Sustainability in Environment
ISSN 2470-637X (Print) ISSN 2470-6388 (Online)
Vol. 11, No. 1, 2026
www.scholink.org/ojs/index.php/se

Original Paper

A Review of In-situ Pyrolysis Research on Tar-rich Coal and Oil Shale

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Received: January 03, 2026 Accepted: February 26, 2026 Online Published: March 15, 2026
doi:10.22158/se.v11n1p271 URL: http://dx.doi.org/10.22158/se.v11n1p271

Abstract

As two important unconventional oil and gas resources, the in-situ pyrolysis technologies of tar-rich Coal and oil shale are key approaches to realize the clean and efficient utilization of resources, alleviate the contradiction between oil and gas supply and demand, and help achieve the "double carbon" goals. Both of them share the core characteristic of being rich in organic matter and convertible into oil and gas products through pyrolysis, yet they exhibit significant differences in resource endowments, pyrolysis characteristics and technological applications. This paper systematically combs the technical principles of in-situ pyrolysis for tar-rich Coal and oil shale, focuses on a comparative analysis of their similarities and differences in pyrolysis kinetics, process routes, key technologies and environmental impacts, and deeply analyzes the common bottlenecks and personalized challenges faced by the current in-situ pyrolysis technologies. It provides theoretical support and practical reference for the optimization and industrial promotion of in-situ pyrolysis technologies for tar-rich Coal and oil shale.

Keywords

Tar-rich Coal, Oil shale, In-situ pyrolysis, Pyrolysis characteristics, Technical comparison, Research review

1. Introduction

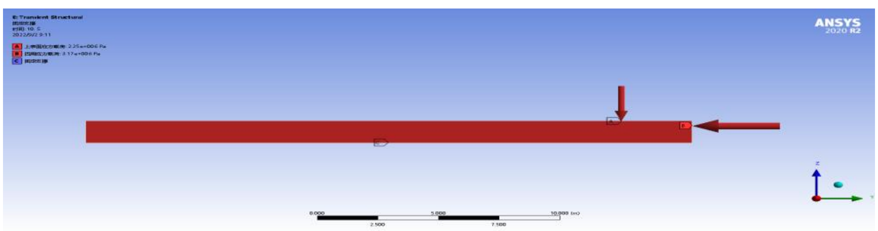
With the accelerated transformation of the global energy structure, the reserves of conventional oil and gas resources are depleting day by day, and the guarantee of energy security is facing severe challenges. China's energy structure is characterized by "abundant coal, deficient oil and scarce gas", with a long-term high external dependence on oil and gas. In 2024, China's external dependence on crude oil still exceeded 70%, and the external dependence on natural gas was more than 40%, making the tasks of energy structure transformation and security guarantee arduous. As unconventional oil and gas resources with abundant reserves and huge potential, the organic matter contained in tar-rich Coal and oil shale can be converted into high value-added products such as tar, coal gas and shale oil through pyrolysis, which serve as an important strategic reserve for replacing conventional oil and gas resources and optimizing the energy structure.

Although tar-rich Coal and oil shale both belong to pyrolyzable unconventional resources, their significant differences in resource endowments and pyrolysis characteristics lead to great variations in their in-situ pyrolysis technical routes, key technical difficulties and application scenarios. At present, relevant researches at home and abroad mostly focus on the technical tackling of in-situ pyrolysis for a single resource, lacking systematic comparison and collaborative research on the two, which makes it difficult to give full play to the development potential of the two resources. Therefore, carrying out a review of in-situ pyrolysis research on tar-rich Coal and oil shale, systematically sorting out their research status, comparing technical differences, analyzing existing problems and prospecting development trends, has important theoretical value and practical significance for promoting the collaborative optimization of in-situ pyrolysis technologies for the two resources, breaking through technical bottlenecks and realizing industrial promotion.

2 Technical Principles and Kinetic Research of In-situ Pyrolysis for tar-rich Coal and Oil Shale

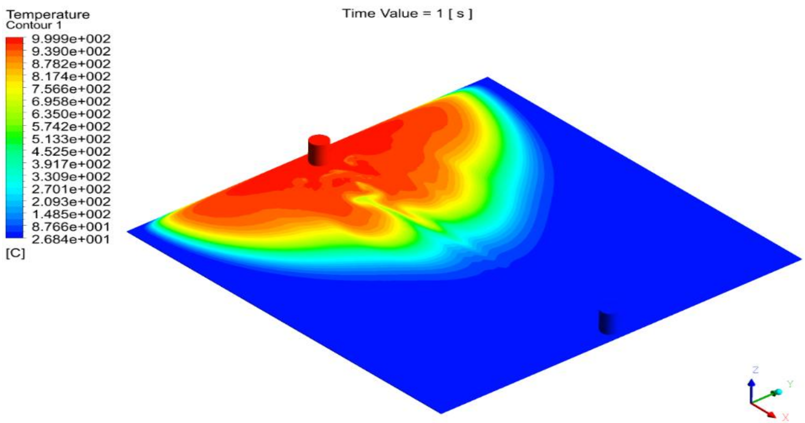
2.1 Technical Principles of In-situ Pyrolysis

The core principles of in-situ pyrolysis for tar-rich Coal and oil shale are common: the temperature of underground reservoirs is raised to the pyrolysis temperature of organic matter through artificial heating, causing the organic matter to undergo thermal decomposition reactions and convert into three types of products—liquid (tar, shale oil), gaseous (coal gas) and solid (semi-coke, oil shale semi-coke). The liquid and gaseous products are then transported to the surface through special channels for separation, purification and utilization, while the solid products remain underground or are mined and utilized in the follow-up. The entire pyrolysis process must be carried out in an oxygen-free environment to avoid oxidative combustion of pyrolysis products, and the heating rate and temperature distribution must be controlled to ensure the stable progress of the pyrolysis reaction.



Schematic diagram of the in-situ pyrolysis model for tar-rich Coal

The pyrolysis processes of both can be divided into three stages: the drying and degassing stage (temperature below 300℃), in which moisture and adsorbed small molecular gases in the reservoir are mainly removed without obvious decomposition of organic matter; the oil and gas production stage via pyrolysis (300~650℃), where the organic matter in tar-rich Coal and kerogen in oil shale begin to decompose in large quantities, producing a large number of products such as tar (shale oil) and coal gas, with the yield of liquid products reaching a peak. At present, 350~450℃ is generally recognized as the high-efficiency pyrolysis temperature range, though the optimal pyrolysis temperature range varies slightly with different coal qualities, oil shale characteristics and pyrolysis atmospheres; the semi-coke formation stage (650~800℃), in which the remaining organic matter decomposes further, the output of coal gas increases continuously, the yield of liquid products decreases gradually, and porous semi-coke (oil shale semi-coke) is finally formed.



Underground pyrolysis diagram of tar-rich coal

Due to the differences in resource characteristics, there are certain differences in the technical principles of their in-situ pyrolysis. tar-rich Coal has good air permeability, so heat carriers (high-temperature flue gas, steam, nitrogen, etc.) can quickly penetrate into the coal seam during pyrolysis. Heat transfer is mainly based on thermal convection and conduction, and pyrolysis products can quickly migrate to production wells through coal seam pores without complex reservoir reconstruction. In contrast, oil shale has extremely poor air permeability, making it difficult for heat carriers to penetrate. Heat transfer is mainly through thermal conduction, and the migration speed of pyrolysis products is slow. Reservoir reconstruction technologies (hydraulic fracturing, controlled shock wave fracturing, etc.) are required to form fracture networks and improve air permeability and product migration efficiency. In addition, tar-rich Coal can adopt the controlled oxidative spontaneous combustion method to provide the heat source for pyrolysis without continuous external heating, while oil shale is difficult to use the spontaneous combustion heating method due to its low organic matter content and poor combustibility, and relies on continuous external heating.

Pyrolysis diagram of tar-rich coal specimen

2.2 Kinetic Research of In-situ Pyrolysis

In-situ pyrolysis kinetics is the core content of studying the pyrolysis reaction rate, reaction mechanism and influencing factors of tar-rich Coal and oil shale, which can provide theoretical support for the optimization of in-situ pyrolysis process parameters and the design of heating systems. At present, domestic and foreign researches on the pyrolysis kinetics of the two mainly adopt thermogravimetric analysis (TG-DTG), differential scanning calorimetry (DSC) and other methods. By establishing kinetic models, key parameters such as activation energy and frequency factor of pyrolysis reactions are calculated to reveal the essential laws of pyrolysis reactions.

Studies on the in-situ pyrolysis kinetics of tar-rich Coal show that the pyrolysis reaction of tar-rich Coal is a complex multi-step reaction process, mainly divided into three continuous reaction stages: volatile release, tar formation and semi-coke formation, with significant differences in the reaction kinetic parameters of each stage. The pyrolysis activation energy of tar-rich Coal is usually between 80~150kJ/mol, which is relatively low, indicating that its pyrolysis reaction is easy to occur, which is closely related to the characteristics of high volatile content and high hydrogen content of tar-rich Coal. The order of pyrolysis reaction is usually between 1 and 2, and the reaction rate is greatly affected by temperature, heating rate, coal seam permeability and other factors, among which temperature is the most critical factor affecting the pyrolysis reaction rate. The increase of temperature can significantly reduce the activation energy and accelerate the pyrolysis reaction process. In addition, the pyrolysis kinetic characteristics of tar-rich Coal are also closely related to coal quality and pyrolysis atmosphere. The pyrolysis activation energy is lower in an inert atmosphere, resulting in higher pyrolysis efficiency.

Studies on the in-situ pyrolysis kinetics of oil shale show that the pyrolysis reaction of oil shale is mainly the thermal decomposition reaction of kerogen, divided into three stages: kerogen cracking, asphaltene formation and asphaltene cracking. Its pyrolysis activation energy is usually between 120~200kJ/mol, higher than that of tar-rich Coal, indicating that the pyrolysis reaction of oil shale is more difficult and requires higher heating temperature and longer heating time. The order of pyrolysis reaction is usually between 1.5 and 2.5, and the reaction rate is significantly affected by temperature, heating rate, reservoir permeability and other factors. The inorganic minerals in oil shale have a certain catalytic effect on the pyrolysis reaction: clay minerals can reduce the pyrolysis activation energy and accelerate the pyrolysis reaction process, while minerals such as quartz and feldspar have no obvious catalytic effect on the pyrolysis reaction and may even hinder heat transfer and reduce pyrolysis efficiency. TG-DTG experiments show that at a heating rate of 20°C/min, the conversion of oxygen-containing compounds to phenolic compounds is inhibited during the co-pyrolysis of tar-rich Coal and oil shale; in the air, an excessively fast heating rate will cause tar to condense in advance and coke on the sample surface, reducing the pyrolysis index from 3.2 to 1.8, while the pyrolysis stability is increased by 6.7 times. The introduction of excessive oxygen-containing groups in the air increases the content of oxygen-containing compounds in the mixed sample by 485.44% and decreases the content of aromatic hydrocarbons by 17.97%.

In general, the pyrolysis activation energy of tar-rich Coal is lower than that of oil shale, making its pyrolysis reaction easier to occur and the pyrolysis rate faster, which is one of the important reasons why the in-situ pyrolysis technology of tar-rich Coal is easier to realize industrialization. At the same time, the pyrolysis kinetic characteristics of both are affected by many factors. The pyrolysis activation energy can be effectively reduced, and the pyrolysis efficiency and product yield can be improved by optimizing pyrolysis process parameters, adding catalysts and improving the pyrolysis atmosphere.

3. Research Status of In-situ Pyrolysis Technologies for tar-rich Coal and Oil Shale

3.1 Research Status of In-situ Pyrolysis Technology for tar-rich Coal

At present, a variety of process routes have been formed for the in-situ pyrolysis technology of tar-rich Coal. According to different heating methods, it is mainly divided into three types: heat carrier injection type, electric heating type and controlled oxidative spontaneous combustion type. The technical characteristics and application scenarios of different process routes are significantly different, and relevant researches focus on process optimization, key technology breakthroughs and field test verification.

Heat carrier injection pyrolysis is the most widely used in-situ pyrolysis process for tar-rich Coal at present. Its core is to inject high-temperature heat carriers (high-temperature flue gas, steam, nitrogen, etc.) into underground coal seams through surface boreholes, and transfer heat to the coal seams by thermal convection and conduction to achieve in-situ pyrolysis. The advantages of this process are high heating efficiency, uniform heat distribution, and the ability to rapidly raise the temperature of coal seams, making it suitable for medium-thick coal seams with good air permeability. The disadvantages are large consumption of heat carriers, the need for supporting construction of surface heating devices, high investment costs, and the risk of environmental pollution caused by heat carrier leakage. Domestic and foreign researches focus on optimizing the type and injection parameters of heat carriers. At present, nitrogen and carbon dioxide are the most widely used heat carriers. Carbon dioxide as a heat carrier can realize the coordination of pyrolysis and carbon sequestration, reduce carbon emissions, and also play a role in oil and gas displacement. In the pilot test of in-situ pyrolysis of tar-rich Coal in Northern Shaanxi, nitrogen was used as the heat carrier for convection heating, and the coal seam permeability was increased by controlled shock wave technology, with an effective permeability enhancement radius of 40~60m, achieving good pyrolysis effects. In addition, studies recommend the adoption of composite heating technology dominated by convection heating and supplemented by other heating methods, and the waste heat of in-situ pyrolysis is used to preheat the heat carrier fluid to reduce the energy consumption of heat injection.

Electric heating pyrolysis realizes in-situ pyrolysis by arranging electrodes in underground coal seams and heating the coal seams by using the thermal effect of electric current. The advantages of this process are precise temperature control, small heat loss, no risk of heat carrier leakage, and strong environmental friendliness. The disadvantages are high energy consumption and great difficulty in electrode arrangement, making it suitable for coal seams with poor air permeability and complex geological conditions. At present, relevant researches focus on optimizing the electrode arrangement, improving heating efficiency, reducing energy consumption, and developing a closed double-shell downhole electric heating device, which solves the problems of combustion reaction stability and repeated ignition reliability existing in downhole combustion heaters. However, this device faces performance challenges such as long service life (3~5 years), high heating temperature (600℃) and large operating power (>2kw/m). At the same time, microwave heating, as a new type of electric heating method, has also been applied to the research of in-situ pyrolysis of tar-rich Coal, which has the advantages of uniform heating, high efficiency and non-contact heating, and can significantly improve pyrolysis efficiency and product yield.

Controlled oxidative spontaneous combustion pyrolysis is a new type of pyrolysis process. Its core is to use the oxidative spontaneous combustion of pyrolysis semi-coke as the heat source for in-situ pyrolysis, establish an efficient heat transfer network through artificial fracturing of coal seams, and improve the heat transfer efficiency and combustion efficiency by using heat-generating agents or catalysts. The advantages of this process are no need for external heat source, low energy consumption, low cost, and the ability to realize the recycling of heat. The disadvantages are that the combustion process is difficult to control and prone to safety risks such as fire, and it is still in the experimental research stage at present. Relevant researches focus on the regulation of spontaneous combustion process and the control of combustion range, and reduce safety risks and improve pyrolysis stability by optimizing fracturing parameters and adding flame retardants.

3.2 Research Status of In-situ Pyrolysis Technology for Oil Shale

The research focus of oil shale in-situ pyrolysis technology is to solve the problems of poor reservoir permeability, difficult heat transfer and slow product migration. According to different heating methods, it is mainly divided into four types: electric heating type, heat carrier injection type, microwave heating type and chemical heating type. At present, relevant researches are in the pilot test stage, and some technologies have realized small-scale industrial application.

Electric heating pyrolysis is the mainstream process of oil shale in-situ pyrolysis, mainly including resistance heating and electromagnetic heating. Resistance heating heats oil shale by arranging heating wells in the oil shale reservoir and using resistance heating, with precise temperature control and small heat loss, suitable for deep oil shale reservoirs. Electromagnetic heating generates heat by using the principle of electromagnetic induction, with high heating efficiency and uniform heating, which can realize rapid pyrolysis of oil shale and has become a research hotspot of oil shale in-situ pyrolysis at present. Shell's ICP technology and ExxonMobil's EIS technology in the United States both adopt electric heating methods, and realize high-efficiency pyrolysis of oil shale by optimizing the arrangement of heating wells and increasing heating power, with a shale oil yield of 5%~8%. Relevant researches in China focus on optimizing electric heating devices, reducing energy consumption, developing a new type of high-efficiency electromagnetic heating device, improving heat transfer efficiency and reducing heating costs.

The heat carrier injection pyrolysis has a similar principle to the relevant process of tar-rich Coal, which injects high-temperature heat carriers (high-temperature flue gas, steam, hot oil, etc.) into the oil shale reservoir through surface boreholes and transfers heat by thermal convection to achieve in-situ pyrolysis. Due to the extremely poor air permeability of oil shale, this process requires first reconstructing the oil shale reservoir to form fracture networks and improve the penetration efficiency of heat carriers. At present, relevant researches focus on optimizing the type of heat carriers, injection parameters and reservoir reconstruction technologies. High-temperature steam is used as the heat carrier, which can not only transfer heat, but also undergo hydrothermal reaction with oil shale, reduce pyrolysis activation energy and improve shale oil yield. At the same time, combined with hydraulic fracturing, high-energy gas fracturing and other technologies, the fracture network structure is optimized to improve the penetration efficiency of heat carriers and the migration speed of products. The oil shale in-situ pyrolysis technology in Estonia adopts the heat carrier circulation heating method, realizing small-scale industrial application with a shale oil yield of 6%~9%.

Microwave heating pyrolysis is a new type of oil shale in-situ pyrolysis process. Its core is to heat oil shale by using the thermal effect of microwaves, which can penetrate the oil shale formation, realize rapid internal heating, with high heating efficiency and uniform heating, which can significantly shorten the pyrolysis cycle and improve shale oil yield. Relevant studies show that microwave heating can reduce the pyrolysis activation energy of oil shale by 20%~30%, shorten the pyrolysis cycle by 30%~50%, and increase the shale oil yield by 10%~15%. At present, the research focus of microwave heating pyrolysis is to develop high-efficiency microwave heating devices, solve the problem of microwave leakage, optimize microwave heating parameters, and improve the feasibility and safety of the technology. Studies have found that optimizing microwave frequency, improving magnetron design and adding microwave absorbents can significantly improve heating efficiency and uniformity. At the same time, this technology has the characteristics of environmental friendliness, and can realize crude oil cracking and desulfurization under the action of catalysts to improve crude oil quality.

Chemical heating pyrolysis heats oil shale by injecting chemical agents (such as oxidants, catalysts, etc.) into the oil shale reservoir and using the heat released by chemical reactions to achieve in-situ pyrolysis. The advantages of this process are no need for external heating equipment, low energy consumption and low cost, suitable for deep and remote oil shale reservoirs. The disadvantages are large consumption of chemical agents, difficult control of the reaction process, and possible secondary pollution. At present, relevant researches focus on optimizing the formula of chemical agents, controlling the reaction process, improving pyrolysis efficiency and reducing the risk of environmental pollution, which are mainly in the laboratory research stage and have not carried out field tests.

The key technologies of oil shale in-situ pyrolysis also include reservoir reconstruction technology, product separation technology, environmental governance technology, etc. In terms of reservoir reconstruction, hydraulic fracturing, high-energy gas fracturing, hydraulic jet fracturing and other technologies are widely used to form fracture networks through reconstruction, improving the air permeability of oil shale and the migration efficiency of products; in terms of product separation, high-efficiency surface separation devices have been developed to realize the separation and purification of shale oil, coal gas and waste residue, improving product quality; in terms of environmental governance, the focus is on solving the treatment of pyrolysis wastewater, waste gas and waste residue, and adopting biochemical treatment, adsorption treatment and other technologies to reduce environmental pollution. At present, the pilot test of oil shale in-situ pyrolysis carried out in the Songliao Basin in China has a shale oil yield of 5%~7% and a coal gas yield of 80~120m^3/t, but there are still problems such as insufficient technical maturity, high investment cost and great difficulty in pollutant treatment.

4. Commonness and Individuality of In-situ Pyrolysis Technologies for the Two Resources

The in-situ pyrolysis technologies of tar-rich Coal and oil shale have certain commonness, mainly reflected in: consistent core principles, both of which realize the thermal decomposition of organic matter into liquid, gaseous and solid products through in-situ heating; similar process routes, including heating system, reservoir reconstruction system, product production system and surface treatment system; common problems faced, such as insufficient technical maturity, high energy consumption, to-be-improved product recovery rate and great difficulty in environmental risk control.

The individual differences between the two are mainly reflected in: first, different heating method selections—tar-rich Coal can adopt controlled oxidative spontaneous combustion heating without continuous external heating, while oil shale is difficult to use spontaneous combustion heating and relies on continuous external heating; second, different reservoir reconstruction demands—tar-rich Coal has good air permeability with low reservoir reconstruction demands, while oil shale has extremely poor air permeability, making reservoir reconstruction the key technology; third, different product characteristics—tar-rich Coal has a high tar yield and low quality through pyrolysis, requiring deep processing to increase added value, while oil shale has a low shale oil yield and high quality through pyrolysis, which can be directly processed into finished oil; fourth, different technical maturity—the in-situ pyrolysis technology of tar-rich Coal has made rapid progress in tests, and some processes are close to industrialization, while the oil shale in-situ pyrolysis technology is more difficult and still in the pilot test stage; fifth, different safety risks—the in-situ pyrolysis of tar-rich Coal faces safety risks such as spontaneous combustion and gas leakage, while the in-situ pyrolysis of oil shale mainly faces safety risks such as heat carrier leakage and uncontrolled fracture propagation. In addition, the safety pre-control of tar-rich Coal in-situ pyrolysis faces challenges such as invisible process, inaccessible personnel and complex underground reaction conditions, and it is necessary to focus on issues such as the combustion and explosion characteristics of oil and gas production, pyrolysis reaction regulation, fracture propagation and oil and gas migration laws.

5. Conclusions

tar-rich Coal has the advantages of abundant reserves, shallow burial, good air permeability and low pyrolysis activation energy. Its in-situ pyrolysis technologies are mainly heat carrier injection type, electric heating type and controlled oxidative spontaneous combustion type, with high technical maturity and rapid test progress. Its environmental friendliness and economic feasibility are superior to those of oil shale in-situ pyrolysis, making it an important choice for the large-scale development of unconventional oil and gas resources in the short term. Oil shale has huge reserves and high-quality shale oil, but with deep burial, poor air permeability and high pyrolysis activation energy. Its in-situ pyrolysis technologies are mainly electric heating type, heat carrier injection type and microwave heating type, with great technical difficulty, still in the pilot test stage, and high environmental risks and costs, serving as an important force to supplement China's oil and gas resources in the long term. TG-DTG experiments show that the co-pyrolysis of the two has a good synergistic effect, which can improve the ignition characteristics of oil shale and enhance pyrolysis efficiency.

At present, the in-situ pyrolysis technologies of tar-rich Coal and oil shale both face common problems such as insufficient technical maturity, great difficulty in environmental risk control and to-be-improved economic feasibility, as well as their own personalized problems. In the future, it is necessary to strengthen the research and development and innovation of core technologies, deepen the research on pyrolysis kinetics and multi-field coupling, optimize environmental governance technologies, improve economic feasibility, strengthen collaborative research and international cooperation, promote the coordinated development and industrial promotion of in-situ pyrolysis technologies for the two resources, give full play to their resource potential, and provide strong support for China's energy structure transformation and security guarantee.

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