The TDES was recyclable for at least five cycles, with treated fabrics (SACF-X) maintaining high performance, highlighting this simple,

eco-friendly process as a scalable route for multifunctional cellulose textiles in protective and sustainable applications [97].

An innovative mechanochemical synthesis process for CP was recently reported by Gao et al. (2025). This work introduces a solvent-free and heat-curing method for producing highly charged and crystalline phosphorylated cellulose nanocrystals (P-CNCs). Using ball milling of MCC, P_2O_5 , and urea (1:0.5:5) at 650 rpm for 30 min, followed by curing at 150 °C for 20 min, washing, and homogenization, the process yielded P-CNCs with ultrahigh charge density (4.03 mmol g⁻¹) and high crystallinity (76.3%). Liquid-state ³¹P NMR confirmed multiple phosphate structures (mono-, poly-, and crosslinked). The method achieved lower energy consumption (0.338 vs. 0.565 kWh g⁻¹) and reduced environmental impact compared to aqueous-based or heat-soaking phosphorylation, offering a scalable, eco-friendly route for industrial P-CNC production [98].

Despite its high technological potential, industrial-scale production of cellulose phosphate remains limited, currently targeting niche sectors in the biomedical and pharmaceutical industries [99]. Only a few biorefineries conduct its synthesis on a large scale due to challenges associated with controlling the degree of substitution and the economic feasibility of the process. However, the growing interest in sustainable and functionalized materials is driving research in this area, indicating significant potential for expanding its applications in industrial hydrogels and advanced biomaterials. The innovations in mechanochemical and green solvent phosphorylation routes make it possible to explore more efficient and sustainable production methods from lignocellulosic waste sources in biorefineries [100].

2.4. Cellulose Acetate (CA)

Cellulose acetate is synthesized through the acetylation of cellulose, generally employing acetic anhydride and sulfuric acid in an acetic acid medium. Based on the degree of substitution (DS), it is classified into cellulose diacetate (CDA), with DS values typically between 2.2 and 2.7, and cellulose triacetate (CTA), with DS exceeding 2.7. The DS strongly influences the physicochemical behavior, processability, and industrial applicability of the material [101]. CDA, which has a lower acetyl content, is more hydrophilic and easier to process, finding use in filtration membranes, biodegradable packaging, plastics, and some textile products [102]. CTA, by contrast, is more hydrophobic and offers enhanced thermal resistance and chemical stability, making it suitable for applications in films, photographic media, linings, and high-performance sportswear.

Advances in nanostructured cellulose acetate have further expanded its applications to tissue engineering, drug delivery platforms, and wound-healing materials [103,104]. The development of cellulose acetate dates back to the late 19th century, when it emerged as a safer, less flammable alternative to cellulose nitrate [105,106]. Initially used for photographic film, textile fibers, and eyeglass frames, the material later evolved into cellulose triacetate to meet the need for heat-resistant products, such as high-performance photographic films and cigarette filters. Today, both CDA and CTA are typically produced via esterification of cellulose with acetic anhydride, catalyzed by sulfuric acid. Globally, cellulose acetate products hold significant commercial value, with 2024 market estimates of approximately USD 1.5 billion for CDA and USD 1.2 billion for CTA, both projected to grow at a CAGR of 5.2% from 2026 to 2033 [107,108].

Sustainable production approaches have recently gained attention, with research exploring agro-industrial residues as feedstocks. For instance, Das et al., 2014 obtained cellulose acetate from sugar palm stem fibers using iodine-catalyzed acetylation, producing a DS suitable for bioplastic manufacture [109]. Similarly, Shaikh et al. (2022) and Bamba et al. (2023) synthesized CTA from date palm trunk and cocoa pod husks, respectively, with

thermal characteristics comparable to conventional products [110,111]. Structurally, CDA and CTA are cellulose esters in which hydroxyl groups (-OH) are replaced by acetate groups (-COCH₃). In CDA, substitution is partial, whereas in CTA it is nearly complete, leading to notable differences in structural organization and physical properties [112–114]. De Freitas et al. (2017) demonstrated an environmentally friendly route by converting corn waste into CTA with a DS of 2.95 using ionic liquids [113]. Both exhibit favorable mechanical and thermal strength, optical clarity, biodegradability, and solubility in solvents such as acetone and chloroform [115–118]. Thanks to its higher DS, CTA provides greater resistance to heat and chemicals, making it ideal for applications where durability is critical [119,120].

The DS depends on reaction parameters such as temperature, reaction time, cellulose source, and reagent ratios [10]. In recent years, research has focused on the production of CDA and CTA from residual biomass, aiming to promote sustainability and valorize agro-industrial waste. Rodríguez-Liébaña et al. (2024) investigated cellulose acetate production using pruned olive tree residues as raw material. The process involved optimized alkaline treatment and bleaching with hydrogen peroxide, yielding purified cellulose pulp that was subsequently acetylated to produce CDA with thermal properties suitable for further processing [121]. Anwar et al. (2024) synthesized cellulose acetate from sugar palm stem fiber residues (*Arenga pinnata*) using a process that included extraction, delignification under pressurized NaOH at 50 °C, followed by bleaching with hydrogen peroxide at 70 °C. Acetylation was performed using acetic anhydride with iodine as a catalyst, producing CDA with a degree of substitution appropriate for biodegradable plastic applications [122].

In the field of cellulose triacetate, Shaikh et al. (2022) synthesized CTA from cellulose extracted from date palm trunk waste (*Phoenix dactylifera* L.), obtaining a polymer with thermal properties comparable to commercial derivatives [110]. Jia et al. (2022) employed corn waste cellulose and an ionic liquid phosphotungstate catalyst, achieving CTA with a DS of 2.95 and good thermal stability [123]. Bamba et al. (2023) produced CTA from cocoa pod husks, reaching a DS of 2.87 and confirming polymer formation via infrared spectroscopy [111]. The use of waste biomass as feedstock, combined with green solvents such as deep eutectic solvents (DES) and ionic liquids, has received increasing attention as a strategy for low-environmental-impact routes in the production of cellulose derivatives. DES, in particular, partially disrupts the crystalline structure of native cellulose, enhancing accessibility and reactivity in subsequent chemical modifications, thereby favoring the achievement of high degrees of substitution. A notable example was reported by Sezali et al. (2023), who obtained cellulose acetate from rice straw using an alkaline DES. FTIR analysis confirmed the replacement of hydroxyl groups by acetyl groups, demonstrating the efficiency of the method. Comparative studies indicated that the cellulose acetate produced via DES exhibited greater thermal stability than a commercial sample, highlighting the potential of these solvents for sustainable chemical modification of cellulose [124].

Furthermore, recent studies have highlighted the use of homogeneous methods in cellulose acetylation. These systems operate under mild conditions, allowing for the production of CA with degrees of substitution ranging from 0.42 to 1.81, thereby eliminating the need for toxic alterations and prior activation steps. These strengths reinforce the role of these promising tools for the sustainable synthesis of biopolymers [57]. While some investigations focus on optimizing synthetic routes, others concentrate on the advanced applications of the obtained results. One example is the fabrication of membranes via phase inversion, in which DES acts as the primary solvent rather than just an additive, expanding the range of potential applications for cellulose-based materials [125]. The synthesis of CA mediated by ILs has received increasing attention, as in the research by Liu et al. (2024). In a

homogeneous system, the combination of 1-butyl-3-methylimidazolium chloride (BmimCl) with 1-butyl-3-methyldihydroimidazolium phosphate (BmimH₂PO₄) showed excellent performance in cellulose acetylation, eliminating the need for a prior hydrolysis step and resulting in degrees of substitution (DS) ranging from 0.83 to 3.0 [126]. Due to the success in obtaining a high DS, the combination of sodium acetate and BmimCl was evaluated. In contrast to commonly used corrosive mineral acids (such as sulfuric acid), sodium acetate is a non-toxic metal salt, which increases operational safety and reduces process costs. Furthermore, it demonstrated excellent catalytic performance even at low temperatures, promoting a significant increase in the degree of substitution, from 2.14 to 2.95. This effect was attributed to the increased nucleophilicity of the hydroxyl groups of cellulose, resulting from the interaction with the acetate ion from the salt [126].

Due to its physicochemical properties, CA is a cellulosic derivative widely used in the industrial fabrication of membranes and filters. Novel cutting-edge applications have been investigated for electrospun and blowspun CA fibers in the format of composite membranes, porous hydrogels, and scaffolds applied in the most diverse technological fields, such as gas separation and water treatment, active packaging, energy harvesting and sensors, wound dressings, and tissue regeneration [117,127–140]. Luo et al. (2025) reported the development of CA composite films reinforced with an ultra-low content (0.1 wt%) of carbonized polymer dots (CPDs), achieving simultaneous enhancement of breakdown strength, energy storage, and mechanical performance [135]. Strong hydrogen bonding interactions between CPDs and the CA matrix create physical crosslinking points, increasing entanglement density. As a result, the films exhibit a remarkable breakdown strength of 520.58 MV/m ($1.62\times$ that of pure CA), a discharge energy density of 2.55 J/cm³ at 450 MV/m ($1.36\times$ that of pure CA), while maintaining a high energy efficiency of 73.3%. The improvements are attributed to the Coulomb-blockade effect of CPDs, which suppresses carrier migration, and the enhanced entanglement density that reduces polarization loss and strengthens mechanical stability. This eco-friendly approach highlights CPDs as an efficient modifier for next-generation sustainable dielectric composite films [135]. CA also plays a key role as a host polymer matrix for the development of a magnesium ion-conducting polymer solid electrolyte, notable for its excellent film-forming properties. Its semi-crystalline structure, when combined with magnesium nitrate salt, becomes more amorphous, facilitating ion mobility and resulting in high conductivity. Furthermore, the interaction between the carbonyl groups of CA and Mg²⁺ ions confers electrochemical stability and enables its application in energy devices, such as magnesium primary batteries [141].

The current body of research on cellulose acetate (CA) underscores its longstanding industrial relevance while highlighting an important paradigm shift toward sustainable production routes and advanced functional applications. Historically synthesized from virgin cellulose with energy- and acid-intensive processes, CA is now increasingly derived from agro-industrial residues and processed using green solvents, thereby reducing environmental burdens and expanding resource circularity. The emergence of nanostructured CA and functional composites illustrates how minimal yet strategic modifications at the molecular level can drastically extend material capabilities. At the same time, these advances raise critical considerations: the scalability and cost-effectiveness of emerging solvent systems, the balance between achieving high degrees of substitution and maintaining biodegradability, and the alignment of laboratory breakthroughs with industrial implementation.

2.5. Cellulose Nitrate (CN)

Cellulose nitrate (CN), or nitrocellulose, is a cellulose derivative in which hydroxyl groups (-OH) are chemically replaced by nitrate groups (-ONO₂), resulting in a polymer

with distinct physicochemical properties such as high flammability and solubility in organic solvents like ether and alcohol. The nitration of cellulose alters its molecular structure by introducing nitrate esters, which significantly reduce the hydrogen bonding within the cellulose chains, leading to changes in thermal behavior and mechanical properties [142,143]. Historically, CN was synthesized in the mid-19th century and found important applications in military explosives, photographic and cinematographic films, and lacquers. The classical method involves nitrating cellulose with a mixture of concentrated nitric acid and sulfuric acid, where sulfuric acid acts as a dehydrating agent to prevent cellulose degradation during the esterification reaction [144]. The degree of nitration can be controlled by varying reaction parameters such as time, temperature, and reagent concentrations, allowing the tailoring of CN for different applications ranging from low-nitrate to highly nitrated cellulose esters. The properties of CN are strongly influenced by the degree of substitution (DS), which reflects the number of hydroxyl groups on the cellulose backbone that are replaced by nitrate groups. At low DS (partial nitration), the material is soluble in alcohol–ether mixtures and finds applications in lacquers, coatings, and inks. In contrast, at high DS (near-complete nitration), cellulose nitrate becomes highly flammable and explosive, historically known as guncotton and widely used in propellants and explosives [144–146].

Recent research has demonstrated the feasibility of producing cellulose nitrate from various biomass sources. Sakovich et al. (2018) investigated the derivation of CN from biomass such as flax, oat, and grass. They used alkaline extraction followed by acid purification, and nitrated the resulting cellulose under controlled conditions, achieving a degree of substitution of approximately 2.3, with biodegradable membranes produced at yields of 85–90% [147]. Meanwhile, Kashcheyeva et al. (2024) studied cellulose sourced from *Miscanthus × giganteus*, applying acid pretreatment and enzymatic purification prior to nitration, achieving degrees of substitution close to 2.5 and yields of 80–88%, positioning the product as a promising candidate for energetic ink formulations. Their research indicated that longer hydrolysis times (up to 24 h) improved CN yield (from 116% to 131%), nitrogen content (up to 11.83%), and viscosity (119 mPa·s), while preserving the fibrous morphology of CN—and producing BC from the same feedstock [148]. Cellulose from oat hulls, *Miscanthus*, oil palm empty bunch, tobacco stalks, and bacterial sources have also been applied to produce this solid biofuel [149–152].

Novel nitration processes aiming at sustainability and higher efficiency have been developed. A method for preparing CN was developed by Duan et al. (2023) using homogeneous esterification of cellulose in the ionic liquid [Bmim]Cl, avoiding the traditional nitric–sulfuric acid system that generates large amounts of waste and requires complex stabilization. The process yielded CN with 12.6% nitrogen content in just 15 min, featuring a 3D honeycomb structure with 200–300 nm pores, uniform nitrogen distribution, and a low polydispersity index (PDI) (1.55). Compared with commercial CN 12.6%, the new material exhibited a 141 J/g higher decomposition heat, 16.4% lower activation energy, and 40.5% faster burning rate, with excellent chemical stability after simple water treatment [153]. This ionic liquid-based approach reduces environmental impact, simplifies processing, and facilitates solvent recycling, offering a cost-effective and greener alternative for this derivatization process.

CN was synthesized using an ultrasound-assisted method from microcrystalline cellulose, aiming to intensify the conventional process [154]. Different ultrasound systems (baths and cup horns) were tested across a wide range of frequencies (20–130 kHz), powers (23–134 W·dm^{−3}), acid mixtures, amplitudes, and reaction times. Optimal conditions—20 kHz, 750 W, 60% amplitude, and 30 °C for 30 min using a mixed acid system (H₂SO₄/HNO₃/H₂O)—produced CN with 12.5% nitrogen content, outperforming silent

reactions under mechanical stirring, which yielded $\leq 11.8\%$. A plasma-assisted route was developed to produce CN from α -cellulose, using vacuum oxygen plasma pretreatment to enhance fiber hydrophilicity, surface roughness, and cross-sectional area [155]. This modification improved acid adsorption, enabling nitration with lower acid-to-cellulose ratios. While conventional synthesis requires a 1:60 acid-to-cellulose ratio to achieve 11.8–12.2% nitrogen, plasma-treated cellulose reached comparable nitrogen content with only a 1:40 ratio, leading to a 15% increase in nitrator capacity. Plasma treatment also facilitated faster acid release and lower viscosity during curing, improving autoclave efficiency. Morphological analyses confirmed nanoscale surface restructuring, correlating with enhanced reactivity. The resulting CN lacquer was transparent and soft, free from yellowing typically caused by residual NO_x .

Han et al. (2016) used cellulose nitrate (CN) as a model of a random and chargeable porous medium, demonstrating, for the first time, the phenomenon of shock electroplating. Its surface, modified with polyelectrolytes, enabled control of surface charge (positive or negative), revealing that negatively charged separators suppress dendritic growth and enable uniform and dense metal deposition even above the limiting current, while positive charges lead to instability. This discovery is crucial for the development of safer and more efficient metal batteries, demonstrating that the structure and surface chemistry of the separator can dominate the metal growth morphology [156]. However, technological research on cellulose nitrate remains largely focused on its role as a biofuel and propellant, underscoring its strategic relevance in both civil and military sectors and its potential contribution to the global energy transition [157]. Due to its high energy density, rapid combustion, and ability to be tailored through controlled nitration, CN is widely studied as a key energetic material for propellants, explosives, and solid rocket propellants. Industrial and academic efforts continue to concentrate on optimizing synthesis methods, improving nitrogen content control, enhancing safety during production, and reducing the environmental impact of effluents, all with the goal of ensuring stable and high-performance combustion [146,158,159]. Future advances in the production of this material are expected to center on greener and more efficient synthesis routes, particularly by minimizing the use of strong mineral acids through recyclable acid systems, catalytic alternatives, and closed-loop recovery processes.

2.6. Cellulose Benzoate (CB)

Cellulose benzoate (CB) is a cellulose-derived ester polymer, produced by the substitution of hydroxyl groups ($-\text{OH}$) in the cellulose backbone with benzoate groups ($-\text{COC}_6\text{H}_5$). This chemical modification significantly alters the physicochemical and mechanical properties of cellulose, imparting CB with enhanced impact resistance, flexibility, transparency, and solubility in various organic solvents. Furthermore, cellulose benzoate maintains biodegradability, positioning it as a promising sustainable material for diverse industrial applications [160,161]. The synthesis of CB commonly involves the esterification of cellulose with benzoic anhydride or benzoyl chloride, typically catalyzed by strong acids such as sulfuric acid. This reaction mechanism leads to the formation of ester linkages along with the release of benzoic acid as a byproduct [162]. The degree of substitution, reaction conditions, and choice of catalyst critically influence the molecular weight, crystallinity, and thermal behavior of the resulting polymer, thereby tailoring its final properties for targeted applications [163]. Cellulose benzoate can be synthesized from various biomass sources. For example, Trisnawati et al. (2023) produced CB from cellulose extracted from *Pandanus tectorius* leaves. They employed benzoyl chloride for the derivatization and subsequently incorporated the CB as a filler into polyvinylidene fluoride (PVDF) membranes. Their study demonstrated that incorporating 0.3 wt% CB significantly enhanced the membrane's

hydrophilicity and anti-fouling properties, leading to an increase in methylene blue rejection efficiency to approximately 82%. This finding highlights CB's potential as an effective membrane modifier for water treatment applications [164]. In a complementary approach, Takao et al. (2022) fabricated microfiltration hollow fiber membranes using cellulose acetate benzoate via thermally induced phase separation (TIPS) and non-solvent induced phase separation (NIPS) methods. These membranes exhibited improved chemical resistance when exposed to sodium hypochlorite (NaClO) solutions compared to conventional cellulose triacetate (CTA) membranes. This was evidenced by a smaller change in the water contact angle after chemical exposure, indicating enhanced durability and potential for filtration in harsh environments [161].

Ci et al. (2025) proposed an innovative route for the homogeneous synthesis of CB using superbase-derived ionic liquids, in which the solvent itself simultaneously acts as both the dissolution medium and reaction catalyst. The ionic liquid employed was 1,8-diazabicyclo(5.4.0)undec-7-ene levulinate ([DBUH]Lev), which was capable of dissolving cellulose and promoting the transesterification reaction with vinyl benzoate. Experiments demonstrated that the optimal reaction conditions were 80 °C for 4 h, with a 1:6 cellulose/vinyl benzoate ratio, yielding a degree of substitution of 2.91. The product was recovered by precipitation in anhydrous ethanol, followed by successive washings and drying at 60 °C for 24 h, resulting in high-purity CB. Beyond synthesis, the authors explored applications of the material in oil–water separation and aerogel production. Electrospun PVDF/CB membranes showed high efficiency in separating emulsions, achieving 96.9% and 97.3% for water/n-hexane and water/toluene systems, respectively, with a significant reduction in oil droplet size from 800–1500 nm to 4–33 nm. These findings demonstrated that the membranes were hydrophobic, oleophilic, and highly porous, underscoring their potential in industrial wastewater treatment processes. In parallel, CB aerogels exhibited low thermal conductivity ($0.044 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$), a value comparable to commercial insulators such as fiberglass and expanded polystyrene. Moreover, they displayed excellent mechanical strength and thermal insulation properties, maintaining a maximum ΔT of ~150 °C after 5 min and ~130 °C after 10 min when exposed to a plate heated to 200 °C, indicating remarkable performance as a thermal and structural insulator. Taken together, these results consolidate CB as a promising cellulose derivative with high added value, particularly for thermal insulation applications in buildings [165].

Liu and Wang (2023) successfully developed cellulose benzoate-based adsorbents for the removal of polystyrene microplastics. Initially, cellulose was dissolved in 1-allyl-3-methylimidazolium chloride (AmimCl) at 100 °C for 1 h. After complete dissolution, benzoyl chloride and pyridine (as a catalyst) were added, and the reaction proceeded for an additional 2 h at the same temperature, leading to the formation of cellulose benzoate. Subsequently, carbon nanotubes (CNTs) or modified carbon nanotubes (MCNTs) were incorporated into the mixture under stirring for 15 min, generating a homogeneous suspension. This suspension was treated with deionized water to remove the ionic liquid and precipitate the cellulose benzoate–CNT composite. The precipitated material was then filtered through a 0.45 μm membrane, thoroughly washed to eliminate residues, and finally lyophilized for 24 h, yielding the final adsorbent. The incorporation of CNTs and MCNTs significantly enhanced the microplastic removal efficiency, increasing it from 68.3% to 97.4% and 97.8%, respectively. Moreover, tests carried out in real river water confirmed the high efficiency of the composites, demonstrating their robustness under complex environmental conditions. These findings highlight the strong potential of functionalized cellulose benzoate as an advanced adsorbent, particularly for use in wastewater treatment plants (WWTPs) and natural water purification systems, positioning it as a promising alternative for mitigating microplastic pollution [166].

Chen et al. (2017) developed the synthesis of cellulose benzoate (CB) from sugarcane bagasse. Initially, cellulose was extracted through washing, drying, dewaxing with toluene/ethanol, and milling until particles of 20–40 mesh were obtained. The resulting cellulose was then dissolved in 1-ethyl-3-methylimidazolium acetate (EmimAc) at 130 °C, yielding a homogeneous solution. For derivatization, vinyl benzoate was added to the solution, and the reaction proceeded at 70 °C for 2 h. The resulting product was precipitated in isopropanol, filtered, washed, and dried, producing CB films. The effectiveness of the synthesis was confirmed through appropriate characterization techniques, demonstrating the successful formation of cellulose benzoate from a renewable lignocellulosic biomass source [167].

Polez et al. (2025) synthesized cellulose benzoate (CB) from cotton linters. Initially, the cellulose underwent a mercerization pretreatment using a 20% (*w/w*) NaOH solution for 1 h at 0 °C to increase the reactivity of the fibers. Subsequently, the cellulose was dissolved in LiCl/DMAc under nitrogen for 90 min at 160 °C, forming a homogeneous solution. The derivatization to cellulose benzoate was carried out via esterification of the dissolved polymer with benzoyl chloride, using different molar ratios (3, 6, and 12 relative to the anhydroglucose unit, AGU), which enabled the production of materials with varied degrees of substitution (DS), allowing for the tuning of the final material's physicochemical and thermal properties [168].

Cellulose benzoate stands out as a cellulose derivative of significant scientific and technological relevance, as the introduction of benzoyl groups into the cellulose chain imparts increased hydrophobicity, thermal stability, and improved processability compared to native cellulose. These features expand its potential applications in films, membranes, and advanced composites, particularly in systems requiring high mechanical strength and stability in organic solvents. However, its synthesis still faces significant limitations, mainly due to the use of aggressive solvents, such as LiCl/DMAc or molten salts, and toxic reagents, like benzoyl chloride, which compromise the feasibility of sustainable, industrial-scale production. In this context, although notable advances have been achieved, the expectation is that new, cleaner, and environmentally compatible synthesis routes will be established, enabling the broader use of cellulose benzoate in higher-value technological applications.

2.7. Cellulose Acetate Butyrate (CAB)

Cellulose acetate butyrate (CAB) is a thermoplastic cellulose-derived copolymer characterized by the substitution of hydroxyl groups with both acetate ($-\text{COCH}_3$) and butyrate ($-\text{COC}_3\text{H}_7$) groups along the cellulose backbone. This unique combination confers CAB with excellent impact resistance, flexibility, optical transparency, solubility in various organic solvents, and biodegradability, making it highly valuable across a range of applications [163,169]. The CAB market is projected to reach USD 1.06 billion by 2030, driven by its renewable origin, thermal stability, UV resistance, and versatility in coatings, plastics, and films [170]. Advances in industrial production have improved sustainability, solvent resistance, and performance customization, positioning CAB as a greener alternative to traditional cellulose esters with broad commercial adoption in coatings, plastics, and printing applications.

The butyrate content in the copolymer plays a crucial role in modulating its hydrophobicity and flexibility, which directly influences its processing behavior and end-use properties [171,172]. The production of CAB is typically carried out as a two-step esterification process to introduce both acetate and butyrate groups onto the cellulose backbone. In the first step, cellulose is reacted with acetic anhydride, usually in the presence of an acid catalyst, to form partially substituted cellulose acetate as an intermediate. In the

second step, the cellulose acetate is further reacted with butyric acid or butyric anhydride to introduce butyrate groups, yielding the final CAB product. This sequential approach provides precise control over the degree of substitution for each acyl group, which is critical for tailoring the polymer's solubility, thermal stability, and flexibility [173,174].

A mechanical activation-assisted esterification (MAE) method was developed by Huang et al. (2020) as a greener and efficient approach to produce CAB from cotton cellulose, offering an alternative to traditional liquid-phase esterification. The process integrates mechanical activation and esterification in a ball mill, eliminating separate pretreatment and enhancing efficiency. Using cellulose, butyric acid, acetic anhydride, and sulfuric acid as a catalyst, CAB was synthesized and purified, with structural confirmation by NMR. Although the molecular structures and molar masses of MAE- and LPE-prepared CAB differed due to distinct reaction systems, the resulting CAB films showed comparable surface and wear properties, including gloss, rub-resistance, and ethanol resistance [175].

The synthesis of CAB using low-environmental-impact solvents has also been the subject of increasing research, although scarce in comparison to other cellulose esters. The IL 1-allyl-3-methylimidazolium chloride (AmimCl) was the first green solvent applied as a recyclable homogeneous medium for the production of CAB in the literature, thus allowing for the production of this copolymer with up to 47% butyrate content at low temperatures and without catalysts [176–178]. In the work developed by Abarkan et al. (2024), ILs act as a reaction medium for the homogeneous modification of cellulose found in plants of the species *Stipa tenacissima*. Among the solvents tested, [C₄mim]OAc demonstrated the greatest cellulose solubilization power (15% by weight) and provided high degrees of substitution, especially for the butyrate component (DS = 2.76). Although the operational benefits of using green solvents with unique properties, such as those observed in ILs and DES, are recognized, the production of highly substituted CABs remains underexplored and requires future investigation [179].

The versatility of this derivative is well demonstrated by its applications explored in the literature. The study by Huang et al. (2018) compared a mechanical activation-assisted (MA) process and a conventional liquid-phase (LP) method to produce CAB, aiming to improve emamectin benzoate (EB) pesticide delivery through controlled-release microspheres. CAB-MA was produced directly in a stirring ball mill via simultaneous mechanical activation and esterification, eliminating the need for pretreatment and extra solvents, while CAB-LP required an initial acetic acid-based pretreatment before esterification. Structural characterization showed that CAB-MA had lower crystallinity, smaller molecular weight, narrower distribution, and higher substitution degree compared with CAB-LP. These features enhanced its pelletizing behavior, polymer–drug interaction, and encapsulation efficiency. When used as wall materials for EB-loaded microspheres, both CABs provided controlled release and photostability; however, CAB-MA offered superior drug loading, entrapment, and photostability performance [180]. Ran et al. (2024) developed an in situ injectable gel formulation for periodontitis treatment, utilizing CAB as a matrix to encapsulate the antimicrobial drugs metronidazole and doxycycline hyclate [181]. Confirming with Edgar et al. (2006), the gel exhibited favorable physicochemical properties, efficient drug release profiles, and promising biocompatibility, indicating its potential as an effective localized therapeutic platform [182].

In the field of materials science, Nejström et al. (2023) used commercial cellulose pulp as the cellulose source, typically derived from wood pulp, which was initially dissolved in a cold alkali/urea aqueous solution, enabling regeneration in alcohols or esters to obtain regenerated cellulose films. For the cellulose acetate butyrate (CAB) films, commercially available CAB grades were used, dissolved in acetone, and processed via the solvent casting

technique to produce self-supporting films. Their results revealed that the film-forming ability of CAB is closely linked to the availability and positioning of hydroxyl groups, while the semi-crystalline nature of the films is influenced by the distribution and quantity of side groups on the polymer chain [183]. Furthermore, the study demonstrated that the crystallinity decreases with lower molecular weights and higher butyrate content, which is also associated with the presence of cholesteric phases in samples with higher acetate-butyrate ratios. CAB was also successfully applied for the production of hard carbon-based electrodes for supercapacitors, crosslinked bionanocomposites with improved mechanical properties, gels with sustained antibacterial activity, and electrospun composite nanofibers for tissue engineering, water treatment, and gas separation [184–188].

CAB stands out as a versatile cellulose ester with unique structural tunability, enabling applications that range from coatings and films to delivery systems and energy materials. Advances in greener synthesis routes, including mechanical activation-assisted esterification and the use of ionic liquids, highlight promising directions for more sustainable and efficient production. Nonetheless, despite its advantageous physicochemical properties and demonstrated potential in biomedical, environmental, and energy-related applications, recent years have seen a relative scarcity of research into CAB production and technological exploitation. Only a limited number of studies report the use of residual biomass sources for CAB synthesis, leaving a clear gap in the exploration of waste valorization and broader application development. This underlines the need for renewed research efforts to expand sustainable production strategies and unlock the underutilized potential of CAB in advanced material technologies.

2.8. Cellulose Propionate (CP)

Cellulose propionate (CP) is a cellulose-derived polymer produced by replacing hydroxyl groups ($-OH$) in cellulose with propionate groups ($-COC_2H_5$). This esterification enhances properties, including high impact resistance, flexibility, transparency, solubility in various organic solvents, and biodegradability, making CP a versatile biopolymer for multiple industrial applications [189–191]. The chemical modification allows the tailoring of physicochemical characteristics by adjusting the degree of substitution, which influences its thermal and mechanical behavior [163,191,192]. The typical synthesis of cellulose propionate involves the esterification of cellulose with propionic anhydride in the presence of acid catalysts such as sulfuric acid, which promotes the reaction and leads to the formation of propionate ester bonds and the release of propionic acid as a byproduct [193]. The process parameters, including reaction time, temperature, and catalyst choice, strongly affect the efficiency and degree of substitution achieved. Additionally, some biorefineries have begun producing CP from agricultural residues such as rice husk and wheat straw, contributing to sustainability efforts by valorizing biomass waste streams [163].

In recent research, Nemr et al. (2021) conducted a comparative study on the propionylation of cellulose sourced from commercial cotton, rice husk, and wheat straw, employing N-iodosuccinimide (NIS) as a catalyst under solvent-free conditions. Reaction times varied from 2 to 6 h, with subsequent treatment involving ethanol and water to quench unreacted reagents. The yields ranged from 71.54% to 88.37%, with DS spanning from 1.32 to 2.80 for cotton, 1.76 to 3.0 for rice husk, and 1.60 to 3.0 for wheat straw cellulose, demonstrating the adaptability of the method across biomass sources [194]. Sandrini et al. (2024) synthesized CP through a homogeneous route, also using biomass as the raw material [195]. For this purpose, sisal cellulose was employed in the synthesis carried out in N,N-dimethylacetamide/lithium chloride (DMAc/LiCl). The process enabled the production of derivatives with different DS values, confirming the efficiency of the homogeneous system in cellulose modification.

The research carried out by Utrera-Barrios et al. (2024) investigated the development of bio-based self-healing elastomeric composites obtained from a blend of epoxidized natural rubber (ENR) with cellulose propionate (CP) reinforced with cellulose fibers. The optimized ENR/CP ratio of 70/30 resulted in an increase in tensile strength, accompanied by a slight reduction in elongation at break. The incorporation of cellulose fibers at medium to high concentrations led to significant mechanical enhancements while also providing self-healing efficiencies above 85%. The healing mechanism was attributed to polymer chain mobility and the formation of hydrogen bonds, highlighting the potential of CP as a key component in the development of high-performance sustainable materials [196]. Lee and Kang (2024) developed microporous CP membranes with glycerol as a plasticizing additive, using hydraulic pressure techniques to form interconnected pores for energy applications. The CP/glycerol membranes exhibited 74.1% porosity and a homogeneous structure, both of which are favorable for ionic transport, in addition to enhanced mechanical strength. These results highlight the potential of CP as a promising polymeric matrix for separators in lithium-ion batteries [197].

CP stands as a highly relevant cellulose derivative, with growing importance across various industrial sectors. In 2024, its global market was estimated at approximately USD 500 million, with projections to reach USD 800 million by 2033, driven by a CAGR of 6.5% between 2026 and 2033. This expansion is supported by rising demand in segments such as coatings, films, paints, automotive interiors, electronic accessories, packaging, and pharmaceuticals. The consolidation of CP in this context stems from its distinctive physicochemical properties, which make it particularly attractive as a sustainable and biodegradable alternative to single-use plastics [198]. This derivative stands out by combining thermoplastic processability with well-balanced mechanical properties, making it a particularly attractive material for high-value-added industrial applications. The presence of propionate groups imparts high chemical resistance, dimensional stability, and greater compatibility with additives and plasticizers, while simultaneously preserving biodegradability. Although traditional heterogeneous routes enable large-scale production, limitations remain regarding substitution uniformity and precise control of final properties. In this context, advances in homogeneous synthesis and the use of biomass as a sustainable raw material have opened new perspectives for obtaining CP with greater selectivity and lower environmental impact.

3. Cellulose Ethers

Cellulose ethers are widely studied cellulose derivatives, obtained through the etherification of hydroxyl groups in the cellulose chain. This modification enhances water solubility, chemical stability, and rheological properties, making them more versatile than native cellulose. Common examples include carboxymethyl cellulose (CMC), hydroxyethyl cellulose (HEC), and methylcellulose (MC), each presenting distinct characteristics suitable for specific applications. These derivatives are extensively used as thickeners, stabilizers, emulsifiers, and film-forming agents in the food, pharmaceutical, cosmetic, and construction industries. Due to their biocompatibility and non-toxicity, cellulose ethers are also applied in drug delivery systems and biomedical formulations. In addition, their renewable origin and biodegradability make them environmentally friendly alternatives to synthetic polymers. The reaction pathways for the production of the polysaccharide ethers are illustrated in Figure 5.

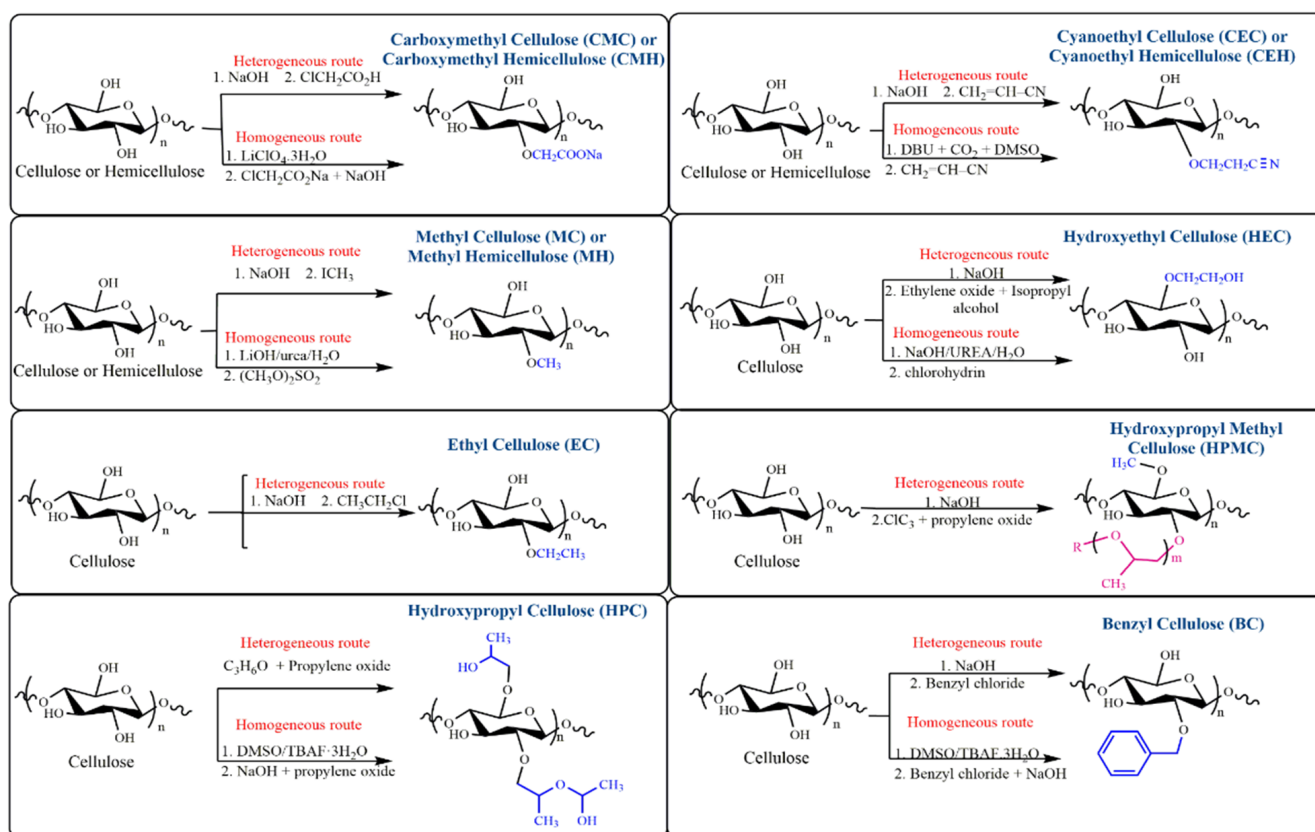


Figure 5. Reaction pathways for the production of the polysaccharide ethers discussed in this review.

3.1. Methyl Cellulose (MC)

Methylcellulose is one of the simplest and most widely used derivatives of cellulose, valued for its enhanced water solubility and thermoresponsive gelation properties. These features make methylcellulose particularly suitable for applications in biomedical fields, construction materials, and cell culture systems [199,200]. The methylation reaction involves replacing hydroxyl groups (-OH) at the C-2, C-3, and C-6 positions of the anhydroglucose unit (AGU) with methyl groups (-CH₃), producing a polymer soluble in water or organic solvents depending on the degree of substitution (DS), which ranges from 0 to 3 [199,201]. Commercial MC is commonly synthesized via a heterogeneous process, where cellulose is first treated with sodium hydroxide (NaOH) to form alkaline cellulose. This intermediate then reacts with etherifying agents such as iodomethane (methyl iodide, MI) and dimethyl sulfate (DMS). DMS is usually preferred for heterogeneous methylation of cellulose (due to better yields and lower cost), but both DMS and MI are highly hazardous; thus, the choice depends on reactivity, scale, cost, and whether strict safety and waste-management measures can be implemented [202,203]. Purification follows through washing with hot water and drying. The use of additional solvents such as acetone or toluene allows modulation of the DS, optimizing the product's solubility and functional properties [204]. Despite its industrial relevance, few studies have focused on preparing methylcellulose derived from waste biomass, an area with potential for value addition to agro-industrial residues.

Viera et al. (2007) produced MC from sugarcane bagasse cellulose using DMS under heterogeneous conditions. The study showed that methylation without solvents yielded a DS of 0.70, whereas the addition of toluene or acetone increased the DS to 1.2. This chemical modification induced notable chemical and thermal changes in the cellulose, expanding its applicability and enhancing the value of sugarcane bagasse as a raw material [205]. Later, Vieira et al. (2012) extended this work by producing methylcellulose from mango

seed fibers (*Mangifera indica* L. Ubá). Using both DMS and iodomethane as alkylating agents in heterogeneous methylation, they found a significantly higher DS with DMS (1.30) compared to iodomethane (0.47), highlighting the impact of the choice of reagent on product properties [206].

Oliveira et al. (2015) produced MC from bacterial cellulose (BC) in a heterogeneous medium using dimethyl sulfate (DMS) as the methylating agent, comparing reactions of 3 h (MC3h) and 5 h (MC5h) with reagent replenishment every hour. Bacterial cellulose (3 g) was treated in an alkaline medium with 50% (*w/v*) NaOH (60 mL) for 1 h at room temperature to increase reactivity by promoting the transition of the structure to the cellulose II allomorph and enhancing the accessible surface area. After removing the excess NaOH, acetone was added as the solvent medium, followed by DMS dropwise, while maintaining the reaction temperature at 50 °C. At the end of the reaction time, the sample was neutralized with 10% (*v/v*) acetic acid, filtered, washed three times with acetone (90 mL each), and dried in an oven at 50 °C. These conditions allowed for high degrees of substitution to be achieved (DS ~2.26 for MC3h and ~2.33 for MC5h), with products exhibiting high crystallinity and thermal stability, as well as the ability to form transparent films and highly porous foams with potential applications in hydrogels and biomaterials [204].

This derivative is well known in the literature for its potential use as a drug excipient and in controlled-release systems [207]. Commercial MC was used for producing thermo-responsive methylcellulose–sodium humate crosslinked hydrogels for localized delivery of cisplatin applied to osteosarcoma treatment via a light-assisted mechanism [208,209]. In vitro tests suggested successful tumor ablation with the synergistic action of both laser emission and cisplatin release by this novel material, resulting in a reduction in the number of viable osteosarcoma cells. MC is also reported as a promising material for the biofabrication of tissue equivalents, using techniques such as extrusion-based 3D (bio)printing, also called 3D (bio)plotting or robocasting, which incorporates cells into the biomaterial during fabrication. It has already been applied for the bioprinting of functional tissues like bone, osteochondral, and cartilage constructs, as well as pancreatic islets from rats [209].

Methylcellulose is widely applied in industry—its global market was valued at approximately USD 1.2 billion in 2023 and is projected to reach nearly USD 2.1 billion by 2032 (CAGR of 6.2%), especially due to its extensive application in the food and beverages industry as a non-toxic and hypoallergenic thickening agent, emulsifier, and stabilizer, enhancing the texture and consistency of various food products, with particular relevance to vegan meat substitutes [210]. It is also employed in pharmaceutical, cosmetic, and construction sectors, where it acts as a binder, film former, controlled-release agent, thickener, and water retention agent.

Despite its outstanding physicochemical properties and established commercial applications, recent research reveals a gap in innovative synthesis strategies and advanced technological applications. Nagel et al. (2010) were pioneers in performing the dissolution of spruce sulphite pulp cellulose in LiOH/urea/H₂O and using this medium for its homogeneous methylation with dimethyl sulfate [211]. MC samples with a DS between 1.07 and 1.59 were obtained by adjusting the reaction temperature (22–50 °C), the molar ratio of dimethyl sulfate (9–15 mol per anhydroglucose unit), and the reaction time (4–24 h). Optimal conditions were achieved at 50 °C using 12 mol DMS/mol AGU. However, while these findings marked an advancement in the study of this derivative, the method requires high energy consumption to dissolve cellulose—employing very low temperatures and high-speed stirring—and a high excess of the methylating agent. Notably, no subsequent studies to date have reported a greener methylation process or the use of

more environmentally friendly solvents—such as ionic liquids (ILs), deep eutectic solvents (DESs), or inorganic molten salts (IMs)—for homogeneous methylation, thus representing a compelling opportunity for novel research.

3.2. Ethyl Cellulose (EC)

Ethyl cellulose is a cellulose derivative obtained by substituting the hydroxyl groups (-OH) of the anhydroglucose units (AGUs) with ethoxy groups (-O-CH₂-CH₃). The synthesis process begins with the deprotonation of cellulose hydroxyl groups in an alkaline medium (mercerization), which facilitates the disruption of its supramolecular structure and increases reactivity. Subsequently, an ethyl halide is introduced to perform the etherification reaction, resulting in the substitution of hydroxyl groups with ethoxy moieties along the cellulose backbone [212,213]. A small number of papers have reported the synthesis of EC in recent years, especially from residual feedstocks. Waste paper was used as a cellulose source by Pratama et al. (2023) to produce EC nanoparticles (NPs) for extracorporeal oxygenation composite membranes [214]. Cellulose was isolated following a procedure involving acetone soaking, alkaline deinking, neutralization, and ball milling to obtain fine cellulose powder. The resulting cellulose was converted into EC nanoparticles via alkalization with NaOH (mercerization), reaction with ethyl chloride in acetone, neutralization, and washing. A sonication step in ethanol was incorporated to achieve nanosized particles before drying. These NPs were subsequently supported onto commercial membranes (nylon and PTFE) and synthetic membranes (SBS and Pebax[®]) via a dip-coating method for enhancing hydrophobicity and membrane oxygenation performance.

In pursuit of more sustainable production methods, Zhou et al. (2024) developed a process that starts with cellulose extraction from rice straw agricultural waste. This cellulose was then converted into ethyl cellulose via an ethylation process where ethyl bromide replaced the conventional ethyl chloride, due to its higher reactivity, affordability, and suitability for room-temperature storage. Initially, the rice straw cellulose was mercerized using a NaOH solution, after which ethyl bromide was introduced to initiate the reaction in a high-pressure reactor. The resulting ethyl cellulose displayed high degrees of substitution, ranging from 2.0 to 2.5, and was successfully formulated into biodegradable films blended with ethanol. The EC was further transformed into biodegradable films for the substitution of fossil-derived plastics, and the waste liquid produced was recycled to avoid environmental pollution. This innovative method not only achieves efficient ethylation but also valorizes agricultural residues, aligning with circular economy principles [215].

Ethyl cellulose is outstanding for its excellent membrane-forming capacity, flexibility, durability, and hydrophobicity [216]. These properties make it highly suitable for applications such as gas separation membranes, evaporation control, and coating materials. Additionally, its non-toxic, biodegradable, and thermoplastic characteristics contribute to its widespread use in pharmaceuticals, food packaging, and other industrial sectors [217]. As with methylcellulose, EC has been applied in composite delivery systems, and recent research demonstrates its use for controlled release of agricultural pesticides, drugs, and fungicides [207,218–226]. Biomedical material applications include the development of composites, hydrogels, and oleogels for functional adipose tissue mimetics, bone regeneration, wound dressings, and ablation therapy of cancer tissues [227–232]. In addition, it has been widely applied in advanced research in the food industry for active packagings, fat replacers, and oleogelator materials, and is an emerging component for composites and membranes employed in water pollution remediation [233–239].

EC has also gained prominence in printed electronics applications. Palmieri et al. (2024) developed EC-based screen-printed electrodes (SPEs) as an environmentally friendly alternative to conventional PET, notable for their water resistance and biodegradability while maintaining performance comparable to traditional substrates. EC eliminates dependence on petroleum-derived plastics, is biodegradable, and overcomes limitations of other biodegradable materials, such as paper, which requires chemical treatments and is water-sensitive [240]. EC can also be combined with different materials to tailor its properties, producing biodegradable, high-performance substrates capable of competing with conventional plastics in flexible electronics. An example of this combination is the blend of ethyl cellulose (EC) and hydroxypropyl cellulose (HPC), which allows the material's mechanical and chemical properties to be synergistically combined, making it suitable for conventional photolithographic processes, traditionally restricted to non-biodegradable substrates such as PET and polyimide. The EC: HPC substrate was used to fabricate capacitive humidity sensors and resistive strain sensors, with titanium and aluminum metal patterns deposited by photolithography. The material has been shown to withstand processes such as metal deposition, UV exposure, and chemical treatments without significant degradation [241].

Ethyl cellulose finds widespread utility across multiple industrial sectors, and its global market is expected to reach \$1.35 billion USD by 2029 (CAGR of 9.0%) [242]. In pharmaceuticals, it is applied as a coating agent, binder, film former, stabilizer, and component in sustained-release formulations for oral and topical dosage forms. In the food industry, it functions as an edible coating, thickening agent, emulsifier, flavor encapsulant, and barrier layer in packaging [242]. It is also employed in cosmetics and personal care products, such as nail polish, where it acts as a film-former, thickener, and stabilizer, as well as in hair care formulations for textural enhancement and shine. Moreover, ethyl cellulose is utilized in printing inks, paints, varnishes, textile sizing, adhesives, and ceramic slurries, where its excellent film-forming capability, chemical resistance, and rheological properties are highly valued. However, further research into synthesis optimization and the development of greener processes is still required for this derivative.

3.3. Hydroxyethyl Cellulose (HEC)

Hydroxyethyl cellulose (HEC) is a versatile cellulose derivative extensively employed in industrial and biomedical applications due to its favorable characteristics, such as biocompatibility, biodegradability, non-toxicity, and excellent water solubility [243]. The derivatization process involves the substitution of the hydroxyl groups (-OH) on the cellulose backbone with hydroxyethyl groups (-CH₂CH₂OH), which imparts improved rheological and film-forming properties to the polymer. Industrially, HEC synthesis typically starts with cellulose mercerization in sodium hydroxide solution to form alkaline cellulose, which subsequently undergoes etherification with gaseous ethylene oxide under controlled conditions to produce hydroxyethyl cellulose [243–245]. Orhan et al. (2018) successfully synthesized HEC from industrial air vacuum dust (APVD), a waste product of the textile industry, demonstrating a sustainable approach to cellulose valorization. Their methodology involved bleaching and mercerization of APVD-derived pulp, followed by etherification with ethylene oxide and isopropyl alcohol under microwave irradiation, with variations in temperature and reaction time. The highest degree of substitution (DS) achieved was approximately 1.09, indicating effective functionalization while utilizing industrial waste as feedstock [246].

Pineapple peel cellulose was employed to produce oxidized HEC (OHEC) for application in pH-sensitive hydrogel composites with *Herichium erinaceus* mushroom residue carboxymethyl chitosan (CMCS) in the research developed by Yin et al. (2022). OHEC was

synthesized from pineapple peel waste through a sequence of cellulose extraction, etherification, and oxidation steps, resulting in an aldehyde-rich polymer. The two components were crosslinked via a Schiff base reaction, forming OHEC/CMCS composite hydrogels. Structural analyses confirmed the formation of a porous 3D hydrogel network with rapid gelation (as fast as 33 s at an optimal mass ratio), high swelling capacity (up to 11.58 g/g), and oxidation-degree-dependent swelling behavior. Drug release experiments with bovine serum albumin demonstrated pH-sensitive and sustained release, with minimal release under acidic gastric-like conditions and rapid release under alkaline intestinal-like conditions, highlighting the potential of waste-derived OHEC hydrogels as eco-friendly carriers for oral protein drug delivery [247].

The homogeneous synthesis of HEC was previously reported by Chinese research groups using NaOH/Urea aqueous solutions [248,249]. Köhler et al. (2010) prepared HEC by dissolving microcrystalline cellulose in [C₂mim][OAc] and reacting with ethylene oxide at 80 °C for 19 h. The product was isolated by precipitation in isopropyl alcohol, redissolution in water, and reprecipitation in isopropyl alcohol, followed by washing and air-drying. The homogeneous process, performed without inorganic bases, produced completely DMSO- and water-soluble HEC with a molar substitution degree of up to 1.07, benefiting from the catalytic effect of acetate ions in [C₂mim][OAc] [250].

The inventors of patent number US8541571B2 (2013) developed a novel approach for producing cellulose ethers by dissolving cellulose in imidazolium salts and quaternary ammonium ionic liquids (ILs), performing etherification directly within the homogeneous medium, without prior alkaline activation of cellulose. This process may optionally include small amounts of water (up to ~25 wt%) or co-solvents (such as DMSO, DMF) to improve dissolution or reaction compatibility. Operating under moderate temperatures (typically 20–130 °C) and pressures (100–2000 mbar), the method enables efficient etherification without polymer degradation. Once the reaction is complete, HEC with DS up to 2.16 is precipitated by adding a non-solvent (e.g., alcohols, acetone, water), isolated, purified, and dried. The ionic liquid can then be recovered and recycled for further use, representing a promising approach within the biorefinery concept [251].

HEC's global market was estimated at USD 0.75 billion in 2024 and is projected to grow to USD 1.11 billion by 2032, registering a CAGR of 5.0% over the forecast period [252]. In the food industry, HEC serves as a stabilizer and thickening agent, enhancing texture and preventing separation in products such as sauces, dressings, and beverages [253]. Its minimal caloric content makes it particularly valuable in low-fat and gluten-free formulations, contributing to healthier food options without compromising quality. Additionally, HEC has been investigated for sustainable active packaging and edible coatings to improve postharvest quality [254–256].

In the cosmetic industry, HEC is widely utilized as a thickener, stabilizer, and film-forming agent in products such as shampoos, conditioners, lotions, and gels [257]. Beyond personal care, it is employed in environmental applications, including water treatment, CO₂ capture, and enhanced oil recovery [258–261]. Its use in soil conditioning also helps improve water retention and reduce erosion, promoting sustainable agricultural practices. Biomedical research has increasingly focused on modifying HEC to enhance gelation and mechanical strength, enabling its use in advanced drug delivery systems, wound dressings, and tissue engineering scaffolds [247,262–267]. Its biocompatibility and hydrogel-forming ability facilitate controlled release of therapeutic agents and support cellular growth. For instance, synergistic combinations of HEC with other polymers, such as chitosan or alginate, have yielded hydrogel composites with tailored swelling, degradation, and controlled drug release profiles [268].

HEC also plays a crucial role in flexible electronic devices, acting as a modifier of the mechanical properties and processability of cellulose-based substrates. As a natural plasticizer, HEC allows cellulose nanofiber (CNF) networks to slide over each other under tension, increasing deformation at break. Films containing HEC are highly transparent, fully biodegradable, and can be printed with silver-based inks for electrical conduction [269]. A particularly promising derivative is quaternized hydroxyethyl cellulose (QHEC), in which the polymer structure is chemically modified to incorporate quaternary ammonium groups. This modification confers unique mechanical, antibacterial, and electronic properties, making QHEC suitable for applications such as wearable bioelectrodes and autonomous sensors [270].

The current state of the art in HEC research reveals a material that is both mature in industrial applications and rapidly evolving through sustainable sourcing and advanced functionalization strategies. While traditional production from commercial eucalyptus cellulose pulp still dominates, recent studies demonstrate a shift toward valorizing agricultural and industrial wastes. Homogeneous synthesis methods have addressed limitations of heterogeneous etherification, achieving higher and more uniform degrees of substitution without severe polymer degradation. Nonetheless, challenges remain in balancing cost-effective large-scale production with the tailored physicochemical properties required for specialized applications.

3.4. Hydroxypropyl Cellulose (HPC)

Hydroxypropyl cellulose (HPC) is a surface-active polymer whose water solubility varies with temperature. This characteristic, combined with its hydrophilicity, phase behavior, and ease of production, makes HPC widely used in the pharmaceutical, food, and cosmetic industries [271]. Currently, industrial HPC production predominantly occurs via a heterogeneous route, in which cellulose is etherified in the presence of solvents such as toluene, acetone, isopropanol, and tert-butanol. Recent advancements have explored homogeneous synthesis to achieve higher etherification efficiency, greater degrees of substitution, and reduced environmental impact, in line with green chemistry principles [272].

Some studies have investigated HPC synthesis from sustainable cellulose sources. Joshi et al. (2019) proposed the use of paper waste as a raw material, employing alkalization followed by etherification. Optimized reaction conditions achieved a DS of 1.15: 1.5 M/AGU NaOH, alkalization at 40 °C for 2.5 h, 27.82 M/AGU propylene oxide, and reaction for 3.5 h at 55 °C [273]. Zhong et al. (2024) produced HPC from residual bamboo powder, using alkaline bleaching followed by heterogeneous etherification with propylene oxide. Optimized conditions yielded HPC samples with DS values of 1.0–1.25, which were applied in thermochromic hydrogels for smart windows. HPC has also been obtained from olive tree branches, banana blossoms, and oil palm empty fruit bunch cellulose via heterogeneous hydroxypropylation [274]. Köhler et al. (2010) reported the first homogeneous synthesis of HPC, dissolving spruce sulfite pulp in [C₂mim][OAc] and reacting it with propylene oxide at 80 °C for 19 h. The product was precipitated in isopropyl alcohol, washed repeatedly, and air-dried. This method, performed without inorganic bases, produced water-soluble HPC with a molar substitution degree of up to 2.79. The catalytic effect of acetate ions in [C₂mim][OAc] was critical for cellulose conversion, whereas other ILs, such as [C₄mim]Cl, showed no reaction [250].

The homogeneous synthesis of HPC was achieved by Wang et al. (2024) through the solubilization of commercial microcrystalline cellulose in a DMSO/TBAF·3H₂O solvent system within 15 min. Cellulose was alkalinized by adding NaOH, followed by the introduction of propylene oxide to initiate the etherification reaction, which proceeded for 6 h at 50 °C. Afterward, the solution was cooled to room temperature, neutralized with

acetic acid, dialyzed, concentrated, and freeze-dried. This method yielded HPC with a high molar degree of substitution (DS up to 12.3), showing potential for applications in force sensors [272].

The HPC market is projected to reach USD 204.5 million by the end of 2025 and grow at a 5.6% CAGR, attaining USD 349.8 million by 2035. In 2024, steady demand from food coatings, ophthalmic solutions, and pharmaceutical excipients supported moderate market growth, with notable expansion driven by oral drug delivery applications, due to HPC's film-forming and mucoadhesive properties [275]. Beyond its industrial applications, HPC has attracted attention for its thermoresponsive behavior, enabling phase transitions in aqueous solutions at specific temperatures, known as the lower critical solution temperature (LCST). This property is particularly valuable for stimuli-responsive materials and smart systems, such as temperature-sensitive drug carriers and adaptive coatings [276,277]. Its reversible solubility switch in response to temperature changes also facilitates use in controlled-release formulations and bioseparation technologies, where environmental responsiveness is essential [278,279]. Additionally, HPC materials can exhibit pH-responsive behavior, useful for targeted drug release [280].

Research efforts have focused on enhancing HPC's functional versatility through blending with other biopolymers or synthetic polymers, yielding composites with improved mechanical properties, stability, and tunable degradation rates, suitable for active packaging, biomedical implants, and tissue engineering scaffolds [277,281–285]. The incorporation of crosslinking agents or physical treatments further expands its application range, enabling the design of hydrogels with tailored porosity and swelling behaviors, ideal for wound dressings, responsive membranes and sensors, and catalytic hydrogels for water decontamination [279,286–291]. A promising HPC application involves gel electrolytes for electrochromic devices (ECDs). ECDs can change color or transparency in a controlled manner via electrical stimuli, with practical examples including electrochromic mirrors in automotive rearview mirrors to reduce night glare. HPC-based electrolytes are attractive candidates due to their transparency, adhesion to diverse surfaces, and compatibility with polymer conductors [292].

HPC can also be combined with poly(2-acrylamido-2-methylpropanesulfonic acid) (PAMPS) to produce anisotropic hydrogels with thermochromic and mechanochromic properties for multifunctional sensor applications. Shi et al. (2023) developed a hydrogel based on this combination that exhibited a vivid interference nuclei display under crossed polarizers, highly sensitive and responsive to external stimuli. The hydrogel changes color over a wide temperature range (25–66 °C) with minimal loss of light transmission (<13%), outperforming conventional hydrogels. The response is reversible and repeatable, with tensile force and pressure altering the color of the aligned hydrogel, whereas the unaligned hydrogel responds only to larger deformations. The color change results from the rearrangement of HPC molecules within the network, modifying birefringence. Therefore, the HPC/PAMPS hydrogel holds potential for temperature sensors with real-time visual feedback and mechanical sensors for pressure or strain detection in soft robotics or human-machine interfaces [293].

Overall, HPC stands out among cellulose ethers for its unique combination of thermoresponsiveness, film-forming ability, and versatility across industrial and biomedical applications. Compared to methylcellulose (MC) and ethylcellulose (EC), HPC offers superior solubility modulation with temperature, enabling functions such as LCST-triggered drug release and adaptive coatings that other derivatives cannot readily match. While its industrial synthesis is still dominated by heterogeneous processes with limited substitution efficiency, recent advances in homogeneous etherification have demonstrated higher substitution efficiency and improved environmental compatibil-

ity. Further research focusing on substitution control, selectivity, and scalable green synthesis, while exploiting HPC's LCST-triggered responsiveness, could reinforce its role as a strategic material for next-generation biomedical, pharmaceutical, and functional materials applications.

3.5. Hydroxypropyl Methylcellulose (HPMC)

Hydroxypropyl methylcellulose (HPMC), also known as hypromellose, is a semi-synthetic cellulose ether obtained by substituting the hydroxyl groups of cellulose with hydroxypropyl and methyl groups through an etherification reaction [294]. Used in hydrophilic tablet matrices for over 60 years, its properties vary according to the degree of substitution and molecular weight, influencing water solubility and swelling capacity depending on the ratio between methoxyl and hydroxypropyl groups [295]. HPMC is a hydrophilic, non-ionic polymer, stable over a wide pH range, biodegradable, and soluble in dipolar aprotic solvents [296]. Its thermal stability and film-forming ability make it widely employed in food biocomposites, particularly in edible films that are transparent, flexible, odorless, and tasteless [297,298].

Structurally, HPMC consists of a cellulose backbone composed of β -(1 $\rightarrow$ 4)-linked D-glucose units, where hydroxyl groups on the anhydroglucose units (C-2, C-3, and C-6 positions) are partially substituted by methoxyl (-OCH₃) and hydroxypropyl (-OCH₂CH(OH)CH₃) side chains. This substitution disrupts the extensive intra- and intermolecular hydrogen bonding typical of native cellulose, increasing the polymer's solubility in water and organic solvents. The balance between methoxyl and hydroxypropyl groups affects not only solubility but also viscosity, gelation temperature, and film-forming properties, allowing tailored applications depending on the substitution pattern [299–301].

Ahmad et al. (2022) synthesized HPMC from oil palm empty fruit bunches (OPEFBs) as a sustainable cellulose source. Initially, OPEFB-derived fibers underwent delignification in 12% (*w/v*) NaOH solution at 90–95 °C for 3 h, followed by bleaching with 10% (*w/v*) H₂O₂ at 80–90 °C for 1.5 h. The resulting cellulose pulp was washed to neutral pH and dried. For etherification, cellulose (5 g) was mixed with isopropanol (100 mL) and 10% (*w/v*) NaOH solution (20 mL) under stirring at 25 °C for 1 h to activate the hydroxyl groups. Subsequently, methylation and hydroxypropylation were carried out using methyl and hydroxypropyl reagents in the same heterogeneous medium. After reaction completion, the product was neutralized, washed, and dried to obtain HPMC. FTIR analysis confirmed the incorporation of methyl and hydroxypropyl groups; although the degree of substitution (DS) was not determined, the modified polymer exhibited properties suitable for food applications. HPMC coatings at 3% (*w/v*) reduced oil uptake in French fries by 16.09%, with changes in texture and moisture content, while a 1% coating provided the best sensory acceptance, indicating its potential for reducing lipid content in fried foods [302].

Rachmawati et al. (2020) synthesized hydroxypropyl methylcellulose (HPMC) from α -cellulose isolated from betung bamboo (*Dendrocalamus asper*) as a renewable lignocellulosic source. Initially, betung bamboo fibers underwent delignification with 17.5% (*w/v*) NaOH solution at 100 °C for 2 h, followed by bleaching with 10% (*w/v*) H₂O₂ in an alkaline medium to obtain high-purity α -cellulose. The α -cellulose was washed to neutral pH and dried. For etherification, α -cellulose (20 g) was dispersed in isopropanol (225 mL) and alkalinized with 20% (*w/v*) NaOH solution (50 mL) under stirring at 40 °C for 1 h to activate hydroxyl groups. Subsequently, methyl chloride (6.8 mL) and propylene oxide (11.2 mL) were added for simultaneous methylation and hydroxypropylation, and the reaction was maintained at 70 °C for 4 h in a heterogeneous medium. After reaction completion, the mix-

ture was neutralized with glacial acetic acid, filtered, washed with hot water and ethanol, and dried at 50 °C to obtain HPMC. The degree of substitution (DS) and molar substitution (MS) were determined as 1.14 and 1.42, respectively, indicating effective incorporation of both methyl and hydroxypropyl groups. The obtained HPMC was evaluated in topical gel formulations, showing good dispersibility, pH stability (5.43–5.57), and desirable viscosity profiles suitable for pharmaceutical applications [303].

In addition to applications in the food industry, HPMC finds widespread use in pharmaceutical formulations, where its biocompatibility, viscosity-enhancing, and gel-forming abilities are exploited for controlled drug release, tablet coating, and as a matrix former in hydrophilic matrix tablets [304,305]. In particular, for ophthalmic formulations, HPMC is employed in eye drops, gels, films, and as a coating agent [306]. HPMC also functions as a mucoadhesive polymer, capable of prolonging the gastric residence time of solid dosage forms. Moreover, one study proposes compressing the polymer with a contrast component to further extend its residence time and potentially enhance drug release [307]. Its ability to swell in aqueous media while maintaining mechanical integrity is particularly valuable for modifying drug dissolution rates and achieving sustained release profiles. Furthermore, due to its non-ionic nature, HPMC is compatible with a wide range of active pharmaceutical ingredients (APIs) and excipients.

Zhang et al. (2022) studied the application of HPMC as a dispersant for synthetic silver nanoflowers (AgNF) in flexible and antibacterial strain sensors composed of a multi-functional conductive polymer (M-CPC) manufactured from thermoplastic polyurethane (TPU). The role of HPMC is to ensure uniform distribution and strong adhesion to the TPU substrate through hydrogen bonding. Practical applications of these sensors include monitoring body movements, such as wrist, elbow, and knee flexions, with stable and accurate responses. The study highlights the superiority of M-CPC compared to traditional composites, such as TPU/AgNF, which exhibit lower stability and performance. The innovative use of HPMC as a connecting bridge between the substrate and conductive nanomaterials opens new possibilities for advanced wearable sensors [308].

The hydroxypropyl methylcellulose (HPMC) market has shown consistent growth, driven by its wide application in the pharmaceutical, food, cosmetics, and construction industries. Globally, the market was valued at approximately USD 2.1 billion in 2023 and is projected to reach around USD 3.5 billion by 2033, growing at a CAGR of ~5.3%. The pharmaceutical sector remains the leading consumer, where HPMC is extensively used as a binder, controlled-release agent, and coating polymer. The food industry represents a significant share, with HPMC employed as a stabilizer, emulsifier, and fat-replacement agent in processed foods. Geographically, Asia-Pacific dominates, largely due to strong pharmaceutical and construction industries in China and India, coupled with increasing demand for processed foods. North America and Europe hold substantial shares, driven by advanced pharmaceutical manufacturing and regulatory acceptance of HPMC in food and drug formulations. Meanwhile, emerging markets in Latin America and the Middle East are expected to experience accelerated growth, supported by expanding pharmaceutical infrastructure and rising urbanization. Overall, HPMC continues to consolidate its role as a multifunctional, sustainable polymer with high industrial relevance across multiple sectors [309].

Cellulose and its derivatives remain at the center of a strategic movement that combines scientific innovation, sustainability, and industrial development. Among the most promising routes are structural modifications of cellulose and hemicellulose—such as oxidation, crosslinking, and grafting—which provide new physicochemical properties and expand applications in functional materials, biomedicine, catalysis, and controlled-release systems. These advances highlight the potential of lignocellulosic biomass as a

renewable, high-value alternative, contributing to a circular bioeconomy. In parallel, the industrial outlook underscores the relevance of these biopolymers: the global market for HPMC, a key derivative, recorded a consumption of around 161.6 thousand tons in 2024, with projections to reach 236.1 thousand tons by 2033, generating between USD 1.2 and 4.6 billion annually, depending on the source. The Asia-Pacific region leads demand, driven primarily by the construction sector, followed by pharmaceutical, food, and cosmetic applications. This sustained growth emphasizes not only the industrial and commercial importance of HPMC but also reinforces the strategic value of research on cellulose polymer modifications. Thus, both scientific and market data converge on the same point: cellulose and its derivatives stand out as materials of both the present and the future. The strengthening of modification methodologies, combined with the expansion of global markets, demonstrates that this research field promotes academic advances while establishing solid foundations for technological innovation and industrial competitiveness on a global scale.

3.6. Carboxymethylcellulose (CMC)

Carboxymethylcellulose (CMC) is an anionic cellulose derivative renowned for its water solubility across a wide temperature range and biocompatibility. This polymer is widely used as a film former with moderate gas permeability. It is commercially applied as its sodium salt (CMCNa), produced by substituting some hydroxyl groups in cellulose with carboxymethyl moieties, mainly at the C2, C3, and C6 positions of glucose units. The sodium cation balances the charge of the deprotonated carboxylic group, increasing hydrophilicity and swelling [310]. Typical degree of substitution (DS) values range from 0.4 to 1.5, influenced by synthesis conditions; however, commercial CMC materials typically exhibit DS values between 0.5 and 1.0 [311].

The carboxymethylation process involves mercerization of cellulose with NaOH in alcoholic media (e.g., isopropanol or ethanol), followed by reaction with monochloroacetic acid under alkaline conditions, resulting in ether bonds that tether the carboxymethyl groups to the cellulose backbone [310,312–314]. CMC stands as one of the most relevant cellulose derivatives in industry worldwide, and its global market is projected to reach USD 1.57 billion, expanding to USD 1.97 billion by 2030 (CAGR of 4.64%) [315]. Market growth is driven by rising demand for clean-label food stabilizers, pharmaceutical coatings with abuse-deterrent properties, oil well drilling fluids, and lithium-ion battery binders. Its strong market position is supported by water solubility, tunable tolerance to heat and salts, and broad regulatory acceptance, ensuring reliable performance across diverse industrial processes. The remarkable physicochemical properties of CMC make it valuable across food, cosmetic, and pharmaceutical sectors as a thickener, viscosifier, gelling agent, adhesive binder, wetting agent, dispersant, and rheology modifier [42,212].

A major challenge in the production of CMC via conventional heterogeneous reactions is the inherently low and uneven degree of substitution, as amorphous domains are more reactive and accessible than crystalline regions, and only the outer surface of cellulose particles interacts directly with the reagents, limiting substitution to surface chains and leaving the core largely unmodified [316]. Additionally, efficient production of water-soluble CMC by heterogeneous carboxymethylation is often time-consuming, requiring harsh conditions and high hydroxide concentration [313]. Ambjörnsson et al. (2013) obtained CMC with a DS of 1.0 after a 48 h reaction at 40 °C using 27.5% (*w/v*) NaOH. Moussa et al. (2019) reported the highest DS in the literature (2.83) after 23 h using a 40% NaOH solution at 80 °C, with the product subsequently washed repeatedly with large volumes of an ethanol/water mixture (80:20 *v/v*%) [310,317].

This cellulose derivative is also the most extensively studied regarding its production from waste biomass and through more sustainable processes. Zininga et al. (2024) produced CMC from sugarcane bagasse using a combined approach of hydrothermal pretreatment, ammonium sulfite alkaline treatment, and bleaching with hydrogen peroxide. The subsequent heterogeneous carboxymethylation employed isopropanol, 28% NaOH, and monochloroacetic acid, yielding a CMC with a DS of 0.44 and 71% purity. This product effectively encapsulated the biocontrol agent *Trichoderma harzianum*, showing performance comparable to commercial high-purity CMC [318]. Similarly, Ndruru et al. (2024) isolated cellulose from coconut fibers via alkaline delignification and bleaching for CMC synthesis aimed at bioplastics. Using 50% NaOH, monochloroacetic acid, and isopropanol, they achieved DS values above 1.0. Bioplastics prepared from this CMC, further modified with carboxymethyl chitosan and glycerol as a plasticizer, displayed enhanced material properties [319].

Recent studies by Moura et al. (2024) reported the optimization of CMC synthesis from sugarcane bagasse and corn cob wastes using a D-Optimal design of experiments (DoE) [13]. The authors developed a more sustainable method in which cellulose extraction employed a mild combined diluted acid/peroxide–alkali (APA) treatment followed by bleaching, producing low-crystallinity pulps rich in amorphous and type II cellulose, which exhibited higher reactivity. The DoE evaluated the effects of activation time, reaction time, NaOH concentration, and cellulose source on CMC yield and water solubility. Statistical analysis (ANOVA, RSM) demonstrated high model precision and accuracy, showing that optimal conditions achieved up to 227% (*w/w*) yield of water-soluble CMC within 2:30 h of total derivatization time under milder conditions than typically reported, reducing costs and improving sustainability within a biorefinery framework. Other residual cellulose sources successfully applied for the production of this derivative include sugarcane leaves, coconut mesocarp, rice straw, oil palm empty fruit bunch, disposable paper cups, Kombucha tea fungal biomass waste, banana pseudostem, paper sludge, cotton stalk and linters, cacao pod husk, textile waste, among others [320–326]. In the early 2000s, Thomas Heinze and his research group were pioneers in the homogeneous carboxymethylation of cellulose using green solvents, especially inorganic molten salts (IMs) and ILs, such as LiCl/N,N-dimethylacetamide (DMAc), LiClO₄·3H₂O, DMSO/tetrabutylammonium fluoride (TBAF), and 1-N-butyl-3-methylimidazolium chloride ([C₄mim]Cl)/DMSO, with DS ranging from 0.4 to 2.09 [327–330].

Casarano et al. (2014) evaluated quaternary ammonium fluoride hydrates in DMSO as efficient solvents for cellulose dissolution and etherification, focusing on BMAF-0.1H₂O/DMSO for producing sodium CMC. Microcrystalline cellulose (MCC) was dissolved in BMAF-0.1H₂O/DMSO after preparing a fine NaOH dispersion, achieving a clear solution in ~20 min at room temperature under N₂. Carboxymethylation was performed by adding NaOH and sodium monochloroacetate, followed by reaction at 70 °C for 4 h. The CMC product was purified via precipitation, manual salt separation, dialysis, and lyophilization. This system achieved a DS of 1.85 for MCC, comparable to previous results with TBAF-3H₂O/DMSO (1.9–1.95), despite the lower water content of BMAF-0.1H₂O, due to the tightly bound water and strong electrolyte–cellulose interactions that maintained reactivity [331].

The study conducted by Akhlaq and Uroos (2025) reports a novel, energy-efficient one-pot method for synthesizing CMC in its acid form using tetrabutylphosphonium hydroxide (TBPH) IL, which simultaneously functions as both a solvent for cellulose dissolution and a base for etherification, replacing sodium hydroxide. MCC (20 wt%) was dissolved in TBPH (40 wt% in water) with isopropanol as a cosolvent, followed by reaction with sodium monochloroacetate at 60 °C for 3 h. The resulting CMC was isolated by filtration, washed,

and dried, achieving a high degree of substitution (DS = 2.1). Structural characterization confirmed successful etherification, while EDX analysis indicated the presence of counterions. Importantly, the TBPH was recovered by distillation and anion exchange, enabling reuse in successive batches, demonstrating the economic and sustainable potential of the method [313].

Due to its notable physicochemical properties, advanced CMC-based materials have been widely explored across diverse technological fields in recent literature. Environmental applications include sandy soil stabilization and stimulation, as well as composites and membranes for adsorption or separation of organic compounds such as fluoroquinolone antimicrobials and dyes, and salts and heavy metals such as Cu^{2+} , U^{4+} , Fe^{3+} , Zn^{2+} , and Pb^{2+} from water [332–339]. In the field of advanced biomedical materials, CMC is extensively employed as a bioink component for the fabrication of three-dimensional scaffolds with controlled architecture, enabling precise cell encapsulation and tissue engineering applications, since its high water-retention capacity and gel-forming ability also make it suitable for scaffolds used in tissue regeneration, where it supports cell adhesion, proliferation, and extracellular matrix deposition [340–343]. In drug delivery systems, CMC serves as an effective carrier for the sustained and targeted release of therapeutics such as antibiotics, anti-inflammatories, and vitamins, benefiting from its adjustable degree of substitution and solubility [344–348]. Beyond its conventional applications in the food industry, it finds use in the composition of active food-preserving edible coatings to increase postharvest quality of fruits and vegetables [349–352].

Optoelectronic devices for optical sensors, solar cells, polarizers, screens, and energy storage are a class of energy materials with notable applications of CMC. This derivative is often employed as the host polymer matrix for the preparation of more eco-friendly composites with nanoparticles, which impart the optical properties to the device [353–355]. It also has promising applications in materials for anodes, separators, and polymer electrolytes in Zn-ion and Li-ion batteries, and in the emerging field of adhesive wearable sensors [312,356–359].

The research carried out by Sun, Xu, and Maimaitiyiming (2025) aimed to develop a biocompatible, self-adhesive, and multifunctional hydrogel sensor based on egg white (EW), gelatin, and NaCMC for applications in wearable electronics, electronic skin, and motion detection. The hydrogel (EWg-CMC0.3) was prepared through a simple, room-temperature physical crosslinking process using varying ratios of EW, gelatin, and CMC, followed by direct-ink-writing (DIW) 3D printing. The resulting hydrogel exhibited strong self-adhesion (255 kPa), rapid self-healing (25 min, healing rate = 224.5%), high swelling capacity, tunable mechanical strength, and excellent printability. Functionally, the sensor demonstrated stable and reproducible strain, stress, temperature, and humidity sensing, maintaining performance under 20% strain for 500 cycles. It enabled reliable detection of complex human motions (e.g., joint bending, facial expressions) with real-time and consistent signal outputs, as well as potential applications for joint disease monitoring and interactive human–computer interfaces. These results highlight the promising potential of EW/gelatin/CMC hydrogels as edible, eco-friendly, and versatile platforms for next-generation wearable sensors and electronic skin technologies [359].

Overall, CMC stands out as the most relevant cellulose derivative due to its versatility, tunable properties, and broad acceptance across industrial applications. However, despite its established position, the field faces critical challenges and opportunities that must be addressed to expand its technological impact. Conventional heterogeneous syntheses remain limited, underscoring the urgency for scalable and greener methodologies. At the same time, the valorization of agricultural residues and industrial byproducts as sustainable feedstocks offers a promising path toward circular bioeconomy strategies,

although further efforts are needed to bridge laboratory-scale demonstrations and industrial deployment. Equally important is the exploration of CMC in its salt forms or as complexes with alternative cations beyond sodium, such as Fe^{3+} , Zn^{2+} , Ca^{2+} , and Al^{3+} , which could unlock novel physicochemical features such as enhanced ionic conductivity, stability, or bioactivity, expanding its use in advanced materials like smart hydrogels, optoelectronics, and next-generation energy storage devices [360–362].

3.7. Benzyl Cellulose (BC)

Benzyl cellulose is a cellulose derivative produced by substituting the hydroxyl groups (-OH) of the anhydroglucose units with benzyl groups ($-\text{CH}_2\text{C}_6\text{H}_5$). This chemical modification introduces bulky hydrophobic aromatic moieties into the cellulose polymer, profoundly affecting its physicochemical properties. The degree of substitution (DS), which represents the average number of substituted hydroxyl groups per glucose unit, is a critical parameter influencing the solubility, thermal stability, and mechanical characteristics of BC. At low DS, benzyl cellulose is soluble mainly in strong polar aprotic solvents such as dimethyl sulfoxide (DMSO) and pyridine. As DS increases, solubility extends to less polar organic solvents such as chloroform and tetrahydrofuran (THF), broadening its processing and application possibilities [363,364]. The benzylation reaction generally involves nucleophilic substitution under alkaline conditions, where cellulose reacts with benzyl chloride to form benzyl ethers and release chloride ions.

Beyond its molecular structure, benzyl cellulose is typically synthesized from purified cellulose extracted from lignocellulosic biomass such as wood pulp, cotton, and agricultural residues. The quality of the cellulose feedstock, determined by its purity and molecular weight, significantly affects the efficiency of benzylation and the properties of the resulting derivative. Traditional production has predominantly used wood pulp due to its abundance and purity. However, recent trends emphasize sustainable biomass sources to improve the environmental footprint and reduce costs [364–366]. Pretreatment methods such as alkaline extraction and bleaching play an essential role in isolating cellulose suitable for modification without compromising polymer integrity.

The benzylation process is typically carried out as a heterogeneous reaction in aqueous or mixed solvent media, where mercerized cellulose is treated with benzyl chloride under controlled alkaline conditions. This method requires optimization of variables like temperature, reagent concentrations, and reaction time to achieve the desired DS while minimizing cellulose degradation. Homogeneous benzylation reactions in solvents such as DMSO provide enhanced reagent accessibility, resulting in more uniform substitution patterns and improved product quality [365,367]. Innovations in catalytic and microwave-assisted benzylation methods also show promise for increasing reaction efficiency and reducing environmental impact, making the process more sustainable [368].

Ramos et al. (2005) synthesized benzyl cellulose via a homogeneous route by dissolving cellulose in DMSO/TBAF·3H₂O and reacting it with benzyl chloride in the presence of NaOH, obtaining products with a degree of substitution of up to 2.85 [369]. In a subsequent study, Casarano et al. (2014) described the dissolution of the BMAF salt in DMSO under stirring at 60 °C in a nitrogen inert atmosphere. Cellulose was then added, ensuring its complete solubilization. Simultaneously, a NaOH dispersion in DMSO was prepared and incorporated into the cellulose solution. Benzyl chloride was subsequently added, maintaining the reaction under nitrogen for 4 h, resulting in the formation of benzyl cellulose [331].

Abe et al. (2017) reported an innovative route for the homogeneous benzylation of cellulose, eliminating the conventional alkaline activation step with NaOH and achieving conversion in only 10 min. The cellulose was initially dissolved in an aqueous solution

of tetra-*n*-butylphosphonium hydroxide (47%) under stirring at 25 °C for 30 min. Subsequently, benzyl bromide was added as the benzylating agent in a controlled dropwise manner, while maintaining gentle stirring under an inert N₂ atmosphere. Upon completion of the reaction, the product was characterized for its DS, yielding values higher than 2.5 for the benzyl cellulose obtained [370].

Saliu et al. (2017) investigated the regeneration of native cellulose that had been previously benzylated, crosslinked, and incorporated with TiO₂ nanoparticles, forming a hydrophobic composite. This material exhibited a high capacity for absorbing organic solvents, including toluene, xylene, chloroform, kerosene, and gasoline, reaching values of up to 27.22 g/g. The results demonstrate its promising potential for applications in the remediation of organic-contaminant spills [371]. Shibano et al. (2020) investigated the use of benzylcellulose derivatives as functional polymers in undoped organic light-emitting diodes (OLEDs). Through a Steglich-type esterification, a series of copolymers designated as TBC-X were synthesized, in which a host component, derived from carbazole (Cz-COOH), and a light-emitting component, derived from phthalimide (BCz-PI-COOH), were chemically incorporated into the structure of 2,3-di-O-benzylcellulose. Among the obtained copolymers, TBC-10 stood out for its good solubility in organic solvents, such as chloroform and toluene, as well as for the formation of thin and homogeneous films via spin-coating. Furthermore, the material exhibited thermally activated delayed fluorescence (TADF) and, when employed as the emitting layer in OLED devices, achieved an external quantum efficiency of 5.9%. These results highlight the potential of benzylcellulose as a versatile platform for the development of advanced optoelectronic materials [372].

Benzyl cellulose stands out among cellulose derivatives for its high chemical modifiability, allowing the introduction of benzyl groups that confer solubility in organic solvents and tunable hydrophobicity. Although classical homogeneous routes offer high degrees of substitution, they may still present limitations in terms of reproducibility and handling of sensitive reagents. Recent advances in homogeneous methodologies and more controlled reaction conditions have promoted greater uniformity in modification. Strategies that reconcile substitution efficiency, selectivity, and sustainability establish benzyl cellulose as a promising material for biomaterials, enantioselective separation systems, and high-performance functional materials.

3.8. Cyanoethyl Cellulose (CEC)

Cyanoethyl cellulose (CEC) is a polymer with excellent physical and chemical properties, including high thermal resistance, remarkable mechanical characteristics, and strong microbial resistance [373]. Its properties vary according to the degree of substitution (DS) of the cyanoethyl group (-CH₂-CH₂-CN), influencing parameters such as the dielectric constant and solubility in organic solvents. CEC is synthesized through a Michael addition reaction, in which pre-alkalized cellulose reacts with acrylonitrile. When the DS is low, CEC exhibits lower thermal stability, good mechanical properties, and high moisture retention capacity. Conversely, a high DS imparts a high dielectric constant and low dielectric loss, making it promising as an insulating medium for capacitor miniaturization [374–376].

In the study by Joshi et al. (2017), office waste paper (OWP) was converted into CEC through a heterogeneous route, involving functionalization in an alkaline medium and etherification with acrylonitrile under different reaction conditions to control the degree of substitution. The authors aimed to optimize the synthesis by varying sodium hydroxide and acrylonitrile concentrations, as well as the time and temperature of the alkalization and cyanoethylation steps. This optimization resulted in a DS of 2.70, higher than that obtained

by conventional methods, reducing the reaction time and increasing the efficiency of the etherifying agent transfer while utilizing waste as a raw material [377].

In a recent and innovative study, Ansari et al. (2023) proposed the synthesis of CEC from pine needle biomass, using an optimized etherification reaction through the Taguchi experimental design to maximize the degree of substitution. The authors investigated six key process variables: alkalization time and temperature, alkali concentration, cyanoethylation time and temperature, and acrylonitrile concentration. To extract cellulose, pine needles were treated with an aqueous solution of NaClO_2 (0.9% *w/v*) and CH_3COOH (3:1 *w/v*). Subsequently, cyanoethylation was carried out via the heterogeneous route. Optimization resulted in a CEC with a high DS (2.67), demonstrating that pine needles are an excellent renewable source for CEC production and that process optimization significantly enhances the efficiency of the cyanoethylation reaction [378].

Wang et al. (2011) investigated the synthesis of this derivative through a homogeneous route using the solvent method. Cellulose was initially solubilized in different systems, with the toluene/isopropanol (85/15) mixture showing the best performance. Under these conditions, the controlled addition of cyanoethylene, followed by heating at 50 °C and stirring for 2 h, enabled the substitution reaction, which was subsequently stopped by neutralization with acetic acid. The product was then washed and dried. In a subsequent step, cellulose reacted with acrylonitrile in the presence of sodium hydroxide, maintained at 50 °C for 4 h, resulting in cyanoethylation. After neutralization, precipitation in boiling water, and drying, the final CEC was obtained [379].

Mi et al. (2022) reported, for the first time, the synthesis of CEC via a Michael addition reaction between acrylonitrile and the activated hydroxyl groups of cellulose, employing a CO_2 -switchable solvent. Initially, cellulose was dissolved in a mixture of CO_2 -switchable solvent and DBU, yielding a homogeneous solution. In the subsequent step, the cellulose intermediate reacted with acrylonitrile through Michael addition, leading to the formation of CEC. Owing to the absence of water in this system, the resulting polymer exhibited high stability, enabling its separation and purification simply by precipitation in methanol [380].

Wang et al. (2019) investigated the dielectric properties of porous CEC membranes, synthesized via a Michael addition reaction and obtained through a solvent-free phase separation method. The study analyzed how variables such as the type of coagulation bath, polymer concentration, and molecular weight of this derivative influence the morphology, structure, porosity, and electrical behavior of the membranes. It was found that increasing the polymer concentration in CEC/DMF solutions reduced porosity, a feature favorable for applications in liquid and gas filtration processes. On the other hand, the decrease in porosity led to an increase in the dielectric constant, highlighting the strong correlation between the porous structure and electrical performance. This relationship endows CEC with great potential for devices that require precise control of dielectric properties, such as sensors, capacitors, and other advanced electronic systems [375].

Wu et al. (2022) investigated the fabrication of flexible composite films based on CEC integrated with boron nitride nanosheets (BNNs), using a solution casting approach. This technique allowed the fabrication of highly uniform and high-quality films, which exhibited remarkable energy storage performance. The composites achieved a discharged energy density of up to 23.5 J/cm^3 , significantly surpassing films based solely on cellulose. Additionally, they demonstrated high conversion efficiency (83.6%) and excellent dielectric strength, withstanding electric fields of up to 680 MV/m without noticeable degradation. Another relevant aspect was the cyclic stability: even after 10^5 charge–discharge cycles, no significant performance loss was observed, indicating robustness for long-term applications. The combination of high dielectric constant, breakdown strength, and rapid discharge

response (≈ 60 ns) underscores the potential of these films for high-efficiency, high-energy-density capacitors, establishing a new paradigm for high-performance natural polymer capacitors with reduced environmental impact [381].

Chen et al. (2025) conducted a pioneering study on the application of this polymer in electrowetting displays, exploring its use as a novel dielectric layer in bilayers with Hyflon (HF). Its incorporation reduced the driving voltage from 17 to 9 V in the HF/CEC system, a value that meets the critical voltage required by integrated driver circuits for liquid crystal displays. This significant voltage reduction highlights the dielectric efficiency of CEC, as well as its potential as a functional hydrophobic material in electrowetting devices, opening new perspectives for the development of low-energy-consumption electronic devices and expanding the scope of cellulose derivatives in advanced optoelectronic technologies [382].

CEC stands out as a cellulose derivative of high scientific and technological relevance, combining the abundance of cellulose with remarkable functional versatility. The introduction of cyanoethyl groups provides significant improvements in solubility, processability, and compatibility with different polymeric matrices, thereby expanding its potential applications. However, its synthetic route still requires further advances, particularly regarding the use of more sustainable solvents and reagents, as well as the precise control of the degree of substitution, since these factors directly affect the final properties of the material. In terms of applications, it has demonstrated great potential: it acts as a matrix in eutectogel electrolytes; it forms flexible hybrid films such as CEC/BaTiO₃/GO; and it contributes to the development of triboelectric nanogenerators [382–384]. Thus, cyanoethyl cellulose is recognized both as a functional derivative of cellulosic biomass and as a strategic material for advancing innovative and environmentally sustainable technologies.

4. Hemicellulose Esters

4.1. Hemicellulose Acetate (HCA)

Hemicellulose acetate is a chemically modified biopolymer obtained through the esterification of hemicellulose hydroxyl groups (-OH) with acetate groups (-COCH₃). This modification introduces acetyl substituents into the sugar units of hemicellulose, such as xylose and arabinose, leading to structural and property changes. The resulting polymer exhibits higher solubility in organic solvents, including acetone and chloroform, as well as improved thermal stability compared to native hemicellulose [8,385]. The primary synthesis method involves acetylation reactions using acetic anhydride or acetyl chloride under basic conditions, typically catalyzed by pyridine or sodium hydroxide (NaOH). In this process, hydroxyl groups are converted into ester linkages, reducing hydrophilicity and altering mechanical behavior [35]. Solvents such as dimethylacetamide (DMAc), dimethyl sulfoxide (DMSO), and water are often employed, depending on the solubility and reactivity of the hemicellulose [6,35]. Reaction parameters—including temperature, reaction time, and catalyst type—play a crucial role in determining the degree of acetylation and, consequently, the properties of the resulting material. For instance, hemicellulose can be treated with acetic anhydride in the presence of sulfuric acid or hydrochloric acid as catalysts, under high-temperature conditions (100–150 °C) for several hours, to promote efficient acetylation and achieve tailored properties [386].

Wang et al. (2017) investigated the phthalate derivatization of hemicellulose extracted from bagasse biomass, achieving degrees of substitution ranging from 16.37% to 52.14%, while employing 1-allyl-3-methylimidazolium chloride (AmimCl) as an ionic liquid to monitor reaction behavior. Their findings confirmed that chemical modification enhanced material properties and broadened potential applications. Furthermore, Sun et al. (2021) proposed a bioconversion strategy using *Saccharomyces cerevisiae* to

metabolize hemicellulose acetate, demonstrating improved Acetyl-CoA generation that facilitates the biosynthesis of high-value compounds such as polyketide triacetic acid lactone (TAL) and vitamin A, revealing the interplay between chemical modification and microbial metabolism [386].

Akkus et al. (2018) synthesized HCA from xylans extracted from corn cobs as a ligno-cellulosic source. Hemicellulose was first isolated via alkaline extraction with KOH, during which potassium acetate (KAc) was generated as a byproduct. Instead of removing this salt, the authors exploited its catalytic effect to promote acetylation using acetic anhydride under reflux, without additional catalysts or solvents. The degree of substitution, expressed as weight gain, reached values up to 23.5% depending on reaction time and temperature. Acetylation markedly reduced water solubility (from 46% to ~2%), decreased moisture uptake, and improved thermal stability (maximum degradation temperature shifted from 283 °C to 349 °C). These modifications resulted in HCA with enhanced hydrophobicity and processing stability, making it a promising candidate for applications in biodegradable packaging and other moisture-sensitive polymer systems [387].

In recent years, several studies have explored applications of hemicellulose and its acetylated derivatives beyond the production of biodegradable films. One promising field is their use as emulsifiers and stabilizers in oil-in-water systems, where the natural degree of acetylation was shown to be critical for the solubility and emulsifying capacity of xylans and arabinoxylans, highlighting their potential in food and cosmetic formulations [388]. In the food sector, the controlled hydrolysis of acetylated xylans has been investigated for the production of xylo-oligosaccharides (XOS) with prebiotic properties, opening new opportunities as functional food ingredients [389].

In the field of biomaterials, acetylated hemicellulose has also shown remarkable versatility. For instance, Berglund et al. (2020) developed biomimetic hydrogels based on acetylated glucuronoxylan incorporated into bacterial cellulose, creating a model that mimics the plant cell wall and enables the investigation of the biomechanical roles of these macromolecules. Similarly, hydrogels prepared from naturally acetylated galactoglucomannan (AcGGM) exhibited thixotropic and strong shear-thinning behavior, features highly valuable for use as rheology modifiers in foods, cosmetics, and biomaterials [390]. In the biomedical context, Wang et al. (2022) reported the development of an injectable, responsive hydrogel obtained from the combination of AcGGM and chitosan through a thiol-ene reaction. The material proved promising for controlled drug delivery, offering biocompatibility and tunable mechanical properties [391]. In colloidal systems, xylan nanocrystals derived from hemicellulose were applied to stabilize highly stable Pickering emulsions, with potential for bioactive encapsulation and the formulation of green emulsions in chemistry and food applications [392].

In recent years, acetylated hemicellulose has been widely investigated for the development of biodegradable films, especially in food packaging and barrier applications. Mugwagwa and Chimphango (2020) demonstrated that hemicellulose films reinforced with acetylated nanocellulose and coated with polycaprolactone exhibited enhanced hydrophobicity, reduced solubility, and lower migration, making them suitable for contact with fatty foods [393]. The use of naturally O-acetylated galactoglucomannan (GGM) has also been explored for latex-based films. Yong et al. (2022) reported that GGM latex films showed high transparency, water contact angles of ~117°, good flexibility, and excellent barrier properties, making them promising as coatings for paper and cardboard substrates [394]. More recently, Nizam et al. (2025) further advanced this approach by developing free-standing films from GGM-derived latex, emphasizing their potential as bio-based barrier materials [395].

For xylan-based derivatives, Martins et al. (2024) produced starch–xylan acetate films with improved oil and water vapor resistance, positioning them as bioplastic candidates for sustainable packaging [396]. Similarly, Zhang et al. (2024) optimized the acetylation degree of xylan to increase “filmability,” producing soluble and freestanding films with promising barrier and mechanical properties [397].

In conclusion, HCA emerges as one of the most versatile and strategically relevant derivatives of hemicellulose. The acetylation process, by introducing acetyl groups into the hydroxyl sites of sugar units such as xylose and arabinose, produces significant structural changes that directly impact solubility, thermal stability, and mechanical performance [398]. These modifications overcome the intrinsic limitations of native hemicellulose, including high hydrophilicity and poor processability, thereby enabling its incorporation into high-value applications. Research advances highlight the ability of acetylated hemicellulose to function across diverse sectors, ranging from functional foods and biomedical systems to advanced biomaterials and packaging technologies. In particular, the development of acetylated hemicellulose films has proven especially promising, as acetylation enhances hydrophobicity, barrier properties, and mechanical resistance—critical factors for food preservation and sustainable packaging applications [399]. Composite systems, such as starch–xylan blends and naturally acetylated galactoglucomannan (GGM) latexes, further demonstrate the adaptability of this derivative, providing renewable alternatives to petroleum-based polymers [400].

Beyond packaging, hemicellulose acetate also shows potential in biomedical and pharmaceutical applications, particularly in controlled drug delivery systems, where its tunable hydrophilicity and biodegradability can be exploited to modulate release profiles effectively. Together, these findings confirm acetylation as a key chemical strategy for tailoring hemicellulose into multifunctional and eco-friendly materials. However, despite notable progress, further research is still required to optimize reaction conditions, control the degree of substitution, and scale up sustainable acetylation processes using greener solvents and milder catalysts.

4.2. Hemicellulose Propionate (HCP)

Hemicellulose propionate is a derivative synthesized by introducing propionate groups ($-\text{COC}_2\text{H}_5$) into the hemicellulose polymer, which significantly modifies its physical and chemical properties. This modification enhances solubility in solvents such as acetone and tetrahydrofuran (THF), while improving thermal stability and flexibility, making it a suitable material for films, coatings, and other industrial applications [401]. The esterification reaction is typically carried out using propionic anhydride in the presence of solvents such as DMAc, DMSO, or water, with catalysts including pyridine or sulfuric acid to promote the formation of ester bonds [55,401]. These reaction conditions allow precise control over substitution degree and polymer properties, adapting the material to specific functional requirements.

Recent studies have explored the biological impact of hemicellulose propionate and its components. Gao et al. (2025) investigated the fermentation behavior of different hemicellulose fractions using fecal inoculum from suckling piglets, finding that specific hemicellulose components influenced the microbial composition, particularly bacteria such as *Prevotella_9* and *Parabacteroides*. These bacteria play essential roles in producing short-chain fatty acids, including acetate, propionate, and butyrate, the latter being critical for host immune function and gut barrier integrity. These findings suggest that dietary supplementation with hemicellulose derivatives could improve animal health by modulating microbiota and metabolite production [401].

In conclusion, hemicellulose propionate emerges as a promising derivative with valuable physicochemical properties and potential biological benefits. However, despite advances in understanding its solubility, thermal behavior, and interactions with gut microbiota, the literature remains limited in systematic studies dedicated to its synthesis. Most reports address the use of propionic anhydride under conventional esterification conditions, yet comprehensive investigations aimed at optimizing reaction parameters, controlling substitution patterns, and tailoring molecular structures for specific applications are still lacking. Therefore, further research focused on synthetic methodologies is essential to fully unlock the potential of hemicellulose propionate as a versatile and high-value biomaterial [402].

4.3. Hemicellulose Sulfate (HCS)

Hemicellulose sulfation occurs through the modification of free hydroxyl groups at the C-6, C-2, and C-3 positions of each anhydroglucose unit (AGU), representing a simple and efficient strategy for producing sulfated analogs with diverse bioactivities [403,404]. In particular, sulfated hemicellulose, such as xylan sulfate, can be obtained from lignocellulosic biomass—for example, corn cobs or sugarcane bagasse—through chemical functionalization of the extracted xylan. In this route, hemicellulose is first isolated via alkaline or oxidative treatments, yielding a high-purity xylan fraction composed predominantly of β -(1 $\rightarrow$ 4)-D-xylose units, along with minor amounts of arabinose and uronic acids [405].

Sulfation is typically achieved via nucleophilic substitution using sulfating agents such as the sulfur trioxide–pyridine complex or chlorosulfonic acid in polar aprotic solvents (e.g., DMF/LiCl), enabling the introduction of sulfate ester groups onto the hydroxyl moieties of the xylan backbone. Process innovations, including continuous-flow microreactor systems, significantly enhance reaction efficiency by reducing diffusion limitations and reaction times, while preserving high molecular weight and achieving controllable and uniform degrees of substitution. Structural characterization confirmed successful incorporation of sulfate groups, evidenced by characteristic S=O and C–O–S absorptions. The sulfated xyans thus produced exhibit enhanced bioactivity, including potent anticoagulant, antimicrobial, and antioxidant properties, highlighting their potential for biomedical, pharmaceutical, and functional material applications [406].

Ragab et al. (2018) synthesized HCS from lignocellulosic biomass by chemical sulfation of extracted xylan fractions. Initially, hemicellulose was isolated through alkaline treatment to obtain high-purity xylan composed mainly of β -(1 $\rightarrow$ 4)-linked D-xylose units with minor arabinose and uronic acid substitutions. The purified xylan was then subjected to sulfation using chlorosulfonic acid (CSA) in an organic solvent medium under controlled temperature and reaction time. This process introduced sulfate groups ($-\text{OSO}_3^-$) at the C-2, C-3, and C-6 hydroxyl positions of the anhydroxylose units. The DS varied between 0.25 and 1.05, depending on the reaction parameters. The sulfated derivative displayed strong anticoagulant activity, comparable to heparin, as well as excellent water solubility and stability. These findings highlight the potential of HCS as a bioactive polysaccharide for pharmaceutical and biomedical applications, particularly in drug delivery and anticoagulant therapy [403].

Both CS and HS derivatives are water-soluble and exhibit biocompatibility while possessing the ability to form polyelectrolyte complexes. These characteristics make them versatile materials with various industrial and biomedical applications, such as drug delivery systems, antiviral, anticoagulant, and antimicrobial agents, stabilizers in cosmetic formulations, water remediation, and in the production of edible films and coatings for food [77,405–411]. While traditional sulfation methods, including heterogeneous and ho-

homogeneous reactions in conventional solvents, remain well established for CS production, they often involve harsh chemicals, long reaction times, and generate significant waste, limiting their environmental sustainability and industrial scalability. The recent adoption of ILs and DES for homogeneous sulfation represents a promising green alternative, offering improved cellulose dissolution, more uniform substitution, and milder reaction conditions [78,412].

The body of research on hemicellulose sulfate (HCS) demonstrates its remarkable potential as a multifunctional derivative with broad biomedical, pharmaceutical, and industrial applications. However, most advances remain concentrated on laboratory-scale sulfation methods that rely on harsh reagents, extended reaction times, and generate significant chemical waste, raising concerns regarding environmental sustainability and large-scale feasibility. Although emerging strategies involving ILs and DES provide greener alternatives with improved efficiency and control over substitution, studies specifically addressing scalable and sustainable synthesis routes for HCS from lignocellulosic biomass are still scarce.

5. Hemicellulose Ethers

5.1. Carboxymethyl Hemicellulose (CMH)

Carboxymethyl hemicellulose (CMH) represents a chemically modified derivative of hemicellulose, a major component of plant cell walls. Hemicellulose itself is a heterogeneous group of polysaccharides that, alongside cellulose and lignin, contribute to the structural integrity and flexibility of plant cells. The carboxymethylation of hemicellulose introduces carboxymethyl groups ($-\text{CH}_2\text{COOH}$) into the polysaccharide backbone, enhancing its solubility in water and organic solvents, and imparting functional groups that can participate in further chemical modifications. This modification not only improves the material's processability but also tailors its physicochemical properties for specific applications.

The synthesis of CMH typically involves the reaction of hemicellulose with chloroacetic acid or its sodium salt under alkaline conditions. Recent studies have optimized this process to achieve high degrees of substitution, which are crucial for enhancing the solubility and reactivity of the resulting product [413]. For instance, a study by Tohamy et al. (2025) developed a rapid method to produce CMH from sugarcane bagasse, utilizing microwave-assisted carboxymethylation. This approach not only shortened the reaction time but also improved the yield and quality of CMH, demonstrating the potential of microwave irradiation in polysaccharide modification [414].

Bai et al. (2023) employed ultrasound-assisted derivatization of bamboo hemicellulose, achieving a degree of substitution (DS) of 0.59 for CMH. This method notably enhanced the solubility and processability of the material, yielding physicochemical properties comparable to those of commercial carboxymethylcellulose (CMC) [415]. The carboxymethylation of holocellulose fractions, encompassing both cellulose and hemicellulose, represents a promising strategy to improve industrial efficiency and promote the valorization of lignocellulosic waste. More recently, de Souza et al. (2025) developed a rapid and environmentally friendly one-pot synthesis of carboxymethyl holocellulose (CMHC) derived from pineapple crown and mango seed tegument. By applying a mild extraction protocol coupled with optimized carboxymethylation conditions designed through a D-Optimal experimental design, the process achieved a high degree of substitution, up to 1.63, and mass yields reaching 246.9%, within a total reaction time of 2 to 15 h [11].

The applications of CMH are diverse, owing to its enhanced solubility, reactivity, and biocompatibility [414]. In the biomedical field, CMHC has been explored as a component in drug delivery systems, wound dressings, and tissue engineering scaffolds. Its ability to form

hydrogels through physical or chemical crosslinking makes it suitable for controlled-release applications. In the environmental sector, CMH-based materials have been investigated for water purification, as they can adsorb heavy metals and organic pollutants from aqueous solutions [416]. Moreover, CMH composites with other biopolymers or nanoparticles have shown promise in food packaging, offering biodegradable alternatives to synthetic polymers [417].

5.2. Methyl Hemicellulose (MHC)

Methyl hemicellulose (MHC) is a chemically modified derivative of hemicellulose, a key heteropolysaccharide found in plant cell walls that contributes to the structural matrix by interacting with cellulose and lignin [418]. Native hemicellulose exhibits limitations such as high hydrophilicity and low thermal stability, which restrict its direct use in industrial and material applications [419]. Methylation, typically performed using methyl iodide or dimethyl sulfate in polar aprotic solvents with alkaline catalysts, modifies hemicellulose by substituting hydroxyl groups with methoxy groups, thus reducing hydrophilicity and enhancing solubility in organic solvents [8,205]. The DS is crucial in defining the physicochemical properties of MHC, including improved compatibility with hydrophobic polymer matrices and altered thermal behavior [420].

Given the increasing demand for renewable and sustainable materials, MHC offers a promising pathway for valorizing lignocellulosic biomass by producing functionalized biopolymers suitable for biodegradable composites, coatings, and films [421]. This modification expands the industrial applicability of hemicellulose, overcoming native material limitations while aligning with green chemistry principles. Advances in methylation techniques and characterization have demonstrated the potential of MHC as a versatile material, driving innovation in biopolymer development and sustainable material science [6].

In the study by Viera et al. (2007), hemicellulose was extracted from sugarcane bagasse through an alkaline treatment aimed at efficiently solubilizing the heteropolysaccharide fraction while minimizing structural degradation. Pretreatment removed extractives and reduced lignin content, followed by controlled sodium hydroxide treatment to cleave ester linkages between hemicellulose and lignin, facilitating polysaccharide release. Hemicellulose was then precipitated from the alkaline extract using ethanol, yielding a material primarily composed of xylans with variable arabinose, glucose, galactose, and uronic acid contents. The purified hemicellulose was methylated in dimethyl sulfoxide (DMSO) using methyl iodide as the methylating agent and sodium hydride as the catalyst. Reaction conditions were optimized to achieve a high degree of substitution (DS), indicating extensive methylation of hydroxyl groups. FTIR and $^1\text{H}/^{13}\text{C}$ NMR spectroscopy confirmed successful methylation, revealing characteristic methyl ether absorptions and uniform substitution along the polymer backbone. Methylation imparted altered physicochemical properties to the hemicellulose, notably reducing hydrophilicity and enhancing solubility in organic solvents, which may improve compatibility with hydrophobic polymer matrices. Thermal analysis indicated modest modifications in degradation behavior, suggesting that substitution influences the thermal stability of the polymer [205].

5.3. Cyanoethyl Hemicellulose (CEHC)

Cyanoethyl hemicellulose (CEH) is a hemicellulosic derivative obtained through the introduction of cyanoethyl groups into the hemicellulosic backbone, typically via nucleophilic addition of hydroxyl functionalities to the vinyl group of acrylonitrile under alkaline conditions. This functionalization significantly modifies the physicochemical

properties of the parent polymer, enabling enhanced reactivity, tunable solubility, and potential for further chemical transformations [422]. Due to the strong electron-withdrawing character and low steric hindrance of the nitrile group, CEH serves as a versatile precursor for the synthesis of advanced functional materials, including carbamoylethyl, carboxyethyl, and amine-functionalized derivatives [423]. These derivatives exhibit potential applications in liquid crystalline materials, biosensors, separation technologies, heavy metal adsorption, and polymer-based composites. The reactivity of nitrile groups toward hydrolysis, reduction, and alkylation broadens the scope of CEH beyond direct utility, positioning it as a valuable intermediate in the design of novel biopolymer-based materials from renewable resources [424].

In the study by Cao et al. (2013), cyanoethyl hemicelluloses were obtained from xylan-rich hemicelluloses of *Dendrocalamus membranaceus* through nucleophilic addition of acrylonitrile in aqueous NaOH. The effects of temperature, reaction time, acrylonitrile/anhydroxylose (AX) ratio, and alkali concentration on the degree of substitution ($DS_{C\equiv N}$, $DS_{C=O}$) were systematically evaluated. Optimal cyanoethylation occurred at $\sim 40^\circ\text{C}$ for 1 h; higher temperatures or prolonged reaction times caused substituent loss and increased carbonyl formation via acrylonitrile hydrolysis. Increasing acrylonitrile concentration raised $DS_{C\equiv N}$ while reducing $DS_{C=O}$, whereas excess NaOH ($>1.5\text{ mol/mol AX}$) decreased $DS_{C\equiv N}$ through alkaline cleavage. ^1H and ^{13}C NMR confirmed successful cyanoethyl incorporation, with preferential substitution at C-3 over C-2, and demonstrated hydration to carbamoylethyl derivatives. Thermal analysis indicated lower stability of CEH compared to native hemicelluloses, attributed to decomposition of the cyanoethyl branches. As observed in previous studies, reaction temperature, time, and the molar ratio between acrylonitrile and anhydroxylose units significantly impacted the DS, which ranged from 0.23 to 1.64 throughout the study [425].

6. Literature Related to Cellulose Derivatives

Selected studies are summarized in Table 1, which presents various applications of the main cellulose derivatives discussed in this work. This compilation highlights the specific potential of each modification, emphasizing how their physicochemical properties directly influence the choice of the most suitable derivative for applications in packaging, biomaterials, pharmaceuticals, and materials engineering.

Table 1. Polysaccharide ester and ether derivatives from several feedstocks and their applications.

Derivative	Biomass Feedstock	Extraction Method	Yield (<i>w/w</i> %)	DS	Application	Ref.
Cellulose Acetate	Olive tree pruning	Acid hydrolysis	43.6	2.4	Dye adsorption	[121]
	Cigarette butts	Soxhlet (acetone/ethanol), alkaline hydrolysis	32.1	2.28	Membrane for water filtration	[426]
	Cotton linters	Acetylation with acetic anhydride and catalyst	85.6	2.45	Biodegradable packaging films	[427]
	Eucalyptus bark	Alkaline + bleaching + acetylation	48.2	2.56	Bioplastics	[428]
	Sugarcane bagasse	Alkaline, bleaching, acetylation	54.0	2.34	Drug delivery systems	[429]
	Butia odorata fruit	Alkaline + NaClO ₂ bleaching + acetylation	49.4	-	Reinforcement in polymer matrices	[430]
	Commercial CA (39.7% acetyl, Mw ~50,000, Sigma-Aldrich, St. Louis, MI, USA)	Dissolution in ternary solvent system (acetone/DMF/water, 3:2:1) followed by electrospinning	–	39.7% acetyl content	Membranes for water treatment, biomedical uses, filtration, and as support for CNF reinforcement	
	Bleached eucalyptus pulp (Suzano, São Paulo, Brazil)	Mechanical refining (PFI mill, 10,000 rev.) + chemical pretreatment with 50% oxalic acid (1 h, 90 °C) + high-pressure homogenization (300 bar, 5 passes) + centrifugation for nano/microfraction separation	48.9% nanofibrillation yield	–	Reinforcement in CA membranes; improved tensile strength (~383%), higher Young's modulus (up to 580 MPa), enhanced hydrophobicity and functionalization (Eu ³⁺ luminescence)	[119]
	Sunflower seed shell	Phase inversion + AC hybrid with CA	-	-	Hybrid membranes for dye removal	[431]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Cellulose Triacetate	Corn Stover	Ionic liquid	82.8	2.8	Synthesis of cellulose triacetate	[123]
	Cocoa Pod Husk	Acid hydrolysis	31.8	-	Nanoparticles applied to chitosan films	[111]
	Sugarcane bagasse	Organosolv + acetylation (acetic anhydride)	62.5	2.8–2.9	Food packaging films	[432]
	Licorice root waste	Alkaline + peroxide bleaching, acetylation	-	2.9	Electrospun nanofibers for filtration	[433]
	Eucalyptus wood	Acid hydrolysis + acetylation	-	2.9	Barrier coatings in multilayer films	[29]
	Corn husk	Alkaline treatment + acetylation	78.6	2.85	Biodegradable composite films	[434]
	Cotton fibers (CF)	Heterogeneous acetylation with glacial acetic acid, acetic anhydride, H ₂ SO ₄ ; isolation by differential solubility	AP: 112%/CTA: 87%	2.86	CTA soluble in chloroform and CHCl ₃ /MeOH (9:1); pore diameter ~256 nm	[435]
	Recycled writing paper (RWP)	Same as above	AP: 94%/CTA: 80%	2.84	CTA soluble in chloroform; pore diameter ~83 nm	[435]
	Recycled newspaper (RN)	Same as above	AP: 84%/CTA: 68%	2.85	CTA soluble in chloroform; pore diameter ~142 nm	[435]
	Macerated woody fibers (MWFL)	Delignification (H ₂ O ₂ + AcOH, 60 °C, 48 h) → heterogeneous acetylation (Ac ₂ O + H ₂ SO ₄ in AcOH) → isolation by solubility	AP: 73%/CTA: 55%	2.89	CTA soluble in chloroform; pore diameter ~108 nm	[435]
	Kapok fibers	Alkaline + bleaching + acetylation	-	2.9	Oil-absorbing membranes (PVDF/CTA blend)	[436]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Cellulose Acetate Propionate	Commercial Cellulose	Chemical	71.5–88.4	1.3–3.0	Cellulose propionate production	[189]
	Commercial Cellulose	Alkaline and bleaching	120.0	2.8	Transparent and flexible films	[51]
	Commercial cellulose	Alkaline	24.3	0.4–0.2	Sustainable packaging	[27]
	Commercial CAP	Solubilization in acetone/DMF + electrospinning	-	-	Nanofibrous membranes for filtration and wound dressing	[437]
	Corncob cellulose	Alkaline delignification + acetylation/propionylation	58.3	2.4–2.7	Bioplastic films for food packaging	[438]
	Bagasse cellulose	Steam explosion + chemical modification	65	2.6	Coating material for controlled drug delivery	[29]
Cellulose Sulfate	Commercial cellulose	Alkaline and bleaching	75.3	2.4	Adsorption of dyes	[74]
	Raw jute fiber	Sulfonation with chlorosulfonic acid (CSA) in DMF	54.1–44.2	-	Ammonium removal	[439]
	Cotton, microcrystalline cellulose	Sulfation with H ₂ SO ₄ /Alc or ClSO ₃ H/pyridine	-	0.58–2.98	Antibacterial agent	[67]
	Cotton (sulfonated)	Sulfonation with chlorosulfonic acid (CSA) in DMF	-	-	Pb(II) removal from water	[440]
Cellulose Phosphate	Commercial CMC	Alkaline	88.1	0.8	Binding agent for drugs	[94]
	Bleached softwood pulp	Phosphoric acid + urea (thermal), comparison of methods	-	0.18–0.20 P/AGU (via ³¹ P NMR)	Fiber modification and structural analysis	[441]
	CNC from cotton	Phosphoric acid + urea (solid-state reaction)	-	High phosphate incorporation	Enhanced dispersion, electrochemical sensors	[442]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Cellulose Phosphate	Rice straw	Delignification + phosphorylation (DAHP/urea)	43–51	Charge density: 1488–2199 mmol/kg	Heat-sealable films, antifizzing cups	[443]
	Wood pulp, MCC	Phosphate-based ILs with H ₂ O (e.g., [Emim]DEP)	-	High swelling ratio 40.1% (no DS reported)	Pretreatment for dissolution	[444]
Cellulose Nitrate	Unconventional feedstocks	Alkaline	Flaxseed: 8.5 Oats: 13.4 Gram: 10.2	Flaxseed: 0.2 Oats: 0.6 Gram: 0.4	Biodegradable packaging	[147]
	<i>Miscanthus × giganteus</i>	Alkaline and acid hydrolysis, nitration	116–131	11.35–11.83% N	Energetic materials, biosensors	[148]
	<i>Miscanthus × giganteus</i> (KAMIS)	Industrial nitric acid method	150	11.18% N	Inks, lacquers, adhesives	[445]
	Date palm fronds (DPF)	Sulfonitric nitration	-	OCN: 12.59%, MCCN: 13.17%	Solid propellants	[446]
	Commercial CA and CN	Electrospinning, carbon black doping	-	12.1% (CN)	Oil spill adsorption	[447]
	Cellulose nitrate (12.6% N)	Acetone-based mixing, milling with ARM	-	12.6% N (in NC)	Combustion enhancement, propellants	[448]
	Screw pine	Ionic liquid	-	0.1	PVDF filler for membranes	[164]
Cellulose Benzoate	Regenerated cellulose	Benzoylation in DMAc/LiCl	-	~2.4	Transparent, moisture-resistant films	[161]
	Cotton-based cellulose	Benzoylation in pyridine	~89	2.40–2.45	UV-shielding, biodegradable films	[165]
	Microcrystalline cellulose	Esterification in AmimCl ionic liquid	-	-	Microplastic removal from water	[166]
	Various cellulose sources	Benzoylation via deep eutectic solvents (DES)	-	DS up to 3.0	Green solvent chemistry, functional films	[49]
	Polysaccharide derivatives	Theoretical modeling of benzoyl/phenyl substitution	-	Substitution at 2,3,6	Chiral selectors in chromatography	[449]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Cellulose Acetate Butyrate	Commercial Cellulose	Solid-phase	-	-	Microsphere material	[180]
	Cellulose (plant-based)	Acetate method (emulsion-solvent evaporation)	-	DS ~2.9 (CAB100: DSAc 0.0; DSBu ~2.9)	Supercapacitor electrodes	[174]
	CAB + CuO nanoparticles	Pulsed laser ablation in liquid (PLAL)	-	-	2-Nitrophenol reduction in wastewater treatment	[450]
	Commercial CAB (various grades)	Solvent casting (acetone)	-	DS range: DSBu 0.76–2.54; DSAc 0.16–2.03	Film development for packaging/coating	[183]
	-	Drop casting on the perovskite layer	-	-	Improves perovskite solar cell stability	[451]
Cellulose Propionate	CP/LA porous film	Solvent casting + lactic acid + water pressure	-	-	Battery separator with straight pores	[452]
	CP/[N4444][SS] film	Solvent casting with ionic liquid	-	-	Barrier film (water/pressure resistance)	[453]
	Cotton linters (>95% cellulose)	Direct propylation with propionic anhydride using N-iodosuccinimide (NIS) as catalyst under solvent-free conditions	61.42–94.19%	1.32–2.85	Industrial use in plastics, coatings, films, optical materials, and pollutant removal	[194]
	Rice husk	Alkali treatment (NaOH), bleaching (NaOCl, H ₂ O ₂), then propylation with propionic anhydride using NIS catalyst	59.59–86.44%	1.76–3.00	Same as above; higher DS makes it suitable for high-value industrial esters	[194]
	Wheat straw	Alkali treatment (NaOH), bleaching (NaOCl, H ₂ O ₂), then propylation with propionic anhydride using NIS catalyst	48.92–69.46%	1.60–3.00	Used in plastics, fibers, membranes, coatings; lower yield but high DS achievable	[194]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (<i>w/w</i> %)	DS	Application	Ref.
Cellulose Propionate	Purchased polymer	Dissolution in DMAc, casting with additives (methanol, oxalic acid, maleic acid, etc.), phase inversion in water	Not reported as <i>w/w</i> % (evaluated by flux/retention)	DS fixed at 2.66 (supplied polymer)	Ultrafiltration membranes: dye removal, salt separation, oil-water separation, protein filtration	[454]
	CP/SMAC/Mo nanocomposite	Pulsed laser ablation (with SMAC)	~91 total mass loss	-	Dielectric and optoelectronic devices	[455]
Hemicellulose acetate	Birchwood xylan	Alkaline aqueous (DMF or solvent-free)	Up to 85	0.9–2.0	Bioplastics, coatings, bioactive polymers	[456]
	Switchgrass	Alkaline extraction with NaOH	27	Up to 2.9	Hydrophobic films (brittle)	[457]
	Sugarcane bagasse	One-pot IL-based transesterification	-	~2.0 (estimated)	Thermoplastics with good flow & strength	[372]
	Lignocellulose	Direct functionalization in ILs	-	~1–3	Functional polymers, recyclable plastics	[458]
Ethyl and hydroxyethyl cellulose	Rice straw cellulose (RSC)	Acid–base extraction of rice straw → ethylation (ethyl bromide, toluene)	79.60	2.0–2.5	Biodegradable films (EC–ethanol films)	[372]
	Commercial ethyl cellulose (purchased)	Microencapsulation of APP with EC (in-pulp addition; coating)	-	-	Flame-retardant cellulose paper	[459]
	Commercial EC + HPMC	Dissolve EC in oil (heat) ± mix with HPMC (oleogelation)	-	-	Low-saturated oleogel shortening (food)	[460]
	Commercial EC	One-step emulsification (microfluidic O/W template) + solvent evaporation	-	-	Microcapsules for sustained curcumin release (drug delivery)	[461]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Ethyl and hydroxyethyl cellulose	Commercial EC	Casting with polysulfide (inverse vulcanization composite)	-	-	Food preservation films; antibacterial/packaging	[234]
	Commercial EC + gelatin	Electrospinning (EC/gelatin matrix with anthocyanin, ε-polylysine)	-	-	Intelligent packaging/pork freshness monitoring	[462]
	Commercial EC	Microencapsulation (EC shells) loaded with octadecyl amine; added to epoxy coatings	-	-	Self-healing/ anti-corrosion coatings	[463]
	Hydroxyethyl cellulose (HEC)	Single-step grafting with (3-aminopropyl) triethoxysilane (APTES)	-	-	Antimicrobial/modified HEC materials	[464]
	Air particle vacuum dust (APVD)	NaOH and H ₂ O ₂	-	1.1	-	[246]
	HEC	Emulsion polymerization/grafting to PVAc (SLS emulsifier)	-	-	PVAc wood adhesive stabilizer/improved wet strength	[465]
Benzyl Cellulose	Commercial cellulose	Modular synthesis from cellulose phenyl carbonate with benzyl amine	-	2.45	Enantioselective membrane filtration	[466]
	Various lignocellulosic sources	Homogeneous benzylation in ionic liquids	>90	2.0–2.5	Optical and thermal materials	[370]
	Microcrystalline cellulose	Solvent-based benzylation using benzyl chloride	85–95	1.8–2.2	Hydrophobic coating materials	[467]
	Wood pulp	Heterogeneous benzylation	~80	1.5–2.0	Compatibilizer in polymer composites	[367]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Carboxymethylcellulose	Coconut mesocarp	Sequential alkaline extraction + organosolv pretreatment	22	NR	CMC production; thickener/emulsifier	[321]
	Commercial/ not specified	Added as a co-stabilizer in AgNP synthesis	-	NR	AgNP stabilizer; antibacterial formulations	[468]
	CMC (substrate)	In situ co-precipitation of Fe salts on CMC (pH 11)	-	NR	Paper coating (magnetic); improved barrier	[469]
	Sugarcane bagasse	Hydrothermal pretreatment + alkaline extraction + MCA etherification	-	0.44	Encapsulation; packaging/coating	[470]
	Waste disposable paper cups	Alkali extraction of fiber + MCA etherification	-	1.21 (max.)	Composites; flexible materials	[324]
	Banana peel	Delignification + bleaching + hydrolysis; MCA etherification	152.65	0.61	Binder/thickener; valorize agro-waste	[471]
	Coconut fibers	Alcohol medium carboxymethylation (alkaline + MCA)	9.45 g (reported; not given as %)	1.82	Superabsorbent; emulsifiers	[472]
	Pineapple leaf cellulose	NaOH swelling + MCA (isopropanol) etherification	136.6	2.3	High-solubility CMC; functional materials	[473]
	Corn (corn husk)	Heterogeneous carboxymethylation; optimized particle size	240	2.41	High-DS CMC for advanced applications	[474]
	Sugarcane bagasse (hemicellulose)	Microwave-assisted carboxymethylation; N-CDs embedding	-	NR	Fluorescent hydrogel biosensor; antibacterial/antifungal	[414]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Hemicellulose propionate	Suckling Piglets	-	-	-	Bioengineering	[401]
	Poplar chips	Delignification with NaClO_2 (75 °C, pH 3.2, 4 h) → extraction with 8% NaOH + 1% $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$ (16 h)	32.9–80.2%	0.32–1.51 (propionate typically ~1.1–1.2)	Biodegradable plastics, resins, films, food coatings, hydrophobic materials	[475]
Hemicellulose hydrolysate succinate	Cassava Starch	Bioconversion	98.0	-	Pure D-(-)-lactic acid production	[476]
	Wheat straw	Extraction with aqueous NaOH , followed by purification and precipitation	~65–85%	0.9–1.3	Biodegradable plastics, films, resins, and compatibilization with hydrophobic polymers	[477]
Cyanoethyl cellulose	Cotton linter pulp	Homogeneous NaOH /urea dissolution → reaction with acrylonitrile (freeze–thaw then AN add, neutralize, lyophilize)	-	1.64–2.12	Lyotropic liquid-crystal studies/optics	[478]
	Sugarcane bagasse (bagasse → HEC → CEC)	Mercerization/HEC formation then cyanoethylation with acrylonitrile in alcoholic NaOH (room temp, 2 h)	-	1.64	Dielectric conducting films, optoelectronics, thermoelectrics	[479]
	Natural cellulose (unspecified source)	Michael addition (AN)/etherification-controlled reaction time; homogeneous route (NaOH /urea style)	-	DS 2.19–2.62 (samples CEC-0.5h, CEC-1.5h, CEC-3h)	Battery binders (LCO cathodes), enhanced adhesion/ion transport	[480]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Cyanoethyl cellulose	Cotton linter pulp (α -cellulose >95%)	Homogeneous synthesis in 7 wt% NaOH/12 wt% urea (pre-chill), then AN addition; phase-inversion to make CEC-based cathode	-	0.42	Phase-inversion cathodes—interpenetrating ion/electron paths (Li batteries)	[481]
	Commercial CEC	Commercial CEC (DS specified by supplier) blended with rGO by solution blending	-	2.55–2.80	rGO/CEC composites for microwave absorption (electromagnetic wave absorbers)	[482]
	Cellulose (used to functionalize garnet filler in PVDF electrolyte)	In situ polymerization/coordination of $C\equiv N$ groups on garnet surface $\rightarrow$ ultrathin PCEC layer on LLZTO	-	-	Interface modification for PVDF/LLZTO composite solid electrolyte (solid-state batteries)	[483]
	Sugarcane bagasse	Synthesis and film casting	-	-	Conducting films	[481]
	Cotton linter pulp	Homogeneous cyanoethylation (NaOH/urea), then phase inversion to form a porous electrode	-	0.42	High-energy lithium metal cathodes (improved porosity, Li-ion diffusion)	[205]
Methylcellulose	Sugarcane bagasse	-	-	0.7–1.2	-	[206]
	Mango seed fibers	Delignification with nitric acid/ethanol	-	1.3–0.5	Mortar additive	[484]
	Commercial cellulose	Etherification	-	Hydroxypropyl & methyl substitution (varies by MW)	Electrospun fibers or electrosprayed particles for food/nano use	[485]
	Cellulose + Satureja khuzestanica oil	Emulsification and casting	-	-	Antimicrobial edible films for food packaging	[482]
	HPMC + Sodium alginate	Surface pretreatment on silk	-	-	Textile printing (enhanced dye uniformity & efficiency)	[486]

Table 1. Cont.

Derivative	Biomass Feedstock	Extraction Method	Yield (w/w%)	DS	Application	Ref.
Methylcellulose	Commercial cellulose	Etherification	-	%Me and %HP (variable)	Controlled drug release tablets	[487]
	Cellulose + PCL + β -carotene	Nanoprecipitation and casting	-	-	Active packaging with antioxidant release	[488]
	Methylcellulose + walnut oil + κ -carrageenan	Emulsion-templated method	97.37	-	Delivery of curcumin	[489]
	HPMC + vinyl chloride	Suspension copolymerization	-	Double-bond functionalization	Antifouling ultrafiltration membranes	[490]
	Methylcellulose + polyethyleneimine	Chemical conjugation	-	Varies by PEI molecular weight	Doxorubicin drug delivery	[491]
Hydroxypropyl cellulose	Commercial cellulose	Etherification with propylene oxide	-	53.4–80.5	Hollow nanocapsules for drug/antioxidant delivery	[492]
	Commercial cellulose	Etherification, then S–O–O emulsion + evaporation	66–81	-	Microcapsules for tryptophan taste-masking	[493]
	Commercial cellulose	Spray freeze drying with DMSO	-	-	Amorphous solid dispersion for aprepitant	[494]
Hydroxypropyl methylcellulose (HPMC)	Commercial HPMC	(Study of commercial product)	-	-	Controlled-release tablets	[487]
	HPMC, Carvedilol	Freeze-drying and inkjet printing	-	-	Poorly soluble drug delivery	[48]
	HPMC, Casein	Electrospinning	-	-	Biodegradable nanofibers	[116]
	HPMC, shellac, CNTs, ZnO-NPs	Crosslinking and loading	-	-	Antimicrobial packaging films	[224]
Hemicellulose succinate	Hemicellulose (oil-palm fruit bunch)	Metabolic engineering of <i>E. coli</i>	-	-	Production of succinic acid	[476]
	Hemicellulose (xylan)	Consolidated bioprocessing (<i>E. coli</i>)	-	-	Production of succinate	[495]

7. Conclusions and Research Prospects

Cellulose- and hemicellulose-derived esters and ethers have demonstrated great potential as sustainable alternatives to petroleum-based polymers, offering biodegradability, tunable properties, and broad applicability in packaging, coatings, biomedical systems, and energy-related materials. Despite these advances, significant challenges remain in optimizing their production, functionalization, and large-scale application. Conventional heterogeneous modification routes often result in limited degrees of substitution, poor regioselectivity, and reduced material uniformity, while homogeneous systems, although more efficient, are still restricted by solvent cost, recyclability issues, and environmental impact.

Future research should prioritize the development of greener, cost-effective, and recyclable solvents, such as ionic liquids, deep eutectic solvents, and molten salt systems, with an emphasis on improving their recovery and reducing toxicity. Valorization of residual biomass as feedstock represents another underexplored area for some derivatives, requiring strategies to efficiently process and fractionate lignocellulosic wastes while maintaining cellulose and hemicellulose integrity. Attention must be given to the development of scalable, well-controlled reactions for hemicellulose derivatives to overcome structural heterogeneity and reactivity challenges, enabling their translation from promising laboratory-scale prototypes to industrial applications comparable to those of cellulose derivatives.

In terms of applications, future studies should explore the structure–property relationships of polysaccharide derivatives, as well as the integration of cellulose and hemicellulose derivatives into advanced material systems, such as nanocomposites, smart hydrogels, stimuli-responsive membranes, and energy storage devices. Biomedical applications such as controlled drug delivery, real-life applied tissue engineering scaffolds, and antimicrobial systems represent particularly promising but still underdeveloped areas. Establishing techno-economic and life cycle assessments will be crucial to determine the industrial viability of these derivatives, ensuring that emerging processes align with circular economy principles.

Addressing these topics will not only expand the scientific understanding of cellulose and hemicellulose functionalization but also accelerate their transition into sustainable, high-performance materials with real industrial and societal impact. Furthermore, regulations for use in sensitive sectors, such as pharmaceuticals and food, still pose barriers. Biomass-derived biopolymers represent a strategic link between innovation and sustainability, but their success depends on overcoming technical, economic, and regulatory obstacles. The convergence of advanced research, clean technologies, and cross-sector collaboration will be essential for their full development in biorefineries.

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