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Lignin-derived bionanocomposites as functional food packaging materials

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Harnessing lignin, which is the second most abundant biopolymer and is cost-effective, biocompatible, and nontoxic, could be a promising alternative to conventional food packaging materials. Each year, millions of tons of lignin are produced, and it is commonly used as a low-value fuel by being burned. However, this inexpensive and abundant bioresource biomass has the potential to be utilized as food packaging materials. It is crucial to explore lignin-based renewable resources to facilitate the shift towards an environmentally friendly materials circular economy. Recent research has shown that lignin-based materials possess excellent anti-oxidant and anti-bacterial properties, in addition to good mechanical and antiviral properties, UV light barrier, and enhanced thermal properties, making them suitable candidates for use as food packaging materials. This study aims to provide current perspectives on the use of lignin based materials for food packaging applications. The article provides a critical analysis of the physicochemical characteristics, processing techniques, and extraction and structural features of lignin from various sources as well as its derived materials. Additionally, it outlines the latest trends in converting lignin into lignin nanoparticles. This comprehensive review concludes with future perspectives on lignin based materials for food packaging applications.

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Sustainability spotlight

In today's world, where environmental concerns are at the forefront, lignin, a sustainable biomass, has the potential to revolutionize various industries and alleviate ecological burdens. In the pulp and paper industry, lignin is produced as a byproduct that can be utilized to reduce waste and develop innovative solutions for food packaging materials. By employing lignin's properties, industries can contribute to a more sustainable future by minimizing the waste of petroleum-based materials and developing eco-friendly packaging alternatives. Additionally, lignin-derived materials in food packaging can significantly reduce the food industry's carbon footprint. Through ongoing research and development, lignin has the potential to catalyze transformative change in various industries and pave the way for a greener and more sustainable future.

Introduction

Lignin, a prevalent aromatic polymer, reinforces plant cell walls alongside cellulose and hemicellulose. Recent data have revealed that the paper sector generates over 50 million metric tons of biomass byproducts yearly.[1,2] However, 98% of this organic matter is burned for power generation or ends up in wastewater systems, which negatively affects the ecosystem.[3,4] Approximately 2% of the biomass material remains suitable for industrial use, particularly for producing beneficial lignin-based products. Additionally, one-fifth of the planet's total biomass consists of lignin.[5] Researchers have discovered that upon combination, these substances form a reinforced structure resembling a supramolecular scaffold, providing cells with chemical and physical resilience to fight against infections and deterioration caused by enzymes and chemicals.[6] Plants like jute, cotton, wood pulp, and industrial hemp possess intermediate lamellae that store a significant amount of lignin (up to 35%). The various types of lignin available in the market include kraft lignin (9%), lignosulfonates (88%), and the newly developed sulfur-free organosolv (2%). If current trends continue, it is projected that lignin will be valued at $913.1 million by 2025.[7,8] Lignin has numerous industrial applications, such as food packaging materials, drug delivery systems, tissue engineering scaffolds, and sterile biomedical device components.[9–11] Its intrinsic UV shielding properties, anti-oxidant capacity, chemical stability, and physical toughness also enhance the value of consumer goods.[12]

Lignin is a polymer composed of three monolignols: syringyl (S) units, guaiacyl (G) units, and *p*-hydroxyphenyl (H) units. Lignin biomolecules include spatially organized phenolic groups, such as sinapyl, coniferyl, and *p*-coumaryl, which enable a variety of functional entities and connections.[13,14] The heterogeneity of lignin arises from the interaction between its H, G, and S units, resulting in a range of functional groups and

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connections. The aryl ether-O-4 linkage accounts for approximately 50% of all connections.[15]

In the past, lignin-containing industrial waste materials and their byproducts were the primary sources of organic components and energy. The structural foundation of lignin is formed by the S, G, and H subunits, which are produced when phenylpropanoid units undergo radical coupling.[16] Due to its heterogeneous nature, recovering lignin in its native form is challenging, making it difficult to obtain structural details. However, recent advancements in near-perfect recovery of milled wood lignin (MWL) and cellulolytic enzyme lignin (CEL),[17] have partially resolved this issue. Previous research on lignin was limited to the pulp and paper industry, as it was typically discarded as waste in that sector. As a result, studies focused on chemical profiling and structural elucidation.[18]

Lignins are polysaccharide-bound compounds that exhibit uneven distribution patterns and are intertwined with cellulose and hemicellulose in their natural state.[18] The physicochemical characteristics of lignin are influenced by various factors, including the plant source, extraction method, treatment parameters, and other elements.[19] Lignin-rich byproducts from industries and agriculture, such as paper and pulp,[20] bagasse,[21] wood, agricultural waste products, and other organic wastes,[22] are abundant in lignin. In contrast, organic wastes like grass, some plant pieces, and other organic wastes produce very little lignin.[15]

Recently, there has been a growing interest in lignin-derived composites due to their nanoforms, which enhance durability and provide significant value to materials such as fabrics and rubber.[23] Native lignin, when considered as a single entity, is unable to provide the expected interaction and performance-boosting properties compared to nano-sized lignins.[24] Nanolignins have demonstrated improved water sensing, mechanical, UV shielding, and thermal performance capabilities when impregnated with gluten-containing nanostructures. In contaminated water systems, chitosan and nanolignins have shown methyl orange scavenging ability up to 83%. According to a study, the lignin nanoparticle shape offers improved stability and dispersibility for up to two months. Without using any chemicals, evenly distributed nanolignins can be produced by sonic irradiation.[25] Silver ion-loaded nanolignins can be filled with biodegradable and environmentally acceptable cationic poly-electrolyte layers to create silver nanoparticle fillers. When combined with silver ions, the poly-electrolyte surface kills a variety of bacterial species, particularly quaternary-amine-resistant *E. coli*, *Pseudomonas aeruginosa*, and *Ralstonia* sp. It also enables interaction with the bacterial cell membrane.[26]

In recent patented work, cryogel is produced by embedding lignin nanoparticles in crosslinked gelatin at subfreezing temperatures, resulting in a densely crosslinked microporous composition with pores size between 50–50 μm. This unique combination of collagen-derived natural polymer (gelatin) and natural biowaste polymer (lignin) enhances mechanical performance and shape recovery rate while exhibiting strong free radical scavenging action and inhibiting bacterial growth.[27] In another patent, developed a novel method to modify lignocellulosic materials, particularly wood, while preserving their original architectural design. By employing partial delignification and filling of the material's structures, a new composite material is produced that exhibits desirable characteristics. This innovative approach creates possibilities for the development of advanced materials with diverse applications, which could significantly impact various industries.[28] Therefore, we believe that it is essential to review the applications of lignin derived environmentally friendly and biodegradable materials with excellent attributes for food packaging. To the best of our current knowledge, a comprehensive review of the lignin structure, sources, extraction and processing techniques, and applications in food packaging materials has not yet been published.

Structure and sources

Lignin, in comparison to cellulose and hemicellulose, possesses a considerably more complex structure.[29] It is composed of three fundamental monolignol building blocks, namely phenylpropanoid units (C9-units) *p*-hydroxyphenyl (H), guaiacyl (G), and syringyl (S), which are polymerized by radicals in plants to form lignin.[30] The C9 formula, which represents the average lignin unit composition, is often written as $C_xH_yO_z(OCH_3)n$. These monolignols are interconnected through a variety of carbon–carbon bonds (b-1, b-5, b–b, 5–5, b-6) and inter-unit connections of ether (b-O-4, a-O-4, 4-O-5) to form a three-dimensional macromolecular structure of lignin.[31] Like cellulose, lignin lacks a regular structure and is highly chemically heterogeneous due to the irregular mixing of varying quantities of monolignols and various patterns of substitution on their phenylpropanoid units.[32] Lignin's polymeric structure has not yet been fully understood. Some researchers propose that lignin should be classified as a random copolymer rather than a biopolymer due to its lack of a repeating monomer in its structure.[33,34] Furthermore, nearly all naturally occurring lignin is connected to carbohydrates, primarily hemicelluloses.[35] Approximately 40–60% of the inter-unit connections in lignin are composed of bond of b-O-4 of the ether.[36] It is important to note that the structure of lignin is significantly influenced by the extraction process, genetic species, culture site, and harvesting season of the parent plant. In terms of the factors that influence the structure of lignin, hardwood lignin is primarily composed of G and S units while a significant portion of G units make up the majority of softwood lignin. The makeup of monolignols and how they are linked together in lignin is determined by the species or source, as demonstrated by the H : G : S ratios in lignins from radish and carrot roots[37] and grass.[38] Additionally, lignins from various sections of a plant have distinct fine structures, as seen in Chinese quince lignin.[39] Furthermore, the method used to remove lignin affects its fine structure, as shown by the differences in the composition of monolignols and their relationships to cellulose enzymatic lignins, ethanol, alkali, and milled wood.[40] The physicochemical differences between lignin and cellulose or hemicellulose are significant. Unlike cellulose, lignin is amorphous in the solid state.

Lignin is a material that resembles approximately spherical particles that only slightly dissolve in solvents.[41,42] Its

insolubility in practically all aqueous solutions is widely recognized.[43] Measurements of polydispersity (M_w/M_n), number average molecular weight (M_n) and weight average molecular weight (M_w) were frequently made in regards to molecular mass. However, these values varied greatly among the various lignins, sometimes by several orders of magnitude. For example, Hatakeyama[44] found that the kraft lignin from beech (*Fagus crenata*) had M_w, M_n, and M_w/M_n values of 8020 g mol^{-1}, 1840 g mol^{-1}, and 4.35, respectively. Nevertheless, Stark, Yelle, and Agarwal[45] reported that the ponderosa pines (*Pinus ponderosa*) kraft lignin was 4.49 × 107, 3.82 × 107, and 1.18, respectively.

The main issue with lignin solutions during molecular mass measurement is their solubility in organic solvents. According to Brunow and Lundquist,[46] a true lignin solution is typically difficult to create and the insoluble portion must coexist. Given that lignin's soluble part is significantly lower than its insoluble portion, there is inevitable ambiguity. Comparably, there is a wide range of documented lignin thermal stabilities concerning the glass transition temperature, with values falling between 89.9 and 174 °C.[47] Mainly, the physicochemical characteristics of lignin are dependent on their source.[48,49]

According to Erdocia *et al.*,[50] the primary functional group found in lignin is the hydroxyl group, which is essential for both its reactivity and how it interacts with surrounding molecules, such as water. Roughly 10% to 13% of all aromatic rings are made up of free phenols.[51] The relevance of lignin's safety for use in a variety of foods is particularly significant for the food industry.[52]

The utilization of agro-processing residues and plants as sources of lignin is of great importance. In India, the production of lignin is facilitated by the large amounts of biowaste generated by the ayurvedic industry. Additionally, other sources of lignin include pine varieties such as red clover, bagasse from sugar cane, lucerne, elephant grass, grass species like *Festuca arundinacea*, Brazilwood, and several bamboo species like *Bambusa vulgaris* & *Chusquea oxylepis*.[53]

There are numerous advantages to using agricultural byproducts for synthesizing lignin, including environmental, financial, and technological benefits. Lignocellulosic materials from crops such as sugar cane, maize, rice, and wheat supply the majority of biomass for the global agro-industry, while other crops make up a small portion of this biomass. The production of bioethanol from lignin-rich agricultural byproducts using enzymes and genetically modified yeasts can contribute to energy security in an environmentally friendly manner. The disposal of biowastes from the Ayurvedic medicinal industry contributes to the large amounts of industrial biomass that accumulate in landfills. Research supports the use of leftover lignin-containing plant-based products. The team's water-miscible bioactive potential has previously been demonstrated using herbal residues left behind by the Ayurvedic medical sector.[22] Traditional solvents, such as water, milk, ghee, oil, *etc.*, listed in Ayurvedic scriptures do not extract all of the phytochemicals from herbal remnants, indicating that there is still potential for further extraction. The research conducted by Vinardell and colleagues focused on the investigation of lignin compounds derived from *Acacia nilotica*, which is commonly referred to as the babul tree, and their biological significance.[54]

Preparation methods

Previously, it was commonly understood that plant-based meals contained higher levels of lignin in their non-edible parts, such as seed coverings, cereal husks, and sugarcane bagasse, than in their consumable parts. As a result, the majority of food-grade lignins are derived from these fibrous by-products. To develop a valuable food ingredient and product with added value, an efficient and cost-effective method of separating lignin from these byproducts must be employed. The conventional lignin isolation process involves pulverizing, separating, purifying, and drying. During the pulping process, lignin is released, and the polysaccharide bonds are broken.[55] The existing lignin isolation technologies may be broadly classified into two groups (Fig. 1) based on the lignin's physical condition during the pulping process.[56] The first group involves extraction techniques that dissolve and completely disperse the lignin in the resultant pulp. In the second group, lignin appears as an insoluble residue in the pulp produced (leaving operations). However, the application scenarios for these two techniques are not well established. Lignin is often measured using a gravimetric technique following isolation due to its low reactivity and inert character.[57] Ideally, the optimal isolation process would yield chemically unchanged lignin that is free of non-lignin impurities. However, no existing technique can meet all of these conditions, despite significant efforts to investigate potential isolation approaches to preserve the original structure of lignin.[58] In other words, the physicochemical characteristics and structure of lignin are largely determined by the isolation process used. The most common way to refer to lignin is by the method used to separate it, such as milled wood lignin (MWL) or lignin that has been hydrolyzed, alkali, sulfite, or organosolv processed.

The process of creating milled wood lignin (MWL) begins with dry biomass being ground in a ball mill until it passes through a 0.50 mm screen, which mechanically breaks the connections between lignin and polysaccharides. Next, the powdered material is dissolved in toluene and extracted using a natural solvent with less than 4% water content, which can be ethanol, acetic acid, or dioxane.

Finally, the lignin found in the natural solvent–water mixture can be recovered as sediments by slowly adding the mixture into deionized water. According to El Hage *et al.*, Holtman *et al.* and Rencoret *et al.*, this method of preparation falls into the first group.[60–62] Although the yield of isolated MWL is often low compared to other techniques, this method has the least impact on lignin structure, as noted by Bjorkman.[63] As a result, MWL is frequently used in structural evaluations but is not suitable for commercial use. On the other hand, lignin can also be produced from unprocessed biomass through substantial acid hydrolysis[64] or hydrolysis with enzymes[40] to break down carbohydrate components and liberate lignin. After the removal of the broken-down carbohydrates, lignin is left as an insoluble residue, which is why this method of preparation falls into the second group.

Cellulase is a commonly utilized enzyme in enzyme-mediated hydrolysis, as reported in a recent study.[40] However,

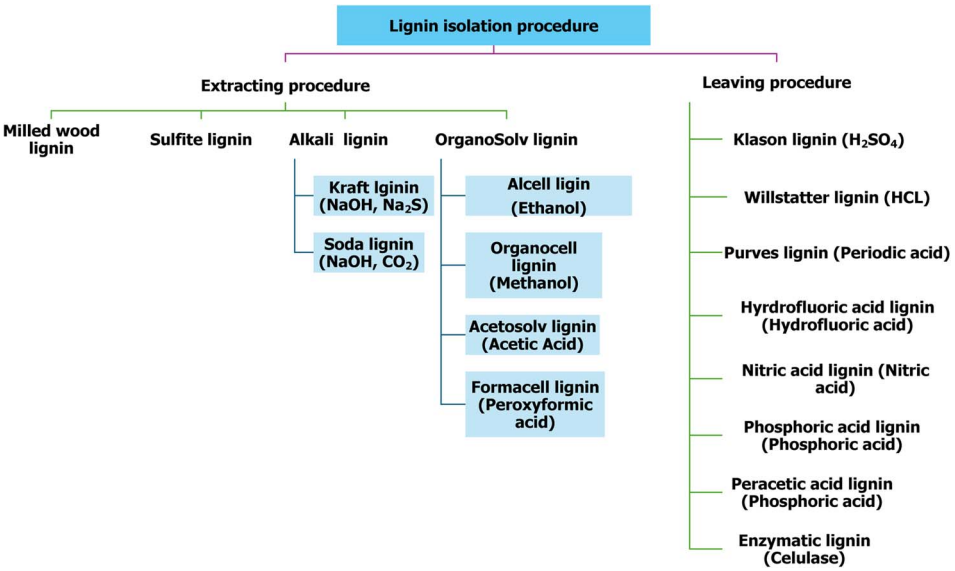

Fig. 1 Procedure of the lignin isolation.[59] Adapted and modified with permission of Francis and Taylor publishers.

other acid hydrolysis agents, such as sulfuric acid,[64] nitric acid,[65] hydrochloric acid,[66] phosphoric acid,[67] oxalic acid,[68] periodic acid,[69] and peracetic acid,[68] are also frequently employed in this process. According to Y. Sun and Cheng (2002),[70] the acid hydrolysis stage is the most crucial step in this process, as it liberates dissolved polysaccharides and yields pure lignin-containing solid residues. Concentrated or diluted acid solutions can be used for acid hydrolysis at typical pressure and temperature conditions, as indicated by Sluiter and coworkers,[71] or at high temperatures (165–195 °C), as reported by Guo and his colleagues.[72] Horst *et al.* mentioned that sulfuric acid and hydrochloric acid are commonly utilized to hydrolyze Klason and Willstätter lignins, which are examples of hydrolytic lignin.[66] In the traditional Klason process, the pretreatment step biomass (milled, dewaxed, defatted, deproteinized, *etc.*) is initially incubated in 72% H_2SO_4 at 37 °C for two hours. To remove all polysaccharides, the mixture is then reduced to 3% H_2SO_4 and refluxed for four hours at 800 °C. The Klason process is considered a standard technique for determining the amount of lignin in plant materials due to its high yield.[73] For a comprehensive understanding of the Klason process, one may refer to the Carrier *et al.*[64] paper. The Klason process is expected to cause structural changes in lignin due to its strong acidic conditions. Specifically, the formation of phenolic groups and the breakdown of certain aryl and alkyl ethers in units of benzyl alcohol are anticipated.[74] Consequently, the Klason procedure is not suitable for adequately preparing lignin for structural characterization. During the acid hydrolysis process, 42% hydrochloric acid was utilized by the Willstatter lignin. The different methods are summarized in Fig. 2.

Regarding the process of enzymatic hydrolysis, lignin's carbohydrates are extracted through the use of enzymes that break down cellulose and hemicelluloses. This results in insoluble leftovers referred to as lignin hydrolyzed enzymatically (EHL), which typically contain approximately 95% of the original lignin and are contaminated with around 10–12% of protein and carbohydrate contaminants, making subsequent analysis more challenging.[57,75] According to Yin and his coworkers,[76] the enzymatic hydrolysis in this process is often carried out under mild conditions, which preserves the majority of the active functional molecules, particularly phenolic and alcoholic hydroxyl groups, in lignin. An example of the EHL process is provided in the paper by Zhang *et al.*[40]

During the pulping stage, lignin is extracted from biomass using a high temperature and a NaOH solution (4–7%).[77,78] This results in the formation of a black liquid, which can be separated from the lignin by acidification with HCl or CO_2. If a solution of NaOH and Na_2S is used as the pulping medium, kraft lignin is generated,[79] while if a NaOH solution is used without Na_2S, soda lignin is produced.[80] These days, softwood and non-wood materials such as peel and pomace are typically processed using the soda process, while hardwood materials are typically processed using the kraft process. During the soda process, lower molecular weight lignin fragments can be produced when alkali hydrolysis partially cleaves the a-ether linkages in lignin.[55]

The use of strong oxidants, such as anthraquinone (AQ) or H_2O_2, is frequently employed in the extraction of linked carbohydrates from lignin in alkali solutions.[81] In the Kraft method, the Na_2S dosage is typically determined to be 16 g per liter of 1 M NaOH. By attaching to the a-carbon atom in the ether linkages, sulfide and bisulfide ions have the potential to break portions of the lignin's ether connections during this process. Unlike soda lignin, kraft lignin contains approximately 1% sulfur in the form of aliphatic groups.[82]

The level of acidity when precipitating alkali lignin has a significant impact on the molecular weight of the end product; the greater the extent of acidity, the lower the product's molecular weight. The sulfite pulping process, which treats biomass with a water-based solution consisting of sulfur

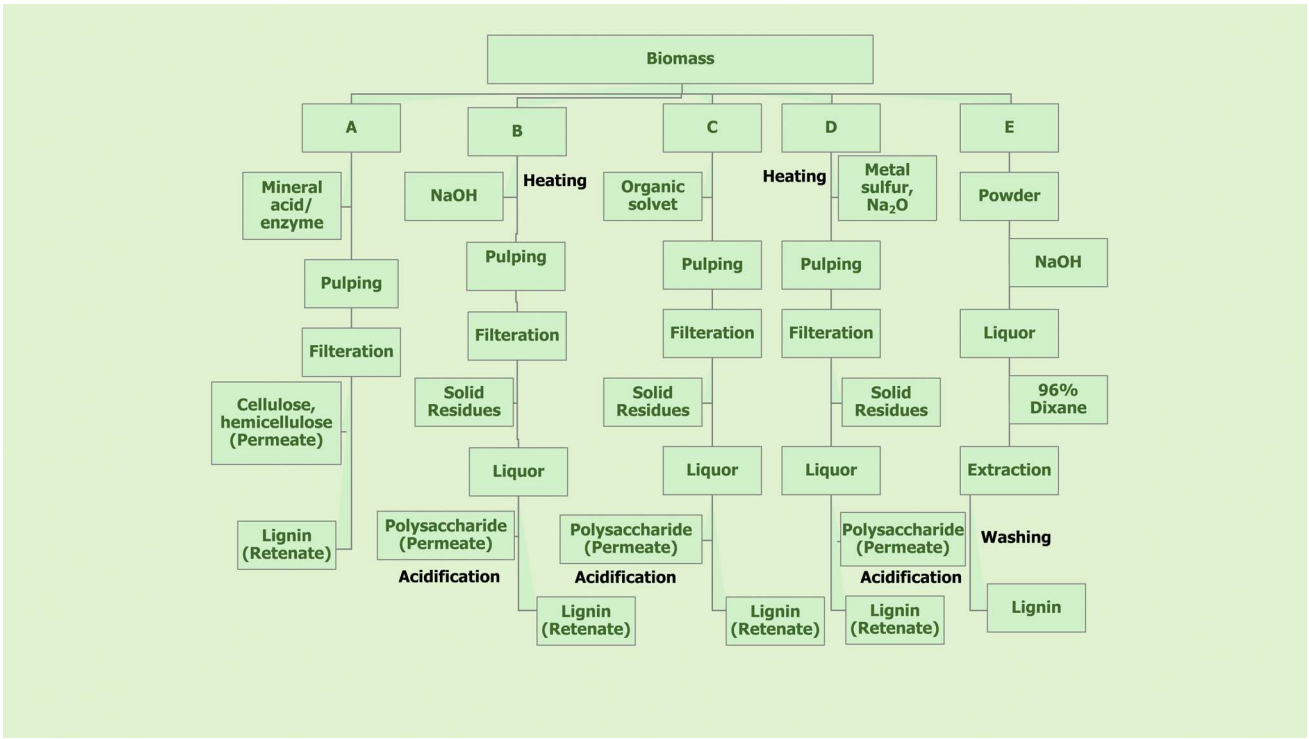

Fig. 2 Flow diagram of preparation processes of milled wood lignin.[59] Adapted and modified with permission of Francis and Taylor publishers.

dioxide and a sulfurous acid salt at temperatures ranging from 125 to 1500 °C, results in sulfonated lignin, also known as lignosulfonate.[55] Acid hydrolysis may be used in this method to cleave the bonds to polysaccharides,[80] and it may also partially destroy the a- and b-ether linkages in lignin.[83] The resulting lignin contains carboxylic groups, phenolic/aliphatic hydroxyl groups, and functional groups such as lignosulfonic acid (LA), lignosulfonate, and carboxylic group, depending on the type of sulfurous acid salt used, such as calcium sulfite,[84] sodium sulfite,[85] or magnesium sulfite.[86] Sulfite lignin is water-soluble due to its high sulfonate content (up to 13%).[82] Lignin can be extracted from pulping liquors through various methods, such as dialysis, membrane filtration, and alcohol precipitation. According to the study reported by Shimizu *et al.*,[83] these techniques may partially degrade the a- and b-ether linkages in lignin. By using different sulfurous acid salts, like calcium sulphite,[87] sodium sulphite,[85] or magnesium sulfite,[86] the resulting lignin contains functional groups such as carboxylic groups, phenolic/aliphatic hydroxyl groups, and lignosulfonic acid (LA), lignosulfonate, and carboxylic group.

Organosolv lignin is produced by heating biomass in a dilute organic solvent (40–80%, v/v) at temperatures ranging from 1400–2200 °C.[88] In this process, the a-ether connections in lignin are broken down using hydrolytic cleavage, and an acid or base catalyst is employed to dissolve the lignin fragments in the solvent.[80] A variety of organic solvents, such as acetone, ethanol, methanol, formic acid, or acetic acid, can be used along with different catalysts like HCl and NaOH-AQ combination.[89] Some of the commonly used techniques include Alkaline sulfite-AQ-methanol (ASAM),[90] Formacell,[91] Alcell,[89] Organocell,[92] and Acetosolv.[93] The Alcell process utilizes a sulfuric acid catalyst (approximately 1.0%) and a dilute ethanol solution (about 65%, v/v) to process biomass.[94] The Organocell method employs a 30% NaOH catalyst in combination with an aqueous methanol solution (40%, v/v), either with or without AQ.[55] The Acetosolv technique involves heating biomass in a 93% (v/v) aqueous acetic acid solution with less than 1% HCl as a catalyst.[55] The Formacell approach uses a water-based solution containing peroxyformic acid, which is created when formic acid and H_2O_2 are combined.[95] Finally, the ASAM procedure utilizes an aqueous NaOH solution (approximately 14%) containing methanol (15%), AQ (0.1%), ethylene diamine tetra acetic acid (0.5%), and Na_2SO_3 (Na2SO3/NaOH $\frac{1}{4}$ 80/20) at 1500 C.[55] Both annual plants and hardwoods can be processed effectively with the organosolv approach. Apart from the ASAM method, organosolv lignin is sulfur-free and has a low molecular weight of roughly 5000.[55] Additionally, the organosolv process can be enhanced by using a novel pulping approach of steam explosion before solvent extraction, which has become more and more common as an organosolv technique in recent years.[83] Furthermore, pretreating biomass with ion solutions such as 1-ethyl-3-methylimidazolium acetate has been shown to lessen the greenhouse gas emissions of volatile organic solvents, making it a more environmentally friendly option.[96]

Strategies for lignin incorporation into polymeric film

Solution casting. Stokes' law forms the basis for the solution casting method. In this process, the prepolymer and polymer

are combined evenly and rendered soluble in the appropriate solution.[97] The lignin is distributed either in the same solution or a separate one, whereas the polymer, acting as the matrix phase, dissolves easily and turns soluble in the solution. Solution casting, also known as the wet processing method, has performed higher compared to other production techniques due to its ease of use and absence of specialized equipment needed for injection-molding or extrusion procedures.[97] Saraf and Glasser used solution casting to create lignin-based polyurethane films by combining hydroxypropyl lignin derivatives with an aliphatic or aromatic isocyanate. The study's experimental materials included two lignins, kraft and steam-exploding lignin, and two diisocyanates, hexamethylene diisocyanate (HDI) and tolylene diisocyanate (TDI).[98] In this process, the lignin and solubilized polymer matrix (epoxy) are continuously mixed and swirled, and then casting, drying, or solvent evaporation is performed.[99] Compared to the hot-pressed method, the solution casting approach yields a lignin-based polymeric film with a higher degree of crystallinity. Solution casting is the most efficient method for producing thermally stable films of polymers from materials that tend to form gels prior to the reaction being complete. Typically, a suitable solvent—such as water, alcohol, or any other organic solvent—is used to dissolve the polymer. Because solution casting polymer composites are made at room temperature, they degenerate and deteriorate less quickly than molten compression molding composites.[100] The physical, aging, and mechanical properties of manufactured polymeric composites are influenced by various factors, including the viscosity of the polymer, the even distribution of the particles, their stereochemistry, conformations, type of hardener, ingredients, and ambient conditions.[101]

Blown film extrusion. Extrusion of blown films is one of the numerous processes utilized in the production of polymers. Specialty polymer films, such as stretch, shrink, and barrier films (which shield deli meat), as well as shopping bags and frozen food packaging, are produced using this method and are widely utilized in packaging.[102] Although there are more types (LDPE, HDPE, and LLDPE), the two polymers that are most frequently used for blown film extrusion are polypropylene and polyethylene. This technology, known as co-extrusion, has the potential to produce multi-layer films that integrate multiple plastics into one film, as well as more complex monolayer films. Chiappero blow-molded LLDPE pellets with sizes of particles ranging from 38 to 75 μm, that include 2.5%, 5%, and 10% lignin to produce flexible films.[103] Melt flow index experiments indicate that 85% of the polymer mix containing kraft lignin may be extrusion molded adequately. Tiny amounts of plasticizing agents are added to blend solutions to modify the cross-linking of lignin molecules. It was noted that 85% of the kraft lignin could possibly be put to use. It was discovered that the use of polymer and the appropriate plasticizers might result in a combination of a glass transition temperature (T_g) which is almost ambient. To obtain the required film thickness and width, this production method can be adjusted. The use of solvent casting and extrusion is prevalent in the production of lignin-based polymers, which have diverse applications such as packaging, drug delivery, biological medicine, wound healing, agricultural applications, seed coatings, and automotive components. The addition of lignin to chitosan films in these processes often results in surface roughening, which can further decrease the contact angle. Studies have demonstrated the benefits of using lignin as twin filler in high-density polyethylene extrusion and blow molding. Two methods were employed to produce polylactic acid nanocomposites with lignin nanoparticles at varying weights: melt extrusion and solvent-based casting. The ability to control the mechanical properties of the final plastic product based on the process settings and base polymer(s) used is a significant advantage of using conventional blown film extrusion techniques. Blown film extrusion is a process in which the polymer is caused to expand radially and move upward, leading to the application of pressures in the draw and transverse directions. As a result, the polymer stretches, giving the film its strength. The quantity of infusion and drawing can be adjusted to provide the desired strength in the cross and draw directions. Furthermore, blown film extrusion is highly versatile, enabling the production of various single or multi-layer films in a range of widths and thicknesses, as reported.[104]

Compression molding. The use of compression molding in the production of thermosetting polymers is a common practice. A standard compression molding procedure involves a curing interval of 3 minutes at a temperature of 350 °F and a pressure of 100 psi (180 °C and 700 kPa). After the material has dried, the plastic package is removed from the mold. This method of production is low-cost and suitable for high-volume production.[105] In one experiment, aluminum foils were coated equally with copolymers based on lignin, and these foils were then placed in a compression mold at a temperature of 110 °C and a pressure of approximately 117 MPa for 3 minutes.[106] It was observed that as the fiber content of the lignin composite film increased, so did its impact and flexural strength. However, for lignin-based polylactic acid films, the solvent casting approach was found to have better dispersion than the compression molding method. Beech wood flour is used to produce Bjorkman lignin, which is then used to make polyethylene films. These films, which were created by compression molding after solution casting, exhibited superior antibacterial properties.[107] By adding 2% of an antimicrobial substance to the polyethylene films through solution casting and then compressing the material, the resulting films had improved antibacterial qualities.[107] Blends of starch, PVA, and lignin with glycerol as a plasticizer were created using a compression molding machine for film manufacturing and the melt compounding process.[108] There are two types of compression molding processes: hot compression molding and cold compression molding. Cold compression molding cures at room temperature, while hot press molding cures by heating the mold, which then transfers heat to the composite components.[105]

Lignin based materials for food packaging applications. Lignin is a promising biopolymer that can be used in food packaging applications. According to recent studies, lignin-derived materials possess excellent physicochemical properties (shown in Table 1) that make them suitable for use in food packaging applications. In this section, we will discuss the

Table 1 Properties of the lignin derived materials for food packaging applications

Lignin derived materials	Barrier and antibacterial properties	Mechanical properties	Thermal durability	Ref.
Lignin—gellan gum-hydroxyethyl cellulose	Enhanced hydrophobicity, UV, antioxidant property	Tensile strength (MPa): 23.0 ± 1.1,39.0 ± 0.8 (respectively, for gellan gum, lignin—gellan gum-hydroxyethyl cellulose)	T_g (°C): 149.2 ± 0.5, 156.9 ± 0.3 (for gellan gum, lignin—gellan gum-hydroxyethyl cellulose, respectively)	111
	Exhibited better antibacterial and non-cytotoxic properties	EB (%): 20.3 ± 0.4,32.5 ± 0.4 (respectively, for gellangum, lignin—gellan gum-hydroxyethyl cellulose)	T_m (°C): 205.6 ± 0.6, 216.0. ± 0.3 (for gellan gum, lignin—gellan gum-hydroxyethyl cellulose, respectively)	
Cellulose–lignin films	Enhance antibacterial and UV-shielding, durability	Tensile strength (MPa): 75.90	First weight loss (room temperature to 160 °C) was about 10%	121
	Enhanced antibacterial property, hydrophobicity		T_{onset} of cellulose film is 270 °C, which increased to 275 °C and 290 °C	
	Good water vapor and oxygen barrier ability of the phenolated lignin/cellulose		T_{max} (maximum mass loss temperature) values are 291, 308 and 320 °C for the cellulose and cellulose-lignin derived films, respectively	
Carboxymethyl cellulose (CMC) and lignin film	Enhanced physical properties, thickness, solubility, moisture content, and water vapor permeability (WVP) were improved from 0.09 to 0.14 mm, 84.75 to 51.03%, 31.34 to 19.30%, and 4.98 to 1.08 × 10^{-10} g m^{-1} s^{-1} Pa^{-1}, respectively	Tensile strength (TS) increased from 18.29 to 32.61 MPa and elongation at break (EAB) of the CMC-lignin films from and 32.5–45.3%	—	122
Poly(butylene succinate (PBS) based lignin films	—	PBS tensile modulus (MPa) 636.8 ± 56.9 (machine direction MD), 794.7 ± 50.4 (transverse direction TD) Tensile strength (MPa) 38.3 ± 3.5 (MD), 35.4 ± 5.3 (TD) Elongation at break (%) 279.3 ± 30.9 (MD), (%) 8.8 ± 2.4 (TD) Tensile modulus 652.0 ± 37.5 (MD), 809.7 ± 69.9 (TD) Tensile strength (MPa) 37.3 ± 1.8 (MD), 33.5 ± 2.5 (TD) Elongation at break (%)282.8 ± 15.8 (MD), 6.7 ± 0.5 (TD)	(PBS melting temperature (T_m) 112.0 °C, crystallization temperature (T_c) 91.0 °C PBS based lignin films T_m 112.3 °C, T_c 90.7 °C	123
Polyvinyl alcohol (PVA)/chitin-lignin nanoparticles (PVA/CI/LNP) films	Enhanced barrier properties	Tensile strength 23.40 ± 1.36	PVA (degradation temperature) (DT) 75.99 °C, (weight loss) (WL) 95.30% at first peak	118
	PVA moisture (%) 16.33 ± 1.70	Elongation (%) 121.6 ± 0.12	PVA/CI/LNP (DT) 96.34 °C, (WL) 97.00%	
	PVA/CI/LNP moisture (%) 5.79 ± 1.81	PVA/Ci/LNP tensile strength 36.82 ± 0.17, elongation (%) 121.2 ± 0.13		
Alkali lignin lignosulfonate—soy protein isolate	Enhanced thermal, mechanical, and UV barrier qualities decreased penetration of the vapor of water	TS (MPa): 4.74 ± 0.34, 8.01 ± 0.89,10.98 ± 1.02 (respectively, for soy protein, 10% lignosulfonate–soy protein, 10% alkali lignin—soy protein) EB (%): 126.33 ± 17.9, 79.95 ± 5.32, 7.45 ± 1.24 (respectively, for soy protein, 10%	First weight loss 50–100 °C. The second weight loss occurred at around 300 °C	112

Table 1 (*Contd.*)

Lignin derived materials	Barrier and antibacterial properties	Mechanical properties	Thermal durability	Ref.
		lignosulfonate—soy protein, 10% alkali lignin—soy protein)		
Lignin—nanocellulose	Improved UV protection and oxygen permeability	Tensile strength (MPa) 22.8 Elastic modulus (GPa) 4.7 Breaking strain (%) 0.7	—	124
Lignin—poly (lactic acid) (PLA)	Strong antioxidative action	TS(MPa): ∼40, ∼30 (respectively, for PLA, PLA—40% lignin) EB(%): ∼15,∼2 (respectively, for PLA, PLA—40% lignin)	T_{onset} (°C): 323.6, 306.1 for PLA, PLA—40% lignin, respectively) T_{max} (°C): 330.2, 320.7(for PLA, PLA—40% lignin, respectively)	125

latest research on lignin based materials for food packaging applications, focusing on their antibacterial, antioxidant, UV, antiviral, and other properties. Plant cell walls are composed of lignin, a complex phenolic substance that serves as a natural UV filter and possesses high antioxidant properties. Despite these advantages, lignin has poor mechanical and separating characteristics. When combined with agar, however, it can improve the film's mechanical and thermal durability, as well as its water vapor barrier and transparency.[109,110] Although the integration of lignin with other biopolymers has the potential to offer numerous advantages, the research conducted in this field is currently limited. For instance, a study by Rukmani Krishnan *et al.*[111] used the solvent casting method to create composite films by combining lignin with gellan gum and hydroxyethyl cellulose at different weight percentages (0, 1, 5, and 10%). The addition of 10% lignin resulted in a 59.2% increase in tensile strength and 100% and 90% protection against UVB and UVA rays, respectively. Another study by Zadeh, O'Keefe, and Kim[112] found that an alkalilignin–lignosulfonate–soy protein isolate film had similar UV barrier properties to lignin, and also exhibited improved thermal and mechanical stability.

A recent study has shown that wood-inspired biopolymeric nanocomposite films offer a promising solution to environmental concerns in food packaging. These films are made from sustainable materials, including cellulose nanofibers, lignosulfonates, and beechwood xylans, and possess exceptional qualities such as UV protection, mechanical strength, and fruit preservation (see Fig. 3). This study emphasizes the potential for cost-effective and eco-friendly packaging solutions that prioritize sustainability.[113]

A study reported that lignin, an abundant biopolymer presents in agricultural biomass and characterized by its sustainability and biodegradability, can be utilized as a viable alternative for the production of biomaterials, particularly polymer packaging. After extracting and analyzing it from wheat straw, the incorporation of lignin into nanocomposite films showcases its potential to enhance antioxidant, antibacterial, and UV protective properties. The resulting films exhibit strong antimicrobial activity against a variety of pathogens and effective UV protection, highlighting their potential as eco-friendly, cost-effective, and sustainable biomaterials. Moreover, the inherent qualities of lignin-based films—such as low cost, biocompatibility, flexibility, and transparency—further support their applicability in the food packaging industry.[114]

Recently, researchers have developed lignin nanoparticles (LNPs) using the deep eutectic solvent (DES) anti-solvent approach and combined them with a polyvinyl alcohol (PVA) matrix to create nanocomposite films. These films exhibit improved mechanical, thermal, and hydrophobic qualities, as well as good UV protection, potent antioxidants, and strong antibacterial activity against *E. coli* and *S. aureus* as exhibited in Fig. 4. The use of LNPs in PVA-based nanocomposite films holds great promise for their application in active food packaging.[115]

Garg and coworkers studied that incorporating lignin's exceptional properties, such as its tensile strength, antioxidant, antibacterial, and UV barrier properties, along with chitosan's film-forming capabilities, could lead to the development of improved food packaging materials. The refined hydrogel formulations show enhanced bioactivity and reduced production costs, suggesting the potential for creating environmentally friendly food packaging solutions. Furthermore, the optimized synthesis method enhances the efficiency of hydrogel film production.[116] According to another study, incorporating lignin into biodegradable polylactic acid (PLA) composite films as a reinforcing filler proves effective. Researchers created lignin-grafted polylactic acid co-polymers and mixed them with PLA through *in situ* polymerization, resulting in films with improved mechanical properties and strong antioxidant capabilities. These findings suggest a promising path for the creation of high-performing, eco-friendly packaging materials.[117]

An intriguing study investigates the use of oxytenanthera abyssinica derived lignin nanoparticles (LNPs) in enhancing the properties of polyvinyl alcohol/chitosan (PVA/CI) and polyvinyl alcohol/chitin (PVA/CH) films for active food packaging. The addition of LNPs at concentrations of 1% and 3% improved the films' mechanical characteristics, antioxidant capacity, and thermal stability. Furthermore, the inclusion of LNPs enhanced the UV-blocking and antimicrobial properties, and prevented the migration of dietary stimulants, making these films promising candidates for use in active food packaging.[118]

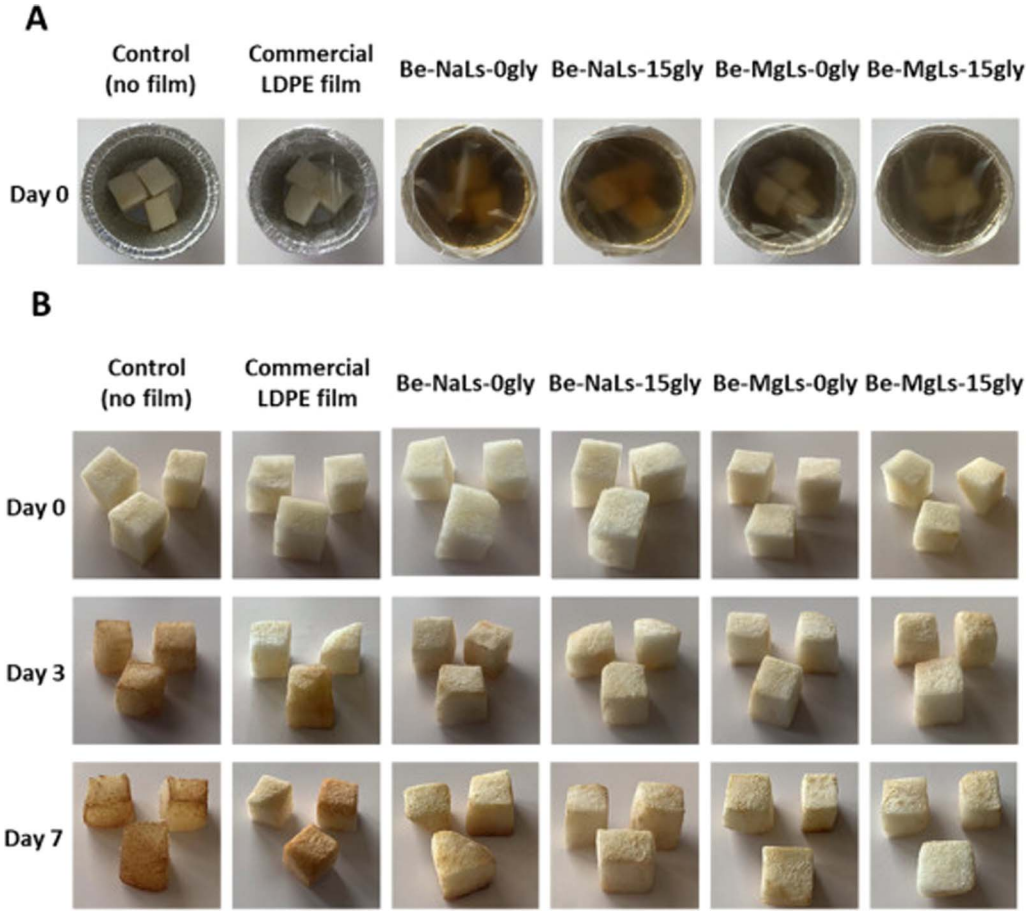

Fig. 3 (A) Digital photos of the packed fruits (B) freshly cut pears before and after storage in the refrigerator at 4 °C (3 and 7 days). Reproduced with the permission of Elsevier.[113]

Researchers have recently uncovered the promising potential of carbon dots (CDs), made from sustainable lignin, for use in intelligent sensing applications in food packaging. By incorporating CDs into a carrageenan biopolymer matrix, a composite film is produced that exhibits increased mechanical characteristics, pH-dependent color change, UV radiation blocking, and enhanced hydrophobicity. Additionally, the film demonstrates good antioxidant and antibacterial qualities, as well as notable decreases in CO_2 gas permeability and oxygen transmission rate as presented in Fig. 5. The practical use of this responsive packaging material in tracking milk deterioration through color changes showcases the potential for sustainable and intelligent food packaging solutions.[119]

A recent study has produced eco-friendly food packaging films by combining lignin with a matrix of potato starch and polyvinyl alcohol (PVA). The addition of lignin significantly improves the tensile strength, UV barrier, antioxidant activity, water vapor barrier, and antibacterial properties of the film. The development of a polymer network structure by hydrogen bonding, as confirmed by structural analysis, increases the interfacial compatibility between polymers. Lignin's high phenolic hydroxyl content facilitates its antioxidant action through a proton-coupled electron transfer pathway. This study presents a promising method for developing multifunctional composite films using elements derived from biomass, which have significant potential for use in food packaging.[120]

Antioxidant properties of lignin based materials. Numerous studies have validated the antioxidant capacity of lignin. Ten years ago, *in vitro* experiments demonstrated that bagasse lignin possessed antioxidant properties.[126] Latif and coworkers research revealed that lignin from oil palm fronds exhibited a stronger Fe^{+3} reducing power than vanillin.[127] According to a study by Li *et al.*, butylated hydroxyl toluene (BHT) had a radical scavenging index of 0.29.[128] On the other hand, bamboo stem lignin treated with organosolv (formic acid/acetic acid/water, 3/5/2, v/v/v) showed a strong ability to scavenge DPPH radicals. Dong and his colleagues reported that the oxygen radical absorbance capacity (ORAC) of the lignins from maize stover obtained by specific ethanol extraction tasks (1741.72–3119.68 lmol TE g^{-1}) was much higher than that of vitamin C (530.64 lmol TE g^{-1}).[129] In another study[130] acetosolv lignin made from bamboo shoot shell showed slightly higher values than BHT in terms of ferric reducing power, DPPH, and ABTS radical scavenging activities. In terms of the half-maximal inhibitory concentration (IC_{50}) of DPPH radical scavenging, the lignins in rice husk by acidolysis (37.2 lg ml^{-1}) and alkaline

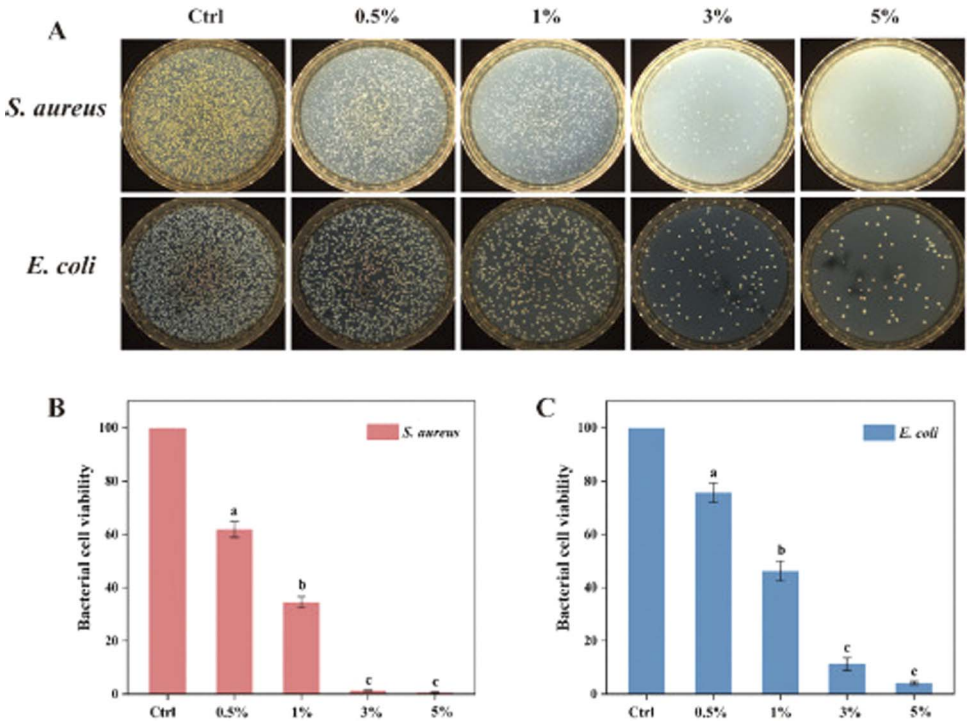

Fig. 4 Plate count images of the bactericidal activity of nanocomposite films (A) corresponding to bacterial cell viability of *S. aureus* (B) and *E. coli* (C). Reproduced with the permission of Elsevier.[115]

enzymatic treatment (52.6 lg ml^{-1}) showed higher values than the reference antioxidants of quercetin (1.8 lg ml^{-1}), rutin (4.3 lg ml^{-1}), BHA (4.5 lg ml^{-1}), and BHT (10.4 lg ml^{-1}).[131] Furthermore, our recent studies[130,132] demonstrated that acetosolv lignins from the shells of sunflower seeds and bamboo shoots have greater antioxidant activity than BHT. A substantial body of research[133,134] has confirmed the *in vitro* antioxidant activity of various lignins. For instance, bagasse lignin, lignosulfonate, curan 100, and steam explosion lignin[135] were able to dose-dependently inhibit red blood cell hemolysis caused by 2,2′-azobis (2-amidinopropane) dihydrochloride (a peroxyl radical catalyst) in preserved human keratinocyte cell line HaCaT and mouse fibroblast cell line 3T3. Additionally, similar results were obtained with human red blood cells.[126] Among all the lignins examined, bagasse lignin showed the highest antioxidant capacity. In human colon cancer CaCo-2 cells, preincubation with kraft lignin from spruce and beech wood lignin from fermentation with yeast at a concentration of 50 lg ml^{-1} may significantly reduce H_2O_2-induced DNA strand breaks.[136] It is demonstrated the transepithelial mechanism of transport of calcium lignosulphonate within CaCo-2 cell layers, even at low levels.[137] A study have shown have shown that the lignin-polycaprolactone (PCL) copolymer has more antioxidant activity than PCL.[138] The lignin-PCL nanofiber exhibited a more robust protective effect against H_2O_2-induced oxidative stress in Schwann nerve cells that had been seeded, in comparison to its PCL counterpart. Furthermore, proliferating on lignin-PCL nanofiber, Schwann cells as well as dorsal root ganglion neurons may facilitate nerve regeneration. Lastly, when beech wood lignin extracted with dioxane/water was fed to rat primary hepatocytes for 21 days at a dosage of 0.8 g/100 g, the hepatocytes demonstrated remarkable resistance to H_2O_2-induced oxidative stress *in vivo* testing. DNA strand breaks were observed to decrease as a result of this.[136]

Catignani and Carter[139] discovered that adding lignin (l–10%) to the rat diet increased the amount of retinol deposited in the liver by 50–100%, despite the lack of data available at the time. Unfortunately, until recently, no clinical data was available on lignin's antioxidant properties. However, it should be noted that the mechanism underlying lignin's *in vivo* antioxidant action remains unknown due to the material's poor absorbability from mice's gastrointestinal tracts.[140] Despite this, the prebiotic benefits of kraft lignins and Alcell for farm animals suggest a feasible *in vivo* approach through gut flora regulation.[141] Within the food chain, it was found that 2.5% of kraft lignin derived from wood sources was just as effective at preventing maize oil oxidation caused by heating it to 1000 C and aerating it at a rate of 180 mm per minute as 0.03% vitamin E.[139] Furthermore, milled wood lignin from green tea leaves may block the autoxidation of linoleic acid by 50%, although it is less efficient than the commercial antioxidants of VE and BHA. Remarkably, lignin has been shown to enhance the antioxidant capacities of a-tocopherol and gallate (EPG).[142] Quercetin and lignin have also been found to have a combined antioxidant impact.[143] As demonstrated, the specific technique employed and the genetic origins of the starting material have a significant impact on the antioxidant capacity of separated lignin. For instance, Dizhbite and coworkers discovered that alkali lignins

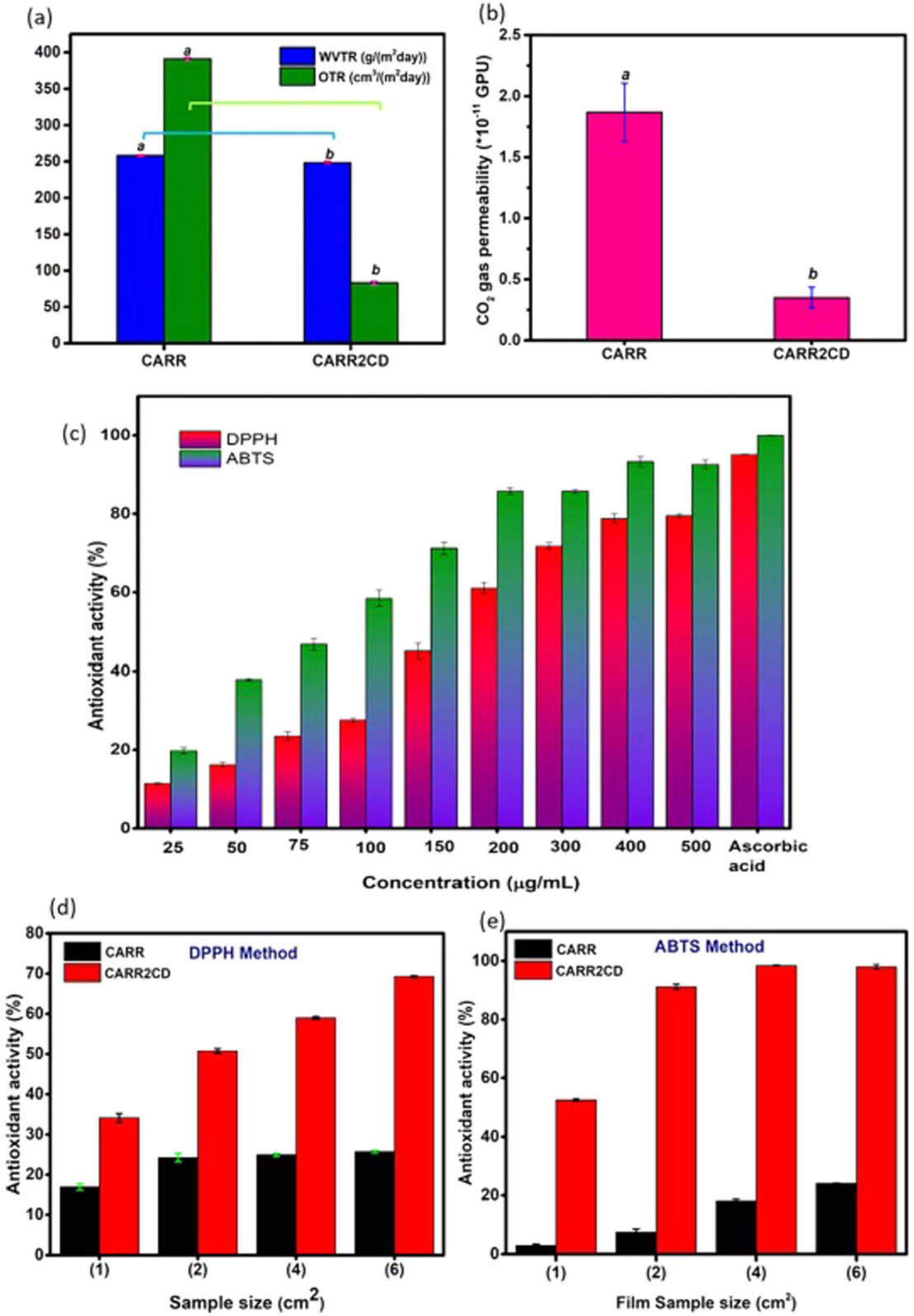

Fig. 5 (a) Water and oxygen barrier properties of Carrageenan and Carrageenan-CD films (b) CO_2 barrier property of Carrageenan and Carrageenan-CD films (c) antioxidant activities of CD solution against ABTS and DPPH radicals (d) antioxidant activity of films against DPPH free radicals (e) antioxidant activity of films against ABTS free radicals. Reproduced with the permission of Elsevier.[119]

from spruce, birch, and aspen—all produced using the same technique—display antiradical capabilities that are, 0.5, 1.0, and 1.1,[144] respectively.

The results of the study conducted by Dong *et al.* indicated that the soda lignins derived from maize stover and prepared under different conditions displayed varying degrees of antioxidant activity. Specifically, the sample extracted at 950 °C for 120 minutes with a solid/solvent ratio of 1 : 4 (w/v) exhibited the highest level of antioxidant activity among the four samples tested.[129] This finding is consistent with previous research demonstrating that lignins derived from the same biomass can have distinct antioxidant properties.[145] It is widely accepted that the structural features of lignin play a critical role in determining its antioxidant activity. For instance, the presence of unbound phenolic hydroxyl, methoxy, aliphatic hydroxyl, and double bonds between the side chain's outermost carbon atoms have been reported to contribute to lignin's antioxidant activity.[144,146] The mechanism by which lignin actsas an antioxidant is depicted in Fig. 6, which includes initiation, propagation, termination, and several advanced oxidized products.

The concentration of phenolic hydroxyl in lignin is a key structural feature that determines its antioxidant capacity. It is worth noting that studies have shown that low molecular weight lignins tend to have greater antioxidant activity than those with higher molecular weights.[147,148] This can be attributed to the fact that lignin fragmentation exposes more phenolic hydroxyls, thereby increasing its antioxidant activity. However, the condensation reaction can harm lignin's antioxidant activity.

Antibacterial properties of lignin based materials. Lignin has been discovered to possess innate antimicrobial qualities (Table 2), which protect plant tissue from microbial harm,.[150] Although the antimicrobial activity of lignin is species-specific, research has shown that it has the potential to serve as a natural antibacterial agent and the mechanism is shown in Fig. 7. It is demonstrated that the antibacterial properties of lignin by successfully preventing the growth of *Staphylococcus epidermidis* in sugarcane bagasse lignin films.[151] Lignin has been reported to be effective against certain Gram-positive bacteria, such as *L. monocytogenes*, *L. innocua*, *S. aureus*, *B. subtilis*, and *B. mycoids*, but not against Gram-negative bacteria like *S. Enteritidis* and *E. coli*.[129,152] The reason for this difference in susceptibility is not entirely clear, but it may be related to the absence of a second cell wall in Gram-positive bacteria.[153] However, this theory does not always hold true for Gram-negative bacteria. Lignin has been found to inhibit the growth of certain Gram-negative bacteria, such as *S. typhimurium* and *E. coli*, as demonstrated by a Medina study.[153] In addition, dietary Alcell lignin may reduce the quantity of *E. coli* in chicken waste and cecum content, as reported by Baurhoo and his colleagues.[154] This finding has important implications for reducing intestinal *E. coli* contamination during poultry slaughter. The precise mechanism by which lignin inhibits bacteria is not yet fully understood. It has been proposed that lignin's polyphenolic compounds damage bacterial cell membranes, causing the cells to lyse and releasing their contents.[141] The effects of lignin on fungi are complex and difficult to interpret universally.

With regard to yeast, spruce organ cells contain lignin that inhibits strains of *R. rubra*, *A. pullulans*, *B. alba*, and *C. tropicalis*, whereas the *C. albicans* strain is unaffected. However, the oil palm empty fruit bunch alkali lignin is ineffective against

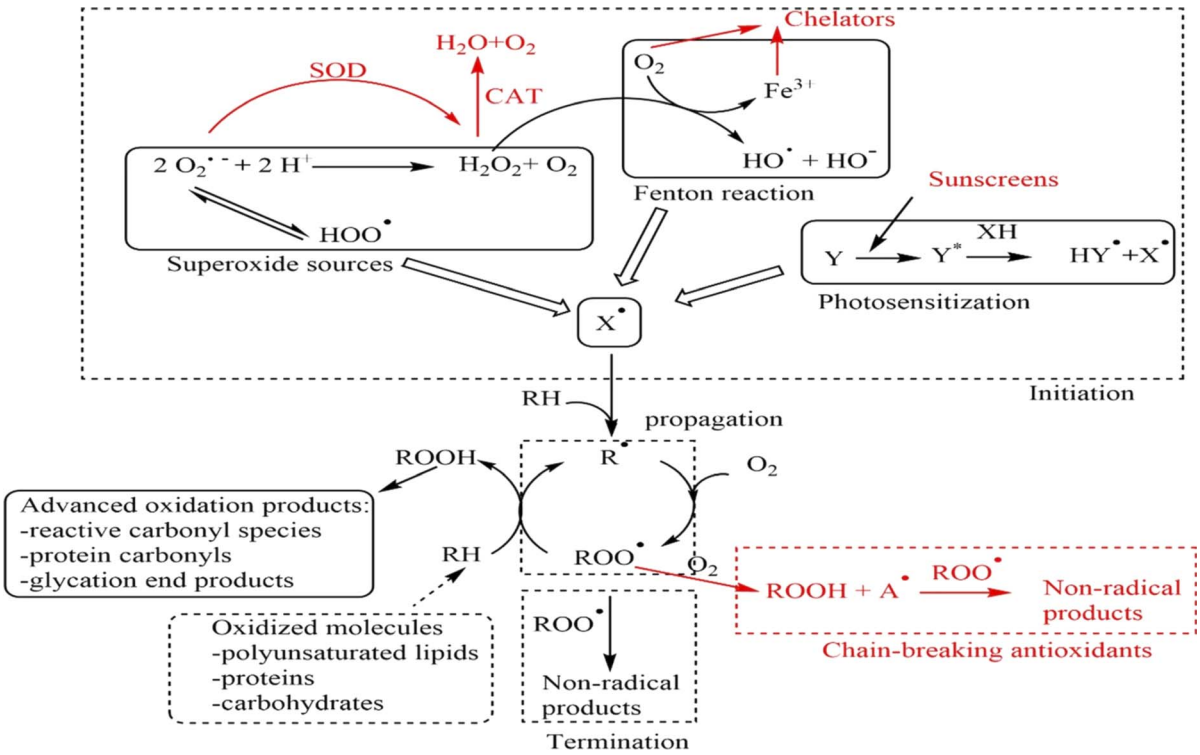

Fig. 6 Various reactions showing lignin antioxidant mechanism. Reproduced with the permission of Nature Springer Publisher.[149]

Table 2 Antibacterial properties of the lignin based materials

Source of lignin	Filler	Matrix	Microorganisms	Ref.
Alkaline lignin	Lignin nanoparticles (LNPs)	Pectin	*S. aureus* and *E.coli*	156
Corn straw	Silver-lignin nanotube	Corn straw	*S. aureus* and *E.coli*	157
Alkaline lignin	Citric acid and acetylated modified LNPs	Polylactic acid	*E. coli* and *M. luteus*	158
Commercial soda lignin (PB1000)	LNPs	Cellulose nanocrystals and cellulose nanofibrils	*S. aureus*	159
Alkali lignin (Sigma-Aldrich)	Nanolignin and 0.5 wt% metal oxide nanoparticles (Ag_2O, TiO_2, WO_3, Fe_2O_3 and $ZnFe_2O_4$)	Polylactic acid (PLA)	*S. aureus* and *E. coli*	160
Acacia mangium pulp mill waste	Starch/polylactic acid	Lignin biofilm	*E. coli*, *Salmonella typhi* and *S. aureus*	161
Lignin (Shandong Longlive co., Ltd.)	Lignin-zinc oxide hybrid particles	(Butyleneadipate-*co*-terephthalate) (PBAT)	Contact type antibacterial property improved bacterial adhesion decreased	162
Bamboo/alkaline	Polyethyleneimine-lignin contained cellulose nanofibers	Poly(vinyl alcohol) (PVA) nanocomposite films	*S. aureus* and *E. coli*	163
Lignosulfonate	Tannic acid @ sodium lignosulfonate–Ag nanoparticles	PVA	*S. aureus* and *E. coli*	164

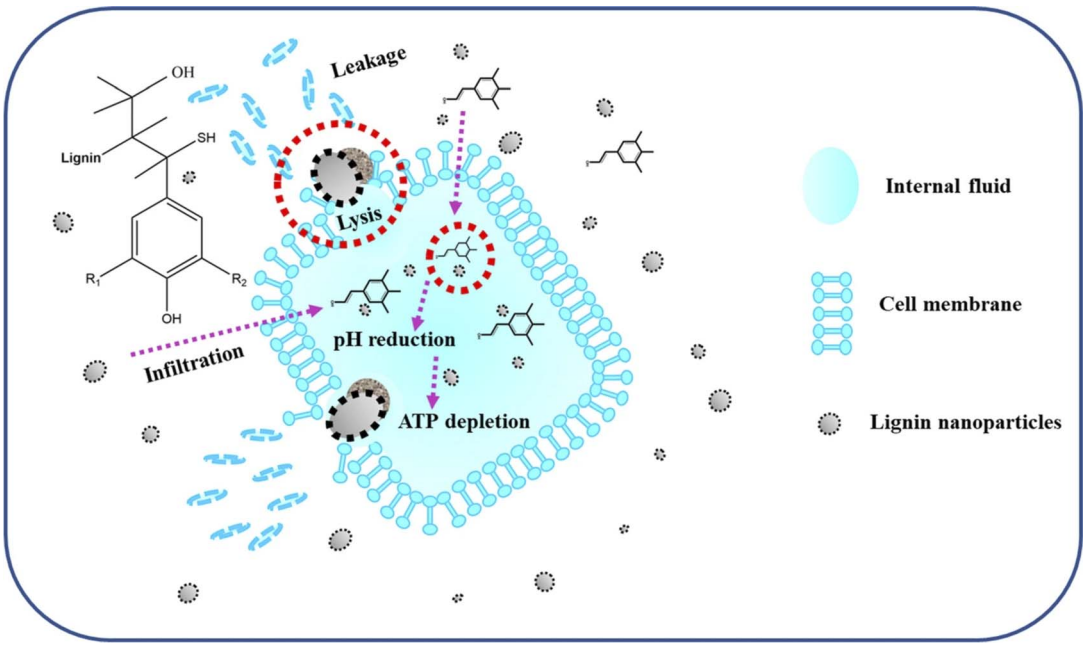

Fig. 7 Mechanism of antimicrobial activity of lignin nanoparticles for food packaging applications. Reproduced with the permission of Nature Springer Publisher.[149]

Candida albicans, but shows inhibitory effects against *Candida lipolytica*. Additionally, alkali lignin from maize stover is effective against some strains of fungi. Apple tree pruning leftovers, on the other hand, are observed to hinder the growth of *S. cerevisiae*. The chemical structure, manufacturing technique, and genetic heritage of lignin all play a role in its antibacterial activity, as do the targeted strain of the microbe and the working concentration. It is clear that lignin's antibacterial activity is not fully established, but its dependence on these factors is evident. In light of the chemical structure of lignin, two primary factors are the side chain structure and its functional moieties. Specifically, the presence of oxygen-containing groups (–OH, –CO, and –COOH) on the side chain is consistently associated with poor inhibitory power. However, isoeugenol, a phenolic component of lignin, displays an excellent inhibitory action due to its double bonds position of the side chain and the methyl group.[141] Furthermore, the aromatic compounds linked to the antibacterial activity of lignin are

similar to antibiotics like methicillin, carbenicillin, and benzyl penicillin.[155]

Antiviral properties of lignin based materials. Several viruses, such as the influenza virus,[165] herpes simplex virus,[166] and human immunodeficiency virus (HIV),[167] are susceptible to the potential antiviral properties of lignins. In a study by Sakagami, it was found that Alcell lignin produced from cacao seed husk displayed anti-HIV and anti-influenza viral activity, with selectivity values of >100 000 and 155, respectively.[168] Furthermore, Matsuhisa and coworkers[169] observed a ten-fold increase in the vitality of hepatitis C virus-infected Huh7.5.1 cells when using the low-molecular-weight lignin from Lentinula edodes mycelia solid culture, compared to its hot-water extract, suggesting the lignin's antiviral activity.

Research has confirmed that lignins possess potential antiviral properties against various viruses, including HIV.[167] According to study reported by Srisapoome *et al.*, intramuscular injection of kraft lignin (from *Penaeus monodon* Linn., black tiger shrimp) and yellow head virus solutions at lignin concentrations of 1–20 mg L^{-1} before incubation significantly reduced shrimp mortality rates at 14–20 days after injection.[170] These findings provide compelling evidence of lignin's antiviral properties. The two main contributing factors for lignin's antiviral activity were found to be lignosulfonic acid (LA) and LCC. As early as 1990, it was discovered that LCC exhibited antiviral properties. Lai *et al.* provided evidence that LCC from pine cones may prevent HIV replication.[171] The LCC from seeds of Pimpinella anisum significantly decreased human cytomegalovirus, measles virus, and herpes simplex virus types 1 and 2 with selectivity index values up to 140, 210, and 3100, respectively.[172] In a study, it is reported that the high selectivity index of up to 77 indicates that LCC generated from cocoa mass demonstrates excellent anti-HIV activity.[173] LA, a prominent member of the lignin-derived macromolecule family, has a well-established antiviral activity. Qiu *et al.*[174] verified LA's bactericidal potential against HIV-1, with EC50 values *versus* the R5 and X4 HIV-1 strains being 6.323 mg mL^{-1} and 1.411 mg mL^{-1}, respectively. Gordts *et al.*[166] reported similar results, demonstrating that LA may significantly inhibit the multiplication of the four HIV strains in MT-4 and peripheral blood mononuclear cells. Furthermore, a clinical trial conducted by Lopez *et al.*[175] found that oral lignin-ascorbic acid supplementation decreased the severity of symptoms and the likelihood of recurrent HSV-1 infection. Vinardell and Mitjans[176] analyzed the extensive data about the antiviral in nature functions of LA and LCC. The literature currently available suggests that the possible antiviral efficacy of lignin is dependent on several factors, such as the lignin's source,[173] the virus strain,[169] and the parameters of treatment (lignin concentration, length of treatment, and stage of virus infection, among others).[170,172] The relationship between lignin concentration and dose dependency is commonly established in research studies.[169,170] Although the exact mechanism underlying lignin's antiviral effect remains unclear, various research proposals have been put forth. Virus infection occurs when viral apolipoprotein E (apoE) interacts with heparin sulfate on the host cell surface.[56] Therefore, lignin's ability to bind to viruses may reduce their adsorption and penetration of host cells. A study suggested that lignin's structural resemblance to heparin sulfate enables it to compete with viruses for cell surface binding, thereby inhibiting their ability to adhere to and infect cells.[177] Secondly, lignin has been demonstrated to inhibit the activities of several viral enzymes, including RNA polymerase, reverse transcriptase, protease, and plaque formation,[171,178,179] thereby inhibiting viral replication. Lastly, lignin's antioxidant properties may strengthen host cells' defense against viral infection by reducing oxidative stress.[175]

UV light blocker properties of lignin based materials. The naturally occurring biopolymer lignin possesses several attractive characteristics, including its three-dimensional, amorphous, renewable, and biodegradable nature. Lignin is derived from the polymer matrix incorporated in active packaging, which is a byproduct of the pulp and paper as well as biorefining industries. As the most abundant natural resource after cellulose, lignin is widely recognized as the greatest naturally occurring phenolic biopolymer. It is composed of three distinct phenol types that are linked *via* ether or single bond linkages, lignin boasts an array of functional groups present in varying concentrations.[180]

Lignin's functional groups, such as phenolic units, ketone molecules, and chromophores, enable it to absorb UV light, making it a potent UV blocker. In addition to its antioxidant and radical-scavenging properties, lignin's functional groups contribute to its exceptional UV blocking capabilities.[181] Lignin can absorb a broad spectrum of UV radiation with wavelengths ranging from 250 to 400 nm and contains chromophore functional groups.[182] Despite its abundance as a byproduct of agriculture, lignin has traditionally had limited utility. However, active packaging, which serves not only as an inert barrier but also as an additional means of preserving food, represents a promising application for this versatile biopolymer.[183] Lignin presents a wide range of frameworks, purities, and related properties, which depend on its sort, source, and extraction method. Some of the distinctive qualities of lignin are its capacity to absorb UV light, low polarity (*i.e.*, hydrophobic qualities), and antioxidant properties.[184] These characteristics make lignin an appealing component in the production of film materials that have UV blocking and reduced hydrophilicity.[185] Lignin has also been studied as a potential additive to polymeric materials to protect them from UV radiation-induced photodegradation, which results in yellowing and a loss of mechanical characteristics.[186]

Lignin's radiation-protective properties have attracted significant attention, particularly its ability to act as a natural UV blocker. Lignin is the second most abundant renewable biomass on Earth and is rich in aromatic rings, making it an attractive candidate for shielding polymeric materials from UV light.[187] Lignin's composition of phenolic units, chromophores, and ketones allows it to naturally block almost the entire UV light spectrum. Additionally, lignin possesses antifungal, antibacterial, and antioxidant qualities.[188] The potential use of lignin as a replacement for artificial absorbers in a composite material is due to its strong UV-shielding ability. The sun produces electromagnetic radiation in the form of UV radiation, which can be divided into three wavelength bands: UVA (315–

400 nm), UVB (280–315 nm), and UVC (100–280 nm).[189] The commercialization of lignin-based UV shield solutions is hampered by their unappealing black appearance, despite their remarkable capacity to absorb UV light.[99] Lignin's complex structure, polydispersity in molecular weight, brownish tint, and numerous impurities make it difficult to utilize as a UV blocker. Further research is needed to render lignin a suitable bio-based UV blocker.[190]

Other properties of lignin based materials. In addition to its well-known properties, recent studies have demonstrated that lignin possesses several other attributes, such as anti-hypercholesterolemia,[191] anticancer,[192] immunomodulating,[193] anticoagulants,[194] and anti-emphysema.[195] With regard to lignin's anticancer properties, an initial study showed that dietary lignin altered fecal dry weight and bile acid excretion, two potential risk factors for colon cancer, while gut transit time, colon pH, and fecal bile acid concentration decreased. However, it failed to reduce colon tumor frequency caused by 1,2-dimethylhydrazine.[196] *In vitro* results showed that dehydrogenate polymer, an enzymatically generated lignin model molecule, inhibited the growth of normal fetal lung fibroblasts and breast cancer cell lines.[197] Furthermore, the findings of Saratale and coworkers[198] confirmed that lignin-silver nanoparticles derived from wheat straw exhibited dose-dependent lethal effects on ovarian carcinoma cell lines ($LD_{50} = 150\ \mu g\ mL^{-1}$). Evidence for lignin's ability to lower blood cholesterol dates back to 1968, when research on its ability to bind bile acids *in vitro* was conducted.[199] Recent studies have demonstrated the excellent ability of bamboo shoot shell lignin to bind a range of bile salts, including sodium deoxycholate, sodium chenodeoxycholate, sodium taurocholate, and sodium cholate.[150] *In vivo* findings by Thiffault *et al.*[200] revealed that lignin was effective in reducing elevated blood cholesterol levels; however, further research showed that lignin was not successful in sequestering conjugated bile acids.[191]

The use of biodegradable materials in food packaging has gained significant attention due to environmental concerns and the depletion of petroleum resources. Cellulose, protein, and starch are commonly used in natural biopolymer packaging sheets, which are more environmentally friendly compared to conventional plastic films. However, these materials suffer from limitations such as poor mechanical and water resistance, which hinder their industrial application. To overcome these limitations, lignin has been incorporated as filler in biodegradable films to enhance their properties. Lignin-incorporated coatings also exhibit antibacterial, antioxidant, and anti-ultraviolet characteristics, making them suitable materials for food packaging. Several studies have investigated the effects of lignin addition on the physical, optical and bioactive properties of various biodegradable materials.

Recent trends of lignin transformation into nanolignin. The most appealing characteristics of lignocellulosic wastes are their accessibility, biological activity, and ecological friendliness. By leveraging the potential of nanotechnology, nanolignin can enhance future treatments, diagnostics, and food sectors that are based on nano-technologies. The growing interest in environmental and nanotechnology research can be attributed to the continuous advancement of biomass containing lignin that is recyclable. The past decade has witnessed a considerable number of scientific breakthroughs that are relevant to the increased utilization of lignin, particularly in the context of nanolignin-based products.[201] The following section discusses the processes that can be employed to transform native lignin into nanolignin.

Milling is a widely adopted method for the production of nanolignins from plant sources, and it is a cost-effective approach to generat lignins with nanomolecular-sized particles, as per Sharma and Kumar study.[202] For instance, 5 g L^{-1} of kraft lignin can be homogenized for 4 hours at 15 K rpm to produce 500 nm of kraft lignin-derived nanolignin. Moreover, an acoustic method based on sonication is used to manufacture stable 10–20 nm nanolignin with a uniform distribution of 0.7% lignin solution generated from wheat straw and Sarkanda grass. The optimal parameters for producing such nanoscale materials without the generation of free radicals are 600 W of power inputs for one hour in a uniform stable nanodispersion with a size of 10–20 nm and 20 kHz sound waves.[203]

The extraction of lignin from biomass sources, such as softwood kraft lignin, has been a challenge due to its complex structure and poor solubility in common solvents. However, by utilizing a well-managed solvent shift approach, mid-sized nanolignins can be produced, despite their relatively low yield. The solvents selected for this method include acetone/water, tetrahydrofuran, dimethyl sulfoxide, and acetone.[202] The solvent-shifting method has been shown to yield lignin nanoparticles with a size of 200–500 nm, using kraft lignin and tetrahydrofuran and water, while the particle size of tiny nanolignin was found to be 221 ± 10 nm.[192]

It has been found that the precipitation of lignin nanoparticles can be influenced by changes in pH, resulting in stable nanostructures with tightly packed lignin moieties. This occurs through two separate processes. In the first process, an ethylene glycol solution containing Indulin AT is added, followed by aqueous HCl, which leads to the formation of biodegradable nanoparticles that are pH stable. In the second procedure, lignin and aqueous sodium hydroxide are mixed, and when coupled with HNO_3, they yield the necessary precipitates. This method produces stable nanolignins below pH 5 that are safe for the environment. Furthermore, model microorganisms including *Saccharomyces cerevisiae* and Chlamydomonas reinhardtii were found to thrive unaffected by lignin-sourced nanostructures, ensuring the viability of cells.[202]

Caicedo *et al.*[204] synthesized nanomaterials derived from lignin by template synthesis. This process involves a Schiff's base reaction between the aldehyde molecules of thioglycolate lignin and its amino counterparts in the alumina-activated template (APTES). Once hydroxycinnamaldehydes, hydroxycinnamates, or hydroxyzine. Antimicrobial silver nanoparticles with a uniform dispersion of 45–55 nm pseudo spherical, facilitated by reductant-assisted template synthesis of lignin molecules, are produced. Although template synthesis offers numerous benefits, it also causes the toxicity factor associated with surfactants like SDS and CTAB. In addition, the removability of source materials and problems with purification are

limiting variables that compromise the overall performance of this technology.

To produce lignin nanofiber scaffolds, a solution containing frozen monomer units is first used. This process results in the formation of ice crystals, which grow and separate the polymer phase. Alkali lignin serves as the source material for homogenous lignin nanofiber scaffolds that are formed under the influence of a rotating drum operating at 300 rpm and 77 K of liquid nitrogen serving as a coolant.

To produce nanolignin *via* electrospinning, a solution of polymers is required as the base substance. The mixture can be fed through a 100 m wide nozzle that acts as an electrode and/or an auxiliary electrode system once it has been mixed with the appropriate solvent. When employing electrospinning to produce nanolignin, the optimal E-field level ranges from 100 and 500 kV m^{-1}.

Supercritical fluids like carbon dioxide (CO_2) are widely utilized in the synthesis of nanolignin due to their non-toxic, non-flammable, and cost-effective nature. Specifically, CO_2 is an ideal choice for supercritical fluid-mediated processes owing to its favorable critical pressure and temperature of 7.4 MPa and 304.3 K, respectively. By reacting lignin with CO_2 and acetone at 30 MPa and 35 °C, fine lignin nanoparticles with a diameter of 144 nm can be produced.[205]

The pressurized CO_2 antisolvent method can also be employed to create environmentally acceptable lignin nanoparticles (LNPs). In this technique, CO_2 is first pumped to a chiller maintained at 258.2 K and then transferred to the precipitator, where it is liquefied. After the precipitating unit has settled, a lignin remedy is added, and the resulting lignin nanoparticles are weighed. The mixture is then exposed to sonication for thirty minutes at room temperature and sprayed through a nozzle controlled by preset flow rates. The particles that pass through the paper filter are collected. It is important to note that this technique does not apply to CO_2 immiscible chemicals.[202]

In this process, an antisolvent such as water is mixed with a lignin solution that has been prepared in an organic solvent. The solvents used are not polar, and water is often employed as an antisolvent because lignin is either immiscible or sparingly soluble.[202] In one study, the antisolvent precipitation method was used to produce lignin particles (ALNP and DLNP) measuring 80–104 nm with strong UV shielding and radical scavenging properties. The researchers used acetone and water, constantly stirring at 20 °C and 300 rpm.[206] However, this method has some drawbacks, including the instability of the colloid system, the morphology of the generated nanoparticles, and the permanent nature of the solvents.[202]

Gilca *et al.*[203] used this method to create stable nanoparticles of lignin from a lignin (0.7%) solution. The aqueous solution was then subjected to an hour-long acoustic treatment using an ultrasonic probe that generated a 20 kHz frequency at 600 W. Mild conditions were applied to allow the uniform nanosuspension to dry.[203]

Another feedstock for LNP synthesis is steam-exploded rice straw lignin (SERSL). To create a homogeneous solution, the SERSL solution mixture and castor oil (20 wt%) were stirred. For four hours, 20 mL of (1 molar) HCl was added in the form of drops at 50 °C with the nitrogen setting in place. The newly generated lignin nanoparticles were thoroughly washed in ethanol and water until the pH reached 7.[207]

Nanoparticles with a diameter ranging from 45 to 250 nm were manufactured through a new method that utilized precursor materials including kraft lignin and organosolv. The synthesis process began with vortexing a kraft solution in ethylene glycol for 30 minutes and then filtering the mixture using a 0.45 μm pore size syringe filter. Subsequently, 1–3 mL of 0.025 M nitric acid was quickly combined with 5 mL of the purified solution in a scintillation vial, and the mixture was shaken continuously, resulting in the formation of particles.[208–210] The second method involved vortexing and filtering the acetone/organosolv solution in stock in the same initial steps. Ultimately, supersaturated lignin was separated into LNPs through the addition of 9.2 mL of water to a 1 mL filtrate solution.[211]

In another study, Popa *et al.* investigated the role of hydroxymethylation in the formation of lignin nanoparticles. The process involved stirring a lignin suspension in 47 mm of water for 120 minutes at room temperature using 10 g of lignin derived from Sarkanda grass and wheat straw. The lignin dispersion was then physically agitated for two hours before being sterilized with a 50% solution containing 1.29 g sodium hydroxide and a 25% solution consisting of 3.14 g ammonium hydroxide. The next step involved allowing a 37% solution with 6.7 g formaldehyde to react for four hours at 85 °C. After employing 1 N HCl (pH 2) in a recovery step, a precipitate was created, which was subsequently centrifuged to form a solid phase. The phase was rinsed twice with water and dried to produce LNPs.[212]

This method transforms lignin-derived non-uniform polymer clusters into uniformly distributed spherical colloidal nanoparticles. In one experiment, alkali lignin was produced by pulping black liquid, which was subsequently purified and acetylated. The acetylated lignin-tetrahydrofuran (ACL-THF) solution (1 mg mL^{-1}) was created by adding tetrahydrofuran to the solution, followed by water. The sudden increase in the ACL-THF solution's scattered light intensity suggests the presence of hydrophobic acetylated lignin molecules in the mixture, which promoted the formation of colloids. The critical water concentration of 44 vol% facilitated the formation of spherical colloids.[213,214]

Rugose wood components can be transformed into lignin-based nanoparticles using the supercritical antisolvent method, which combines several methods such as centrifugation, dissolution, precipitation, and the use of a CO_2 supercritical apparatus. In their patented process for creating industrial-grade xylogen NPs, the researchers chose carbonic acid as their preferred antisolvent.[215] The process of creating xylogen nanoparticles, which are particle sizes of less than 0.2 mm that have been alkali treated and freeze-dried to create particles as small as 26 nm. The resulting xylogen NPs have an average particle size of 30 nm.[216]

These xylogen NPs, when used as fillers in organic rubber, can improve vulcanization and create stable 100 nm lignin

nanoparticles. To synthesize the NR/LPCs (natural rubber/lignin-poly (diallyldimethylammonium chloride) (PDADMAC) complexes) composites, a colloidal LPC solution is mixed with a 2% PDADMAC solution at an alkaline pH. The resulting electrostatically assembled solution is then coprecipitated with H_2SO_4 (pH 2) to create the final product. The necessary steps include filtration, cleaning, and drying under vacuum at 50 °C to produce the NR/LPCs nanocomposites.[217]

Future perspectives and summary. Lignin possesses several promising applications, particularly in biopolymers, but several variables hinder its integration into polymer matrices. The structural and compositional diversity of lignin is impacted by factors such as the plant's source and extraction technique. To fully comprehend the potential of lignin in various sectors, it is crucial to characterize both the raw materials and final products comprehensively. The many types of lignin emphasize the significance of understanding the structure–activity relationship.

Incorporating lignin into polymer films for food packaging can enhance mechanical strength, gas barrier properties, and provide antioxidant and anti-UV benefits. However, compatibility issues and the risk of phase separation pose challenges when adding lignin to these materials. To effectively address this issue, it is crucial to functionalize lignin or its nanoparticles in order to achieve uniformity in the film. Furthermore, additional research is needed to investigate the relationships between lignin and other compounds, as well as the digestibility of lignin-based films. Assessing the influence of lignin on the compostability and degradability of the final product is also essential, considering its limited degradability in composting environments.

The exact mechanism by which lignin acts as an antibacterial agent is still uncertain and is the subject of ongoing research. In pharmaceutical and biological contexts, lignin's inconsistent molecular weight, impurities, and reactive group composition present both opportunities and challenges, which complicates the evaluation of its efficacy and safety. Despite research demonstrating lignin's antibacterial properties in solution, there is still much to learn about using it as a coating material for antimicrobial surfaces. The development of lignin coatings with the ability to directly inactivate microorganisms appears promising, given the significant role that surface transmission plays in the spread of bacteria. Further studies should focus on refining coating preparation and assessing effectiveness against a diverse range of infections.

In summary, lignin offers a wide range of potential applications, particularly in the food industry, due to its versatility and availability. Its ability to serve as a sustainable material has garnered interest owing to its antioxidant and antibacterial properties. However, further research is necessary to address safety concerns and better understand its interactions with other food components. Moreover, there is a need for more research to demonstrate its effectiveness in real-world food systems and to clarify its *in vivo* bioactivities. To fully exploit lignin's potential in various sectors such as materials science, pharmaceuticals, and packaging, affordable valorization technologies must be developed. By utilizing lignin's unique properties, we can enhance revenue streams, improve environmental performance, and accelerate the transition toward a more sustainable and eco-friendly economy.

Conflicts of interest

There are no conflicts to declare.

References

1 R. H. Branco, L. S. Serafim and A. M. Xavier, *Fermentation*, 2018, **5**, 4.
2 E. Mnich, N. Bjarnholt, A. Eudes, J. Harholt, C. Holland, B. Jørgensen, F. H. Larsen, M. Liu, R. Manat and A. S. Meyer, *Nat. Prod. Rep.*, 2020, **37**, 919–961.
3 A. Rahman, O. Farrok and M. M. Haque, *Renewable Sustainable Energy Rev.*, 2022, **161**, 112279.
4 R. K. Srivastava, N. P. Shetti, K. R. Reddy and T. M. Aminabhavi, *Sci. Total Environ.*, 2020, **722**, 137927.
5 A. Boarino and H.-A. Klok, *Biomacromolecules*, 2023, **24**, 1065–1077.
6 M. Xie, W. Muchero, A. C. Bryan, K. Yee, H. B. Guo, J. Zhang, T. J. Tschaplinski, V. R. Singan, E. Lindquist, R. S. Payyavula, J. Barros-Rios, R. Dixon, N. Engle, R. W. Sykes, M. Davis, S. S. Jawdy, L. E. Gunter, O. Thompson, S. P. DiFazio, L. M. Evans, K. Winkeler, C. Collins, J. Schmutz, H. Guo, U. Kalluri, M. Rodriguez, K. Feng, J. G. Chen and G. A. Tuskan, *Plant Cell*, 2018, **30**, 1645–1660.
7 C. D. Pham, M. D. T. Dang, T. B. Ly, K. D. Tran, N. T. Vo, N. H. N. Do, P. T. Mai and P. K. Le, *Int. J. Biol. Macromol.*, 2023, **230**, 123175.
8 D. S. Bajwa, G. Pourhashem, A. H. Ullah and S. G. Bajwa, *Ind. Crops Prod.*, 2019, **139**, 111526.
9 E. M. Zadeh, S. F. O'Keefe and Y.-T. Kim, *ACS Omega*, 2018, **3**, 7388–7398.
10 D. de Oliveira Begali, L. F. Ferreira, A. C. S. de Oliveira, S. V. Borges, A. R. de Sena Neto, C. R. de Oliveira, M. I. Yoshida and C. I. Sarantopoulos, *Int. J. Biol. Macromol.*, 2021, **180**, 262–271.
11 R. Shorey, A. Salaghi, P. Fatehi and T. Mekonnen, *RSC Sustainability*, 2024, **2**(4), 804–831.
12 A. Akbarian, A. Andooz, E. Kowsari, S. Ramakrishna, S. Asgari and Z. A. Cheshmeh, *Bioresour. Technol.*, 2022, **362**, 127774.
13 H. Lu, V. Yadav, M. Bilal and H. M. Iqbal, *Chemosphere*, 2022, **288**, 132574.
14 B. Abraham, V. Syamnath, K. Arun, P. F. Zahra, P. Anjusha, A. Kothakotta, Y.-H. Chen, V. K. Ponnusamy and P. Nisha, *Sci. Total Environ.*, 2023, **881**, 163316.
15 S. Sethupathy, G. Murillo Morales, L. Gao, H. Wang, B. Yang, J. Jiang, J. Sun and D. Zhu, *Bioresour. Technol.*, 2022, **347**, 126696.
16 Z. Ma, J. Wang, H. Zhou, Y. Zhang, Y. Yang, X. Liu, J. Ye, D. Chen and S. Wang, *Fuel Process. Technol.*, 2018, **181**, 142–156.

17 S. Bertella and J. S. Luterbacher, *Trends Chem.*, 2020, **2**, 440–453.
18 S. Sharma, A. Sharma, S. I. Mulla, D. Pant, T. Sharma and A. Kumar, *Lignin: biosynthesis and transformation for industrial applications*, 2020, pp. 1–15.
19 D. Huang, R. Li, P. Xu, T. Li, R. Deng, S. Chen and Q. Zhang, *Chem. Eng. J.*, 2020, **402**, 126237.
20 M. Tortora, F. Cavalieri, P. Mosesso, F. Ciaffardini, F. Melone and C. Crestini, *Biomacromolecules*, 2014, **15**, 1634–1643.
21 M. Bertolo, L. B. Brenelli de Paiva, V. Nascimento, C. Gandin, M. Neto, C. Driemeier and S. Rabelo, *Ind. Crops Prod.*, 2019, **140**, 111591.
22 B. Abraham, T. R. Reshmitha, M. M. Navami, L. George, V. V. Venugopalan and P. Nisha, *Ind. Crops Prod.*, 2020, **151**, 112451.
23 M. P. Ajith, M. Aswathi, E. Priyadarshini and P. Rajamani, *Bioresour. Technol.*, 2021, **342**, 126000.
24 P. G, S. As, J. S. Jayan, A. Raman and A. Saritha, *Process Saf. Environ. Prot.*, 2021, **145**, 395–410.
25 B. Abraham, V. L. Syamnath, K. B. Arun, P. M. Fathima Zahra, P. Anjusha, A. Kothakotta, Y.-H. Chen, V. K. Ponnusamy and P. Nisha, *Sci. Total Environ.*, 2023, **881**, 163316.
26 A. P. Richter, J. S. Brown, B. Bharti, A. Wang, S. Gangwal, K. Houck, E. A. Cohen Hubal, V. N. Paunov, S. D. Stoyanov and O. D. Velev, *Nat. Nanotechnol.*, 2015, **10**, 817–823.
27 A. Memic and T. Abudula, Antioxidant, antibacterial, injectable lignin-gelatin composite cryogels for wound healing and tissue engineering, *US pat.*, 10881760, King Abdulaziz University, 2021.
28 B. Timotebo and P. A. C. Uduso, Process method for treating structures of lignocellulosic material and method for manufacturing composite structures, *JP Pat.*, 7275335B2, 2015, **https://patents.google.com/patent/JP7275335B2/en?q=(Lignin+use+in+Packaging)&oq=Lignin+use+in+Packaging+**.
29 J. Deng, T. Xiong, H. Wang, A. Zheng and Y. Wang, *ACS Sustain. Chem. Eng.*, 2016, **4**, 3750–3756.
30 Q. Zhao, H. Wang, Y. Yin, Y. Xu, F. Chen and R. A. Dixon, *Proc. Natl. Acad. Sci. U. S. A.*, 2010, **107**, 14496–14501.
31 R. Vanholme, B. Demedts, K. Morreel, J. Ralph and W. Boerjan, *Plant Physiol.*, 2010, **153**, 895–905.
32 M. Baucher, B. Chabbert, G. Pilate, J. Van Doorsselaere, M.-T. Tollier, M. Petit-Conil, D. Cornu, B. Monties, M. Van Montagu and D. Inze, *J. Plant Physiol.*, 1996, **112**, 1479–1490.
33 H. Hatakeyama, T. Hatakeyama and J. Advances, *Polym. Sci.*, 2009, **232**, 1–63.
34 A. Yanez-McKay, W. Li, R. Mabon and L. Broadbelt, *Energy Fuels*, 2016, **30**, 5835–5845.
35 P. Bajpai, *Carbon Fibre from Lignin*, Springer, 2017.
36 K. Gabov, R. J. Gosselink, A. I. Smeds and P. Fardim, *J. Agric. Food Chem.*, 2014, **62**, 10759–10767.
37 M. Bunzel, A. Seiler and H. Steinhart, *J. Agric. Food Chem.*, 2005, **53**, 9553–9559.
38 K. Ross and G. Mazza, *Int. J. Mol. Sci.*, 2010, **11**, 4035–4050.
39 H.-S. Yin, H.-M. Liu and Y.-L. Liu, *Molecules*, 2017, **22**, 890.
40 Y. Zhang, L. Yang, D. Wang and D. Li, *Int. J. Biol. Macromol.*, 2018, **107**, 1193–1202.
41 K. Holtman, H. M. Chang and J. Kadla, *J. Wood Chem. Technol.*, 2007, **27**, 179–200.
42 N. Terashima, T. Awano, K. Takabe and M. Yoshida, *C. R. Biol.*, 2004, **327**, 903–910.
43 H. Ohi, Y. Ju and K. Ken-Ichi, *Jpn. Tappi J.*, 1997, (10), 578–586.
44 H. Hatakeyama and T. J. B. l. Hatakeyama, *Proteins, Bioactive Nanocomposites*, 2010, pp. 1–63.
45 N. M. Stark, D. J. Yelle and U. P. Agarwal, in *Lignin in Polymer Composites*, ed. O. Faruk and M. Sain, William Andrew Publishing, 2016, pp. 46–66, DOI: **10.1016/B978-0-323-35565-0.00004-7**.
46 G. Brunow and K. Lundquist, *Lignin Lignans: Advances in Chemistry*, 2010, vol. 1.
47 R. J. Sammons, D. P. Harper, N. J. Labbe, J. J. Bozell, T. J. Elder and T. Rials, *J. Bioresour.*, 2013, **8**, 2752–2767.
48 Y.-C. Lu, Y. Lu and X. Fan, *Lignin: Biosynthesis and Transformation for Industrial Applications*, 2020, pp. 17–75.
49 O. Y. Abdelaziz and C. P. Hulteberg, *Waste Biomass Valorization*, 2017, **8**, 859–869.
50 X. Erdocia, R. Prado, M. Corcuera and J. Labidi, *J. Ind. Eng. Chem.*, 2013, **20**, 1103–1108.
51 M. Ek, G. Gellerstedt and G. Henriksson, *Ljungberg Textbook: Pulp and Paper Chemistry and Technology*, De Gruyter Open, 2007.
52 P. Guiraud, R. Steiman, F. Seigle-Murandi and J. L. Benoit-Guyod, *Ecotoxicol. Environ. Saf.*, 1995, **32**, 29–33.
53 S. Haghdan, S. Renneckar and G. D. Smith, *Lignin in Polymer Composites*, 2016, vol. 1, pp. 1–11.
54 M. P. Vinardell and M. Mitjans, *Int. J. Mol. Sci.*, 2017, **18**, 1219.
55 H. Chung and N. R. Washburn, in *Lignin in Polymer Composites*, ed. O. Faruk and M. Sain, William Andrew Publishing, 2016, pp. 13–25, DOI: **10.1016/B978-0-323-35565-0.00002-3**.
56 J. Jiang, W. Cun, X. Wu, Q. Shi, H. Tang and G. Luo, *J. Virol.*, 2012, **86**, 7256–7267.
57 E. P. Feofilova and I. S. Mysyakina, *Prikl. Biokhim. Mikrobiol.*, 2016, **52**, 559–569.
58 S. Y. Lin and C. W. Dence, *Methods in Lignin Chemistry*, Springer Science & Business Media, 2012.
59 J. Tao, S. Li, F. Ye, Y. Zhou, L. Lei and G. Zhao, *Crit. Rev. Food Sci. Nutr.*, 2020, **60**, 2011–2033.
60 K. Holtman, H.-M. Chang, H. Jameel and J. Kadla, *J. Wood Chem. Technol.*, 2006, **26**, 21–34.
61 R. El Hage, N. Brosse, L. Chrusciel, C. Sanchez, P. Sannigrahi and A. Ragauskas, *Polym. Degrad. Stab.*, 2009, **94**, 1632–1638.
62 J. Rencoret, G. Marques, A. Gutiérrez, L. Nieto, J. Jiménez-Barbero, A. T. Martinez and J. del Río, *Ind. Crops Prod.*, 2009, **30**, 137–143.
63 A. J. Bjorkman, *Ind. Eng. Chem.*, 1957, **49**, 1395–1398.

64 M. Carrier, A. Loppinet-Serani, D. Denux, J.-M. Lasnier, F. Ham-Pichavant, F. Cansell and C. Aymonier, *Biomass Bioenergy*, 2011, **35**, 298–307.
65 D. L. Brink, Method of Treating Biomass Material, *US Pat.*, 5366558, 1993.
66 D. J. Horst, J. J. R. Behainne, P. P. de Andrade Júnior and J. L. Kovaleski, *J. Energy Sustain. Dev.*, 2014, **23**, 78–84.
67 X. Ma and G. Zhao, *J. Appl. Polym. Sci.*, 2011, **121**, 3525–3530.
68 V. Chaturvedi and P. Verma, *3 Biotech*, 2013, **3**, 415–431.
69 I. M. Cabott, Experiments on the pulping of periodate lignin by the sulfite process, *PhD thesis*, 1951, **https://escholarship.mcgill.ca/concern/theses/v979v615m**.
70 Y. Sun and J. Cheng, *Bioresour. Technol.*, 2002, **83**, 1–11.
71 A. Sluiter, B. Hames, R. Ruiz, C. Scarlata, J. Sluiter, D. Templeton and D. Crocker, Determination of structural carbohydrates and lignin in biomass, in *Laboratory Analytical Procedure (LAP)*, National Renewable Energy Laboratory, 2008, 1617, pp. 1–16.
72 G.-L. Guo, W.-H. Chen, W.-H. Chen, L.-C. Men and W.-S. Hwang, *Bioresour. Technol.*, 2008, **99**, 6046–6053.
73 M. Bunzel, A. Schu∈ßler and G. H. Saha, *J. Agric. Food Chem.*, 2011, (23), 12506–12513.
74 L. E. Beramendi-Orosco, M. Castro-Díaz, C. E. Snape, C. H. Vane and D. J. Large, *Org. Geochem.*, 2004, **35**, 61–72.
75 Z. Hu, T.-F. Yeh, H.-M. Chang, Y. Matsumoto and J. Kadla, *Holzforschung*, 2006, **60**, 389–397.
76 Q. Yin, W. Yang, C. Sun and M. Di, *Bioresources*, 2012, **7**, 5737–5748.
77 J. Domínguez-Robles, R. Sánchez, E. Espinosa, D. Savy, P. Mazzei, A. Piccolo and A. Rodríguez, *Int. J. Mol. Sci.*, 2017, **18**, 327.
78 J. Gierer, *Wood Sci. Technol.*, 1980, **14**, 241–266.
79 A. Maček, *Prog. Energy Combust. Sci.*, 1999, **25**, 275–304.
80 G. Jiang, D. Nowakowski and T. Bridgwater, *Thermochim. Acta*, 2010, **498**, 61–66.
81 K. Lundquist, R. Simonson and K. Tingsvik, *Pulp Pap. J.*, 1981, 63.
82 N.-E. Mansouri and J. Salvadó, *Ind. Crop. Prod.*, 2007, **26**, 116–124.
83 K. Shimizu, K. Sudo, H. Ono, M. Ishihara, T. Fujii and S. Hishiyama, *Biomass Bioenergy*, 1998, **14**, 195–203.
84 A. Corey, K. Wamsley, T. Winowiski and J. Moritz, *J. Appl. Poultry Res.*, 2014, **23**, 418–428.
85 L. Bosong, L. Wei, Z. Qi, W. Tiejun and M. Longlong, *J. Anal. Appl. Pyrolysis*, 2014, **108**, 106355.
86 S. Kang, B. Li, J. Chang and J. Fan, *J. Bioresour.*, 2011, **6**, 243–252.
87 X. Ouyang, X. Qiu and P. Chen, *Colloids Surf., A*, 2006, **282**, 489–497.
88 F. Ferdosian and C. Xu, *Conversion of Lignin into Bio-Based Chemicals and Materials*, Springer, 2017.
89 E. Muurinen, *Organosolv pulping: A review and distillation study related to peroxyacid pulping*, 2000, **https://urn.fi/URN:ISBN:9514256611**.
90 H. Kirçi, S. Bostanci and M. K. Yalinkilic, *Technology*, 1994, **28**, 89–99.
91 S. Hidayati, A. Zuidar, W. Satyajaya, M. Murhadi and D. Retnowati, *IOP Conf. Ser.: Mater. Sci. Eng.*, 2018, **344**, 012006.
92 A. Lindner and G. Wegener, *J. Wood Chem. Technol.*, 1988, **8**, 323–340.
93 G. Vázquez, G. Antorrena, J. González, S. Freire and S. López, *J. Bioresour. Technol.*, 1997, **59**, 121–127.
94 X. Pan, D. Xie, R. Yu, D. Lam and J. Saddler, *Ind. Eng. Chem. Res.*, 2007, **46**, 2609–2617.
95 M. Suchy and D. S. Argyropoulos, in *Oxidative Delignification Chemistry*, ACS Symposium Series, American Chemical Society, 2001, vol. 785, ch. 1, pp. 2–43.
96 K. Saha, J. Dasgupta, S. Chakraborty, F. Antunes, J. Sikder, S. Curcio, J. Santos, H. Arafat and S. da Silva, *Cellulose*, 2017, **24**, 1–17.
97 E. A. Agustiany, M. Rasyidur Ridho, M. Rahmi DN, E. W. Madyaratri, F. Falah, M. A. R. Lubis, N. N. Solihat, F. A. Syamani, P. Karungamye and A. Sohail, *J. Polym. Compos.*, 2022, **43**, 4848–4865.
98 V. P. Saraf and W. G. Glasser, *J. Appl. Polym. Sci.*, 1984, **29**, 1831–1841.
99 Y. Zhang and M. Naebe, *ACS Sustain. Chem. Eng.*, 2021, **9**, 1427–1442.
100 T. Wang, M. Wang, L. Fu, Z. Duan, Y. Chen, X. Hou, Y. Wu, S. Li, L. Guo and R. Kang, *Sci. Rep.*, 2018, **8**, 1557.
101 S. Li, J. Zhou, G. Chen, S. Qin, H. Li and Y. Chen, *Trans. Chin. Soc. Agric. Eng.*, 2021, **37**, 279–286.
102 Y. Wang and G. W. Padua, *Macromol. Mater. Eng.*, 2003, **288**, 886–893.
103 L. R. Chiappero, S. S. Bartolomei, D. A. Estenoz, E. A. Moura and V. V. Nicolau, *J. Polym. Environ.*, 2021, **29**, 450–459.
104 S. Edebali, *Iran. J. Chem. Chem. Eng.*, 2020, **39**, 245–251.
105 D. Dixit, R. Pal, G. Kapoor and M. Stabenau, *Lightweight Ballistic Composites*, Elsevier, 2016, pp. 157–216.
106 Y. Kang, Z. Chen, B. Wang and Y. Yang, *Ind. Crops Prod.*, 2014, **56**, 105–112.
107 A. Gregorova, M. Hrabalova, R. Kovalcik and R. Wimmer, *J. Polym. Eng.*, 2011, **51**, 143–150.
108 R. Wulandari, *J. Phys.: Conf. Ser.*, 2019, **1295**, 012058.
109 F. S. Mostafavi and D. Zaeim, *Int. J. Biol. Macromol.*, 2020, **159**, 1165–1176.
110 N. M. Sá, A. L. Mattos, L. M. Silva, E. S. Brito, M. F. Rosa and H. M. Azeredo, *Int. J. Biol. Macromol.*, 2020, **161**, 1337–1345.
111 B. Rukmanikrishnan, S. Ramalingam, S. K. Rajasekharan, J. Lee and J. Lee, *Int. J. Biol. Macromol.*, 2020, **153**, 55–62.
112 E. M. Zadeh, S. F. O'Keefe and Y.-T. Kim, *ACS Omega*, 2018, **3**, 7388–7398.
113 J. M. Silva, C. Vilela, A. V. Girão, P. C. Branco, J. Martins, M. G. Freire, A. J. Silvestre and C. S. Freire, *Carbohydrate Polym.*, 2024, **337**, 122112.
114 S. Kirar, D. Mohne, M. Singh, V. Sagar, A. Bhise, S. Goswami and J. Bhaumik, *Sustain. Mater. Technol.*, 2024, e00864.
115 Q. Zheng, P. O. Osei, S. Shi, S. Yang and X. Wu, *Food Biosci.*, 2024, **59**, 104022.
116 S. Garg and A. Avanthi, *bioRxiv*, 2024, preprint, DOI: **10.1101/2024.05.03.592363**.

117 P. Beniwal, D. Guliani and A. P. Toor, *J. Polym. Res.*, 2024, **31**, 68.
118 L. A. Worku, M. G. Tadesse, A. Bachheti, D. Pandey, A. K. Chandel, A. W. Ewuntu and R. K. Bachheti, *Int. J. Biol. Macromol.*, 2024, **254**, 127644.
119 U. Sangeetha, N. Sudhakaran, P. Parvathy, M. Abraham, S. Das, S. De and S. K. Sahoo, *Int. J. Biol. Macromol.*, 2024, **266**, 131005.
120 S. Wang, Y. Hao, Q. He and Q. Gao, *J. Thermoplast. Compos. Mater.*, 2024, 08927057241233566.
121 Y. He, H.-C. Ye, T.-T. You and F. Xu, *Food Hydrocolloids*, 2023, **137**, 108355.
122 A. Riaz, H. Mostafa, K. G. Lawal, N. Sivapragasam, T. Ramachandran, F. Hamed, I. Manikas, B. Sundarakani, C. Stathopoulos and S. Maqsood, *Food Biophys.*, 2024, 1–13.
123 P. Nuamduang, R. Auras, C. Winotapun, B. Hararak, W. Wanmolee and P. Leelaphiwat, *Int. J. Biol. Macromol.*, 2024, 131185.
124 J. Dou, T. Vuorinen, H. Koivula, N. Forsman, M. Sipponen and S. Hietala, *ACS Appl. Nano Mater.*, 2021, **4**, 2921–2929.
125 E. S. Esakkimuthu, D. DeVallance, I. Pylypchuk, A. Moreno, M. H. Sipponen and J. Frontiers, *Biotechnol. Bioeng.*, 2022, **10**, 1025076.
126 M. Vinardell, V. Ugartondo and M. Mitjans, *Ind. Crops Prod.*, 2008, **27**, 220–223.
127 N. H. A. Latif, A. A. Rahim, N. Brosse and M. H. Hussin, *Int. J. Biol. Macromol.*, 2019, **130**, 947–957.
128 M. F. Li, S. N. Sun, F. Xu and R. C. Sun, *Food Chem.*, 2012, **134**, 1392–1398.
129 X. Dong, M. Dong, Y. Lu, A. Turley, T. Jin and C. Wu, *Ind. Crops Prod.*, 2011, **34**, 1629–1634.
130 W. Gong, Z. Xiang, F. Ye and G. Zhao, *Ind. Crops Prod.*, 2016, **91**, 340–349.
131 A. Salanti, L. Zoia, M. Orlandi, F. Zanini and G. Elegir, *J. Agric. Food Chem.*, 2010, **58**, 10049–10055.
132 W. Gong, Z. Ran, F. Ye and G. Zhao, *Food Chem.*, 2017, **228**, 455–462.
133 S. S. Qazi, D. Li, C. Briens, F. Berruti and M. M. Abou-Zaid, *Molecules*, 2017, **22**, 372.
134 A. Arshanitsa, J. Ponomarenko, T. Dizhbite, A. Andersone, R. Gosselink, J. Putten, M. Lauberts and G. Telysheva, *J. Anal. Appl. Pyrolysis*, 2013, **103**, 78–85.
135 V. Ugartondo, M. Mitjans and M. P. Vinardell, *Bioresour. Technol.*, 2008, **99**, 6683–6687.
136 B. Košíková, J. Lábaj, A. Kovalcik and D. Slameňová, *Holzforschung*, 2006, **60**, 166–170.
137 E. P. o. F. Additives and N. S. a. t. Food, *EFSA J.*, 2011, **9**, 2319.
138 J. Wang, L. Tian, B. Luo, S. Ramakrishna, D. Kai, X. J. Loh, I. H. Yang, G. R. Deen and X. Mo, *Colloids Surf. B Biointerfaces*, 2018, **169**, 356–365.
139 G. L. Catignani and M. E. Carter, *J. Food Sci.*, 1982, **47**, 1745.
140 Joint FAO and WHO Expert Committee on Food Additives, Safety evaluation of certain food additives/prepared by the sixty-ninth meeting of the Joint FAO/WHO Expert Committee on Food Additives (JEFCA), in *Safety evaluation of certain food additives/prepared by the sixty-ninth meeting of the Joint FAO/WHO Expert Committee on Food Additives (JEFCA)*, 2009, **https://kohahq.searo.who.int/cgi-bin/koha/opac-detail.pl?biblionumber=31771&shelfbrowse_itemnumber=56006**.
141 B. Baurhoo, C. Ruiz-Feria and X. Zhao, *Anim. Feed Sci. Technol.*, 2008, **144**, 175–184.
142 K. Toh, H. Yokoyama, H. Noda and Y. Yuguchi, *J. Food Biochem.*, 2010, **34**, 192–206.
143 D. Liu, Y. Li, Y. Qian, Y. Xiao, S. Du and X. Qiu, *ACS Sustain. Chem. Eng.*, 2017, **5**, 8424–8428.
144 T. Dizhbite, G. Telysheva, V. Jurkjane and U. Viesturs, *Bioresour. Technol.*, 2004, **95**, 309–317.
145 A. García, G. Spigno and J. Labidi, *Ind. Crops Prod.*, 2017, **104**, 242–252.
146 R. Y. Nsimba, N. West and A. A. Boateng, *J. Agric. Food Chem.*, 2012, **60**, 12525–12530.
147 L. An, G. Wang, H. Jia, C. Liu, W. Sui and C. Si, *Int. J. Biol. Macromol.*, 2017, **99**, 674–681.
148 X. Pan, J. F. Kadla, K. Ehara, N. Gilkes and J. N. Saddler, *J. Agric. Food Chem.*, 2006, **54**, 5806–5813.
149 Anushikha and K. K. Gaikwad, *Biomass Convers. Biorefin.*, 2023, 1–13.
150 E. Gogo, A. Opiyo, K. Hassenberg, C. Ulrichs and S. Huyskens-Keil, *Postharvest Biol. Technol.*, 2017, **129**, 107–117.
151 J. Sunthornvarabhas, S. Liengprayoon, T. Lerksamran, C. Buratcharin, T. Suwonsichon, W. Vanichsriratana and K. Sriroth, *Sugar Technol.*, 2018, **21**, 355–363.
152 A. M. A. Nada, A. I. ElDiwany and A. M. Elshafei, *Acta Biotechnol.*, 1989, **9**, 295–298.
153 J. D. C. Medina, A. L. Woiciechowski, A. Zandona Filho, L. Bissoqui, M. D. Noseda, L. P. de Souza Vandenberghe, S. F. Zawadzki and C. R. Soccol, *Ind. Crop. Prod.*, 2016, **94**, 630–637.
154 B. Baurhoo, A. Letellier, X. Zhao and C. A. Ruiz-Feria, *Poultry Sci.*, 2007, **86**, 2509–2516.
155 M. T. Madigan, J. M. Martinko and J. Parker, *Brock Biology of Microorganisms*, Prentice hall, Upper Saddle River, NJ, 2015.
156 S. Zhang, X. Cheng, Q. Fu, Y. Li, P. Wu, Y. Qiao, J. Yan, L. Si, G. I. Waterhouse and H. Li, *Food Hydrocolloids*, 2023, **143**, 108783.
157 Y. Lu, M. Jiang, Y. Pan, F. Wang, W. Xu, Y. Zhou and X. Du, *Int. J. Biol. Macromol.*, 2024, **254**, 127630.
158 E. Cavallo, X. He, F. Luzi, F. Dominici, P. Cerrutti, C. Bernal, M. L. Foresti, L. Torre and D. Puglia, *Molecules*, 2020, **26**, 126.
159 E. Gerbin, G. Rivière, L. Foulon, Y.-M. Frapart, B. Cottyn, M. Pernes, C. Marcuello, B. Godon, A. Gainvors-Claisse and D. Crônier, *Int. J. Biol. Macromol.*, 2021, **181**, 136–149.
160 E. Lizundia, I. Armentano, F. Luzi, F. Bertoglio, E. Restivo, L. Visai, L. Torre and D. Puglia, *ACS Appl. Bio Mater.*, 2020, **3**, 5263–5274.
161 F. Falah, D. Zulfiana, M. Septiano, D. S. Nawawi, F. P. Sari, W. Fatriasari and N. N. Solihat, Antimicrobial activity of food packaging biofilms derived from lignin-starch-poly

(lactic acid), in *AIP Conference Proceedings*, AIP Publishing, 2024, vol. 2973, p. 1.
162 L. Xiao, Z. Yao, Y. He, Z. Han, X. Zhang, C. Li, P. Xu, W. Yang and P. Ma, *Ind. Crops Prod.*, 2022, **187**, 115515.
163 Y. Li, Y. Chen, Q. Wu, J. Huang, Y. Zhao, Q. Li and S. Wang, *Polymers*, 2022, **14**, 1705.
164 X. Zhang, W. Liu, D. Sun, J. Huang, X. Qiu, Z. Li and X. Wu, *ChemSusChem*, 2020, **13**, 4974–4984.
165 H. Sakagami, T. Kushida, T. Oizumi, H. Nakashima and T. Makino, *Pharmacol. Therapeut.*, 2010, **128**, 91–105.
166 S. C. Gordts, G. Férir, T. D'huys, M. I. Petrova, S. Lebeer, R. Snoeck, G. Andrei and D. Schols, *PLoS One*, 2015, **10**, e0131219.
167 H. Sakagami, K. Asano, K. Satoh, K. Takahashi, M. Kobayashi, N. Koga, H. Takahashi, R. Tachikawa, T. Tashiro, A. Hasegawa, K. Kurihara, T. Ikarashi, T. Kanamoto, S. Terakubo, H. Nakashima, S. Watanabe and W. Nakamura, *In Vivo*, 2007, **21**, 499–505.
168 H. Sakagami, K. Satoh, H. Fukamachi, T. Ikarashi, A. Shimizu, K. Yano, T. Kanamoto, S. Terakubo, H. Nakashima, H. Hasegawa, A. Nomura, K. Utsumi, M. Yamamoto, Y. Maeda and K. Osawa, *In Vivo*, 2008, **22**, 327–332.
169 K. Matsuhisa, S. Yamane, T. Okamoto, A. Watari, M. Kondoh, Y. Matsuura and K. Yagi, *Biochem. Biophys. Res. Commun.*, 2015, **462**, 52–57.
170 P. Srisapoome, K. Hamano, I. Tsutsui and K. Iiyama, *Fish Shellfish Immunol.*, 2018, **72**, 494–501.
171 P. K. Lai, J. Donovan, H. Takayama, H. Sakagami, A. Tanaka, K. Konno and M. Nonoyama, *AIDS Res. Hum. Retrovir.*, 1990, **6**, 205–217.
172 J.-B. Lee, C. Yamagishi, K. Hayashi and T. Hayashi, *Biosci. Biotechnol. Biochem.*, 2011, **75**, 459–465.
173 H. Sakagami, M. Kawano, M. M. Thet, K. Hashimoto, K. Satoh, T. Kanamoto, S. Terakubo, H. Nakashima, Y. Haishima, Y. Maeda and K. Sakurai, *In Vivo*, 2011, **25**, 229–236.
174 M. Qiu, Q. Wang, Y. Chu, Z. Yuan, H. Song, Z. Chen and Z. Wu, *PLoS One*, 2012, **7**, e35906.
175 B. S. Lopez, M. Yamamoto, K. Utsumi, C. Aratsu and H. Sakagami, *In Vivo*, 2009, **23**, 1011–1016.
176 M. P. Vinardell and M. Mitjans, *Int. J. Mol. Sci.*, 2017, **18**, 1219.
177 A. Raghuraman, V. Tiwari, Q. Zhao, D. Shukla, A. K. Debnath and U. R. Desai, *Biomacromolecules*, 2007, **8**, 1759–1763.
178 T. Ichimura, T. Otake, H. Mori and S. Maruyama, *Biosc. Biotech. Biochem.*, 1999, **63**, 2202–2204.
179 K. Nagata, H. Sakagami, H. Harada, M. Nonoyama, A. Ishihama and K. Konno, *Antivir. Res.*, 1990, **13**, 11–21.
180 W. Yang, Y. Weng, D. Puglia, G. Qi, W. Dong, J. M. Kenny and P. Ma, *Int. J. Biol. Macromol.*, 2020, **144**, 102–110.
181 R. Kaur, S. Bhardwaj, S. Chandna, K.-H. Kim and J. Bhaumik, *J. Clean. Prod.*, 2021, **317**, 128300.
182 H. Sadeghifar, R. Venditti, J. Jur, R. Gorga and J. Pawlak, *ACS Sustain. Chem. Eng.*, 2016, **5**, 625–631.
183 Q. Xing, P. Buono, D. Ruch, P. Dubois, L. Wu and W. Wang, *ACS Sustain. Chem. Eng.*, 2019, **7**, 4147–4157.
184 Y. Kim, J. Suhr, H.-W. Seo, H. Sun, S. Kim, I.-K. Park, S.-H. Kim, Y. Lee, K.-J. Kim and J.-D. Nam, *Sci. Rep.*, 2017, **7**, 43596.
185 P. Posoknistakul, C. Tangkrakul, P. Chaosuanphae, S. Deepentham, W. Techasawong, N. Phonphirunrot, S. Bairak, C. Sakdaronnarong and N. Laosiripojana, *ACS Omega*, 2020, **5**, 20976–20982.
186 R. Kaur, N. S. Thakur, S. Chandna and J. Bhaumik, *J. Mater. Chem. B*, 2020, **8**, 260–269.
187 A. Raman, A. Sankar, A. SD, A. Anilkumar and A. Saritha, *Macromol. Mater. Eng.*, 2022, **307**, 2200114.
188 Q. Xing, D. Ruch, P. Dubois, L. Wu and W.-J. Wang, *ACS Sustain. Chem. Eng.*, 2017, **5**, 10342–10351.
189 F. Avelino, D. R. de Oliveira, S. E. Mazzetto and D. Lomonaco, *Int. J. Biol. Macromol.*, 2019, **125**, 171–180.
190 Y. Chai, Y. Wang, B. Li, W. Qi, R. Su and Z. He, *Langmuir*, 2021, **37**, 7219–7226.
191 D. Barnard and K. Heaton, *Gut*, 1973, **14**, 316–318.
192 P. Figueiredo, K. Lintinen, A. Kiriazis, V. Hynninen, Z. Liu, T. Bauleth-Ramos, A. Rahikkala, A. Correia, T. Kohout, B. Sarmento, J. Yli-Kauhaluoma, J. Hirvonen, O. Ikkala, M. A. Kostiainen and H. A. Santos, *Biomaterials*, 2017, **121**, 97–108.
193 H. Sakagami, *J. Pharmacol. Sci.*, 2014, **126**, 92–106.
194 A. Y. Mehta, B. M. Mohammed, E. J. Martin, D. F. Brophy, D. Gailani and U. R. Desai, *J. Thromb. Haemostasis*, 2016, **14**, 828–838.
195 B. Saluja, J. N. Thakkar, H. Li, U. R. Desai and M. Sakagami, *Pulm. Pharmacol. Therapeut.*, 2013, **26**, 296–304.
196 I. L. Cameron, W. E. Hardman and D. W. Heitman, *Nutr. Cancer*, 1997, **28**, 170–176.
197 L. Andrijevic, K. Radotic, J. Bogdanovic, D. Mutavdzic and G. Bogdanovic, *Journal of B.U.ON. : official journal of the Balkan Union of Oncology*, 2008, **13**, 241–244.
198 R. G. Saratale, G. D. Saratale, G. Ghodake, S. K. Cho, A. Kadam, G. Kumar, B. H. Jeon, D. Pant, A. Bhatnagar and H. S. Shin, *Int. J. Biol. Macromol.*, 2019, **128**, 391–400.
199 M. A. Eastwood and R. H. Girdwood, *Lancet*, 1968, **2**, 1170–1172.
200 C. Thiffault, M. Bélanger and M. Pouliot, *Can. Med. Assoc. J.*, 1970, **103**, 165–166.
201 N. Dey, G. Kumar, A. S. Vickram, M. Mohan, R. R. Singhania, A. K. Patel, C. D. Dong, K. Anbarasu, S. Thanigaivel and V. K. Ponnusamy, *Bioresour. Technol.*, 2022, **344**, 126171.
202 S. Sharma and A. Kumar, *Lignin*, Springer, 2020.
203 I. A. Gilca, V. I. Popa and C. Crestini, *Ultrason. Sonochem.*, 2015, **23**, 369–375.
204 H. M. Caicedo, L. A. Dempere and W. Vermerris, *Nanotechnology*, 2012, **23**, 105605.
205 A. A. Myint, H. Lee, B. Seo, W.-S. Son, J. Yoon, T. J. Yoon, H. J. Park, J. Yu, J. Yoon and Y.-W. Lee, *Green Chem.*, 2015, **18**, 2129–2146.
206 S. R. Yearla and K. Padmasree, *J. Exp. Nanosci.*, 2016, **11**, 289–302.

207 O. ur Rahman, S. Shi, J. Ding, D. Wang, S. Ahmad and H. Yu, *New J. Chem.*, 2018, **42**, 3415–3425.
208 W. Yang, J. Owczarek, E. Fortunati, M. Kozanecki, A. Mazzaglia, G. M. Balestra, J. M. Kenny, L. Torre and D. Puglia, *Ind. Crops Prod.*, 2016, **94**, 800–811.
209 W. Yang, E. Fortunati, F. Bertoglio, J. Owczarek, G. Bruni, M. Kozanecki, J. Kenny, L. Torre, L. Visai and D. Puglia, *Carbohydrate Polym.*, 2018, **181**, 275–284.
210 M. Yang, W. Zhao, S. Singh, B. Simmons and G. Cheng, *Nanoscale Adv.*, 2019, **1**, 299–304.
211 A. P. Richter, B. Bharti, H. B. Armstrong, J. S. Brown, D. Plemmons, V. N. Paunov, S. D. Stoyanov and O. D. Velev, *Langmuir*, 2016, **32**, 6468–6477.
212 V. I. Popa, A.-M. Capraru, S. Grama and T. Malutan, *Cellul. Chem. Technol.*, 2011, **45**, 221.
213 Y. Qian, Y. Deng, X. Qiu, H. Li and D. Yang, *J. Green Chem.*, 2014, **16**, 2156–2163.
214 Y. Qian, Q. Zhang, X. Qiu and S. Zhu, *J. Green Chem.*, 2014, **16**, 4963–4968.
215 S. Zhiqiang, S. Wenfeng, Z. Jun, Z. Baoyou, Z. Bin, Y. Lei, Z. Baishi, Z. Xiuhua, Z. Jian and H. Luming, *Chinese Pat.*, 102002165, 2011.
216 L. Zhiming, L. Guochao and W. Haiying, *Chinese Pat.*, CN 103145999, 2013.
217 C. Jiang, H. He, H. Jiang, L. Ma and D. Jia, *eXPRESS Polym. Lett.*, 2013, **7**, 480–493.