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Sustainable approaches to lignin recovery from kraft black liquor and its application in environmental remediation

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Black liquor, the alkaline effluent of kraft pulping, is one of the largest underutilized sources of technical lignin globally. This chemically valuable aromatic biopolymer is predominantly incinerated for energy recovery, foregoing the considerable chemical value embedded in lignin's aromatic backbone and reactive functional groups. Conventional lignin isolation via acid precipitation is constrained by high energy demands, equipment corrosion, and acidic waste generation. These limitations collectively undermine the large-scale valorization of lignin. This review examines conventional and emerging approaches for lignin recovery from kraft black liquor, with particular emphasis on ionic liquids and deep eutectic solvents. Beyond extraction, the potential of lignin-derived materials for environmental applications is discussed, highlighting their role in promoting a circular economy. This review discusses lignin precipitation mechanisms and evaluates key barriers, including solvent recovery, scalability, and techno-economic feasibility. Addressing these challenges positions the pulp and paper industry at the forefront of sustainable chemical manufacturing.

KEYWORDS

environmental applications, green solvents, lignin, surface functionalities, sustainable approaches

1 Introduction

Lignocellulosic biomass has become a cornerstone of renewable energy research in response to the depletion of fossil fuel resources. Among its components, lignin stands out as a complex and heterogeneous biopolymer found within the secondary cell walls of vascular plants (Yu et al., 2025; Sun, 2020; Zhang et al., 2022). Lignin is crucial to structural integrity, water transport, and defense mechanisms (Li et al., 2022a). Despite its essential role in plant physiology, lignin is a major impediment in industrial applications such as pulp and paper production. Residual lignin darkens pulp, reduces pliability, and compromises final product quality (Mandal et al., 2023). Delignification thus defines the kraft pulping process, but at a substantial chemical and environmental cost (Saadan et al., 2024; Rullifank et al., 2020).

A direct consequence of delignification is the generation of kraft black liquor, an alkaline aqueous waste stream generated during the kraft pulping process. Lignin constitutes 25%–35% of the dry matter in black liquor, alongside 30%–35% aliphatic carboxylic acids, 5%–10% other organic compounds, and 30%–40% inorganic compounds (Zhou et al., 2022; Argyropoulos et al., 2023). However, the precise composition of black liquor varies

significantly depending on factors such as wood species and delignification conditions (Pola et al., 2022). Softwood-derived black liquor generally contains higher lignin content and lower proportions of aliphatic acids than hardwood-derived streams. Traditionally, kraft black liquor is primarily incinerated in recovery boilers to regenerate pulping chemicals and produce energy (Hao, 2021). However, this approach overlooks the potential value of lignin itself. Incineration converts a potentially high-value chemical stream into low-grade steam. The opportunity cost of this conversion has been underappreciated by industry for decades.

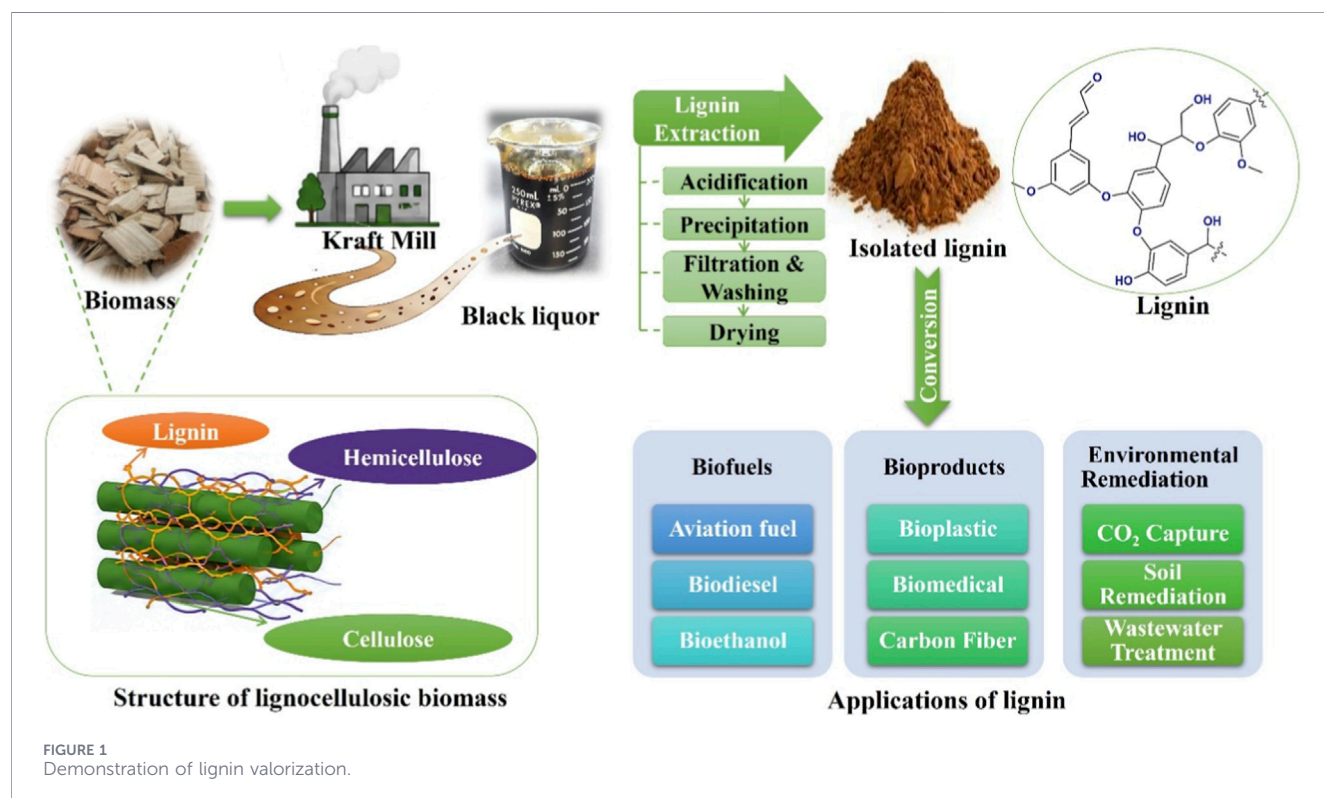
During kraft pulping, wood chips are treated with white liquor, a mixture of sodium hydroxide and sodium sulfide, under high temperature and pressure. These conditions chemically degrade lignin and separate it from cellulose and hemicellulose, yielding kraft lignin (Jardim et al., 2022; Woiciechowski et al., 2020). Unlike the polysaccharide fractions, lignin is a complex, three-dimensional, amorphous biopolymer (Sun, 2020; Ralph et al., 2019; Ralph et al., 2008; Lu et al., 2017; Sharma and Kumar, 2020). Its aromatic nature, thermal stability, and renewability make it an attractive feedstock for the development of bio-based materials and chemical transformations (Shah et al., 2023; Li et al., 2022b; Li et al., 2025).

Notably, the same reasons that necessitate the removal of lignin in pulp production also highlight its potential for high-value applications (Li et al., 2019). The pulp and paper industry has explored lignin recovery via acidification since the 1960s, recognizing its potential for producing value-added materials (Hasan et al., 2023). Figure 1 outlines the valorization pathway for lignin, from biomass pulping and lignin isolation to structural characterization and various industrial applications.

Conventional lignin recovery from black liquor relies on sequential acid precipitation. Initially, the pH is adjusted to approximately 9 using carbon dioxide (CO₂) precipitation, followed by sulfuric acid (H₂SO₄) precipitation to a pH of 2. The recovered lignin is subsequently separated from the black liquor by membrane filtration (Zhu et al., 2016; Wang et al., 2023a). Despite its industrial prevalence, this method is associated with significant drawbacks (Tanis et al., 2023). Equipment corrosion, high capital expenditure, energy-intensive operation, and hazardous chemical disposal collectively limit its sustainability and economic viability.

Different pulping processes yield distinct technical lignin types with unique structural and chemical characteristics. Kraft, lignosulfonate, soda, and organosolv lignins each exhibit different molecular-weight distributions, functional group profiles, and degrees of condensation (Melro et al., 2018). Furthermore, the structure and chemical properties of lignin vary significantly with extraction method, directly influencing its potential applications. Conventional extraction techniques include grinding, hydrolysis, enzymatic degradation, and solvent-based extraction (Gillet et al., 2017; Yoo et al., 2020; Allegretti et al., 2019; Roy et al., 2022). Understanding this structure-property-application relationship is hence critical for rational valorization of lignin.

There is therefore a pressing need for alternative recovery strategies from kraft black liquor. This review evaluates advanced solvent systems, including deep eutectic solvents (DESs), ionic liquids (ILs), and supercritical fluids, as emerging alternatives for lignin isolation and valorization. These systems generally operate under milder processing conditions and may better preserve lignin functional groups than conventional mineral acid precipitation (Roy et al., 2020). The downstream potential of recovered lignin for environmental remediation, including water treatment and soil



restoration, is critically assessed within a circular economy framework. Current limitations, particularly the lack of pilot-scale validation, are explicitly addressed. This work aims to bridge existing gaps and advance lignin's role as a key material in sustainable, zero-waste industrial systems.

2 Lignin recovery approaches

Sustainable recovery of lignin from kraft black liquor is a critical step toward the development of integrated biorefineries and circular bioeconomy strategies. The heterogeneous nature of lignin, together with its sensitivity to pH, temperature, ionic strength, and black liquor composition, makes selective recovery challenging (Zhu et al., 2016; Yoo et al., 2020; Reyes et al., 2020). Conventional recovery technologies, including acid precipitation, have been successfully implemented at industrial scale because they integrate well with existing kraft mill operations and enable efficient chemical recovery (Morya et al., 2022). However, these processes may increase energy consumption, promote lignin condensation, and require corrosive chemicals (Alam et al., 2024). Consequently, considerable research has focused on developing alternative recovery strategies that better preserve lignin structure while reducing environmental impacts (Sharma et al., 2023; Lappalainen et al., 2020). Rather than replacing conventional technologies, these emerging approaches aim to complement existing processes by improving lignin quality, preserving functional groups, and expanding downstream applications.

2.1 Conventional lignin recovery processes

Several methods have been explored for extracting lignin from black liquor. These include the Westvaco process, the LignoBoost process, the LignoForce process, and the more recent Sequential Liquid-Lignin Recovery and Purification (SLRP) process (Kienberger et al., 2021; Fitzgerald and Thies, 2022). Although these technologies share the common objective of precipitating lignin through acidification, they differ in process configuration, lignin purity, ash removal, filtration performance, and compatibility with the chemical recovery cycle of kraft mill. Understanding these differences is essential for selecting suitable recovery routes for specific lignin applications.

2.1.1 Westvaco process

In the Westvaco process, black liquor with solid content of approximately 20–30 wt% is acidified with CO₂ in alternating tanks until the pH reaches approximately 9. The carbonated liquor is then heated to 71 °C–93 °C to enhance filterability before cooling, filtering and drying. Further processing of this lignin may include acid hydrolysis to remove hemicellulose and sodium, leaving sulfur from the digestion process on the lignin (Kienberger et al., 2021; Xie et al., 2015).

In contrast, continuous processes using H₂SO₄ rapidly lower the pH, promoting protonation of lignin functional groups and molecular aggregation. Excessively low pH may also induce acid-catalyzed condensation. However, this method presents challenges during filtration and may produce lignin with relatively high levels

of residual sulfur and inorganic impurities, limiting its use in certain high-value applications.

2.1.2 LignoBoost process

LignoBoost was developed to address the purity limitations of the Westvaco process. It operates as a two-stage process using black liquor drawn from the evaporation plant (Tomani et al., 2011). In the first stage, the pH is reduced using CO₂. As the pH decreases, ionized phenolic and carboxylate groups become protonated, reducing the negative surface charge and promoting lignin aggregation and precipitation. In the second stage, the filter cake is re-dispersed and re-acidified. The resulting slurry is filtered and washed by displacement washing. This two-step approach significantly improves lignin purity compared with single-stage acid precipitation (Jeffri et al., 2024).

2.1.3 LignoForce process

The LignoForce process introduces two additional steps before lignin precipitation, namely black liquor filtration and oxidation. The first commercial-scale implementation of the LignoForce process was launched in 2016 by West Fraser in Canada (Kouisni et al., 2012). Kraft black liquor releases total reduced sulfur (TRS) compounds, including hydrogen sulfide, methyl mercaptan, dimethyl sulfide, and dimethyl disulfide, particularly below pH 11 (Kouisni et al., 2016). These volatile compounds have a strong odor and are more likely to be released in acidic conditions. At higher pH values (above ~12), sulfides remain in ionic forms such as HS[−] or S^{2−}, reducing their volatility (Kreitman et al., 2019). The LignoForce oxidizes black liquor under controlled conditions prior to acidification. This improves lignin filterability by promoting colloidal agglomeration into particles with a more uniform size distribution. It also reduces acid consumption and substantially reduces TRS emissions (Kouisni et al., 2014).

2.1.4 Sequential liquid-lignin recovery and purification process

The Sequential Liquid-Lignin Recovery and Purification (SLRP) process operates under elevated pressure (5–15 bar) and temperature (100 °C–150 °C) (Kienberger et al., 2021). These conditions maintain lignin in a liquid state, distinguishing SLRP from earlier precipitation-based recovery methods. CO₂ is introduced at the base of a column reactor, where it contacts the black liquor in a counter-current flow from above, reducing the pH to approximately 9–10 (Lake and Blackburn, 2014). The resulting liquid lignin phase is separated from the lignin-lean liquor, which is returned to the evaporation train. The liquid lignin is then dispersed in water, acidified with H₂SO₄, filtered, and washed to reduce ash content (Lake and Blackburn, 2014).

2.1.5 Sulfonation of lignin

Sulfonation is one of the most widely used post-precipitation modification methods for producing water-soluble lignin derivatives (Borrega et al., 2020). The resulting product, lignosulfonate, is the most commercially available technical lignin, with global production

exceeding one million tonnes annually (Strassberger et al., 2014). During this process, sulfonic acid groups ($-\text{SO}_3\text{H}$) are introduced onto the lignin backbone through reactions with sulfite- or sulfur-based reagents (Anukam et al., 2021). The incorporation of sulfonate groups significantly improves water solubility, colloidal stability, and dispersibility while increasing the density of negatively charged functional groups.

Despite its advantages, sulfonation increases the sulfur content and processing cost of lignin, limiting its use in carbon materials and high-temperature applications (Borrega et al., 2020; Fatehi and Chen, 2016). Consequently, sulfonated lignin is preferred for aqueous applications, whereas native kraft lignin is generally more suitable for carbon materials and polymer composites (Eraghi Kazzaz and Fatehi, 2020).

2.1.6 Comparison of commercial technical lignin

Although these recovery technologies have been successfully implemented at industrial scale, they produce lignin with markedly different physicochemical properties. Molecular-weight, ash content, sulfur content, solubility, and functional group composition vary depending on the recovery and purification processes. These properties strongly influence downstream applications. For example, low-ash and low-sulfur lignin are preferred for carbon materials, polymer composites, and catalytic applications, whereas water-soluble lignosulfonates are better suited for dispersants, binders, and soil remediation (Belgacem et al., 2003). Consequently, evaluating lignin properties is as important as understanding the recovery process itself. Table 1 summarizes the representative properties, indicative commercial prices, major suppliers, and typical applications of commercially available technical lignin.

In addition to precipitation-based processes, membrane separation has emerged as an important downstream purification technique. Ultrafiltration and nanofiltration are widely used to fractionate lignin into defined molecular-weight ranges while reducing ash and inorganic impurities. Commercial suppliers frequently combine membrane processes with precipitation to produce application-specific lignin fractions with improved

consistency for carbon materials, polymers, and specialty chemicals. However, membrane fouling and capital costs remain major challenges to large-scale implementation.

2.1.7 Issues with and alternatives to conventional processes

Conventional lignin recovery technologies remain the benchmark for industrial implementation because of their proven scalability and integration with existing kraft mill infrastructure. Processes such as LignoBoost and LignoForce produce high-purity lignin while maintaining the sodium and sulfur balance required for efficient chemical recovery (Kienberger et al., 2021).

Despite their effectiveness, these processes rely on acidification, which increases chemical consumption and may promote lignin condensation under strongly acidic conditions. Emerging solvent systems, including deep eutectic solvents, ionic liquids, and organic acids, have demonstrated the potential to preserve lignin structure under milder conditions. However, challenges associated with solvent cost, viscosity, recovery efficiency, recyclability, and large-scale implementation continue to limit their commercial adoption. Future research should therefore focus on integrating the robustness of conventional processes with the structural advantages offered by potential lower-impact recovery technologies, while improving solvent recovery, recyclability, and techno-economic feasibility.

2.2 Emerging methods for sustainable lignin recovery

Recent advancements in solvent-based lignin precipitation have introduced promising alternatives that may reduce hazardous chemical consumption and better preserve lignin structure under selected processing conditions (Lu and Gu, 2022; Gigli and Crestini, 2020; Wang L. et al., 2024). These methods have the potential to reduce hazardous chemical inputs, lower energy demands, and generate fewer harmful by-products (Álvarez et al., 2024; McMichael, 2023). They may also preserve the functional group framework of lignin, which conventional methods frequently degrade (Mersmann et al., 2025; Wang R. et al., 2024). However,

TABLE 1 Chemical and physical properties of different commercial lignin.

Process	Commercial product	Typical MW (g/mol)	Ash (%)	Sulfur (%)	Water solubility	Moisture content (%)	Prices (US\$/t)	Ref
Kraft Pulping	BioPiva	5,000–6,000	0.5–3.0	<1.5–3.0	No	<6.0	250–600	Borrega et al. (2020)
LignoBoost	LignoBoost lignin	1,500–25,000	0.02–1.0	1.5–3.0	No	8.0–35.0	250–500	De Assis et al. (2018)
LignoForce	LignoForce lignin	3,000–10,000	<1.0–2.0	1.0–2.5	No	~8.0	250–600	Hu et al. (2016), Dessbesell et al. (2020)
Organosolv	Organosolv lignin	<5,000	0–1.7	0	Slight	4.0–6.0	300–520	Vishtal and Kraslawski (2011)
Lignosulfonate	Borregaard	<15,000	4.5–18.0	3.0–8.0	Yes	<5.0	180–500	Ludmila et al. (2015)
Soda Pulping	Soda Lignin	1,000–3,000	0.5–2.3	0	No	2.5–5.0	200–300	Vishtal and Kraslawski (2011), Ludmila et al. (2015), Tribot et al. (2019)

their overall sustainability depends on solvent recovery, process integration, recyclability, and life-cycle performance.

2.2.1 Inorganic acids

As previously mentioned, weak acids such as CO_2 and strong acids such as H_2SO_4 remain the most widely used agents for lignin precipitation from black liquor. Both acids primarily function by protonating ionized phenolic functional groups, reducing the negative surface charge of lignin molecules and promoting aggregation and precipitation (Sewring and Theliander, 2019). Strong acids can further lower the pH below approximately 2, resulting in protonation of carboxylate groups and increased lignin recovery. Consequently, the choice of acid directly affects lignin recovery, functional group composition, molecular-weight, and process cost (Pajer et al., 2024).

One key advantage of using CO_2 is that it does not disrupt the sodium (Na) and sulfur (S) balance in the kraft recovery cycle. Studies have demonstrated that CO_2 effectively precipitates lignin by lowering black liquor pH to approximately 7–9.5 (Nagy et al., 2010; Kosa et al., 2011; Yiamsawas et al., 2023; Chen et al., 2013). Yiamsawas et al. demonstrated that CO_2 precipitation at pH 9–10 produces lignin with high purity and structural integrity, forming uniform particles with good dispersibility (Yiamsawas et al., 2023). Stoklosa et al. extended this approach using sequential CO_2 acidification for pH-controlled fractionation of softwood lignin (Stoklosa et al., 2013). They found that phenolic hydroxyl content increased as pH decreased, while molecular-weight and $\beta\text{-O-4}$ linkages correlated positively with pH. This enables production of tailored lignin fractions for specific applications.

H_2SO_4 is often considered more effective than CO_2 for lignin precipitation due to its ability to lower pH more rapidly and completely, leading to faster and higher lignin recovery. It is also relatively inexpensive and fully dissociates in aqueous solution, allowing efficient protonation of lignin functional groups (Jung and Kim, 2015). However, the use of H_2SO_4 may disrupt the sodium and sulfur balance of the kraft recovery cycle and increase chemical consumption and wastewater-treatment requirements (Hubbe et al., 2019).

2.2.2 Ionic liquids

Ionic liquids (ILs) are organic salts with tunable chemical and physical properties that melt below 100 °C (Introduction, 2017). They consist of cations such as ammonium, pyridinium, phosphonium, sulfonium, and cholinium, and organic or inorganic anions (e.g., BF_4^- , PF_6^- ; CH_3COO^- ; $\text{N}(\text{CN})_2^-$) (Melro et al., 2018; Galbe and Wallberg, 2019). One significant benefit of ILs is their ability to combine different cations and anions, allowing precise control over solvent power, polarity, and hydrophobicity (Hasanov et al., 2020). ILs possess remarkable properties, including low melting points, negligible vapor pressure, wide electrochemical windows (~6 V), high electrical conductivity, and broad liquid ranges, making them ideal solvents for lignin precipitation.

ILs precipitate lignin by disrupting the hydrogen bonding and electrostatic interactions that maintain its solubility in the highly alkaline black liquor. Depending on the IL chemistry, lignin

precipitation may occur through pH reduction, disruption of intermolecular interactions, or anti-solvent addition, resulting in lignin aggregation and phase separation (Yinghuai et al., 2013). Cations can interact with aromatic and oxygen-containing functional groups through electrostatic, hydrogen-bonding, and $\pi\text{-}\pi$ interactions. Anions influence lignin solubility through hydrogen-bonding interactions and changes in solution chemistry (Mukesh et al., 2024).

This method not only improves lignin recovery but also offers more control over its purity and molecular structure. By selecting specific ILs, lignin recovery can be tailored for specific downstream applications. Furthermore, the low volatility and potential recyclability of ILs make them a promising alternative to traditional acidification methods, with potential environmental and economic benefits. Recent studies have identified highly effective ILs for lignin precipitation (Bagh et al., 2021; Chambon et al., 2020). Bagh et al. extracted lignin from black liquor using five acetate-based ILs, achieving up to $75.3\% \pm 3.4\%$ efficiency with ethanolanmonium acetate [Eth][Ac] at a solvent-to-black liquor ratio of 20:1 (Bagh and Ray, 2022). The lignin recovery achieved with [Eth][Ac] was also comparable to that obtained using the conventional IL [Emim][Ac] ($77.6\% \pm 2.1\%$). These findings demonstrate the potential of protic ILs as a promising alternative for kraft black liquor lignin recovery.

Despite these advantages, the industrial implementation of IL-based recovery remains limited by solvent cost, high viscosity, solvent recovery, and process economics (Welton, 2018). The strong $\pi\text{-}\pi$ interactions between ILs and lignin can complicate solvent regeneration, increasing solvent consumption and operating costs during repeated recovery cycles (de Maria and Maugeri, 2011).

2.2.3 Organic acids

Organic acids, characterized by their carboxyl ($-\text{COOH}$) functional groups, typically contain ten or fewer carbon atoms, distinguishing them from fatty acids (Wang Y. et al., 2020). These acids occur naturally in plants and animals. The acidity is influenced by additional functional groups, including alcohols, phenols, thiols, and enols. Citric, lactic, and acetic acids are the most commonly used organic acids for lignin precipitation, with citric acid being the most widely studied.

The precipitation mechanism is similar to that of inorganic acids but generally operates under milder conditions. Organic acids protonate phenolic and carboxyl groups on lignin, reducing electrostatic repulsion between dissolved lignin molecules and promoting aggregation. The mild acidity of organic acids generally reduces the extent of acid-catalyzed condensation reactions during precipitation. Condensation, which involves the formation of new carbon-carbon or carbon-oxygen bonds between lignin fragments, is a well-documented consequence of aggressive acid treatment (Li et al., 2018; Huang et al., 2025). It increases molecular-weight heterogeneity, reduces reactive functional group content, and limits downstream applications. Organic acid precipitation suppresses this effect, yielding lignin with more intact $\beta\text{-O-4}$ linkages and higher phenolic hydroxyl content (Rahimi et al., 2014). These structural features are directly valuable for applications in polymer synthesis, antioxidant

formulation, and catalytic conversion. Furthermore, lignin precipitation using organic acids offers a cleaner alternative to sulfuric acid-induced precipitation.

Silva et al. investigated the isolation of kraft lignin from hardwood black liquor using acetic, lactic, and citric acids (da Silva et al., 2020). They reported that these organic acids yielded high-purity lignin with low ash and carbohydrate contents. The recovered lignin exhibited greater methoxyl carbonyl group contents, strong antioxidant activity, and a high energy potential (21–24 MJ/kg). Namane et al. reported that lignin precipitated with organic acids had lower sulfur content and different thermal properties than lignin precipitated using CO₂ or H₂SO₄ (Namane et al., 2016). It also exhibited higher char yields and lower weight loss during thermal degradation, making it a promising candidate for bio-based chemical applications.

However, unlike sulfuric acid, organic acids cannot reduce pH below approximately 2–3 due to their weak acidity. This limits the extent of protonation and may reduce lignin recovery. Their lower acid strength may also require higher acid consumption or longer contact times to achieve comparable precipitation. This can influence process economics and large-scale implementation.

Nevertheless, organic acids offer an important advantage from a process-integration perspective. Unlike sulfuric acid, they do not disrupt the sodium and sulfur balance of the kraft recovery cycle. This reduces chemical make-up requirements at the mill level, simplifies waste-stream management, and improves compatibility with existing kraft mill chemical-recovery operations.

2.2.4 Supercritical fluid

A supercritical fluid exists above its critical temperature and pressure simultaneously, exhibiting both liquid- and gas-like properties. It possesses liquid-like density together with gas-like diffusivity and low viscosity. Supercritical fluid technologies have been investigated for lignin precipitation from black liquor by utilizing these unique transport properties. Depending on the process conditions, supercritical fluids may also reduce undesirable lignin condensation by facilitating selective separation under relatively mild conditions.

Carbon dioxide is the most widely used supercritical fluid in lignin processing. It becomes supercritical above 304.25 K and 72.9 atm (Ali et al., 2023). Supercritical CO₂ (scCO₂) is non-flammable and leaves no solvent residue upon depressurization, a significant advantage over organic solvents in large-scale processing environments.

Unlike strong mineral acids, scCO₂ lowers the pH of black liquor through the formation of carbonic acid (H₂CO₃), which subsequently establishes bicarbonate (HCO₃²⁻) and carbonate (CO₃²⁻) equilibria. The resulting decrease in pH protonates ionized phenolic groups, reducing lignin solubility and promoting precipitation (Wang X. et al., 2017; Yang et al., 2018). The high diffusivity of CO₂ may also facilitate water displacement from porous lignin flocs during depressurization. However, this dewatering effect depends strongly on pressure, temperature, black liquor composition, and reactor configuration. Consequently, scCO₂ remains a promising proof-of-concept technology, although further pilot-scale validation is required before industrial implementation.

Adjaye et al. developed a novel scCO₂ process for lignin precipitation, where scCO₂ acidifies black liquor below the pK_a of lignin phenolic groups (Asafu-Adjaye et al., 2022). This facilitates precipitation while simultaneously facilitating dewatering of the flocculated solids. Compared with conventional thermal evaporation, this approach has the potential to improve lignin separation while reducing thermal energy requirements. However, its overall techno-economic feasibility depends on CO₂ compression energy, pressure maintenance, reactor design, and process integration within existing kraft mills.

2.2.5 Deep eutectic solvents

Deep eutectic solvents (DESs) represent one of the most promising alternative solvent systems for lignin recovery. Unlike ILs, which require complex multi-step synthesis and costly purification, DESs form spontaneously when two or more components are combined at specific molar ratios. Their unique property, a pronounced melting-point depression at specific eutectic compositions, was first observed by Abbott et al. in a 1:2 mixture of choline chloride and urea (Abbott et al., 2003). Choline chloride has a melting temperature of approximately 302 °C, whereas urea melts at approximately 133 °C. This mixture, known as reline, forms a liquid at room temperature with a eutectic melting point of 12 °C. This dramatic melting point depression arises from the formation of an extensive hydrogen-bonding network between the chloride anion of choline chloride and the hydrogen-bond donors of urea. The same principle governs all DESs systems, in which a hydrogen-bond acceptor is paired with one or more hydrogen bond donors to form a structurally reorganized liquid (Abbott et al., 2003; Ashworth et al., 2016; Stefanovic et al., 2017; Arunan et al., 2011). These solvents exhibit physicochemical properties that are distinct from those of their individual components (Nabhai et al., 2026). DESs have the potential to revolutionize solvent applications due to their low cost, biodegradability, non-toxicity, and ease of synthesis compared to ILs (Xu P. et al., 2017; Khandelwal et al., 2016; Radošević et al., 2015).

Choline chloride is the most widely used hydrogen-bond acceptor because of its low cost, commercial availability, and biocompatibility. Common hydrogen bond donors include urea, glycerol, and carboxylic acids (Pinho et al., 2024). The choice of these donors determines the DESs polarity, viscosity, hydrogen bonding capacity, and ultimately the functional groups of the recovered lignin. Within these systems, choline chloride promotes lignin solubility and dispersion through hydrogen-bond interactions (Chen et al., 2023; Liu et al., 2019; Abbott et al., 2001). Hydrogen-bond donors influence the acidity of the DESs and thus the extent of lignin modification during recovery. Depending on the DESs composition, acidity, temperature, and residence time, treatment may either preserve ether linkages under mild recovery conditions or promote partial β-O-4 cleavage under more acidic or severe conditions (Yang et al., 2023; De Santi et al., 2021). Therefore, the extent of structural preservation is highly dependent on DESs composition and operating conditions.

Liu et al. demonstrated this advantage directly by isolating lignin from black liquor using a DES through a microwave-assisted process with a lactic acid and choline chloride (10:1 M ratio) (Liu et al., 2023). This process yielded 1.58 g of lignin with 88.12% purity, outperforming conventional acid precipitation

(1.18 g, 78.54% purity). The DES also retained more than 95% of its recovery efficiency over three reuse cycles, demonstrating promising solvent recyclability. Similarly, Bagh et al. recovered lignin from black liquor using lactic acid:choline chloride (11:1 M ratio) and achieved a recovery efficiency of 79.7%, comparable to that obtained using IL [Emim][Ac] (Saadat et al., 2022). These studies demonstrate the potential of DESs as alternative media for kraft black liquor lignin recovery. Although DESs generally exhibit recovery efficiencies comparable to those of ILs, they may offer advantages in terms of lower solvent cost, simpler preparation, and lower processing temperatures. However, these advantages depend on efficient solvent recovery and process integration and should not be considered universal.

A major limitation of many DESs systems is their relatively high viscosity, particularly for glycerol-based or high-lactic-acid formulations (Pires et al., 2022). High viscosity may hinder mass transfer during precipitation, slow filtration, and complicate lignin cake washing. Consequently, reducing viscosity while maintaining solvent performance remains an important objective for future development of DES-based solvents.

Direct comparison of lignin recovery methods is challenging because reported values depend on the feedstock, black liquor composition, operating conditions, and analytical methods. Therefore, the values presented in Table 2 should be regarded as representative ranges rather than directly comparable. Table 2 summarizes molecular-weight, ash content, and inter-unit linkage information, which strongly influence lignin reactivity and downstream applications.

Conventional recovery processes have demonstrated superior industrial maturity and seamless integration with kraft mill operations. Emerging solvent systems may better preserve the ether linkages and oxygen-containing functional groups in kraft lignin compared with aggressive mineral acid precipitation. The extent of structural preservation depends on the severity of the kraft pulping process and the subsequent recovery conditions. However, their commercial implementation remains constrained by solvent cost, recovery efficiency, viscosity, and scale-up challenges. Consequently, the selection of a lignin recovery method should consider not only recovery performance but also the desired physicochemical properties of the recovered lignin and its intended end-use application.

3 Mechanisms and influences of lignin recovery from black liquor

Native wood lignin is an amorphous, branched, and polydisperse macromolecular polymer composed of phenylpropane units (Vieira et al., 2022). It functions as a binding agent within the lignocellulosic matrix, filling spaces between cellulose and hemicellulose to confer structural rigidity. Its primary structural units are p-coumaryl alcohol (H-unit), sinapyl alcohol (S-unit), and coniferyl alcohol (G-unit) (del Río et al., 2020). Together, lignin accounts for approximately 15–35% of the dry weight of lignocellulosic biomass, depending on the biomass source. The dominant inter-unit linkages include β -O-4 aryl ether, β -5

phenylcoumaran, and β - β' pinosresinol, with β -O-4 linkages being the most abundant (Liu et al., 2024; Schoofs et al., 2024; Balakshin et al., 2020). Table 3 summarizes the distribution of these linkages across softwood and hardwood lignin. The monolignol units and dominant interunit linkages of lignin (Figure 2) play a critical role in determining its structural properties and influence its behavior during recovery from black liquor.

Hardwood and softwood lignin differ in monomeric composition. Hardwood lignin contains both coniferyl and sinapyl alcohol units, whereas softwood lignin is composed predominantly of coniferyl alcohol (G units) (Erfani Jazi et al., 2019). The higher methoxy-group content of hardwood lignin reduces carbon-carbon condensation and results in a less condensed structure. The principal functional groups in lignin include phenolic hydroxyl, aliphatic hydroxyl, carbonyl, and carboxyl groups (Balakshin et al., 2014). However, only a fraction of the phenolic hydroxyl groups are free, whereas the majority participate in inter-unit linkages within the lignin polymer. During enzymatic dehydrogenation, carbonyl and aliphatic hydroxyl groups are incorporated into the lignin structure, further influencing its physicochemical properties (Zhu and Theliander, 2015).

During the kraft pulping process, wood is subjected to temperatures exceeding 160 °C, reaction times of two to 3 hours, and strongly alkaline conditions containing hydroxide and hydrosulfide nucleophiles (Fearon et al., 2020). These conditions selectively fragment and solubilize lignin while partially degrading hemicelluloses. During treatment, hydroxide and hydrosulfide anions cleave lignin-carbohydrate linkages and break inter-unit bonds, liberating lignin from the carbohydrate matrix (Zakzeski et al., 2010; Geller and stedt, 2015). Structural changes in native lignin involve the cleavage of β -O-4 ether bonds, the introduction of thiol groups and stilbene linkages, and modifications to carbohydrate linkages (Fearon et al., 2020). These changes define the chemical character of kraft lignin and directly influence its precipitation behavior (Gillet et al., 2017).

At alkaline pH (Woiciechowski et al., 2020; Ralph et al., 2019), the weakly acidic carboxyl and phenolic groups of lignin are predominantly deprotonated (Fearon et al., 2020). Electrostatic repulsion between the resulting negatively charged species maintains lignin in solution. Above pH approximately 13.8, lignin is highly soluble. Ionic-dipole and hydrogen-bonding interactions with the solvent dominate over intermolecular associations. Dissolved lignin interacts with sodium ions, behaving as a polyelectrolyte. Understanding this dissolved state is essential as precipitation is, mechanistically, the reversal of these stabilizing interactions.

Lignin precipitation from alkaline solutions is closely related to the protonation of ionized phenolic groups (-O^-). The dissociation equilibrium of the phenolic group in lignin can be expressed by Equation 1:



where L represents the lignin molecule, and -OH refers to the phenolic hydroxyl group on the aromatic ring of a lignin phenylpropane unit.

TABLE 2 Comparison of lignin recovery methods from black liquor based on recovery performance, structural characteristics, and physicochemical properties.

Solvent type	Feedstock/ black liquor	Conditions	Lignin purity (%)	Extraction efficiency (%)	Molecular weight (g/mol)	Ash content (%)	Energy consumption	Structural integrity	Scale	Ref.
CO ₂	Kraft black liquor	60–80 °C/pH 9–10	65–75	60–70	2,500–5,000	1–3	Low–Moderate (no neutralization waste; integrates with mill cycle)	Moderate (limited condensation; β-O-4 partially preserved)	Industrial	Robles et al. (2016) , Tomani (2010)
H ₂ SO ₄ (Strong acid)	Kraft black liquor	pH < 2	60–70	65–75	3,000–7,000	2–5	High (corrosive, neutralization, effluent treatment needed)	Low (acid-catalyzed condensation; structural degradation)	Industrial	Nadányi et al. (2024)
Organic Acids	Kraft black liquor	pH 2–4	80–90	Moderate to High	1,500–3,500	<1	Low, cleaner than strong acids	High (β-O-4 linkages preserved; low sulfur contamination)	Laboratory	Sharma et al. (2024) , Duret et al. (2025)
Ionic Liquids	Kraft/non-kraft	IL dependent	75–90	70–80	1,500–4,000	<1	High (due to multi-step recovery and heating)	High (selective precipitation; controlled molecular weight)	Laboratory	Chambon et al. (2020)
Deep eutectic solvents	Kraft black liquor	DES dependent	79–88	79–84	1,500–3,500	<1	Low–Moderate (mild temperatures, simple recovery)	Condition-dependent	Laboratory	Li P. et al., (2023) ; Song et al. (2020)
Supercritical fluids	Kraft black liquor	>31 °C, >7.4 MPa	70–85	70–80	-	-	Moderate–Low (no thermal evaporation needed)	-	Laboratory	Asafu-Adjaye et al. (2022)

TABLE 3 Lignin linkages connecting the phenylpropane units and their approximate units (Eswaran et al., 2022).

Type of linkage	Dimer structure	Structure type	Approximate percentage (%)	
			Softwood	Hardwood
β -O-4	Arylglycerol- β -aryl ether	Linear	45–50	50–65
β -5	Phenylcoumaran	Linear	9–12	3–11
5–5	Biphenyl	Branched	10–11	1–5
β - β	Pinoresinol	Linear	2–6	3–16
β -1	1,2-Diarylpropane	Branched	7	7
4-O-5	Diaryl ether	Branched	4–8	7
α -O-4	Noncyclic benzyl aryl ether	Linear	9–12	3–11
Others	-	-	13	5

The dissociation constant (K_a) of the phenolic group is expressed by Equation 2:

$$K_a = \frac{[L - O^-][H^+]}{[L - OH]} \quad (2)$$

Assuming that protonated lignin exists as a solid phase with unit activity and that ion activity coefficients are approximately unity, Equation 2 simplifies to Equation 3:

$$K_a = [L - O^-][H^+] \quad (3)$$

The logarithmic form of the dissociation constant, pK_a , is defined by Equation 4:

$$pK_a \equiv -\log_{10} K_a \quad (4)$$

which is widely used to describe phenolic group dissociation (Zhu et al., 2015). As the solution pH decreases toward the pK_a of a given functional group, protonation becomes increasingly favorable. Consequently, lignin precipitation occurs progressively as ionized phenolic and, under more acidic conditions, carboxylate groups become protonated, reducing electrostatic repulsion and promoting molecular aggregation. The apparent pK_a is influenced by temperature, ionic strength, solvent composition, and lignin molecular structure. The influence of these factors on lignin precipitation is discussed in the following subsections.

3.1 Influence of pH

pH is the dominant control variable in lignin precipitation from black liquor (García et al., 2009; Helander et al., 2013; Lourençon et al., 2015; Wang and Chen, 2013). The ionization state of lignin functional groups is governed by their dissociation constants (pK_a), as summarized in Table 4. In general, oxygen-containing functional groups adjacent to the aromatic ring lower the pK_a , increasing the acidity of lignin (Wei et al., 2024). As the pH decreases, functional groups become progressively protonated as the solution pH approaches and falls below their pK_a values (Zhou et al., 2023).

The reduction in surface charge weakens electrostatic repulsion, allowing attractive intermolecular interactions, including van der Waals forces and π - π stacking, to promote molecular aggregation

and precipitation. Lignin recovered at lower pH generally exhibits higher recovery as demonstrated in Figure 3 (García et al., 2009). However, excessive acidification (typically below pH 2) may promote acid-catalyzed condensation, increasing structural heterogeneity and reducing the abundance of reactive β -O-4 linkages. Therefore, precipitation pH must be carefully optimized to balance lignin recovery and structural preservation (Wang and Chen, 2013).

Ela et al. systematically investigated the influence of precipitation variables on lignin recovery from hardwood kraft black liquor (Andeme Ela et al., 2020). The titration curve revealed a buffering region between pH 5.5 and 8.5, where limited lignin protonation occurred and precipitation was minimal. As the pH decreased below this region, protonation of phenolic groups reduced the negative surface charge (ζ -potential), promoting molecular aggregation and increasing lignin recovery. The authors also observed that precipitation at very low pH increased lignin yield but promoted acid-catalyzed condensation, leading to greater structural heterogeneity and a lower proportion of β -O-4 linkages. These findings demonstrate that careful control of precipitation pH is essential for balancing lignin recovery with product quality.

3.2 Influence of temperature

Temperature affects lignin precipitation through three concurrent mechanisms, it modifies lignin solubility, alters aggregation kinetics, and shifts the apparent pK_a of phenolic groups. These effects do not act independently, making temperature one of the most complex and least predictable process variables in lignin recovery. Particle formation is governed by temperature-dependent local supersaturation. As temperature rises, nucleation kinetics accelerate, but particle morphology also changes.

Two key findings from the literature reveal an important apparent contradiction. Helander et al., using a 1,000 Da membrane cutoff to fractionate weak black liquor, found that low molecular-weight lignin could not be extracted at 70 °C and pH 9 (Borisova et al., 2015). They attributed this to the formation of relatively large lignin agglomerates, which retained low-molecular-weight lignin within the aggregates. Wallmo et al., by contrast, found

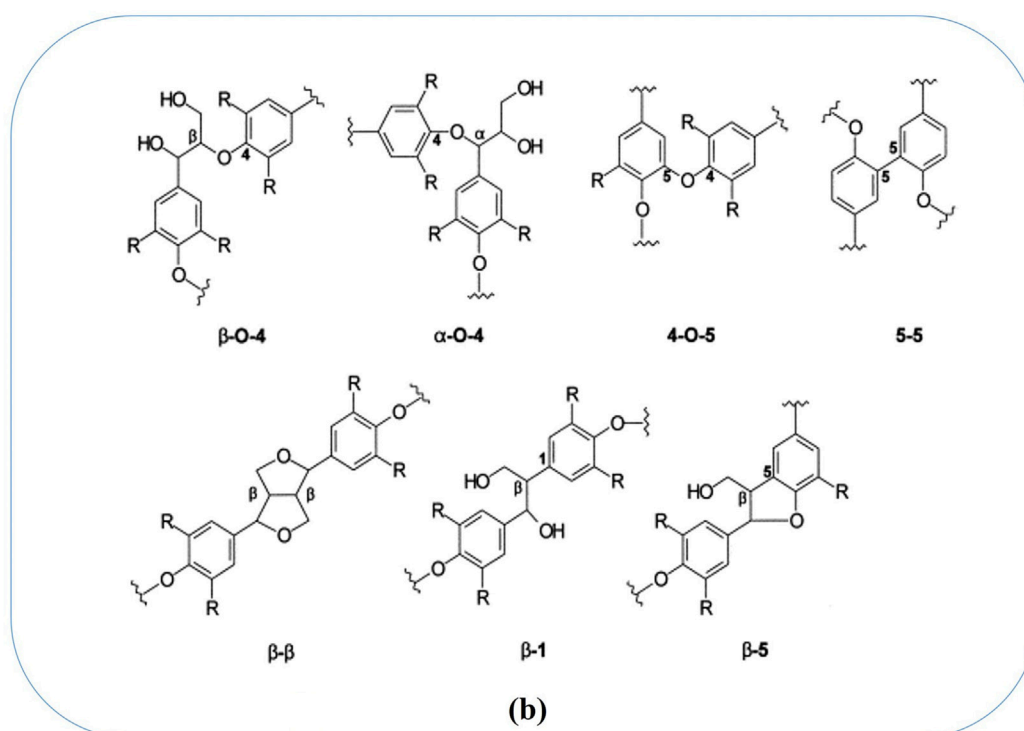
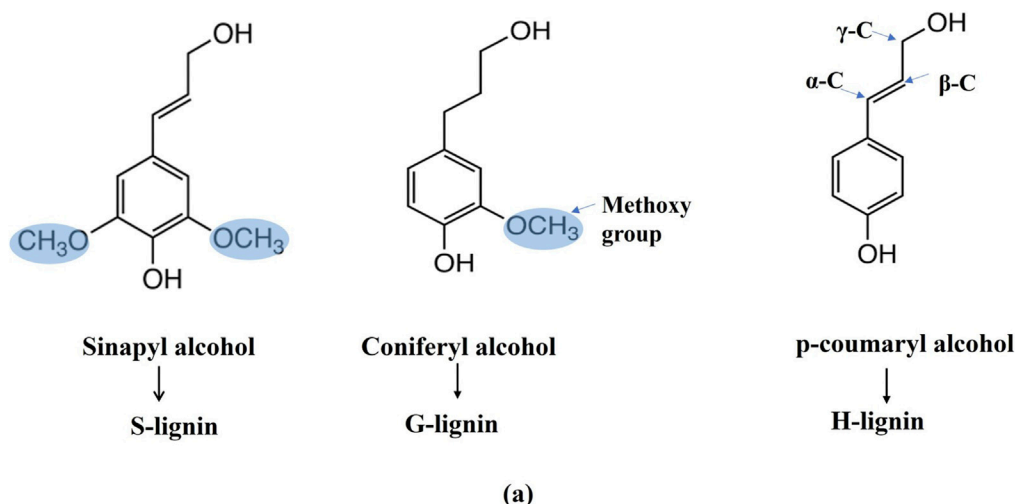


FIGURE 2
(a) Monolignol species: p-coumaryl alcohol (H unit), coniferyl alcohol (G unit), sinapyl alcohol (S unit) and (b) the most dominant linkages of lignin.

that CO_2 -based lignin precipitation increased with decreasing temperature (Wallmo et al., 2009). These findings are not necessarily contradictory but reflect the sensitivity of precipitation behavior to the specific method employed.

Higher temperatures also led to more compact, sticky, and closely packed particles, reducing xylose content and promoting particle growth (Aja et al., 2018). The underlying thermodynamic explanation involves pK_a shifts. Higher temperatures lower the apparent pK_a of kraft lignin phenolic groups, increasing their degree of dissociation and therefore increasing lignin solubility. Increased thermal energy also enhances electrostatic repulsion

between dissolved lignin molecules. Consequently, above a critical temperature, further heating may suppress rather than enhance lignin precipitation. Despite its importance, the relationship between temperature, lignin structure, and particle formation remains insufficiently understood and requires further investigation (Mishra and Ekielski, 2019).

3.3 Influence of ionic strength

Modifying the ionic strength (IS) of lignin solutions is another effective approach for lignin precipitation (Sabaghi and Fatehi, 2020;

TABLE 4 Lignin dissociation constants of various functional groups.

Type of group	pK _a Value
R – CH(OR')COOH	3–4
R – COOH	4–5
RCO – phenyl – OH	7–8
phenolic – OH	9.5–10.5
Hemiacetalic	12–12.5
Alcoholic	13.5–17

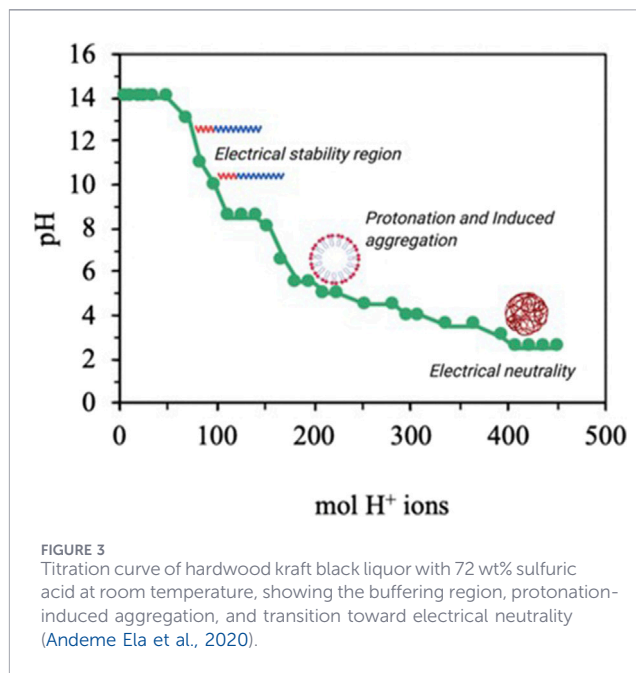
(Figueiredo et al., 2021). Black liquor contains primary inorganic ions such as Na⁺, K⁺, Cl[−], HS[−], SO₄^{2−}, SO₃^{2−}, S₂O₃^{2−}, and CO₃^{2−} (Hao, 2021). Due to its highly alkaline nature, the organic components in black liquor remain predominantly dissolved. As ionic strength increases, the electrical double layer surrounding charged lignin molecules is compressed (Jacobs et al., 2023). Consequently, electrostatic repulsion between lignin macromolecules decreases, reducing the energy barrier for aggregation. Additional ions also partially neutralize surface charges, promoting floc formation and precipitation. Therefore, increasing ionic strength enhances lignin recovery by improving the precipitation process yield.

This effect is strongly influenced by changes in temperature and pH. Zhu et al. confirmed this relationship experimentally, demonstrating that lignin yield increased systematically with ionic strength (Zhu and Theliander, 2015). Among the variables studied, pH was found to have the most significant effect. However, ionic strength amplified precipitation but could not substitute for adequate acidification. In industrial black liquor, the naturally high ionic strength may reduce the amount of acid required to achieve target precipitation yields compared to dilute model systems.

The interaction between ionic strength, pH, and temperature represents the most undercharacterized aspect of lignin precipitation science. Each variable is typically studied in isolation, yet in industrial black liquor they operate simultaneously and interdependently. A better understanding of these coupled effects will be important for optimizing industrial lignin recovery processes.

4 Applications of lignin in environmental remediation

Lignin has considerable potential for environmental remediation, including water treatment and soil restoration. Its performance depends on functional groups, molecular-weight, structural integrity, and surface properties. These properties are influenced by both the original lignin source and the recovery method. Emerging recovery systems, including organic acids, DESs, and ILs, may alter lignin chemistry during isolation and thereby influence its downstream remediation performance. However, direct studies linking alternative-solvent recovery of kraft lignin to improved remediation remain limited. Most available studies use commercial lignin, lignosulfonates, non-kraft lignin, or materials subjected to additional modification. The following



section evaluates the performance of lignin-based materials across different environmental applications.

4.1 Removal of inorganic compounds from water

Lignin-based materials remove inorganic contaminants mainly through surface complexation, electrostatic interactions, ion exchange, and chelation (Yang et al., 2019; Sharma et al., 2022). Phenolic hydroxyl and carboxyl groups provide oxygen-donor sites for metal coordination, while adjacent functional groups may enable multidentate interaction (Kayan and Kayan, 2021; Feng et al., 2018). The dominant mechanism depends on lignin chemistry, solution pH, metal speciation, and competing ions (Amonette and Joseph, 2012; Luo et al., 2018). Sulfonic, amine, and thiol groups can provide additional binding sites, but these functionalities are often introduced through post-recovery modification rather than during the original isolation process (Li et al., 2022b; Ge et al., 2014a; Wang et al., 2023b; Yan and Li, 2016).

Recovery conditions may influence this behavior by changing the abundance and accessibility of functional groups. Carboxylic acid-based DESs systems may increase the density of oxygen-containing functional groups during precipitation and recovery. This may improve the metal-binding affinity of the recovered lignin compared with conventional sulfuric acid precipitation. Organic acid-mediated precipitation has similarly been reported to increase carbonyl and methoxyl groups relative to mineral acid precipitation, potentially improving adsorption toward inorganic contaminants.

Unmodified lignin provides intrinsic metal-binding sites but generally exhibits limited surface area and heterogeneous site accessibility. Functionalization can increase affinity and selectivity by introducing carboxyl, amine, sulfonate, or sulfur-containing groups. For example, the high adsorption capacities of carboxyl-functionalized lignin (COOH-lignin) and dithiocarbamate-modified lignin arise primarily from these additional binding

TABLE 5 Functionalized lignin for heavy metal removal and its efficiency.

Functionalized lignin-based adsorbents	Lignin isolation technique	Temp (°C)	pH	Residence time (min)	Pollutant	Adsorption capacity (mg g ⁻¹) /Removal efficiency (%)	Water matrix	Ref.
Amino and sulfonic groups Biofunctionalized lignin	Precipitation with H ₂ SO ₄	25	6	90	Cu (II) and Pb (II)	Cu (II) 71/76.1% and Pb (II) 26/94.5%	Syn ¹	Ge et al. (2014b)
Cu/N doped lignin	Commercial	25	8	30	As (V)	253.5/86.44%	Syn	Shi et al. (2020)
Aminated lignin	Precipitation using HCl	25	5.5	240	Cu (II)	37.14/70.71%	Syn	Xu et al. (2017b)
Polyethylenimine lignin	Commercial	25	6	60	Pb (II)	96.60/99.73%	Syn	Zhang et al. (2019)
COOH- lignin	Klason method	25	6	50	Pb (II)	269	Syn	Yu et al. (2020)
Cross-linked lignosulfonate	-	25	6	-	MB and Pb (II)	358/91.8% (MB) and 100.9/85% Pb (II)	Syn	Zhang et al. (2020)
Lignin-CNTs	Precipitation with H ₂ SO ₄	25	5.8	120	Pb (II)	251	Syn	Li et al. (2017)
Lignosulfonate/ N-methylaniline composite	Industrial	44	2	10	Cr (VI)	1,264.8/99.7%	Syn	Zhou et al. (2020)
Lignosulfonate/chitosan	Commercial	-	-	60	Co (II) and Cu (II)	Co (II) 386/99% and Cu (II) 283	Syn	Mu et al. (2020)
Amino-functionalized lignin	Kraft Pulping	22–44	6.5	90	Cd (II), Cr (VI) Ni (II) and As (V)	74.84 53.49 49.40 and 51.53	Syn	Popovic et al. (2020)
Organosolv lignin	Commercial	35	1	100	Cr (VI)	46.2/94.7%	Syn	Chi et al. (2020)

¹Synthetic Aqueous Solution.

sites. Table 5 summarizes representative lignin-based adsorbents for heavy metal removal. Because the reported adsorption performances were obtained under different experimental conditions, the values are intended for qualitative comparison only.

Adsorption capacity depends strongly on initial contaminant concentration, adsorbent dosage, pH, contact time, ionic strength, and water composition. High adsorption capacities measured in concentrated single-solute systems may not translate to effective treatment of dilute or complex effluents. Competing ions and natural organic matter can occupy binding sites, while changes in pH may promote desorption and contaminant remobilization. Regeneration can reduce material consumption, but repeated acid or base treatment may alter lignin structure and generate secondary waste. Modified materials also require assessment of reagent or metal leaching before practical deployment.

The key trade-off is therefore between material simplicity and adsorption performance. Unmodified kraft lignin is abundant and requires minimal processing, but its adsorption capacity and

selectivity are generally lower than those of functionalized lignins. Functionalized lignin offers stronger and more selective binding but requires additional reagents and processing. Lignin-derived composites often exhibit the highest performance, but their higher cost, potential component leaching, regeneration, and end-of-life management may reduce the overall sustainability benefits of using lignin. Consequently, recovered kraft lignin is currently most promising as a low-cost precursor for targeted functionalization rather than as a universal adsorbent.

4.2 Removal of organic compounds from water

Lignin-based materials have been widely studied for the removal of organic contaminants, including dyes and per- and polyfluoroalkyl substances (PFAS). Pollutant removal occurs through electrostatic attraction, hydrogen bonding, π - π interactions, hydrophobic interactions, and, in porous materials,

pore filling (Tkachenko et al., 2024). Aromatic contaminants can interact with the lignin backbone through π - π interactions, whereas hydroxyl, phenolic, and carboxyl groups contribute to hydrogen bonding and electrostatic interactions. Consequently, molecular-weight, degree of condensation, surface area, porosity, and functional group accessibility strongly influence removal performance.

Dyes are the most widely studied organic contaminants for lignin-based adsorbents. Unmodified lignin can adsorb dyes through its aromatic framework and oxygen-containing functional groups. However, adsorption strongly depends on dye charge and solution pH. At low pH ($\text{pH} < \text{pH}_{\text{pzc}}$), protonation of surface functional groups results in a positively charged lignin surface, promoting the adsorption of anionic dyes through electrostatic attraction. Conversely, under alkaline pH conditions ($\text{pH} > \text{pH}_{\text{pzc}}$), deprotonation of phenolic and carboxylic groups generates a negatively charged lignin surface that enhances adsorption of cationic dyes (Tkachenko et al., 2024; Jiang et al., 2023). Several studies have demonstrated the benefits of targeted functionalization, with additional charged or reactive groups improving the adsorption of both cationic and anionic dyes.

For example, Yu et al. developed a lignin-betaine polymer (LB) as a renewable water treatment material (Yu et al., 2023). The polymer achieved 98% removal of methylene blue (MB), primarily through electrostatic attraction, supplemented by hydrogen bonding and π - π interactions. Furthermore, the adsorbent exhibited good regeneration and reusability, highlighting its potential for practical wastewater treatment applications. Similarly, Han et al. prepared polyethylenimine-functionalized lignin/SiO₂ porous composite microspheres (LSPGs) through inverse emulsion polymerization (Han et al., 2023). The highest adsorption capacities were 355.21 mg/g for Cr (VI), 313.48 mg/g for PO₄³⁻, and 588.24 mg/g for Congo red. Adsorption occurred primarily through electrostatic attraction, along with redox reactions, hydrogen bonding, and π - π interactions. The adsorbent retained over 80% of its adsorption capacity after five regeneration cycles, demonstrating excellent structural stability and reusability. These studies demonstrate the benefit of targeted material modification rather than a universal advantage of recovered lignin.

Beyond dye remediation, lignin-based systems have also demonstrated considerable potential for removal and catalytic degradation of persistent fluorinated organic contaminants such as PFAS (Buck et al., 2011). PFAS exhibit exceptional chemical stability due to the strong C-F bond (heat of formation of C-F is 486 kJ/mol in CF₄), resulting in significant persistence within aquatic environments (Meegoda et al., 2020; Sunderland et al., 2019). Their interaction with lignin depends on fluorocarbon chain length, functional head group, hydrophobicity, and water chemistry.

Mel et al. studied PFAS adsorption and desorption using kraft lignin in simulated pulp and paper wastewater (Mel et al., 2024). PFAS containing sulfonate groups exhibited higher affinity for lignin than carboxylate-containing PFAS. Adsorption also increased with fluorocarbon chain length, with long-chain PFAS ($\text{C} \geq 6$) exhibiting stronger adsorption than short-chain PFAS ($\text{C} \leq 5$). These findings

provide useful insights into designing lignin-based PFAS treatment systems.

More complex lignin-derived materials have also been developed for PFAS degradation. For instance, Hou et al. prepared a lignin/polyvinyl alcohol (PVA)/Co/Fe metal-organic framework (lignin/PVA/bi-MOFs) composite membrane for the catalytic degradation of perfluorooctanoic acid (PFOA) (Hou et al., 2021). The membrane achieved approximately 89.6% degradation within 3 h under optimized conditions. The lignin-based membrane promoted both adsorption and catalytic activation of PFOA while maintaining high activity after repeated use.

Biomimetic lignocellulosic systems have also been explored for combined PFAS adsorption and biotransformation. For example, in a novel study, Li et al. developed a renewable artificial plant for *in-situ* microbial environmental remediation (RAPIMER), a lignocellulosic platform for PFAS remediation (Li et al., 2022b). RAPIMER exhibited high adsorption capacity for PFAS and co-contaminants, while facilitating *in-situ* biotransformation through the fungus, thereby enhancing PFAS detoxification. Constructed from renewable lignocellulosic materials, RAPIMER represents a promising strategy for integrated adsorption and biodegradation of persistent fluorinated contaminants. As summarized in Table 6, functionalized lignin systems exhibit strong adsorption and catalytic remediation performance toward a wide range of organic contaminants, including dyes and PFAS.

Collectively, these studies highlight the considerable potential of functionally engineered lignin-based composites as sustainable multifunctional materials for next-generation wastewater treatment applications.

4.3 Soil remediation

Soil contamination by heavy metals presents a remediation challenge fundamentally different from water treatment. Metals persist indefinitely in the soil matrix, accumulating over time through agricultural, industrial, and atmospheric deposition. They reduce biological activity, impair food production, leach into groundwater, and enter the food chains through plant uptake. Conventional remediation approaches, including excavation, soil washing, and electrokinetic extraction are effective but prohibitively expensive and ecologically disruptive at field scale.

In the quest for sustainable and efficient soil-remediation techniques, lignin-based materials have demonstrated significant potential, primarily through *in situ* solidification and stabilization. These methods are widely recognized for their operational simplicity, high efficiency, and cost-effectiveness. Phenolic, carboxyl, and other oxygen-containing groups can interact with metal ions through complexation, ion exchange, and electrostatic interactions (Wang D. et al., 2020; Ge and Li, 2018). This interaction effectively reduces the bioavailability and toxicity of heavy metals, thereby mitigating their environmental impact.

Lignin addition may also influence soil properties beyond metal stabilization. Depending on its composition and application rate, lignin may affect soil aggregation, water retention, organic carbon

TABLE 6 A list of important research reports on lignin-based materials for the removal of organic compounds from water.

Adsorbent	Design strategy	Dye name	Adsorption mechanism	Adsorption capacities (mg g ⁻¹)	Adsorption efficiency (%)	Water matrix	Ref
Lignin-grafted-(acrylamide and N-isopropyl acrylamide)/montmorillonite	Grafting	MB	Electrostatic interaction	9,646.92	96	Syn	Wang et al. (2017b)
Acetic acid lignin	Precipitation	MB	Electrostatic attraction	63.3	95.78	Syn	Feng et al. (2016)
Organosolv lignin	Extraction	MB	Hydrogen bonding, ion-exchange	40.02	-	Syn	Zhang et al. (2016)
Chitosan-alkali lignin composite	Chemical integration	Remazol Brilliant Blue R	Electrostatic interaction, chemical interaction	111.11	97	Syn	Nair et al. (2014)
Aminated lignin	One-pot amination reaction	CR	Electrostatic attraction	129.87	94.6	Syn	Chen et al. (2022)
Fe ₃ O ₄ @TEOS@APTES@lignin	Mannich reaction	CR	Chemical adsorption	229	95.5	Syn	Fang et al. (2021)
Chitosan-lignin-titania	Precipitation, sol-gel, blending	Brilliant black	Electrostatic interaction	15.8	83	Syn	Masilompane et al. (2018)
Cu (II) -doped lignin-based adsorbent	Amination, carbonization, Cu (II)-doping	Azure B	Electrostatic interaction, π - π interaction, hydrogen bonding	1,168	80	Syn	Li L. et al. (2023)
Quaternized lignin	Chemical grafting	Rhodamine B	π - π interaction, hydrogen bonding	41.85	99	Syn	Du et al. (2023)
Eucalyptus lignin	Klason method	MB	π - π interaction, hydrogen bonding	31.9	90	Syn	Cemin et al. (2021)
Carboxy-methylated lignin-Fe complex	Etherification, complexation	Brilliant Red 2BE	Electrostatic attraction	73.6	99.5	Syn	Da Silva et al. (2011)
Wheat straw lignin		Brilliant Red HE-3B	Ion exchange	10.173	-	Syn	Suteu et al. (2010)
Carboxy-methylated lignin-Al	Chemical grafting, complexation	Procion Blue MX-R	Electrostatic attraction	73.52	93.97	Syn	Adebayo et al. (2014)
Magnetic acetylated modified lignin	Self-assembly, electrostatic interactions	MB	Electrostatic attraction, hydrogen bonding	142.33	90	Syn	Zhao et al. (2022)
Carbonized and Bare Lignin	-	PFOA PFBS PFBA	hydrophobic interactions	5.3 4.3 1.6	-	Syn	Roman et al. (2025)
Lignin/PVA nanofiber membrane	Electrospinning method	PFOA	Catalytic activation through radical generation	-	89.6%	Syn	Hou et al. (2021)

content, and nutrient availability (Bolan et al., 2022). Lignin also serves as a carbon source for beneficial microorganisms, stimulating microbial activity and nutrient cycling, which are vital for sustainable land use and ecological restoration. This co-benefit profile makes lignin uniquely positioned for remediation contexts. A recent study developed biodegradable kraft lignin-based agro-hydrogels for soil hydration and slow urea release (Fihurka and Budnyak, 2025). The hydrogels improved water management while providing controlled nutrient delivery and showed 9.5%–14.5% weight loss after 40 days of soil burial. This demonstrates that kraft lignin-based materials can combine soil-conditioning functions with gradual biodegradation. Such multifunctionality may be valuable in remediation systems where contaminant stabilization is accompanied by soil restoration and revegetation. Similarly, Yang et al. developed modified waste lignin as a carrier for functional microorganisms (Yang et al., 2024). The lignin-based carrier increased microbial activity by 34.3%–41.0% and enhanced soil total nitrogen and available phosphorus by 70.8% and 64.4%, respectively, while promoting plant growth. These findings show that lignin-based materials can support soil restoration through complementary functions involving water management, nutrient delivery, and microbial activity.

Long-term stability nevertheless requires further evaluation. Lignin biodegradation and changes in soil pH or redox conditions may alter contaminant binding and potentially promote remobilization. Future studies should thus assess metal mobility, plant uptake, groundwater leaching, soil microbial responses, and long-term field performance. Consequently, a thorough understanding of the mechanisms governing lignin modification and stabilization is essential. Standardized preparation methods are also needed to minimize heterogeneity and batch-to-batch variability, ensuring reproducible material properties. Overall, lignin is particularly promising for *in situ* stabilization and restoration of contaminated soils because it combines metal-binding functionality with soil-conditioning benefits.

The suitability of lignin-based materials varies with both pollutant and lignin type. Metal-ion removal is particularly compatible with lignin's intrinsic oxygen-containing functional groups, although regeneration and contaminant remobilization remain concerns. Dyes and organic micropollutants often require additional functionalization or carbonization, increasing processing costs, while soil stabilization offers added soil-conditioning benefits but requires long-term validation. Kraft lignin, one of the most readily available technical lignin, is therefore particularly suitable for insoluble adsorbents and soil stabilization, whereas lignosulfonates are better suited to applications requiring water solubility and dispersion. Overall, the most practical applications are those that exploit the intrinsic chemistry of abundant technical lignins while minimizing extensive post-recovery modification.

4.4 CO₂ capture and carbon sequestration

Lignin-derived porous carbons have gained increasing attention for CO₂ adsorption applications because of their abundant and cost-effective raw material sources (Zhao et al., 2024; Cui et al., 2023; Nabhai et al., 2025). CO₂ adsorption primarily occurs through interactions between CO₂ molecules

and surface functional groups, including Lewis acid-base interactions, hydrogen bonding, and electrostatic interactions (Nabhai et al., 2025; Barker-Rothschild et al., 2025). These interactions are generally limited in unmodified lignin but can be enhanced through surface modification, which introduces additional adsorption sites and improves CO₂ adsorption capacity and CO₂/N₂ selectivity. Figure 4 illustrates the modification of lignin-derived porous carbons, their CO₂ adsorption mechanisms, and the regeneration and utilization of captured CO₂.

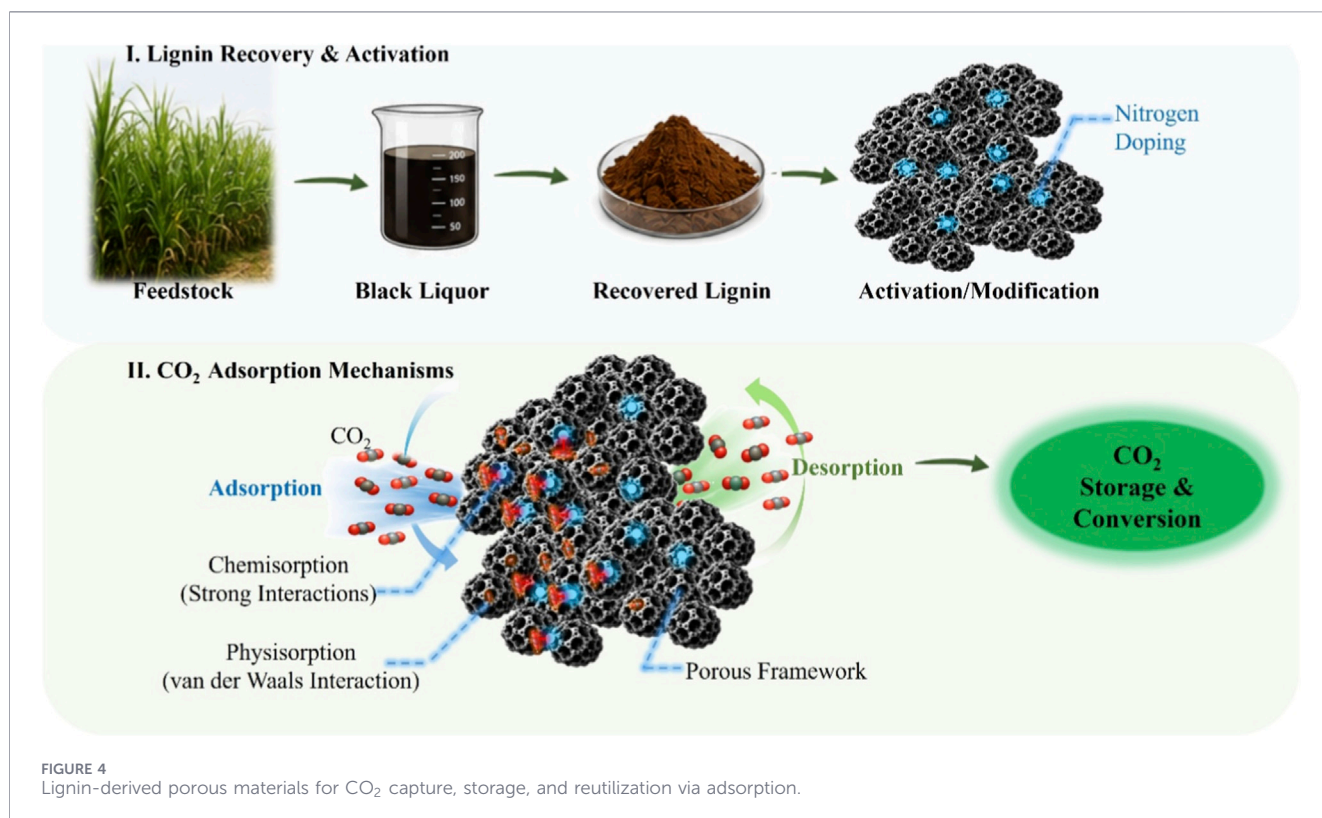
Nitrogen doping is one of the most effective modification strategies for improving CO₂ capture. Nitrogen-doped lignin introduces basic nitrogen functionalities (such as pyridinic and amino nitrogen), which strengthen interactions with CO₂ and improve both adsorption capacity and CO₂/N₂ selectivity (Liu et al., 2022). Activation further enhances adsorption performance by increasing surface area and pore development. For example, ZnCl₂ activation produced porous carbons with BET surface areas of 1,125–1,998 m²/g, and enhanced CO₂ adsorption and CO₂/N₂ selectivity (Zhao et al., 2024). Saha et al. demonstrated that KOH and NH₃ co-activation produced BET surface areas of 1,631–2,922 m²/g and demonstrated high CO₂ adsorption capacities of 5.48 mmol/g at 298 K and 8.6 mmol/g at 273 K (Saha et al., 2017). The combined approach outperforming most nitrogen-doped carbons and exhibited excellent CO₂/N₂ selectivity. Multi-cycle stability and effective dynamic column performance confirmed the relevance of these materials to industrial deployment.

Although these materials show considerable promise for carbon capture, chemical activation increases energy and reagent consumption. Therefore, efficient recovery and recycling of activating agents, together with NH₃ capture and reuse where applicable, are essential to minimize secondary waste and improve the overall sustainability of lignin-derived CO₂ adsorbents.

5 Circular economy and environmental impact

Biomass will undoubtedly play a pivotal role in the transition toward a sustainable bioeconomy as efforts to reduce CO₂ emissions intensify, and the cost of fossil-derived resources continues to increase. When lignin is removed from the recovery boiler, it transitions from being a component of black liquor into a value-added product with diverse applications. Given its tunable physicochemical properties, lignin has considerable potential as a renewable feedstock for sustainable materials and chemicals. However, its environmental and economic viability must be evaluated using life-cycle assessment (LCA) and techno-economic analysis (TEA) (Li et al., 2025; Osman et al., 2021).

LCA and TEA are therefore essential for determining whether emerging lignin recovery methods provide advantages over established processes. Although DESS- and ILS-based systems may reduce strong mineral acid use and improve lignin quality, their overall performance depends on solvent production, solvent loading, energy demand, recovery efficiency, water use, and waste generation. To date, the net environmental benefit of DESS- and ILS-based recovery has not been comprehensively quantified for kraft black liquor lignin.



Kulas et al. conducted TEA and LCA on the Aqueous Lignin Purification with Hot Agents (ALPHA) process for fractionating and purifying lignin from biorefineries and kraft pulp mills (Kulas et al., 2021). The process separates lignin into different molecular-weight fractions for applications including polyurethane foams, activated carbons, and carbon fibers. Scenario analysis revealed that solvent choice significantly affects process economics. Ethanol-based processing (US\$463/ton) produced substantially lower costs than acetic acid-based processing (US\$838/ton) while also generating higher-value products. The ethanol process yielded an estimated annual profit of US\$151 million and a net present value of US\$533 million over 30 years. LCA further showed that lignin-derived products produced using the ethanol process had greenhouse-gas emissions comparable to or lower than those of fossil-derived alternatives. These findings highlight that solvent selection influences both the economic and environmental performance of lignin recovery. However, comparable LCA and TEA studies for DESs- and ILs-based recovery from kraft black liquor remain unavailable, representing an important research gap.

Culbertson et al. evaluated the environmental impact of kraft pulp mills with and without kraft lignin recovery using both mass and economic allocation methods (Culbertson et al., 2016). Key findings showed that incorporating lignin extraction reduced the global warming potential from 722 kg CO₂ eq to 602 kg CO₂ eq per air-dried metric tonne of pulp. Under system expansion, the global warming potential further decreased to 234 kg CO₂ eq because lignin displaced petroleum-derived phenolic resins. Sodium chlorate and natural gas were identified as major contributors to environmental burdens. Overall, this study demonstrated that lignin recovery can reduce the environmental footprint of kraft pulp production.

Lignin valorization nevertheless faces structural challenges that LCA alone cannot resolve. Lignin composition varies with wood species, pulping conditions, and recovery method, resulting in variability in molecular-weight and functional-group distribution that can affect downstream product performance (Rinaldi et al., 2016). Several solvent-based recovery methods have been reported to produce lignin with smaller molecular-weight distributions and improved preservation of functional groups compared with conventional acid precipitation, potentially improving product consistency. However, whether these advantages outweigh the additional solvent and energy requirements remains to be established.

From a TEA perspective, conventional acid precipitation currently retains a clear advantage because of its industrial maturity and integration with existing kraft mill infrastructure. Tiller et al. compared CO₂-based lignin precipitation with the reuse of sodium bisulfate spent acid from pulp bleaching (Tiller et al., 2025). For a model mill producing 1,000 metric tons of bleached pulp and 140 metric tons of lignin per day, recycling bleaching spent acid reduced the estimated minimum lignin selling price from US\$426 t⁻¹ to US\$398 t⁻¹. However, the integrated process also increased natural-gas consumption by 2.3 t h⁻¹, illustrating that lower chemical costs do not necessarily correspond to lower environmental impacts. This study demonstrates the importance of mill integration and internal waste-stream utilization in lignin recovery economics. In comparison, the economic feasibility of DESs- and ILs-based recovery remains strongly dependent on solvent price, solvent-to-black-liquor ratio, solvent recovery, recyclability, viscosity, and

regeneration energy. Direct TEA and LCA using identical kraft black liquor feedstock and consistent system boundaries are therefore needed to determine whether improved lignin quality with emerging recovery systems can justify their additional processing costs.

Overall, positioning lignin as a cornerstone material in the circular bioeconomy requires comprehensive LCA and TEA comparing emerging solvent systems with conventional acid precipitation under consistent industrial conditions.

6 Perspectives, challenges and future prospects

Future research should focus on establishing direct relationships between kraft black liquor composition, recovery conditions, lignin structure, and downstream remediation performance. Although alternative recovery systems have shown considerable potential for preserving lignin quality, their industrial implementation remains constrained by lignin heterogeneity, solvent recovery, process integration, and scalability.

Looking ahead, DESs represent a promising opportunity for the circular utilization of black liquor. However, their viscosity and other physicochemical properties, including density and surface tension, must be optimized for large-scale applications. The use of water as a co-solvent offers a potential solution for modifying these properties, making DESs more adaptable for large-scale applications. Standardized comparisons are needed using the same black liquor feedstock to evaluate conventional acid precipitation, organic acids, DESs, ILs, and membrane-assisted recovery. These studies should report lignin yield, molecular-weight, ash and sulfur content, functional groups, solvent consumption, and recovery efficiency under comparable conditions.

Additionally, pilot-scale validation is particularly important for emerging solvent systems. DESs- and ILs-based solvents should be evaluated over repeated solvent-recycling cycles to determine solvent loss, impurity accumulation, energy demand, and changes in lignin quality. In addition, membrane fouling, solvent recovery, and integration with existing kraft mill chemical-recovery systems require further investigation.

A major knowledge gap is the limited understanding of how recovery-induced structural changes influence downstream performance. Future studies should compare lignin recovered from the same black liquor under identical conditions for pollutant removal, regeneration, and long-term stability. Ecotoxicity, modifier leaching, contaminant remobilization, and end-of-life management should also be evaluated. Finally,

comprehensive kraft-black-liquor-specific LCA and TEA are needed to determine whether emerging recovery technologies provide measurable environmental and economic advantages over conventional acid precipitation.

Author contributions

AN: Conceptualization, Visualization, Writing – review and editing, Writing – original draft. OW: Writing – review and editing. HG: Writing – review and editing. ZC: Writing – review and editing. HP: Writing – review and editing, Validation. JS: Resources, Writing – review and editing. QW: Writing – review and editing, Resources. JL: Conceptualization, Supervision, Writing – review and editing.

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