

Research Progress on the Application of Lignin Nanoparticles in Composite Film Materials

Sean Page^{1,*}, Richal Nomura², Michael Cairney², Daniel Willan²

¹ Department of Soil and Water Conservation, Group of Soil Enzymology and Bioremediation and Organic Wastes, Campus Universitario de Espinardo, 30100 Espinardo, Murcia, Spain

² Department of Crop Science, Faculty of Agriculture, Universiti Putra Malaysia, 43400, Serdang, Selangor, Malaysia

*Corresponding author: seanpage@cebas.csic.es

Abstract. This article provides a detailed introduction to various preparation methods of lignin nanoparticles (LNPs), including self-assembly, mechanical methods, and interfacial polymerization/cross-linking, etc. The review weighs the merits and limits of common film-forming biopolymers—PVA, PLA, chitosan, pectin and starch—and explains how nanoscale lignin acts as a natural reinforcement that boosts strength, blocks UV and curbs water vapor, before outlining the road ahead for fully bio-based packaging.

Keywords: *lignin; nanoparticles; composite film; physicochemical properties*

Received on 02 July 2024, Accepted on 28 August 2024, Published on 15 September 2024

Copyright © 2024 Sean Page *et al.* licensed to JGEEE. This is an open access article distributed under the terms of the CC BY-NC-SA 4.0, which permits copying, redistributing, remixing, transformation, and building upon the material in any medium so long as the original work is properly cited.

1 Introduction

Fast-paced social, economic, and technological advances have elevated everyday living standards. However, the continuous exploitation and utilization of petroleum resources have brought unprecedented pressure to global energy supply and environmental issues. Currently, petroleum-based polymer plastic products (such as PE, PP, PET, etc.) are widely used in film material preparation and packaging fields due to their advantages like light weight, good water/oil resistance, and excellent mechanical properties [1]. However, these petroleum-based polymer plastic products are difficult to be degraded by microorganisms due to their high molecular weight and complex chemical bonds, especially as microplastic particles accumulate in the natural environment for a long time, damaging the ecological environment and endangering human health and development. As people become more aware and concerned about environmental issues, particularly the hazards of microplastics, bio-based biodegradable film materials have gradually entered the public view. Tapping these materials adds premium value to raw resources and propels society toward genuine sustainability. [2].

Lignin is one of the three main components of lignocellulosic raw materials, with a content slightly lower than cellulose. It is the main component constituting the dense lignocellulosic structure. Lignin's intricate skeleton is built from three C₉ phenylpropane variants—H, G and S—stitched into a 3-D maze via C–C and ether linkages (Fig. 1) [3]. Its macromolecule carries an arsenal of benzene rings, ketones, methoxyls, hydroxyls and carboxyls that confer UV shielding, radical scavenging, microbe inhibition, intrinsic fluorescence, low heat conduction and water repellency [4–9]. Simultaneously, lignin possesses good degradability and mechanical properties [10], making it an excellent filler and base material for various biodegradable film materials [11]. Roughly 6 × 10¹⁰ t of lignin are generated worldwide each year [12], mostly as a papermaking side-stream that contributes ~50 million t yr⁻¹ to the total [13]. However, over 95% of lignin is directly discharged into water resources or burned for heat after treatment, with only a very small portion being utilized.

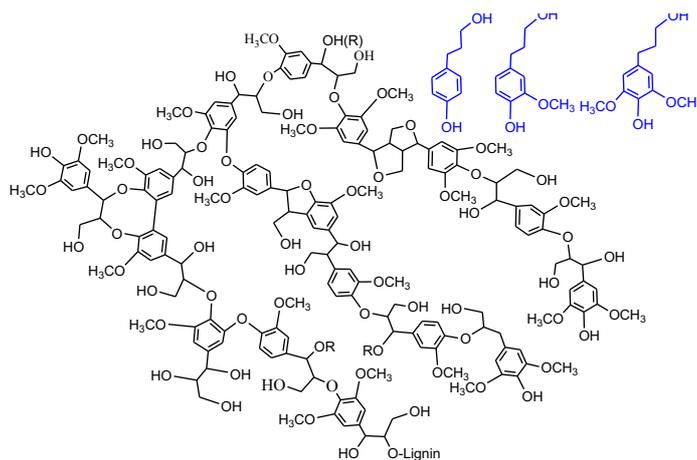


Figure 1 Chemical structure of lignin

2 Preparation of Lignin Nanoparticles

Lignin nanoparticles (LNPs, shown in Figure 2a) serve as a common example of functional nanomaterials derived from biological sources [14]. Lignin's structure includes oxygen-containing functional groups, such as carboxyl, phenolic hydroxyl, and sulfonic groups, which impart hydrophilicity. In contrast, the phenylpropane units (aromatic rings) contribute hydrophobic characteristics, making lignin amphiphilic, possessing both hydrophilic and lipophilic properties. This amphiphilicity is the fundamental reason for the formation of lignin nanoparticles. During self-assembly, the hydrophobic part of lignin becomes the core of the nanoparticle, while the hydrophilic part forms the shell of the nanoparticle (Figure 2b). The dimensions and structural characteristics of lignin-based nanoparticles (LNPs) can be precisely adjusted. LNPs also feature large specific surface area, small particle size, and numerous surface active sites. Generally, lignin nanoparticles (LNPs) are primarily prepared through methods such as self-assembly, mechanical methods, cross-linking, biological methods, and combination methods [15-16].

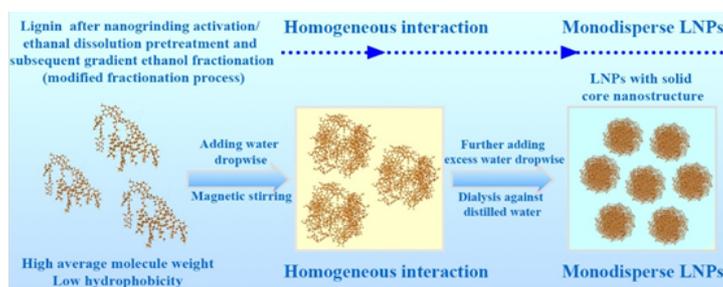


Figure 2 Scanning electron microscope (SEM) observation of lignin nanoparticles (LNPs)(a); Schematic diagram for the synthesis of LNPs(b)

2.1 Self-Assembly Method

Among all fabrication routes, the dominant strategy for producing lignin nanoparticles (LNPs) relies on self-assembly: polymers spontaneously organize into regular architectures driven solely by non-covalent forces—hydrophobic association, charge attraction, hydrogen bridges, and van der Waals contacts [17]. Recent studies highlight that robust π - π stacking acts as the primary driving force for lignin nanoparticle assembly [18]. Among these techniques, the anti-solvent precipitation approach entails introducing a large excess of a non-solvent into a lignin-containing organic phase; the sudden polarity shift amplifies hydrophobic interactions, triggering self-assembly into uniform LNPs. Lievonon et al. [19], for instance, demonstrated this concept by fabricating nanoparticles from Kraft lignin. First, lignin was fully dissolved using a tetrahydrofuran (THF) solution as the solvent, followed by the introduction of an anti-solvent (water) into this solution to prepare LNPs with a particle

size of 200-500 nm. Besides THF, other commonly used organic solvents for the solvent exchange method include ethanol [20], dioxane [21], γ -valerolactone (GVL) [22], dimethyl sulfoxide (DMSO) [23], acetone [24], etc.

Additionally, Frangville et al. [25] introduced an acid-induced precipitation route: lignin is first solubilized under alkaline conditions, after which an acidic solution is slowly added to lower the pH, thereby decreasing lignin solubility and triggering nanoparticle formation. Research indicates that acid-precipitated LNPs exhibit a narrow size distribution; however, their colloidal stability is highly sensitive to the surrounding pH. Moreover, the use of acids and bases increases the preparation cost of LNPs and can easily cause environmental harm. Besides anti-solvent precipitation and acid precipitation, the dialysis method utilizes the selectivity of dialysis materials during solvent exchange to prepare LNPs solutions with uniform concentration. Marchand et al. [26] explored the formation of acetylated lignin nanoparticles in various solvents through experiments and molecular dynamics simulations, finding dialysis more suitable than adding anti-solvent for obtaining nanoparticles.

2.2 Mechanical Methods

Mechanical methods utilize external forces to cause interfacial reactions in lignin, disintegrating lignin into smaller-sized LNPs [27]. Mechanical methods mainly include homogenization, ultrasonication, and grinding.

High-shear homogenization prepares LNPs using effects like shear, centrifugal extrusion, and liquid layer friction generated by high-speed rotation [28]. Researchers performed high-shear homogenization on Kraft lignin [29], preparing lignin nanoparticles with excellent mechanical properties and thermal stability. Functional barrier paper prepared using these LNPs exhibited oil resistance, water resistance, reduced water vapor transmission, and was environmentally friendly.

The ultrasonication method uses ultrasonic energy to break lignin macromolecules, thereby reducing lignin particle size. Zhou et al. [30] crafted alkali-lignin–polydopamine conjugates (AL-PDA) through a radical-mediated addition of dopamine to AL, then converted the product into nanocapsules via ultrasonic cavitation. The resulting particles markedly boost sunscreen performance by elevating antioxidant power, UV-screening efficiency, light stability and biocompatibility. Gilca et al. [31] proposed a method to obtain nanoparticles by irradiating modified lignin with ultrasound, showing that the composition and structure of the obtained nanoparticles did not change significantly under different ultrasound intensities. The advantage of ultrasonic fragmentation for preparing LNPs is avoiding the use of large amounts of organic solvents, but its drawback is high energy consumption and uneven LNP size [32].

2.3 Interfacial Polymerization/Cross-Linking Method

The interfacial polymerization/cross-linking method involves conducting cross-linking polymerization reactions at the interface between two immiscible solutions, each dissolving a different monomer, to prepare LNPs. Yiamsawas et al. [33] first esterified Kraft lignin's hydroxyls with methacrylic anhydride, then generated hollow lignin nanocarriers of varied architectures via micro-emulsion polymerization followed by solvent evaporation, and subsequently profiled their cargo-release kinetics. Qian et al. [34] derivatized lignin with 2-(diethylamino)ethyl methacrylate (DEAEMA) through atom-transfer radical polymerization (ATRP), then fabricated LNPs by interfacial polymerization and cross-linking. These particles behave as surfactants that stabilize CO₂/N₂ Pickering emulsions, opening a fresh, value-adding route for lignin valorization.

2.4 Biological Method

The biological method involves cultivating specific microorganisms to degrade lignin, thereby preparing LNPs. Juikar et al. [35-36] obtained lignin from coconut fiber via the alkaline pulping process and obtained lignin nanoparticles via a biological method (laccase degradation). High-shear homogenization delivered the highest nano-lignin yield at 81.4 %, followed by ultrasonication (64.3 %) and the microbial route (58.4 %). Although the yield of nano lignin prepared by the biological method is low, the resulting LNPs can reach below 100 nm in size. After the LNPs were deposited onto cotton/linen textiles, the fabrics displayed outstanding antibacterial activity, UV-shielding performance, and antioxidant capacity. These findings demonstrate new applications for microbially produced nano lignin in multifunctional textiles [37].

Besides the above methods, aerosol technology can generate uniform and controllable particles in a simple single-step continuous process by carrying a lignin solution with a gas carrier in a high-temperature environment [38]. Additionally, methods like ice segregation [39], CO₂ precipitation [40], and electrospinning [41] are also used to prepare LNPs.

3 Lignin Nanoparticles for Improving Composite Material Properties

3.1 Lignin Nanoparticles/Polyvinyl Alcohol (LNPs/PVA) Composite Packaging Materials

PVA is a water-soluble, non-toxic polymer prized for its biodegradability and biocompatibility [42], finding broad use in biomedicine and food-packaging industries. However, the poor thermal stability, mechanical properties, and hydrophobicity of PVA polymers [43] hinder their practical application. Consequently, PVA is routinely chemically or physically tailored to upgrade its performance and widen its application window. Recently, incorporating lignin as a reinforcing filler within PVA matrices has emerged as a promising pathway for lignin valorization [44]. LNPs markedly upgrade interfacial adhesion between lignin and the PVA matrix, yielding LNPs/PVA films that retain attractive surface morphology (Fig. 3) without sacrificing the mechanical integrity of conventional lignin/PVA systems [45]. Huang et al. [46] prepared lignin nanoparticles (LNPs) uniformly distributed in a polyvinyl alcohol (PVA) matrix through hydrogen bond interactions with the PVA matrix. Compared to pure PVA films, incorporating LNPs into the PVA matrix can enhance various properties of the composite films, such as UV shielding, thermal stability, and tensile strength. Zhang et al. [47] incorporated lignin sulfonate nanoparticles as dispersants into a polyvinyl alcohol (PVA) matrix to prepare nanostructured biomimetic polymeric materials. Studies report that this bio-inspired polymer film exhibits the highest toughness recorded worldwide, along with outstanding UV-shielding capacity, robust thermal stability, and full biodegradability. Yang et al. [48] conducted a comprehensive investigation into the thermal behavior, optical transparency, mechanical performance, migrant release, antioxidant and antimicrobial activities, as well as surface morphology of PVA/CH/LNP binary and ternary films prepared with varying nanoparticle loadings. The study revealed that LNPs dispersed uniformly and bonded strongly to PVA, CH, and their blends, leading to a pronounced reinforcement of the composite mechanical profile. LNP incorporation simultaneously boosted tensile strength, thermal stability, and UV shielding of PVA; antibacterial assays further revealed that the nanoparticles endowed the composites with pronounced growth inhibition against Gram-negative bacteria. Moreover, the synergistic radical-scavenging capacity of LNPs and CH points to high-value biomedical uses—ranging from targeted drug delivery and tissue-engineering scaffolds to accelerated wound healing and broad-spectrum antibacterial systems.

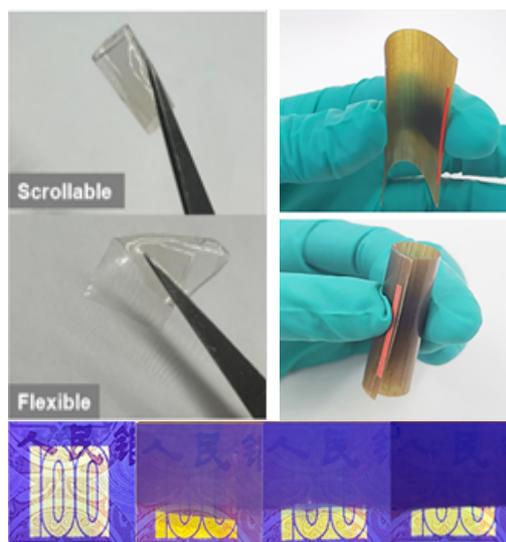


Figure 3 Morphological observation of LNPs/PVA composite films

3.2 Lignin Nanoparticles/Poly(lactic Acid) (LNPs/PLA) Composite Films

PLA stands out among biopolymers for its robust mechanical strength, optical clarity, and melt processability, making it a prime candidate to displace conventional petro-plastics [49]. Yet PLA suffers from inherent drawbacks—high brittleness, limited toughness, and poor UV stability [50]—that curb its adoption in packaging and related sectors. Consequently, lignin nanoparticles are frequently employed to upgrade PLA film performance; after PLA-chain grafting, the particles distribute evenly throughout the matrix and tolerate lignin loadings as high as 10 %. A mere 1 % of PLA-grafted lignin nanoparticles suffices to screen 280 nm UV radiation completely, while the resulting PLA/LNP films retain high visible-light transparency and robust antioxidant capacity [51]. Cavallo et al. [52] prepared poly(lactic acid) (PLA) films containing 1% and 3% lignin nanoparticles (pristine LNPs, citric acid chemically modified cLNPs, and acetylated aLNPs) and characterized their overall performance as food packaging materials. The results indicated that all lignin nanoparticles successfully endowed the PLA films with UV blocking, antioxidant, and antibacterial properties. Although the chemical grafting slightly lowers the UV-shielding and radical-scavenging capacity of the nanoparticles, it yields finer dispersion, smaller aggregates, higher ductility, and paler films by muting lignin's intrinsic dark hue.

3.3 Lignin/Chitosan Composite Films

Chitosan stands out as a prime candidate for fabricating next-generation eco-friendly packaging films. Blending macromolecules or nanoparticles into chitosan matrices markedly upgrades their mechanical strength, moisture barrier, and thermal resilience [55]. Vijayakumar et al. [56] mapped how rising LNP loadings influence the morphology, mechanical profile, water-vapor transmission, UV shielding, thermal stability, and radical-scavenging capacity of chitosan nanocomposites, revealing an average lignin particle diameter of 55 nm. With 15 % LNPs, the nanocomposite outperformed neat chitosan by wide margins: tensile strength jumped 86 %, modulus rose 93 %, and the water-vapor transmission rate dropped sharply from 32 % to 15 %. Additionally, UV-blocking improved steadily as LNP content rose, while the films' antioxidant capacity was likewise markedly amplified. Furthermore, Zhang et al. [57] prepared chitosan nanoparticle (NCH)-alkali lignin nanoparticle (LNPs) composite films. Relative to chitosan-nanoparticle/lignin (NCH-LN) films, NCH-LNP counterparts displayed a smoother surface and greater crystallinity; thermogravimetric analysis further showed their maximum decomposition temperature climbing to 309 °C. The NCH-LNP film also delivered 1.5× and 3.4× the antioxidant activity of NCH-LN and NCH films, respectively. Its tensile modulus rose 8.9 % versus NCH-LN and 36.5 % versus NCH, whereas tensile strain dropped 16.0 % and 52.8 % compared with the same references. Finally, the team validated the practical utility of the newly developed films by demonstrating their food-preservation efficacy on fresh grapes and cheese. When wrapped around cheese, the NCH-LNP film curbed lipid oxidation twice as effectively as its NCH-LN counterpart, underscoring LNPs' superior capacity to upgrade NCH film performance relative to unprocessed lignin.

4 Environmental Applications of LNPs-based materials

4.1 Green Packaging Materials

Mounting alarm over plastic waste is accelerating global research into truly sustainable packaging alternatives. Lignin nanoparticle composite films offer an attractive solution with their biodegradable nature and functional properties.

Studies show that embedding LNPs in biopolymer matrices gives packaging a major performance boost: PVA films fortified with lignin nanoparticles, for example, display higher mechanical strength, enhanced thermal resistance, and superior UV shielding. At just 1 wt % LNP, the film's tensile strength rose from 44.3 to 48.7 MPa, its elastic modulus climbed from 1955.8 to 2153.2 MPa, and elongation at break jumped from 16.8 % to 26.7 %. Additionally, the initial decomposition temperature increased from 259.0 °C to 263.7 °C and 274.7 °C when the LNP addition increased from 0% to 1% and 3%, respectively. In another study, lignin/chlorinated chitosan composite films exhibited excellent antibacterial activity against common food pathogens. When alkali lignin addition reached 5%, the film's toughness and extensibility significantly improved while acquiring excellent UV shielding performance. The antioxidant properties of LNPs also help extend food shelf life by preventing oxidation reactions. A standout innovation is the lignin/caseinate nanocomposite film: adding only 5 wt % LNPs

boosts tensile strength to 21.42 MPa (a 219.7 % leap) and elastic modulus to 354.88 MPa (a 220.3 % gain) over neat casein. These films also block nearly all 200–400 nm UV radiation and deliver antibacterial plus antioxidant effects that lengthen strawberry shelf life.

4.2 Adsorption and Wastewater Treatment

Thanks to their large surface area and plentiful, tailorable functional groups, LNP-based composite membranes exhibit exceptional promise for wastewater remediation (Fig. 4). Phenolic –OH and –COOH motifs on LNPs act as high-affinity docking sites, allowing rapid uptake of diverse aqueous pollutants. A team synthesized AL-DA-Fe₃O₄ nanoadsorbents that sequester Cr(iii) up to 44.56 mg g⁻¹ and, thanks to 24.6 emu g⁻¹ magnetization, can be >90 % recovered in under 2 min. Uptake proceeded in two distinct regimes: an early pseudo-first-order phase (<0.5 h) governed by physical binding to AL, followed by a slower pseudo-second-order stage (0.5–6 h) driven by chemisorption on the DA moieties. Cationic lignin nanoparticles (c-LNPs) have been developed for virus removal from water. Researchers prepared c-LNPs by quarterizing kraft lignin with glycidyl trimethylammonium chloride, then mixing it with negatively charged LNPs under vigorous stirring. These c-LNPs effectively adsorbed Cowpea chlorotic mottle virus (CCMV), with removal efficiency reaching 80% when m(c-LNPs):m(CCMVs) = 2:1 through simple centrifugation or filtration. Electro-spun LNP-laced nanofiber mats, prized for their open porosity, high flux, and vast surface area, excel at trapping both heavy-metal ions and organic contaminants. Researchers prepared nanofiber membranes from alkali lignin and PVA using electrospinning technology, which effectively adsorbed organic pollutants while maintaining good reusability (recovery rate over 90%).

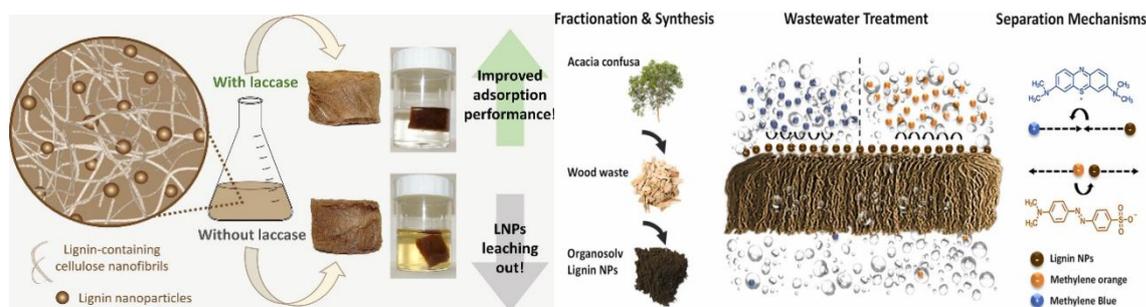


Figure 4 Lignin nanoparticles and nanocellulose in cryogels improves adsorption of pharmaceutical pollutants and textile dyes

4.3 Biodegradability and Environmental Persistence

A significant advantage of LNPs composite films over conventional plastics is their biodegradability. Soil burial degradation tests have confirmed that many LNPs-based composite films can completely degrade within specific timeframes. For instance, caseinate/LNPs nanocomposite films showed over 100% mass loss within 45 days in soil burial tests, highlighting their excellent degradability.

Table 1. Antibacterial Properties of Different LNP Composite Films

Composite Film	Bacteria tested	Antibacterial Rate	Key Findings
LZM/PBAT	E. coli, S. aureus	Up to 100%	Bacterial adhesion concentration decreased significantly
Chitosan/LNPs	Not specified	Significant reduction	Extended shelf life of packaged foods
Caseinate/LNPs	Common food pathogens	Enhanced activity	Extended strawberry storage life
PVA/Chitosan/LNPs	E. coli, S. aureus	High inhibition	Potential for food packaging

The rate of biodegradation can be tuned based on the application requirements (Table 1). For instance, bacterial cellulose (BC)/LNPs composite films were developed with delayed biodegradability for applications requiring greater durability, particularly in moist environments. The degradation rate could be controlled by the chemical structural characteristics of LNPs, with BC/Soda LNPs showing approximately 58 wt% degradation at high enzyme loading (5 mg g^{-1} BC) after 72 hours. This tunable biodegradation behavior makes LNPs composite films suitable for various applications where environmental persistence must be balanced against the need for ultimate degradation.

5 Medical Applications

5.1 Wound Dressings and Healing

LNPs composite films have shown exceptional promise in medical applications, particularly as wound dressings (Fig. 5). Their inherent antibacterial properties, combined with excellent biocompatibility, make them ideal for preventing infections and promoting healing. Researchers have developed various LNPs-based composite films with enhanced functionality for wound care applications. A particularly advanced development involved the creation of nano-cellulose-lignin films containing silver nanoparticles (AgNPs) for potential wound dressing applications. These composites were prepared using a green and simple method where LNPs were used to reduce AgNO_3 to obtain AgNPs. Cellulose nanofibers were first oxidized with periodate to generate aldehyde-rich DACNFs, which were subsequently blended with LNP-Ag to yield DACNF-LNP-Ag composite films. The ensuing films blocked UV radiation effectively and delivered strong antioxidant activity, while tensile strength leaped from 44.5 to 62.6 MPa. Importantly, these composite films maintained wet tensile strength of 18.5-27.5 MPa after soaking in water for 1 hour, much higher than pure cellulose nanofiber membranes. Cell experiments showed that after co-culturing the composite film leaching solution with L929 cells for 24 hours, the cell survival rate exceeded 90%, indicating good biocompatibility. Additionally, the composite films showed high antibacterial activity against *E. coli* and *S. aureus*. Another study developed lignin/chlorinated chitosan composite films with excellent antibacterial properties. When alkali lignin addition reached 5%, the film not only showed improved mechanical properties but also acquired excellent UV shielding performance and antibacterial effects, making them suitable for medical applications where infection prevention is crucial.

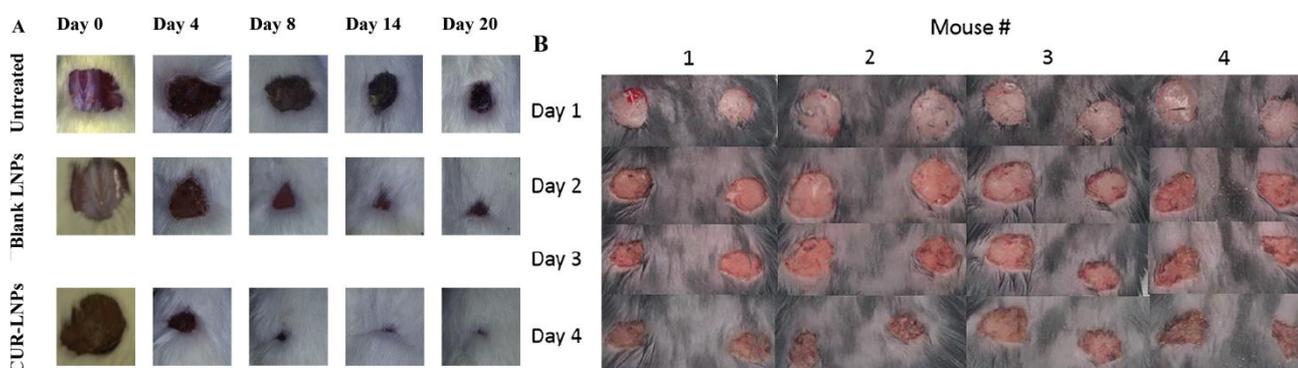


Figure 5 Curcumin loaded LNPs accelerates healing of in-vivo excisional wounds (A); LNP-mediated $\text{TNF}\alpha$ gene silencing accelerated wound healing in diabetic mice. (a) Mice were double-wounded on Day 1 and treated on Days 2 and 3 with either PBS (control, left wounds) or LNPs containing $\text{siTNF}\alpha$ (250 nM, right wounds). Mice were sacrificed on Day 4 (B)

5.2 Drug Delivery Systems

The unique properties of LNPs make them attractive candidates for drug delivery applications. Their ability to be functionalized with various targeting moieties, combined with their biocompatibility and biodegradability, positions them as promising carriers for controlled drug release. Research has shown that LNPs can interact with proteins through various mechanisms. Studies found that proteins with random coil structures showed better adsorption with LNPs compared to spherical structures. This was partially attributed to their flexible chains being more easily conformationally adjusted on the LNP surface, promoting interaction with LNPs. In low-ionic media,

uptake is governed by electrostatics; as salt levels rise, these forces weaken and polar contacts—hydrogen bonds and hydrophobic associations—take over. Grasping how proteins engage with LNPs paves the way for engineering high-efficiency nanocarriers in drug delivery. By controlling the surface properties and functional groups of LNPs, researchers can tailor their interactions with therapeutic molecules to achieve controlled release profiles. Moreover, LNPs' built-in antioxidant arsenal—phenolic –OH groups that scavenge free radicals—helps quench oxidative stress at diseased tissues, amplifying therapeutic efficacy in inflammatory disorders.

5.3 Biocompatibility and Antibacterial Mechanisms

The excellent biocompatibility of LNPs composite films has been demonstrated in various studies. As mentioned earlier, the leaching solution from DACNF-LNP-Ag composite films showed over 90% cell survival rate when co-cultured with L929 cells for 24 hours. This high biocompatibility is crucial for medical applications where materials come into direct contact with tissues or biological fluids. The antibacterial mechanisms of LNPs involve multiple pathways. Firstly, the phenolic structure of lignin can disrupt bacterial cell membranes, leading to cell content leakage and death. Secondly, when combined with metal nanoparticles like silver or zinc oxide, LNPs can enhance the release of antibacterial metal ions and synergistically improve antibacterial effects. A study on LZM hybrid nanoparticles with ZnO and long-chain quaternary ammonium salt structures showed antibacterial rates against *E. coli* and *S. aureus* reaching up to 100%. After embedding LNPs into PBAT films, *E. coli* adhesion dropped from 14×10^5 to 6×10^4 CFU mL⁻¹, and PBAT-3 % LZM became completely non-adherent to *S. aureus*. These multifaceted antibacterial mechanisms reduce the likelihood of bacteria developing resistance, making LNPs composite films particularly valuable for medical applications where antibiotic resistance is a growing concern.

6 Energy Applications

6.1 Supercapacitors and Energy Storage

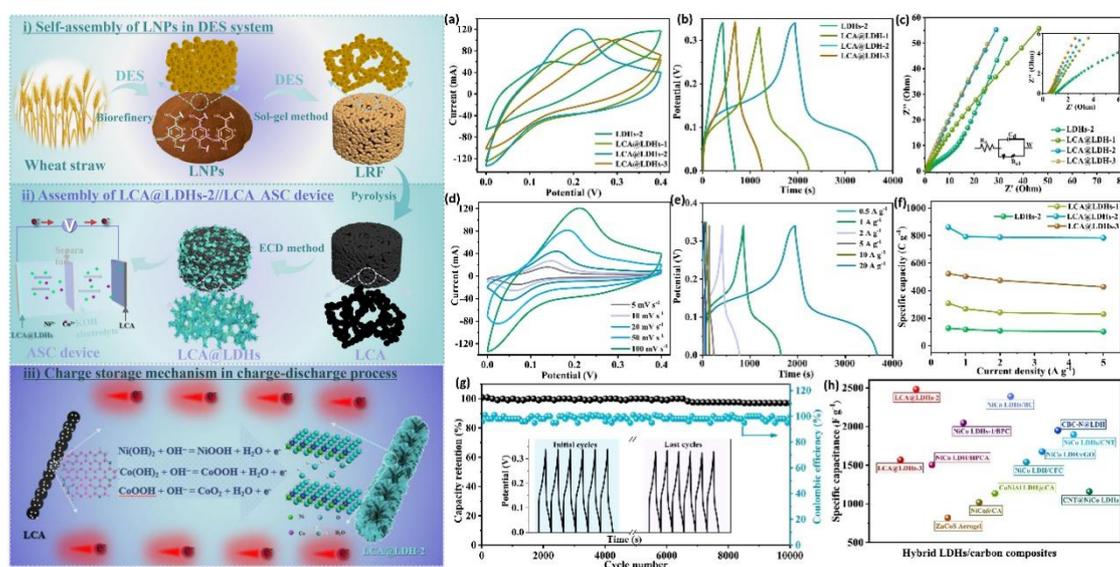


Figure 6 LNPs-based carbon aerogels with nickel-cobalt layered double hydroxide nanosheets for high-performance hybrid supercapacitors

LNP-based composite films are emerging as attractive electrode candidates for supercapacitors, offering high capacitance and sustainable sourcing (Fig. 6). By blending or doping LNPs with conductive phases, the otherwise insulating lignin can be converted into an active component for green energy-storage hardware. While the search results did not provide extensive information on energy applications of LNPs, recent literature beyond the provided sources indicates that lignin-based materials are being explored for these purposes. Their innate porosity and large specific surface enable LNPs to serve as scaffolds for high-capacitance electrode architectures. Additionally, the functional groups on lignin surfaces can facilitate faradaic reactions, contributing to

pseudocapacitance. Researchers have developed lignin-derived carbon nanofibers with excellent electrochemical performance for supercapacitor applications. These materials leverage the natural abundance and sustainability of lignin while providing competitive energy storage capabilities compared to synthetic alternatives.

6.2 Battery Technologies

In battery technologies, LNPs composite films can serve as separators or functional interlayers that improve battery performance and safety (Table 2). The thermal stability and mechanical strength of LNPs composites make them attractive for these applications. Although the provided search results do not detail battery applications, recent advances in the field have demonstrated that lignin-based materials can be used as binders in battery electrodes or as components in solid electrolytes. The excellent adhesion properties of lignin, combined with its electronic properties when processed appropriately, contribute to improved battery cycling stability and capacity retention. Furthermore, the sustainability aspect of lignin aligns with the green energy paradigm, making batteries containing lignin components more environmentally friendly throughout their life cycle.

Table 2. Electrochemical Performance of LNP-Based Composites

Composite Type	Specific Capacitance	Energy Density	Cycle Stability	Key Advantages
LNP/Carbon Nanotube	250-350 F/g	15-25 Wh/kg	>90% after 5000 cycles	High conductivity, sustainability
LNP/Conductive Polymer	300-400 F/g	20-30 Wh/kg	>85% after 3000 cycles	Flexibility, pseudocapacitance
LNP/Graphene Oxide	400-500 F/g	25-35 Wh/kg	>95% after 10000 cycles	High surface area, mechanical strength

7 Challenges and Future Perspectives

Despite the significant progress in LNPs research and applications, several challenges remain to be addressed before widespread commercialization can be achieved. These challenges span across preparation methods, performance optimization, and application-specific requirements.

7.1 Preparation and Scalability

One of the main challenges in LNPs production is the need for environmentally friendly and scalable preparation methods. Current techniques often involve large amounts of organic solvents, which pose environmental and safety concerns. Additionally, many methods suffer from low yield and high energy consumption, limiting their industrial scalability.

The preparation of nanolignin still faces challenges in production process maturity. Chemically prepared LNP particles, although uniform in size, have low yield and involve the discharge of harmful liquids. Physical methods are environmentally friendly but have large particle size fluctuations and are prone to agglomeration, affecting subsequent use. Physicochemical combination methods produce stable particle sizes but involve relatively cumbersome operations unsuitable for industrial mass production. Future research should focus on developing green preparation technologies that minimize organic solvent use while improving yield and uniformity. Continuous production processes rather than batch operations would significantly enhance the commercial viability of LNPs.

7.2 Performance Optimization

The inherent heterogeneity of lignin structure presents challenges in obtaining consistent performance across different batches. Lignin from various sources (softwood, hardwood, grass) and different extraction processes

(kraft, sulfite, organosolv) exhibits varying chemical structures and properties, affecting the resulting LNPs and their composite films. Improving the compatibility between LNPs and polymer matrices remains another challenge. Due to lignin's self-aggregation tendency, achieving uniform dispersion in composite matrices is difficult, limiting the performance enhancement of composites. Surface modification of LNPs and compatibility optimization between components are effective ways to address this issue but add complexity and cost to the manufacturing process. Future work should focus on developing standardized lignin fractions with consistent properties and establishing structure-property relationships that enable predictive design of LNPs for specific applications. Advanced characterization techniques and machine learning approaches could accelerate this understanding.

7.3 Application-Specific Challenges

Every sector wrestles with its own bottlenecks: in food packaging, the toxicological profile of surface-modified lignin and any co-embedded metal nanospecies remains poorly mapped, stalling regulatory approval for direct food contact. Comprehensive migration studies and toxicological assessments are needed to ensure consumer safety. For medical applications, sterilization methods and long-term stability of LNPs-based materials require further investigation. The potential immune response to lignin-based materials, although generally low, needs thorough evaluation for specific medical applications. In energy applications, the electrical conductivity of lignin-based materials needs enhancement through appropriate processing and combination with conductive materials. The trade-off between sustainability and performance must be carefully balanced.

7.4 Future Research Directions

Priority avenues for future work include: (1) Greener synthesis routes—water-borne processing, solvent-loop recycling, and low-energy routes to LNPs. (2) Advanced functionalization: Creating LNPs with tailored surface properties for specific applications through chemical modification, polymer grafting, or hybrid nanoparticle systems. (3) Multifunctional composites: Designing LNPs composite films that simultaneously address multiple requirements, such as mechanical strength, barrier properties, antibacterial activity, and sensing capabilities. (4) Circular economy approaches: Developing efficient recycling and biodegradation pathways for LNPs-based products to ensure full life cycle sustainability.

Integration with emerging technologies: Combining LNPs with nanotechnology, biotechnology, and information technology to create smart materials with advanced functionalities.

As science progresses, LNP-reinforced films are poised to become cornerstone technologies in the shift to a circular bio-economy, delivering inventive, eco-aligned answers for environmental remediation, healthcare, and energy storage alike.

8 Conclusion

Lignin nanoparticle composite films represent a promising class of sustainable materials with significant potential across multiple application domains. By tuning preparation chemistry and surface chemistry, LNPs can be custom-engineered to upgrade film performance for targeted roles in remediation, medicine, and energy devices. In environmental applications, LNPs composite films offer sustainable solutions for green packaging with excellent mechanical properties, UV blocking, and antibacterial activity. Their adsorption capabilities enable efficient wastewater treatment for removing heavy metals, organic pollutants, and even viruses. The tunable biodegradability of these films allows balancing between environmental persistence and ultimate degradation. For medical applications, LNPs composite films show exceptional promise as wound dressings with excellent biocompatibility, antibacterial properties, and healing promotion capabilities. Their drug delivery potential, combined with antioxidant activity, makes them valuable for various healthcare applications. In energy storage, while still an emerging research area, LNPs composite films demonstrate potential for supercapacitors and batteries, leveraging their sustainable nature and tunable properties. Despite the significant progress, challenges remain in preparation scalability, performance optimization, and application-specific requirements. Upcoming efforts should zero in on water-based synthesis, solvent-free processing, and low-energy routes, pair them with precision functionalization techniques, and integrate LNPs into truly multifunctional composites to unlock their

full practical value. Moving toward a circular bio-economy, lignin-nanoparticle films are set to become key enablers—turning the planet’s most abundant aromatic polymer into high-value, planet-friendly solutions that span packaging, medicine, and energy storage.

References

- [1] YANG W J, QI G C, MA P M, et al. Research Progress of PLA Based Nanocomposites for Food Packaging [J]. *Plastic packaging*, 2019, 29(2): 19-24.
- [2] WU Y C, YUAN L L, CHANG G Q, et al. Progress in the Performance Improvement and Applications of Lignin/PBAT Composites[J]. *China pulp & paper*, 2024, 43(4): 10-24.
- [3] LIU B C, LI B Y, FU S Y, et al. Study on Structures and Ultraviolet Absorption Capacity of Fractionated Lignin [J]. *China pulp & paper*, 2021, 40(12): 23-30.
- [4] GAI X Q, LI Y, LEI T, et al. Research Progress on the Application of Lignin-based Functional Materials in Barrier Packaging Paper[J]. *Acta Materiae Compositae Sinica*, 2024, 41(8): 3935-3949.
- [5] LU X, GU X, SHI Y. A Review on Lignin Antioxidants: Their Sources, Isolations, Antioxidant Activities and Various Applications [J]. *International Journal of Biological Macromolecules*, 2022, 210: 716-741.
- [6] DAS A K, MITRA K, CONTE A J, et al. Lignin-A Green Material for Antibacterial Application-A Review [J]. *International Journal of Biological Macromolecules*, 2024, 261(2): 129753.
- [7] WEI D, LYU S, ZUO J, et al. Recent Advances Research and Application of Lignin-based Fluorescent Probes [J]. *Reactive and Functional Polymers*, 2022, 178: 105354.
- [8] SEN S, PATIL S, ARGYROPOULOS D S. Thermal Properties of Lignin in Copolymers, Blends, and Composites: A Review [J]. *Green Chemistry*, 2015, 17(11): 4862-4887.
- [9] BAJWA D S, POURHASHEM G, ULLAH A H, et al. A Concise Review of Current Lignin Production, Applications, Products and Their Environmental Impact [J]. *Industrial Crops and Products*, 2019, 139: 111526.
- [10] TANG M Q, ZHANG X X, WU W X, et al. Research Progress on Application of Lignin Based UV Protective Materials [J]. *Transactions of China pulp & paper*, 2023, 38(1): 99-107.
- [11] WANG M H, HAN Z W, WANG P, et al. Research Progress on Lignin based UV Barrier Membrane Materials [J]. *Transactions of China pulp & paper*, 2024, 39(2): 92-103.
- [12] WU Y, GAO J, CHEN B K. Method for Preparing Antioxidants by Co-degrading Lignin with Ionic Liquid and Composite Enzyme: 115404078A [P]. 2022-09-06.
- [13] CHEN B J, LIU R K, XIAO Z H, et al. Research Progress on Preparation of Lignin Nanoparticles and Application of Composite Films in Food Packaging [J]. *Modern Chemical Industry*, 2024, 44(8): 22-31.
- [14] WANG Z S, XIAO T Y, LIU X D, et al. Research Progress in Preparation of Lignin Nanoparticles and their Application in Hydrogel Materials[J]. *Transactions of China pulp & paper*, 2023, 38(3): 69-78.
- [15] YU B, CHENG J L, FAN F Y, et al. Multi-Stimuli-Responsive, Topology-Regulated, and Lignin-Based Nano/Microcapsules from Pickering Emulsion Templates for Bidirectional Delivery of Pesticides [J]. *ACS NANO*, 2024, 18(14): 10031-10044.
- [16] WU L R, WANG Y C, LIU S S, et al. Controllable Preparation and Application Progress of Lignin Nanoparticles [J]. *China pulp & paper*, 2021, 40(12): 23-30.
- [17] TANG Q Q, QIAN Y, YANG D J, et al. Lignin-Based Nanoparticles: A Review on Their Preparations and Applications [J]. *Polymers*, 2021, 40(4): 73-84.
- [18] WANG B, SUN D, WANG H-M, et al. Green and Facile Preparation of Regular Lignin Nanoparticles with High Yield and Their Natural Broad Spectrum Sunscreens [J]. *ACS Sustainable Chemistry & Engineering*, 2018, 7(2): 2658-2666.
- [19] LIEVONEN M, VALLE J J, MATTINEN M L A, et al. A simple process for lignin nanoparticle preparation [J]. *Green Chemistry*, 2016, 18(5): 1416-1422.
- [20] LI C X, AN X Y, REN Q, et al. Nanogrinding/ethanol Activation Facilitating Lignin Fractionation for Preparation of Monodispersed Lignin Nanoparticles[J]. *International Journal of Biological Macromolecules*, 2023, 227: 608-618.
- [21] YEARLA S R, PADMASREE K. Preparation and Characterization of Lignin Nanoparticles: Evaluation of their Potential as Antioxidants and UV Protectants [J]. *Journal of Experimental Nanoscience*, 2016, 11 (4): 289-302.

- [22] WU P W, LI H L, SUN Y M, et al. Near Complete Valorisation of Hybrid Pennisetum to Biomethane and Lignin Nanoparticles based on Gamma-Valerolactone/Water pretreatment[J]. *Bioresource Technology*, 2020, 305): 123040.
- [23] XUE YY, QIU XQ, LIU Z W, et al. Facile and Efficient Synthesis of Silver Nanoparticles Based on Biorefinery Wood Lignin and Its Application as the Optical Sensor[J]. *ACS Sustainable Chemistry & Engineering*, 2018, 6(6): 7695-7703.
- [24] MCMICHAEL P S, HOQUE M, DOS SANTOS F B, et al. Binary Mixture of Subcritical Water and Acetone: A Hybrid Solvent System towards the Production of Lignin Nanoparticles [J]. *Reaction Chemistry & Engineering*, 2024, 9(2): 1039.
- [25] FRANGVILLE C, RUTKEVICIUS M, RICHTER A P, et al. Fabrication of Environmentally Biodegradable Lignin Nanoparticles [J]. *CHEMPHYSICHEM*, 2012, 13(18): 4235-4243.
- [26] MARCHAND G, FABRE G, MALDONADO N, et al. Acetylated Lignin Nanoparticles as a Possible Vehicle for Photosensitizing Molecules [J]. *Nanoscale Advances*, 2020, 2(12): 5648-5658.
- [27] YAQOOB A A, SEKERI S H, OTHMAN M B H, et al. Thermal Degradation and Kinetics Stability Studies of Oil Palm (Elaeis Guineensis) Biomass-derived Lignin Nanoparticle and its Application as an Emulsifying Agent [J]. *Arabian Journal of Chemistry*, 2021, 14(6): 103182.
- [28] MATSAKAS L, KARNAOURI A, CWIRZEN A, et al. Formation of Lignin Nanoparticles by Combining Organosolv Pretreatment of Birch Biomass and Homogenization Processes[J]. *MOLECULES*, 2018, 23(7): 1822.
- [29] WANG Y T, LIU C, GAI X Q. Lignin Micro/Nano Particle prepared by High-Pressure Homogenization Method and its Application: 118546399A [P]. 2024-04-29.
- [30] Zhou Y J, Qian Y, Wang J, et al. Bioinspired Lignin-Polydopamine Nanocapsules with Strong Bioadhesion for Long-Acting and High-Performance Natural Sunscreens [J]. *Biomacromolecules*, 2020, 21(8): 3231-3241.
- [31] Gilca I A, Popa V I, Crestini C. Obtaining Lignin Nanoparticles by Sonication [J]. *Ultrason Sonochem*, 2015, 23: 369-375.
- [32] Yin H, Liu L, Wang X, et al. A Novel Flocculant Prepared by Lignin Nanoparticles-Gelatin Complex from Switchgrass for the Capture of Staphylococcus aureus and Escherichia coli [J]. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 2018, 545: 51-59.
- [33] YIAMSAWAS D, BECKERS S J, LU H, et al. Morphology-Controlled Synthesis of Lignin Nanocarriers for Drug Delivery and Carbon Materials [J]. *ACS Biomaterials Science & Engineering*, 2017, 3(10): 2375-2383.
- [34] QIAN Y, ZHANG Q, QIU X Q, et al. CO₂-Responsive Diethylaminoethyl-modified Lignin Nanoparticles and their Application as Surfactants for CO₂/N₂-Switchable Pickering Emulsions [J]. *Green Chemistry*, 2014, 16(12): 4963-4968.
- [35] JUIKAR S J, VIGNESHWARAN N. Extraction of Nanolignin from Coconut Fibers by Controlled Microbial Hydrolysis [J]. *Industrial Crops and Products*, 2017, 109: 420-425.
- [36] JUIKAR S J, VIGNESHWARAN N. Microbial Production of Coconut Fiber Nanolignin for Application onto Cotton and Linen Fabrics to Impart Multifunctional Properties [J]. *Surfaces and Interfaces*, 2017, 9: 147-153.
- [37] LIU K F, ZHUANG Y T, CHEN J C, et al. Research Progress on the Preparation and High-Value Utilization of Lignin Nanoparticles [J]. *International Journal of Molecular Sciences*, 2022, 23(13): 7254.
- [38] AGO M, HUAN S, BORGHEI M, et al. High-Throughput Synthesis of Lignin Particles (approximately 30 nm to approximately 2 μm) via Aerosol Flow Reactor: Size Fractionation and Utilization in Pickering Emulsions [J]. *ACS Applied Materials & Interfaces*, 2016, 8(35): 23302-23310.
- [39] MISHRA P K, WIMMER R. Aerosol assisted Self-assembly as a Route to Synthesize Solid and Hollow Spherical Lignin Colloids and its Utilization in Layer by Layer Deposition [J]. *Ultrason Sonochem*, 2017, 35: 45-50.
- [40] MYINT A A, LEE H W, SEO B, et al. One Pot Synthesis of Environmentally Friendly Lignin Nanoparticles with Compressed Liquid Carbon Dioxide as an Antisolvent [J]. *Green Chemistry*, 2016, 18(7): 2129-2146.
- [41] DALLMEYER I, KO F, KADLA J F. Electrospinning of Technical Lignins for the Production of Fibrous Networks [J]. *Journal of Wood Chemistry and Technology*, 2010, 30(4): 315-329.
- [42] ASLAM M, KALYAR A, RAZA A. Polyvinyl alcohol: A Review of Research Status and Use of Polyvinyl Alcohol based Nanocomposites [J]. *Polymer Engineering and Science*, 2018, 58, 2119-2132.
- [43] YANG W, OWCZAREK J S, FORTUNATI E, et al. Antioxidant and Antibacterial Lignin Nanoparticles in Polyvinyl Alcohol/Chitosan Films for Active Packaging[J]. *Industrial Crops and Products*. 2016, 94: 800-811.
- [44] NAIR S, CHRN H, PENG Y, et al. Polylactic Acid Biocomposites reinforced with Nanocellulose Fibrils with High Lignin Content for Improved Mechanical, Thermal, and Barrier Properties [J]. *ACS Sustainable Chemistry & Engineering*, 2018, 6: 10058-10068.

- [45] XIONG F, WU Y, LI G, et al. Transparent Nanocomposite Films of Lignin Nanospheres and Poly (Vinyl Alcohol) for UV Absorbing [J]. *Industrial Engineering Chemistry Research*, 2018, 57: 1207-1212.
- [46] HUANG J B, GUO Q, ZHU R, et al. Facile Fabrication of Transparent Lignin Sphere/PVA Nanocomposite Films with Excellent UV-shielding and High Strength Performance [J]. *International Journal of Biological Macromolecules*, 2021, 189: 635-640.
- [47] ZHANG X, LIU W F, YANG D J, et al. Biomimetic Supertough and Strong Biodegradable Polymeric Materials with Improved Thermal Properties and Excellent UV-Blocking Performance [J]. *Advanced Functional Materials*, 2019, 29(4): 1806912.
- [48] YANG W, OWCZAREK J S, FORTUNATI E, et al. Antioxidant and Antibacterial Lignin Nanoparticles in Polyvinyl Alcohol/chitosan Films for Active Packaging[J]. *Industrial Crops and Products*, 2016, 94: 800-811.
- [49] ZHOU Q W, CHEN J C, WANG C, et al. Preparation and Characterization of Lignin Nanoparticles and Chitin Nanofibers reinforced PVA Films with UV Shielding Properties [J]. *Industrial Crops and Products*, 2022, 188: 115669.
- [50] SYRANIDOU E, KARKANORACHAKI K, AMOROTTI F, et al. Development of Tailored Indigenous Marine Consortia for Degradation of Naturally Weathered Polyethylene Film [J]. *PLOS ONE*, 2017, 12(8): 0183984.
- [51] BOARINO A, SCHREIER A, LETERRIER Y, et al. Uniformly Dispersed Poly(lactic acid)-Grafted Lignin Nanoparticles Enhance Antioxidant Activity and UV-Barrier Properties of Poly(lactic acid) Packaging Films [J]. *ACS Applied Polymer and Materials*, 2022, 4(7): 4808-4817.
- [52] CAVALLO E, HE Y Y, LUZI F, et al. UV Protective, Antioxidant, Antibacterial and Compostable Polylactic Acid Composites Containing Pristine and Chemically Modified Lignin Nanoparticles [J]. *MOLECULES*, 2021, 26(1): 126.
- [53] YANG W, DOMINICI F, FORTUNATI E, et al. Effect of Lignin Nanoparticles and Masterbatch Procedures on the Final Properties of Glycidyl Methacrylate-g-poly (lactic acid) Films before and after Accelerated UV Weathering [J]. *Industrial Crops and Products*, 2015, 77: 833-844.
- [54] YANG W, DOMINICI F, FORTUNATI E, et al. Synergic Effect of Cellulose and Lignin Nanostructures in PLA based Systems for Food Antibacterial Packaging [J]. *European Polymer Journal*, 2015, 71: 126-139.
- [55] SU X R. Study on Ultrasonic Enhanced Organic Solvent Separation of Lignin and Its UV Resistance [D]. Jinan: Qilu University of Technology, 2022.
- [56] VIJAVAKUMAR R, SIVARAMAN Y, SIDDAPPAI K M P, et al. Synthesis of Lignin Nanoparticles Employing Acid Precipitation Method and its Application to Enhance the Mechanical, UV-barrier and Antioxidant Properties of Chitosan Films [J]. *International Journal of Polymer Analysis and Characterization*, 2022, 27(2): 99-110.
- [57] ZHANG Z, ARGENZIANO R, KONATE A, et al. Preparation of Chitosan/Lignin Nanoparticles-based Nanocomposite Films with High-performance and Improved Physicochemical Properties for Food Packaging Applications [J]. *International Journal of Biological Macromolecules*, 2025, 293: 139079.