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Advances in Low-Rank Coal Pyrolysis: Strategies for Optimizing Product Quality and Expanding Applications in Clean Energy and Environmental Protection

Lijun Bai[1, a*]

[1]*College of Environmental Science and Engineering, Tongji University, Shanghai, China*

[a]*bailijun218@163.com*

[*]*corresponding author*

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Abstract: Low-rank coal, a widely available fossil fuel with abundant reserves, has significant potential for clean energy conversion. Pyrolysis is an effective method for utilizing low-rank coal sustainably. This study summarizes the characteristics of low-rank coal and provides an in-depth analysis of its pyrolysis process and product characteristics. It reveals that the low quality of pyrolysis products is a key factor limiting the development of low-rank coal pyrolysis technology. To address this issue, three product control strategies are proposed to enhance product quality and efficiency. The application of these strategies can effectively optimize the pyrolysis process and improve product performance, thereby expanding its potential applications in energy, chemicals, environmental protection, and new materials. Additionally, the study explores the broad application prospects of low-rank coal pyrolysis products, demonstrating the significant role of low-rank coal pyrolysis technology in optimizing energy structures and promoting environmental protection. The study provides solutions to the key bottlenecks constraining the development of pyrolysis technology for this low-rank coal, which is of great value for optimizing the energy structure and promoting environmental protection.

1. Introduction

Low-rank coal, a widely distributed and abundant conventional fossil fuel, has long been regarded as an inefficient energy source due to its high volatile matter, high moisture content (25%-65%), and high oxygen content. It is typically combusted directly, resulting in energy waste and exacerbating environmental pollution [1, 2]. However, with the growing demand for clean energy transition, the efficient and clean conversion of low-rank coal through pyrolysis has become a research focus. Pyrolysis, a simple and efficient thermochemical conversion method, can

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decompose low-rank coal into high-value products such as gases (e.g., hydrogen, methane), liquids (coal tar), and solids (semi-coke) under anoxic or low-oxygen conditions, providing an important route for coal resource utilization and sustainable development [3].

The pyrolysis process of low-rank coal is significantly influenced by its composition and structure. The abundant side chains and oxygen-containing functional groups (e.g., methoxy, carboxyl) in its chemical structure are prone to cleavage during the initial pyrolysis stage, releasing volatile components, while the condensation stage forms a stable semi-coke structure [4]. Parameters such as pyrolysis temperature, heating rate, atmosphere, and catalysts play a decisive role in the product distribution and quality. For example, low-temperature pyrolysis favors tar formation, while high temperature promotes the yield of syngas and high-quality semi-coke [5]. Additionally, the introduction of novel catalysts (e.g., metal oxides) can reduce activation energy and optimize product selectivity, while advanced reactor designs such as fluidized beds and rotary kilns enhance heat transfer efficiency and process continuity [6, 7].

Despite significant progress in low-rank coal pyrolysis technology, its industrial application still faces multiple challenges, including the control of pyrolysis reaction conditions, product stability and quality, and the subsequent processing and utilization of pyrolysis products. These issues require further research and resolution. To promote the industrialization of low-rank coal pyrolysis technology, cooperation with research institutions should be strengthened, focusing on technological breakthroughs and demonstration projects. Through the accumulation of practical experience and continuous technological improvements, the industrial application of low-rank coal pyrolysis can be gradually advanced.

This review aims to systematically analyze the core reaction mechanisms and key influencing factors of low-rank coal pyrolysis, exploring its application potential in clean energy technologies, with the goal of providing theoretical support for future research directions and technological upgrades. By integrating product control strategies for low-rank coal and analyzing the application potential of pyrolysis products, this paper aims to facilitate the transformation of low-rank coal from a "low-efficiency fuel" to a "high-value resource," contributing to the low-carbon and sustainable development of the global energy structure.

2. Characteristics of low-rank coal

2.1. Physico-chemical characterization of low-rank coal

Low-rank coal deposits are located near the surface, offering advantages such as easy accessibility, low extraction costs, high reactivity, and low prices. These attributes make the use of low-rank coal highly feasible for energy supply and industrial feedback[4]. The moisture, volatile matter, and fixed carbon content of coal vary due to factors such as temperature, burial pressure, and the duration of formation. According to the international coal classification standard ISO 11760-2018, low-rank coal has a bed moisture content of less than 75% and a mean random vitrinite reflectance ($\overline{R_r}$) of less than 0.5%. The subcategory classification standards for low-rank coal are shown in Table 1.

Table 1. subcategory classification standards of low-rank coal

Type	Parametric standards
C-grade low-rank coal (C grade lignite)	$\overline{R_r}$<0.4%, bed moisture 35%-75%, no ash base
B-grade low-rank coal (B grade lignite)	$\overline{R_r}$<0.4%, bed moisture less than 35%, no ash base
A-grade low-rank coal (subbituminous coal)	0.4%≤$\overline{R_r}$<0.5%

Typical low-rank coal is a soft, brittle material with a dull and simple appearance. It has a relatively high volatile matter content, and due to the abundance of C-C and C-O bonds, its pyrolysis, gasification, and other depolymerization reactions are significantly more reactive than those of higher-rank coals [8]. For example, during pyrolysis, free radical fragments are more easily generated and can enhance the yield of light tar through hydrogenation and catalysis [9]. Additionally, low-rank coal generally has a higher moisture content, especially in lignite, leading to low combustion efficiency and high pollution. Pre-drying is required to reduce processing energy consumption [2]. Furthermore, metal ions, such as sodium and calcium, accumulated in the ash due to carboxyl groups, can act as solvents in ash chemical reactions, affecting the ash fusion point and slagging behavior [10].

2.2. Organic structure characteristics of low-rank coal

The aromatic core structure of low-rank coal is relatively loose, with smaller aromatic nuclei and lower aromaticity. Each aromatic layer contains fewer carbon atoms, and the aromatic cores are not highly condensed. Instead, they are connected through biphenyl, naphthalene skeletons, as well as polymethylene and ether bridge bonds, forming a more open macromolecular network. This structural characteristic results in a higher proportion of amorphous carbon in the organic matrix of low-rank coal, with overall lower compactness [4, 8].

Low-rank coal also has abundant branched chains and functional groups. There are many long branched structures around the aromatic cores, and the content of oxygen-containing functional groups is significant. Oxygen atoms, as the main heteroatoms, exist in the form of hydroxyl (including phenolic), carbonyl, carboxyl, and methoxy groups. Some oxygen-containing functional groups are directly connected to the aromatic cores or participate in the linkage of different aromatic cores. Carboxyl functional groups also possess ion-exchange properties, which can accumulate alkali metal and alkaline earth metal ions, releasing them during combustion and affecting the ash's chemical behavior [4, 8].

3. Pyrolysis process, product characterization and regulation strategy

3.1. Pyrolysis process of low-rank coal

Pyrolysis is a simple and efficient method for the clean conversion of low-rank coal, enabling the direct production of clean fuels and high-value chemicals from the carbon and hydrogen compounds in low-rank coal. Due to its relatively low carbon content, improving the processing methods of low-rank coal and enhancing product quality can reduce greenhouse gas emissions, making it one of the key approaches to achieving carbon peak and carbon neutrality. The pyrolysis process is relatively complex, occurring under high temperatures in the absence of oxygen or in an inert atmosphere, involving both physical changes and chemical reactions. The pyrolysis classification of low-rank coal, according to different standards, is shown in Table 2 [8].

Table 2. Low-rank coal pyrolysis category

Temperature	Heating rate	Pyrolysis form
Low temperature pyrolysis (450-650℃)	Slow pyrolysis (1℃/s)	Traditional pyrolysis
Medium-level temperature pyrolysis (600-900℃)	Medium-speed pyrolysis (5-100℃/s)	Plasma pyrolysis
High temperature pyrolysis	Rapid pyrolysis	Hydropyrolysis

(900-1200℃)	(500-106℃/s)	
Ultra-high temperature pyrolysis (>1200℃)	Flash pyrolysis (>106℃/s)	Catalytic pyrolysis

The pyrolysis process of low-rank coal involves complex sequential chemical reactions, primarily consisting of a series of parallel free radical reactions. The free radical fragments produced during pyrolysis can react with each other to form volatiles and residues, such as tar or semi-coke [11]. Due to the diversity in the stability of the organic structure of low-rank coal, the primary driving factor of the pyrolysis process is temperature. As temperature increases, the thermal cracking of large organic molecules in the coal intensifies, leading to changes in the composition of pyrolysis products. The pyrolysis of low-rank coal can be divided into three stages: drying and degassing (room temperature 300℃), organic decomposition (300-600℃), and coking and polymerization (600-1000℃)[8, 12]. At temperatures below 300℃, the main hydrocarbon structures in low-rank coal undergo little change, but non-covalent bonds dissociate, and volatile components are released. Below 120℃, drying and dewatering primarily occur, while after this point, gases such as CO_2 , CH_4 , and N_2 are removed[12]. In the second stage of pyrolysis, organic depolymerization and decomposition reactions dominate, with the release of the majority of volatiles occurring after 350℃. At this point, polymethylene or ether bonds, side chains, and oxygen-containing functional groups are significantly broken down by heat, while some hydrogenated aromatics begin to dehydrogenate, generating highly reactive free radicals [8, 13]. These free radicals can stabilize through hydrogen transfer, rearrangement, crosslinking reactions, condensation reactions, or by capturing hydrogen through collisions with other fragments or free radicals. Primary products directly generated from macromolecules undergo secondary reactions during the devolatilization process [8, 13]. The third stage of low-rank coal pyrolysis mainly involves the polymerization of semi-coke into coke, while the tar further decomposes into gases such as H_2 and CO[12].

3.2. Pyrolysis products of low-rank coal

3.2.1. Coal gas

Coal gas primarily consists of combustible gases such as H_2 , CH_4 , CO, and CO_2 , with the proportion of hydrogen and methane increasing as the temperature rises. Additionally, sulfur- and nitrogen-containing compounds (e.g., H_2S, NH_3) may be produced due to the decomposition of impurities in the coal [14]. The coal gas obtained from low-rank coal pyrolysis has a high calorific value, typically ranging from 12-20 MJ/m^3; making it suitable as industrial fuel or synthesis gas feedstock for the production of chemicals such as methanol and ammonia [15]. Compared to direct combustion, coal gas results in lower pollutant emissions (SO_2 , NOx), aligning with the needs of clean energy transition [3].

However, coal gas derived from low-rank coal pyrolysis faces challenges such as severe dust carryover and unstable composition. When processing fine coal particles, the gas carries a large amount of dust, reducing the quality of subsequent products and affecting the calorific value and downstream utilization of the gas [5]. Moreover, traditional pyrolysis processes often dilute the coal gas with inert gases, lowering its calorific value, and imprecise temperature control can lead to fluctuations in gas composition. Additionally, coal gas is prone to secondary cracking under high temperatures, resulting in a reduction of light hydrocarbons and an increase in hydrogen content, which negatively impacts its economic feasibility as a fuel or chemical feedstock [16].

3.2.2. Coal tar

Coal tar mainly consists of light and heavy oils (collectively referred to as tar), containing macromolecular aromatic hydrocarbons such as phenols, naphthalenes, and asphaltenes [8]. In the vacuum distillation fraction of coal tar, components with a boiling point below 450℃ account for about 70%, and at high temperatures, secondary reactions may occur, forming coke particles [4]. Coal tar can be used as a chemical raw material for producing plastics, dyes, and pharmaceutical intermediates, or as a fuel oil to replace traditional petroleum products. Some tar can be further processed to extract high-value chemicals, such as phenol.

The heavy coal tar obtained from low-rank coal pyrolysis has a high proportion and is characterized by high viscosity and the tendency to form coke, which can easily lead to pipeline blockages and increase subsequent processing difficulties. Additionally, the presence of dust in the tar requires treatment through technologies such as centrifugation or electrostatic precipitation. However, existing technologies (e.g., high-temperature electrostatic dust removal) are costly and have limited efficiency [5]. The oil also contains heteroatom compounds, such as sulfur, nitrogen, and oxygen, as well as metal impurities, which require hydrorefining. However, hydrorefining processes involve high investment costs and expensive catalysts, limiting the economic feasibility [9].

3.2.3. Coal tar

Semi-coke obtained from low-rank coal pyrolysis is characterized by high carbon content and low ash content, with a calorific value reaching 25-30 MJ/kg. It has the advantages of low sulfur and low volatile matter, resulting in minimal combustion pollution [17]. However, its calorific value decreases naturally after pyrolysis. Additionally, semi-coke has a developed pore structure, which can be further activated to be used as an adsorbent or catalyst support [8]. However, as a solid fuel, the calorific value of semi-coke is lower than that of raw coal, and it requires supporting clean combustion technologies, which leads to low market acceptance. When used as a chemical raw material, further processing (e.g., gasification) is necessary, increasing the overall cost.

3.3. Product regulation strategy

3.3.1. Catalytic pyrolysis

The role of catalysts in coal pyrolysis is mainly reflected in two aspects: reducing the reaction conditions of coal pyrolysis and regulating the distribution and composition of pyrolysis products. Metal-based catalysts are commonly used in the coal pyrolysis process. Catalysts containing alkali metals (e.g., Na, K) or alkaline earth metals (e.g., Ca, Mg) can enhance the gasification reactivity of pyrolysis semi-coke, promoting its thorough pyrolysis [18]. Alkali metal carbonates, such as K_2CO_3 and Na_2CO_3, can increase the yield of pyrolysis coal gas and improve the gasification reaction rate of semi-coke [19]. Alkaline earth metal oxides, such as CaO and MgO, can promote the decomposition of tar and increase the content of H_2 and CH_4 in coal gas [20]. Furthermore, some transition metals (e.g., Co, Mo, Ni) loaded on molecular sieves can facilitate the directional conversion of low-rank coal into light aromatics, increasing the value of the tar [21]. However, during pyrolysis, direct contact between the catalyst and low-rank coal can lead to catalyst deactivation.

3.3.2. Atmosphere

Changing the pyrolysis atmosphere has been proven to effectively regulate the quality of pyrolysis products. The pyrolysis atmosphere participates in the thermal decomposition reactions of low-rank coal, altering its reaction mechanism and significantly improving the quality of the pyrolysis products. For example, using a hydrogen-rich atmosphere (e.g., H_2 , coke oven gas) can promote hydrogenation reactions, increasing the proportion of light components in the tar. However, due to the high cost of hydrogen, its use as a pyrolysis atmosphere for low-rank coal presents a lower economic benefit [9]. Therefore, replacing pure hydrogen with cheaper coke oven gas or synthesis gas for coal hydrogenation pyrolysis is not only feasible but also shows significant advantages [22]. In addition, pyrolysis of low-rank coal in a CO_2 atmosphere can increase the tar yield. CO_2 reacts with alkanes in the volatiles to undergo reforming reactions, generating hydrogen radicals, which further promote the breaking of ether bonds, Car-Car bridge bonds, and aliphatic C-C bonds. This reaction mechanism increases the tar yield from 17.98% to 20.68% [23]. Regulating the pyrolysis atmosphere not only improves product quality but also offers economic advantages, providing new insights for optimizing low-rank coal pyrolysis technology.

3.3.2. Pyrolysis parameter

The pyrolysis behavior of low-rank coal is closely related to temperature and pyrolysis residence time. Increasing temperature promotes the secondary cracking and condensation reactions of coal tar, but it can also have adverse effects. Xu et al. [24] demonstrated that when the pyrolysis temperature is raised from 500℃ to 900℃, the tar yield is reduced by half. While high temperatures help achieve thorough pyrolysis of low-rank coal, they also increase costs and may lead to carbon deposition on the pyrolysis furnace walls, which in turn affects heat transfer efficiency [25]. Furthermore, the length of the pyrolysis residence time is correlated with changes in product yield. At temperatures below 600℃, the residence time has a minimal effect on coal tar yield. However, when the temperature reaches 600℃, extending the residence time significantly reduces the tar yield and increases the gas yield [25]. When the residence time is extended from 0.4 seconds to 14 seconds, its effect is comparable to increasing the temperature by 100℃ [24]. Additionally, selecting an appropriate pyrolysis reactor type can regulate the residence time of volatile products. For instance, using a fluidized bed reactor can shorten the residence time of volatiles, but it is important to carefully balance the pyrolysis temperature and heat transfer efficiency.

4. Application potential of low-rank coal pyrolysis products

Low-rank coal pyrolysis products exhibit significant application potential across multiple fields. In the energy sector, coal gas can be used as a clean fuel for power generation and heating, replacing traditional coal combustion and reducing pollutant emissions. After processing, coal tar can be converted into liquid fuels, alleviating petroleum resource shortages. Additionally, the oil and gas produced through pyrolysis can be transformed into liquefied natural gas (LNG) and liquefied petroleum gas (LPG), reducing dependence on foreign oil and gas resources. In the chemical industry, coal tar can be hydrogenated or catalytically reformed to produce high-value chemicals such as benzene, toluene, and xylene. The CO and H_2 in coal gas are key raw materials for synthesizing methanol, dimethyl ether, and other chemicals. Pyrolysis technology also offers environmental benefits by reducing pollutant emissions and aligning with carbon-neutral goals, with carbon capture technology enabling resource recycling.

Furthermore, low-rank coal pyrolysis products show immense potential in the fields of new materials and other industrial applications. The recondensed components of coal tar can be used to produce high-performance plastics, rubber, and carbon materials. Semi-coke can serve as a metallurgical reducing agent or be used as battery electrode materials. With technological innovation and industry chain integration, low-rank coal pyrolysis technology holds vast prospects for improving resource utilization, optimizing energy structures, and facilitating green low-carbon transformation. National policy support and technological breakthroughs will further unleash its market potential, driving the industrialization of low-rank coal fractionation utilization demonstration projects.

5. Conclusion

This study summarizes the characteristics of low-rank coal, providing an in-depth analysis of its pyrolysis process and product characteristics. It reveals that the low quality of pyrolysis products is a key factor limiting the development of low-rank coal pyrolysis technology. To address this issue, the study proposes three product control strategies aimed at improving the quality and efficiency of pyrolysis products. The application of these strategies can effectively optimize the pyrolysis process, enhance product performance, and expand its potential applications in the energy, chemical, environmental, and new materials sectors. Additionally, the study explores the broad application prospects of low-rank coal pyrolysis products, particularly in alternative energy, chemical synthesis, and low-carbon transformation, highlighting the significant role of low-rank coal pyrolysis technology in optimizing energy structures and environmental protection.

Low-rank coal pyrolysis technology, as an important coal processing method, is crucial for enhancing coal utilization efficiency and reducing energy consumption. However, the current low efficiency of the pyrolysis process severely restricts the promotion and application of this technology. To solve this issue, technological innovation and research and development are needed to continuously optimize the pyrolysis process, develop new equipment, and improve the pyrolysis efficiency of low-rank coal. At the same time, actively expanding the downstream markets for pyrolysis products, especially in industries such as pharmaceuticals, agriculture, and power generation, will help further enhance the market value of the technology. A well-developed industrial chain structure is key to the long-term and stable development of low-rank coal pyrolysis technology. By optimizing the various links in the industrial chain and establishing full-chain cooperation from raw material supply to pyrolysis process and product application, the commercialization of the technology can be effectively promoted, providing strong support for the sustainable development of the industry. Therefore, driving technological innovation and market expansion, and building a complete industrial chain, will not only improve the application efficiency of low-rank coal pyrolysis technology but also lay a solid foundation for the long-term and stable development of the industry.

Data Availability

Data sharing is not applicable to this article as no new data were created or analysed in this study.

Conflict of Interest

The author states that this article has no conflict of interest.

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