

## Green Modification of Lignin Nano Particles (LNPs) For Environmental Remediation: A Review



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### ABSTRACT

The increasing global concern over environmental pollution caused by industrial effluents has driven the search for sustainable and eco-friendly materials for pollutant removal. Lignin, a naturally abundant biopolymer, has emerged as a promising candidate for the development of adsorbents due to its renewable nature, low cost, and functional groups that facilitate pollutant binding. Recent advancements in the green modification of lignin nanoparticles (LNPs) have significantly enhanced their adsorption capacity for various pollutants, including heavy metals, dyes, and organic compounds. Green modification of LNPs enhances adsorption efficiency by introducing functional groups and improving compatibility with various pollutants. Therefore, this review provides a comprehensive overview of the green modification of lignin nanoparticles and their potential for industrial effluent treatment, highlighting their application in pollutant adsorption, and the mechanisms involved. The environmental benefits, challenges and future perspectives of using green modified LNPs for industrial effluent treatment are also discussed.

**Keywords:** Green Modification, Lignin Nano Particles (LNPs), and Environmental Remediation.

### 1.0 Introduction

Lignocellulose biomass is a strategic natural renewable resource generated by different sectors such as agriculture, forestry, and industry. Lignocellulose is derived from plant and wood, which mainly consists of cellulose, hemicellulose, and lignin. Lignin, a complex aromatic polymer, is a major component of lignocellulosic biomass, a renewable and abundant resource [1]. Lignin is the second most abundant natural biopolymer after cellulose [2] [3]. It is primarily obtained as a by-product from the pulp and paper industry [4]. The conversion of lignin into value-added products has garnered significant attention due to its potential to replace fossil-based materials. Among these products, lignin nanoparticles (LNPs) have emerged as promising candidates for

various applications owing to their unique properties such as biodegradability, structural diversity, and functional properties [2] [5]. However, their inherent properties such as macro structure and insolubility in many solvents often limit their direct application in certain areas [6]. Consequently, modification of LNPs has become a crucial strategy to tailor their physicochemical characteristics and expand their applicability.

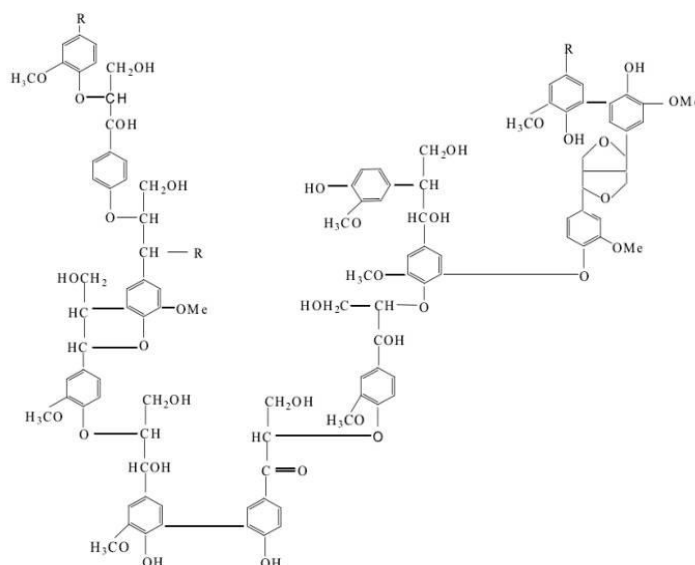


Fig 1. Structural depiction of Lignin

**Citation:** Ifiok O, Ekwere, Aniekan E. Akpakpan, Iniobong S, Enengedi, Ito E. Udo, Nsima A. Akpan, Ukponobong E. Antia and Itohowo G. Asuquo (2026). Green Modification of Lignin Nano Particles (LNPs) For Environmental Remediation: A Review.

*Journal of e-Science Letters.*

DOI: <https://doi.org/10.51470/eSL.2026.7.1.80>

Received: 04 November 2025

Revised: 06 December 2025

Accepted: 03 January 2025

Available: February 05 2026

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## 2.0 Lignin Nanoparticles (LNPs)

Lignin nanoparticles (LNPs) are an emerging class of biopolymeric nanoparticles that are derived from lignin, a complex aromatic polymer found abundantly in the cell walls of plants. The nanoscale conversion of lignin has improved the lignin valorisation process efficiency [7]. Compared to lignin, lignin nanoparticles (LNPs) possess a higher surface-to-volume ratio and better dispersibility, leading to improved mechanical performance and thermal stability [8 -10]. Furthermore, LNPs also showed greater antioxidant activity [11-12] and good UV shielding properties [13 - 14]. The growing interest in LNPs stems from their renewable nature, biodegradability, and ability to replace petroleum-based nanomaterials. With their unique chemical structure and properties, Lignin nanoparticles have found applications in various fields such as biomedicine, environmental science, and material engineering.

### 2.1 Synthesis

LNPs can be synthesized using various methods, which are broadly classified into two categories:

- **Top-down approaches:** These methods involve breaking down bulk lignin into smaller particles using techniques such as milling, ultrasonication, and solvent shifting.
- **Bottom-up approaches:** These methods involve building up LNPs from lignin molecules through self-assembly or precipitation techniques, such as antisolvent precipitation and nanoprecipitation.

The synthesis of Lignin nanoparticles typically involves various methods that enable the controlled preparation of nanoscale lignin materials. The main synthesis techniques include:

#### 1. Solvent Evaporation/Solvent Exchange Method

This method involves dissolving lignin in an appropriate solvent and subsequently evaporating the solvent under controlled conditions, leading to the formation of nanoparticles. Solvent evaporation is a simple and cost-effective approach that provides good control over particle size and distribution. The use of ethanol as a solvent for lignin leads to the formation of uniform-sized nanoparticles with low polydispersity.

Solvent Exchange method proposed by Lievonen *et al.* [15] involves the dissolution of lignin in THF and the placement of the obtained lignin/THF solutions in a dialysis bag, which was subsequently immersed in excess water. The lignin nanoparticles were formed after the dialysis process continued for at least 24 h.

#### 2. Antisolvent Precipitation

In this technique, lignin is dissolved in a solvent, and an antisolvent is gradually added to the solution. This change in solvent conditions promotes lignin precipitation, resulting in nanoparticle formation. The precipitation of lignin in water after dissolving it in dimethyl sulfoxide (DMSO) has been widely used to produce nanoparticles with high stability.

#### 3. High-Energy Ball Milling

This mechanical method involves grinding lignin in a ball mill to break down its particles into nanoscale dimensions. The method is environmentally friendly as it avoids the use of solvents and is scalable for industrial purposes. Mechanical treatments, such as dry and wet milling techniques, are widely used to reduce particle size down to nanometer scale [17]. According to Malcolmson & Embleton [18] the milling process has the disadvantage of non-uniformity in particle size and broad particle size distributions but is still a simple process for

nanoparticle production. These nanoparticles are very effective in adsorption of pollutant compounds due to their large surface area.

#### 4. Coacervation/Phase Separation

Coacervation involves inducing a phase separation in a lignin solution by altering temperature or pH, leading to the formation of nanoparticles. This method is typically used for producing nanoparticles with a well-defined size. Jiang *et al.* (2020) successfully employed the coacervation method to prepare stable lignin nanoparticles with tunable surface properties and sizes.

#### 5. Electrospinning

Electrospinning is a technique used to create nanoparticles or nanofibers by applying a high voltage to a polymer solution. Lignin-based nanoparticles can be synthesized by electrospinning, offering a controlled way to tailor particle morphology and size. Lignin nanoparticles were synthesized by electrospinning lignin solutions, resulting in nanoparticles that exhibit good dispersion and stability (Martín *et al.*, 2021).

#### 6. Ultrasonication

Ultrasonication requires no organic solvents or chemical modification on lignin and requires no post-treatment. During the ultrasonication, lignin particles may face both physical (ultrasound-induced acoustic cavitation, intense local heating and pressure) and chemical effects (generation of hydrogen and hydroxyl radicals, which tackle the functional groups available on a particle) that generate highly monodisperse nanoparticles [19]. Compared to the other methods, the overall preparation time of LNPs via ultrasonication is relatively short, about an hour [20 -21]. However, the economic feasibility of sonication depends on sonication time, frequency, the wattage of the sonicator, amplitude and lignin concentration [21]. Long ultrasonication time, high frequency and amplitude, and low lignin concentration ultimately leads to increased energy consumption.

#### Properties of Lignin Nanoparticles (LNPs)

- **Biocompatibility:** LNPs are derived from lignin, a natural biopolymer, making them generally non-toxic and compatible with biological systems. This property is beneficial for biomedical applications.
- **Biodegradability:** Being organic in nature, LNPs can degrade in the environment, which reduces the risk of pollution associated with synthetic nanoparticles.
- **Antioxidant Activity:** Lignin contains various phenolic compounds that exhibit antioxidant properties, making LNPs useful in food preservation and cosmetic formulations.
- **UV Absorption:** LNPs can absorb ultraviolet light, providing potential applications in UV protection in coatings and sunscreens.
- **Adsorption Properties:** LNPs can absorb various molecules due to their high surface area, which is advantageous for applications in drug delivery and wastewater treatment.
- **Mechanical Strength:** Lignin's inherent strength contributes to the mechanical properties of LNPs, making them suitable for reinforcing materials.

## 2.2 Uses/Application of Lignin and Lignin nanoparticles

Lignin nanoparticles possess a range of properties that make them useful in various fields; which include; Biocompatibility and Biodegradability, Antioxidant and Antimicrobial activity [22], Biomedical Applications and Food Industry [23 -24], Materials Engineering [14, 20] and in soil remediation [25].

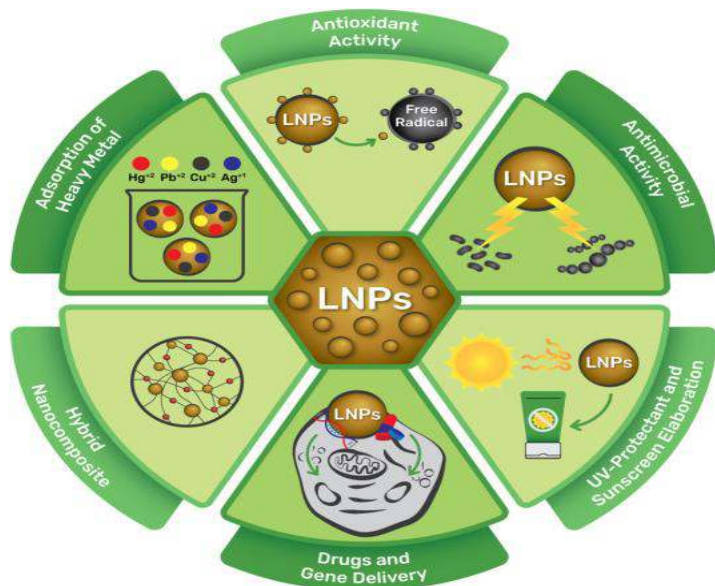


Fig 2: Applications of Lignin nanoparticles (LNPs) (Schneider et al., 2021) [26]

### Environmental Applications

Lignin nanoparticles can be employed in environmental remediation processes, particularly for wastewater treatment. Their surface can be functionalized to adsorb heavy metals and organic pollutants from contaminated water. Numerous studies have utilized Lignin nanoparticles to adsorb pollutants such as dyes, heavy metals, and pesticides, offering an eco-friendly solution for environmental cleanup [27-28]. The use of lignin, a renewable resource, to produce nanoparticles ensures that lignin-based products have a lower environmental impact compared to traditional plastic nanoparticles derived from fossil fuels.

Lignin nanoparticles are being explored for various industrial, biomedical, and environmental applications due to their unique properties. Below are some notable applications:

### 3.0 Modification of Lignin Nanoparticles for the adsorption of pollutants from Industrial Effluents

Lignin nanoparticles, derived from the abundant lignin polymer in plant biomass, are emerging as sustainable adsorbents due to their high surface area, functional group availability, and environmental friendliness. However, their inherent properties often limit their direct application in certain areas. Consequently, modification of LNPs has become a crucial strategy to tailor their physicochemical characteristics and expand their applicability. Some limitations of LNPs, which necessitate their modification, include Limited dispersibility, low surface reactivity. Several chemical and physical modification methods have been developed to address these limitations:

#### 3.1. Chemical Modification:

- **Esterification:** This involves the chemical reaction of LNPs with carboxylic acids or anhydrides. This introduces ester groups, altering hydrophobicity and improving compatibility with hydrophobic polymers. (e.g., acetylation, butyrylation).

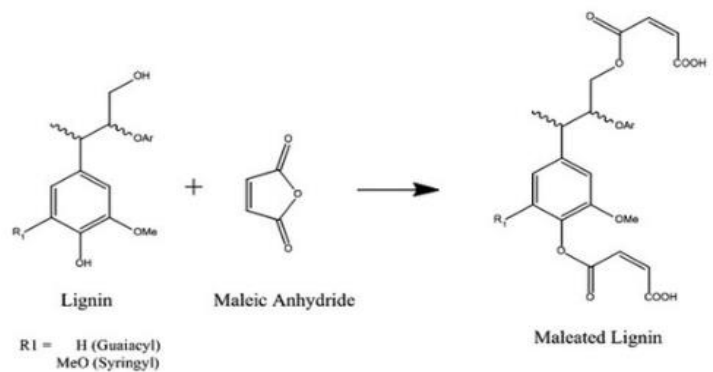


Fig 3: Esterification reaction of Lignin with maleic acid anhydride [29]

- **Etherification:** Introducing ether linkages through reactions with alkyl halides or epoxides can enhance hydrophobicity and thermal stability. (e.g., alkylation, hydroxyalkylation)
- **Oxidation:** Oxidizing LNPs using oxidizing agents like hydrogen peroxide or potassium permanganate introduces carbonyl and carboxyl groups, increasing hydrophilicity and reactivity.
- **Grafting:** Grafting polymers (e.g., polyethylene glycol (PEG), polymethyl methacrylate (PMMA)) onto LNPs can improve dispersibility, biocompatibility, and provide specific functionalities.

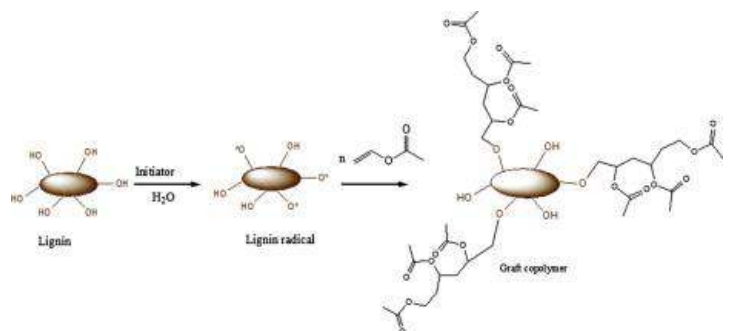


Fig 4: Graft copolymerization of Lignin with vinyl acetate [30]

- **Amination:** Introducing amino groups onto LNPs can enhance their reactivity and enable further functionalization for applications like drug delivery.
- **Phenolation:** Reacting lignin with phenol under acidic conditions introduces phenolic hydroxyl groups, enhancing antioxidant activity and reactivity.

### 3.2. Physical Modification:

- **Coating:** Coating LNPs with other materials, such as polymers or inorganic nanoparticles, can alter their surface properties and provide protection against degradation.

Table 1: Some empirical studies on the modification of lignin for pollutant removal application

Modification agent	Application	Adsorption capacity (mg/g), %	Reference
Fe nanoparticles	Cr <sup>6+</sup> removal	65.83%	[31]
Polyethyleneimine	Sb <sup>3+</sup>	371.7	[32]
Reinforced Chitosan	Cr <sup>6+</sup> removal	100%	[33]
Chlorohexanoic acid	Ni <sup>2+</sup>	70.3	[34]
Fe <sub>3</sub> O <sub>4</sub>	Cu <sup>2+</sup>	353.36	[35]
Fe <sub>3</sub> O <sub>4</sub>	As <sup>5+</sup>	>77.6%	[36]
Fe(0)	Naphthalene	97.81%	[37]
MIL-101-NH <sub>2</sub>	Tetracycline	40	[38]
Polyethyleneimine	Indigo disulfonate	1099.01	[39]
Chitosan	Tartrazine	436.68	[39]
Amidogen	Naphthalene	97.81%	[31]

Some of the general properties of modified lignin particles include;

- **Enhanced Solubility:** Chemical modifications can improve the solubility of LNPs in various solvents, expanding their applicability in different formulations.
- **Functionalization:** Modification processes can introduce functional groups (e.g., amine, carboxyl) that enhance reactivity and allow for specific interactions with other molecules, increasing their utility in targeted drug delivery.
- **Tailored Release Profiles:** Modified LNPs can be designed to achieve controlled or sustained release of therapeutic agents, which is critical in pharmaceutical applications.
- **Improved Mechanical Properties:** Modifications can lead to improved mechanical properties, such as tensile strength and elasticity, enhancing their use in composite materials.
- **Increased Thermal Stability:** Chemical modifications can enhance the thermal stability of lignin nanoparticles, making them suitable for high-temperature applications.
- **Targeted Applications:** Functionalized LNPs can be tailored for specific applications, such as targeting cancer cells or enhancing antimicrobial activity in coatings.

Industrial effluents pose a significant threat to environmental health due to the presence of various pollutants, including heavy metals, dyes, pharmaceuticals, and other organic contaminants. Conventional treatment methods often face limitations in terms of efficiency, cost-effectiveness, and environmental impact [40]. The use of modified lignin nanoparticles (LNPs) for pollutant adsorption has gained attention due to their sustainability, cost-effectiveness, and high functional versatility [6]. Traditional chemical modification methods, while effective, can involve hazardous chemicals and generate toxic by-products [41]. To address these concerns, green modification techniques, which prioritize environmental sustainability and safety, have gained attention. Green modification focuses on sustainable processes that minimize environmental harm. It involves using non-toxic reagents, energy-efficient methods, and renewable resources to achieve desired modifications. For LNPs, green modification aligns with their eco-friendly origins and enhances their compatibility with green technologies.

#### 4.0 Benefits of Green Modification:

- 1. Reduced Environmental Impact:** Minimizes hazardous waste and toxic by-products.
- 2. Energy Efficiency:** Employs methods that consume less energy, such as enzymatic or microwave-assisted processes.
- 3. Biocompatibility:** Ensures the modified nanoparticles are safe for biomedical and environmental use.
- 4. Alignment with Circular Economy Principles:** Utilizes renewable resources and supports sustainable material cycles. Green modification can improve the adsorption performance of LNPs by:

- **Increasing surface area:** Modification can lead to changes in the morphology of LNPs, increasing their surface area and providing more adsorption sites.
- **Introducing specific functional groups:** Modification can introduce functional groups with high affinity for specific pollutants, enhancing adsorption selectivity.
- **Improving dispersibility:** Modification can improve the dispersibility of LNPs in aqueous solutions, facilitating their interaction with pollutants.

#### 4.2. Green Modification Techniques for Lignin Nanoparticles

There various physical and chemical techniques utilized in green modification of lignin nanoparticles. These processes enhance the capacity of lignin nanoparticles for the adsorption of organic and inorganic pollutants. These techniques include;



Fig 5: Green Modification of Lignin NPs

##### 4.2.1 Enzymatic Functionalization

Enzymes like laccase and peroxidase catalyze the introduction of reactive functional groups, such as hydroxyl (-OH) and carboxyl (-COOH) groups, onto lignin nanoparticles. These groups improve the adsorption efficiency of pollutants like heavy metals and dyes through electrostatic interactions and complexation [42].

##### 4.2.2 Microwave-Assisted Modification

Microwave irradiation promotes rapid and energy-efficient chemical modifications, such as sulfonation and esterification, under environmentally friendly conditions. The process enhances LNP dispersibility and interaction with pollutants. Xiao *et al.* [43] achieved the microwave-assisted sulfonation of lignin with multiple chemicals for the production of a lignin-based dye dispersant.

##### 4.2.3 Plant-Based Functionalization

Plant-derived compounds, such as tannins and flavonoids, can bind to LNP surfaces, introducing natural antioxidant and adsorptive properties. These modifications are particularly effective for adsorbing organic pollutants. A study by Guo *et al.* [44] on co-self-assembly of lignin and tannin successfully achieved the enhanced removal of metal ions from aqueous solution. The study found that the abundant ortho-phenolic hydroxyl groups of tannin caused the efficient adsorption and partial reduction of Pd<sup>2+</sup> ions.

##### 4.2.4. Functionalization with Natural Polymers

LNPs can be functionalized with natural polymers such as chitosan, starch, or cellulose to improve their stability and adsorption properties. For example, chitosan-modified LNPs have shown enhanced adsorption of heavy metals, like Cu(II) and Pb (II) due to the chelating ability of chitosan [28].

##### 4.2.5. Crosslinking with Biobased Crosslinkers

Crosslinking LNPs with biobased agents like genipin or epichlorohydrin enhances their mechanical strength and reusability. Crosslinked LNPs have been effective in adsorbing organic pollutants such as phenols and pharmaceuticals [45].

#### 4.2.6. Incorporation of Magnetic Nanoparticles

Magnetic LNPs, synthesized by embedding iron oxide nanoparticles, enable easy separation of adsorbents from treated water using an external magnetic field. This approach reduces the risk of secondary pollution and improves the practicality of LNPs in industrial applications [23].

#### 4.2.7. Surface Grafting with Eco-Friendly Agents

Surface grafting involves attaching functional groups such as carboxyl, amino, or sulfonate groups to LNPs using green reagents like citric acid or amino acids. This modification increases the number of active sites for pollutant binding. A study by Tang *et al.* [42] demonstrated that carboxyl-functionalized LNPs exhibited high adsorption capacity for cationic dyes.

### 4.3. Mechanisms of Pollutant Adsorption on GMLNP

#### 4.3.1 Physical Adsorption

Green-modified lignin nanoparticles offer a high surface area for physical adsorption. This mechanism relies on van der Waals forces and hydrophobic interactions, making it effective for removing non-polar organic pollutants.

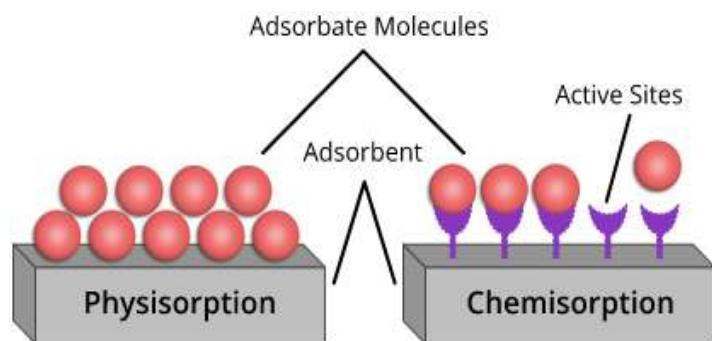


Fig 6: Physisorption vs Chemisorption

#### 4.3.2 Chemical Adsorption

Functional groups introduced during green modification, such as carboxylic acids, phenols, and sulfonic acids, enable chemical bonding with pollutants. The chemical adsorption of pollutants by modified LNPs involves various mechanisms, including:

- Electrostatic interactions between charged pollutants and functional groups on LNPs. An example is when sulfonated LNPs bind with cationic dyes through electrostatic interactions. According to Li *et al.* [35], sulphonation improved electrostatic interactions, as well as  $\pi$ - $\pi$  stacking, in the adsorption process.
- Hydrogen bonding and van der Waals forces for non-ionic pollutants. The hydroxyl and carbonyl groups on lignin nanoparticle surfaces can form hydrogen bonds with organic compounds, promoting their adsorption [46].
- Chelation and coordination for heavy metal ions. The interactions between heavy metal ions and lignin nanoparticles primarily involve complexation and chelation processes [47]. This may be attributed to the formation of coordination complexes between anionic groups in the lignin, such as  $-\text{COOH}$  and  $-\text{OH}$ , and the metal ions.
- $\pi$ - $\pi$  interactions for aromatic compounds. Aromatic organic contaminants often undergo  $\pi$ - $\pi$  stacking interactions with lignin nanoparticles, facilitated by the presence of phenolic rings in lignin structures [47].

#### 4.3.3 Ion Exchange

Ion exchange occurs when functionalized LNPs replace ions on their surface with pollutant ions in solution. This mechanism is highly effective for removing heavy metals and ammonium ions from industrial effluents.

### 4.4. Applications in Pollutant Removal

#### 4.4.1 Heavy Metal Adsorption

Green-modified lignin nanoparticles have shown exceptional potential for removing heavy metals such as chromium, lead, and mercury. The high surface area to volume ratio of lignin nanoparticles, combined with the multitude of active sites offered by their functional groups, results in a substantial adsorption capacity for heavy metals [3]. The interactions between heavy metal ions and lignin nanoparticles primarily involve complexation and chelation processes. According to Zhang *et al.* [48], functionalized lignin-based hybrid magnetic nanoparticles exhibited high adsorption capacities for lead and copper ions from simulated industrial wastewater. Studies on the utilization of green modified LNPs for enhanced adsorption of metal ions have also been reported [9], [42], [48], [50].

#### 4.4.2 Dye Removal

Dyes from textile and paper industries are major water pollutants. Sulfonated and tannin-functionalized LNPs have demonstrated high adsorption capacities for cationic and anionic dyes. A study by Li *et al.* [35] indicated that sulfonated LNPs removed over 90% of methylene blue from dye effluents within 60 minutes, which is higher than the removal efficiency of unmodified LNP. Xiao *et al.* [43] utilized microwave-assisted sulfonation of lignin nanoparticles in dye dispersion application, with efficient results. Furthermore, Tang *et al.* [42] reported that carboxyl-functionalized lignin LNPs were very effective for dye removal.

#### 4.4.3 Organic Pollutant Adsorption

Persistent organic pollutants, including phenols and polycyclic aromatic hydrocarbons, can be effectively removed using green-modified LNPs. Pasini *et al.* [51] reported that lignin nanoparticles have shown remarkable versatility in adsorbing various organic contaminants, including pesticides, pharmaceuticals, dyes, and volatile organic compounds (VOCs). According to Tan *et al.* [46] the properties of lignin such as their porous structure, large surface area, and abundant functional groups enable effective interactions with organic molecules.

### 5.0 Application of lignin nanoparticles in soil remediation

Soil pollution remains a serious environmental challenge, threatening ecological balance and public health [52]. As the demand for sustainable and effective remediation strategies increases, lignin nanoparticles have gained attention as an innovative solution. Produced from lignocellulosic biomass, these nanoscale particles possess distinctive characteristics that make them suitable for treating contaminated soils [53].

Their high surface area and abundance functional groups such as hydroxyl and phenolic groups give lignin nanoparticles remarkable adsorption capacity [54]. This enables them to bind and immobilize a wide range of pollutants, including heavy metals, organic contaminants, and synthetic dyes [55]. Through these interactions, they reduce the mobility and toxicity of hazardous substances in soil environments [55].

A major advantage of lignin nanoparticles is their sustainability material, making them consistent with environmentally friendly remediation approaches.

In addition to their strong pollutant-binding ability, lignin nanoparticles also play a meaningful role in improving overall soil quality. They enhance soil structure by encouraging particle aggregation and increasing the soil's capacity to retain moisture [56].

Furthermore, as carbon-rich materials, they provide an energy source for beneficial soil microorganisms. This stimulates microbial activity and promotes nutrient cycling processes that are essential for sustainable agriculture, land rehabilitation, and ecological restoration [57]. Lignin nanoparticles represent a sustainable and versatile solution for soil remediation [26]. Their remarkable adsorption capabilities, environmental friendliness, and potential for improving soil quality make them a valuable material in the current effort to remediate soil contamination and safeguard environmental health [58].

### 6.0 Air Pollution Control with Lignin Nanoparticles:

Lignin nanoparticles (LNPs) have emerged as promising materials for air pollution control due to their versatile pollutant-removal mechanisms. They are capable of capturing gaseous contaminants such as volatile organic compounds (VOCs) and industrial emissions through surface adsorption, physical filtration, and chemical interactions including hydrogen bonding and  $\pi$ - $\pi$  stacking, electrostatic interactions and/or hydrogen bonding [59]. Their nanoscale size provides a high surface-area-to-volume ratio, which significantly enhances their reactivity and adsorption efficiency compared to bulk lignin [60]. In addition to passive adsorption, engineered lignin-based composites can exhibit photocatalytic properties, enabling the degradation of organic air pollutants under visible light. This makes them not only absorptive but also actively degradative materials. Furthermore, lignin is a renewable byproduct of the pulp and paper industry, making LNP-based air purification systems environmentally friendly and sustainable alternatives to conventional materials derived from non-renewable resources [61]. To further improve their performance in industrial air filtration systems, lignin nanoparticles can be functionalized for example, by incorporating magnetic iron oxide to enhance their structural stability, ease of recovery, and recyclability.

## 7. Challenges and Future Directions

### 7.1 Challenges

- 1. Scalability:** Green modification methods, such as enzymatic processes, may be cost-prohibitive at an industrial scale.
- 2. Selectivity:** While GMLNPs can adsorb a wide range of pollutants, their selectivity for specific contaminants requires optimization.
- 3. Stability:** Maintaining nanoparticle stability in complex wastewater matrices remains a challenge.

### 7.2 Future Directions

- 1. Hybrid Materials:** Combining GMLNPs with other adsorbents, such as activated carbon or biochar, can improve pollutant removal efficiency.
- 2. Advanced Modification Techniques:** Further development of energy-efficient methods, such as plasma treatment and supercritical CO<sub>2</sub> modification, could enhance the scalability of green modification.
- 3. Circular Economy Integration:** Exploring the recovery and reuse of GMLNPs from treated wastewater can enhance their economic and environmental viability.

## 8. Conclusion

Green-modified lignin nanoparticles present a promising solution for addressing the environmental challenges posed by industrial effluents. Their sustainable origin, combined with advanced green modification techniques, enables efficient adsorption of a wide range of pollutants, including heavy metals, dyes, and organic compounds. While challenges such as scalability and stability remain, ongoing research into hybrid materials and advanced green processes will likely unlock the full potential of GMLNPs. As industries move towards more sustainable practices, GMLNPs stand as a cornerstone for eco-friendly wastewater treatment solutions and soil remediation.

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