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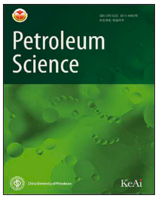

Review Paper

Lignin in oilfield application: Current trends and future perspectives

Na-Na Song [a], Mei-Chun Li [a,b,c,*], Xue-Qiang Guo [a], Ya-Xuan Zhang [a], Ming-Yuan Xin [a], Jin-Sheng Sun [a,c], Kai-He Lv [a,c], Chao-Zheng Liu [b]

[a] School of Petroleum Engineering, China University of Petroleum (East China), Qingdao, 266580, Shandong, China
[b] Co-Innovation Center of Efficient Processing and Utilization of Forest Resources, College of Materials Science and Engineering, Nanjing Forestry University, Nanjing, 210037, Jiangsu, China
[c] State Key Laboratory of Deep Oil and Gas, China University of Petroleum (East China), Qingdao, 266580, Shandong, China

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ABSTRACT

With the depletion of shallow and mid-depth oil and gas resources, exploration and development have increasingly shifted toward deep and ultra-deep reservoirs. However, high bottom-hole temperatures and pressures, along with complex geological conditions, pose significant challenges. Additionally, growing environmental regulations restrict the use of conventional petroleum-derived chemicals such as polyacrylamide, sulfonic acid resins, and sulfonic acid asphalt. In recent years, lignin has demonstrated significant potential in petroleum exploration and development due to their sustainability, biodegradability, and excellent thermal, chemical, mechanical, and rheological properties. This article reviews the structure, classification, preparation, and modification methods of lignin, followed by a systematic discussion of its applications in oilfield operations. In drilling fluids, lignin and its derivatives serve as rheological regulators, fluid loss control agents, and shale inhibitors, contributing to enhanced cuttings transport and wellbore stability. In fracturing fluids, they function as thickeners and breaker agents, improving fracturing efficiency while protecting the reservoir. In enhanced oil recovery, lignin-based polymers act as surfactants and profile control agents, reducing interfacial tension between water and rock surfaces and increasing the effective permeability of sandstone reservoirs. Furthermore, in oilfield wastewater treatment, lignin-based materials effectively remove oil-water mixtures, heavy metal cations, and solid particles through filtration, adsorption, and flocculation.

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1. Introduction

In the past few decades, global energy demand has been increasing and is expected to continue the same trend in the coming years (Jürgen and Rebecca, 2020). Despite the contribution of renewable energy sources in recent years, petroleum still remains the primary source of energy worldwide (Aftab et al., 2017; Sukmana et al., 2019). However, the oil industry is facing the reality of a scarcity of new large oil fields and a decline in production from existing reservoirs (Aftab et al., 2017). To meet demand, the oil industry has been moving toward drilling deeper to maximize the depth of the reservoir (Aftab et al., 2017). This requires the use of a large number of chemical additives to ensure smooth drilling under different conditions (Li et al., 2023). At present, a wide range of polymers, (e.g., polyacrylamide, sulfonate resin, and sulfonate asphalt), are often used as chemical additives in oilfields. However, most of them are derived from fossil fuels (Clemons and Caulfield, 2005), which have adverse effects on ecosystems and human health (Luong et al., 2012). Therefore, the development of environmentally friendly and cost-effective oilfield chemical additives has become an urgent task.

In recent years, biomass resources have garnered significant attention (Liu et al., 2024). Among a wide spectrum of biomass resources, lignin is considered to be the second most abundant natural material in plants (after cellulose) (Da et al., 2023; JamJam et al., 2019; Wei and Liu, 2023). Additionally, lignin as an industrial waste byproduct of the pulp and paper industry is currently not being fully utilized (Altam et al., 2022; Sathawong et al., 2018). It is

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* Corresponding author.
E-mail address: mli@upc.edu.cn (M.-C. Li).
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reported that only a small fraction (about 2%) is effectively utilized, while the majority of lignin is incinerated or discharged into the land, causing environmental pollution and significant waste of renewable resources (Balk et al., 2023; Clemons and Caulfield, 2005). It is worth noting that lignin has superior environmental protection, sustainability, biodegradability, thermal stability, and good mechanical properties (Kandola et al., 2025; Mahmood et al., 2016; Zhu et al., 2016). Furthermore, lignin is an irregular three-dimensional complex amorphous polyphenol molecule, and it holds crucial value in the development of renewable oilfield chemical additives (Gouveia et al., 2019). Compared with traditional petroleum-based polymers, lignin offers several advantages. As a biopolymer extracted from plants, lignin is widely available and renewable, whereas petroleum-derived polymers rely on non-renewable fossil fuels (Li et al., 2024). Utilizing lignin can reduce dependence on fossil resources and lower environmental impact, aligning with the growing demand for sustainability. Additionally, lignin exhibits excellent thermal, physical, and chemical properties, making it highly suitable for applications in harsh reservoir conditions (Liu W. et al., 2023). Its inherent thermal stability allows it to withstand high temperatures commonly encountered in reservoirs, preventing degradation and ensuring long-term performance (Luo et al., 2019). Furthermore, the diverse chemical structure of lignin allows for various modifications to impart specific functionalities, making it highly adaptable for a wide range of oilfield applications, e.g., rheological modifier, fluid loss reducer, shale inhibitor, and surfactant (Muhammed et al., 2021).

However, the application of lignin in oilfield has some limitations. For example, because lignin is a natural product, its extraction and purification process are complicated, which limits the feasibility of its large-scale application. High salinity conditions exist in some oilfield environments, and lignin may precipitate or lose activity at high salt concentration, which reduces its effectiveness as an oilfield chemical additive. In addition, lignin may decompose under extra-high temperatures in the ultra-deep formation, leading to poor performance. To promote the effective application of lignin in oilfields, it is necessary to modify lignin. Generally, lignin is chemically modified by etherification, crosslinking, and polymer grafting to enhance its thermal resistance and salt tolerance for more efficient oilfield application. The etherification of lignin introduces hydrophobic or hydrophilic groups, enhancing its solubility, chemical stability, and thermal stability while reducing sensitivity to acids and bases. However, precise control of reaction conditions is necessary to avoid uneven modification. Etherification may also lead to functional group loss and increased costs due to expensive etherifying agents. Crosslinking significantly enhances lignin's mechanical property, toughness, and heat resistance, making it suitable for structural materials. It improves solvent resistance and enables sustained release applications. However, crosslinking requires complex processes, high temperatures, and long curing times. Polymer grafting improves lignin's thermal stability and water resistance, allowing tailored material properties. However, reaction conditions are complex, and interactions between the grafted polymer and lignin may lead to unpredictable properties. Each modification method has advantages and limitations, requiring careful selection based on application needs, target performance, and economic feasibility.

Current applications of lignin and its derivatives in oilfields include drilling fluids, fracturing fluids, enhanced oil recovery fluids, and wastewater treatments. In drilling fluids, lignin and its derivatives can be used as rheological regulators, fluid loss reducing agents, and shale inhibitors (Chen et al., 2018). For example, Zhang et al. (2019) prepared a series of ferric chloride lignin sulfonates (FCLS) as shale inhibitors using ferric chloride and lignin as raw materials, and experiment results proved that FCLS significantly inhibited the swelling ratio of clay. In fracturing fluids, lignin and its derivatives can be employed as thickener and breaker, promoting the fracturing efficiency and reservoir protection. For example, Cong et al. (2022) immobilized β-mannanase with lignin as substrate (MIL), and then coated MIL with gelatin to prepare gel-breaking agent Ge@MIL. Experimental results indicated that during the backwashing process, MIL was released from Ge@MIL. Due to MIL's biodegradation effect on guar gum (GG), the viscosity of GG-based fracturing fluid gradually decreased, reaching the allowable discharge standard (5 Pa·s) after 10 min (Cong et al., 2022). However, the substrate immobilization method may reduce the activity and stability of the enzyme, resulting in low catalytic efficiency, and affect the degumming effect of Ge@MIL. In addition, the binding of lignin and enzyme may not be strong enough, resulting in unstable fixing effect, affecting the durability of Ge@MIL breaker. In EOR, lignin and its derivatives can be used as surfactants and profile control agents. Lignin-based polymers can form a dense and durable support network in fractures, improving the effective permeability of sandstone reservoirs. This lowers the tension between the aqueous phase and the rock surface, ultimately increasing oil and gas recovery rate. Dong et al. (2023) synthesized heat-resistant modified lignin (CRF) through sulfonation. They combined CRF with foaming agent CX-5 to form a steam-driven temperature control system, resulting in the formation of PCS (the compound of CRF and CX-5). PCS effectively improved the heterogeneity of the reservoir and increased the recovery rate by 28.7%. However, the thermal stability of CRF has not been studied. As a result, CRF may degrade under extremely high-temperature conditions, compromising its stability and performance and limiting its application in high-temperature reservoirs. Additionally, in some cases, the sulfonated derivatives of CRF may precipitate, affecting their dispersion and stability at the oil-water interface, thereby reducing their effectiveness in enhancing oil-water interactions. In the treatment of oilfield wastewater, lignin and its derivatives can effectively remove the oil-water mixture, heavy metal cations, and solid particles in wastewater through filtration, absorption and flocculation approaches. Yu et al. (2021) successfully prepared a fluorine-free micro-scale porous material composed of silane-modified lignin particles with strong super-oleophilic and super-hydrophobic properties. The experimental results indicate that the oil separation efficiency reached over 99%, and the separation efficiency of the surfactant stabilized oil-water emulsion reached over 98%.

In recent years, several review articles have explored the diverse applications of lignin in the oil and gas industry. For example, Negi and Singh (2023) provided a review on the applications of lignin in petroleum exploration, biofuel production, and oil spill cleanup, highlighting both the direct use of lignin and its chemical modifications to enhance performance in these areas. Juyal and Bhadauriya (2024) specifically focused on the role of lignin-based nanoparticles in enhancing oil recovery through electroflooding. Their work is pioneering in proposing lignin-derived nanoparticles as an environmentally friendly alternative for the efficient extraction of heavy oil during electric enhanced oil recovery (EOR) processes. Ghazali et al. (2018) provided an insightful overview of the potential advantages and future prospects of lignin-based deflocculants for clay-based drilling fluids, particularly in challenging high-temperature and high-pressure (HPHT) environments, such as deepwater drilling operations. Despite these valuable contributions, a comprehensive review covering the full spectrum of lignin's preparation, modification, and application across various oilfield operations, including drilling fluids, fracturing fluids, EOR, and wastewater treatment, is still lacking. Such an overview would provide crucial insights into the challenges and opportunities associated with lignin's integration into these diverse applications, paving the way for more sustainable and efficient oilfield technologies.

This review provides an overview of the recent advances in the applications of lignin and its derivatives in oilfields (Fig. 1). Initially, the structure, classification, and preparation of lignin are briefly introduced. Then, the strategies used to enhance the thermal resistance and salt tolerance of lignin are discussed. Subsequently, the latest progress of lignin and its derivatives in a wide range of oilfield applications, including drilling fluid, fracturing fluid, enhanced oil recovery, and oilfield wastewater treatment, are reviewed. Finally, the challenges and future research opportunities of lignin and its derivatives in oilfield applications are prospected.

2. Structure, classification, and preparation of lignin

2.1. Structure of lignin

Lignin is a polyphenolic biopolymer with a complex three-dimensional network, present in the cell walls of all terrestrial plants and some aquatic plants (Fig. 2(a)) (Su et al., 2022). In nature, lignin is second only to cellulose in abundance and is the only renewable aromatic biopolymer (Bruijnincx et al., 2015). The three main precursors of lignin polymers are para-coumaryl alcohol (H-unit), coniferyl alcohol (G-unit), and sinapyl alcohol (S-unit) (Fig. 2(b)) (Li and Xu, 2023). Lignin is synthesized by enzymatic dehydrogenation of these units, which form C–O and C–C bonds, resulting in heterogeneous and three-dimensional structures (Katahira et al., 2018).

Lignin from different plants is composed of varying percentages of these precursors (Fig. 2(c)) (Chio et al., 2019). According to the source or extraction process, lignin can be classified into different categories. For example, based on the source, lignin can be divided into softwood lignin (e.g., pine, spruce, and fir), hardwood lignin (e.g., oak, maple, and birch), and grass lignin (e.g., wheat straw and corn stover) (Fig. 2(d)).

Softwood lignin is mainly composed of G units (about 90%) and a small number of H and S units, making its structure more linear than that of hardwood lignin (Mei et al., 2019; Ragauskas et al., 2014; Vanholme et al., 2010). Such homogeneity of composition and structure is conducive to the increase of lignin yield during the separation process (Gellerstedt, 2015). In addition, softwood lignin contains more β–5′, 5–5′, β–β′, and β–1′ bonds. These bonds have high dissociation energy, making softwood lignin more resistant to degradation than hardwood lignin (Suota et al., 2021). Furthermore, the guaiacyl (G) unit in softwood lignin is easy to condense at the free C_5 position, resulting in a more heat-stable C–C bond. The interunit structure of C–C increases the average molecular weight, enhances the thermal stability, and creates a strong cross-linking structure, which makes it widely used in the preparation of adhesives (Suota et al., 2021).

Hardwood lignin is derived from the leaves or wood chips of angiosperms (hardwood, deciduous or broad-leaved species) and contains different proportions of S-type and G-type unit (Mei et al., 2019; Ragauskas et al., 2014; Vanholme et al., 2010). Hardwood

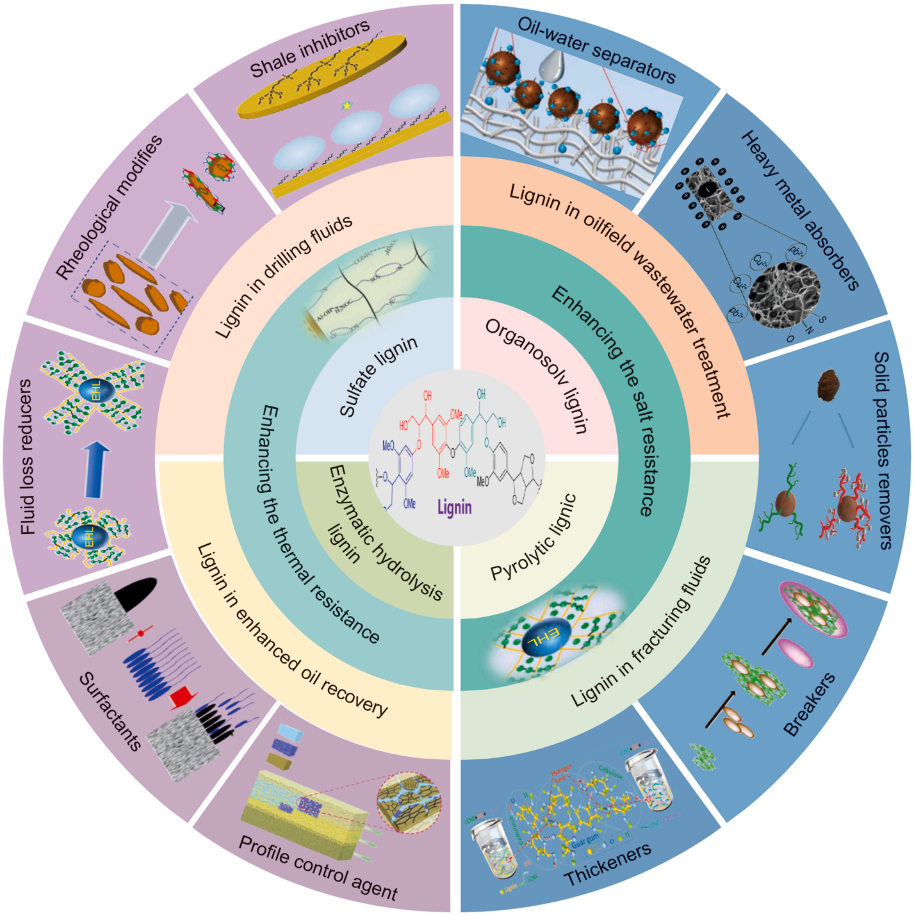

Fig. 1. An overview of the topic of this review, including the classification and modification of lignin, and its application in oilfields, including drilling fluid, fracturing fluid, enhanced oil recovery, and oilfield wastewater treatment.

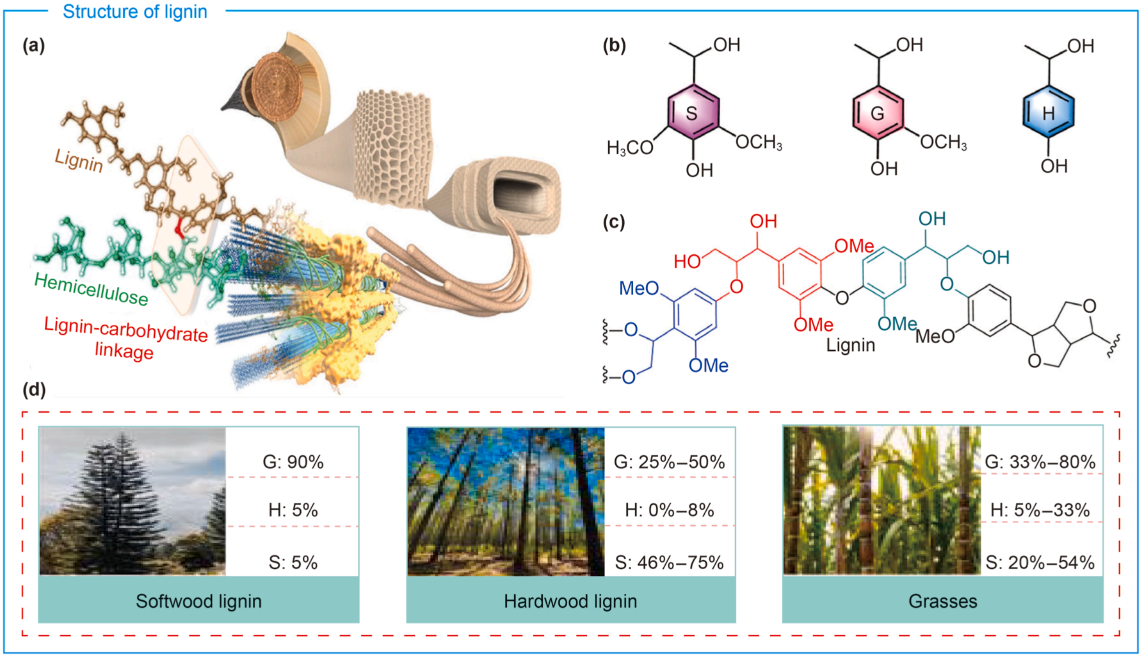

Fig. 2. Structure of lignin: **(a)** Three-dimensional view of lignin carbohydrate complex (Li and Takkellapati, 2018); **(b)** The three main types of natural lignin and their corresponding lignin units (S, G, H) (Yoo et al., 2020); **(c)** Molecular structure formula of lignin (Menezes et al., 2023); **(d)** The origin of lignin and the proportion of S, G and H monomers (Yoo et al., 2020).

lignin has more β–O–4 bonds than softwood lignin, therefore during its extraction, hardwood lignin exhibits lower thermal resistance compared to softwood lignin (Zhao et al., 2014). Suota et al. (2021) observed that the initial mass loss occurred at 212.8 and 232.6 °C for hardwood lignin and softwood lignin, respectively. With the increase of temperature, half of the initial mass of softwood lignin was lost at 539.5 °C, whereas the initial mass of hardwood lignin took place at 446.9 °C.

Lignin from grass usually contains S, H, and G units, which are synthesized by the phenylpropanoid and flavonoid pathways in the secondary cell wall (John and Gosta, 2008) In addition to differences in the relative amount of monolignols, another major difference between grass lignin and woody lignin is the presence of lignin-carbohydrate complex (LCC) bonds. In woody biomass, LCC is mainly composed of phenylglucosides, phenyl ethers, and gamma-ester bonds. In the grass, LCC bonds connect hemicellulose and lignin through ferulic acid (FA) bridges (Balakshin et al., 2011; Zeng et al., 2013).

2.2. Classification and preparation of lignin

The raw, unmodified lignin in lignocellulose is called natural lignin. It is a natural polymer found in the cell walls of wood and plants that often combines with cellulose and hemicellulose to form LCC. Lignin has a complex structure and is not easily separated from other components because of its covalent and hydrogen bonds with carbohydrates (Zhao et al., 2017). Therefore, it is impossible to completely isolate the neat lignin without destroying its primary structure. In recent years, a wide range of methods have been developed to extract lignin from biomass. According to its extraction process, lignin can be divided into four categories, i.e., sulfate lignin, organic solvent lignin, enzymatic hydrolysis lignin, and pyrolytic lignin (Chio et al., 2019).

2.2.1. Sulfate lignin

Sulfonated lignin is a by-product of pulp production, currently incinerated in the recovery boiler in the chemical recovery cycle, generating valuable bioenergy (Dimitris et al., 2023). The production of sulfate lignin is carried out in an acidic environment where NaOH and Na_2S are used for cooking, followed by acidification, neutralization, filtration, and washing to obtain the final product (Fig. 3(a)). During the reaction, bonds between lignin molecules are fractured and recombined under the action of basic ions (OH^-, HS^- and S^{2-}) to form water-soluble sulfate lignin. Furthermore, β–O–4 and α–O–4 bonds are broken during the reaction, resulting in the formation of a large number of phenolic hydroxyl groups in the lignin (Florbela et al., 2009). The incorporation of phenolic hydroxyl and sulfate groups in lignin structure improves the solubility and reactivity of lignin (Malyar et al., 2022).

Claudia et al., (2017) used carbon dioxide precipitation method to continuously cook coniferous wood chips and extracted coniferous lignin sulfate from the black liquor obtained by sulfate pulping. The sawdust of southern pine was suspended in NaOH solution, and then Na_2S solution was added. The active alkalinity of the suspension was 17%, the vulcanization degree was 25%, and the liquid lignin ratio was 6/1. The pulp was then processed using a Parr reactor. When the H-factor reached 1750, the reaction mixture was cooled to around 70 °C, then filtered to remove unreacted lignin particles. The filtrate was collected and acidified with CO_2 gas until the pH reached 9. Then, the suspension was acidified to pH = 2 using a 50% v/v H_2SO_4 aqueous solution. After filtration, the filter cake was rinsed with water and finally dried to obtain softwood sulfate lignin. According to the methoxy signal in ^{13}C NMR and QQ-HSQC spectra, it was suggested that in the process of sulfate pulping, the hydrogen sulfide anion nucleophilic attacked the methoxy group in the lignin, causing demethylation and producing malodorous methyl hydrogen sulfide and dimethyl hydrogen sulfide. By infrared analysis, it was found that sulfate lignin contains small amounts of β–O–4′ and small amounts of stable carbon-carbon groups, such as phenyl coumaran (β–5′) and pinoresinols (β–β′), and also contains stilbenes, aryl enol ether, lignin-carbohydrate alpha-phenyl ether, and second iso-rotarinol structure. Sulfate lignin has been widely employed for a variety

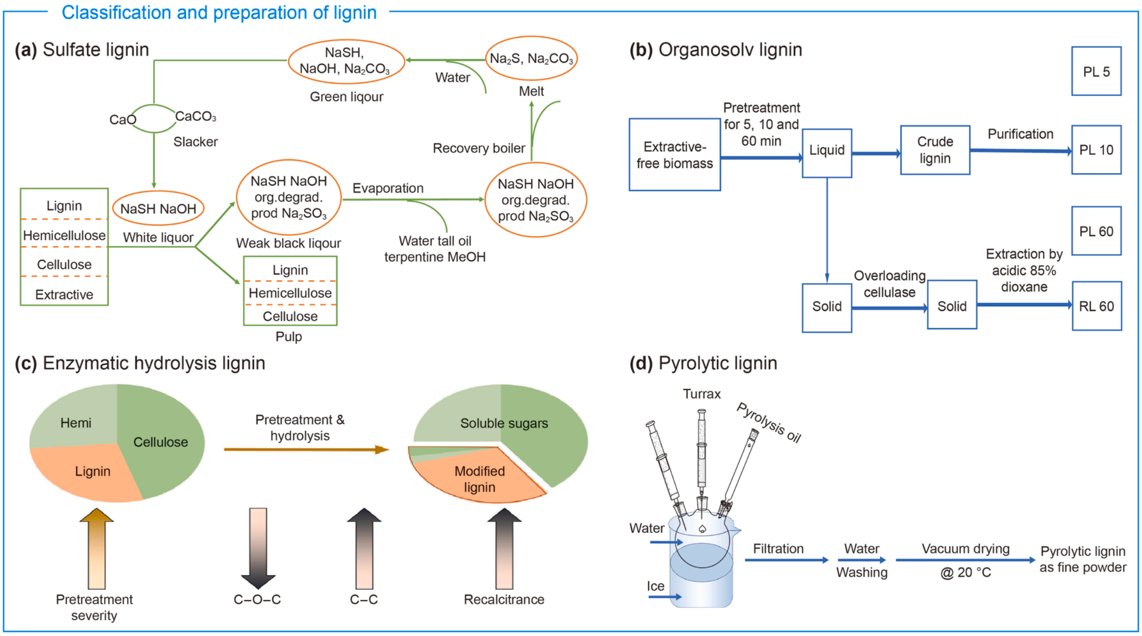

Fig. 3. Classification and preparation of lignin: **(a)** Preparation process of sulfate lignin (Yao et al., 2018); **(b)** Preparation process of organosolv lignin (Scholze and Meier, 2001); **(c)** Preparation process of enzymatic hydrolysis lignin (EHL) (Yoo et al., 2020); and **(d)** preparation process of pyrolytic lignin (PL) (Claudia et al., 2017).

of applications, including polymer composites (Wang C. et al., 2016, Wang Z.H. et al., 2016), adhesives (Dávid and Pukánszky, 2017), carbon fibers (Doherty et al., 2011), biofuels, and biochemicals (Sen et al., 2015).

2.2.2. Organosolv lignin

The pretreatment alters certain properties of the plant cell walls via removing and/or redistributing hemicellulose and lignin, disrupting the ultrastructure of cellulose, and increasing the accessible surface area of cellulose, thereby reducing the biomass resistance (Li et al., 2024b; Xu et al., 2021; Zhou et al., 2024). Among various biomass pretreatment methods, organic solvent pretreatment is one of the most widely used delignification methods (Liu K. et al., 2022; Zheng et al., 2024). It is most commonly used as a pre-treatment step before enzymatic hydrolysis of cellulose fractionation (Camilla et al., 2018; Kangas et al., 2015; Jang et al., 2016). Organic solvents can dissolve a large amount of lignin and produce a solid residue rich in cellulose (Yao et al., 2018). Through adding an antisolvent into the lignin solution, the dissolved lignin can be precipitated. Therefore, the lignin extracted from biomass using an organic solvent is called organosolv lignin (OSL) (Teramoto et al., 2012).

The OSL extraction process involves the dissolution and extraction of lignin and hemicellulose with an organic solvent, leaving behind insoluble solid cellulose fibers (Fig. 3(b)) (Schwiderski et al., 2014; Timilsena et al., 2013). Ethanol is one of the cheapest and most readily available green solvents for biomass production. It has the advantages of low boiling point, easy recyclability, and the ability to produce sulfur-free lignin, making it widely used in organic solvent pretreatment (Bassem et al., 2010; Arato and Gordon, 2005; Zhao et al., 2009). The contents of hydroxyl and carbonyl groups of OSL is higher than that of conventional processed lignin, which is beneficial to the chemical modification and processing of lignin (Dan et al., 2016; Torre et al., 2013). OSL has a similar structure to the original lignin and is generally considered as a substitute for natural lignin (Chen H. et al., 2019; Li et al., 2024a).

Yao et al. (2018) studied the structural changes of lignin during the pretreatment of papyrus with ethanol organic solvent, and characterized the structure of ethanol-soluble lignin by FT-IR, GPC, ^{31}P and HSQC NMR techniques. The extraction of lignin by organic solvent method is divided into two processes: i) pretreatment, and ii) separation and purification. The first step involved pretreating the papyrus. The pulp was pretreated with approximately 50% v/v ethanol/water solution and 0.9 wt% sulfuric acid, and the solid residue was collected after filtration. The solid residue was then washed with a mixture of ethanol/water (1/1, v/v) to neutrality and dried at room temperature for 24 h. The second step was to purify the pre-treated lignin by separation. The solid residue obtained in the above steps was subjected to rotary evaporation and freeze-drying to obtain coarse lignin. The recovered solid was then dissolved in 1,2-dichloroethane/ethanol (2/1, v/v), precipitated with ethyl ether, washed with petroleum ether, centrifuged, and dried under vacuum at 40 °C to obtain OSL. The results of GPC showed that as the pretreatment time increased from 5 to 60 min, and the mass average molecular weight (Mw) of lignin in ethanol organic solvent increased. ^{31}P NMR analysis showed that the aliphatic OH content increased in the order of $OSL_{60\ \mathrm{min}} < OSL_{5\ \mathrm{min}} < OSL_{10\ \mathrm{min}} <$ MWL (Milled Wood Lignin). $OSL_{60\ \mathrm{min}}$ had the highest average molecular weight, and stilbene units were formed in the pretreatment process, which is suitable for the synthesis of plant antiviral drugs. By contrast, $OSL_{10\ \mathrm{min}}$ exhibited the highest content of fatty and phenolic hydroxyl groups, which is ideal for the synthesis of polyurethanes for thermoplastic and thermosetting materials.

2.2.3. Enzymatic hydrolysis lignin

Enzymatic hydrolysis lignin (EHL) is a by-product of fuel ethanol production from lignocellulosic raw materials (Sandeep et al., 2014; Qian et al., 2014). The extraction of EHL is preceded by enzymatic treatment (Fig. 3(c)). The degree of enzymatic treatment of biomass is related to the loss of C–O–C bonds and the formation of C–C bond, which is conducive to the enhancement of chemical resistance of lignin (Fabricia et al., 2023).

Zhang et al. (2022) used cellulase and xylanase to separate and purify lignin from the residues of corn stover after enzymatic hydrolysis, and then employed pH gradient precipitation for lignin fractionation to produce structurally intact lignin. Laccase (Lac), lignin peroxidase (LiP), and manganese peroxidase (MnP) underwent enzymatic reactions at 30 °C for 24 h using different combinations. The structural changes of lignin after degradation were characterized by SEM, nitrogen adsorption, and Fourier transform infrared spectroscopy. The results showed that during Lac enzymatic hydrolysis, the methoxy–O–CH_3 bond attached to the benzene ring was broken, resulting in lignin depolymerization. The content of methoxy group was reduced, and acetyl syringone (a specific product of Lac enzyme) was formed. Therefore, Lac primarily catalyzed the decomposition of phenolic lignin substrates. On the other hand, LiP has a very high REDOX potential and can self-oxidize non-phenolic lignin. When lignin is used alone, the C–H vibration peaks of $–CH_3–$ and $–CH_2–$ at 1462, 2854 and 2925 cm^{-1} were weakened compared to the original lignin, indicating the cleavage of benzene ring side chain groups (Perveen and Noreen, 2022). Furthermore, MnP contains abundant Mn^{2+} ions, which are oxidized to Mn^{3+} in the enzymatic reaction. Then, Mn^{3+} oxidizes phenol to produce phenoxy residue. However, MnP has a low REDOX potential and can only oxidize and degrade phenolic lignin (Wong, 2009). After treatment of lignin with MnP alone, the peak strength at 834, 1225, 1510, 1711, and 2925 cm^{-1} decreased, indicating that MnP destroyed the structure of lignin, albeit less effectively than Lac and LiP. Overall, Lac can act synergistically initiate enzymatic reactions with LiP and MnP, enhancing lignin degradation beyond what is achievable with Lac alone (Aleksandar et al., 2013).

2.2.4. Pyrolytic lignin

Pyrolytic lignin (PL) mainly originates from the pyrolysis of lignin components in biomass, and is a major constituent of heavy organic compounds in bio-oil (Fig. 3(d)) (Zhang et al., 2021). Bio-oil contains 15%–23% of PL depending on the biomass and pyrolysis condition (Stankovikj et al., 2017; Mortaza et al., 2019; Tuya et al., 2004; Qu et al., 2013; Han et al., 2019). Due to the high content of PL in bio-oil, extracting PL from bio-oil is the most commonly used method for obtaining PL. PL is mainly composed of phenolic compounds with benzene ring structure and is relatively hydrophobic, resulting in poor solubility in water. During the aging process of bio-oil, PL easily settles in solid form or separates as a heavy, thick liquid at the bottom of the container (Zhang et al., 2021). Therefore, cold water precipitation is an effective method for separating PL from bio-oil (Zhang and Wu, 2019; Scholze and Meier, 2001; Wen et al., 2016; Wang et al., 2015). PL is rich in benzene rings, which can be used to produce resins by condensation with aldehydes (Arjan and Hero, 2014).

The water-insoluble part of the oil is called PL (Dobele et al., 2009); Dolele et al. prepared pyrolysis oil in a two-stage ablative pyrolysis reactor using wood chips of Populus tremula, Alnus incana, Fraxinus excelsior and mixture of hardwood species (HM) (technological waste) as raw materials. The water-soluble (Py-C) and insoluble (Py-L) products of pyrolysis oil from different wood sources were obtained, and the water-soluble products were mainly fractionated. In the experiment, wood chips were cooked in an autoclave with 5% NaOH for 3 h to prepare alkaline lignin, and then pyrolyzed in a two-chamber pyrolysis device to obtain pyrolytic oil. The pyrolytic oil was dropped into the ice water during a stirring process at a rate of 6000 rpm, followed by filtering, washing, and drying in a vacuum to obtain PL (Scholze and Meier, 2001). The ABTS+ (cation-radical obtained from 2,20-azino-bis (3-ethylbenzo-thiazoline-6-sulphonic acid) diamonium salt), DPPH (free-radical obtained from 2,2-diphenyl-2-picrylhydrazyl hydrate), and hypoxanthine-xanthine oxidase assays confirmed the high antioxidant capacity of PL. Therefore, PL can be considered a promising natural macromolecular antioxidant, capable of competing with other low molecular commercial antioxidants.

The above discussed four extraction methods have different advantages and disadvantages: (1) Sulfate lignin extraction is performed under high temperature and pressure, where sulfate reacts with the phenylpropanoid structure in lignin, leading to its separation. However, it generates significant waste, such as “black liquor,” which is highly alkaline and contains corrosive and polluting substances, requiring costly treatment processes like neutralization, precipitation, and filtration. (2) Organic solvent lignin extraction relies on the affinity between organic solvents and specific lignin functional groups to selectively dissolve and extract lignin. While this method preserves lignin structure and yields high-purity lignin, its high cost and complex process limit large-scale applications. (3) Enzymatic lignin extraction uses specific enzymes (e.g., ligninase, cellulase, and hemicellulase) to hydrolyze lignin under mild conditions, allowing selective degradation while maintaining structural integrity. Though environmentally friendly, this method is expensive and time-consuming. (4) Pyrolytic lignin extraction involves high-temperature decomposition, breaking chemical bonds (e.g., C–C and C–O) to form gases, bio-oils, and solid residues, offering a rapid process that converts lignin into valuable chemicals. However, high temperatures may degrade lignin's structure, affecting its properties and applications. Ultimately, selecting the appropriate extraction method depends on the intended application, ensuring optimal utilization of lignin's multifunctional potential.

3. Modification and functionalization of lignin

With the depletion of shallow and middle-deep oil and gas resources, the exploration and development of oil and gas from deep and ultra-deep reservoir have become an important direction (Dou et al., 2022). However, due to the high bottomhole temperature and pressure of deep and ultra-deep reservoirs, as well as complex geological conditions (including the presence of salt and salt gypsum rock) of deep and ultra-deep reservoir, the performance of lignin deteriorates under these harsh conditions. Therefore, further improving the temperature and salt resistance of lignin under harsh formation conditions is a necessary requirement for efficient and safe exploration and development of deep and ultra-deep oil and gas resources. To address these challenges, chemical modification of lignin, such as etherification, crosslinking, and polymer grafting methods have been explored (Dou et al., 2022).

3.1. Enhancing the thermal resistance

The thermal resistance of lignin can be enhanced through various strategies, including ionic complexing, chemical crosslinking, and surface grafting of temperature resistant polymers. For instance, lignosulfonate is widely used as treatment agents in water-based drilling fluid (WBDF). It is a by-product of the papermaking industry, a natural polymer material containing aromatic, phenolic hydroxyl, alcohol hydroxyl, carbonyl, carboxyl, and sulfonic acid groups. It possesses renewable, biodegradable, non-toxic, and can meet the requirements of environmentally friendly drilling fluid rheology, filtration and shale inhibition additives (Abdollahi et al., 2018). However, the thermal instability of lignosulfonates causes them to lose effectiveness at high temperatures.

To improve its temperature resistance, ferric chloride was added to the lignosulfonate, and a series of ferric chloride-lignosulfonate (FCLS) were prepared as shale inhibitors by complexing the lignosulfonate with Fe^{3+} (Fig. 4(a)) (Zhang et al., 2019). As a result, FCLS effectively inhibited the hydration expansion of shale at 105 °C (Kusrini et al., 2020; Jiang et al., 2020; Li et al., 2018; Luo et al., 2010). In another example, lignosulfonate was chemically cross-linked with starch and polyphenols to synthesize the copolymer SPL, which helped stabilize the dispersion of bentonite and maintain sufficient viscosity of WBDF at a high temperature of 150 °C (Li X.L. et al., 2021). Furthermore, functional monomers, such as 2-acrylamide-2-methylpropanesulfonic acid (AMPS), acrylonitrile (AN), and N, N-dimethylaminoethyl isobutyric acid (DMAEMA) were grafted on the molecular chain of lignin by grafting copolymerization, and the resulting grafted copolymer reduced the filtration loss of WBDF to 8 mL at an extremely high temperature of 260 °C (Chang et al., 2019).

3.2. *Enhancing the salt resistance*

In the process of oil exploitation, the intrusion of cationic electrolyte in salt-gypsum layer can make drilling fluid ineffective and hinder the smooth progress of drilling operations. Therefore, it is necessary to adjust the composition of drilling fluid and add specific chemicals to render drilling fluid with superior salt resistance. The aromatic ring in lignin possesses a strong electron cloud density, which gives the lignin molecules a strong electron cloud affinity (Sun et al., 2021). This feature enables lignin to interact with the ions in salt, forming a stable complex, that maintain its stability and function in a certain salt environment (Laurichesse and Avérous, 2014).

In order to further enhance the salt resistance of lignin, the incorporation of salt-resistant groups (e.g., acrylamide, 2-acrylamide-2-methylpropane sulfonic acid, and sodium p-styrene sulfonate) into lignin molecules have been widely reported. Using methanol as the reaction medium, ferrous ions as the catalyst, and benzoyl peroxide as the initiator, lignin-acrylamide copolymers (PLA) have been prepared via free radical polymerization. When the drilling fluid contained 0.7% NaCl and 0.3% PLA, the bentonite neither hydrated nor dispersed, which improved the yield point and apparent viscosity of drilling fluid to a certain extent (Chen G. et al., 2014; Abdollahi et al., 2018). Alternatively, EHL nanoparticles were prepared by intramolecular self-assembly method, followed by surface modification using silane coupling agent. Then, EHL nanoparticles reacted with acrylamide, 2-(N-3-Sulfopropyl-N, N-dimethyl ammonium) ethyl methacrylate and N-Vinyl-ε-caprolactam monomers to produce an environmentally friendly salt-responsive fluid loss reducer (EHL-ASN) with anti-polyelectrolyte effect (Fig. 4(b)) (Sun et al., 2021). When the content of electrolyte sodium chloride was 6.0 mol/L, the fluid loss of EHL-ASN/WBDF was only 4.0 mL, and it still had a low fluid loss value of 13.8 mL after treated at 150 °C for 16 h, showing a strong salt tolerance. This is because that in the presence of electrolyte, the ionic network structure of EHL-ASN was altered, and the EHL-ASN underwent a transition from collapsed spherical structures to extended chain structures. The sulfonic acid groups and cationic groups on the side chains of the molecular chains were adsorbed onto the clay particles, forming stable ionic bonds and hydrogen bonds with the clay. This effect reduced the aggregation of clay particles, leading to the formation of dense mud cakes under the

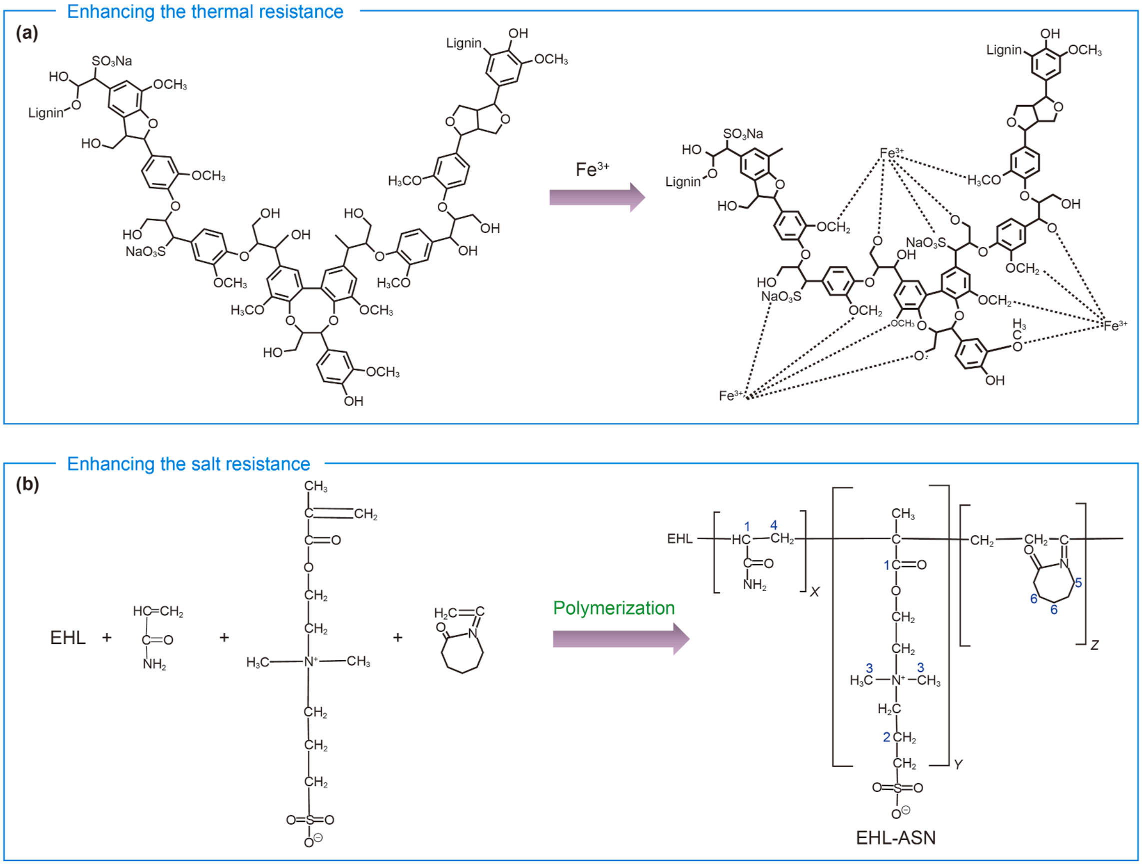

Fig. 4. Modification and functionalization of lignin: **(a)** Improved the thermal resistance mechanism (Zhang et al., 2019); **(b)** Improved the salt resistance mechanism (Sun et al., 2021).

action of pressure differential, thereby reducing the fluid loss. This study provided a pathway to address the instability and collapse issues of wellbore caused by the deterioration of WBDF performance due to the invasion of electrolyte during drilling in highly saline reservoirs (Sun et al., 2021).

4. Applications of lignin in oilfields

Owing to the excellent sustainability, superior biodegradability, and facile surface modification, lignin and its derivates have found wide applications in oilfields, including drilling fluids, fracturing fluids, enhanced oil recovery, and wastewater treating. In the drilling fluids, lignin and its derivatives can serve as rheological modifiers, fluid loss reducers, and shale inhibitors, leading to superior drill cutting carrying capacity and enhanced wellbore stability. In the fracturing fluid, lignin and its derivates are used as thickeners and breakers, increasing fracturing efficiency and protecting reservoirs. In enhanced oil recovery, lignin and its derivates function as surfactants and profile control agents. By forming dense and durable support networks in fractures, lignin-based polymers can improve the effective permeability of sandstone reservoirs and reduce the tension between water phase and rock surface, thus improving oil and gas recovery rate. In the treatment of oilfield wastewater, lignin and its derivates can effectively eliminate the oil-water mixture, heavy metals cations, and solid particles in wastewater by filtration, absorption, and flocculation. Table 1 summarizes the recent advances in the application of lignin and its derivatives in oilfields, including the role and source of lignin, modification method, and main findings.

4.1. Lignin in drilling fluids

Drilling fluid, also known as drilling mud, is used to aid the oil and gas drilling process. During drilling operations, drilling fluid is used for cooling the drill bit, cleaning the bottom hole, suspending cuttings, and balancing formation pressures (Li M.C. et al., 2021; Liu X.Y. et al., 2023). Water-based drilling fluid is a colloidal system that uses water as the dispersing medium, clay as the dispersed phase, and various chemical materials (e.g., rheological modifier, fluid loss reducer, and shale inhibitor) as functional additives (Khodja et al., 2010; Sun et al., 2020). It was reported that lignin and its derivates can serve as effective rheological modifier, fluid loss reducer, and shale inhibitor in water-based drilling fluids.

4.1.1. Rheological modifies

Cleaning the bottom of the well and suspending cuttings are the most important functions of drilling fluid, which are closely related to the rheological properties of drilling fluid (such as viscosity, shear force, shear thinning, thixotropy) (Ahmad et al., 2018). If the viscosity of drilling fluids is too high, it may lead to challenges and issues during the drilling process, such as reduced drilling efficiency, increased pumping pressure, and sticking and dragging of drill string (Saboorian-Jooybari et al., 2016). However, if the viscosity of drilling fluids is too low, it can also pose serious challenges and issues during the drilling process, such as poor hole cleaning, reduced cuttings suspension, and increased risk of blowouts (Li et al., 2015). Therefore, controlling the rheological properties of the drilling fluid is crucial. In recent years, a variety of natural and synthetic polymers have been used as rheological modifiers to achieve the required performance of drilling fluids (Allahvirdizadeh et al., 2016; Li et al., 2015; Saboorian-Jooybari et al., 2016). Tackifier and viscosity reducer are the two most commonly used rheological modifiers (Gudarzifar et al., 2020).

Lignin and its derivatives have many active groups, including aromatic group, phenol hydroxyl group, alcohol hydroxyl group, carbonyl group, carboxyl group, and sulfonic acid group (Kim et al., 2016; Shorey et al., 2021). The presence of these functional groups in lignin makes it easy to disperse into the drilling fluid and adsorb onto the surface of clay particles, forming a strong structure, thus enhancing the rheological properties of the drilling fluid. Therefore, lignin and its derivatives have been used as rheology modifiers for drilling fluids to improve their rheological characteristics . Chemical crosslinking and radical graft polymerization have been reported to modify lignin as high-performance tackifiers. For instance, Li X.L. et al. (2021) prepared a temperature-resistant additive (SPL) through chemical reaction of lignosulfonate, starch, and tea polyphenols using the glutaraldehyde as a crosslinking agent (Fig. 5(a)). SEM observation revealed that the SPL polymer exhibited an irregular vesicular structure with a particle size of 10 μm. When the shear rate was 0.1 s^{-1}, the viscosity of pure BT-WBDF after aging at 120 °C was 9137.158 mPa·s (Fig. 5(b)), which decreased significantly to 594.681 mPa·s after aging at 150 °C. After adding SPL additive, SPL promoted the hydration of clay particles and enhanced the colloidal stability. Throughout the entire range of shear rates, the viscosity of BT-WBDFs with SPL was consistently higher than that of pure BT-WBDFs. Specifically, at a shear rate of 0.1 s^{-1}, the viscosity of BT-WBDFs containing 2.0% SPL after aging at 120 °C and 150 °C reached 30186.35 and 18991.04 mPa·s, respectively. The addition of SPL maintained the high viscosity of BT-WDFs well at high temperatures. Alternatively, Chang et al. (2019) grafted N, N-dimethylacrylamide (DMAM), 2-acrylamide-2-methylpropanesulfonic acid (AMPS), and diallyldimethylammonium chloride (DMDAAC) onto nano-lignin to synthesize a novel amphoteric polymer (Nano-LS-g-DAD). It was found that the presence of 2.0% Nano-LS-g-DAD increased the AV, PV and YP of WBDFs from 6.8 mPa·s, 3.5 mPa·s, and 3.25 Pa to 30 mPa·s, 20 mPa·s, and 10 Pa, respectively; which was changed to 31 mPa·s, 24 mPa·s, and 6 Pa even after aging at 260 °C for 16 h. These results indicated the superior thickening performance of Nano-LS-g-DAD at room and high temperatures. The surface grafting of DMAM, AMPS and DMDAAC not only enhanced the anti-curling characteristics and heat resistance of lignin, but also made lignin have certain hydrophobic properties, which improved the thickening performance of lignin at room and high temperatures.

If the viscosity and shear stress of the drilling fluid are too high, it increases the flow resistance, leading to excessive energy consumption and significantly affecting the drilling speed. In addition, it can cause problems such as drilling bit stucking, difficulty in removing drilling chips from the ground, and challenges in degassing the drilling fluid. Therefore, it is essential to add additives into the drilling fluid to reduce the viscosity of the system (Chen et al., 2011). It was reported that the modified lignin with a low molecular weight can also serve as a viscosity reducer in BT-WDFs. For instance, Xu et al. (2022) prepared a lignin-based viscosity reducer (VR-H) through a three-step procedure, which includes 1) a polycondensation between lignin, humic acid, and formaldehyde solution to produce humic acid-modified lignin, 2) a free-radical polymerization between humic acid-modified lignin and a variety of vinyl monomers (i.e., acrylic acid, acrylamide, and N-(3-triethoxymethiloxanyl) propyl acrylamide) to generate vinyl polymer-grafted lignin, and 3) an amidation reaction between the vinyl polymer-grafted lignin and monoethanolamine to synthesize VR-H (Fig. 5(c)). Due to the presence of vinyl polymers, the VR-H possessed a significant quantity of hydroxyl, amide, and silicon hydroxyl groups on its surface. These groups allowed VR-H to be effectively absorbed onto the surface of clay particles. Consequently, the negatively charged carboxyl groups on the surface of VR-H enhanced the electrostatic repulsion between clay particles, thus breaking the “face-to-edge” and “edge-to-edge”

Table 1
Summarizes the recent advances in the applications of lignin in oilfields.

	Roles of lignin	Sources of lignin	Modification methods	Main findings	References
Drilling fluid	Fluid loss reducer, shale inhibitor	Sodium lignin sulfonate	SPL was obtained by chemical cross-linking of corn starch, tea polyphenols and sodium lignosulfonate at 100 °C.	The filtration loss reduction value at 150 °C is 17.5 mL, which can meet the general deep well drilling. SPL can disperse BT particles at high temperature, prevent the agglomeration of particles, and play a role in hydration and dispersion of clay particles.	Li X.L. et al. (2021)
	Fluid loss reducer, viscosity reducer	Sodium lignin sulfonate	DMAM, AMPS and DMDAAC were grafted to nano-LS in aqueous solution using ammonium persulfate and sodium bisulfite as initiators. A novel amphoteric polymer LS-g-DAD was synthesized.	The filtration loss of WBDF containing 2% LS-g-DAD at room temperature is only 4 mL, and the filtration loss after aging at 260 °C is 8 mL. When the NaCl content is 20%, the liquid loss at 200 °C for 16 h is 16.6 mL, indicating that the synthesized product has good temperature resistance and salt corrosion resistance	Chang et al. (2019)
	Fluid loss reducer, viscosity reducer	Organic solvent lignin	In the organic solvent process, formic acid was used as a solvent to extract lignin from LS, LOPBF and LCH fibers.	The apparent viscosity of LOPBF, LS, LCH lignin, and LASS (control group) was 11, 13, 12, and 8 mPa·s, respectively, before hot pressing, and increased to 40, 40, 27, and 42 mPa·s after hot pressing at 90 °C. The filtration volumes before hot pressing were 8, 9, 8, and 7 mL, respectively, and increased to 24, 24, 22, and 23 mL after hot pressing at 90 °C. Lignin from different sources exhibited a rapid increase in viscosity after the hot rolling process. However, with increasing lignin concentration, it effectively functioned as a diluent to reduce viscosity and also exhibited a certain degree of filtration reduction.	Mohd and Abdullah (2024)
	Fluid loss reducer, tackifier	Lignosulfo-nate	A series of lignosulfonate-based treating agent was prepared by Mannich reaction of lignosulfonate with formaldehyde and primary/secondary amine.	At room temperature, 0.3% lignosulfonate was added, and the filtration loss was 9.8 mL. The lignosulfonate-based residue in the waste drilling fluid after drilling operation can be biodegraded into organic fertilizer, which has certain environmental protection performance.	Chen et al., (2010)
	Fluid loss reducer	Lignosulfo-nate	A new chromium-free lignosulfonate diluent FTLS was prepared by cross-linking LS with natural tannin extract in the presence of formaldehyde and further chelating with ferrous ions.	When 0.5% FTLS was added into the base fluid, the API fluid loss is 23.6 mL at 170 °C. When the content of sodium chloride is 1.2% and the addition amount of FTLS is 0.5%, the API fluid loss is 26.7 mL. Compared with the widely used FeCr-Lignosulfonate diluent, the FTLS has better fluid loss reduction performance and high salinity tolerance.	Zhang and Yin (1999)
	Tackifier	Lignosulfo-nate	Graft copolymer LA was prepared by grafting AAm onto lignosulfonate using $H_2O_2/CaCl_2$ REDOX system and potassium persulfate thermal initiation system.	When the additive amount of LA-2 is 2.45 g, AV of drilling fluid changes from 1.75 to 3 cP, PV changes from 1.0 to 2.5 cP, and YP changes from 1.5 to 2.0 cP. At 36 % NaCl concentration, AV, PV and YP values all increase slightly, except for a slight decrease in YP value.	Mahdi Abdollahi (2018)
	Filtrate reducer	Sodium lignin sulfonate	With ammonium persulfate and sodium bisulfite as initiators, DMAA and γ-methylpropenoxy trimethoxysilane as reaction monomers were grafted with lignosulfonate to obtain fluid loss reducer LSDK.	When the copolymer concentration is 3%, the fluid loss after aging at 180 °C is 9.8 mL. When NaCl content is 2.40 mol, the viscosity of drilling fluid is 13.5 mPa·s and the filtration loss is 11.2 mL, indicating that the copolymer has strong salt tolerance.	Liu et al. (2020)
	Filtrate reducer	Enzymatic hydrolysis lignin	EHL produces intramolecular self-assembly nanoparticles in	When the content of electrolyte sodium chloride is 6.0 mol/L, the fluid loss of EHL-ASN/WBDF is only 4.0 mL,	Sun et al. (2021)

Table 1 (*continued*)

	Roles of lignin	Sources of lignin	Modification methods	Main findings	References
			tetrahydrofuran, and lignin nanoparticles are modified with silane coupling agent under acidic conditions. The modified lignin nanoparticles were reacted with acrylamide, 2-(N-3-Sulfopropyl-N, N-dimethyl ammonium) ethyl methacrylate and N-Vinyl-ε-caprolactam monomers in the presence of potassium persulfate as an initiator.	and it still has a low fluid loss value of 13.8 mL after treated at 150 °C for 16 h, showing a strong salt tolerance. The results of biotoxicity evaluation showed that the maximum half effective concentration (EC_{50}) of EHL-ASN was 51900 mg/L, which met the direct emission standard.	
	Filtrate reducer	Sodium lignin sulfonate	The fluid loss agent LSMP was synthesized using MIL-100(Fe)-NH_2 (20) as a photocatalyst, following the activation of sodium lignosulfonate and subsequent free radical polymerization with acrylamide (AM) and 2-acrylamide-2-methylpropanesulfonic acid (AMPS).	Due to its strong hydrogen bonding interactions, LSMP exhibited excellent rheological and filtration properties. The filtrate volume of drilling fluid containing 2.0 wt% LSMP was only 3.6 mL before aging and 4.6 mL after aging at 150 °C, representing reductions of 85.0% and 88.5%, respectively, compared to the pure liquid phase. Even under high-temperature and high-salinity conditions, LSMP demonstrated exceptional performance, with filtrate volume reductions of 96.58% and 86.52% in the presence of 25 wt% NaCl and 2.0 wt% $CaCl_2$, respectively.	Li Q. et al. (2024)
	Filtrate reducer	Borregaard	Lignin-based polymer (BioDrill FM400) was kindly provided by Borregaard, Norway.	When BioDrill FM400 was combined with xanthan gum, the plastic viscosity (PV) increased by 10% and 20%, while the yield point (YP) increased by 11% and 6% at ambient and hot rolling temperatures, respectively. Similarly, the filtrate volume and cake thickness were reduced, forming a filter cake less than 2 mm on the filter paper at both room and high temperatures. The filtrate volume and cake thickness were reduced by 63% and 42%, respectively.	Ali et al. (2025)
Fracturing fluids	Thickeners	Energy cane bagasse	Lignin containing-CNFs were prepared by DES pretreatment and high-pressure homogenization.	The presence of 0.5 wt% lignin containing-CNFs increases the viscosity of GG-VFF by 10%. Lignin containing-CNF/GG-VFF has excellent proppant suspension ability. After 24 h at 25 and 80 °C, the sand particles are evenly dispersed in the fluid without settling.	Liu C.Z. et al. (2022)
	Breakers	Hybrid poplar	Gelatin coated β-mannanase immobilized lignin (Ge@MIL) was prepared by using lignin from biomass as substrate for β-mannanase immobilization (MIL).	Ge@MIL can reduce the viscosity of fracturing fluid from 210 to 25 Pa·s at 25 °C, showing strong salt tolerance. The morphological integrity of the fracturing fluid did not change significantly after soaking in saturated NaCl solution for 8 h. The prepared Ge@MIL can release enzymes at specific temperature, rapidly degrade guar gum and reduce its viscosity at the same time. It is an excellent rubber breaker with delayed release function and has broad application prospect in low permeability oilfield.	Cong et al. (2022)
Enhanced oil recovery	Surfactants	Alkali lignin	Lignin salt betaine zwitterionic surfactant (LSBA) was synthesized by three steps of sulfonation, quaternization, and alkylation.	The hydrophile lipophile balance (HLB) values of LSBA-1, LSBA-2 and LSBA-3 were 14, 11 and 10, respectively. LSBA surfactants exhibited high surface activity with low surface tension and the critical micelle concentration (cmc) of LSBA-1, LSBA-2 and LSBA-3 were 4.36×10^{-4}, 4.45×10^{-4} and 1.04×10^{-3} mg/L. LSBA surfactants	Chen et al. (2018)

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Table 1 (*continued*)

	Roles of lignin	Sources of lignin	Modification methods	Main findings	References
				reduced the IFT between Xinjiang crude oil and brine down to ultra-low (10^{-3} mN/m) under weak alkaline conditions.	
	Surfactants	Alkali lignin	Oxidized sulfonated alkali lignin was synthesized from alkali lignin by sulfonation method. After sulfonation, lignin was aminated under different conditions. Finally, LS-A1 was synthesized from lignin amine and bromohexadecane by alkylation reaction.	The results show that ethylenediamine is more suitable for amination, and the optimal molar ratio of alkali lignin/ethylenediamine/formaldehyde is 1/2/1.5. The Hydrophile-Lipophile Balance (HLB) value of the synthesized product is 10. LS-A1 and NaOH can reduce the interfacial tension between Daqing crude oil and synthetic water to 10^{-2} mN/m at 45 °C.	Chen et al. (2018)
	Surfactants	Alkali lignin	A series of environmentally friendly anion-nonionic surfactant lignin polyether sulfonates (LPES) were prepared by alkoxylation, allylation, and sulfonation of alkali lignin as raw materials.	The critical micelle concentration (CMC) of LPES surfactants increases with the increase of ethylene oxide (EO) groups in the molecule, and the corresponding surface tension value is about 32–38 mN/m. HLB test shows that LPES surfactants have excellent emulsifying properties in oil/water (O/W) systems. In the presence of alkali, LPES surfactants can reduce the interfacial tension (IFT) between Daqing (North China) crude oil and brine to 10^{-3} mN/m.	Chen S.Y. et al. (2019)
	Surfactants	Kraft lignin	Kraft lignin was mixed with an amine (polyacrylamide or hexamethylenetetramine) and a surfactant sodium dodecyl benzene sulfonate in NaCl brine to prepare lignin-based surfactants.	The best formulation of lignin-based surfactant is hexamethylenetetramine (HMTA) as amine, and the concentration of lignin and sodium dodecylbenzene sulfonate (SDBS) is 2 wt%, showing good stability. The experimental results show that when the surfactant is injected at 1.0 PV, the oil recovery rate is 15% and the interfacial tension is 0.75 mN/m.	Ganie et al. (2019)
	Surfactants	Alkali lignin	Lignin nanoparticles (LNPs) were synthesized from alkali lignin using an antisolvent method, where an aqueous solution of the anionic surfactant nonpolyolefin sulfonate (AOS) served as the antisolvent. The prepared LNPs were then combined with AOS and coconut diethanolamide (CDEA) to formulate nanofluids.	These nanofluids exhibited excellent stability under high-salinity conditions and demonstrated significant properties, including reduced oil-water interfacial tension and enhanced emulsion stability. At a salinity of 5000 ppm, the AOS solution reduced the interfacial tension (IFT) to 10^{-1} mN/m, while the AOS/LNP system further decreased it to 10^{-2} mN/m.	Zhang et al. (2024)
	Profile control agent	Reed paper black liquor	Lignin was sulfonated to obtain high temperature resistant modified lignin (CRF), and the composite solution containing CRF and CX-5 at a certain concentration was used as a modulated system (PCS).	The optimal formulation of PCS is 0.3% CRF as oil displacement agent and 0.5% CX-5 as blowing agent. The experimental results show that PCS can effectively improve the reservoir heterogeneity and increase the recovery rate by 28.7%.	Dong et al. (2023)
Oilfield wastewater treatment	Oil-water separators	Acetonitrile extracted Lignin (AEL)	The preparation of AEL-FP membrane consists of two steps:1) coating a thin layer of lignin on the surface of the filter paper, and 2) adhering silica nanoparticles to the pre-coated filter paper to form a micro-nano binary structure.	The separation efficiency of AEL-FP is greater than 90%, and it has good applicability to various oils. The recyclability of AEL-FP was evaluated. After 5 cycles, the oil removal efficiency of AEL-FP remained at 96.6%.	Gong et al. (2021)
	Oil-water separators	Kraft lignin	The fluoride-free microscale porous material composed of silane-modified lignin particles has strong superlipophilic	The porous materials composed of silane-modified lignin can not only selectively absorb oil from immiscible oil-water mixtures, but also effectively separate oil-water emulsions stabilized by surfactants. The results showed that	Yu et al. (2021)

Table 1 (*continued*)

Roles of lignin	Sources of lignin	Modification methods	Main findings	References
		and superhydrophobic properties.	the separation efficiency of the material was higher than 99%, and the separation efficiency of the surfactant-stabilized oil-water emulsion was higher than 98%.	
Heavy metal absorbers	Sodium lignin sulfonate	Lignin-based adsorbent (SLCA) was prepared by free radical polymerization of acrylic acid with sodium lignosulfonate and citric acid.	The optimized SLCA adsorbent has a maximum adsorption capacity of 276 mg/g for Cu^{2+} and 323 mg/g for Pb^{2+}.	Wang et al. (2021)
Solid particles removers	Sulfate lignin	The water-soluble lignin-based cationic copolymer was prepared by polymerization of AM, DMC and sulfate lignin (KL) in aqueous solution.	Kaolin particle size increased from 4.7 to 22.3 μm and bentonite particle size increased from 6.1 to 31.9 μm after KAD1 treatment. In addition, the interfacial tension between water and kaolin and water and bentonite decreased from 12.3 to 5.8 mN/m.	Agha et al. (2019)
Oil absorbent	Alkali lignin	The oil absorber LPU-RGO-ODA foam was synthesized by grafting polydopamine-reduced graphene oxide (rGO) and octadecylamine (ODA) onto the internal matrix of LPU foam substrate.	PU-rGO-ODA foam is an excellent oil absorber, and its oil absorption is 26–68 times of its own weight, which is much larger than commercial non-woven polypropylene oilabsorber. LPU-rGO-ODA foam also showed high oil absorption selectivity and good reusability in the repeated adsorption-extrusion cycle. By squeezing the oil-containing sponge, the oil absorbed in the adsorbent can be removed and collected, and the foam adsorbent can be reused 20 times in the next adsorption cycle.	Oribayo et al. (2017)
COD remover	Eucalyptus waste liquid	Calcium lignin adsorbent was prepared by direct lime calcification method using chemical mechanical pulping waste liquid as raw material.	The COD removal rate can reach 85.83% when lignin calcium is used to treat pulp and papermaking wastewater. The adsorption isotherm and kinetics agree with Langmuir model and pseudo-second-order kinetics model respectively. The conversion rate of calcium ions can reach 83.56% when the waste lignin calcium is calcined at high temperature to regenerate lime.	Li Y.J. et al. (2023)

microstructures formed between clay particles in drilling fluid, resulting in the remarkable viscosity reduction. In addition, the VR-H also interacted with other viscosity thickeners, which impaired the interaction between viscosity thickeners and clay particles as well as between the molecular chains of viscosity thickeners, leading to reduction of viscosity. Overall, the VR-H not only disassembled the rigid, interconnected microstructure of clay particles, it also deteriorated the interaction between other polymers and clay particles, and thereby lowered the drilling fluid viscosity (Huang et al., 2015). As a result, the addition of 1.0% VR-H decreased the plastic viscosity of BT-WBDFs from 71 to 55 mPa·s after aging at 200 °C (Fig. 5(d)), demonstrating a good viscosity reduction effect.

4.1.2. Fluid loss reducers

When the drill passes penetrates a high-permeability reservoir, the pressure differential caused by the column pressure of the drilling fluid exceeds the formation pore pressure. As a result, a large amount of free water from the water-based drilling fluid penetrates into the reservoir through the wellbore, causing damage to the oil and gas reservoir, leading to adverse consequences such as wellbore collapse, pressure difference plugging during the drilling process (Aftab et al., 2020; Raghuraman et al., 2007). The function of the fluid loss reducers is to minimize filtration loss of the water-based drilling fluid and prevent wellbore collapse by forming a low-permeability, flexible, thin, and dense filter cake on the wellbore (Chang et al., 2024).

Fluid loss agents used in WBDFs maintain hydration and dispersion of drilling fluids by introducing strong hydrophilic groups, such as sulfonic acid and carboxyl groups (Kusrini et al., 2020; Jiang et al., 2020; Li et al., 2018). Lignosulfonate has been widely used in lime water-based drilling fluids because of its low price and wide source, as well as the presence of a large amount of sulfonic acid and carboxyl groups on the surface (Gu et al., 2017). It is one of the most used drilling fluid treatment agents (Li and Liu, 2004). Chen et al. found that lignosulfonate can increase the apparent viscosity of drilling fluid by 3%, the dynamic shear force by 14%, and the static shear force by 47% at room temperature, and reduce the filtration loss (Chen et al., 2010). The fluid loss of the base drilling fluid at room temperature was 12.5 mL, and when 0.3% lignosulfonate was added, the fluid loss decreases to 9.8 mL. However, lignosulfonate products used in drilling fluids are often used in combination with systems containing iron, chromium and other components, which inevitably leads to environmental pollution (Yu, 2012).

In recent years, it has been proven that grafting functional polymers onto lignin surfaces is an effective strategy for developing high-performance lignin-based filtration reagents. For instance, Chang et al. (2019) grafted N, N-dimethylacrylamide (DMAM), 2-acrylamide-2-methylpropanesulfonic acid (AMPS) and

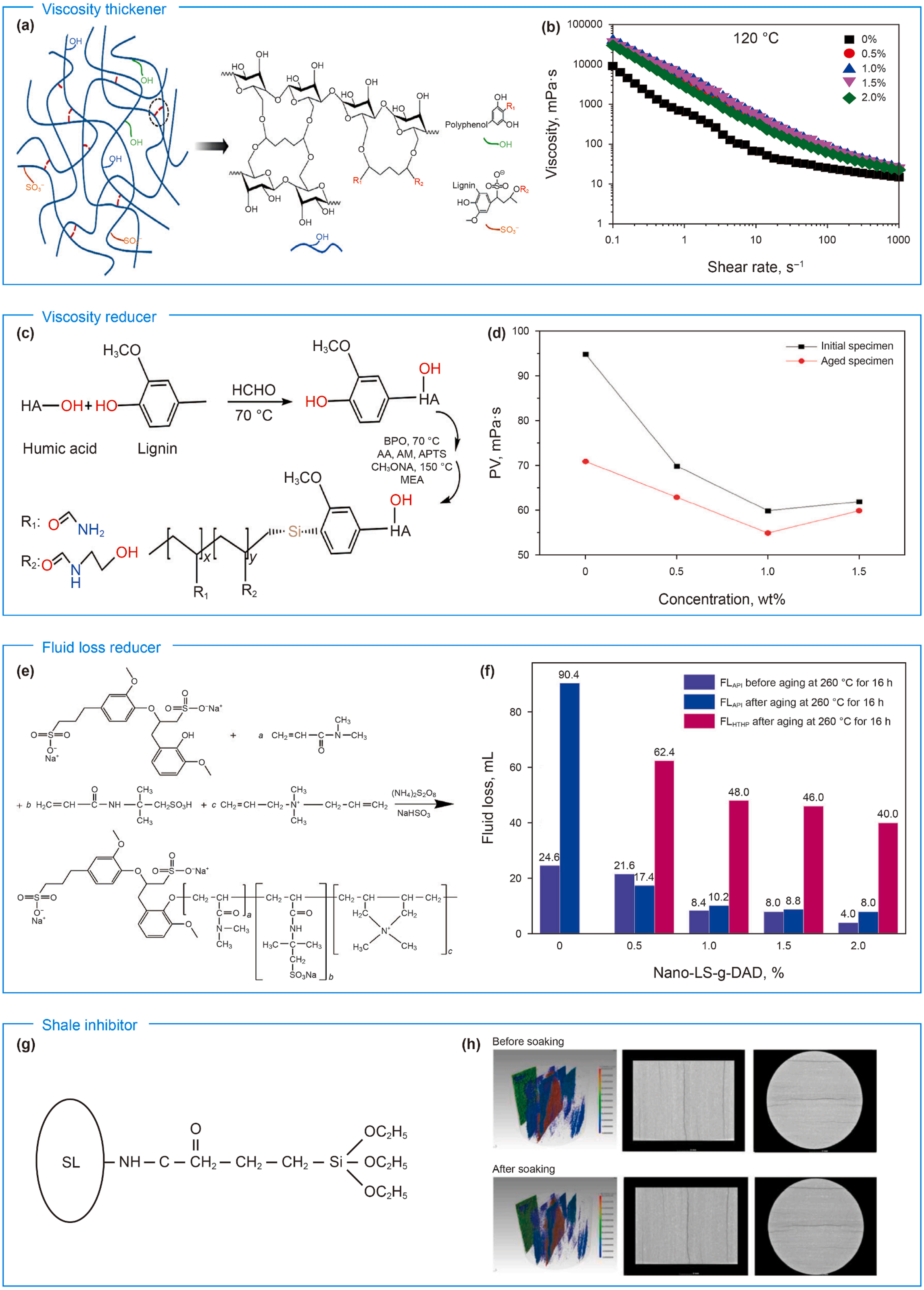

Fig. 5. Application of lignin in drilling fluid: **(a)** The molecular structure diagram of SPL (Li X.L. et al., 2021); **(b)** The relationship between viscosity and shear rate of BT-WDFs after aging at 120 °C under different SPL concentrations (Li X.L. et al., 2021); **(c)** Synthesis route of VR-H (Xu et al., 2022); **(d)** The impact of VR-H concentration on the rheological properties of drilling fluid: plastic viscosity (PV) (Xu et al., 2022); **(e)** Graft copolymerization of monomer LS, DMAM, DMDAAC, and AMPS (Chang et al., 2019); **(f)** FL values of WBDF containing Nano-LS-g-DAD at different concentrations before and after aging at 260 °C for 16 h (Chang et al., 2019); **(g)** Molecular structure of Si-SL (Su et al., 2022); **(h)** Tomographic images of shale in Si-SL-P solution (Su et al., 2022).

diallyl dimethylammonium chloride (DMDAAC) onto nano-lignin to synthesize a novel nano-lignin-based amphoteric copolymer-Nano-LS-g-DAD (Fig. 5(e)) (Chang et al., 2019). Due to the interaction of π–π bond in Nano-LS-g-DAD, the thermal decomposition of Nano-LS-g-DAD was observed above 281 °C. The filtration properties of Nano-LS-g-DAD were evaluated before and after hot rolling at 260 °C for 16 h. The results showed that after hot rolling, the filtration loss of Nano-LS-g-DAD/BT-WDFs was only 8.0 mL (Fig. 5(f)), demonstrating the superior capacity of Nano-LS-g-DAD in maintaining good filtration performance of BT-WBDFs under ultra-high temperature conditions (Chang et al., 2019). It is suggested that introducing DMDAAC into the polymer enhanced the rigidity of the molecular chain, thereby improving the temperature resistance of the copolymer (Li et al., 2014). However, when the polymer encountered salt, the molecular chain curled, which impaired its filtration performance (Yan, 2013). To overcome this bottleneck, Sun et al. have developed an environmentally friendly salt-resistant zwitterionic polymer based on enzyme-promoted lignin nanoparticles (EHL-ASN) as a filtration and reduction additive for WBDF (Sun et al., 2021). When electrolytes were added to EHL-ASN/WDFs, the EHL-ASN molecular chains expanded. In addition, the sulfonic acid and cationic groups formed stable ionic bonds with clay particles, while the amide groups formed hydrogen bonds with hydroxyl groups on the clay particles. The ionic and hydrogen bonds formed between EHL-ASN and clay particles ensured the tight adsorption of EHL-ASN on the clay particle surface, forming a stable spatial network structure. Therefore, in the presence of electrolytes, the clay particles maintained a stable dispersion structure, preventing aggregation and flocculation. This formed a dense mud cake under the action of pressure differential, and hence reduced the filtration loss of the WBDFs. When it was heated for 16 h at 150 °C, the filtration loss of WBDFs containing 2.5% EHL-ASN under saturated salt condition was significantly reduced from 194 to 4 mL, with a decrease of 97.9%. In addition, EHL-ASN also exhibited a certain salt tolerance. For instance, when the electrolyte content was 0, the fluid loss of EHL-ASN/WDFs before and after hot rolling at 150 °C was 25.0 and 47.0 mL, respectively. However, when the sodium chloride concentration was 6.0 mol/L, the fluid loss of EHL-ASN/WBDFs before and after hot rolling was only 4.0 and 13.8 mL, respectively. The mud cakes of EHL-ASN/WBDFs were also studied from both macroscopic and microscopic aspects. With the increase in the amount of EHL-ASN added, the mud cakes of EHL-ASN/SSBM became smoother and thinner, in the absence of micro-cracks and pores, demonstrating good filtration reduction performance.

Alternatively, under high temperature and high salinity conditions, lignosulfonate has been grafted with different functional polymers to improve its filtration performance. For instance, Abdollahi et al. used $H_2O_2/CaCl_2$ REDOX and potassium persulfate (KPS) thermal initiation systems to graft acrylamide (AAm) onto lignosulfonate (LS) to obtain lignin grafted copolymer (LA) (Abdollahi et al., 2018). Bentonite based tap water (TWF), seawater (SWF) and NaCl salt-saturated water (SSWF) were compounded with LS and LA to prepare different drilling fluids, respectively. The results indicated that the graft copolymer LA exhibited excellent thermal stability and salt resistance. The LS chain in the graft copolymer was responsible for thermal stability, while the PAAm chain improved rheological and filtration performance under high salinity. The graft copolymer synthesized by adding 20% AAm exhibited a gel-like structure in SSWF fluid, which effectively improved the filtration property of drilling fluid at high temperatures. It was reported that the addition of 2.45 g (i.e., 0.7 wt%) LA to WBDFs reduced the fluid loss value from 23 to 18 mL, demonstrating their effectiveness as fluid loss reducers in WBDFs. With ammonium persulfate and sodium bisulfite as initiators, Liu et al. (2020) grafted N, N-dimethylacrylamide (DMAA) and γ-methylacryloxy-trimethoxy-silane (KH570) onto sodium lignosulfonate (LS) in aqueous solution, and synthesized a novel LS/KH570/DMAA grafted copolymer as fluid loss reducers (Liu et al., 2020). The filtration performance of water-based drilling fluids with different concentrations of LS/KH570/DMAA grafted copolymers were evaluated. The results showed that when the copolymer concentration is 3%, the fluid loss of BT-WDFs is only 6.2 mL after aging at 180 °C for 16 h. Then, the filtration mechanism was investigated by measuring the distribution of clay particles in the filtrate and the zeta potential of the water-based drilling fluid. The copolymers were adsorbed onto the surface of clay particles and connected the clay particles tightly, which is beneficial to improve the compactness of the filter cake and reduce the filtration loss. The water-based drilling fluid containing LS/KH570/DMAA graft copolymer showed excellent filtration performance under calcium salt attack. The filtration loss of WBDF was only 14.4 mL when the concentration of $CaCl_2$ was 0.036 mol and aging at 180 °C for 16 h. The grafted copolymer can be used as an efficient fluid loss reducer in deep well operations.

4.1.3. Shale inhibitors

Wellbore instability is considered as one of the most challenging issues during the drilling process. Wellbore instability mostly occurs in shale rock, because about 50% of shale rock consists of clay minerals, which are mainly divided into kaolinite, illite, and montmorillonite. Clay minerals can easily hydrate due to water absorption, thus causing clay expansion and affecting wellbore stability. In addition, when water-based drilling fluids are used in shale formation, the risk of shale hydration expansion is further exacerbated. Hydration can further exacerbate issues such as contamination of underground aquifers, accumulation of solids in mud, increased torque and drag, and stuck pipe, leading to reducing in drilling efficiency in shale formations. To address these challenges, drilling fluid additives known as shale inhibitors are often employed to mitigate shale hydration effects and maintain wellbore stability. These inhibitors help prevent clay dispersion, reduce water absorption by shale, and improve overall drilling performance. Several types of shale inhibitors have been developed, including inorganic salts (e.g., NaCl, KCl, $CaCl_2$, and NH_4Cl (Balaban et al., 2015)), low molecular weight organic compounds (e.g., potassium formate-HCOOK, ethylene glycol, diamine, cyanoprosilanol, and ionic liquids (Ahmed et al., 2019; Chu et al., 2020; Luo et al., 2017), as well as biopolymers and derivatives (e.g., cationic starch, graphene-grafted glucose glucose, polyarginine, chitosan grafted arginine, and Zizyphus spina-christi extract (Li et al., 2020; Ma et al., 2019; Rana et al., 2020). However, few of these materials have shown a balance between good shale inhibition capacity and eco-friendliness.

With the increasing demand for sustainable development in oil and gas industry, a series of studies have been conducted on using lignin as an eco-friendly shale inhibitor in recent years. Zhang et al. (2019) prepared a series of ferrichloride lignin sulfonates (FCLS) as shale inhibitors using ferrichloride and alkali lignin (LS) as raw materials, and studied their mechanism of action. The study showed that FCLS was comparable with conventional shale inhibitors. When immersed in 0.3% FCLS-2 solution for 12, 24, and 36 h, the change in the mud balls was minimal, and the surface remained smooth after 36 h, except for a few cracks. The significant inhibitory effect of FCLS-2 on the swelling process of clay was mainly due to the adsorption effect of FCLS-2 on the surface of the clay, which blocked or reduced the water infiltration into the clay, thus preventing the hydration and swelling of the clay. The driving forces for absorption included hydrogen bond, electrostatic interaction, and anchoring effect between FCLS and clay particles.

Although Zhang et al. proved that FCLS has good inhibitory properties as a shale inhibitor, the swelling rate was still high. In addition, the shale inhibition mechanism was not studied comprehensively. To address this concern, Su et al. (2022) modified alkaline lignin (LS) with silane coupling agent to produce silane-modified LS (Si-LS) (Fig. 5(g)), and investigated the shale inhibition mechanism of through a wide spectrum of analytic techniques, including shale microfracture propagation synthesis, X-ray diffraction (XRD) measurement, adsorption measurement, contact angle measurement, Scanning Electron Microscope (SEM) and Atomic Force Microscope (AFM) observation. Three types of silane-modified sulfonated lignin derived from masson pine (Si-SL-M), eucalyptus (Si-SL-E), straw (Si-SL-S), and phyllostachys bissetii (Si-SL-P) were used and compared (Su et al., 2022). They found that the adsorption performance of Si-SL-P was the best, with an adsorption capacity of 38.89 mg/g at 60 °C. The phenyl groups in the Si-SL molecular chain were beneficial for improving the thermal stability and reducing the hydrophilicity of the clay particles surface (Akpan et al., 2019). By capturing the X-ray attenuation information, 2D and 3D structural images of the interior of the rock before and after flooding are obtained (Ma et al., 2016), it is found that cracks of different widths are produced inside the core after the shale is immersed in water, and the shale soaked in the Si-SL series solution only produces tiny cracks of limited length and width (Fig. 6(h)), indicating that the solution of Si-SL series can effectively inhibit the formation and expansion of cracks caused by the hydration expansion of water-sensitive clay minerals in the shale. Since Si-SL can strongly adsorb on the shale surface, the formation and expansion of cracks can be prevented. A hydrophobic film was formed on the shale surface, which reduced the flow rate of water molecules in the liquid phase into the shale interior. The field evaluation of Si-SL-P as a shale inhibitor further validated the potential of Si-SL as a biodegradable shale inhibitor for drilling fluids.

The rheology, filtration, and shale inhibition properties of lignin-filled drilling fluids are intricately linked to the molecular structure and functional groups present in lignin, as well as the specific environmental conditions in which these fluids are used. The molecular structure of lignin, which includes a complex arrangement of phenolic rings and various side chains, plays a crucial role in determining how lignin interacts with other components in the drilling fluid. Functional groups such as hydroxyl, methoxy, and carboxyl groups influence the lignin's ability to interact with water, salts, and other additives, thereby affecting the fluid's viscosity, filtration rate, and stability. Moreover, the environmental conditions, such as temperature, pressure, and salinity, can significantly alter lignin's behavior in the fluid. For example, high temperatures or the presence of salts may lead to the modification or degradation of lignin, which in turn can impact its ability to inhibit shale swelling, control fluid loss, or maintain the desired rheological properties. Therefore, understanding the interplay between lignin's molecular characteristics and the operating environment is essential for optimizing lignin-filled drilling fluids for specific oilfield applications.

4.2. Lignin in fracturing fluids

Oil is an important resource worldwide and extensively used in various industries. With the increasing demand and the depletion of conventional oil reservoirs, attention has turned to the development of low-permeability reservoirs (Sun et al., 2011). In order to increase oil production, fractures are created near the wellbore. The most commonly used technique for this purpose is hydraulic fracturing (Barati and Liang, 2014), which employs fracturing fluid

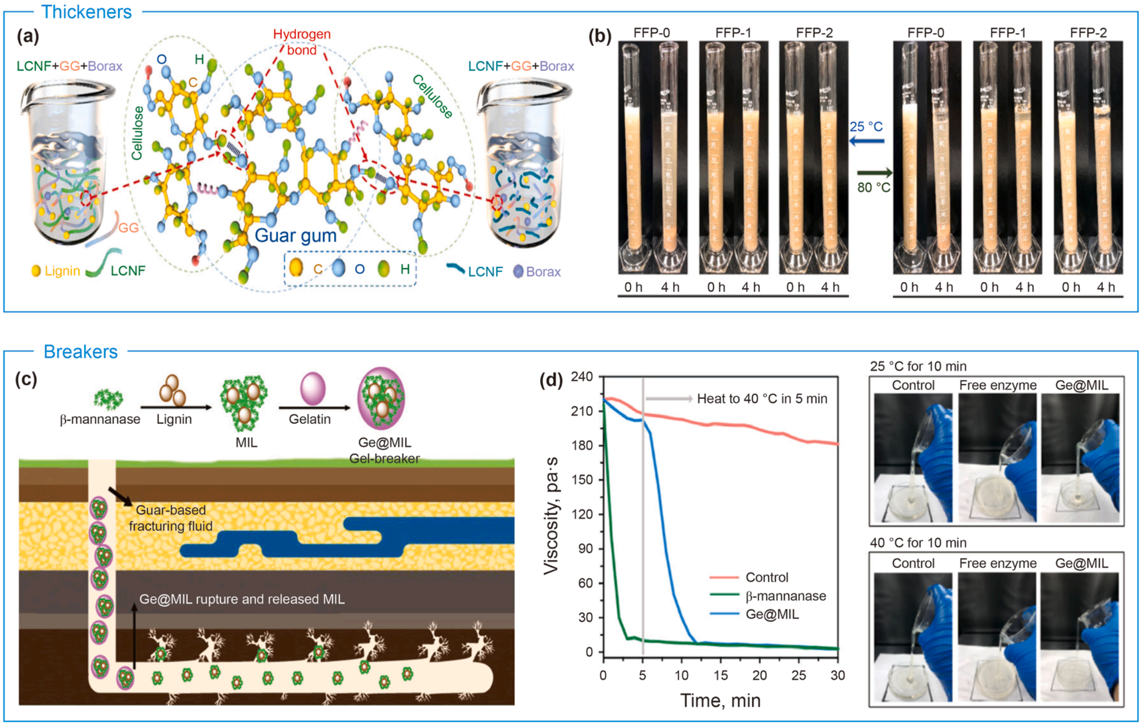

Fig. 6. Application of lignin in fracturing fluids: **(a)** Hydrogen bond interaction diagram of LCNMs and GG in VFF (Liu C.Z. et al., 2022); **(b)** The suspension performance and rheological properties of sand-containing LCNM/GG-VFF at 25 and 80 °C (Liu C.Z. et al., 2022); **(c)** Functional diagram of gelatin coated β-mannanase immobilized lignin (Ge@MIL) in the formation (Cong et al., 2022); **(d)** The impact of free β-glucosidase and Ge@MIL on the viscosity curve and apparent viscosity of guar-based fracturing fluid (Cong et al., 2022).

to crack the underground rock layers and form channels for the oil and gas flow. According to the function stage of fracturing fluid in the construction process, fracturing fluid can be divided into pre-fluid, sand-carrying fluid, and displacement fluid. Additionally, fracturing fluid systems include water-based fracturing fluid, oil-based fracturing fluid, emulsified fracturing fluid, acid-based fracturing fluid, and foam fracturing fluid. Water-based fracturing fluids are extensively used because of their safety, cleanliness, cost-effectiveness, and easy to control properties. Thickeners and breakers are commonly used in fracturing fluids. It was reported that lignin-based composites can be used as both thickener and breaker in fracturing fluids.

4.2.1. Thickeners

Thickeners are essential components of fracturing fluids, contributing to the success of the hydraulic fracturing process by controlling viscosity, aiding in fracture propagation, suspending proppants, and preventing fluid loss into the formation. Currently, the vast majority of water-based fracturing fluids use polymers as thickeners (Al-Muntasheri, 2014; Cao et al., 2021). Polymers can be divided into natural and synthetic polymers, which can be linear (non-crosslinked) or gelated (cross-linked). In natural polymers, the main polysaccharides used include guar gum and its derivatives (hydroxypropyl guar and carboxymethyl hydroxypropyl guar), xanthan gum, cellulose derivatives (carboxymethyl hydroxyethyl cellulose, carboxymethyl cellulose and hydroxyethyl cellulose). Crosslinkers include various multivalent metal ions (Cr^{3+}, Al^{3+}, Zr^{4+}, Ti^{4+}, $Fe^{2+}$and Fe^{3+}) or borate ions (Xia et al., 2020). In the synthetic polymer, polyacrylamide and its derivatives are mainly used, including hydrophobic modified polyacrylamide (HM PAAm) (Cadix et al., 2015; Wei et al., 2014). In natural polymers, the branched polysaccharide guar gum (GG), extracted from the Guar plant Cyamopsis tetragonolobus, has attracted attention due to its high molecular weight and excellent thickening ability. GG can be further cross-linked with different agents such as borax to form a viscoelastic fracturing fluid (VFF) (Wang et al., 2014; Wang S.B., 2016). However, the rheological properties of GG-based VFF are usually not suitable for harsh environments (e.g., poor stability when transporting large proppants). Therefore, it is crucial to further improve the rheological properties of GG gel, such as zero-shear viscosity, shear thinning behavior and yield stress (Fan et al., 2017).

In order to address this issue, Liu C.Z. et al. (2022) addressed this issue by enhancing the rheological properties of GG-based viscoelastic fracturing fluids (VFF) through the incorporation of lignin-containing cellulose nanomaterials (LCNMs) (Fig. 6(a)). Using bleached wood pulp as raw material, lignin-containing cellulose nanocrystals (LCNCs) and lignin-containing cellulose nanofibers (LCNFs) were prepared by microwave-assisted deep eutectic solvent (MV-DES, lactic acid and choline chloride) pretreatment combined with high-pressure homogenization (HPH). The suspensions of LCNF and LCNC both exhibited typical shear-thinning behavior and high viscoelasticity similar to gels. Compared with LCNC suspension, LCNF suspension had higher viscosity and dynamic modulus due to its higher aspect ratio. Subsequently, LCNFs were used as a rheological modifier for GG-VFF. Rheological analysis showed that the presence of 0.5 wt% LCNFs increased the viscosity of GG-VFF by 10%, attributed to the strong hydrogen bonding between LCNFs and GG in the VFF. In addition, borax could cross-link LCNFs and GG, resulting in a more tangled network structure, and therefore enhancing the rheological properties (Sun and Boluk, 2016). Subsequently, using 50-mesh sand as proppant, the proppant suspension capability of LCNF/GG-VFF was directly evaluated through settling experiments. The sand in the VFFs without LCNFs settled to the bottom of the measuring cylinder, while the sand grains within cavities with LCNFs remained well suspended in the measuring cylinder (Fig. 6(b)). The addition of LCNFs not only improved the rheological characteristics of GG-VFF, but also further enhanced its proppant suspension capability, thereby facilitating the uniform transport of proppants into the fractures.

4.2.2. Breakers

Once the fracturing process is finished, it becomes essential to remove the fracturing fluids from the fractures. However, due to their high viscosity, these fluids tend to linger in the formation. Biopolymers like guar gum, in particular, can remain in the fractures in significant quantities and pose a risk of causing substantial damage to the formation. Hence, breakers capable of reducing the viscosity of fracturing fluids are needed. However, the breakers are usually injected into the well along with the fracturing fluids, and the traditional breakers can only reduce the viscosity of the fluids during the fracturing process, which cannot break the fracturing fluids at the later stage of the fracturing. Therefore, it is important to develop high-performance breakers with superior delayed release functions.

The β-mannanase is an enzyme breaker, which can break the 1,4-glycosidic bond in the molecular chains of GG efficiently even under low temperatures. Lignin can be not only used as a substrate to immobilize β-mannanase, it also inhibits the activity of β-mannanase due to the hydrophobic nature of lignin. Furthermore, some polymers can be coated on the surface of lignin/β-mannanase composites to achieve the superior delayed release function. For instance, Cong et al. used lignin as substrate for β-mannanase immobilization (MIL), and then coated the MIL with gelatin to prepare the breaker Ge@MIL (Fig. 6(c)) (Cong et al., 2022). The SEM results indicated that the shape of Ge@MIL was roughly spherical, with an average size between 60 and 70 μm. At a gelatin concentration of 10% (10% Ge@MIL), the gelatin exhibited poor coverage of MIL, and it was difficult to form a spherical shape. As the gelatin concentration increased, the coating effect became more uniform and tended towards a spherical shape. The experimental results showed that the viscosity reduction effect of Ge@MIL after a 5-min delay was similar to that of free β-glucosidase, both decreasing from 210 to 25 Pa·s at 25 °C. This result indicated that Ge@MIL had a positive delayed effect on viscosity reduction of fracturing fluid. Subsequently, the sample was heated to 40 °C and kept warm for 5 min. Thanks to the release of MIL from Ge@MIL as well as the strong biodegradation effect of MIL on GG, the viscosity of GG-based fracturing fluid gradually decreased. After 10 min, the viscosity of the GG-based fracturing fluid reached the allowable discharge standard (5 Pa·s) (Fig. 6(d)). In addition, the Ge@MIL exhibited superior salt tolerance. Even after soaking in saturated NaCl aqueous solution for 8 h, there was no significant change in its structural integrity.

4.3. Lignin in enhanced oil recovery (EOR)

For most oil fields in the world, after the primary (artificial lift) and secondary (water flooding) oil recovery methods, at least half of the oil is still left in the reservoir (Porges, 2006). Tertiary oil recovery, also known as enhanced oil recovery (EOR), increases oil recovery by improving macro-sweep efficiency or micro-displacement efficiency. Chemical flooding mainly involves injecting oil displacement agents, such as profile control agents, surfactants, polymers, etc. (Olajire, 2014). Lignin can be modified by various chemical reactions and applied as surfactants and profile control agents to enhance oil recovery.

4.3.1. *Surfactants*

Surfactant flooding is a promising chemical method to improve oil recovery (dasani and Bai, 2011). Surfactants not only reduce the interfacial tension (IFT) between oil and water, but also increase the capillary number, and thereby significantly improved oil recovery efficiency (Fig. 7(a)) (Chen S.Y. et al., 2014; Jiang et al., 2014; Li et al., 2012). Currently, surfactant flooding is facing serious issues such as high adsorption loss, large consumption, and high cost, which significantly limit the widespread application of surfactants in enhancing oil recovery. Therefore, the developing efficient and economical surfactants has become an urgent task. Lignin contains a variety of functional groups in its molecular structure, such as carbonyl ($-CO^-$), hydroxyl ($-OH$), methoxy ($-OCH_3$), alkyl, etc., which can be modified through various chemical reactions (Alonso et al., 2004; Nadja Cachet, 2014). For instance, the introduction of hydrophilic and lipophilic groups can affect the water solubility and interfacial activity of lignin (Wu et al., 2024). Therefore, the synthesis of surfactants for enhancing oil recovery using biodegradable and low-cost lignin as raw materials has received widespread attention in recent years.

Lignosulfonates are the first lignin-based surfactant active component used in surfactant flooding. However, industrial lignosulfonates have strong hydrophilic groups and lack lipophilic long chain groups, which hinders their interfacial activity (Jiao et al., 2004). Chen et al. (2018) synthesized lignin sulfite betaine zwitterionic surfactant (LSBA) by three steps of sulfonation, quaternization and alkylation. By altering the carbon chain length of tertiary amines, LSBA-1, LSBA-2 and LSBA-3 were synthesized using (2, 3-epoxypropyl) decyl dimethyl ammonium chloride, dodecyl dimethyl tertiary amine and tetradecyl dimethyl tertiary amine as raw materials. The results indicated that when the surfactant concentration was 0.4%, the dynamic interfacial tension (IFT) of the Huabei crude oil reached its lowest value. Additionally, when the mass fraction of Na_2CO_3 was 0.4%, LSBA-1 effectively reduced the IFT between Huabei crude oil and saltwater, with lowest value reaching 10^{-3} mN/m (Fig. 7(b)). Although the surfactant prepared by this method has better application, the complex preparation process and high equipment requirements for quaternization hinder large-scale production applications in oil fields.

4.3.2. *Profile control agent*

During the reservoir development process, due to prolonged chemical flooding, the injected displacement agents tend to protrude outward along the high-permeability layers, leading to reduced flow resistance. Consequently, it becomes challenging to increase production in low-permeability areas (Valluri et al., 2017). The inconsistent oil recovery performance between high and low permeability zones inhibits uniform liquid production and water uptake, resulting in interlayer conflicts, particularly pronounced in heterogeneous reservoirs (Yang et al., 2007). Chemical technology is an effective way to control interlayer cross-flow in reservoir (Feng et al., 2020). By injecting high-temperature and anti-surge plugging agents into the target formation to block surge channels and selectively block high-permeability formations, the injected steam is diverted towards the medium and low-

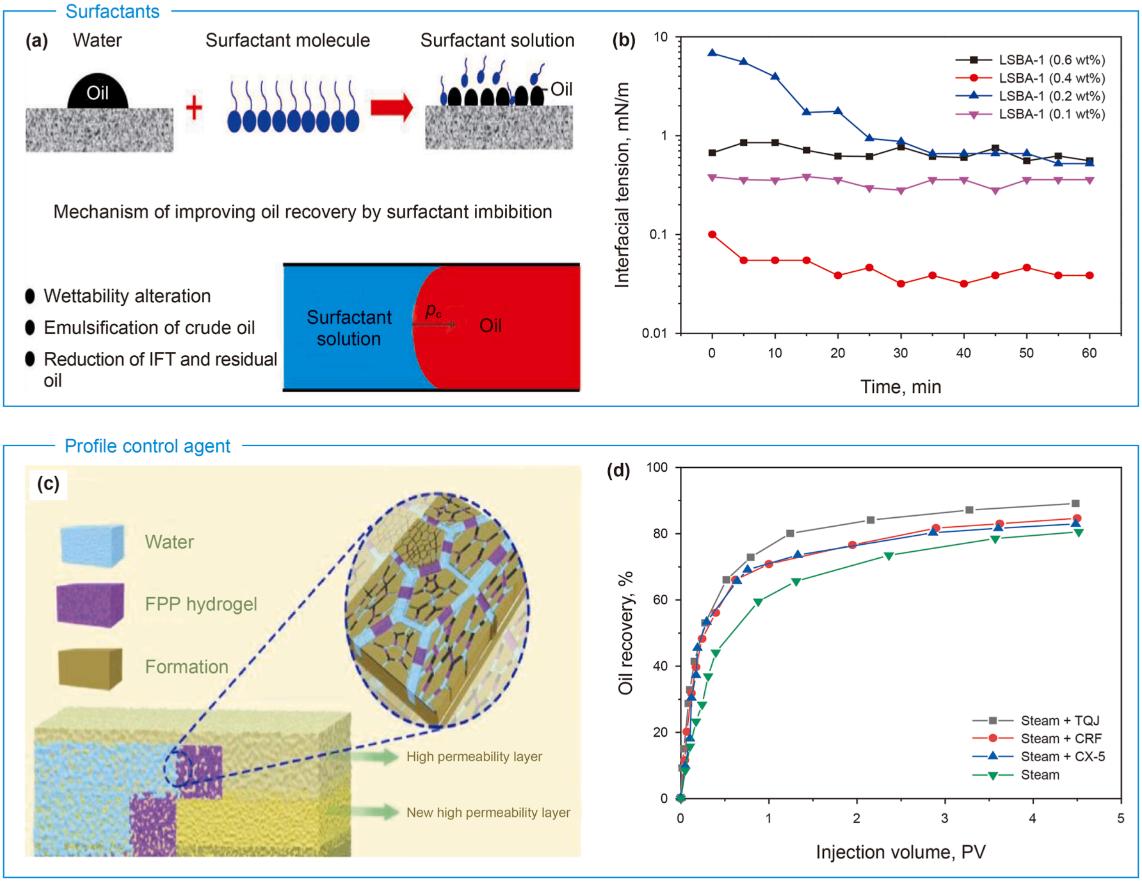

Fig. 7. Application of lignin in EOR: **(a)** Diagram of surfactants reducing interfacial tension between oil and water (Wang et al., 2023); **(b)** Concentration effects of LSBA-1 on the IFT between Huabei crude oil and saltwater at 54 °C (Chen et al., 2018); **(c)** Action mechanism diagram of profile control agent to enhance oil recovery in formation (Bai et al., 2020); **(d)** Oil displacement efficiency curve at 270 °C (Dong et al., 2023).

permeability formations, improving the gas invasion profile and increasing the oil recovery (Fig. 7(c)). Conventional plugging agents are ineffective in addressing the steam channeling issue during steam injection processes, leading to limited improvement in oil recovery.

Dong et al. (2023) developed a high-temperature resistant modified lignin (CRF) using the sulfonation method and combined it with foaming agent CX-5 to form a steam-driven temperature control system (PCS). Through single-point sand-packed tube displacement, three parallel sand-packed tube displacement, and large-scale sand-pack model displacement experiments, the effect of PCS-assisted steam flooding on improving oil recovery was evaluated. The optimal formulation for PCS was determined to be 0.3% CRF as the displacement agent and 0.5% CX-5 as the foaming agent. The experimental results showed that the recovery rate with PCS-assisted steam flooding at 270 °C was 7.9% higher than that with steam flooding alone. PCS effectively improved reservoir heterogeneity and increased the oil recovery by 28.7% (Fig. 7(d)).

4.4. Lignin in oilfield wastewater treatment

During the process of oil extraction and treatment, a large amount of oilfield wastewater is generated. Improper handling of oil field wastewater not only increases the cost of oil extraction, but also causes serious pollution (Ozgun et al., 2013). The composition of oilfield wastewater is quite complex, including surfactants, drilling cuttings, heavy metals, salts, oil, heavy metals, and more (Hedayatipour et al., 2017; Caliani et al., 2022; Meng et al., 2016). Currently, the most practical methods for treating oil field wastewater are coagulation, filtration, and oxidation. Lignin, as a biopolymer, can be separated from various lignocellulosic waste materials. Lignin contains various functional groups in the molecular structure of the benzene ring and side chain, such as hydroxyl (–OH), methoxy (–OCH_3), and carbonyl (–CO^-), can be modified through a variety of chemical reactions, and applied to the treatment of oilfield wastewater in various forms, such as particles, membranes, and aerogels via flocculation, filtration, and absorption mechanisms. This helps in ensuring that oilfield wastewater can meet industrial discharge standards and reduce pollution to soil and surface water (Alonso et al., 2004; Cachet et al., 2014).

4.4.1. Oil-water separators

Oilfield wastewater comes from a variety of processes, including drilling, fracturing, enhanced oil recovery, and well washing in oil and gas exploitation, all of which can introduce oils into oilfield wastewater. In addition, oilfield equipment, pipelines, and storage tanks may develop leaks or experience spills, leading to the contamination of groundwater or surface water with oil. Fatty acids and colloid asphaltenes in oil can act as natural emulsifiers, which form emulsified oil in oilfield waster in the process of thermal recovery, tertiary oil recovery and formation migration. Therefore, oil in oilfield wastewater mainly exist in the forms of emulsified oil, free oil, and oil slick. Among them, the oil slick and free oil have larger sizes, accounting for 70%–80% of the total oil. They can be removed by standard sedimentation processes or absorption methods. Emulsified oil and dissolved oil, on the other hand, have smaller size of less than 10 μm. They exhibit minimal merging between droplets, maintaining long-term stability and posing challenges in separation. This characteristic complicates the treatment of oilfield wastewater (Wang et al., 2020).

Filter paper is a type of paper-based composite material that can separate impurities from gas or liquid and recover useful substances. In recent years, lignin superhydrophobic modification methods have been applied to filter paper, giving it special functions such as oil-water separation capability (Tang et al., 2017). Gong et al. developed an oil-water separation membrane (AEL-FP) based on filter paper, acetonitrile extracted lignin (AEL) and silica nanoparticles (Fig. 8(a)) (Li et al., 2010). The preparation of AEL-FP membrane involved two impregnation steps: 1) coating a thin layer of lignin on the surface of the filter paper, and 2) adhering silica nanoparticles to the pre-coated filter paper to form a micro-nano binary structure, changing the filter paper's wettability from hydrophilic to superhydrophobic and oleophilic. The oil-water separation efficiency of *n*-hexane, petroleum ether, and chloroform was tested, and the results showed that the separation efficiency of AEL-FP was all above 90% (with the highest separation efficiency of 98.6% for trichloromethane) (Fig. 8(b)), indicating good applicability to various oils. The recyclability of AEL-FP was also evaluated. After five cycles, the oil removal efficiency of AEL-FP remained at 96.6%, indicating that AEL-PF has good recyclability. The above method is mainly for oil slick and dispersed oil in oilfield wastewater. However, the wastewater from oilfield often contains oil-in-water emulsion, this method remained challenging for the separation of emulsion oil (Ventikos et al., 2004). To address this concern, Yu et al. (2021) successfully prepared a fluoride-free micro-scale porous material composed of silane-modified lignin particles, which has strong super-oleophilic and super-hydrophobic properties. It was found that the silane-lignin coating can rapidly adsorb oil (dodecane) droplets (≈5 μL) within about 50 ms, which is due to the strong oleophilic property of silane-modified lignin. It was also found that a large number of nanometer to micron-sized pits were deposited on the surface of silane-lignin, which further induced strong capillary force to absorb oil. As a result, highly efficient separation of oil-water emulsion was achieved by a simple one-step method (i.e., by placing particles or clumps into the oil-water emulsion). The porous material composed of silane-modified lignin not only had the ability to selectively absorb oil from immiscible oil-water mixtures but also effectively separated oil-water emulsions stabilized by surfactants. The results indicated that the separation efficiency of the material reached over 99%, and the separation efficiency of surfactant-stabilized oil-water emulsions can reach over 98%. The recyclability experiment and SEM showed that the silane-modified lignin-based porous material still maintained its weight and microstructure after 20 cycles of separation, and had good separation efficiency.

4.4.2. Heavy metal absorbers

Oilfield wastewater has a complex composition, containing not only crude oil and water, but also a large amount of heavy metals, e.g., Cr^{6+}, Cu^{2+}, Pb^{2+}, Ca^{2+} and other heavy metal ions. Traditional techniques for removing heavy metals from wastewater include biological treatment (Raul and Guieysse, 2006), electrochemical techniques (Tonni et al., 2006), precipitation (Fu and Wang, 2011), oxidation (Matilainen and Sillanpaa, 2010), membrane filtration (Ventikos et al., 2004) and adsorption (Ulusoy and Simsek, 2013; Ngah and Hanafiah, 2008). Among them, adsorption is a commonly used method for removing toxic heavy metals from wastewater.

Wang et al. (2021) used sodium Lignosulfonate acid sodium and citric acid for free radical polymerization of acrylic acid to prepare an adsorbent (SLCA). The structure and properties of SLCA were characterized by TGA, XPS, FTIR, SEM and other characterization methods. After the adsorption of Pb^{2+} and Cu^{2+} on SLCA, it was observed that the pores on the surface of SLCA were covered with grooves, demonstrating the successful adsorption of heavy metal ions (Fig. 8(c)). At the same time, the adsorption performance of SLCA was investigated through adsorption experiments. The optimized SLCA adsorbent exhibited a maximum adsorption

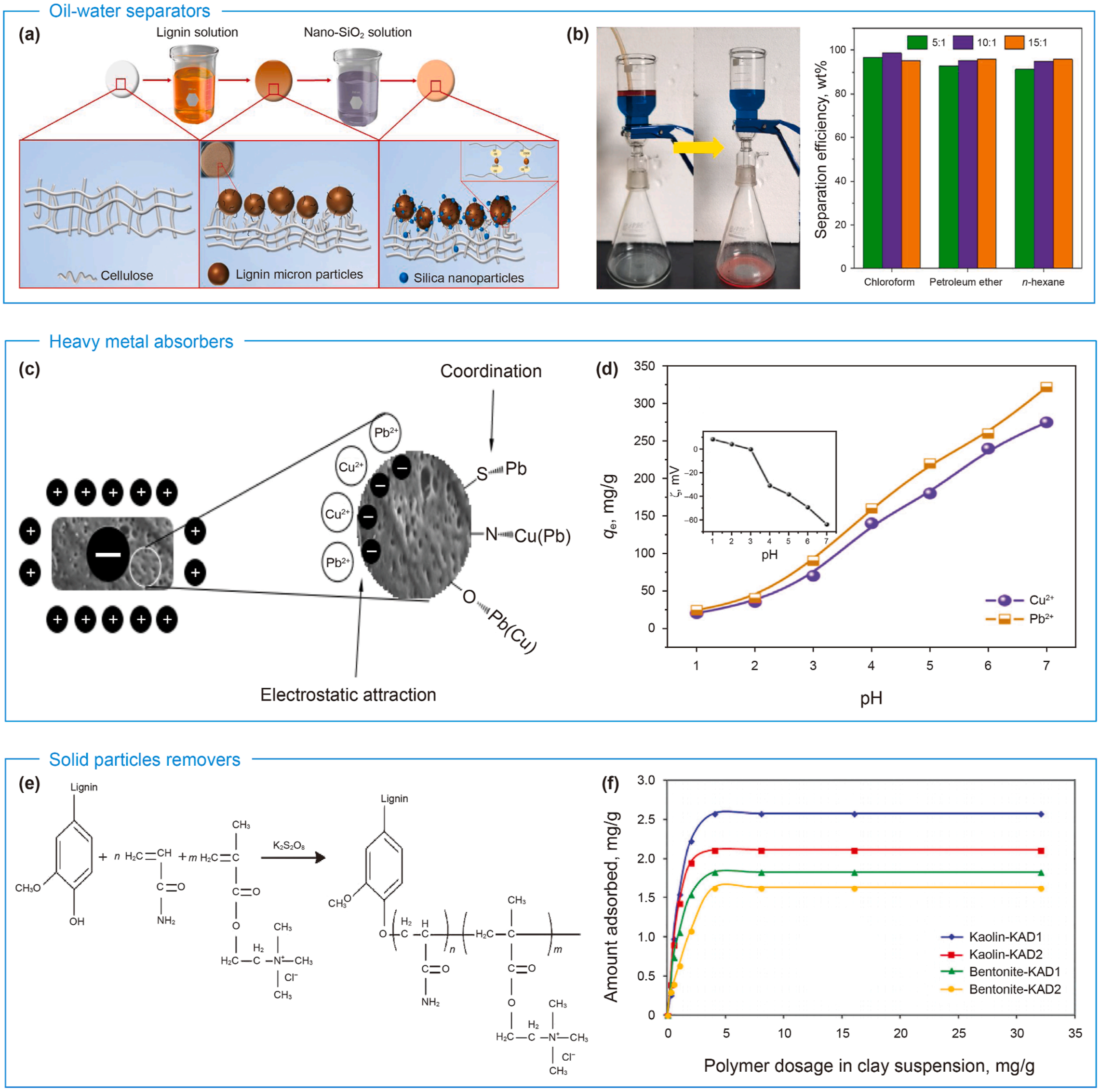

Fig. 8. Application of lignin in oil field wastewater cleaning: **(a)** Illustration on the filtration mechanism of paper/lignin/SiO_2-based membrane (Gong et al., 2021); **(b)** The separation efficiency of modified filter paper for different oil-water emulsions (water:oil mass ratio = 5:1, 10:1, 15:1) (Gong et al., 2021); **(c)** XPS spectra and adsorption mechanism of SLCA, SLCA/Cu, SLCA/Pb (Wang et al., 2021); **(d)** Effect of different solution pH on SLCA adsorption capacity (Wang et al., 2021); **(e)** Synthesis of copolymerization using KL, DMC and AM (Agha et al., 2019); **(f)** The adsorption amount of KL and KAD on the surface kaolin and bentonite (Agha et al., 2019).

capacity of 323 mg/g for Pb^{2+} and 276 mg/g for Cu^{2+} (dose = 0.01 g, $T = 20$ °C, pH = 7) (Fig. 8(d)). Additionally, Langmuir isotherm and second-order kinetic model were also established, indicating that the adsorption of Pb^{2+} and Cu^{2+} by SLCA belonged to monolayer chemical adsorption. XPS analysis confirmed that complexation and electrostatic attraction were the adsorption mechanisms. Oilfield wastewater contains both trivalent chromium (III) and hexavalent chromium (VI), and Cr (VI) can exist in the forms of chromate (CrO^{4-}), hydrochromate ($HCrO^{4-}$), or dichromate (Cr_2O^{7-}), with concentrations ranging from tens to hundreds of milligrams per liter. Due to repulsive electrostatic interactions, these Cr (VI) anions are usually poorly adsorbed by negatively charged soil particles in the environment, leading to their free migration in water environments (Park et al., 2008; Li et al., 2009). Therefore, the removal of hexavalent chromium from oilfield wastewater is considered a serious environmental problem. Albadarin et al. (2011) studied the use of H_3PO_4-activated lignin as a potential low-cost adsorbent for the removal of Cr (VI) from aqueous solutions at different concentrations (Albadarin et al., 2011). The results indicated that as the concentration of Cr (VI) increased from 10 to 100 mg/L, the equilibrium adsorption capacity (q_e) increased from 8.87 to 78.5 mg/g. Acid-base adaptation experiments found that pH value had a significant influence on the adsorption capacity of Cr (VI), and H_3PO_4-activated lignin had better adsorption performance for Cr (VI) under acidic conditions, with the maximum adsorption capacity at pH = 2.

4.4.3. Solid particles removers

Oilfield wastewater discharge contains a lot of suspended particles (e.g., bentonite, kaoline, sepiolite, palygorskite and clay mineral mixture), which can easily cause blockage and pollution to the formation (Ali, 2017). Flocculation method has been used for decades to treat suspended particles in wastewater, since most naturally occurring colloids carry negative charges, adding cationic polymers provides an effective strategy for removing suspended particles in wastewater (Petrak et al., 2015; Guo et al., 2018).

Agha et al. (2019) prepared water-soluble lignin-based cationic copolymers in aqueous solution by polymerizing acrylamide (AM), (2-methacryloyloxyethyl) trimethylammonium chloride (DMC), and sulfate lignin (KL) (Fig. 8(e)). Two cationic KAD polymers with the same molecular weight but different charge densities were used as flocculants to simulate clay suspensions. Two cationic KAD polymers with the same molecular weight but different charge densities were used as coagulants to simulate clay suspensions. The copolymer with higher charge density (KAD1) demonstrated superior adsorption capacity on kaolin particles (2.58 mg/g) and bentonite particles (1.83 mg/g) at a dosage of 4 mg/g (based on clay) compared to the copolymer with lower charge density (KAD2) (Fig. 8(f)). Additionally, the higher charge density of KAD enhanced its ability to alter the zeta potential and turbidity of clay suspensions. When the copolymer dosage was 4 mg/g, the kaolin particle size treated by KAD1 increased from 4.7 to 22.3 μm, and the bentonite particle size increased from 6.1 to 31.9 μm. The interfacial tension between water and kaolin, as well as water and bentonite, decreased from 12.3 to 5.8 mN/m. KAD1 exhibited stronger flocculants at different shear rates, showing higher regeneration affinity, and hence was more favorable for the flocculation of large particle contaminants in oilfield wastewater.

5. Conclusions

Lignin, a hydroxylated aromatic biopolymer found in plant cell walls, has demonstrated significant potential for oilfield applications due to its sustainability, biodegradability, and excellent rheological, mechanical, chemical, and thermal properties. Its incorporation in drilling fluids, fracturing fluids, enhanced oil recovery (EOR), and oilfield wastewater treatment offers an environmentally friendly alternative to traditional petroleum-based additives. However, its widespread application remains constrained by the harsh conditions of oil reservoirs, including high salinity, pressure, and temperature. To enhance its performance under these conditions, chemical modifications such as etherification, crosslinking, and polymer grafting have been explored, improving lignin's thermal stability, salt tolerance, and functional versatility.

In drilling fluids, modified lignin exhibits multifunctional capabilities as a rheology modifier, fluid loss reducer, and shale inhibitor, enhancing wellbore stability and optimizing drilling fluid performance. In fracturing fluids, it functions as both a thickener and a breaker, improving proppant transport while enabling controlled degradation of guar-based fluids. In EOR, lignin-based surfactants and profile control agents reduce interfacial tension and regulate permeability to enhance oil recovery efficiency. Additionally, in oilfield wastewater treatment, modified lignin demonstrates superior adsorption capacity for oil-water separation, heavy metal removal, and suspended particle flocculation.

6. Challenges and future prospects

Despite these advancements, further research is required to develop large-scale extraction methods, as well as optimize lignin's molecular design for improved reservoir compatibility, long-term stability, and multifunctional performance. Future efforts should focus on developing tailored lignin-based materials through advanced modification techniques, integrating lignin with nanotechnology, and exploring its synergistic effects with other oilfield additives. In particular, the development of lignin with superior thermal resistance and salt tolerance is critical to meet the practical demands of harsh formation conditions.

To address these challenges, research should focus on optimizing large-scale lignin extraction techniques, such as improving the efficiency of green solvents or enzymatic processes to make them more cost-effective and scalable. Furthermore, advanced modification methods, such as atom transfer radical polymerization (ATRP), reversible addition–fragmentation chain transfer (RAFT), and click chemistry, could be employed to precisely modify lignin with thermally resistant and salt-tolerant polymers (e.g., polyether-ether-ketone, polyamide, fluorine polymer), thereby enhancing lignin's stability under high-temperature and high-salinity conditions. Additionally, artificial intelligence can be implemented to design the molecular structure, leading to optimized performance and tailored functionalities. Last but not least, interdisciplinary collaboration between material scientists and petroleum engineers will be crucial for accelerating the transition of lignin-based technologies from laboratory research to large-scale industrial applications. Addressing these challenges will facilitate the large-scale adoption of lignin in oilfield applications, paving the way for more sustainable and efficient oil and gas development.

CRediT authorship contribution statement

Na-Na Song: Investigation, Formal analysis, Writing – original draft. **Mei-Chun Li:** Project administration, Conceptualization, Supervision, Writing – review & editing. **Xue-Qiang Guo:** Data curation, Formal analysis. **Ya-Xuan Zhang:** Investigation. **Ming-Yuan Xin:** Formal analysis. **Jin-Sheng Sun:** Project administration, Resources. **Kai-He Lv:** Methodology, Resources. **Chao-Zheng Liu:** Methodology.

Conflict of 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.

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