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Efficient and environmentally friendly techniques for extracting lignin from lignocellulose biomass and subsequent uses: A review

Mohammad Mahbubul Alam [a,b,*], Antonio Greco [a], Zahra Rajabimashhadi [a], Carola Esposito Corcione [a]

[a] Department of Engineering for Innovation, University of Salento, Lecce 73100, Italy
[b] Department of Textile Engineering, Ahsanullah University of Science and Technology, Dhaka 1208, Bangladesh

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ABSTRACT

The development of sustainable and effective methods for extracting lignin is crucial for achieving the advantages and promoting the shift towards a more sustainable and circular bioeconomy. This study addresses the use of environmentally friendly processes, including organosolv technique, supercritical fluid (SCF), non-thermal plasma (NTP), ionic liquids (ILs), deep eutectic solvents (DES), and microwave assisted extraction (MAE) techniques for lignin extraction. Organosolv treatment offers high selectivity and purity of lignin make this process economically feasible. Using supercritical water, carbon dioxide, or ethanol to extract lignin without harmful solvents is successful and customizable. NTP technologies break down lignin, simplifying processing and increasing its value. Whereas ILs may boost lignin synthesis and change its properties via solvent design. DES-based extraction methods can efficiently and specifically extract lignin. The rapid and effective MAE method employs microwave radiation to reduce extraction times and boost yields for lignin extraction. These methods feature high selectivity, little environmental impact, and the capacity to target lignin fractions. The study describes the fundamentals, benefits, and drawbacks of each extraction process, focusing on their ability to extract lignin on a large scale and its future usage. Additionally, this review explores the most recent advancements in the application sector, as well as the challenges and potential advantages of valorizing streams derived from extraction, thereby fostering the development of environmentally friendly and sustainable solutions. This research concludes that to overcome future challenges, need to address scale concerns, cost, emissions, and efficient lignin use.

1. Introduction

Plant cell walls are rich in lignin, a complex phenolic molecule found together with cellulose and hemicellulose (Balk et al., 2023). Lignin, the second most prevalent natural polymer globally, is mostly derived as a byproduct from the pulp and paper industry and ethanol manufacturing. Today, most biorefineries value cellulose and hemicellulose, a sugar-based platform. Though considered a low-value residual product, it has great potential for the generation of environmentally friendly, cost-effective, and biodegradable goods (Calvo-Flores and Martin-Martinez, 2022; Ferrari et al., 2022). Lignocellulose (LC) biomass fibers are utilized as reinforcing fillers and substitutes in polymers to create green

Abbreviations: APIL, Aprotic ionic liquid; BHT, Butylated hydroxytoluene; CAGR, Compound annual growth rate; CF, Carbon fiber; ChCl, Choline chloride; C-lignin, Catechyl Lignin; CPT, Cold plasma treatment; CXLF, CO_2-expanded liquid extraction; DES, Deep eutectic solvent; G, Guaiacyl; GLY, Glycerol; HBA, Hydrogen bond acceptor; HBD, Hydrogen bond donor; HCL, Hydrochloric acid; H_2SO_4, Sulfuric acid; IL, Ionic liquid; KL, Kraft lignin; LA, Lactic acid; LC, Lignocellulose; LNP, Lignin nanoparticles; LPR, Lignin-based phenolic resin; MAE, Microwave assisted extraction; $MgCl_2$, Magnesium chloride; MW, Microwave; MWA, Microwave assisted; MWL, Milled wood lignin; NTP, Non-thermal plasma; NaOH, Sodium Hydroxide; Na_2S, Sodium sulphide; NaDES, Natural deep eutectic solvents; OSL, Orgnosolv lignin; PC, Potassium carbonate; PBS, Polybutylene succinate; PIL, Protic ionic liquids; PLA, Polylactic acid; PR, Phenolic resins; PU, Polyurethane; S, Syringyl; SCF, Supercritical fluid; SC-CO_2, Supercritical carbon dioxide; SCES, SC-CO_2 ethanol solution; TPU, Thermoplastic polyurethane; UV, Ultra-violet; VOC, Volatile organic compounds; $ZrOCl_2$, Zirconyl chloride.

* Corresponding author at: Department of Textile Engineering, Ahsanullah University of Science and Technology, Dhaka 1208, Bangladesh.
E-mail addresses: mohammad.alam@studenti.unisalento.it (M.M. Alam), antonio.greco@unisalento.it (A. Greco), zahra.rajabimashhadi@unisalento.it (Z. Rajabimashhadi), carola.corcione@unisalento.it (C. Esposito Corcione).

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composites, owing to their environmentally friendly properties (renewable, fully or partially recyclable) (Bartos et al., 2020), affordability (Arrieta et al., 2020), abundant availability (Dixit et al., 2021), low density and biodegradability (Jiang et al., 2021). Lignin, a varied and intricate polymer, is present in the cellular structures of several plant species, including wood, annual plants, and agricultural leftovers (Pei et al., 2023). The origins of lignin may be impacted by several factors, including the kind of LC material, plant species, genetics, and environmental circumstances (Vásquez-Garay et al., 2021).

The projected value of the global lignin market in 2023 was USD 1.08 billion, with a projected compound annual growth rate (CAGR) of 4.5 % from 2024 to 2030. Europe and North America dominate the markets for lignin-based dispersants because of their highly developed pulp and paper industries, which produce substantial amounts of lignin as a secondary product. In 2023, lingosulphonates had the highest market share in the United States, accounting for nearly 75.0 % of the demand. In 2023, Europe had the dominant position in the global lignin consumption market, accounting for more than 35 % of the total revenue. The demand is expected to increase due to regulatory restrictions that limit greenhouse gas emissions and the significant manufacturing base of biopolymers in France, Belgium, Germany, and the Netherlands (Lignin Market Size, Share And Growth Analysis Report, 2030, n.d.). Lignin is categorized as lignosulphonate (88 %) and Kraft lignin (KL) (9 %), with organosolv (2 %), a novel category gaining prominence for second-generation biofuels (bioethanol synthesis). The organosolv lignin category is predicted to expand the most from 2016 to 2025, at over 5 % (Bajwa et al., 2019). Currently around 70 million tons are derived from the paper and pulp sector. However, just 2 % of the whole 70 million tons is extracted and exploited for commercial purposes, whilst a majority of 98 % is incinerated as fuel (Covinich and Area, 2024). According to estimates (Fig. 1), the quantity of lignin that is now being processed via industrial processes is somewhere between 70 and 80 million tons on an annual basis. Furthermore, estimates suggest that an additional 730 Mton/year of lignin might be collected from stalks and straws as leftovers from agricultural streams. There is a need for less than half of this lignin to be returned to the land. It is estimated that only 0.075 Mt/year of hydrolysis lignin from biorefineries and soda lignin from the pulping of annual plants on a limited scale are generated and separated around the world (Tardy et al., 2023).

The most integrated biologically based biorefinery ideas include four main basic sections: biomass feedstock handling and preparations; biomass pretreatment; enzymatic hydrolysis and microbial fermentation; product recovery and purification. For an integrated biorefinery to effectively convert biomass into useful products and produce biofuels and biochemicals sustainably, several basic parts are essential (Dale and Ong, 2012). Improved processing methods and bioenergy crops with required chemical and physical qualities are needed to develop sustainable lignin-based polymers (Kocaturk et al., 2023). Thermochemical and homogeneous/heterogeneous catalysis can depolymerize and upgrade lignin for fuels and chemicals (Ramzan et al., 2023). Lignin multifunctionality has traditionally produced many product streams that need substantial separation and purification, but structuring plant feedstocks for structural homogeneity and customized functionality minimizes this difficulty (Zhou et al., 2022).

Conventional pretreatment methods require significant energy consumption and are not ecologically sustainable, resulting in the production of several unwanted chemicals and causing disposal problem. However, environmentally friendly techniques can effectively substitute the carbon footprint and expenses associated with traditional procedures, resulting in reduced emissions and environmental impact. This factor inspired the substitution of conventional, non-sustainable pretreatment with sustainable pretreatment known as “green or environmentally friendly” method (Hamada et al., 2023; Pereira et al., 2021b). Recent methods for isolating cellulose and lignin from lignocellulosic biomass are gaining increasing interest in research and development. Chemat et al. (2012) defined the green technology as “Green extraction is based on the discovery and design of extraction processes which will reduce energy consumption, allows use of alternative solvents and renewable natural products, and ensure a safe and high quality extract/product”. Pre-treatment of biomass may be achieved by the use of non-thermal plasma, primarily aiming to eliminate lignin from lignocellulosic material. An individual extraction of lignin, cellulose, and hemicellulose is made possible by this procedure. Each of these three components has the potential to be used as an important raw material in the manufacturing of high-value products (Pereira et al., 2021b).

LC pretreatment helps break down the complicated structure of LC biomass, which is mostly cellulose, hemicellulose, and lignin, to provide liquid fuel substitutes for petroleum (Garlock et al., 2011; Kammoun et al., 2023). In this review, several efficient environmentally friendly techniques such as organosolv treatment, supercritical fluids (SCF), non-thermal plasma (NTP), Ionic liquids (ILS), deep eutectic solvents (DES), and Microwave assisted extraction (MAE) techniques are described. Also describes the application of lignin where biobased manufacturing process elaborated. Also described its numerous applications in many

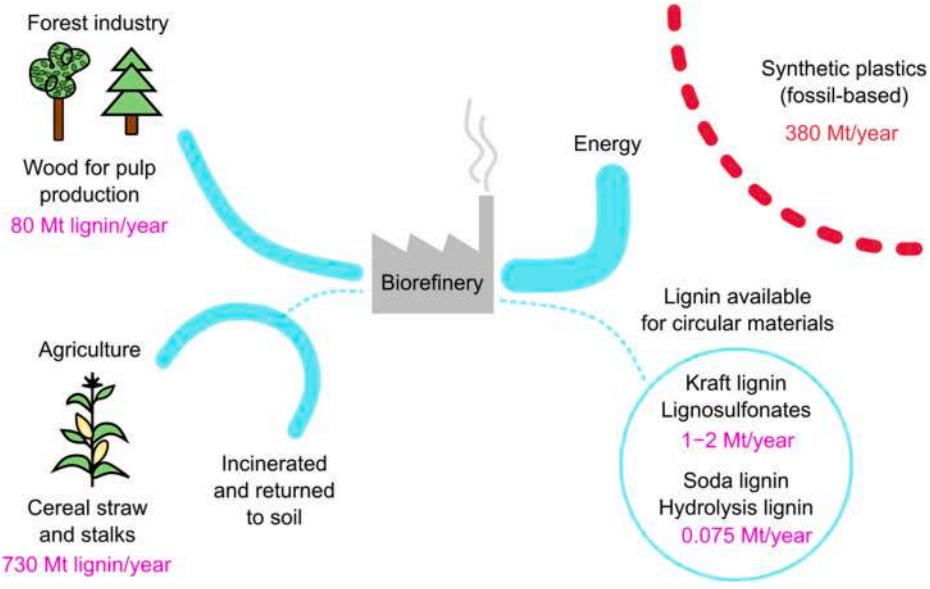

Fig. 1. Overview of annual lignin production and availability in isolated grades of various types and purities. Reused with the permission from (Tardy et al., 2023).

sectors due to its unique properties, such as textiles, food packaging, carbon fiber manufacture, polyurethane foam, phenolic resins, 3-D printing, and biomedical applications. Finally, the environmentally friendly processes of lignin extraction and application are discussed.

2. Chemical structure and properties of lignin

Lignin has a chemical structure that is very complicated due to the fact that it changes depending on the source and the purification process, resulting in the fact that the precise chemical structure of lignin is completely unknown (Wang et al., 2022b). Lignin, a high-molecular-weight polymer, contains phenolic hydroxyl, carboxyl, benzyl alcohol, and methoxy groups (Jin et al., 2021). The cell wall has a structure where phenolic chemicals and carbohydrates are strongly bonded together by covalent bonds (Ahmad et al., 2021). Lignin, acting as a cementing agent (Vásquez-Garay et al., 2021), provides the cell wall with mechanical strength, stiffness, and support for plant development (Mariana et al., 2021). The lignin backbone consists mostly of three distinct forms of phenyl propane monomer units, sometimes referred to as monolignols. The specific components of this natural polymer are *para*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol (Angelini et al., 2016). These components consist of p-hydroxyphenyl (H), guaiacyl (G), and syringyl (S) residues, respectively. Softwood lignin, mostly derived from gymnosperms, often consists of over 95 % G units. Hardwood lignin, mostly derived from angiosperms, consists predominantly of G and S units in variable proportions. In contrast, straw, grass, and other monocots are primarily made of H, G, and S units (Christensen and Rasmussen, 2019; Suota et al., 2021). Fig. 2 represents the chemical structure and combination of H, G, and S components in various plant categories.

The units are connected by several chemical bonds, such as ether linkages (β-O-4, 4-O-5, β-5), carbon–carbon connections (β-β, 5-5, β-1), and other interunit couplings (Gu et al., 2023). The existence of these connections adds to the intricacy and diversity of lignin's composition, rendering it difficult to analyze and incorporate into blends for the advancement of polymers and other substances (Lizundia et al., n.d.; Vásquez-Garay et al., 2021). The β-O-4 linkage is the most prevalent in natural lignin, accounting for 40–60 % of connections generated during biosynthesis (Wang et al., 2023a; Zhang et al., 2023). It is highly flexible and easily broken by pretreatment or depolymerization processes (Fabbri et al., 2023). Additional links that have a somewhat high occurrence include the 5–5 linkage (10–20 %) and b-5 linkage (10–12 %) (Wang and Deuss, 2023).

Lignin has several advantageous characteristics, including antioxidant and antibacterial properties (Sanchez et al., 2023),biodegradable and compatible properties (Gan et al., 2022), outstanding thermal reactivity (Fazeli et al., 2024), and adhesive capabilities (Mili et al., 2022). These traits render it a versatile chemical that holds significant financial benefits for companies.

3. Sources of lignin

The main sources of lignin comprise: Woody biomass, which is plentiful in trees and woody plants, including coniferous trees (such as pine, fir, and spruce) and hardwood trees (such as birch, aspen, eucalyptus, acacia, and oak). Non-woody biomass which includes agricultural byproducts, LC industrial and municipal wastes, as well as energy crops like switchgrass and Miscanthus. Additional sources of lignin include agricultural byproducts like coconut coir, straw from crops such as wheat, maize, and rice, as well as empty fruit bunches from palm trees (Abolore et al., 2024; Chen et al., 2020a). Lignin can also be obtained from municipal and industrial processes, such as byproducts from the food and beverage industry, brewery spent grains, pressed sugarcane bagasse, and papermaking sludge (Beluhan et al., 2023; Gbenebor et al., 2023). Table 1 displays the main components of several sources of LC biomass where the amounts of cellulose, hemicellulose, and lignin in different biomasses varied based on the origins of the biomasses (Abolore et al., 2024). Typically, it comprises around 30–50 % cellulose, 15–30 % hemicellulose, 15–30 % lignin, and 0–5 % other minor constituents (such as proteins, esters/fatty acids, and inorganic elements) based on its dry weight (Tanis et al., 2024). Lignin diverse sources underline its existence in many biomass forms, needing particular

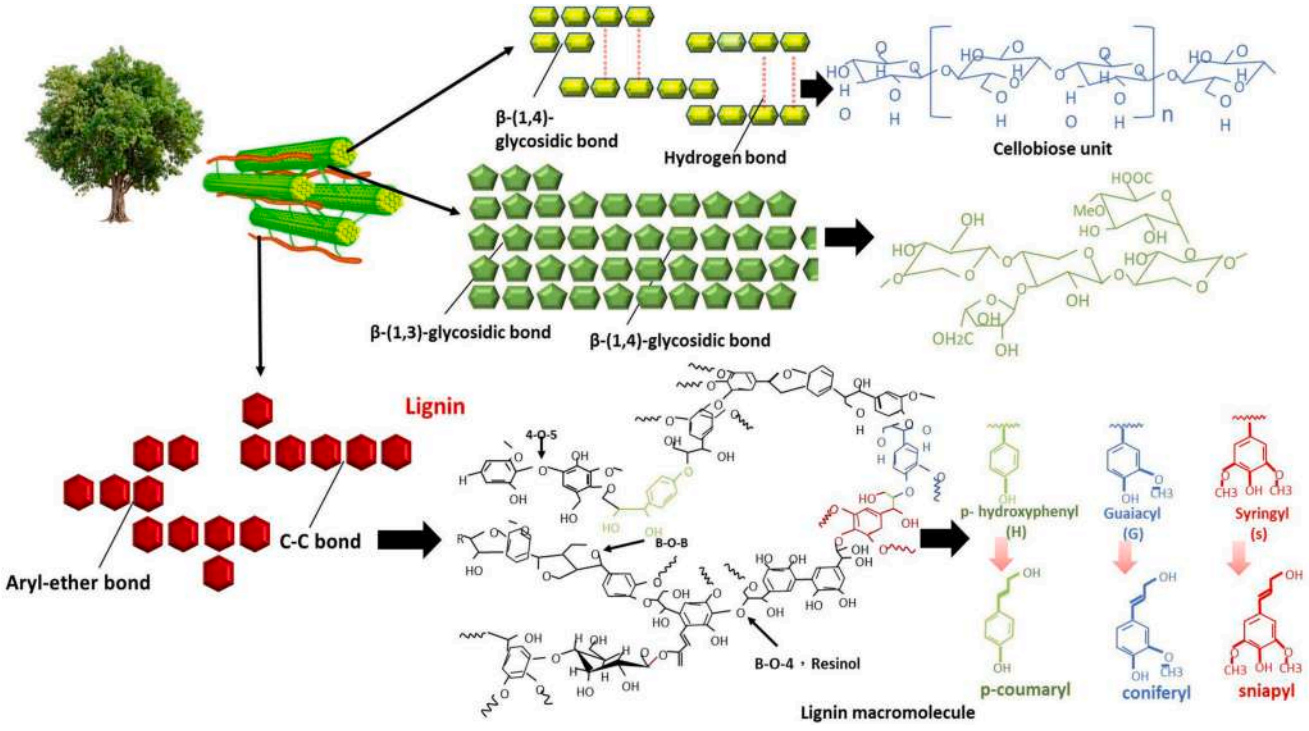

Fig. 2. The main connections and chemical structure, including the combination of H, G, and S components in various plant categories: Reused with permission from (Singhania et al., 2022).

Table 1
Lignocellulose composition of biomass derived from different sources.

Source	Type	Cellulose (%)	Hemicellulose (%)	Lignin (%)	References
Pine	Softwood	32	14	31	(Cotana et al., 2014)
Sugarcane bagasse	Herbaceous	44	28	21	(Ajala et al., 2021)
Olive tree	Hardwood	38	28	17	(Requejo et al., 2012)
Oil palm rachis	Softwood	38	21	16	(Solarte-Toro et al., 2018)
Rice straw	Herbaceous	37	38	22	(Oun and Rhim, 2016)
Wheat straw	Herbaceous	37	23	20	(Díez et al., 2020)
Oak tree	Hardwood	41	34	28	(Laskowska et al., 2018)
Poplar sawdust	Hardwood	48	16	27	(Bai et al., 2022)
Cotton	Herbaceous	94	2	3	(Xiao et al., 2024)
Jute	Herbaceous	67	14	13	(Ekundayo and Adejuyigbe, 2019)
Spruce wood	Softwood	39	35	24	(Čabalová et al., 2021)
Corn stover	Herbaceous	32	21	14	(Vergara et al., 2018)
Switchgrass	Herbaceous	45	31	12	(Khalid et al., 2017)
Tiger nut (*Cyperus esculentus* L)	Herbaceous	47	13	29	(Pelegrín et al., 2022)

attention in developing sustainable and efficient extraction methods (Boarino and Klok, 2023).

3.1. Woody biomass

Woody biomass comes from trees, sawmill leftovers, and forests. Woody biomass, the most abundant organic substance on Earth and renewable source of energy, contains cellulose, hemicellulose, and lignin depending on the tree type (Konstantinavičienė and Vitunskienė, 2023). Wood may be categorized into softwood and hardwood based on variations in plant cell structure and plant development characteristics (Stagno et al., 2022). Softwood and hardwood derive from coniferous and deciduous trees, respectively. Softwoods include pine, fir, and spruce, whereas hardwoods include birch, aspen, eucalyptus, acacia, and oak (Abolore et al., 2024). Hardwood fiber is shorter and finer than softwood fiber, which is the main difference. The lignin content in softwood ranges from 27 to 32 % including higher amount of extractives, such as resin (Tarasov et al., 2018). Hardwood is a kind of wood that typically has a less lignin content ranging from 18 to 25 % than softwood. The lignin content of agricultural biomass is quite low, typically ranging from 17 to 24 %. The use of lignin in plant fiber may be optimized at higher quantities following extraction, where some extractions techniques can more effectively remove lignin and provide a highly efficient conversion product from LC biomass (Xiao et al., 2024).

3.2. Non-woody biomass

Non-woody biomass sources of lignin including agricultural residues, LC industry, municipal waste, and energy crops (Blasi et al., 2023). Recent years have seen a rise in the number of individuals interested in the process of extracting lignin from non-wood biomass (Arni, 2018). Agricultural residues, such as straw obtained from the production of wheat, maize, and rice, are generated as byproducts of agricultural operations and operate as a significant source of non-woody biomass. These leftovers contain significant amounts of lignin, which may be extracted and used for various purposes (Kumar et al., 2023; Owonubi et al., 2021). An example of this is the use of lignin extracted from rice straw to produce bio-oil, which may be effectively used as a sustainable fuel or as a primary ingredient for the production of other chemicals and goods (Vu et al., 2022). LC industrial and municipal wastes, such as byproducts from the food and beverage sector, brewery spent grains, and papermaking sludge, also contain significant amounts of lignin.

These waste products may be used as raw materials for the extraction and utilization of lignin, therefore minimizing waste and fostering the sustainable utilization of resources (Vasić et al., 2023). An example of this is the use of lignin extracted from waste produced during the papermaking process to produce adhesives and composites that are sourced from biological sources (Zhong et al., 2024). Switchgrass and Miscanthus are cultivated as energy crops because of their capacity as non-woody biomass resources (Guragain et al., 2015). These crops contain significant amounts of lignin, which may be extracted and used for various purposes. An instance of using lignin generated from switchgrass is in the production of bio-based polymers and composites (van der Cruijsen et al., 2021). There are a few advantages associated with the use of lignin that is obtained from non-woody biomass sources. These advantages include cost-effectiveness, accessibility, adequate supply, and the reduction of waste via the recycling of materials that would otherwise be disposed through cremation or landfilling activities.

4. Pretreatment methods

Technical lignin may be classified into four separate kinds depending on the pulping technology used: kraft lignin (KL), lignosulfonate, soda, and organosolv lignin (OL) (Ejaz and Sohail, 2022; Ruwoldt et al., 2023). The process of extracting lignin often included the use of toxic chemicals, such as sulfuric acid and sodium hydroxide. These compounds have the potential to have negative impacts on the environment and need a significant amount of energy to be consumed (Brienza et al., 2024; Liu et al., 2022a). However, in recent years, there has been a growing interest in the development of environmentally safe and sustainable methods for extracting lignin. This is something that has gained a substantial amount of attention. When compared to conventional procedures, these environmentally friendly approaches to lignin extraction provide significant advantages in terms of sustainability, reduced effect on the environment, and increased capacity for energy efficiency (Kang et al., 2023; Sternberg et al., 2021). Typical green techniques used in the preparation of lignocellulose biomass include the use of organosolv (OS), supercritical fluid (SCF), Non-thermal plasma (NTP), ionic liquids (ILs), deep eutectic solvents (DESs), and MW assisted extraction (MAE) etc.

Researchers are developing a new generation of tailored solvents with improved properties, efficiency, and sustainability. OS pretreatment can make three separate product streams: a pulp that is high in cellulose, an aqueous fraction that is high in hemicellulose derivatives, and an organic fraction that is high in lignin derivatives (polyphenols). This is a significant advantage that organosolv pretreatment offers in comparison to other fractionation methods. This characteristic makes it possible to extract value from all of the elements of lignocellulose, provided that the pretreatment does not result in an excessive amount of degradation (Brienza et al., 2024). SCF extraction is another approach that is utilized for the separation of lignin from lignocellulose compounds. This method involves the process of separating one component (the extractant) from another component (the matrix) by employing supercritical fluids as the extracting solvent (Neata et al., 2015). Furthermore, NTP is a technology that is increasingly being investigated as a means of treatment and/or pre-treatment of lignocellulosic

materials. This NTP approach is regarded to be ecologically beneficial (Pereira et al., 2021b).

ILs have been used to dissolve polysaccharides (such as hemicelluloses and cellulose), lignin, and even all structural polymers at the same time. ILs provide many benefits, however, they do have drawbacks (Paulsen Thoresen et al., 2023). Furthermore, there has been a growing interest in DESs as biomass pretreatment alternatives to organic solvents. DESs are an eco-friendly way to separate lignin from LC biomass. However, DESs may be utilized as solvents, reaction media, or catalysts to convert lignin into useful compounds. The extracted lignin may be utilized to generate energy or make clever biomaterials. The potential applications of DES-extracted lignin in new fields remain uncertain (Lobato-Rodríguez et al., 2023). The term "microwave-assisted extraction" (MAE) refers to the use of microwave radiation to heat solvents and plant tissues during the pretreatment process in order to accelerate the kinetics of pretreatment.

Table 2 represents a short overview of environmentally friendly methods for lignin extraction with the uses of solvent and yield percent.

4.1. Conventional methods

4.1.1. Kraft process

The Kraft process is the most common way pulp and paper companies remove lignin from wood. Over 80 % of cellulosic pulp production worldwide employs Kraft process wood delignification (Jardim et al., 2020). The primary active agents in the Kraft process are sodium hydroxide and hydrosulfide anions, which operate at temperatures ranging from 150 to 170 °C (Hu et al., 2023). The Kraft process involves the removal of lignin from cellulose by breaking α-O-4 and β-O-4 bonds, leading to a delignification rate of over 90 % for the production of bleachable-grade cellulose (Abolore et al., 2024; Lahtinen et al., 2021). Breaking the β-O-4 ether connections leads to a substantial amount of phenolic hydroxyl groups in the recovered Kraft lignin (KL) (Wang et al., 2023a; Yue et al., 2023). During the pulping process, the large lignin molecules are broken down, resulting in a drop in molecular weight and exhibits variation from hardwoods and softwoods. The lignin is then dissolved in an alkaline solution, causing the solution to darken, and become brown (Chen, 2015). KL may be recovered by precipitating black liquor using mineral acids or carbon dioxide at a controlled pH. When the pH goes up, the phenolic hydroxyl groups in lignin become ionized. This makes the lignin dissolve in the lignocellulosic biomass. The extraction of Kraft lignin may be accomplished by either batch or continuous techniques, which are both widely used. The batch system primarily consists of a series of digesters that receive lignocellulosic resources and exchange liquors with the tank farm until they achieve pulp. The discharge tank receives the liquors after that. In a continuous system, the air is extracted from the lignocellulosic resource using chip steaming. The resource is then delivered to the digester using a liquid solution, and the chemical impregnation process takes place by applying temperature for cooking. This is followed by cleaning and unloading. This mechanism plays a crucial role in the depolymerization and repolymerization of lignocellulosic biomass (Lobato-Peralta et al., 2021a). Although the Kraft process has a high sulfur concentration, the recovered lignin only includes a residual sulfide of 1–2 % (Jardim et al., 2022; Yiamsawas et al., 2023). Typically, the lignin obtained from the Kraft process exhibits low purity and may include impurities such as sulfur compounds and sugars. However, this lignin may be subjected to further processing and modification to improve its quality for various applications, such as adhesives, binders, and biofuels (Vasile and Baican, 2023).

4.1.2. Lignosulfonate process

The lignosulfonic method is a major industrial source of lignin. Depending on the cationic content of the pulping/cooking liquid, pulping is done at pH 2–12. Sulfite pulping is the process of breaking down lignocellulosic biomass in a water-based solution that has salts from sulfurous acid in it. The temperature range for this process is 130–180 °C (Sutradhar and Fatehi, 2023; Vinod et al., 2023). Most lignosulfonic procedures use magnesium or calcium counterions and are acidic owing to the sulfonic group (Ruwoldt, 2020). Two methods, sulfonation and hydrolysis, are used during sulfite pulping to accomplish biomass delignification. Under acidic circumstances, bisulfite ions react as nucleophiles with benzylic carbenium ions, resulting in the creation of α-sulfonic acid structures. However, under neutral or basic circumstances, sulfonation only engages phenolic units, resulting in the

Table 2
Source of lignin, various environmentally friendly extraction methods, type of solvent and yield.

Biomass source	Extraction methods	Solvent type	Lignin yield (%)	Reference
Rice straw	DES	Triethylbenzyl ammoniumchloride/ lactic acid (LA)	24.40	(Lim et al., 2019)
Hazelnut skin	Organosolv process	Methanol	34.73	(Oliva et al., 2021)
Corn Stalk	NTP	Argon gas (Fenton process)	26.00	(Grbić et al., 2022)
Sugarcane bagasse	Organosolv process	Butylated Hydroxytoluene	45.28	(Schmatz et al., 2022)
Rice husks	DES	Choline chloride (ChCl)/ Formic acid (FA)	19.90	(Owhe et al., 2021)
Oil palm mesocarp fiber	MAE	γ-valerolactone	19.10	(Azlan et al., 2022)
sawmill chips (Spruce and pine)	DES	ChCl/LA	39.30	(Provost et al., 2022)
Sugarcane bagasse	DES	Triethylbenzyl ammonium chloride/LA	11.46	(Liu et al., 2021)
Corn stover	ILs	Tetra-butyl-phosphonium acetate	13.50	(Glińska et al., 2021)
Eucalyptus wood	Organosolv process	Glycerol (GLY)	65.00	(Romaní et al., 2016)
Switchgrass	DES	ChCl-GLY-PTSA (p-toluenesulfonic acid)	18.84	(Chen et al., 2019b)
eucalyptus fiber	Super critical fluid (SCF)	SC-CO_2, Ethanol and water	49.70	(Jiang et al., 2020)
Wheat straw	NTP	O_3 containing gas	14.90	(Schultz-Jensen et al., 2011)
Rice straw	SCF	SC-CO_2	5.80	(Gao et al., 2010)
Coconut coir	MAE-DES	ChCl/LA	82.00	(Mankar et al., 2022b)
Eucalyptus urophylla	ILS-MAE	1-ethyl-3-methylimidazolium acetate	45.80	(Sun et al., 2019)
Corn stover	DES	ChCl/AA	20.6.00	(Owhe et al., 2021)
Oak wood	MAE-DES	ChCl-FA	16.00	(Mattonai et al., 2022)
Banana peel	NTP	oxygen	48.00	(Pereira et al., 2023)
Raw ash tree	ILs	pyrrolidinium acetate	18.00	(Hasanov et al., 2022)
Brewer's spent grains	DES	ChCl/LA	54.40	(Cassoni et al., 2023)
Olive tree pruning	DES	ChCl/LA	37.80	(Cassoni et al., 2023)
Potato crop residues	Organosolv process	Ethanol	43.90	(Zolfaghari et al., 2024)
Cocoa bean shells	DES-MAE	p-toluenesulfonic acid, choline chloride and glycerol	95.5	(Mao et al., 2023)

formation of quinone methides. Being around sulfite ions causes quinone methides to be attacked by nucleophiles, which makes α-sulfonic acids. In addition, sulfonation can happen at other connections between units, which adds β- and γ-sulfonic groups to lignin units. This can happen in both acidic and alkaline environments. Sulfonation adds polar sulfonate groups to lignin structures, which enhances the solubility of lignin in water. Hydrolysis, on the other hand, breaks down the carbohydrate contents and intermolecular ether connections (namely β-O-4′ and α-O-4′ bonds) between lignin units (Brienza et al., 2024). In general, sulfite pulping has progressively declined in popularity as a pulping technique because of the relatively weak strength of the resulting pulps(Shevchenko et al., 2023). Additional drawbacks include of the significant deterioration of hemicellulose and the poor nature of the resulting lignin streams (lignosulfonates), which are highly condensed and tainted with sulfur (3–8 %), carbohydrates, and other impurities (such as minerals and extractives) (Margarida Martins et al., 2022; Ojo, 2023). One of the most renowned examples of lignin valorization on an industrial scale is the use of lignosulfonates, which currently account for over 90 % of the entire market for commercial lignin.

4.1.3. Soda pulping process

The soda pulping process is a procedure that is used for the purpose of chemically pulping a variety of non-wood species, including bagasse, wheat straw, hemp, kenaf, and sisal (Margarida Martins et al., 2022). Using a solution of sodium hydroxide (NaOH) as the cooking solvent, the lignin is extracted from the plant material. This process is known as plant material extraction (Shah et al., 2023). This approach is generally recognized as the first chemical technique for extracting lignin. The method utilizes alkaline hydrolysis, namely sodium hydroxide at concentrations ranging from 13–16 wt%. The process is carried out in pressurized reactors at temperatures ranging from 140 °C–170 °C. The goal is to cleave the chemical bonds of α-aryl ether inside the phenolic units of lignin. Lignin may be extracted from the liquor by adding sulphuric acid until the black liquor reaches a pH of around 2–3. The lignin is recovered from the liquor using a basic centrifuge or filter process. Furthermore, it is possible to extract the lignin using precipitation. Alternatively, it is possible for the in-situ silica to precipitate concurrently with the lignin (Lobato-Peralta et al., 2021a).

Therefore, in contrast to the Kraft process, the soda pulping method does not include the incorporation of sodium sulfide (Na_2S) into the alkali solution. When it comes to the degradation of lignin, the soda pulping approach is not as effective as the Kraft process since it does not include a powerful nucleophile on its own (Fernández-Rodríguez et al., 2019). Soda lignin, on the other hand, undergoes very minor structural modifications during the recovery process and may be used without the need for purification. Because of its characteristics, soda lignin is an excellent candidate for chemical modification, which may then be used in a variety of applications that add value (Komisarz et al., 2023; Lobato-Peralta et al., 2021a). One significant benefit of soda pulping is the generation of sulfur-free lignin, which may be extracted during precipitation. Compared to KL or lignosulfonates, soda lignin is a more appealing raw material. However, the significant level of condensation remains a barrier to the improvement of soda lignin (Brienza et al., 2024).

4.1.4. Alkaline treatment

Alkaline pretreatment is a widely used and economical procedure for treating lignocellulosic biomass. It involves using an alkaline reagent to dissolve lignin and hemicellulose in the biomass. The swelling reaction in the cell wall is caused by alkaline pretreatment, which is done in order to increase the internal surface area of the cellulose, as well as to reduce the degree of polymerization and crystallinity of the cellulose. The dissolution of lignin and hemicellulose occurs as a result of the decomposition of the ester bonds that exist between the molecules of lignin and hemicellulose (Hernández-Beltrán et al., 2019; Oriez et al., 2020). The dissolution of lignin and hemicellulose, as well as the de-esterification of intermolecular connections, are two of the essential activities that are included in the alkaline pretreatment process. The alkaline pretreatment often use chemicals such as sodium hydroxide (NaOH), sodium carbonate (Na_2CO_3), potassium hydroxide (KOH), and ammonium hydroxide (NH_4OH), these activities take place under mild conditions. The effectiveness of the process is significantly influenced by a number of factors, including the proportion of solid to liquid, temperature, pressure, and length of time spent with the substance. Moreover, instead of employing higher concentrations of the alkaline reagent, this method can operate at room temperature and atmospheric pressure, to avoid breakdown and degradation of polysaccharides (Tanis et al., 2024). Nevertheless, NaOH is widely recognized as the most effective alkali for the pre-treatment of lignocellulosic biomass. Additionally, it is the most often used alkali for the purpose of dissolving lignin and hemicellulose. In addition, sodium hydroxide is readily available and affordable, has a high degree of alkalinity, and can be managed with relative simplicity. These characteristics make it an attractive choice for widespread industrial applications. It is often used in combination with other chemicals and techniques to enhance the efficiency of pretreatment. Several experiments have shown the efficacy of NaOH in cellulose pretreatment (Abolore et al., 2024). The lignin obtained by alkaline fractionation does not include sulfur, unlike the lignin generated through Kraft and sulfite pulping methods. This lack of sulfur is very beneficial for subsequent chemical activation procedures, as it allows for various valorization paths, such as the production of fuel additives or bio-based polymers (Oriez et al., 2020). Although, the alkaline pretreatment of lignocellulose biomass is efficient, it has several limits. One of the drawbacks of using alkaline for lignin removal is its lack of selectivity, which results in the removal of not just lignin but also certain cellulose and hemicellulose components. In addition, it necessitates the use of strong alkaline substances such as NaOH or NH_4OH, the creation, retention, and elimination of which could result in environmental concerns. Moreover, the alkaline treatment and high pH conditions can have a negative impact on the stability and effectiveness of cellulase enzymes used in the hydrolysis process. Additionally, the alkaline treatment can lead to the formation of inhibitory compounds like phenolics, furans, and organic acids derived from lignin (Abolore et al., 2024).

4.1.5. Acid treatment

Acid pretreatment is a commonly used method for altering the lignocellulosic structure by breaking glycosidic linkages, which leads to the conversion of polysaccharides into smaller sugar molecules (Hernández-Beltrán et al., 2019). This approach mainly focuses on selectively extracting the hemicellulose fraction while simultaneously eliminating the acid-soluble lignin component. In addition, it breaks down cellulose into cello-oligosaccharides. Organic acids, such as formic acid or acetic acid, and inorganic acids, such as nitric acid and sulfuric acid, are used in acid pretreatment (Tanis et al., 2024). The primary choice for pretreating various types of lignocellulosic biomass (wood and agriculture waste) is dilute sulfuric acid, primarily owing to its effectiveness in dissolving lignin and hemicellulose. The use of organic acids in biomass pretreatment is growing due to their high efficacy in breaking down lignocellulosic biomass, resulting in less degradation byproducts and a higher yield of oligomeric sugars. Acetic acid pretreatment has the benefit of dissolving a greater amount of lignin compared to hot water pretreatment and other acid pretreatment methods. Acetic acid has been shown to function as a solvent for lignin in the treatment process, resulting in the formation of acetic acid lignin (Chen et al., 2020b). The treatment may be conducted in either a weak acid solution (0.1–10 %) at elevated temperatures (100–250 °C) or in a strong acid solution (30–70 %) at lower temperatures (<100 °C). The intensity of the pretreatment can be quantified using a severity factor (Brienza et al., 2024). The acid pretreatment method has many benefits over other pretreatment procedures in terms of disrupting the

lignocellulosic matrix and converting amorphous cellulose. However, this pretreatment method also has several drawbacks related to the creation of inhibitory substances and the manufacture of wastewater with a significant potential for acidification, aquatic toxicity, and human toxicity via consumption or exposure (Solarte-Toro et al., 2019). So that Concentrated acid pretreatment have significant downsides, including strict safety and equipment requirements, as well as the generation of a substantial quantity of neutralization waste. Typically, using weak acid solutions is a preferable choice for preparing lignocellulose on a large scale in industry. This method significantly enhances the ability of enzymes to break down the biomass, while also being more cost-effective and ecologically benign (Baruah et al., 2018).

4.2. Environmentally friendly methods

4.2.1. Organosolv process

Recently, advanced biorefinery techniques using organosolv fractionation have been developed to extract cellulose, lignin, and hemicellulosic carbohydrates from lignocellulosic biomass (Zhang et al., 2022). Organosolv pretreatment uses water and organic solvents such as methanol, ethanol, acetone, ethylene glycol, and triethylene glycol, with or without a catalytic agent (Beluhan et al., 2023; Broda et al., 2022). Pretreatment helps break down cellulose, remove hemicellulosic materials, and produce hydrolysate with less compounds from sugar and lignin breakdown (Nair et al., 2023). Fig. 3 illustrates the process by which lignocellulose is broken down in the organosolv method. This method removes lignin by breaking α and β-O-aryl ether bonds and eliminating most hemicellulose carbohydrates. After hydrolysis breaks lignin bonds and lignin-carbohydrate linkages, cellulose and a tiny quantity of hemicellulose form a solid residue. Generating lignin is usually pure, high-quality, and less condensed than KL (Schmatz et al., 2022). This method avoids the use of hazardous chemicals and extreme working conditions, such as those found in the kraft and sulfite processes (Vaidya et al., 2022). Organosolv lignin (OSL) has an advantage over KL because it does not include sulfur, which forms unpleasant organosulfur compounds that deactivate subsequent catalysts. The technology to turn sulfur-free high-quality lignin into usable goods is absent (Bhattacharyya et al., 2020; Latham et al., 2021). The direct separation of hemicellulose without the need for delignification is one of the most advantageous aspects of organosolv extraction. Because the acetyl groups that are present in the cell walls of LC biomass are not converted to acetic acid and subsequently shed off, the structure of hemicellulose is maintained in a satisfactory manner (Lu et al., 2021).

In a conventional organosolv method, lignocellulosic biomass is subjected to high temperatures while being exposed to an appropriate organic solvent. This enables the lignin to maintain its original structure. Remarkably, the pace of pulping may be expedited by using an appropriate acid or alkali during this procedure. Lignin may be precipitated from black liquor by introducing acidified water, followed by centrifugation of the liquor. The precipitated lignin is then separated, rinsed with acidified water, and dried in an oven (Prado et al., 2013). Subsequently, the solvent may be recovered using several methods such as water washing and subsequent distillation, leading to the isolation of pure lignin as a solid residue. The potential explanation for the separation of pure lignin may be attributed to its hydrophobic nature, which renders it insoluble in water and so prevents it from being washed away. Due to the preservation of its molecular structure, lignin generated from this technique has significant potential to serve as a feedstock for biorefineries in the production of valuable phenolic and aromatic compounds (Ahmad and Pant, 2018, p. 14). Researchers have examined the use of Lewis acids as catalysts in the organosolv treatment of wheat straw. The lignocellulosic biomass has been fractionated and the lignin has been fragmented using aqueous ethanol in the presence of $FeCl_2$, $CuCl_2$, $FeCl_3$, $ZrOCl_2$. The level of delignification and the amount of KL obtained increased in proportion to the strength of the Lewis acid. The presence of a suitable catalyst, such as mineral acids and ethanol, favors the organosolv process, resulting in the production of antioxidant chemicals (Constant et al., 2015). Acetic acid pretreatment is often conducted at elevated temperatures ranging from 135 to 200 °C, with relatively short retention durations of 30–120 min. According to reports, the temperature range in the procedure may be decreased to 110 °C by adding sulfuric acid (Wei Kit ChinChin et al., 2020). Typically, oxalic acid has been used as a substitute for sulfuric acid. It is also used as an acid catalyst in pretreatment. The process is ecologically beneficial for generating. Extremely low levels of substances that hinder fermentation. It is also recognized to possess superior catalytic efficiency compared to sulfuric acid (Nair et al., 2023).The organosolv procedure dissolves lignin via acid-catalyzed solvolysis of β-ether bonds. In situ hemicellulose deacetylation releases acetic acid in the reaction mixture, or it is added (HCl, H_2SO_4, oxalic acid, formic acid, and acetic acid) (Brienza et al., 2024; Liu et al., 2022b; Luo and Abu-Omar, 2017). The external acid enhances β-ether acidolysis, resulting in decreased lignin

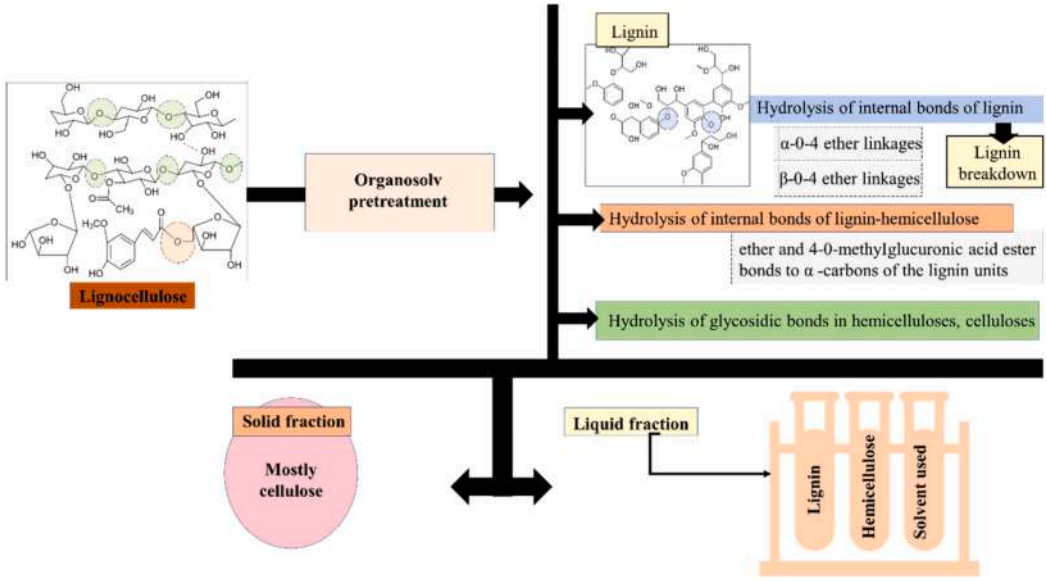

Fig. 3. The mechanism and process of organosolv pretreatment on lignocellulosic biomass: Adopted from (Nair et al., 2023).

concentration in cellulose pulp. In contrast, in situ acid production lignin may retain more β-ether connections than native lignin (Chen et al., 2023; Sun et al., 2018). Mou and Wu (2016) examined eucalyptus that underwent the organosolv process, where a combination of water and ethanol with a ratio of 60:40 (w/w) was used, and the L/S ratio was 6:1. The lignin concentration was found to be 22.1 % after the organosolv treatment was carried out at a temperature of 160 °C for a period of 90 min. The study demonstrated that organosolv pretreatment was a very effective method for removing lignin from the cell wall and surface of the fiber. Schmatz et al. (Schmatz and Brienzo, 2022) conducted research on organosolv pretreatment at a temperature of 121 °C, using butylated hydroxytoluene (BHT) and ethanol. The results demonstrated significant effectiveness in removing lignin from biomass. When the ethanol concentration is 50 %, the use of BHT results in a more significant elimination of lignin in comparison to not using BHT. This discovery implies that BHT might serve as an alternative technique for extracting lignin, hence facilitating the production of important substances such as resins and phenols.

Researchers studied the efficacy of chemical compounds, such as antioxidants and surfactants, was assessed in the elimination of the lignin component from sugarcane bagasse. The additions did not cause any disruption in the process of hemicellulose solubilization, which was identical to the organosolv pretreatment without any additives. The use of antioxidants shown an effect on the elimination of lignin, compared to the organosolv pretreatment without any addition. This surfactant has the ability to provide advantages and facilitate biotechnological processes. Additionally, it has antioxidant properties that aid in the elimination of lignin. Produce raw materials as alternatives to petroleum derivatives, namely for the production of resins and biomaterials (Schmatz et al., 2022).

The solvents used in an organosolv technique are much more expensive than the chemicals used in traditional pulping methods. Prior research assessed the effectiveness of combining an organosolv technique with membrane filtering. The study found that the manufacturing cost of the resulting lignin was around 52 €/ton, which exceeded the cost of KL (33 €/ton). A separate study evaluated the economic and environmental consequences of various lignin extraction techniques. It was determined that the production costs and potential environmental effects of organosolv extraction were greater compared to alternative methods such as kraft extraction, lignosulfonate extraction, and soda extraction (Karagoz et al., 2023).

Organosolv approach offers many benefits, including the delicate extraction of an exceptionally pure lignin fraction and the easy recovery of the solvent. Due to the very high cost of the procedure, it is less feasible in the sectors that it is targeted for.

4.2.2. *Supercritical fluid methods*

The development of an eco-friendly alternative technique with low ecological consequences is crucial to produce targeted goods derived from lignin. Thus, energy efficiency, lignin conversion, and fewer hazardous compounds must be prioritized. Supercritical solvent-based lignin depolymerization technologies limit condensation and are ecologically beneficial as the less time required for sample preparation, improved extraction efficiency and speed (Roy et al., 2022). A combination of very high temperatures and pressures is required for the formation of subcritical and supercritical fluids (da Silva et al., 2016). The structure of the lignin may be penetrated by supercritical fluids, which have special qualities such as low viscosity, low dielectric constant, and high diffusivity. These properties allow the supercritical fluids to readily dissolve the depolymerized products (Escobar et al., 2020; Gosselink et al., 2011). Supercritical procedures have been used since the 1990s, and during their early implementation, KL and OL were subjected to treatment with supercritical methanol (Yong and Yukihiko, 2013). Various kinds of subcritical and supercritical fluids, including water, ethanol, and carbon dioxide, have been used in this experimental setup (Roy et al., 2022). Depolymerisation activities in the lignin processing industry have used subcritical and supercritical fluids (Nardella et al., 2023; Roy et al., 2022). A supercritical fluid is a substance that is exposed to temperatures and pressures that exceed its critical point. Carbon dioxide (CO_2) is the preferred supercritical fluid for pretreating lignocellulose biomass because of its low critical temperature (31.1 °C) and pressure (73.8 bar), which make it easily accessible and safer to use. CO_2 has characteristics of both a gas and a liquid in its supercritical form, making it very effective as a solvent for supercritical fluid extraction (Abolore et al., 2024). For the lignin extraction, the process involves placing the ground biomass into a specialized extraction tank, designed to withstand high pressure and temperature. The process of compression uses supercritical carbon dioxide to subject the extraction vessel to a specific pressure and temperature higher than its critical point. This step involves subjecting the biomass to supercritical carbon dioxide, thereby simplifying the lignin extraction process. Co-solvents or modifiers can increase the solubility of lignin in supercritical carbon dioxide. Depressurization or other separation techniques separate the extracted lignin from the supercritical carbon dioxide (Srinivasan and Ju, 2010). Fig. 4 depicts the basic process for the extraction of Lignin via SFE and CXLE.

Yong and Matsumura (2012) found that the rapid transformation of lignin and the full depolymerization process took occurred within five seconds of the lignin's residence period when using supercritical water conditions. Additionally, the findings of the research indicated that the production of phenolic compounds took place within a short period of residence time. This occurs due to the fact that the ether bonds found in lignin are readily broken down when subjected to supercritical conditions. Later, Machmudah et al. (2015) showed that using hot compressed water in a continuous flow reactor, lignin was extracted from Japanese rice straw at 170–230 °C and 1.42–4.67 mL/min under 4 MPa pressure for 60 min. Subcritical water during the chemical phase caused heteroatom-carbon bond breakage and hydrolysis. This study found that greater temperatures and flow rates increased lignin recovery. At 170 °C and 200 °C and 4.67 mL/min, Japanese rice straw lignin removal reached 85 %.

By increasing the flowrate, dissolved lignin was extracted from the reactor without reacting or precipitating. Lignin recovery improved with this improvement. This shows how fast subcritical reactions happen. Lignin recovery dropped to 64 % at 230 °C under the same conditions. Ether and C-C bonds break at high temperatures, reducing lignin recovery. Hydrothermal degradation of these linkages produced phenols and methoxy phenols.

SC-CO_2 has attracted significant attention in comparison to other supercritical solvents owing to its distinctive features, such as its designation as an environmentally friendly and non-polluting solvent. It is non-flammable, has reduced thickness, lower polarity, is non-toxic, promotes particle dispersion, and is chemically inert. The chemical may be easily recovered from the reaction solution by simply decreasing the pressure, thereby minimizing the need for further processing

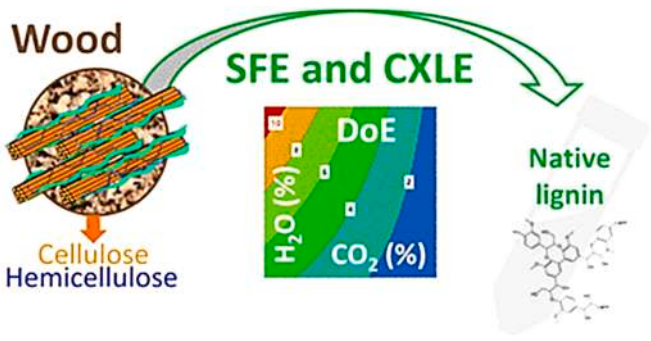

Fig. 4. Basic process for the extraction of Lignin via SFE and CXLE: Reproduced from (Nardella et al., 2023) with permission from the Royal Society of Chemistry.

downstream. During the pre-treatment processes, it is of the utmost importance to investigate the best conditions that influence the lignin content, cellulose, and hemi-cellulose hydration. Using SC-CO_2 as a pre-treatment for biomass not only results in a large increase in glucose production, but it also effectively eliminates lignin, breaks down hemicellulose, and extracts a wide variety of compounds from the biomass (Badgujar et al., 2021). Although sub- and supercritical technology has showed promise for extracting and isolating lignin, it must be improved for greater quality and usability.

Neata et al. (2015) shows that the supercritical fluid extraction with CO_2 to reduce lignin content by 85 %. The authors used also simplifies solvent recovery, reduces pressure drops, and reduces mass-transfer impediment compared to traditional liquid extraction. Modifiers may boost nonpolar supercritical CO_2 selectivity, allowing a wide range of solvent power modifications. Furthermore, Yang et al. (2022) showed that SC-CO_2 was increased the solubility of soluble compounds, break lignin ether linkages by delivering hydrogen ions, and chemically bind ethanol to lignin particle fragments in the SC-CO_2 ethanol solution (SCES) method. The SCES approach yielded optimal results at 206.63 ℃, 10.29 MPa pressure, and 58 % ethanol content ratio, resulting 75.41 % lignin extraction rate.

The study demonstrated that the SCES technique produced lignin with higher antioxidant activity with SCES-extracted lignin might be a natural antioxidant. Later, Nardella et al. (2023) highlighted the effectiveness of supercritical fluid extraction and CO_2-expanded liquid extraction, both of which make use of organic co-solvents that are favorable to the environment. The primary objective of the research was to find ways to improve the extraction procedure in order to acquire substantial quantities of lignin monomers and oligomers that are viable for extraction. In another study, Ho Seo et al. (2019) used four organic solvents to separate crude supercritical lignin via supercritical water oxidation from hardwood, resulting in five lignin fractions (F1 − F4 and F_{IN}). The research examined supercritical lignin's molecular weight distribution, separation, and characterization. This involved studying fractionated lignin functional groups. The findings provide valuable insights on fractionated lignin' molecular weight and functional groups during fractionation. According to authors, these discoveries may enhance lignin-based biomaterials and biochemicals by using woody biomass lignin.

According to Daza Serna et al. (2016) studies, the reuse of solvents, such as carbon dioxide and water–ethanol, in the supercritical pre-treatment process is not particularly challenging. The supercritical pretreatment process incurs higher total utility costs, mostly because of the demanding operating conditions, such as high pressures and the energy needed for carbon dioxide liquefaction. Supercritical pretreatment offers optimal economic and environmental performance. Nevertheless, several limitations must be overcome, such as minimizing residual energy use in order to save expenses. The viability of using carbon dioxide as a green solvent for continuous processing of biomass by supercritical pretreatment is dependent upon the outcomes seen for various lignocellulosic residues that possess distinct composition characteristics. As a result, it becomes possible to comprehend the genuine potential of this pre-treatment in the future. Scientists are searching methods to enhance the separation of lignin using SC-CO_2. This includes using co-solvents or surfactants to increase lignin solubility, fractionation techniques to generate fractions with different properties, and process parameter effects on yield and purity. The molecular interactions between SC-CO_2 and lignin are also being studied to better regulate the separation process. In the future, there will be an increased use of SC-CO_2 for the purpose of separating lignin. The demand for sustainable materials and chemicals and the desire to reduce fossil fuel use are driving LC biomass technology development. Using SC-CO_2 to separate lignin is intriguing due to its eco-friendliness and ability to produce high-quality lignin for many uses.

4.2.3. Non-thermal plasma treatment

Plasma technology has recently been investigated as an alternate method for pretreating biomass in a biorefining process (Meoli et al., 2023). Plasma technology works on a fundamental physical concept. When energy is applied to matter, solids become liquids and liquids become gases. Gases become plasma, the fourth state of matter, when given greater energy. Plasma technology may be thermal or non-thermal (Merche et al., 2012). Establishing thermodynamic equilibrium among generated organisms differentiates them. All organisms in thermal or hot plasma have thermal equilibrium temperatures. In non-thermal plasma (NTP), heavy species and electrons are not thermally balanced. Since the gas temperature is low enough to touch, it is sometimes called NTP or cold plasma. In cold plasma, electrons absorb energy as in heat plasma. However, low pressure reduces collisions, preventing electrons from transferring energy to heavier species. Thus, electrons remain hotter than other species (Wu et al., 2013). Plasma has a notable ability to cleave the C-C and C-H bonds in organic compounds (Lim and Zulkifli, 2018), while also having a strong impact on guaiacol in the results of biomass pyrolysis. The use of NTP may enhance the process of biomass pretreatment. The process of extracting lignin from LC materials results in the formation of "holocellulose," a substance abundant in cellulose and hemicellulose. Each component may then be used to produce valuable goods. Fig. 5 presents a schematic illustrating the potential degradation pathway of lignocellulosic biomass using NTP. The process of NTP for the extraction of lignin involves a synergistic interaction between direct reactions, indirect oxidation, the production of reactive species, changes in the structure of the biomass, and enhanced enzymatic hydrolysis. Through the combination of these actions, the components of lignin are broken down in a synergistic manner, which assists in the extraction of these components from lignocellulosic biomass (Pereira et al., 2021b).

Lignin may undergo thermal conversion to provide valuable commodities, including activated carbon and chemical compounds like vanillin. Additionally, the portion that consists of cellulose and hemicellulose may also be used to acquire items of significant value (Meoli et al., 2023; Shao et al., 2022). Pereira et al. (2021a) experimented with NTP resulted in a significant reduction in lignin content, attaining about 50 % delignification during a 5-minute pretreatment duration while the kinetic experiments showed that a significant reduction in both soluble and insoluble lignin during the first 2.5 min of the process. This demonstrates the effectiveness of NTP as a better approach for using lignin. However, longer periods of treatment did not result in higher removal of lignin, suggesting that shorter treatment durations are more beneficial for the delignification process while still preserving the integrity of the sugar components. Later, Grbic´et al. (Grbić et al., 2022) showed that during the treatment of biomass with NTP, the quantity of lignin in the biomass was decreased. They showed that when NTP treatment combined with the Fenton reagent, has shown remarkable success in removing large amounts of lignin from LC substrates, such as maize stalks and discarded coffee waste, which was a noteworthy achievement. They suggested that in order to obtain the desired outcome of separating lignin while minimizing the impacts on other components of the biomass, it is essential to take into consideration the specific conditions of the NTP treatment whereas the ratio of biomass to Fe_2^+ to H_2O_2 is an important factor to take into consideration.

NTP is a favourable, environmentally friendly and renewable technique that operates at low temperatures and air pressure and may reduce the resistance of lignocellulose biomass by eliminating lignin (Qin and Li, 2023; Shao et al., 2022). It is reasonably easy to use and has minimal running costs. The NTP process generates reactive species and concurrently triggers chemical activities, including ionization, dissociation, and excitation. It also induces physical processes like as electric fields, UV radiation, and shock waves (Pereira et al., 2021b).

Plasma that does not thermally change is ecologically friendly since it may function at regular temperatures and pressures without harsh chemicals. Furthermore, this emerging technology has several

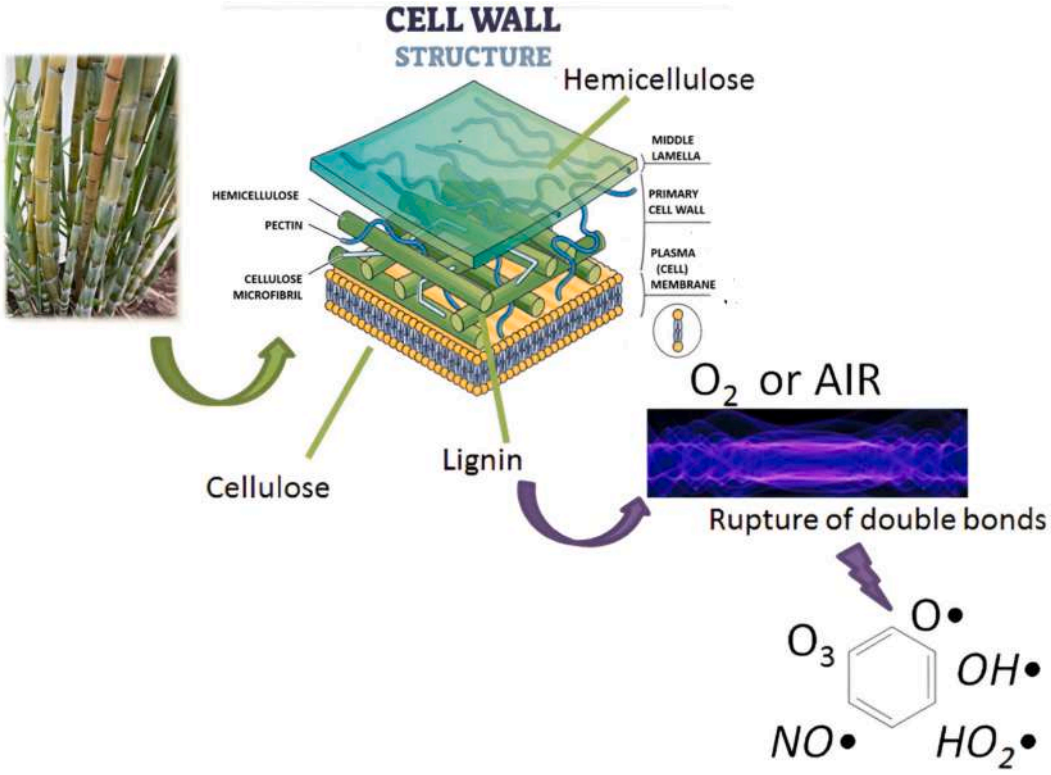

Fig.5. A potential process for the breakdown of lignocellulosic biomass using NTP: Adopted from Processes, MDPI 2023 (Meoli et al., 2023).

advantages, including a short processing time, friendly processing conditions, no chemical waste formation, and no fermentation inhibitors (Pereira et al., 2021b). This quality is consistent with the principles of green chemistry and sustainable manufacturing. Even though NTP shows potential for promising results in the delignification process relates to a number of possible problems such as initial investment, energy consumption, process optimization, scale–up challenges, and safety considerations. Therefore, researchers are still developing NTP processes with lab- or pilot-size installations. Researchers need to conduct further research to fully understand and evaluate the technology. As it scales up, energy efficiency challenges arise, necessitating the prediction of energy losses and the management of oxidizing species formation to control the energy demand for treating a specific feedstock (Meoli et al., 2023). It is possible that current research and development activities may be able to overcome some of these problems and further improve the feasibility and efficiency of NTP for delignification operations.

4.2.4. Ionic liquid method

Ionic liquids (ILs) are organic salts having a melting point below 100 °C that are often composed of an organic cation (such as imidazolium, pyridinium, aliphatic ammonium, alkylated phosphonium, and sulfonium ions) and a corresponding inorganic or organic anion. These liquids have arisen as a very environmentally friendly option for the pretreatment of biomass (Margarida Martins et al., 2022). One factor that leads to the broad identification of ILs is the fact that they may function as both solvents and reagents (Norfarhana et al., 2024). ILs exhibit characteristics such as low vapor pressures (Hasanov et al., 2020), high thermal and chemical stability (Azimi et al., 2022), powerful dissolving capabilities (Norfarhana et al., 2024), non-flammability (Abushammala and Mao, 2020), and non-toxicity (Yin et al., 2021). They may adjust to process needs by changing their anions and cations, resulting in variable characteristics (Eqbalpour et al., 2023). These solvents disintegrate biomass entirely (Acosta et al., 2014). ILs may selectively dissolve biomass components like cellulose, hemicellulose, and lignin depending on the IL. An anti-solvent or acidification may remove dissolved lignin from the liquid stream for high purity recovery (Bernardo et al., 2019).

Currently, two distinct methods are being explored for the treatment of LC biomass utilizing ILs. An alternative method is the disruption of the lignocellulose composite, namely the crystalline cellulose, by a process known as dissolution pretreatment. This technique is obtained by directly dissolving cellulose using ionic solutions. The second, more modern, method employs ILs to extract lignin and hemicellulose from lignocellulose while leaving cellulose as a solid that can be filtered. This process is referred to as ionosolv pretreatment. This procedure is similar to organosolv processing; however, it takes place at standard atmospheric pressure. IL-based pretreatment offers several distinct benefits in comparison to conventional pretreatment procedures. These factors include the use of reduced process pressures, diminished friction and abrasion, and the capability to accomplish inventive product separations (Brandt-Talbot et al., 2017).

In recent years, research has shown that some ILs are effective for delignifying LC biomass. They are LC fractionation solvents because of this. ILs dissolve LC biomass, increasing access to lignin and perhaps valorizing it. Different techniques may be used during or after IL-aided fractionation to create fuels, chemicals, and materials. Lignin extraction from lignocelluloses, catalytic delignification from biomass, and saccharification of polysaccharides connected to lignin pieces are examples. Delignification disrupts lignin structure, causing biomass swelling, which is impossible for natural lignin (Hasanov et al., 2020). Fig. 6 shows the ILs-MAE Process, which was used for the extraction of lignin and the dissolution of biomass. ILs have a unique capability to dissolve LC biomass, hence enhancing the availability of lignin inside the biomass structure. ILs use this procedure to remove lignin from lignocellulosic biomass. ILs are specifically engineered to exploit this characteristic. By dissolving the biomass in ILs, the lignin molecules are extracted and exposed, allowing for further processing. The contact between the IL and lignin molecules causes a disruption in the structure of lignin. Due to this disruption, the intermolecular connections that

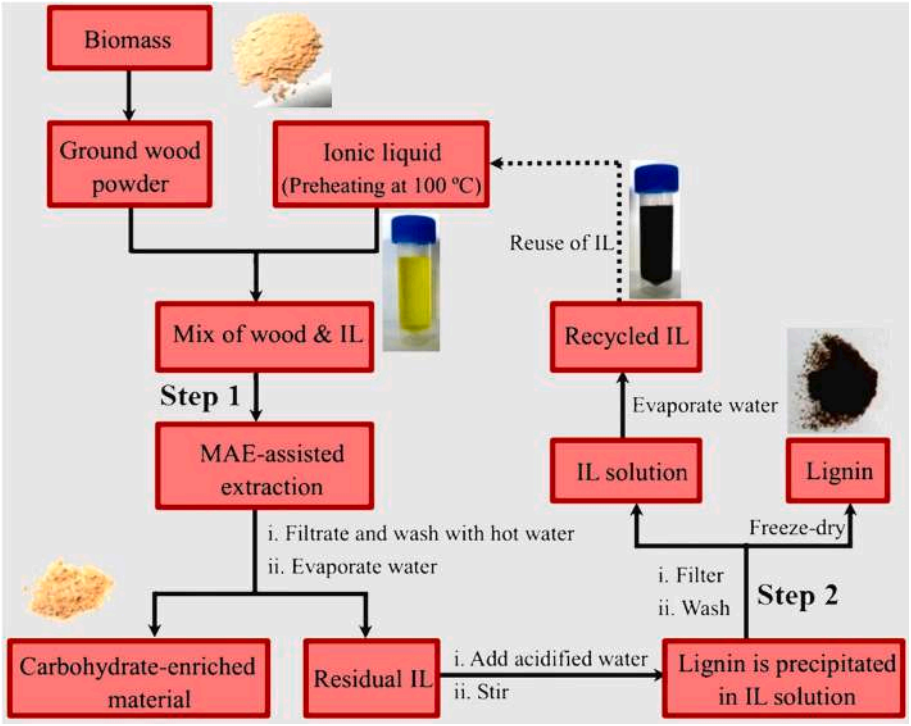

Fig 6. The ILs-MAE Process, which is used for the extraction of lignin and the dissolution of biomass. Reproduced with the permission from ACS Sustainable Chemistry and Engineering 2019 (Sun et al., 2019).

hold the lignin inside the matrix of the biomass weaken, facilitating its separation from the other biomass components. Delignification is a selective technique that specifically focuses on separating and isolating the constituents of lignin, leading to a higher efficacy in extracting lignin. The choice of certain cations and anions in the IL is directly related to how lignin molecules are dissolved, leading to their extraction from the biomass matrix. By adhering to these techniques, it is feasible to separate lignin from biomass in a very efficient way (Norfarhana et al., 2024).

The classification of ILs into protic and aprotic categories. Aprotic ILs (APILs) primarily interact with cellulose, causing a decrease in cellulose crystallization. On the other hand, protic ILs (PILs) break down lignin, allowing for its removal from the structure (Haykir et al., 2023). In order to enhance the selectivity and efficiency of the reaction system, it is important to note that researchers have the ability to build functionalized ILs based on the requirements of the system.

Sun et al. (2019) studied with the use of ionic liquids and MAE to isolate lignin from eucalyptus. Lignin samples were analyzed and compared to determine the extraction method's success in terms of lignin yield, sugar content, and S/G ratio. They found ILs-MAE extracted 97.6 % (w/w) eucalyptus biomass and 24.6 % lignin (Klason lignin) at 140 °C. It was also investigated that using low-intensity MWs, they found half of lignin (45.8 %) content. Variations in MW power affect biomass decomposition and lignin extraction by ILs. Later, Ovejero-Pérez et al. (2020) suggested that PILs (1-methylimidazolium chloride) is an efficient solvent for the specific extraction of lignin from wood. This study investigates the impact of treatment severity on the process of lignin dissolution by using the PILs to extract lignin from biomass at various loadings, temperatures, and durations. The highest amount of lignin recovered (82.35 g lignin/100 g biomass lignin) was obtained while using a biomass loading of 10 % (w/w), a temperature of 135 °C, and a reaction time of 6 h.

The researchers also demonstrated that by balancing depolymerization and repolymerization, 1-methylimidazolium chloride lignin extraction severity affects molecular weight and lignin variety. It also facilitates ether bond breakage and recombination, which may improve the thermal stability of removed lignin. Recently, Yang et al. (2023) aimed that production of high content lignin fibers with lignin levels reaching upto 90 %. This was achieved utilizing an affordable protic ILs called *N,N*-dimethylbutylammonium hydrogen sulfate [DMBA][HSO4], together with 88 % hydrolyzed PVA as a spinning aid. The study demonstrated the applicability of the approach to iono-solvlignins and industrial KLs, which exhibit limited solubility in volatile organic solvents. The findings suggest that it is possible to get a high carbon yield (35–40 %) while maintaining a superior fiber shape. They also found that the spinning method utilizes unaltered industrial lignin, a low-cost and non-flammable water/IL mixture that can potentially be recycled.

So that the ILs method incorporates a commercially available polymer additive that is cost-effective, non-toxic, biodegradable, and possibly derived from biological sources. Although ILs have a number of exceptional benefits, as was noted before, they also have a number of drawbacks. These disadvantages include not only a high cost of some solvents, a complex retrieval procedure, and reutilization of these liquids are also crucial. The amount of solvent loss and purge that may be allowed before it has a major impact on the economics of the process is determined by the cost of the solvent. These difficulties have a substantial impact on the large-scale industrial use of ILs for the processing of LC biomass. So that the toxicological and environmental studies for scaling up IL-biomass dissolution are crucial. Innovative research in ILs has led to sustainable technology development, including the extraction of biopolymers from any biomass source.

4.2.5. *Deep eutectic solvents*

Organic solvents are dangerous and expensive due to their production and purification. Considering and assessing material safety in manufacturing and use is crucial. Sustainable development is the major reason for switching to green solvents, along with economics. Nature invites us to foster industrial success via environmentally appropriate and sustainable actions. Despite their small limitations, green solvents must be used as heat transfer fluids with greater attention. A solution to the negative effects of dangerous substances on science and business is needed. Give eco-friendly alternatives a priority in industrial chemical operations (Jafari et al., 2021). In recent years, there has been a

substantial increase in the use of deep eutectic solvents (DESs) for fractionation, owing to their exceptional capacity to dissolve lignin from LC biomass (Mankar et al., 2022b; Zhou et al., 2022b). DESs are a kind of environmentally friendly solvents that have a low tendency to evaporate (Chen and Mu, 2021; Smith et al., 2014), strong resistance to heat (Wang et al., 2021), low levels of toxicity (Suthar et al., 2023), and the capacity to break down naturally (Jagirani and Soylak, 2022). These characteristics make them appealing solvents for the processing of LC biomass, particularly for the valorization of lignin (Sosa et al., 2020).

In general, there are four types of DESs (Table 3) (Jablonský et al., 2019). Type I, DES are attributed to a limited number of unhydrated metal halides that possess sufficiently low melting temperatures for their manufacture, so that this type of DESs have the limited applicability. Despite this, the incorporation of hydrated metal halides with ChCl in the creation of Type II DESs enables them to possess a broader spectrum of uses. Due to their cost-effectiveness and air and moisture resilience, hydrated metal halides make good DESs for industrial usage (Smith et al., 2014). Type III DESs have been extensively researched and used because to their straightforward synthesis, biodegradability, and very low cost. Usually, this particular form of DES includes a quaternary ammonium salt as the hydrogen bond acceptor (HBA) (specifically ChCl) and amides, carboxylic acids, urea and alcohols as the hydrogen bond donors (HBDs). The primary mode of interaction between HBA and HBD is via hydrogen bonds, but electrostatic and Van der Waals forces may also play a role (Almeida et al., 2023). Type IV DESs consist of metal salts (e.g., ZnCl2) and HBDs such as urea, acetamide, and ethylene glycol (Tanaka et al., 2022). Additionally, Abranches et al. (2019) found similar effect in a combination of thymol and menthol, and they proposed that this mixture be classified as a new form of DES (Type V), which is a non-ionic DES in nature. This finding broadens the capabilities of DESs as well as the applications that may be used for them, and it has sparked a great deal of curiosity among academics working in both experimental and computational fields. Fig. 7 shows that the characteristics features of DES and its multidirectional application areas. DESs are often created by mixing HBDs and HBAs to produce a eutectic mixture with a significantly reduced melting point compared to the separate components (Andrew P. Abbott et al., 2004).

DESs are preferred over traditional ionic liquids (ILs) because to the greater accessibility and lower prices of basic ingredients, as well as the simplicity of storage and production (Bo et al., 2022; Grillo et al., 2021). Fig. 8 shows the development of DES for the use of biomass pretreatment. The use of DES pretreatment on hardwood plant fiber enhances the dissolution of the interlayer inside the cell wall, leading to an increased release of lignin and hemicellulose. The acidic DES, in particular, has superior ability to dissolve bonds in lignocellulose (Wu et al., 2022). Lignin passes through depolymerization and repolymerization routes that are comparable to those outlined for the use of ILs. The primary mechanism of depolymerization is the cleavage of inter-unit ether linkages (Mankar et al., 2022a; Wang et al., 2023b). When it comes down to it, the residual content of β-O-4′ linkages and the degree of condensation of the isolated lignin are contingent upon the

Table 3
Examples of various compositions of DES.

DES Type	Composition	Example	Reference
Type I	Metal salt + Organic salt	$Zncl_2$ + ChCl	(Smith et al., 2014)
Type II	Metal salt hydrate + Organic salt	$CoCl_2{\cdot}6H_2O$ + ChCl	(Smith et al., 2014; Xiao et al., 2024)
Type III	Organic salt + HBD	ChCl + Urea	(Zhang et al., 2012)
Type IV	Metal salt + HBD	Urea + $ZnCl_2$	(Tanaka et al., 2022)
Type V	Non-ionic DES, (Phenolic group + aliphatic hydroxyl group)	Thymol + Menthol	(Abranches and Coutinho, 2022; Tolmachev et al., 2022)

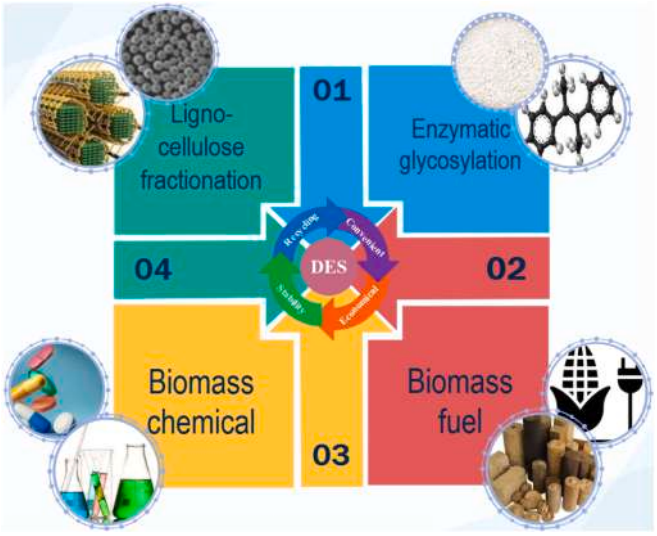

Fig. 7. The characteristics of DES and its areas of application: Reproduced with the permission from (Xiao et al., 2024).

characteristics of the solvent and the temperature at which the process is occurring (Brienza et al., 2024). In order to successfully valorize lignin as a by-product in biorefinery operations, it is essential to have a solid understanding of the structural alterations that occur in lignin during DES pretreatment. A typical DES-extracted lignin purity is 75–98 %. Lignin impurities consist mainly of DESs and polysaccharide residues. With more severe treatment conditions and anti-solvent or recovery strategies, lignin purity rises (Lobato-Rodríguez et al., 2023). Cronin et al. (2020) shows that how DESs remove high-quality lignin from corn stover hydrolysate. The process delivered up to 75 % lignin with purity exceeding 90 %. The composition of the DES substantially affects lignin extraction yield and purity.

DES formulations affected extraction efficiency and purity. The study also found that DES treatment factors including reaction temperature, time, and composition may govern extracted lignin properties like molecular weight distribution and crucial functional group abundance. The high-purity lignin recovered from maize stover hydrolysate by DES extraction may be utilized to make sustainable and biodegradable composite materials, expanding its usage in various sectors. Later, Fernandes et al. (2021) formulated a novel DES by combining lactic acid (LA), tartaric acid (TA), and choline chloride (ChCl) in a molar ratio of 4:1:1. The purpose was to extract lignin effectively and selectively from maritime pine sawdust. The most favorable extraction circumstances were a duration of 1 h at a temperature of 175 °C, resulting in a 95 % yield of lignin with a purity of 89 %. The lignin generated exhibited comparable characteristics to those documented in the existing literature, suggesting its potential use in bioplastics, adhesives, and carbon fibers. Table 4 describes the pretreatment conditions with various composition of DES.

Ma et al. (2022) showed wheat straw fractionation by treating it with a chlorine-lactic acid DES system. DES pretreatment considerably increased lignin extraction, cellulose retention, and hemicellulose removal. The investigation was carried out at 150 °C for 6 h that was good. The technique preserved 49.94–73.60 % of the processed biomass's cellulose and made the residue 89.98 % digestible. The experiment showed that DES pretreatment can fractionate LC biomass into high-quality cellulose and lignin with several uses. Fig. 9 shows the extraction of lignin by DES.

Mankar et al. (2022b) developed an efficient delignification process of coconut coir was carried out employing DESs. Specifically, a range of DESs based on ChCl and carboxylic acid were used to remove lignin from coconut coir with the assistance of MW irradiation. Out all the DESs that were created, the combination of ChCl and LA at a ratio of 1:4 resulted in

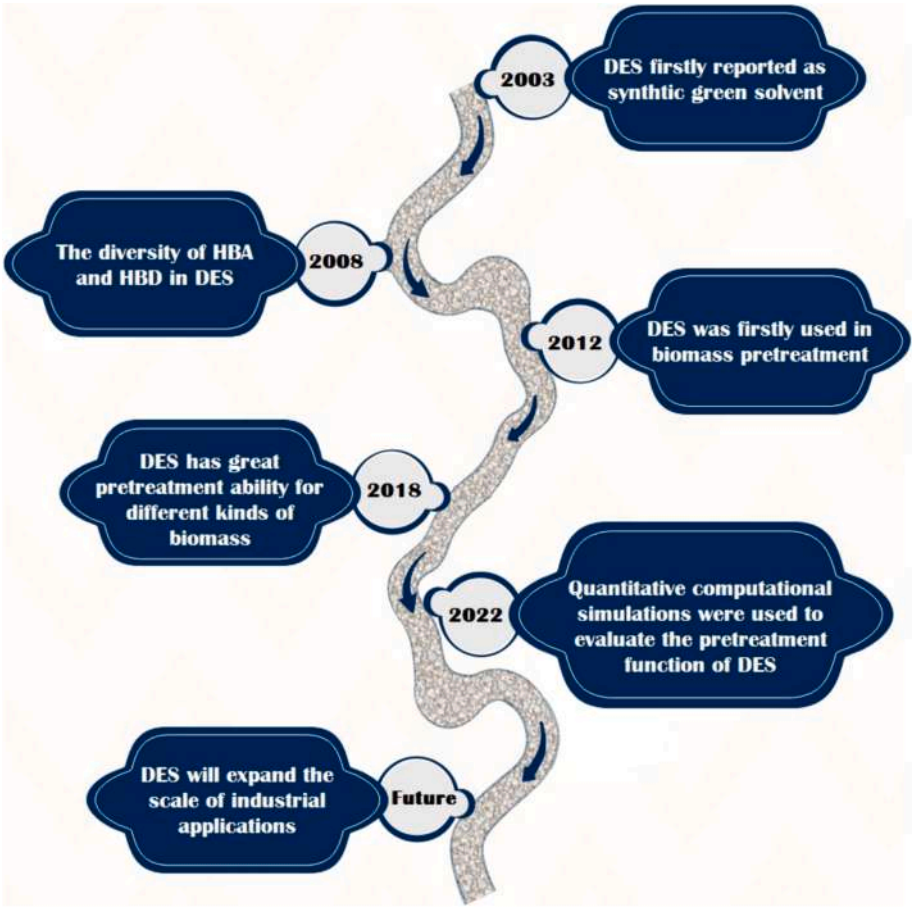

Fig. 8. Progress of DES solvent for use the use of biomass pretreatment (Xiao et al., 2024).

Table 4
Composition of DES with pretreatment conditions.

Feedstock	HBA	HBD	Molar Ratio	Pretreatment Temp (^{0}C)	Pretreatment Time (hour)	Reference
Rice husk	ChCl	LA	1:10	155	2	(Owhe et al., 2021)
Rice husk	ChCl	FA	1:2	155	2	(Owhe et al., 2021)
Rice husk	ChCl	AA	1:2	155	2	(Owhe et al., 2021)
Oil palm	ChCl	LA	1:5	120	8	(Tan et al., 2018)
Oil palm	ChCl	Gly	1:2	120	8	(Tan et al., 2018)
Eucalyptus chips	ChCl	LA	1:10	120	8	(Smink et al., 2020)
Beech wood	ChCl	KOH	1:4	80	8	(Mamilla et al., 2019)
Beech wood	ChCl	OA	1:2	80	8	(Mamilla et al., 2019)
Beech wood	ChCl	Urea	1:2	80	8	(Mamilla et al., 2019)
Wheat straw	ChCl	LA	1:2	150	3	(Lou and Zhang, 2022)
Poplar saw dust	ChCl	LA	1:2	130	1.5	(Su et al., 2021)
Sugarcane bagasse	ChCl	LA	1:5	80	12	(Chourasia et al., 2021)
Sugarcane bagasse	ChCl	MA	1:1	80	12	(Chourasia et al., 2021)
Sugarcane bagasse	ChCl	GLY	1:2	80	12	(Chourasia et al., 2021)
Rapeseed	ChCl	GA	1:1	100	8/16	(Suopajärvi et al., 2020)
Rapeseed	ChCl	GA	1:1	80	24	(Suopajärvi et al., 2020)
Corn stalk	ChCl	MA	1:1	100	8/16	(Suopajärvi et al., 2020)
Corn stalk	ChCl	MA	1:1	80	24	(Suopajärvi et al., 2020)
Red Pine	ChCl	LA	1:1	100/120/130	24/24/24	(Park et al., 2022)
Red Pine	BE	LA	1:1	100/120/130	24/24/24	(Park et al., 2022)
Red Pine	PC	GLY	1:4	100/120/130	24/24/24	(Park et al., 2022)
Bamboo	K_2CO_3	GLY	1:6	130	6	(Guo et al., 2022)
Miscanthus	ChCl	GLY	1:2	90/130	12	(Hassan and Mutelet, 2022)
Miscanthus	ChCl	EG	1:2	90/130	12	(Hassan and Mutelet, 2022)
Miscanthus	ChCl	UREA	1:2	90/130	12	(Hassan and Mutelet, 2022)
Bamboo	ChCl	BDO, $AlCl_3$	25:50:1	80–140	1	(Cheng et al., 2022)
Saw dust	ChCl	LA	1:2	120–180	4	(Kumar et al., 2020)
Wheat straw	K_2CO_3	GLY	1:5	100	16	(Yue et al., 2020)
Wheat straw	ChCl	LA	1:2	100	16	(Yue et al., 2020)
Poplar	ChCl	LA	1:9	120	6	(Chen et al., 2019a)

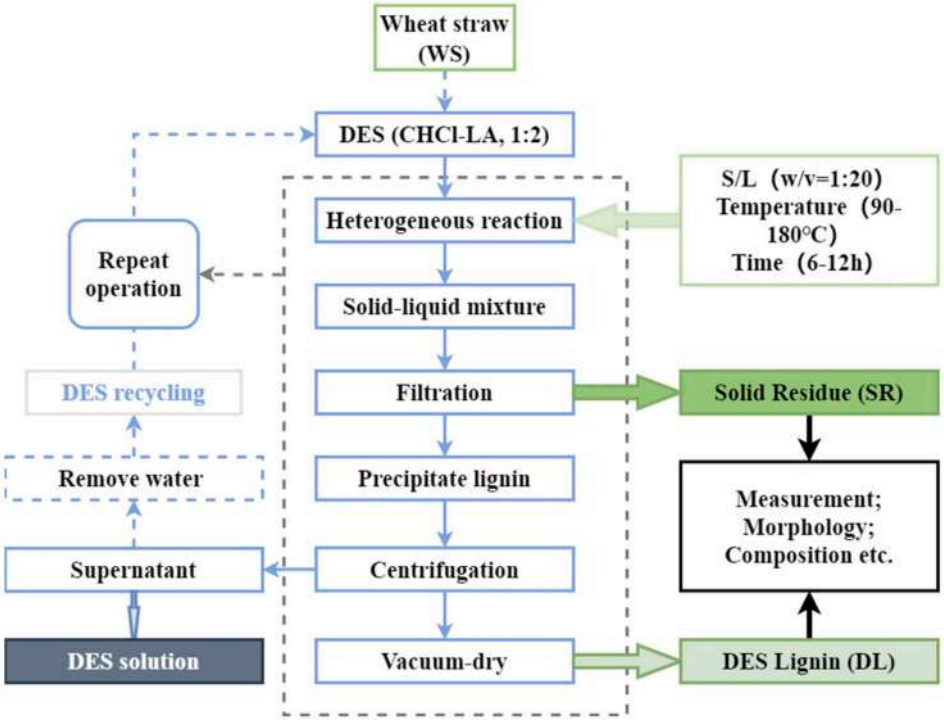

Fig. 9. Process flow chart of the lignin extraction by using DES:
Adopted from Molecules, MDPI 2022 (Ma et al., 2022).

the highest lignin yield of 82 % with a purity level above 95 %. Furthermore, Cui et al. (2023) shows that, DES and ethanol are efficient for lignin extraction and enzymatic hydrolysis, increasing glucose output under ideal pretreatment conditions. High temperature pre-treatment was shown to be beneficial for lignin with strong antioxidant performance. Recently, Wang et al. (2024) revealed that the use of DES pretreatment leads to a substantial degradation of poplar cell walls. Pre-treatment with DES eliminates non-cellulose components, lowering the "anti-degradation barrier" and enhancing enzymatic saccharification from 12.36 % to 90.56 %. In addition, DES pre-treatment breaks the β-O-4 connection between lignin units, resulting in a drop in molecular weight from 3187 to 1112 g/mol in 0–6 h. Regenerated lignin samples repolymerize after long treatments. The study found that pretreatment at temperatures over its melting point turns lignin into a liquid, enabling it to leak out of the cell wall. When the temperature drops, some dissolved lignin in deep eutectic solvent (DES) solutions condenses and deposits on cellulose microfibrils. DES pre-treatment of poplar breaks down the cell wall, improving sugar enzymatic conversion and lignin structure. These findings are crucial for biorefinery and lignin use.

Catechyl lignin, often known as C-lignin, is a sub-class of lignin that was recently identified in the seed coverings of certain plant species. These plant species include castor, vanilla orchid, and cactus (Sathasivam et al., 2023). The uniformity and consistency of C-lignin make it a perfect model of lignin for the process of valorization. In the last ten years, significant progress has been achieved in the study of C-lignin, both via experimental and theoretical approaches, driven by the fascinating characteristics of this biopolymer (Wang et al., 2022b). Several separation techniques have been developed to separate C-lignin, with

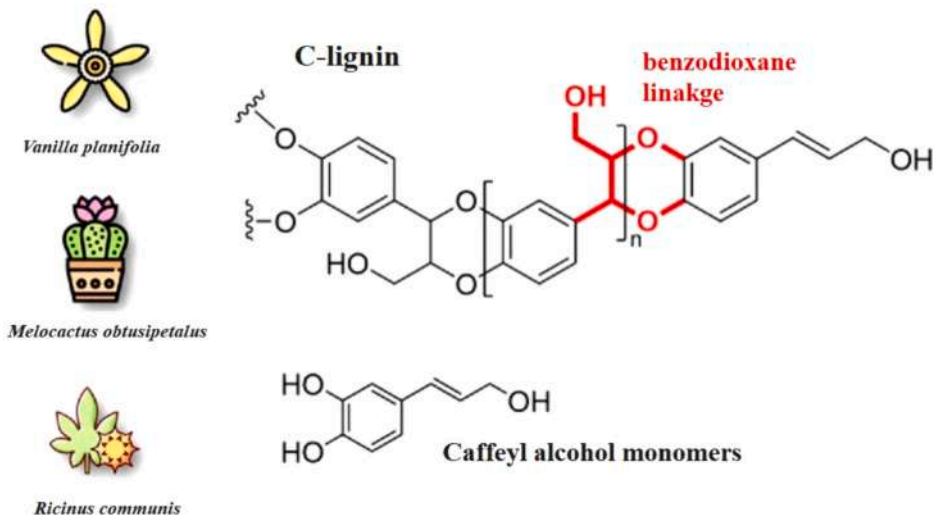

Fig. 10. Chemical structure of C-lignin with the highlighted benzodioxane linkages: Reused with permission from (Sathasivam et al., 2023).

DESs treatment being a particularly promising way for extracting C-lignin from biomass sources. Given that C-lignin consists of uniform catechyl units, the process of depolymerization to generate catechol monomers presents a very encouraging approach for the beneficial usage of C-lignin (Li et al., 2023b). Fig. 10 shows that chemical structure of C-lignin with the highlighted benzodioxane linkages. Wang et al. (2020a) shows that DESs effectively extract C-lignin from castor seed coats for the first time using an environmentally friendly, economically viable, and reusable process where C-lignin was extracted in huge amounts and with high purity. It was demonstrated that the halogen anion Cl^- has the ability to establish hydrogen bonds with –OH groups present in lignin, leading to the breakdown of lignin. DES functions as both a solvent and an acid catalyst for the breakage of β-O-4 bonds. After catalytic depolymerization with the combination of Pd/C and ChCl/LA, DES, C-lignin generated 29.6 % monomeric catechol derivatives with 86 % catechyl propanol. The findings demonstrate the potential of DESs to extract C-lignin from castor seed coatings and convert it into catechol molecules in a sustainable manner.

The use of C-lignin has the potential to be advanced via the development of extraction techniques that are both successful and environmentally friendly, as well as through the extension of the variety of raw materials that contain C-lignin. Both developments have the ability to advance the usage of C-lignin. In order to reduce the cost of C-lignin feedstock, the objective is to achieve genetic control over the production of C-lignin in plants. A method that has shown to be effective in the extraction of C-lignin is the use of DES therapy. On the other hand, the particular interaction that takes place between DES and C-lignin during the extraction process is not well known at this time. So that more research is needed to improve C-lignin extraction.

Pretreating LC biomass using DESs has several advantages. Unlike ionic liquids (ILs), DESs include inexpensive, readily available ingredients (Amini et al., 2021; Scelsi et al., 2021). DESs also inhibit cellulolytic enzymes and fermentative microorganisms less (Beluhan et al., 2023). After DES treatments, pulp purification from solvent may not be necessary. So that, DESs provide superior performance in extracting lignin, yielding larger quantities, and achieving more purity compared to conventional methods (Hu et al., 2023a). DESs are eco-friendly solvents, cost-effective, recyclable, and may increase pretreatment efficiency when paired with MW and ultrasonic technology (Roy et al., 2020; Xiao et al., 2024).

DES pretreatment of biomass has some drawbacks, including the high cost of DES synthesis, poor biomass penetration, poor compatibility with downstream processes etc. To improve lignin extraction, DES composition and conditions may be customized for each lignin source. Recently, Peng et al. (2023) studied the environmental efficacy of several pretreatment methods, namely ChCl:LA, ChCl:Urea, and NaOH, in the synthesis of bioethanol from rice straw. The findings demonstrated that the use of ChCl:LA and ChCl:Urea resulted in a detrimental effect on the net energy ratio in comparison to the utilization of NaOH. Nevertheless, the use of ChCl:LA led to a decrease in greenhouse gas emissions. Zaib et al. (Zaib et al., 2022) was performed life cycle assessment to evaluate the environmental footprint of ChCl:Urea with that of conventional organic solvents in the manufacture of acetophenone. The findings indicated that ChCl:Urea had a reduced environmental footprint compared to dichloromethane and ethyl acetate but a greater effect than methanol and ethanol. This method can effectively extract lignin from agricultural waste, woody biomass, and waste streams. However, further research is needed to understand extraction processes and find the best DESs for different lignin sources. It is necessary to conduct process optimization and scale-up studies in order to ascertain whether or not this method is feasible for use in industrial settings. As a result of the complexity of solvent design and process optimization, the high cost of DES synthesis, the low biomass penetration, the low compatibility with downstream processes, the difficulties associated with scaling up owing to the need for large amounts of DESs, the problems associated with handling and disposal, and the low solvent recovery or recycling that is available. It is still essential to do more research on the pretreatment technologies and solvent recycling methods in order to achieve the objective of increasing the number of DES-based biorefinery platforms.

4.2.6. Microwave assisted extraction

Microwave assisted extraction (MAE) has gained significance in the last 15 years owing to its environmentally friendly and efficient extraction capabilities (Reddy et al., 2020). MAE is a technology that enables the rapid and efficient degradation of lignin and hemicelluloses, while also being ecologically beneficial (Abolore et al., 2024). MAE offers a significant advantage compared to conventional extraction methods due to its use of MW radiation. This radiation speeds up the evaporation of remaining water in the raw materials and subsequently weakens the cell walls of plants, making it easier to extract substances by internal diffusion (Wang et al., 2020b,c). The mechanism of MAE is influenced by several physical and chemical processes, bincluding as the solvent's penetration into the plant material, its reflection and refraction of MWs, and the solubility and diffusion of cellulose in the solvent (López-Salazar et al., 2023; Mamilla et al., 2019).

MWA pretreatment of lignocellulose biomass involves the interaction between the microwave electromagnetic field and the polar molecules in the biomass. This reaction results in rapid heating and breakdown of the lignocellulosic structure. Additionally, the microwave radiation may directly heat and partly degrade lignin, resulting in improved accessibility of cellulose to enzymes or chemical hydrolysis during following processing stages. The efficacy of the microwave aided treatment process is influenced by several factors, such as the nature of the biomass, level of moisture present, particle size, type of solvent used, as well as the strength and duration of the microwave application (Abolore et al., 2024). In addition, due to the selective and focused nature of the heating in MAE, less energy is wasted in the surrounding environment (Reddy et al., 2020). Considering the above advantages, the MAE process is more ecologically sustainable. Consequently, the extraction yield is enhanced, while the time and solvent consumption are reduced (Isci et al., 2020). Investigations have been conducted to assess the viability of using MW power as a pre-treatment technique for enhancing various LC by-products. The MW power generates interior warmth more efficiently than traditional heating, therefore surpassing the sluggish and inefficient energy transmission of conventional systems (Abbott et al., 2004; Kohli et al., 2020).

MW possess perpendicular magnetic and electric fields due to their electromagnetic nature. Dipolar rotation and ionic conduction are two simultaneous mechanisms via which the electric field generates heat (Rodríguez-Rojo et al., 2012). Dipole rotation refers to the mutual influence of polar molecules due to the MW's swiftly alternating electric field (Francisco et al., 2012). The alignment of a molecule electric field with a dipole moment, which can be either permanent or even produced by the electric field in both the solid samples and the solvent, causes dipolar rotation to happen. The ion motion produced by an electric field causes an instantaneous superheating of ionic substances as part of the ion conduction mechanism. Energy transfer is more effective as temperatures rise (Kwon et al., 2021). This oscillation heats the sample liquid by clashing with nearby molecules. MW heat samples consistently and simultaneously, unlike other procedures. MW disrupt weak hydrogen bonds, which dipole rotation promotes in cellulose and lignin isolation (Ninomiya et al., 2014; Sun et al., 2019). Fig. 11 shows the scheme of the MAE of lignin from LC biomass. Zhou et al. (2012) found that the using MAE for lignin is that it is a better and more efficient approach for extracting lignin from birch in comparison to the standard oil bath heating methodology. The study demonstrated that MAE resulted in considerably higher lignin yield and delignification degree compared to oil bath heating. Achieving a MW heating time of 30 min resulted in a significant maximum delignification degree of 89.77 %. The extracted lignin sample had a slightly lower molecular weight than milled wood lignin (MWL) and exhibited improved antioxidant activity

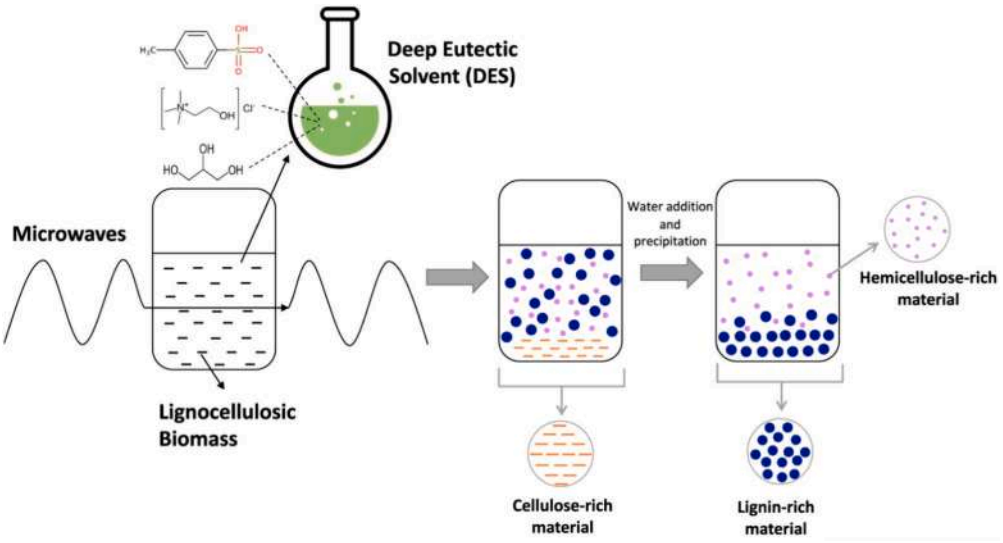

Fig. 11. Scheme of the MAE of lignin from LC biomass. Reproduced with the permission from (Mao et al., 2023).

compared to MWL. In addition, the research observed that MAE necessitated a lower energy input compared to oil bath heating and had the ability to rapidly enter the solution, resulting in time savings. Later, Zhou et al. (2017) found that MW-assisted acidolysis is a promising method for efficiently and specifically removing lignin from softwood. The technology enables the attainment of greater amounts of lignin purity and yield by a direct operation, which sometimes takes several days or even weeks when using conventional processes using milled wood lignin. The residual lignin obtained during MW-assisted acidolysis is a material characterized by a high concentration of aromatic chemicals. This technique leads to reduced degradation of lignin in comparison to conventional acidolysis conducted under same conditions.

Kohli et al. (2020) found that MW technology and DESs broke down miscanthus and birchwood biomass feedstocks efficiently. The maximum amount of miscanthus and birchwood lignin was extracted using ChCl-formic acid and ChCl-oxalic acid DES. Lignin extraction from biomass feedstocks increased with temperature from 60 to 130 °C. Extended reaction times improved biomass feedstock delignification. Grillo et al. (2021) showed a unique method of biomass pretreatment by mixing natural deep eutectic solvents (NaDESs) with MW technology. The results indicate that NaDESs have significant promise as environmentally friendly solvents for the separation and recovery of lignin, as well as for the release of sugars during subsequent enzymatic hydrolysis. Furthermore, an examination was conducted on a novel category of NaDESs formed from lignin, known as LigDESs. The results of this investigation demonstrated encouraging outcomes in the process of removing lignin from wheat straw. MW irradiation facilitated a rapid pretreatment at a low temperature (120 °C) for a short duration (30 min). In order to get a deeper comprehension of the interaction between MW and these environmentally friendly solvents, an analysis was conducted on the dielectric characteristics of NaDESs. A "closed-loop" biorefinery process was enabled by a NaDES that used biomass pretreatment lignin as a hydrogen bond donor. Later, Meindl et al. (2022) discovered the best ChCl based DESs for MW lignin extraction. A normal household MW was used to remove lignin quickly and effectively from untreated larch bark using DES. Lignin was eliminated 96 % after 30 min of MW irradiation. In comparison to traditional deep eutectic extraction by heating, reaction time was cut by 87 % and energy costs by 93.5 %. Switching hydrogen bond donors and testing acid, hydroxyl, and amide-based donor systems for MW lignin extraction. The investigation described a novel ecological and energy-efficient lignin valorisation approach without pricey apparatus.

In the end, comprehensive control and monitoring capabilities optimize operations in real time, improving productivity and product quality (Abolore et al., 2024). MAE provides benefits but also has drawbacks such inconsistent heating, limited penetration depth, safety, scalability, material restrictions, and response control (Priecel and Lopez-Sanchez, 2019). Although MW equipment requires a large initial investment but has lower operational costs than traditional heating techniques or a short pre-treatment period (Sharma et al., 2023). Furthermore, DES-MAE analysis of the extracted lignin indicated that it has bigger particle sizes and a greater structural diversity compared to commercial Kraft lignin. Additionally, the lignin obtained by using the combination of DES-MAE has a higher degree of methylation and a higher H/G sub-unit ratio. So that, this type of extracted lignin has significant potential for innovative value-added applications (Mao et al., 2023).

4.2.7. Comparative analysis of the different pretreatment techniques

Extracting lignin is an essential stage in the pretreatment procedure for turning lignocellulosic biomass into biofuels and other valuable products. Table 5 shows a comparison of conventional and environmentally friendly pretreatment methods, showing the yield, purity, cost, and emissions of lignocellulosic biomass for each.

5. Application of lignin

There are several applications that might be found for lignin in a variety of industries, including as the textile application, the food packaging industry, the biomedical application and also it offers a sustainable and environmentally friendly alternative to traditional materials, which include biobased polymers and functional compounds like phenolic resin, polyurethane foams, and carbon fibers. An area of research that is particularly exciting is 3-D printing, which has the potential to employ lignin as a manufacturing material, so opening new prospects for environmentally responsible industrial methods. Lignin contribution to the creation of environmentally friendly technologies and materials is becoming more significant as researchers continue to investigate the various benefits that lignin offers the scientific community. Furthermore, it is crucial to acknowledge that lignin has several other roles. The possibilities seem to be limitless, including a wide range of applications such as energy and coatings sectors as a feasible substitute for fossil-derived compounds, providing several environmental advantages. With the growing need to reduce carbon emissions and adopt more sustainable practices, lignin presents itself as a promising solution to meet the demands of a rapidly changing world. By

Table 5
Comparative analysis of the different pretreatment techniques.

Extraction method	Yield	Purity	Cost	Emissions	Reference
Kraft process	Moderate	Purity is higher than lignosulfonate process	Varied depending on scale and efficiency	High	(Brienza et al., 2024)
Lignosulfonate process	Moderate	Low which contains sulfur and other impurities	Relatively high	High	(Brienza et al., 2024)
Soda Pulping	Moderate to high	Produced sulfur free lignin	Varied depending on scale and efficiency of operations.	Less emissions than Kraft and sulfite pulping	(Brienza et al., 2024)
Alkali treatment	Higher in agricultural residues than woody biomass.	Sulfur free lignin than Kraft and sulfite pulping	Cost effective	Less than acid treatment.	(Oriez et al., 2020)
Acid treatment	Moderate to high	Good	Relatively high	High	(Solarte-Toro et al., 2019)
Organosolv	Moderate to high	High due to the specific type of organic solvent	Cost effective	Lower emissions compared to acid treatment	(Tanis et al., 2024)
SC-CO_2	Moderate to high	High	Initial investment cost is higher than conventional method	Considered as green and sustainable process	(Yang et al., 2022)
NTP	Good (though depending on the process parameters)	High	Initial investment and operational cost are higher	Considered as environmentally friendly process	(Pereira et al., 2021b)
ILS	High due to the efficient dissolution of lignin in ILs	High due to the capacity of ILs to specifically dissolve lignin from the biomass matrix.	Lower than dilute acidic treatment	Lower than conventional methods	(Brandt-Talbot et al., 2017)
IL-MAE	High	High	Initial investment is higher, but the faster extraction process causes lower costs.	Low	(Sun et al., 2019)
DES	High due to the efficient fractionation	High	20 % less than ILS	Low	(Lobato-Rodríguez et al., 2023)
MAE	Good	High	The initial investment is higher, but the energy savings and faster extraction make it cost-effective (93.5 % reduction).	Low	(Meindl et al., 2022)

consistently conducting research and producing innovative ideas in lignin, everyone may enhance its intrinsic value and contribute to a more environmentally friendly and sustainable world.

5.1. Lignin used in textile area

A growing number of studies have indicated that biopolymers produced from lignin have the potential to serve as an alternative to synthetic polymers in the production process of textiles (Vasile and Baican, 2023). Using lignin in textiles has several benefits. Renewable, plentiful, and able to reduce petroleum reliance. Lignin-based polymers offer decreased density, enhanced toughness, superior thermal characteristics, and biodegradability. Therefore, these materials are ideal for various textile applications (Jin et al., 2021). Several spinning techniques may create high-quality lignin textile fibers. The amorphous form, fragility, and inability to interact with other chemicals make lignin challenging to utilize in textiles. Scientists changed the structure and mixed polymers to increase lignin fiber spinning and physical qualities (Elfaleh et al., 2023; Raman et al., 2022).

Lignin and lignin-based bio adsorbent are adaptable, affordable, and high absorbent of heavy metals and synthetic colourants, making them promising textile wastewater treatment solutions. Multiple research studies have demonstrated that lignin-based biomaterials remove contaminants from wastewater, making them useful in wastewater treatment. Lignin nanomaterials provide appealing and adaptable water treatment methods (Islam and Hyder, 2022). Yao et al. (2020) demonstrated that the production of carbonized lignosulfonate-based porous nanocomposites as adsorbents for contaminants and dye particles demonstrates the effective use of lignin-based materials with exceptional adsorption characteristics. The lignosulfonate was transformed into carbon spheres using a sustainable technique known as MWA hydrothermal carbonization. The spheres underwent oxidation to be converted into nanographene oxide (nGO) carbon dots. The materials were then merged to produce self-supporting three-dimensional porous composites, eliminating the need for organic solvents or chemical crosslinking agents. Recently, lignin has been recognized as a nontoxic, biodegradable, UV-protective biopolymer. Lignin provides broad UV protection due to its phenolic, ketone, chromophoric, and auxochromic structures (Zhang and Naebe, 2021).

Kwon et al. (2023) studied MWA-DES pretreated lignocellulose nanofibrils (LCNF) shown ability to UV radiation and transmit UV light depending on lignin levels. Due to its phenolic nature, lignin, found in biomass with cellulose, absorbs UV light. UV protection is necessary in fabrics, and cosmetics to prevent UV damage. UV rays may damage, discolor, and degrade materials, shortening their lifespan. Researchers suggested that by adding UV-blocking LCNF to textiles, manufacturers may protect their products from UV rays. This precaution improves the longevity and efficiency of fabrics and compositions by keeping their quality, appearance, and functionality. Recently Nisar and Raza (2024) examined a bio-finish made from lignin was used to produce a superhydrophobic cellulose fabric with a water contact angle of up to 157.2°. This shows excellent water repellency and self-cleaning. The fabric also blocks 92.24 % and 98.62 % of UV-A and UV-B light which is ideal for UV-protection applications. The coating of lignin may increase cellulose fabric's water-repellent and UV-blocking properties which might potentially be used in the area of technical and functional textiles (Fig. 12).

Similarly, Juikar and Nadanathangam (2020) introduces a unique microbiological method for synthesizing nanolignin from cotton stalks. After application of nanolignin into cotton and linen textiles, get diverse advantages. The treated materials exhibited antioxidant characteristics, provided UV protection, and effectively eliminated *Staphylococcus aureus* and *Klebsiella pneumoniae*. The findings show that nanolignin may improve textile performance sustainably. Nanolignin may enhance textiles in an ecologically friendly and customizable way.

The diverse composition and inherent resistance to degradation of lignin render it unsuitable for a wide range of applications. This characteristic may be useful for protecting natural fiber geotextiles from

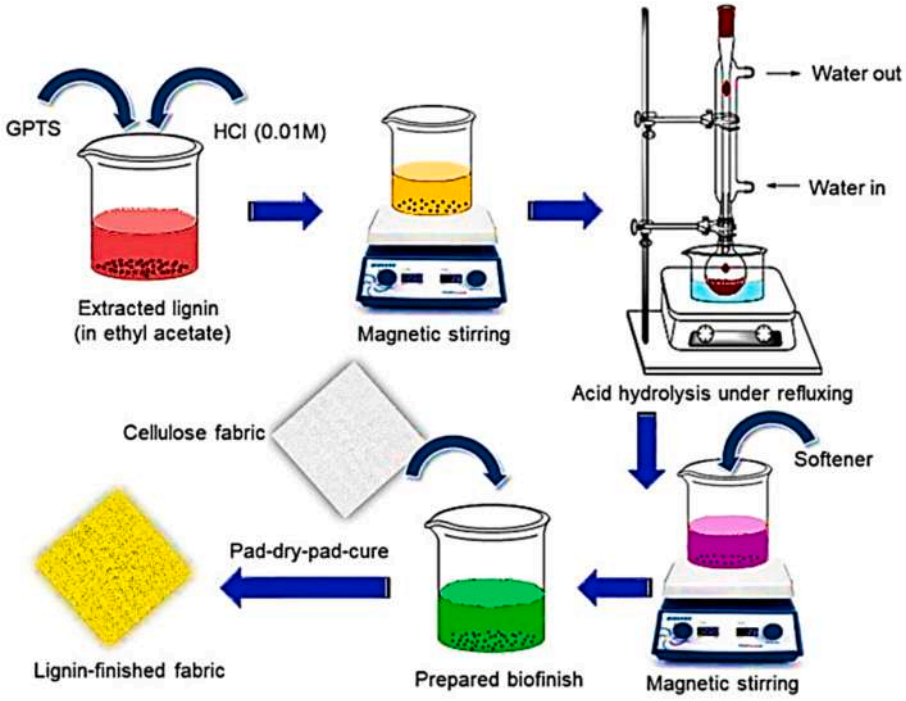

Fig. 12. Application of lignin-based coating on cellulose fabric: Reused with permission from (Nisar and Raza, 2024).

biodegradation (Kaya et al., 2023). Due to its unique structure, lignin exhibits antioxidant properties like hindered phenols utilized in polymers (Pouteau et al., 2003). Hindered phenols are widely utilized in geotextiles because they work from ambient to processing temperatures. These antioxidants remain effective after landfill disposal because they considerably limit polymer waste breakdown. Lignin's biodegradability and long-lasting antioxidant capabilities make it a good antioxidant (Kaya et al., 2023). Kaya et al. (2023) showed that lignin may preserve natural fiber geotextiles, prolonging their lifespan. Textile auxiliary lignin blends organic fiber strength with lignin coating protection. This research supports sustainable fabrics and eliminates artificial material pollution.

Lignin has attracted considerable interest as a bio-derived flame retardant component owing to its notable production of char as it decomposes (Raman et al., 2022). Cayla et al. (2016) showed that lignin has been included as an ingredient in intumescent flame retardant (IFR) systems for textiles. When lignin used in conjunction with ammonium polyphosphate (AP), has the potential to enhance the fire resistance of polylactic acid (PLA) textiles. These results suggest that adding lignin to intumescent systems may improve textile fire retardancy, making them suitable for many applications. In addition, Lignin as a bio-derived carbonization agent in intumescent formulations answers textile industry need for eco-friendly products. This method generates eco-friendly and sustainable textile resources and satisfies flame retardant requirements, enabling a "green concept".

5.2. Lignin in food packaging

Packaging film prolongs food shelf life and prevents spoiling. Today, most food packaging films are made from petroleum-based polymers. Due to their stability and degradation resistance, their widespread usage has caused severe environmental damage. Thus, researchers must quickly find biodegradable plastic film alternatives (Anushikha and Gaikwad, 2023). Lignin, a byproduct of cellulose, has drawn an interest in the packaging industry due to its capacity to withstand free radicals, water, and biodegrade properties. Lignin's complex phenolic structure gives it antioxidant properties, making it a useful biopolymer. The water repellency of film is improved due to the presence of lignin's hydrophobic functional groups (Boarino and Klok, 2023). Utilizing lignin in food packaging may reduce plastic waste and give sustainable alternatives (Basbasan et al., 2022). Lignin-derived nanofillers or nanoparticles have the potential to enhance the mechanical strength, gas barrier, UV resistance and antioxidant characteristics of biodegradable polymers (Chelliah et al., 2023). Lignin may also be used as a covering for controlled-release packaging or as a reservoir for active ingredient release. Lignin may also store antioxidant and antibacterial bioactive molecules like lignans (Boarino and Klok, 2023).

Zolfaghari et al. (2024) emphasized the possibility of using OSL obtained from potato crop leftovers as a suitable ingredient for creating environmentally friendly films intended for sustainable food packaging. Incorporating extracted lignin led to a decrease in the oxygen permeability, an improvement in the thermal stability of the film, and an elevation in its antioxidant capabilities. The water vapor transfer rate was minimized by increasing the quantity of lignin. According to researchers' suggestion, MWA-DES extracted lignin may enhance the UV resistance of their packaging goods. This measure ensures the preservation of the quality, appearance, and effectiveness of packaged items, hence enhancing their longevity and performance (Kwon et al., 2023). Zadeh et al. (2018) indicated that adding alkali lignin to biopolymeric packaging sheets may enhance additional benefits on food, packaging, agriculture, and medicine industries. Enzymatically modified soy protein isolate films with lignin had superior mechanical properties, radical scavenging activity, smoother surfaces, and water absorption. The study also demonstrated that UV rays was blocked by alkali lignin, which increased with concentration.

The intrinsic characteristics of lignin impact the UV-blocking effectiveness of packaging films (Tran et al., 2021). As an antioxidant, lignin contains phenolic compounds that absorb and block UV radiation (Lin et al., 2021). Lignin blocks UV light owing to its chromophoric nature, which gives it color and light absorption. Lignin-modified films' UV-blocking ability aids food packing by protecting UV-sensitive goods from UV-induced oxidation. This boosts the packaging films' efficacy and protects the packaged items (Lee et al., 2020).

5.3. Lignin-based carbon fibers

The development of carbon fiber that uses lignin as the feedstock has been the subject of ongoing research and development for more than half a century. During the 1960 s and 1970 s, the Nippon Kayaku Company was the company that brought the lignin-based carbon fiber known as Kayacarbon to the market. This product was taken off the market in the 1970 s due to the availability of superior carbon fibers that were derived from a variety of precursors (Li and Takkellapati, 2018). Despite cellulose being the most abundant renewable carbon source on Earth, lignin is the second. Its main sources are wood and plant biomass (Sokolov et al., 2018). The need for affordable carbon fiber in the production of key items like automobiles and wind turbines has made lignin-based carbon fiber a desirable target (Yadav et al., 2023). In addition to its cheap cost and renewable nature, lignin is anticipated to provide other advantages such as a high carbon yield and the removal of harmful compounds used in the manufacturing of carbon fiber with PAN (Qu et al., 2022). When it comes to the transformation of biomass into carbon-based products that are beneficial to the environment, organosolv pretreatment is a strategy that shows a great deal of promise. However, there is a lack of complete understanding on how the existing extraction methods impact the acceptable use of these compounds (Paulsen Thoresen et al., 2021).

The process of producing carbon fiber from lignin involves three distinct steps: spinning, thermostabilization, and carbonization (Sun et al., 2022; Yadav et al., 2023). Melt-spinning and solvent-assisted spinning are the two most often used ways for preparing precursor from lignin. The spinning temperature is contingent upon the specific kind of lignin and other constituents present in the mixture (Bengtsson et al., 2022; Li and Takkellapati, 2018). Mainka et al. (2015) showed that Lignin-based carbon fiber may be used in mass series automotive applications, since the research describes the main processes involved in its conversion and the results suggest lignin-based carbon fiber might be a sustainable and cost-effective option for industrial uses, notably in the automobile sector. Lignin-based carbon fibers have the potential to produce vehicle body panels, chassis, and interior components that are both lightweight and possess high strength (Wang et al., 2022a). Because of their extensive surface area and electrical conductivity, they may potentially serve as electrodes for both supercapacitors and batteries (Wei et al., 2017). Lignin-based carbon fibers are used in the aerospace and military industries as ablative materials after thermal stability improvement (Xu et al., 2014). Additionally, it has the capability to identify gases, chemicals, and biological substances because to its expansive surface area and heightened sensitivity (Wang et al., 2020a). Lignin-based carbon fibers possess adsorption properties that enable them to effectively cleanse both water and air (Song et al., 2017). Dai et al. (2020) created hydrophobic lignin/TiO_2 carbon nanofibers with superior photocatalytic efficiency and cycle performance. The produced carbon nanofibers degraded methylene blue more than three times faster than commercial TiO_2 powder. Using these fibers, methylene blue removal rate reached 91.5 % after four cycles at 200/min. The improved characteristics of lignin-based carbon nanotubes are due to their strong electrical conductivity and hydrophobicity.

5.4. Lignin-based polyurethane foams

Lignin is used in the production of polyurethane (PU) foams, among other diverse applications. PU foams are highly adaptable materials with a wide range of uses, including transportation, medicine, and insulation (Kaikade and Sabnis, 2023). Lignin has the capacity to diminish reliance on limited resources and improve the sustainability of materials created in the manufacturing of PU foams (Duval et al., 2022). One of the most prevalent methods for incorporating lignin-based green composites into PU products is via integration, which includes foams, elastomers, adhesives, and coatings (Alinejad et al., 2019). It has been observed that incorporating lignin into rigid PU foams improves their characteristics in comparison to conventional PU foams. Some of the advantages include enhanced crosslinking density, enhanced biodegradability, heightened antioxidant properties, UV resistance, and better thermal stability and mechanical strength of the final product (Kaur et al., 2022; Ruwoldt et al., 2023).

Xue et al. (2015) was developed MWA liquefaction to improve the optimization of biobased polyols obtained from lignin liquefaction. The untreated liquefied lignin products may be immediately used in the one-shot process for producing stiff polyurethane foams. Gaining knowledge about the yield, content, and molecular structures of liquid lignin polyol would be advantageous for manufacturing certain stiff polyurethane foam products. Huang et al. (2017) analysed switchgrass lignin samples obtained by fractionation of MW liquefaction were used in the production of semirigid PU foams without undergoing purification. The PU foam samples were made by combining lignin with polyol and isocyanate in different amounts. This technology allowed the researchers to test switchgrass lignin as a polyurethane foam reinforcing filler. They also examined how lignin concentrations affected foam properties. Lignin's high hydroxyl functional groups enhance crosslinking density, resulting in hard foam, making it an issue in PU flexible foam compositions (Gondaliya and Nejad, 2021). Lignin's high Tg, which ranges from 90 to 180 °C, limited copolyol solubility, and sulphur-based odor further prevent its application in flexible PU foam (Agustiany et al., 2022). Fig. 13 shows the flow process to produce lignin-based polyurethane foam in both flexible and stiff forms.

Gondaliya and Nejad (2021) found that adding unmodified lignin to PU flexible foam improved its compression force deflection, tear resistance, and support factor. Lignin increased foam tensile strength and thermal stability. The study suggests that using lignin instead of polyol in PU foam may benefit the environment and economics.

5.5. Lignin-based phenolic resin

Kraft and organosolv lignin provide distinct benefits for phenolic resin applications. KL is cost-effective and reactive, making it ideal for large-scale industrial activities. However, OSL purity, chemical composition, and structure make it suitable for high-purity, specific applications (Salleh et al., 2023). OSL may be used as a suitable raw material in certain resin compositions, by substituting up to 30 % of the phenol in lignin-modified phenolic resins (PR). Various techniques have been used to incorporate organosolv lignin into PR (Çetin and Özmen, 2002). Yao et al. (2023) studied the MAE to get lignin with a more native-like structure provides several benefits when used as a filler in biobased resins. The advantages include higher mechanical qualities, increased toughness and ductility, and facilitation of sustainable material development. When combined, these advantages result in the production of environmentally friendly high-performance materials that may be used in many applications.

Some studies suggested that BHT might serve as an alternative technique for extracting lignin, hence facilitating the production of important substances such as resins and phenols (Schmatz and Brienzo, 2022). PRs are widely employed in plastic molding, foam manufacture, coating application, and semiconductor packaging due to their resistance to corrosion, heat, flame, and electrical insulation. Numerous studies have sought cost-effective, abundant, sustainable, and high-performing PR-making alternatives to phenol. As the lignin is the most abundant natural polyphenol possesses chemical structures similar to phenol. Sustainable fuels and compounds from lignin are gaining popularity. Replace phenol with lignin to cut PR costs and improve performance, resulting in low-toxicity residues and environmental friendliness (Li et al., 2023a). Fig. 14 shows the LPR synthesis with the sources of lignin.

Lignin-based phenolic resin (LPR) synthesis offers several benefits. Firstly, lignin contains a complex three-dimensional network of phenol or aldehyde structural units. These units may give aldehyde and hydroxyl groups during phenol–formaldehyde reactions, making phenolic

Fig. 13. Flow process to produce lignin-based polyurethane foam in both flexible and stiff forms: Reused with permission from (Agustiany et al., 2022).

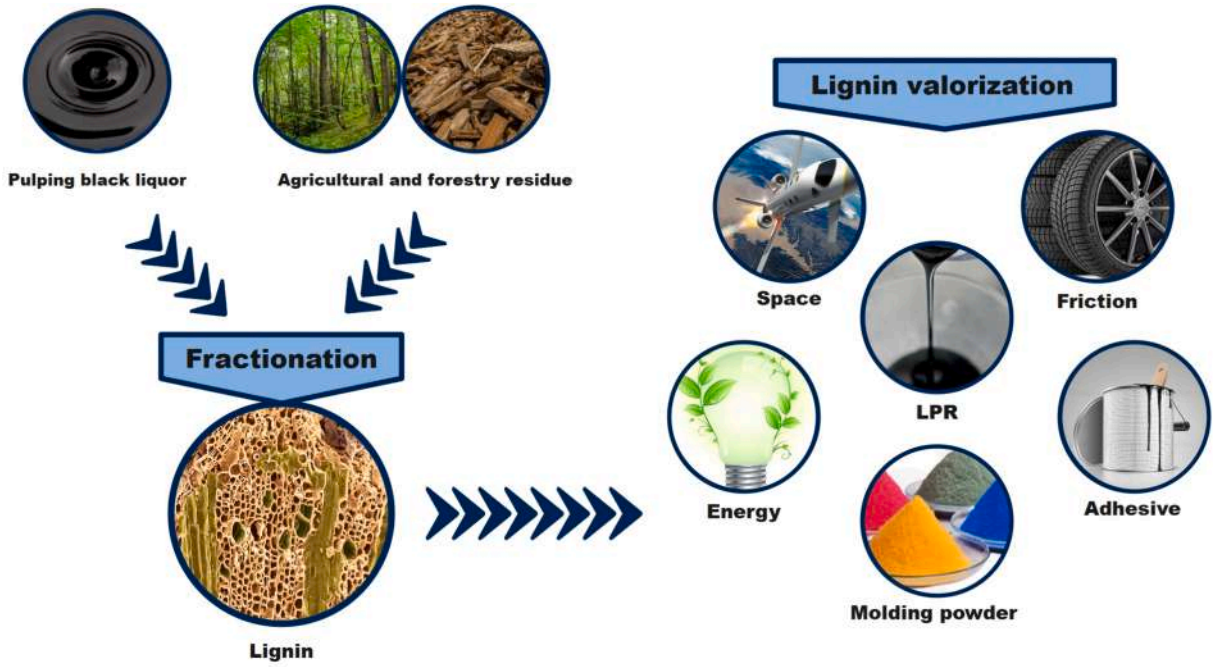

Fig. 14. Origins of lignin and its uses in LPR synthesis. Reused with the permission from GC 2023 (W. Li et al., 2023).

resin easier to synthesize. Secondly, this reduces the usage of non-renewable, ecologically harmful phenol and formaldehyde. OSL improves phenolic resin adhesiveness, solidification rate, and thermal/mechanical properties (Gao et al., 2021).

The incorporation of lignin into phenolic-based resin systems offers a significant opportunity for the advancement of high-performance bio-based products (Lu and Gu, 2023). Lignin-based phenolic resins offer improved features such lower free formaldehyde and phenol levels, higher temperature resistance, and mechanical endurance. These improvements may help develop eco-friendly resin products by reducing volatile organic compounds (VOCs) and their environmental impact (Melro et al., 2022; Xie et al., 2023). Fig. 15 shows that the Manipulation of TPR with a high degree of *ortho*-substitution in lignin, and the mechanism of PBS toughening.

Xie et al. (2023) studied the thermoplastic lignin phenolic polymers were synthesized through the utilization of lignin, phenol, and formaldehyde as precursor substances. In order to mitigate the brittleness and fragility of the lignin thermoplastic phenolic resin, a toughening modification was implemented involving Polybutylene succinate (PBS). Lignin contributes to the principles of green chemistry in phenolic resin production by using sustainable feedstocks, minimizing waste generation, and mitigating environmental consequences.

5.6. *Lignin-based 3-D printing*

Lignin and its derivatives are attractive materials for 3-D printing due to their renewable nature and ability to develop chemicals that are beneficial to the environment. The use of lignin in 3-D printing has several advantages, including its abundant availability as a byproduct of the pulp and paper industry, its inherent biodegradability, and its capacity to replace petroleum-based polymers in various applications (Vasile and Baican, 2023). Lignin may be utilized in 3-D printing via

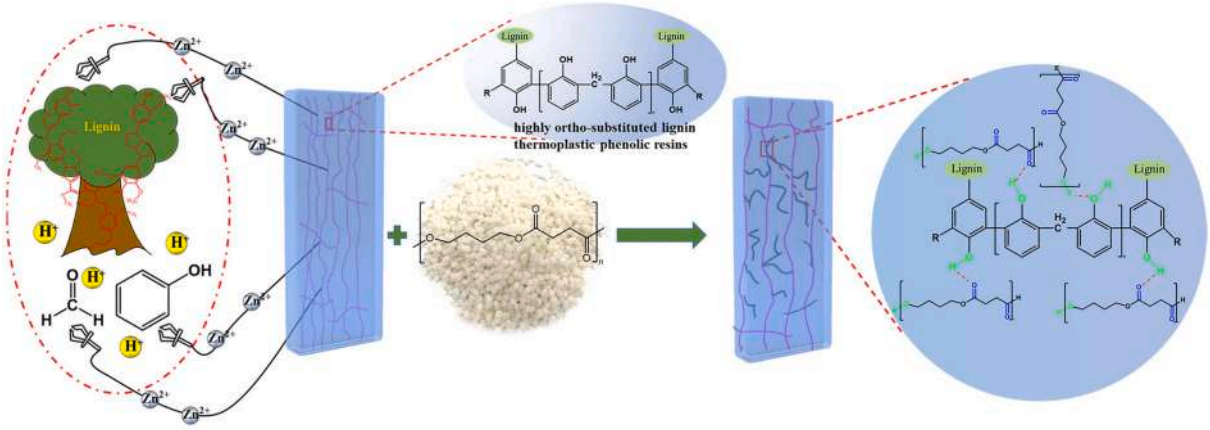

Fig. 15. Manipulation of TPR with a high degree of *ortho*-substitution in lignin, and the mechanism of PBS toughening. Adopted from Molecular Science, MDPI 2023 (Xie et al., 2023).

extrusion (Wan et al., 2023), direct ink writing (DIW) (Ebers et al., 2021), fused filament fabrication (FFF) (Jiang et al., 2023), and vat photopolymerization(Vasile and Baican, 2023). These technologies allow the printing of feedstocks with pure lignin, derivatives, or other polymers or monomers as fillers or additives (Abdullah et al., 2023). Lignin improves mechanical characteristics, thermal stability, and biocompatibility in 3-D printing materials, making them acceptable for many applications (Fazeli et al., 2024; Wan et al., 2023). Lignin is used in 3-D printing not only for creating materials, but also for producing functional biomaterials that possess distinct features (Abdullah et al., 2023).

Polylactic acid (PLA), which is recognized for its flexibility and usefulness in 3D printing (Fico et al., 2024). PLA composites loaded with organosolv lignin create continuous filaments during 3-D printing due to their rheology. Whereas PLA composites loaded with KL found that the melts flow poorly, making higher concentrations difficult to print. This is usually due to material flow issues. Compared to PLA and acrylonitrile butadiene styrene (ABS), OSL-filled PLA composites exhibit higher tensile strength and ductility. However, PLA composites loaded with KL become increasingly brittle as lignin concentration rises. This may limit their usage in heavy-load situations. OSL is extracted differently from KL, which may alter biocomposite sustainability and biodegradability. Sustainable biomass is used to make OSL, which promotes the manufacturing of ecologically friendly products (Ebers et al., 2021). Yao et al. (2023) showed that the use of methacrylated MW-extracted lignin in resin compositions for digital light processing 3-D printing results in an improvement in the toughness of the products through the enhancement of crosslinking, structural integrity, flexibility, and reinforcing effects. These characteristics, when combined, contribute to significant improvements in the tensile strength and elongation at break of the printed materials, which ultimately results in greater durability and resilience of the printed materials. The primary barrier to using lignin for 3-D printing includes its inherent fragility and lack of thermoplastic properties (de Freitas et al., 2021; Ebers et al., 2021). In order to tackle this difficulty, scientists have investigated the use of thermoplastic polyurethane to control the flow characteristics of lignin, with the objective of enhancing its capacity to be processed and printed in 3D printing (Zhou et al., 2022c). Fig. 16 shows that representation of the fused deposition 3D printing process for CF/Lignin/TPU composites.

Furthermore, the use of lignin-coated cellulose nanocrystal/methacrylate composites in 3D stereolithography printing has shown improved mechanical strengthening and resistance to changes in temperature (Zhang et al., 2019). This advancement broadens the range of possible uses for lignin-based materials in the area of 3-D printing.

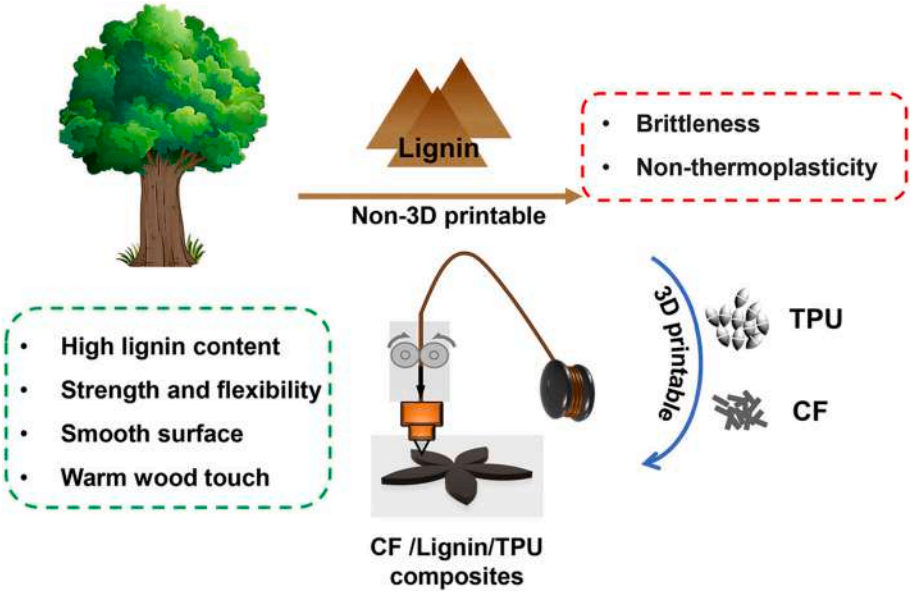

Fig. 16. Representation of the fused deposition 3D printing process for CF/Lignin/TPU composites: Reused with the permission from (Zhou et al., 2022c).

Moreover, lignin may be combined with other components, either by physical or chemical means, such as cellulose or its derivatives. This allows for the use of the unique properties of each component in the creation of advanced 3-D bio-feedstocks for the production of 3-D materials (Yang et al., 2020). Nguyen et al. (2018) developed a novel category of printed renewable composites containing 40–60 % lignin. Lignin added to nylon, a thermoplastic matrix, increases stiffness and decreases flow when melted, improving 3-D printability. Carbon fibers (CFs) increase the material's stiffness and endurance, making it better than thermoplastics like high-impact polystyrene (HIPS) resins for mechanical qualities and 3-D printing.

5.7. Lignin in biomedical applications

Lignin is a biowaste macromolecule that is not being used to its full potential and is attracting significant attention in the field of biomedical research. Nonetheless, the origin of lignin and the method of extraction significantly impact its chemical composition, hence affecting a cell's response to lignin. OSL is obtained using a sustainable method from residual waste material. There is a limited amount of research that has examined the compatibility of OSL with human cells (Menima-Medzogo et al., 2022). Lignin has great potential in several biomedical applications, namely in bone tissue engineering, wound healing, and drug delivery systems. Lignin is mostly used in biomedical applications due to its antibacterial and antioxidant characteristics, which effectively inhibit bacterial growth and mitigate oxidative stress, respectively (Gaspar and Fardim, 2023).

Bone tissue engineering has a close connection to the induction of bone and cartilage development, whether it arises from bone depletion or specific disorders such as osteoarthritis. The remarkable biocompatibility and biodegradability of lignin, together with its antioxidant and antibacterial properties, make this basic material a vital component in the formation of bone tissue (Hachimi Alaoui et al., 2023). The use of lignin in the process of wound healing has the potential to effectively limit the development of bacteria and reduce oxidative stress, consequently accelerating the healing process and reducing the amount of pain experienced by the patient (Balaji et al., 2023; Gaspar and Fardim, 2023). Jaiswal et al. (2020) showed that the carrageenan-based hydrogel with lignin-mediated silver nanoparticles and $MgCl_2$ greatly improved Sprague-Dawley rats' wound healing. The histological examination revealed that the hydrogel reduced the wound area to below 3 % after a span of two weeks. The findings suggest that adding lignin to the carrageenan matrix increased silver nanoparticle production for wound treatment. Lignin may increase hydrogel wound healing properties.

Menima-Medzogo et al. (2022) researched on fractionated OSL was analyzed to determine its structural and physico-chemical properties. The results revealed that as the molecular weight decreased, the phenolic biopolymer exhibited a less branching conformation and an increasing amount of aliphatic hydroxyl functionalities. Common biomaterials are known to be compatible with low molecular weight organosolv lignin. High cell viability was obtained by using low molecular weight lignin. Higher concentrations of the substance resulted in a decrease in cell viability, suggesting that these quantities had a harmful effect on the cells. The study suggested that low molecular weight OSL has the potential to be used in many tissue engineering applications.

Some studies have employed OSL as a surface treatment to reduce implant-related bone infection, a significant issue in orthopedic and trauma surgery and uncommon in dental implants. These studies found that coating titanium bone implants with 1 % (w/v) OSL increased antimicrobial efficacy against Staphylococcus aureus, Pseudomonas aeruginosa and Candida famata, which can colonize skin and mucous membranes and cause prosthesis-related infections and joint and bone inflammation (Erakovic et al., 2014). In the field of biomedicine, lignin is used extensively for the purpose of developing nano and microcapsules that contain specific drugs for the purpose of developing targeted therapies (Stanisz et al., 2022). Lignin have the capability to encapsulate drugs that are either hydrophobic or hydrophilic (Vasile and Baican, 2023).

Furthermore, limited research indicates that OSL may be used to produce cell scaffolds. It has been shown that the inclusion of lignin in the scaffolds enhances their rigidity and thickness, as well as promotes cell adhesion. These findings indicate that OSL has potential applications in creating tissue-like biomaterial-based structures for tissue regeneration. Additional research is required to explore eco-friendly pretreatment techniques such as ILs and DESs even with microwave. These methods seek to extract lignin from lignocellulosic biomass without damaging it or creating byproducts. Pharmaceutical discharge from biorefinery waste streams needs additional investigation.

6. Challenges of lignin extraction and uses

The quality and value of cellulose and hemicellulose fractions are affected by residual lignin, hence efficient and selective separation is crucial. Developing cost-effective, environmentally friendly, and byproduct-free extraction techniques is the challenge. Traditional lignin extraction procedures need high temperature, pressure, organic solvents, and acidic/alkaline conditions. These processes may utilize a lot of energy and toxic materials. Sustainability requires exploring energy and chemical-efficient extraction methods such employing ecologically friendly solvents or gentle extraction conditions (Lu and Gu, 2022). LC biomass from different sources and cultivars may vary greatly in lignin content and composition. Standardized and effective biomass extraction procedures are problematic due to biomass diversity. Sustainable and economically viable lignin extraction requires customizing and enhancing extraction methods for each feedstock. Wastewater, byproducts, and possibly hazardous waste from lignin extraction must be handled to minimize environmental impact. Environmentally acceptable methods for managing and reusing lignin extraction leftovers and trash are needed. To guarantee extraction sustainability, water usage and wastewater discharge must be minimized. The Perspective emphasizes that the fundamental heterogeneity in the structure and content of lignin, which is impacted by the plant source and extraction process, is the fundamental limitation that prevents the full use of the potential of materials that are based on lignin.

A comprehensive characterisation study must be carried out on both the initial reagents and the final products. Furthermore, since there is such a wide variety of lignin, it is necessary to develop a clear and direct link between its structure and activity for the bulk of the applications that it is used for. Effective valorization of lignin is difficult due to its complex structure. Lignin's resistance to breakdown and chemical modification complicates high-value product extraction and conversion. The resistance hinders lignin's use in many industrial applications. Sustainable valorization requires an interdisciplinary strategy that blends circular economics, green chemistry, and industrial ecology (Tardy et al., 2023). This technique considers the whole lignin life cycle, decreasing inefficiencies, optimizing resource utilization, and minimizing environmental impact. Industrial ecology attempts to create resource-efficient and sustainable industrial systems that mimic natural ecosystems, while green chemistry ensures valorization processes are environmentally safe. Sustainable lignin usage solutions may be created by combining these strategies to reduce environmental impact and resource consumption.

Utilizing environmentally friendly techniques for extracting lignin has many benefits, including cost-effectiveness. The use of toxic chemicals and the substantial energy consumption in conventional procedures, such as the Kraft process, may lead to high costs. Conversely, green processes often include the use of renewable resources and extraction approaches that are less severe. This may facilitate cost reduction and enhance the economic viability of lignin extraction. Despite the potential for economic savings, green processes still face some utilization

barriers that require resolution. One of the major challenges is the limited scalability of green extraction technologies, which are less efficient than conventional extraction techniques. Environmentally sustainable processes may rely on extraction technologies that are more intricate, posing challenges for scaling up to industrial levels. The limited availability of green extraction technology and equipment poses another barrier to its utilization. There are a number of businesses that still rely on traditional methods for lignin extraction, and there is a lack of infrastructure and expertise about ecologically acceptable extraction technology. There is a possibility that businesses may have challenges while trying to transition towards more environmentally friendly methods of lignin extraction.

Those goods that are generated from lignin do not have established procedures or precise standards, for this reason, lignin makers struggle to maintain quality and performance, which limits their market appeal. Lignin-derived products are only starting to develop in popularity. Current infrastructure and supply systems are largely for fossil fuels. Thus, establishing a sustainable and resilient market for lignin-based alternatives needs significant investments and stakeholder engagement.

7. Conclusion

The purpose of this review article is to introduce lignin as well as an in-depth study of its environmentally friendly methods that are used for its extraction now a days. DES, MAE and ILs recover lignin faster and retain its structure better than conventional solvents. Lignin extraction with high purity and low degradation has also showed promise using NTP and SCF methods. These methods allow extracted lignin to be customized to match industrial needs, such as molecular weight and functional groups. In addition to this, it investigates the many uses of lignin in a variety of different sectors has been extensively documented. Many industries employ biobased lignin, including polymers, adhesives, transportation fuels, and carbon compounds. Lignin aromatic structure and complicated chemical composition make it a promising precursor for high-value commodities, reducing fossil fuel use. LC biomass delignification research develops technological methods to turn LC feedstocks into commercially viable and competitive products. The huge potential and flexibility of lignin as a promising raw material is highlighted by ongoing study into the efficient extraction of lignin and its numerous applications across a variety of industries. Nevertheless, there are other obstacles that need to be controlled, such as the development of extraction techniques, the expansion of lignin's adaptability to specific industrial processes, and the implementation of manufacturing methods that are both economically feasible and substantial.

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CRediT authorship contribution statement

Mohammad Mahbubul Alam: . **Antonio Greco:** Writing – review & editing, Validation, Supervision, Formal analysis, Conceptualization. **Zahra Rajabimashhadi:** Writing – review & editing, Resources, Formal analysis. **Carola Esposito Corcione:** Writing – review & editing, Validation, Supervision, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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