

Advances in catalytic pyrolysis of lignin toward aromatic hydrocarbon production

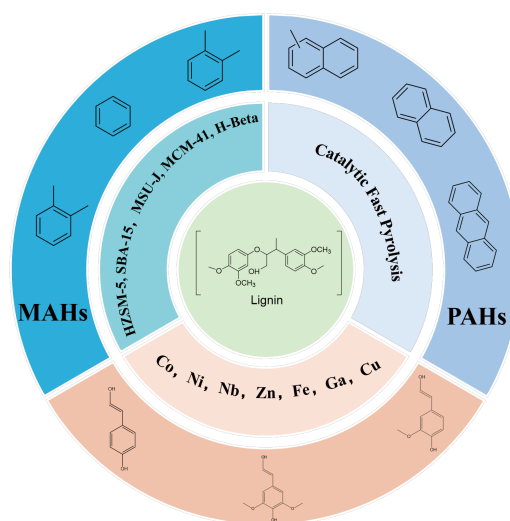
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Abstract: Aromatic hydrocarbons, as crucial chemical feedstocks, are widely utilized in the production of fuels, synthetic fibers, and pharmaceuticals, mainly originate from petroleum refining. The catalytic conversion of lignin to produce aromatic hydrocarbons represents one of the important pathways for achieving green production of these chemicals. The core of catalytic pyrolysis of lignin to produce aromatic hydrocarbons lies in the regulation of catalyst acidity and pore structure, optimization of reaction conditions, and the understanding of the conversion mechanisms. Therefore, this review summarizes the influence of pyrolysis methods, catalysts, and reaction conditions on the catalyzed pyrolysis of lignin for aromatic hydrocarbon production, systematically outlines the catalytic reaction mechanism, and provides an outlook on future key research directions and ideas, in order to give theoretical and technological guidance for this process.

Key words: lignin; aromatic hydrocarbons; pyrolysis; catalysts



Aromatic hydrocarbons, as essential chemical feedstocks that are widely utilized in the production of plastics, synthetic fibers, pigments, and dyes, are extensively applied in energy, daily chemical, and pharmaceutical industries. Currently, they are primarily derived from naphtha reforming and petroleum cracking^[1]. However, the scarcity, non-renewable nature, and environmental pollution associated with fossil resources have created an urgent demand for the development of a green, efficient, environmentally friendly, and renewable route for the production of aromatics. Lignocellulosic biomass, as an important component of renewable feedstocks, is mainly composed of cellulose (40%–50%), hemicellulose (20%–

30%), and lignin (10%–25%)^[2–3]. As shown in Figure 1^[4], lignin, one of the main constituents of lignocellulose, is composed of three phenylpropanoid structural units: guaiacyl (G), syringyl (S) and *p*-hydroxyphenyl (H), which are connected through C–O–C bonds (β -O-4, α -O-4, γ -O-4), C–C bonds (5–5, β -1, β -5) and a small proportion of ester bonds^[5]. It is widely recognized as the primary source of biomass-derived aromatic materials^[6]. Moreover, conventional pulping processes generate large quantities of black liquor, which is mainly composed of lignin. Currently, this by-product is discharged directly or incinerated for power generation, leading to environmental pollution and the waste of carbon resources.

Received: 2025-08-16; Revised: 2025-09-19

Foundation items: Supported by the National Natural Science Foundation of China (Youth Program, 22308178), the Shandong Provincial Natural Science Foundation (ZR2023MB040 and ZR2024MB071), the Jinan “New 20 University Policies” Project (202228018), the Pilot Project of Integration of Science, Education and Industry (2024ZDZX07) and the Jinan University-Local Integration Development Strategic Project (JNSX2024040).

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DOI: 10.1016/S1872-5813(25)60618-9 CSTR: 32157.14.2097-213X.2025.JFCT.0168

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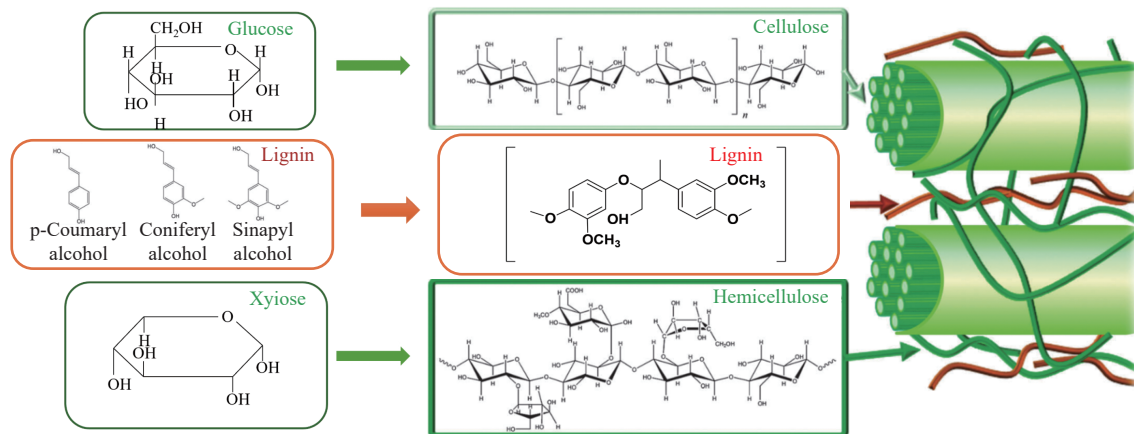


Fig. 1 Structure of lignocellulosic biomass depicting cellulose, hemicellulose and lignin^[4]
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The conversion of lignin in the black liquor into aromatic hydrocarbons not only mitigates environmental pollution but also enables the high-value utilization of lignin^[7–8]. Therefore, the development of lignin-based technologies for the production of aromatic hydrocarbons holds significant environmental and economic importance.

Pyrolysis, as a key strategy for the thermochemical conversion of biomass, is effective for the depolymerization of lignin macromolecules under high-temperature and oxygen-free conditions to produce bio-oil that is mainly composed of phenolic derivatives and aromatic hydrocarbons. To increase the content of benzenoid compounds in the lignin-derived bio-oil, the catalytic fast pyrolysis (CFP) process has been employed to selectively depolymerize lignin and remove oxygen-containing functional groups. This process facilitates the conversion of lignin into high-value light aromatic hydrocarbons, such as benzene, toluene and xylene^[9–10]. Therefore, this review systematically summarizes the recent advances in the catalytic pyrolysis of lignin for the production of aromatic hydrocarbons, with particular emphasis on the sources and structural characteristics of lignin, the pyrolysis pathways, the catalysis systems, the reaction conditions, and the conversion mechanisms. In addition, the key research directions and challenges associated with the catalytic pyrolysis of lignin are discussed, to provide theoretical guidance and technical support for its industrial-scale application.

1 Sources and structural characteristics of lignin

1.1 Sources of lignin

The global plants have been reported to sustainably synthesize approximately 5×10^{10} t of lignin annually through the process of photosynthesis^[11]. In addition, the conventional pulp and paper industries generate more than 5.0×10^7 t of lignin as by-products every year, and the cellulosic ethanol industry is projected to contribute an additional 2.2×10^8 t of lignin waste by the year 2030^[12].

1.2 Structure of lignin

As shown in Figure 2, the chemical bonds in lignin are primarily classified into three types of linkages, including ether bonds accounting for 60%–70%, carbon–carbon bonds accounting for 30%–40%, and a small proportion of ester bonds that are mainly present in the lignin derived from herbaceous plants. The ether bond network is composed of three major substructures: the β -O-4, α -O-4 and γ -O-4 linkages between the propyl side chains and the aromatic rings, the 4-O-5 linkages between the aromatic units, and the α -O- β and α -O- γ cross-linkages between the side chains. The carbon–carbon bond framework mainly consists of rigid structures such as the 5–5' biphenyl linkages and the β -1 and β -5 diphenylpropane linkages, together with a small proportion of special bonding motifs, including the β -6 and α - β -O linkages^[10]. In addition, lignin is a complex aromatic biopolymer composed of three types of phenylpropanoid monomers, *p*-hydroxyphenyl (H), guaiacyl (G) and syringyl (S), which are interconnected in a non-linear and random manner through the ether and carbon–carbon bonds^[13]. As illustrated in Figure 3, lignin macromolecule exhibits significant structural complexity and contains a large number of oxygen-containing functional groups, such as hydroxyl, carbonyl, carboxyl, and methoxyl groups, thereby imparting both aliphatic and aromatic characteristics to lignin^[13–14].

2 Pyrolysis pathways of lignin

2.1 Direct pyrolysis of lignin

Lignin pyrolysis can be divided into three sequential stages, including the dehydration stage, the volatile intermediate formation stage, and the further degradation of cross-linked compound stage, ultimately yielding the gaseous, liquid, and solid products. At relatively low pyrolysis temperatures (150–400 °C), the weak linkages within the lignin macromolecule begin to break, and the most abundant ether bonds are cleaved first.

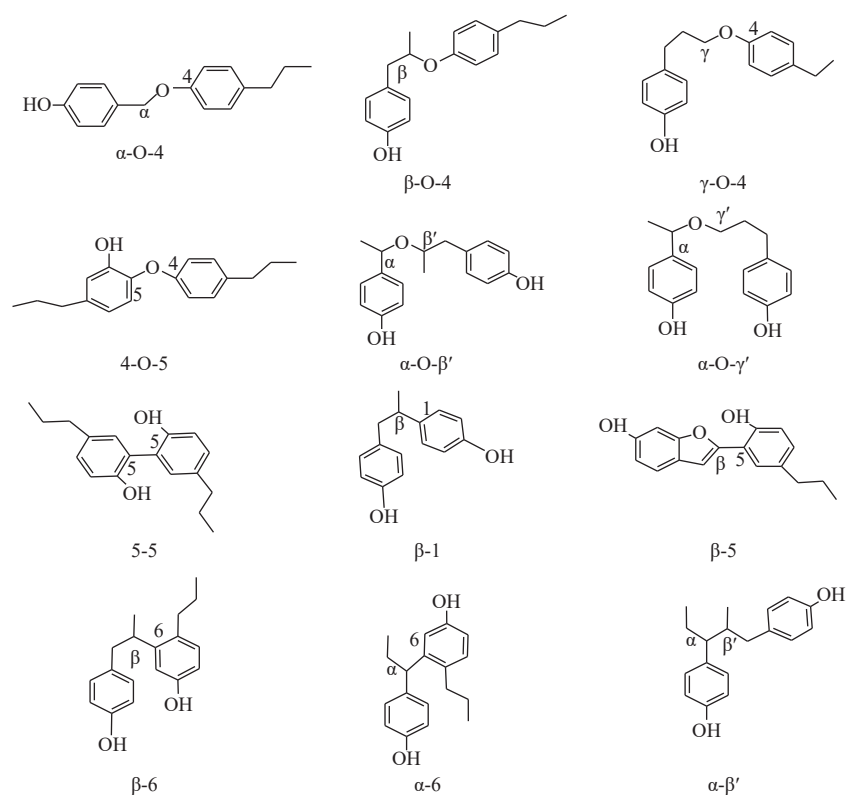


Fig. 2 Various connecting bonds in lignin structure^[10]
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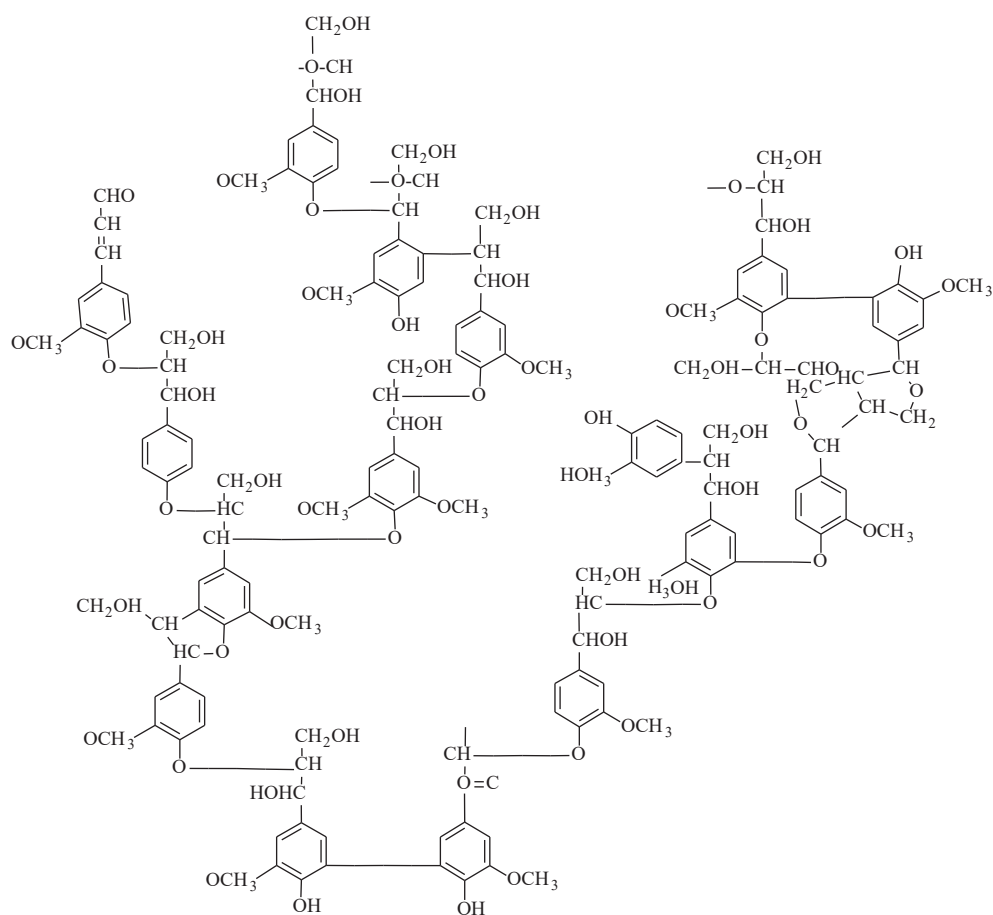


Fig. 3 Schematic diagram of lignin structure^[15]
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Meanwhile, a small proportion of carbon–carbon bonds, such as the β -1 and biphenyl linkages, also undergo cleavage, leading to the formation of low-molecular-weight volatile compounds including gases and oils^[16]. Below the temperature of 350 °C, the benzyl and methoxyl groups remain relatively stable, and the major compounds formed during the pyrolysis correspond to the basic structural units of lignin, such as 4-methylguaiacol, eugenol and *p*-coumaryl alcohol. When the temperature increases to 350 °C or higher, the hydrogen radicals released from the lignin molecules can stabilize the intermediates or depolymerized fragments, thereby enhancing the yield of lignin-derived monomeric products^[17]. At pyrolysis temperatures above 450 °C, the methoxyl groups attached to the aromatic rings undergo cleavage, and the hydroxyl or methylated groups can react with the aromatic moieties, resulting in the rearrangement and stacking of the aromatic ring structures to form the highly ordered solid carbon structures^[18]. The pyrolysis products are mainly composed of compounds such as vanillin, guaiacol, and methoxyquinone, which gradually decompose and undergo carbonization under high-temperature conditions. When the pyrolysis temperature further increases to 500 °C, the aromatic ring structures begin to decompose and are converted into the non-condensable gaseous products. As the temperature rises, the secondary cracking, carbon deposition, and condensation reactions proceed more extensively, leading to the enhanced release of volatile species and a high content of the residual solid carbon (biochar)^[19].

Overall, the liquid products obtained from the fast pyrolysis of lignin possess a high oxygen content, which makes them unsuitable for direct utilization. Therefore, catalysis is required for the upgrading of the oxygenated products to enhance the yield of aromatic compounds. The properly designed catalysts can promote the degradation of lignin matrix, stabilize the reactive intermediates, and regulate the reaction pathways, thereby significantly improving the selectivity of the target aromatic products^[20–22].

2.2 Catalytic pyrolysis of lignin

The core of the catalytic pyrolysis of lignin lies in the directed regulation of its thermal decomposition pathways through the catalytic action, enabling the efficient conversion of the complex biomass polymer into high-value chemicals and fuels. The commonly used catalysts include metal oxides, zeolites, metal-modified zeolites, and the composite molecular sieve catalysts.

2.2.1 Metal (oxide) catalysts

The composite metal oxides exhibit excellent catalytic performance due to the tunable physical and electronic structures arising from the interactions among the components. In addition, they are cost-effective and have

been widely applied in the catalytic pyrolysis of lignin for the production of the aromatic hydrocarbons^[23]. Yang et al.^[24] systematically investigated the catalytic performance of six catalysts, including the HZSM-5, the c-Nb, the c-Al, the c-Nb₃Al₁, the c-Nb₃Al₂ and the c-Nb₃Al₃, in the catalytic fast pyrolysis (CFP) of lignin for the generation of the monoaromatic hydrocarbons (MAHs). The experimental results showed that the c-Nb₃Al₂ catalyst possessed a large surface area of 90 m²/g, a uniform mesoporous structure of 16.15 nm, and a high acidity of 2.00 mmol NH₃/g, exhibiting excellent activity in the catalytic pyrolysis of lignin, with the liquid products containing no detectable oxygenated compounds. Compared with the HZSM-5 catalyst, the c-Nb₃Al₂ achieved a selectivity of 77.0% for the MAHs, demonstrating both high MAHs selectivity and efficient deoxygenation performance. Zhang et al.^[25] investigated the effects of MoO₃, HZSM-5, and α -Al₂O₃ catalysts on the product yield from lignin catalytic pyrolysis using a one-pot technique, and it was found that under the reaction temperature of 340 °C and the H₂ atmosphere, MoO₃ exhibited significantly superior catalytic performance compared with others, achieving a yield of 9.2% for aromatic hydrocarbons and 7.9% for phenolic compounds. Ryu et al.^[26] investigated the co-pyrolysis of lignin and low-density polyethylene (LDPE) for the production of aromatic hydrocarbons under MgO/C catalysis, and found that the high activity of the MgO/C in the formation of aromatic hydrocarbons could be attributed to the synergistic effect between the basic sites in the MgO and the acidic sites in the carbon. When the lignin-to-LDPE mass ratio was 3:1, the yield of the benzene-type compounds (BTEx) reached a maximum of 56.40%, whereas when the ratio was 1:1, the total yield of the aromatic hydrocarbons, including the alkyl-substituted MAHs and the polycyclic aromatic hydrocarbons (PAHs), reached a maximum of 62.52%. Mao et al.^[27] investigated the effects of Co-Mn-O (CMO) metal oxide catalysts on the production of monomeric aromatics from lignin pyrolysis, and found that in a continuous-flow reaction system, the CMO exhibited excellent redox activity due to the presence of the oxygen vacancies (Ov). When corn cob lignin was used as the substrate, the use of the CMO increased the production efficiency of the target aromatic aldehydes by more than 40% as compared with the uncatalyzed system, with the total yield of the aromatic monomeric products reaching 14.23%. Klockhorst et al.^[28] investigated the effect of formic acid as hydrogen donor and Ru/C as catalyst on the composition and the yield of bio-oil from lignin pyrolysis, and it was found that the lignin-derived oil primarily consisted of the small molecules, including aromatic hydrocarbons (10.5%), alkyl phenols (6.0%), catechol (8.7%), guaiacol (1.3%) and alkanes (5.2%), with the others being soluble high-molecular-weight compounds. This process successfully achieved the efficient conversion of lignin, with the yield of the lignin-derived oil reaching as high as 71% based on the mass of lignin feedstock.

In summary, metal (oxide) catalysts can effectively tune the deoxygenation pathways and suppress the condensation

side reactions by promoting the selective cleavage of the C–O bonds and the hydrolysis reactions. Through the provision of active sites and well-defined pore structures, these catalysts significantly reshape the pyrolysis routes, inhibit the onn-selective cracking and the condensation reactions, and direct the conversion of lignin macromolecules into the high-value small molecules such as aromatic hydrocarbons and phenolic compounds. The subsequent mechanisms, including deoxygenation, shape-selective catalysis, and coke suppression, further enhance the yield of aromatic hydrocarbons and improve the product selectivity. However, the limited coke resistance and deactivation stability of these catalysts remain significant drawbacks. In practical applications, they are therefore commonly supported on high-surface-area carriers or combined with zeolitic molecular sieves to improve durability and catalytic performance.

2.2.2 Zeolite-based catalysts

Zeolite-based catalysts have gained considerable importance in catalysis due to their high surface area, shape-selective properties, intrinsic acidity, and easily tunable active sites. Custodis et al.^[29] systematically investigated the effects of mesoporous molecular sieves (MCM-41, SBA-15 and MSU-J) and microporous molecular sieves (HZSM-5 and H-USY) on the yield and the composition of the bio-oil produced from lignin catalytic pyrolysis. The results indicated that when HZSM-5 was employed as the catalyst, the bio-oil yield reached a maximum of 56%, with the relative content of the light aromatic hydrocarbons in the bio-oil reaching 79%. Kim et al.^[30] investigated the effects of milled wood lignin (MWL), organosolv ethanol lignin (OEL), and alkali lignin (AL) on the yield and composition of the bio-oil from the pyrolysis in the presence of HZSM-5 catalyst. The study revealed that under the reaction conditions of a HZSM-5 catalyst with a Si/Al ratio of 30, a catalyst-to-lignin mass ratio of 2:1, and a pyrolysis temperature of 600 °C, the yield of the light aromatics from MWL pyrolysis reached 35.7 mg/g. Paysepar et al.^[31] investigated the influence of hydrolyzed lignin (HL) pyrolysis over low Si/Al ratio faujasite (zeolite X), high Si/Al ratio faujasite (zeolite Y) and ZSM-5 and found that ZSM-5 exhibited the highest efficiency in converting HL into monomeric aromatics and phenolic compounds. At a reaction temperature of 450 °C, the catalytic pyrolysis of HL yielded a maximum bio-oil yield of 57.4%, with the content of monomeric aromatic/phenolic compounds reaching 110 mg/g. Zhao et al.^[32] investigated the product distribution from the pyrolysis products of alkali lignin/alkali lignin char in the presence of ZSM-5. It was found that under an alkali lignin:alkali lignin char:ZSM-5 ratio of 1:1:1, a reaction temperature of 500 °C, and a reaction time of 10 min, the yield of aromatics increased from 17 mg/g (without char) to 33 mg/g (with 1 g char addition). Shen et al.^[33] investigated the effect of HZSM-5 on the catalytic pyrolysis of lignins derived from fir and rice straw at different reaction temperatures. Owing

to its relatively large BET surface area (383.96 m²/g) and appropriate acidity, HZSM-5 exhibited the highest catalytic activity at 650 °C with a Si/Al ratio of 25, where the absolute peak area of toluene reached 1.4×10^9 a.u./mg for both lignin types.

ZSM-5 zeolite catalysts, owing to their high surface area and strong acidic centers, facilitate the deep cleavage of the C–O and C–C bonds in lignin macromolecules. In addition, the shape-selective microporous channels promote cracking, decarbonylation, dehydration, and aromatization reactions, thereby enhancing the formation of the aromatic compounds. However, the narrow pore size of ZSM-5 limits mass transfer, and its strong acidity often leads to coke deposition, resulting in rapid catalyst deactivation.

2.2.2.1 Effect of zeolite acidity

Among the acidic sites of zeolites, Lewis acids primarily promote dehydration and oligomerization reactions, whereas Brønsted acids facilitate cracking, hydrogen transfer, cyclization, and aromatization processes. The acidity strength of HZSM-5 is a critical factor influencing the yield of aromatics during lignin pyrolysis, and its acidity is largely determined by the Si/Al ratio of the zeolite. HZSM-5 with a low Si/Al ratio (high acidity) enhances the formation of aromatics by facilitating demethylation, demethoxylation, dehydroxylation, and methyl transfer reactions on its acidic sites^[33]. Kim et al.^[30] investigated the effect of HZSM-5 zeolites with different Si/Al ratios at various reaction temperatures on the aromatic yield from the lignin catalytic pyrolysis. The results revealed that both reaction temperature and Si/Al ratio significantly affected aromatic yields. Specifically, as the Si/Al ratio decreased from 280 to 30, aromatic product yield increased from 0.76% to 2.62%. In addition, within a pyrolysis temperature range of 500–700 °C, aromatic yield continuously increased with temperature. Han et al.^[34] investigated the effect of β -zeolites (BZ) with different Si/Al ratios on the catalytic pyrolysis of Kraft lignin (KL). The results showed that BZ with different Si/Al ratios could effectively catalyze KL pyrolysis. Among them, BZ with a Si/Al ratio of 45 exhibited stronger acidity, a pronounced layered porous structure, and well-ordered mesopores, resulting in the best catalytic upgrading performance. Under these conditions, the total product yield reached 58.4%, the oxygen content decreased to 14.19%, and the degree of deoxygenation in the heavy oil was 37.1%. Kurnia et al.^[35] investigated the catalytic pyrolysis behavior of lignin over five types of high-alumina zeolites, namely H-hematite, H-zeolite, H-ZSM-5, H-Beta and H-USY molecular sieves. The results revealed that pore structure, pore size, and acidity significantly affected product distribution, coke formation, and deoxygenation performance. Under the strong acidity of HZSM-5, the maximum aromatic content in the light oil reached 88.7 mg/g, whereas the H-Beta with a weaker acidity exhibited the highest selectivity toward the monoaromatic hydrocarbons (MAHs).

The strong acidity of HZSM-5 facilitates demethylation and demethoxylation reactions, thereby enhancing lignin conversion efficiency and facilitating the formation of benzene and naphthalene. Notably, benzene was consistently produced in higher yields than naphthalene, which can be attributed to the distinct influence of HZSM-5 acidity on dealkylation and oligomerization reactions^[36].

2.2.2.2 Effect of zeolite pore structure

Different pore structures of zeolites exhibit distinct shape-selective effects on both reactants and products, thereby influencing the selectivity and yield of individual components in lignin-derived bio-oil^[37]. Jae et al.^[38] investigated the influence of zeolites with different pore sizes and internal pore volumes on lignin pyrolysis products, as illustrated in Figure 4. It was found that small-pore zeolites, such as ZK-5 and SAPO-34 (a microporous molecular sieve composed of Si, Al and P), produced a higher proportion of oxygenated compounds without aromatic hydrocarbon formation. In contrast, large-pore zeolites (SSZ-55, β and Y) generated more coke and only a small amount of aromatics. Zeolites with medium pore sizes of 5.2–5.9 μm (e.g., ZSM-5) yielded 35.5% aromatic hydrocarbons and the lowest coke (30.4%). Custodis et al.^[29] investigated the effect of Al-MCM-41 in nanoparticle form (Al-MCM-41(50)-nano) on product yields from lignin catalytic pyrolysis. The results showed that nano-Al-MCM-41, owing to its high-textured porosity and engineered external surface, significantly improved reactant accessibility to acid sites while reducing the residence time of reactants and intermediates, thereby achieving the highest liquid yield (54%) among the mesoporous catalysts. Shafaghat et al.^[39] investigated the effect of zeolites with different pore sizes on the yield of aromatics from lignin catalytic pyrolysis. It was found that small-pore zeolites,

such as ZSM-5, were less suitable for lignin conversion because their narrow channels limit the diffusion of reactants and intermediates, resulting in high yields of oligomeric by-products. In contrast, zeolites with larger pore sizes facilitated the diffusion of reactants and the intermediates, exhibiting better catalytic performance and achieving higher yields of aromatic hydrocarbons. Lazaridis et al.^[40] investigated the pyrolysis performance of Kraft lignin under the catalysis of ZSM-5 with different pore structures. The results indicated that conventional microporous ZSM-5 zeolites (Si/Al = 11.5, 25 and 40) exhibited higher selectivity toward monocyclic aromatic hydrocarbons, whereas nanoscale (≤ 20 nm) and mesoporous (9 nm, prepared *via* mild basic treatment) ZSM-5 zeolites demonstrated better selectivity for the alkylphenols.

2.2.3 Metal-modified zeolite catalysts

The acidic sites of HZSM-5 zeolites tend to strongly interact with phenols and their derivatives, leading to catalyst deactivation, coke deposition, and reduced deoxygenation performance^[41]. In contrast, the metal-modified catalysts exhibit optimized pore structures, well-distributed acidic sites, and improved catalytic performance. According to the literature, single-metal-modified zeolites, such as Zn, Ga, Mg and Co, exhibit strong deoxygenation, hydrogen transfer, and aromatization performance for lignin pyrolysis. Huang et al.^[42] investigated the variation of light aromatic hydrocarbons from catalytic pyrolysis of poplar, fir, and corn stover lignin over bifunctional HZSM-5 catalysts modified with Zn, Ga and Mg. The study found that under Zn-modified catalysts, all three lignins produced high yields of light aromatic hydrocarbons, whereas Mg-modified catalysts suppressed aromatic formation.

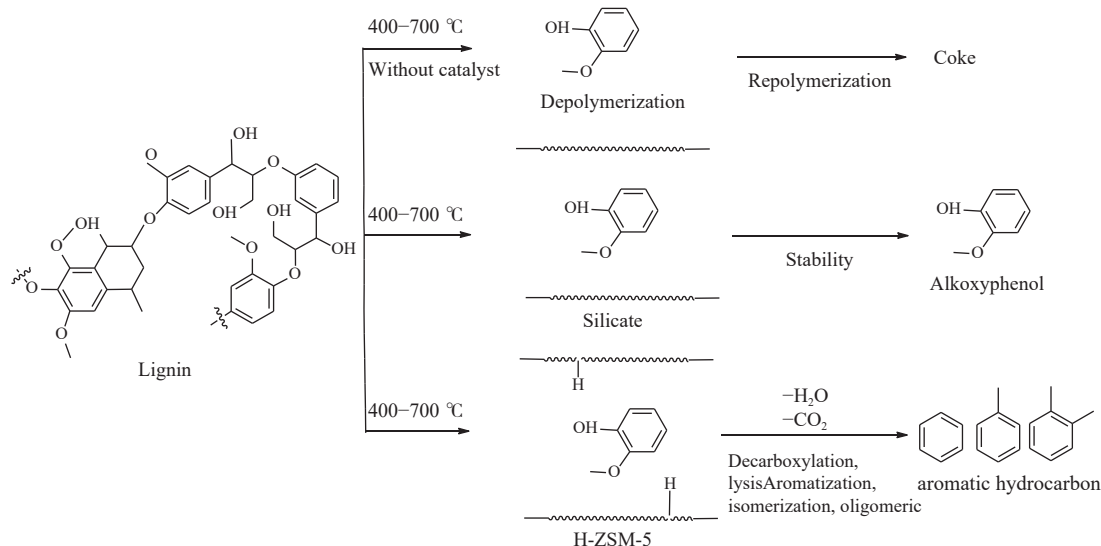


Fig. 4 The synergistic effect of molecular sieve porosity and acidity on the distribution of products from lignin catalytic pyrolysis^[36] (with permission from ACS Publications)

Notably, the BTX (benzene, toluene, and xylene) selectivity of milled fir lignin reached a maximum of 65.02%. Jin et al.^[43] investigated the effect of Co-modified HZSM-5 on the yield of monocyclic aromatic hydrocarbons (MAHs) from lignin catalytic pyrolysis. The study revealed that 1% Co-modified HZSM-5 (1% Co-HZ) significantly increased the mesoporous surface area and the average pore size while retaining strong acid sites, thereby enhancing mass transfer efficiency and raising MAHs yield from 17.3% to 29.3%. Yang et al.^[44] investigated the effect of ZSM-5 catalysts modified with different Fe loadings on the distribution of lignin pyrolysis products. The study revealed that with increasing Fe loading, the specific surface area, pore volume, and number of acidic sites of the catalyst changed significantly, while the overall structure of the catalyst remained stable. The formation of light olefins correlated closely with moderate acidity, and the enhanced total acidity after Fe modification resulted in an aromatic hydrocarbon yield of 12.8%. Huang et al.^[45] investigated the effects of various metal-modified HZSM-5 catalysts on the yield and composition of aromatic hydrocarbons from lignin pyrolysis. The study found that BTX selectivity under 4Zn/HZSM-5 and Ga-Zn/HZSM-5 catalysts reached 65.19% and 64.89%, respectively, both significantly higher than that of 4Ga/HZSM-5 (52.77%). This result indicates that the construction of Ga-Zn bimetallic active sites can effectively enhance the selectivity of BTX aromatic hydrocarbons during lignin conversion. Li et al.^[46] investigated the effects of Ni-, Cu-, and Ru-co-modified HZSM-5 catalysts on the yield of aromatic hydrocarbons from lignin catalytic pyrolysis. The study revealed that HZSM-5 catalyst co-loaded with the three metals possessed a micro-mesoporous structure, which was conducive to enhancing the aromatic hydrocarbon yield, with the relative content of aromatics reaching up to 28.95%.

Overall, metal-modified HZSM-5 catalysts can regulate the type, loading and dispersion of metals to optimize catalyst acidity and pore structure, thereby influencing the product distribution of bio-oil, enhancing aromatic hydrocarbon selectivity, suppressing coking, and substantially increasing the yield and selectivity of target BTX aromatics. However, large metal particles or clusters may block zeolite micropores, especially when noble metals (Pt and Pd) or low-melting-point metals (e.g., Zn) are loaded. During high-temperature reactions or regeneration (coke burning), these metal particles may migrate and sinter, leading to a loss of active sites.

2.2.4 Composite zeolite catalysts

The catalytic pyrolysis of lignin over composite zeolite catalysts has been systematically investigated to enhance the production of aromatic hydrocarbons. These composite zeolites are synthesized through the co-crystallization of two or more zeolites, thereby combining the pore structures and acidic properties of individual components. Such

catalysts exhibited multiple catalytic sites, strong synergistic effects, high selectivity, excellent thermal stability, and good reusability. Wang et al.^[47] investigated the catalytic performance of Ni- and Mo-bimetallic co-modified micro-mesoporous composite zeolite catalyst (denoted as HM, formed by combining HZSM-5 with MCM-41) for the production of monocyclic aromatic hydrocarbons (MAHs) from lignin catalytic pyrolysis. The study revealed that the Mo₁Ni₂-HM catalyst, prepared *via* the co-impregnation method, achieved a MAHs yield of 39.2%, representing an increase of 21.52% compared with that of the unmodified HM catalyst (17.69%). Lee et al.^[48] investigated the effect of a dual-catalyst system composed of natural zeolite and HZSM-5 on the product distribution of lignin pyrolysis. The results showed a pronounced synergistic effect, where the natural zeolite suppressed the formation of high-molecular-weight oxygenates through *in situ* pre-catalytic reactions, while providing an optimized environment for subsequent aromatization over HZSM-5. As a result, the selectivity of aromatic hydrocarbons increased from 32.1% to 47.8%, and coke deposition on the HZSM-5 surface was significantly reduced. Zheng et al.^[49] investigated the catalytic behavior of a Co-Ni@H/M core-shell catalyst and its synergistic effects during lignin catalytic pyrolysis. The study revealed that the use of Co-Ni@H/M catalyst increased the selectivity of enzymatic lignin-derived products by 11.68%, reaching 33.02%. The bimetallic Co-Ni synergy mainly promoted the dealkylation of phenolic -OH groups and oxygenated intermediates, while enhancing oligomerization, cyclization, and aromatization of short-chain alkyl compounds toward MAHs and suppressing polymerization of phenolic and oxygen-containing species. Liu et al.^[50] further examined the CuZS (Cu-ZSM-5/SBA-15) catalyst at various catalyst-to-feed ratios and reaction temperatures, focusing on the distribution and composition of the pyrolysis products. The results showed that at the reaction temperature of 800 °C, CuZS promoted decarboxylation and decarbonylation reactions of alkaline lignin (AL), thereby facilitating the cleavage of aliphatic side chains and exhibiting the optimal catalytic activity. Furthermore, when the catalyst-to-feed ratio was 10:1, the selectivity of total hydrocarbons (THs) increased to 92.25%, with the MAHs accounting for 66.07% and the BTEX fraction reaching 61.72% of the total MAHs.

In summary, composite zeolite catalysts exhibit excellent potential for enhancing the catalytic pyrolysis of lignin to produce aromatic hydrocarbons. The introduction of composite zeolite structures effectively optimizes the catalytic reaction environment and redistributes acidic sites, thereby enhancing reaction selectivity, reducing by-product formation, and improving catalyst stability. Experimental and reported results have demonstrated that composite zeolites provide superior resistance to coking compared with the conventional microporous zeolites and maintain high catalytic activity during pyrolysis. However, issues related to deactivation and regeneration of these catalysts

remain unresolved, and the high regeneration cost still limits the large-scale industrial application of these composite zeolite catalysts.

The commonly used feedstocks and catalysts for lignin catalytic pyrolysis to produce aromatic hydrocarbons are summarized in Table 1. As shown in the table, metal and metal oxide catalysts exhibit high catalytic activity and achieve high aromatic yields. However, their limited selectivity, poor stability, and high cost restrict large-scale application. Zeolite catalysts, owing to their unique shape-selective properties and excellent structural stability, effectively promote aromatization reactions. However, their microporous structures hinder the mass transfer of bulky lignin-derived molecules, and continuous operation often results in coke deposition, leading to pore blockage and catalyst deactivation. Metal-modified zeolites, through the synergistic interaction between metal active sites and the acidity and pore structure of zeolites, significantly enhance the yield and selectivity of target aromatic hydrocarbons (BTX). Nevertheless, catalyst deactivation caused by pore blockage and surface coking remain a major challenge. Future research should focus on regulating the interaction between metal species and zeolite framework, optimizing mesoporous structure, and improving metal loading techniques to develop composite zeolite catalysts with high activity, selectivity, stability, and cost efficiency. By constructing dual-functional synergistic sites and hierarchical pore structures, such catalysts can

simultaneously address the three key challenges of deoxygenation efficiency, hydrogen transfer management, and mass transfer limitation, thereby achieving high yield and selectivity of aromatic hydrocarbons.

3 Reaction conditions

3.1 Effect of catalyst-feed contact mode

In pyrolysis reactors, systems in which the catalyst is directly mixed with the feedstock are referred to as *in situ* catalytic pyrolysis, whereas the configurations where the pyrolysis vapors generated from the thermal decomposition of the feedstock are subsequently transported to a separate downstream catalyst bed are defined as *ex situ* catalytic pyrolysis^[51], as illustrated in Figure 5^[52].

Fan et al.^[53] systematically investigated the effects of HZSM-5 zeolite on the product composition of lignin pyrolysis vapors under both *in situ* and *ex situ* catalytic conditions. The experimental results revealed that during *ex situ* catalytic pyrolysis, lignin feedstock tended to aggregate and experience uneven heating, thereby promoting char formation. In contrast, for the *in situ* catalytic pyrolysis, lignin and catalyst were homogeneously premixed prior to the reaction, thereby mitigating lignin aggregation and reducing carbonization, which consequently enhanced the yield of bio-oil and syngas.

Table 1 Common raw materials and catalysts for lignin pyrolysis to produce aromatic hydrocarbons

Raw material	Catalyst	Preparation method	Aromatic	Quote
Corn straw	Mo ₁ Ni ₂ -HM	co impregnation method	39.24%	[47]
Corn straw	c-Nb ₃ Al ₂	acid precipitation method	77%	[24]
Lignin	1% Co-HZ	wet impregnation method	29.3%	[43]
Coniferous wood (Chinese fir), Broad-leaved wood (poplar), and herbaceous plants (corn stover)	Zn/HZSM-5	immersion method	3.122×10 ⁹ a.u./mg (CF-MWL), 2.916×10 ⁹ a.u./mg (P-MWL) and 2.865×10 ⁹ a.u./mg (CS-MWL)	[42]
Cork alkali lignin	HZSM-5	physical mixing method	44.6%	[29]
Natural lignin (ML)	HZSM-5		35.7 mg/g	[30]
Hydrolyzed lignin (HL)	ZSM-5		0.11 g/g HL	[31]
Mixed hardwood	Ru/C		10.5%	[28]
Alkali lignin	6%Fe/ZSM-5	immersion method	12.8%	[42]
Alkali lignin	ZSM-5		33 mg/g	[32]
Sulfate lignin and LLDPE	MgO/C	equal humidity Immersion method	56.4%	[26]
EHL (enzymatic lignin hydrolysis)	Co-Ni@H/M	solvent immersion method	33.02%	[49]
Masson pine wood pulp lignin (MWL)	Ga-Zn/HZSM-5	wet impregnation method	64.89%	[43]
Corn cob lignin	CMO	citric acid sol gel method	14.23%	[27]
Alkali lignin (AL)	CuZS	wet impregnation method	66.07%	[50]
Bio oil (LDB)	Ni-Cu-Ru/HZSM-5	wet impregnation method	28.95%	[46]

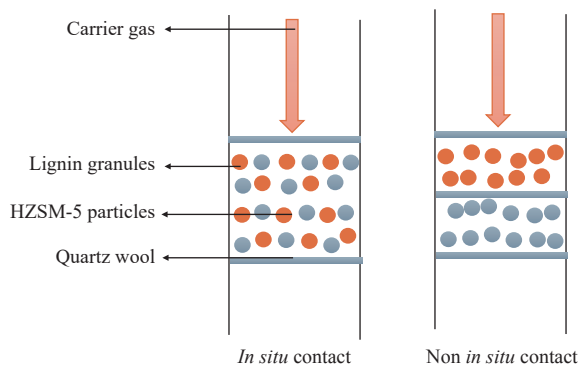


Fig. 5 Contact method between catalyst and raw materials^[52]

As a result, compared with the *in situ* catalytic pyrolysis yielding 28.95% bio-oil and 39.45% char, the *ex situ* configuration produced 23.3% bio-oil and 43.63% char. Li et al.^[54] examined the distribution of lignin pyrolysis products over various metal-modified ZSM-5 catalysts under different catalytic modes. The results indicated that, compared with the *in situ* catalysis, the *ex situ* catalysis exhibited a higher selectivity for guaiacol. Under H@ZSM-5 catalysis, the guaiacol selectivity increased from 27.52% to 42.61%, while under the Fe@ZSM-5 catalyst, it increased from 27.18% to 42.46%. In contrast, the Mg@ZSM-5 catalyst, owing to its intrinsic basicity, inhibited the deoxygenation reactions and preserved the methoxy groups under both catalytic modes, resulting in guaiacol selectivities of 49.77% (*in situ*) and 55.48% (*ex situ*). Lee et al.^[48] investigated the pyrolysis characteristics of lignin using the natural zeolite (NZ) in the *in situ* catalysis and the HZSM-5 in the *ex situ* catalysis. The study found that the *in situ* catalysis using the NZ had no significant influence on the formation of aromatic hydrocarbons, but increased the yields of alkylphenols and catechol while reducing the yields of guaiacol and syringol. Compared with the single-stage *ex situ* HZSM-5 catalysis, the two-stage NZ-HZSM-5 catalytic pyrolysis produced less char and resulted in a higher-quality bio-oil with increased contents of BTEX and alkylphenols.

The *in situ* catalytic contact facilitates the decomposition of lignin macromolecules and enhances the formation of volatile intermediates, resulting in a higher yield of bio-oil compared with *ex situ* catalytic contact, whereas the yields of the solid and gaseous products exhibit the opposite trend^[53]. In contrast, *ex situ* catalytic contact provides better control of the reaction parameters during both the lignin pyrolysis and catalytic upgrading stages. Moreover, after reaction, catalyst separation from biochar is simplified, and the recovered biochar can be directly utilized as a renewable carbon resource.

3.2 Effect of reaction temperature

The catalytic pyrolysis of lignin at elevated temperatures has been systematically investigated to elucidate the effect of the reaction temperature on lignin depolymerization, deoxygenation, and aromatic hydrocarbon formation.

Increasing the reaction temperature accelerates the decomposition of lignin macromolecules and promotes the deoxygenation process, thereby enhancing the production of aromatic hydrocarbons. However, excessively high temperatures induce secondary cracking of aromatic compounds into small gaseous molecules^[55] and facilitates the oligomerization of monocyclic aromatic hydrocarbons (MAHs) into polycyclic aromatic hydrocarbons (PAHs). Jiang et al.^[56] investigated the effect of reaction temperature on the catalytic conversion of guaiacol to aromatic hydrocarbons using a fixed-bed reactor. The study revealed that increasing the temperature from 500 to 800 °C significantly enhanced the yield of aromatic hydrocarbons, with the mass fractions of benzene and toluene reaching 14.07% and 4.04%, respectively. The combined mass fractions of aromatic hydrocarbons (benzene, toluene, styrene, naphthalene, and 2-methylnaphthalene) and char increased from 1.83% and 1.17% to 24.27% and 12.81%, respectively. At the reaction temperature of 600 °C, a high Si/Al ratio (200) HZSM-5 catalyst further increased the mass fractions of benzene and toluene by 2.94% and 1.94%, respectively. Nishu et al.^[57] examined the effect of different pyrolysis temperatures (450, 500, 550 and 600 °C) on the selectivity of aromatics in the pyrolysis products of rice straw lignin using ZSM-5, NaOH-modified ZSM-5 (0.4 mol/L), and Ni-modified ZSM-5 (8%) analyzed *via* Py-GC-MS. The results showed that under the catalysis of ZSM-5, small pyrolysis molecules underwent polymerization, yielding the highest aromatic selectivity (49%) at 550 °C. At higher temperatures, selectivity decreased due to dominant oligomerization and exothermic hydrogen-transfer reactions. In contrast, under the catalysis of NaOH-modified ZSM-5 (0.4 mol/L), the aromatic selectivity increased with temperature, reaching a maximum of 62% at 550 °C. When Ni/ZSM-5 (8%) was employed, the selectivity of the aromatics was highly sensitive to temperature, gradually increasing from 450 to 550 °C and achieving the highest (73%) at 550 °C. Overall, due to the high acidity of ZSM-5 promoting aromatization and oligomerization reactions, 550 °C was identified as the optimal reaction temperature for rice straw lignin conversion into aromatic hydrocarbons. Zhang et al.^[58] investigated the effect of the reaction temperature on the product distribution of lignin catalytic pyrolysis under a nitrogen flow rate of 50 mL/min and a methanol-to-guaiacol molar ratio of 25. The study revealed that at 300 °C, the catalytic conversion of guaiacol predominantly produced aromatic oxygenates, with a relatively low selectivity toward aromatic hydrocarbons. Increasing the temperature to 400 °C markedly improved the conversion efficiency of guaiacol and methanol, and the combined yield of key intermediates *p*-methoxybenzene (PMB) and hydroxymethylbenzene (HMB) reached 48.31%, representing an optimal balance among deoxygenation, aromatization, and hydrogen-transfer reactions. Under these conditions, the aromatic hydrocarbon yield reached 57.93%. Further increasing the temperature to 450 °C caused excessive deoxygenation and C–C bond cleavage, which

destabilized the intermediates and sharply reduced the aromatic hydrocarbon selectivity to 24.12%. Based on the comprehensive analysis of product distribution and catalyst stability, 400 °C was determined to be the optimal reaction temperature for guaiacol conversion into aromatic hydrocarbons. Shen et al.^[33] investigated the influence of reaction temperature on the aromatic formation pathways and selectivity during the pyrolysis of cedarwood and rice straw lignin. For cedarwood lignin, the depolymerization-derived aromatic hydrocarbons (such as benzene and naphthalene derivatives) reached their maximum peak areas at 650 °C. In contrast, the pyrolysis of rice straw lignin exhibited a staged temperature-dependent behavior, in which the yields of toluene, ethylbenzene, *p*-xylene and methyl naphthalene peaked at 650 °C, whereas the formation of benzene, *o*-xylene and naphthalene required higher temperatures (around 800 °C) to activate deep deoxygenation and cyclization pathways. Therefore, 650 °C was identified as the optimal reaction temperature to achieve the highest overall aromatic hydrocarbon yield.

A moderate increase in reaction temperature can enhance decarboxylation and dehydration activities, thereby promoting the formation of aromatic hydrocarbons. However, excessively high temperatures tend to trigger over-cracking reactions and facilitate the conversion of monocyclic aromatic hydrocarbons (MAHs) into polycyclic aromatic hydrocarbons (PAHs), which in turn accelerates the deposition of carbonaceous species on the catalyst surface and leads to catalyst coking. This phenomenon ultimately causes catalyst deactivation and a significant reduction in the catalytic activity for aromatic hydrocarbon production.

3.3 Effect of the feedstock-to-catalyst ratio

The amount of catalyst is a crucial parameter in lignin pyrolysis. An appropriate catalyst-to-lignin ratio can facilitate the cleavage of active bonds in lignin and accelerate macromolecule depolymerization by reducing the activation energy of these bonds, thereby directing the conversion toward aromatic compound formation. Yang et al.^[59] investigated the product distribution of lignin pyrolysis over a Ni_3Al_3 mixed metal oxide (MMO) catalyst. The results showed that at a lignin-to-catalyst mass ratio of 1:5, the major products were oxygenated compounds, including 31.5% simple phenols, 0.4% phenol derivatives, and 7.4% other oxygenates. As the catalyst amount increased, the content of aliphatic hydrocarbons initially rose from 14.1% (1:5) to 22.6% (1:10), then decreased to 21.3% (1:15) and 18.0% (1:20). A higher Ni_3Al_3 -MMO loading accelerated the deoxygenation of phenol and their derivatives and extended the contact time with pyrolysis vapors, resulting in a gradually increasing of monocyclic aromatic hydrocarbons (MAHs) to 78.2% at a lignin-to-catalyst ratio of 1:20. Li et al.^[60] investigated the fast pyrolysis of enzymatic hydrolysis lignin (EHL) over Nb_2O_5 catalyst for the production of aromatic hydrocarbons. The study revealed that the yield of MAHs decreased with the

catalyst-to-lignin ratio (C/L ratio). Specifically, at a C/L ratio of 1:5, MAHs yield was 6.5%, which increased to 9.7% at 1:7 and further rose to 10.6% at 1:9, with MAHs accounting for 94% of the total aromatic fraction. Huan et al.^[61] investigated the effect of different lignin-to-catalyst ratios on the hydrothermal liquefaction of alkali lignin over Zr-, Mo- and Ce-impregnated red mud (RM) catalysts. Using the bimetallic 5% Ce-Mo/RM catalyst, a C/L ratio of 1:5 yielded 81.8% bio-oil, which increased to 91.6% at 1:2. Conversely, at 1:10, the alkali lignin was effectively depolymerized into guaiacol derivatives, achieving vanillin selectivity of 65.4%. Zou et al.^[62] investigated the influence of lignin-to-catalyst ratios on the pyrolysis products over a hierarchical ZSM-5/MCM-41 catalyst. The MCM-41 component, owing to its weak acidity and large pore size, facilitated depolymerization of macromolecular intermediates. Without the catalyst, the conversion of lignin and waste oil was approximately 50%. As the catalyst ratio increased, conversion rose steadily to 76% at a 1:10 ratio. Beyond this point, conversion slightly decreased and the bio-oil proportion remained nearly constant. Thus, a lignin-to-catalyst ratio of 1:10 was identified as the optimal condition balancing conversion efficiency and product selectivity.

The effect of catalyst loading on lignin pyrolysis has been systematically investigated to evaluate its influence on bio-oil yield and quality. At a low catalyst loadings, lignin undergoes insufficient catalytic cracking, resulting in high contents of macromolecular organics including phenolic compounds, high bio-oil viscosity, and elevated oxygen content. At moderate catalyst loadings, catalytic activity improves, promoting deoxygenation reactions, such as hydrodeoxygenation (HDO), and cracking reactions, thereby generating high amounts of monomeric phenols including guaiacol and syringol, and low-molecular-weight aromatic hydrocarbons, which improves the quality of bio-oil. However, at excessively high catalyst loadings, the liquid products can be further cracked into gaseous molecules, leading to a decline in bio-oil yield. These results collectively demonstrate that an optimal catalyst loading is essential to achieve a balance between deoxygenation efficiency, molecular weight reduction, and the maximization of bio-oil yield and quality.

3.4 Effect of reaction time

The effect of reaction time on lignin catalytic pyrolysis has been systematically investigated to evaluate its influence on the yield and selectivity of aromatic hydrocarbons. At short reaction times, lignin underwent incomplete pyrolysis, and the macromolecular structure was not fully cleaved into small fragments including phenolic monomers and ketones, resulting in insufficient aromatic precursors and low yields of aromatic hydrocarbons. At moderate reaction times, lignin experienced sufficient depolymerization to generate reactive intermediates including phenols and olefins, which were further deoxygenated and aromatized over the catalysts such as

HZSM-5, producing benzene, toluene, and xylene (BTX). Conversely, at excessively long reaction times, aromatic products underwent secondary reactions including polymerization and coking, generating polycyclic aromatic hydrocarbons (PAHs) or coke that covered the catalytic active sites, thereby reducing the yields of aromatic hydrocarbons. Experimental results from Zhao et al.^[32] demonstrated that under conditions of alkaline lignin, lignin char, and ZSM-5 mixed in a 1:1:1 mass ratio at 500 °C, the formation of aromatic hydrocarbons initially increased with reaction time and reached a maximum of 33.8 mg/g at 10 min. Further increased in reaction time promoted the formation of CO and CH₄, indicating a decline in aromatic hydrocarbon production. Similarly, studies by Ma et al.^[63] showed that for reaction times below 2.5 s, heat transfer delays prevented the system from reaching the effective reaction temperature, yielding predominantly sulfur-containing non-aromatic compounds. At reaction times exceeding 2.5 s, the acidic sites of the large-pore FAU zeolite were fully activated, facilitating the activation of aromatic units within the lignin structure, with aromatic hydrocarbon selectivity reaching over 50% at 20 s. Lin et al.^[64] further investigated pyrolysis times of 15, 30 and 60 s at 450 °C for enzymatically hydrolyzed corn stover lignin. The results showed that the phenolic compound content was highest (≈35%) at 15 s, decreased to ≈25% at 30 s as the aromatic hydrocarbon content increased from <10% to ≈20% and at 60 s, deoxygenation and aromatization were further enhanced, making aromatic hydrocarbons the predominant products (≈30%) while phenolic content dropped below 15%. At this stage, the combined content of hydrocarbons and char rose to ≈25%, indicating that prolonged reaction times promote condensation and coking reactions.

Overall, the reaction time exerts a significantly influence on the progress and efficiency of the catalytic process. Short duration lead to incomplete pyrolysis and low aromatic hydrocarbon yield, whereas moderate durations maximize yield and selectivity. Excessively long reaction times, however, promote secondary reactions and coke formation, diminishing catalytic activity. These findings collectively demonstrate that optimizing reaction time is essential to maximize both yield and selectivity of aromatic hydrocarbons during lignin catalytic pyrolysis.

4 Reaction mechanism

4.1 *In situ* pyrolysis mechanism of lignin

The *in situ* pyrolysis mechanism of lignin was systematically investigated by Lu et al.^[65] During pyrolysis, chemical bonds, such as C–C and C–O–C linkages are cleaved, generating phenolic compounds and aromatic hydrocarbons, while functional groups, including carboxyl, carbonyl, and methoxy groups, are released from aromatic rings to form light gaseous products. Finally, lignin-derived char, which is rich in functional groups such as –OH and

–OCH₃, is produced through carbonization. On one hand, with increasing temperature, functional groups in the biochar are further cleaved, producing light gaseous products. On the other hand, the reactive aromatic species in the bio-oil can be converted into biochar through re-polymerization. Zhong et al.^[66] investigated the influence of lignin pyrolysis pathways on bio-oil production, as illustrated in Figure 6. Lignin, as a macromolecular component of biomass, undergoes bond cleavage at elevated temperatures, producing intermediate phenolic units such as guaiacyl, syringyl, and *p*-hydroxyphenyl. These intermediates subsequently undergo rearrangement, demethylation, and demethoxylation reactions to form various phenolic compounds and light products. The aforementioned products can further undergo condensation, polymerization, or cleavage reactions, ultimately yielding char, gaseous products (e.g., CO and CH₄), and liquid products such as phenolics and aromatic hydrocarbons.

4.2 Mechanism of lignin pyrolysis to aromatics over zeolite catalysts

The distinct reaction pathways involved in the catalytic fast pyrolysis of lignin were systematically investigated by Luo et al.^[67], as illustrated in Figure 7. Initially, lignin undergoes thermal decomposition into small oxygenated molecules, such as acids, aldehydes, furans, phenols and dehydrated sugars. These intermediates then diffuse into the zeolite pores, where the deoxygenation occurs to produce hydrocarbon precursors, such as ethylene, propylene, and butenes, which subsequently undergo aromatization to form aromatic hydrocarbons (Pathway 1). Phenol, guaiacol, and other aromatic oxygenates are converted over the catalyst *via* demethoxylation and demethylation reactions into the phenol, cresols and ethylphenols, which are further dehydrated to yield the benzene, toluene, and xylene (Pathway 2).

The mechanism of NaOH-assisted catalytic pyrolysis of lignin over HZSM-5 for the production of aromatic hydrocarbons was systematically investigated by Wu et al.^[68], as illustrated in Figure 8. Initially, lignin undergoes direct thermal pyrolysis to produce char, hydrogen, and small oxygenated molecules such as phenols and aldehydes. Under the catalysis of HZSM-5, these oxygenates are further converted into monoaromatic hydrocarbons (MAHs, e.g., the benzene and toluene) *via* deoxygenation, aromatization, and cracking reactions, accompanied by the release of CH₄ and H₂. The phenolic intermediates may undergo demethylation and demethoxylation to produce additional MAHs, but they can also condense to form polycyclic aromatic hydrocarbons (PAHs) or large oxygenated molecules, leading to coke formation. Upon the addition of NaOH, demethylation, demethoxylation, and condensation reactions are promoted, leading to the formation of more PAHs, H₂ and CH₄, accompanied by the formation of coke.

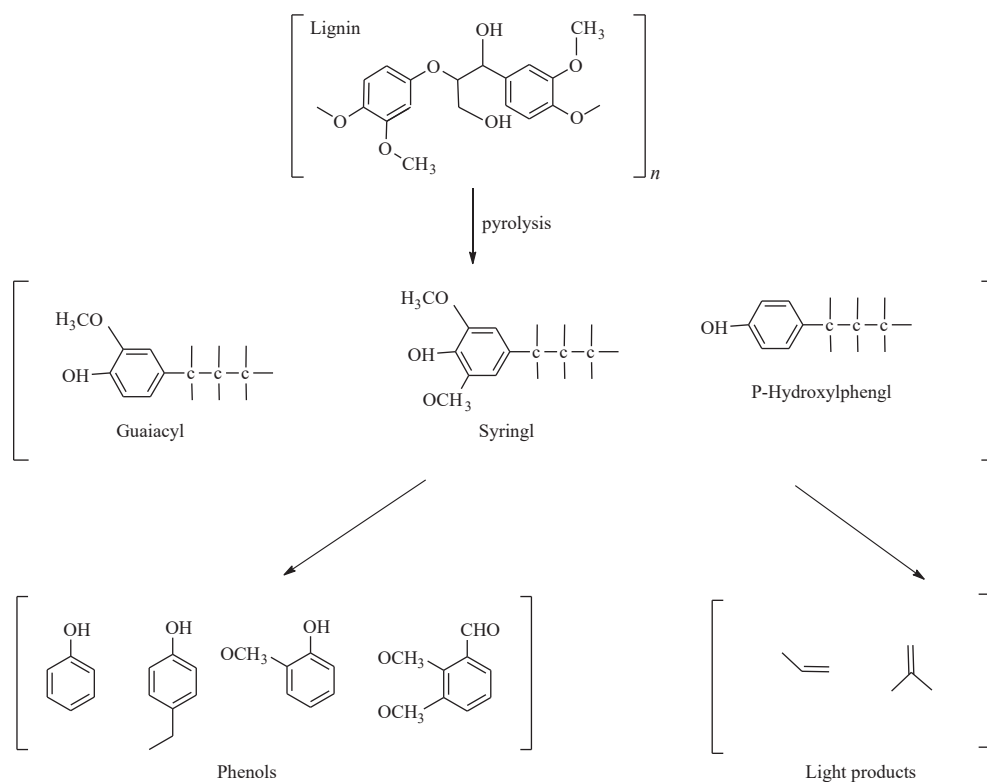


Fig. 6 Transformation pathways of lignin pyrolysis^[66]
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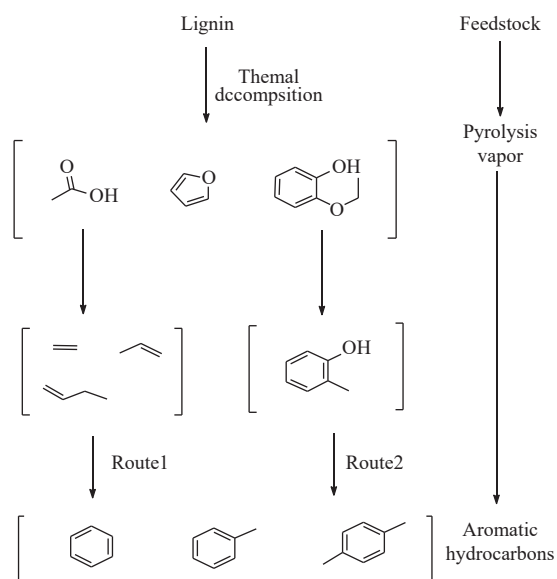


Fig. 7 Different reaction pathways of lignin catalytic fast pyrolysis^[67]
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Additionally, the volatile products of lignin pyrolysis (e.g., H_2 , CH_4 and phenolic compounds) under the influence of NaOH can enhance *in situ* hydrogen transfer, releasing more hydrogen radicals into zeolite channels. This process facilitates the hydrodeoxygenation of phenolics and promotes deoxygenation and polymerization reactions on both the internal and external surfaces of the HZSM-5, ultimately generating aromatic hydrocarbons.

The reaction mechanism of lignin pyrolysis catalyzed by HZSM-5 was systematically investigated by Wan et al.^[52], as illustrated in Figure 9. It was found that upon heating to a specific temperature, lignin undergoes pyrolysis, primarily producing phenolic compounds and other intermediate species. These intermediates subsequently diffuse and interact with HZSM-5 zeolite. Under the catalytic influence of the surface acidic sites of HZSM-5, these intermediates undergo structural transformations, including demethoxylation and dehydroxylation, leading to the formation of aromatic hydrocarbons. Due to the spatial selectivity of the zeolite pore channels, the reaction system preferentially produces monoaromatic hydrocarbons (MAHs) and other accompanying products. However, if some MAHs are not promptly desorbed from the catalyst surface, they may condense on the HZSM-5 surface to form polycyclic aromatic hydrocarbons (PAHs), which can further polymerize into highly condensed coke.

The synergistic catalytic mechanism of alkaline lignin char over ZSM-5 was systematically investigated by Zhao et al.^[32], as illustrated in Figure 10. It was found that, initially, at high temperatures, alkaline lignin undergoes the cleavage of chemical bonds such as β -O-4, β - β and α -O-4, generating numerous monomers, including methoxylated phenols, *via* radical reactions. Subsequently, the C-O bonds in these methoxylated phenols are further cleaved under the catalytic effect of lignin char, yielding large amounts of alkyl phenols and other small molecules (e.g., acetic acid), accompanied by decarboxylation, decarbonylation, and demethylation reactions that produce the gases such as CO_2 , CO and CH_4 .

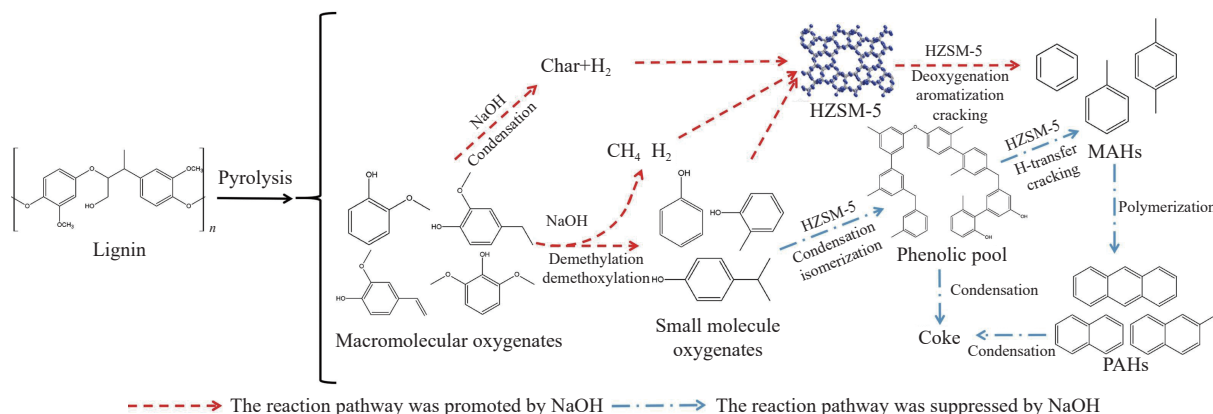


Fig. 8 Effect of NaOH on the reaction pathway of lignin/HZSM-5 catalytic pyrolysis^[68]
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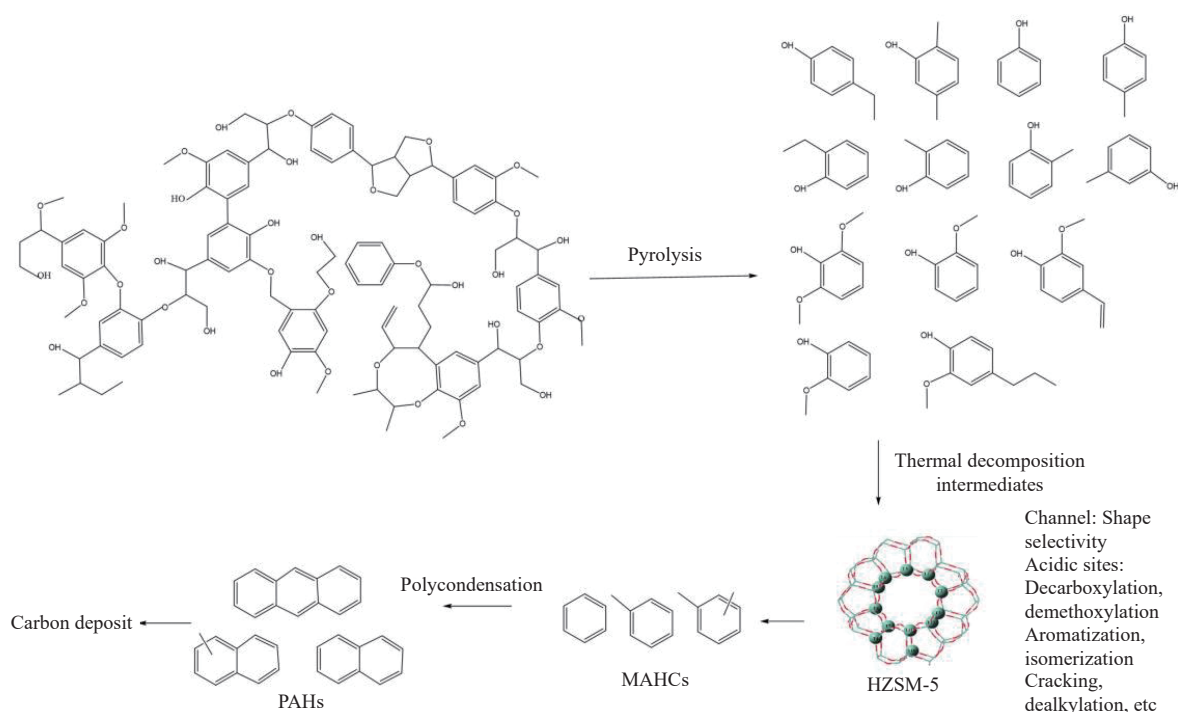


Fig. 9 Mechanism of lignin pyrolysis catalyzed by HZSM-5^[52]

Finally, the small-molecule intermediates, including methoxylated and alkyl phenols, undergo shape-selective catalytic conversion over ZSM-5, involving decarbonylation, dealkylation, isomerization, and aromatization reactions, ultimately forming aromatic hydrocarbons.

4.3 Mechanism of lignin catalytic pyrolysis to aromatics over metal-modified zeolites

The formation mechanism of mono-aromatic hydrocarbons (MAHs) from milled wood lignin (MWL) via a combined torrefaction-deoxygenation pretreatment (TDP) and dual-catalyst catalytic fast pyrolysis (DC-CFP) approach was systematically investigated by Huang et al.^[69], as illustrated in Figure 11. The study revealed that, following the cleavage of β -O-4, β - β and β -5 linkages, heavy phenolic intermediates (e.g., oligomers, guaiacyl

units, and syringyl phenols) were generated, and the reaction pathway proceeded predominantly in two stages. First, the heavy phenolics passed through the mesoporous Nb-modified Al_2O_3 catalyst bed, where the weak acid sites facilitated C-O and C-C bond cleavage, converting the heavy phenolics into light phenols (e.g., phenol and alkylphenols), aliphatic hydrocarbons (e.g., ethylene and propylene), and gaseous products. Subsequently, the light phenols and aliphatic hydrocarbons entered the microporous HZSM-5 catalyst bed, where they underwent secondary catalytic reforming reactions, including deoxygenation and aromatization at the strong acid sites, ultimately yielding light aromatics. The Lewis acid sites of Al_2O_3 contributed to molecular-weight reduction by eliminating side chains and oxygen-containing groups, while the introduction of Nb further enhanced the activation and cleavage of both aliphatic and aromatic C-O bonds, thereby increasing the yield of light phenols.

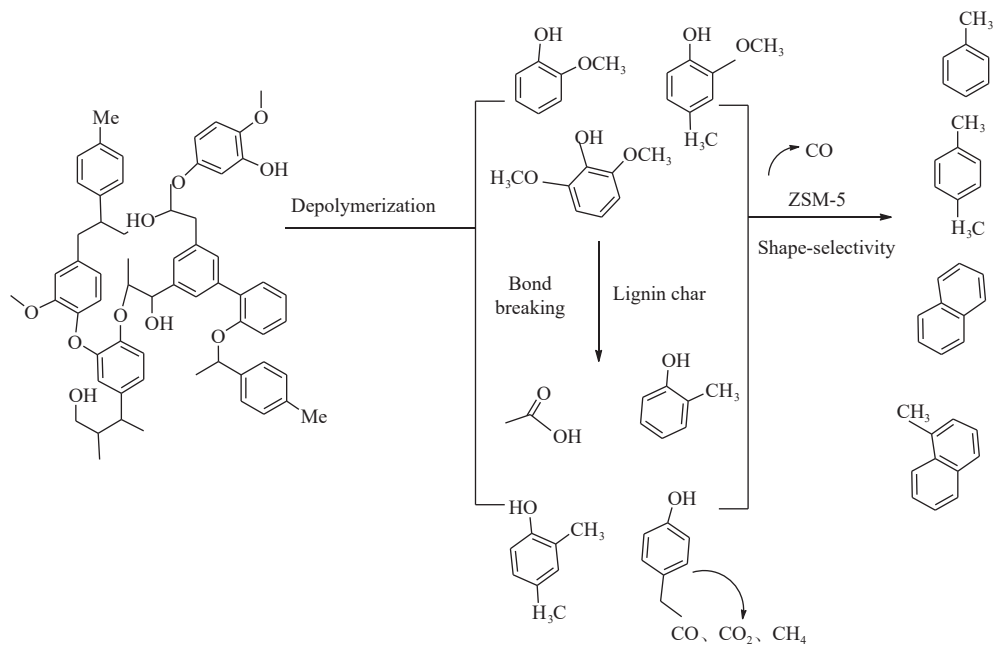


Fig. 10 Speculation on the mechanism of synergistic catalysis between alkaline lignin carbon and ZSM-5^[32] (with permission from Elsevier)

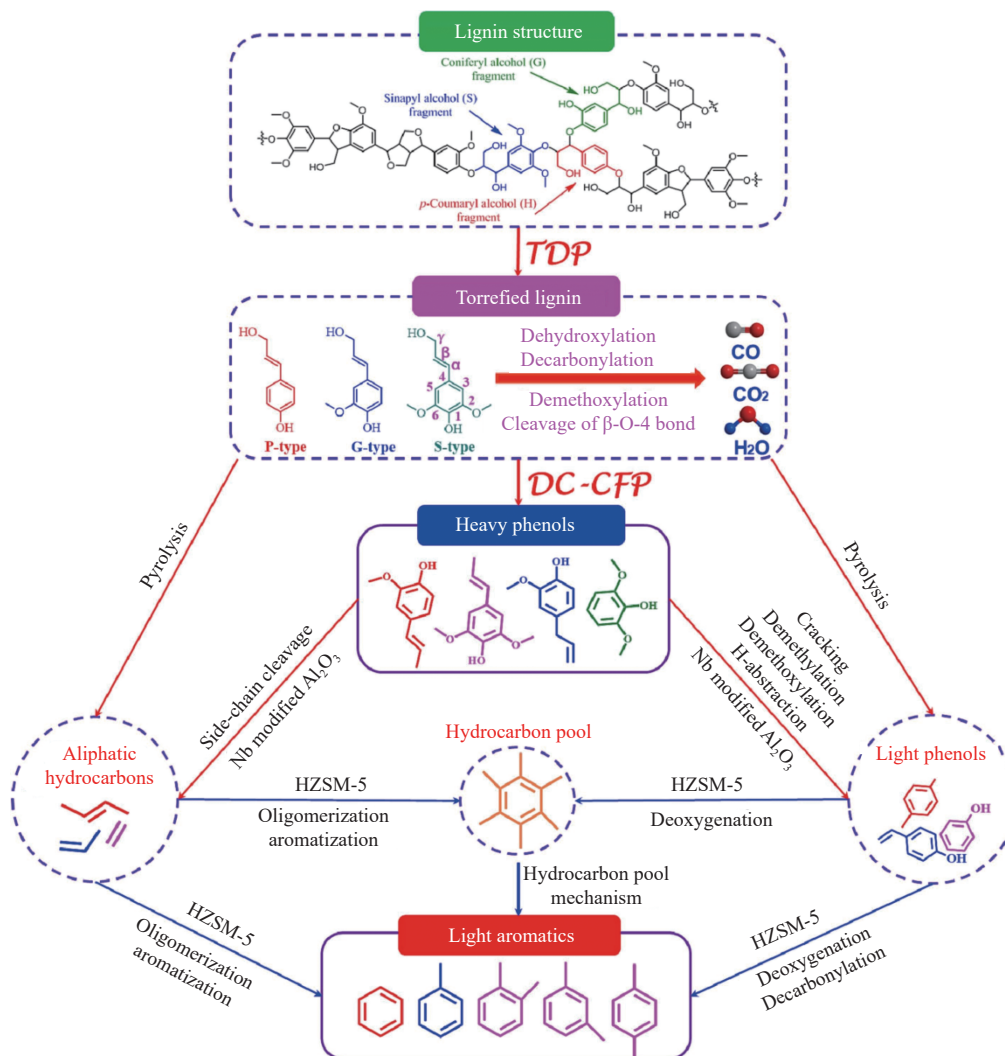


Fig. 11 Proposed formation mechanism of light aromatics during integrated approach of TDP and DC-CFP of MWL^[69] (with permission from Elsevier)

The conversion pathways of the main pyrolysis products of enzymatic hydrolysis lignin (EHL) over the Co-Ni@H/M catalyst were systematically investigated by Zheng et al.^[49], as illustrated in Figure 12. In this dual Co-Ni metal-loaded catalytic system, the increased number of the metal active sites significantly optimized the reaction pathways. On one hand, the dealkylation of phenol-O1 species (phenol and the monoalkylphenols such as *o*-cresol and *p*-cresol) and other oxygenated compounds produced short-chain alkyl groups (e.g., methyl, ethyl and propyl), which subsequently underwent oligomerization, cyclization, and aromatization to form MAHs. On the other hand, the synergistic effect of Co and Ni further enhanced the dealkylation efficiency of phenolic and oxygenated compounds and accelerated the directed conversion of short-chain alkyl groups into aromatics *via* the hydrocarbon pool mechanism, thereby increasing the yield of the aromatics while suppressing the coke formation.

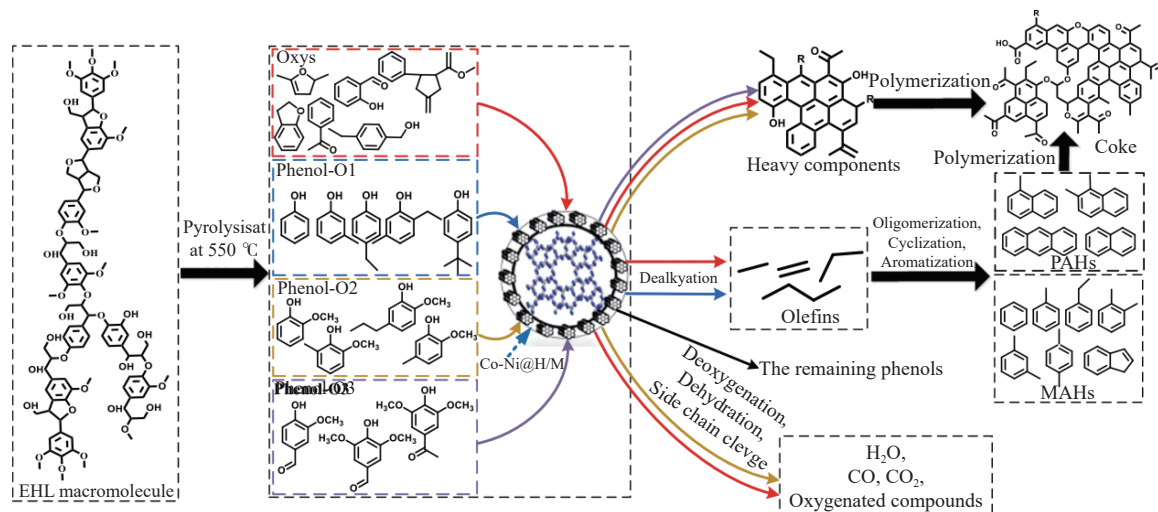


Fig. 12 Plausible conversion pathway of main pyrolysis products from EHL over Co-Ni@H/M^[49]
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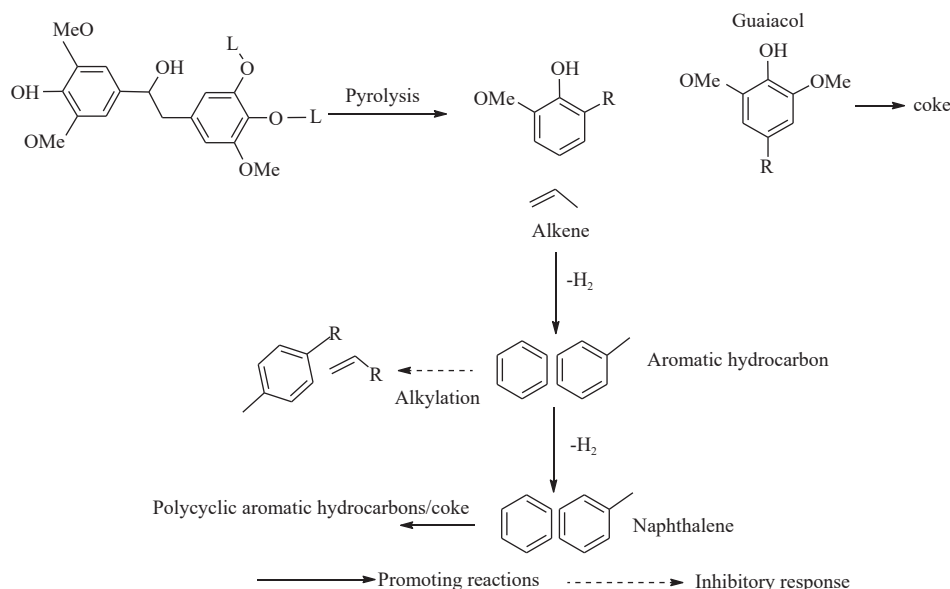


Fig. 13 Fe/H-ZSM-5 catalytic pyrolysis of lignin to produce aromatic hydrocarbons^[70]
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It was found that, the methoxy-substituted phenols underwent a series of intermediate transformations, including alkyl transfer and dealkylation reactions, and were subsequently converted into MAHs such as the BTEX within the molecular sieve hydrocarbon pool through dehydration, decarboxylation, and deoxygenation–dealkylation pathways. The synergistic effect of the metal oxides not only reduced the dissociation energy barriers of C–O and C–C bonds, facilitating the cleavage of lignin side chains, but also stabilized the valence state of Ni through coordination, thereby maintaining its catalytic activity and promoting hydrogen transfer and demethoxylation reactions. The mechanism of efficient lignin deoxygenation over the Ni–Mo bimetallic catalysts supported on NZ–HZSM-5 was further investigated by Lee et al.^[48]. The catalyst characterization indicated that metal modification significantly enhanced the textural properties and the acid site distribution of both the *in situ* natural zeolite (NZ) and the *ex situ* HZSM-5. The supported catalysts exhibited a substantially higher specific surface area, enhanced structural stability, and more accessible acid sites compared with the pristine supports. It was observed that the Ni active centers preferentially activated the C–O bonds in phenolic hydroxyl groups, whereas the Mo component selectively cleaved the methoxy structures. Together, these effects facilitated the directed transformation of radical intermediates and promoted deep deoxygenation, effectively suppressing coke formation. By regulating the molecular weight distribution of pyrolysis intermediates, the *in situ* depolymerization of the high-molecular-weight oxygenated compounds was promoted, providing dual benefits of high aromatic selectivity and sustained MAH productivity.

5 Conclusion and outlook

Lignin catalytic pyrolysis for the production of aromatic hydrocarbons offers a promising route to alleviate the current energy crisis, reduce dependence on fossil fuels, and valorize black liquor from the paper-making industry. This review systematically summarizes the effects of different pyrolysis strategies, catalyst types, and reaction parameters on lignin catalytic pyrolysis for the production of aromatic hydrocarbons, and provides a comprehensive overview of the corresponding catalytic reaction mechanisms. Future studies should prioritize the rational design of bifunctional catalyst systems that can simultaneously optimize deoxygenation efficiency and aromatization activity, thereby overcoming the persistent technical challenges related to aromatic selectivity and catalyst coking. Ultimately, such strategies aim to achieve the efficient and sustainable synthesis of lignin-derived aromatic hydrocarbons. The key perspectives can be summarized as follows:

Development of highly efficient, stable and reusable catalysts, coupled with the optimization of the reaction

conditions based on the properties of the feedstock, desired products and reactor configuration, to enhance the overall efficiency, selectivity and yield of catalytic pyrolysis.

Advancement of precise product quantification through the application of soft-ionization GC-MS techniques, standardization of analytical methods, establishment of comprehensive databases, and integration of machine learning-assisted quantitative analysis.

Investigation of optimized catalyst preparation routes and scalable process technologies, alongside the development of mature reactor systems for lignin catalytic pyrolysis to aromatics, to achieve the industrial implementation of high-value lignin conversion.

Declaration of Competing Interest

The authors declare no conflict of interest.

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木质素催化热解制芳烃研究进展

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摘要: 芳烃作为重要的化工原料广泛应用于能源、日用化工和医药等领域, 目前, 主要来源于石油炼制和催化重整, 木质素催化转化制芳烃是实现芳烃绿色生产的重要路径之一。木质素催化热解制备芳烃的核心在于催化剂酸性和孔径的调控、反应条件的优化和芳烃转化机理的探究, 因此, 本工作综述了热解方式、催化剂和反应条件等对木质素催化热解制备芳烃的影响, 系统概述了其催化反应机理, 并展望了未来重点研究方向和思路, 以为木质素催化热解制备芳烃提供理论和技术指导。

关键词: 木质素; 芳烃; 热解; 催化剂

中图分类号: TK-6

文献标识码: A

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