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Review on in situ conversion of oil shale

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Abstract. *In situ conversion technology for oil shale is an innovative method of energy extraction that involves the underground heating of oil shale reservoirs to thermally crack kerogen into oil and gas. This approach avoids the environmental damage and high energy consumption associated with traditional mining methods and offers advantages such as reduced environmental impact, small ecological footprint, and low development costs. In this paper, a variety of in situ conversion technologies, including in situ conversion process (ICP), Electrofrac, geothermic fuels cells process (GFC), and supercritical water heating, are discussed in detail. The paper also analyzes the current state and development trends of in situ conversion technologies in China, and highlights the importance of advancing basic theoretical research and overcoming key technologies to enable large-scale utilization of oil shale resources. Continued research and improvement in in situ conversion technologies will enhance both the efficiency and sustainability of the energy industry.*

Keywords: *oil shale, in situ conversion, dry distillation, underground heating, shale oil, supercritical water.*

1. Introduction

With the rapid development of the global economy, the supply of traditional oil resources has been unable to meet demand, and oil prices fluctuate frequently and increase gradually. Oil shale is a type of sedimentary rock with high ash content and combustible material. It is composed of organic matter and a variety of minerals. These components have a direct influence on the oil storage capacity and pyrolysis efficiency of oil shale. After being heated, products such as hydrocarbons and solids are generated, and through

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a series of processing steps, energy products such as shale oil and shale gas can be obtained. This brings great challenges to global energy security. In this context, shale oil, as an important unconventional oil and gas resource, has gradually entered public attention and become a research hotspot in the global energy field [1].

The oil shale mining industry can be traced back to the last century. After half a century of development, the production process has become relatively mature, mainly divided into above-ground dry distillation and underground dry distillation. In China, oil shale production is primarily based on above-ground dry distillation. However, because most of the oil shale reservoirs in China are continental and of low maturity, continuous reliance on surface retorting technology can no longer meet growing demand. Therefore, it is imperative to select appropriate in situ conversion technologies [2–5].

The world is extremely rich in oil shale resources. According to the U.S. Energy Information Administration and other institutions, the world has tens of billions of tons of technologically recoverable resources [5]. By 2022, China's oil shale reserves totaled 7.2×10^{11} tons, but annual production was only 7.8×10^{5} tons [5]. Among them, the United States, as a pioneer in shale oil development, has accounted for half of total shale oil production, having a profound impact on the global energy market. Russia and China, also rich in oil shale resources, are among the world's leading energy powers and are actively promoting the exploration and development of shale oil [6].

China is extremely rich in oil shale resources, with about 47.644 billion tons [7, 8], ranking second in the world. However, due to complex geological conditions and the late start of exploration and research, the exploitation of shale oil in China is still at a critical stage of exploration [9–11]. In recent years, with growing national attention to unconventional oil and gas resources and continuous progress in exploration and development technology, China's shale oil exploration and development has made phased progress, laying a solid foundation for large-scale exploitation in the future [12–14].

Looking ahead, with the continuous growth of global energy demand and the increasing shortage of conventional oil resources, increasing attention is being paid to unconventional oil and gas resources [15]. With the continuous progress and development of exploration and development technologies and the gradual reduction of costs, shale oil production will continue to expand, and its position in the global energy market will continue to increase [16]. At the same time, with growing global emphasis on low-carbon development and increasingly strict environmental regulations, the environmental problems associated with shale oil processing and utilization will be more effectively addressed [17]. As a result, shale oil has promising prospects and is expected to become an important force in the global energy sector [18].

In this regard, this paper focuses on the current research status of the shale pyrolysis process and proposes the future development directions for of oil shale [19]. Based on the current research trends in electric heating for in situ exploitation of oil shale, this paper outlines future development directions and offers specific guidance and reference value for further research on in situ conversion technologies for oil shale.

2. Oil shale in situ conversion technologies

2.1. Classification of in situ conversion technologies

Table 1. Features of in situ conversion technologies

Type of technology	Heating time	Environmental impact	Economic cost
ICP	2–4 years	Medium	Electricity cost Facility
Electrofrac	3–5 years	Medium	Electricity cost Facility
GFC	None	Medium	Electricity cost Facility
HVF	0.5–3 years	Slight	Electricity cost
Fluid heating	2–3 years	Slight	Heating cost
Radiation heating	None	Slight	Radiation energy

In situ conversion technology is derived from the three modes of heat transfer. A representative in situ conversion technology based on heat conduction is electric heating. In situ conversion using electric heating involves heating the reservoir through direct contact with an electric heater, transferring heat via conduction [20]. A representative technology based on thermal convection is fluid heating in situ conversion. This method uses heat convection to transfer thermal fluids from the ground into the reservoir. Thermal radiation, on the other hand, refers to the emission of electromagnetic waves from a heated object. The representative in situ conversion technology based on thermal radiation is the radiant heater, which heats the reservoir through the heat emitted by the radiant source [21, 22]. In addition, there is a more specialized combustion heating technology, which combines both radiation and conduction to achieve effective heat transfer [22]. As shown in Table 1, the environmental impact of all in situ conversion technologies is mainly reflected in their effects on groundwater, the broader underground environment, and greenhouse gas emissions.

2.2. In situ conversion technology using electric heating

In situ conversion technology using electric heating refers to placing an electric heater in the reservoir and using the thermal effect of electric current to heat the reservoir. Compared with other heating methods, electric conduction heating is more versatile, has higher heat transfer efficiency, and is more environmentally friendly [23]. Therefore, both international and domestic mining technologies are being improved in this direction. For example, oil companies such as Shell and ExxonMobil have launched ICP and Electrofrac technologies based on conduction heating [24].

2.2.1. ICP in situ conversion technology

Shell proposed the in situ conversion process (ICP) technology, which first involves drilling freezing wells into the ground and injecting freezing liquid to form an underground freezing wall surrounded by the heated oil shale reservoir (Fig. 1). This freezing wall isolates the reservoir from energy exchange with the surrounding environment, thereby improving heating efficiency and protecting groundwater and the surrounding environment. The environmental protection benefits of ICP technology have been evident since its introduction in China in 2002. With more than 20 years of application and through experimental and practical engineering work, most of the technological and equipment-related challenges have been resolved. In particular, ICP technology shows high adaptability for low-maturity shale oil, such as that found in the Ordos Basin [25–27].

Meanwhile, ICP technology is one of the few in situ conversion technologies that has undergone field trials. The test site is located in the Piceance

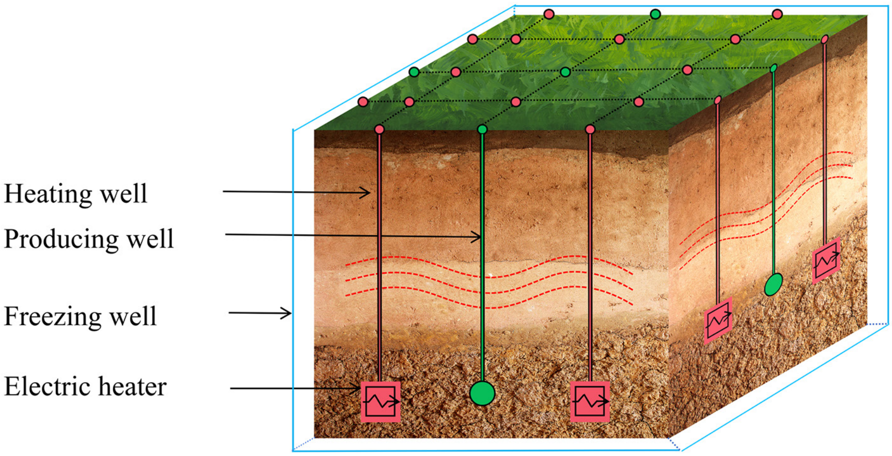

Fig. 1. Schematic diagram of ICP in situ conversion technology.

basin in northwest Colorado, USA, where approximately 150–200 freezing wells were drilled in preparation. These wells were fully protected by freezing throughout the process. After the cold wells were completed, cold fluid was injected, initially freezing the injection area, and then, once fluid circulation began, freezing the entire operational area to create a frozen wall barrier. After the freezing process was completed, a pattern of wells was drilled at intervals of 20–40 feet, with a total of 70–100 wells. An upright, bare electrode heater was installed in each well. The bare electrode heater consisted of a 410SS alloy electrode rod with a diameter of 1.5 inches, offering high resistance to both temperature and corrosion.

As shown in Fig. 1, three electrode heaters are arranged in a three-phase Y-type circuit, forming a "three-in-one unit." Since each unit has a separate three-phase transformer, this design significantly reduces the initial investment costs, saving billions of dollars compared to conventional isolated single-phase transformer setups.

Although the heating performance and efficiency of electric in situ conversion technology are already considered high, research shows that it can be further optimized through improvements in the heating method. In the ICP field tests, the spacing between heating wells was 10–20 feet, which led to uneven reservoir heating. A potential solution could involve designing a heat-conductive material with minimal thermal loss within a certain distance, which could be used to encase the reservoir and promote more uniform heat effects.

Although ICP technology has significant advantages such as high thermal conductivity efficiency and suitability for low-permeability formations, its shortcomings are also obvious. Notably, it requires high energy input, and its thermal conductivity is greatly affected by the conductivity of the formation itself. These factors mean under certain geological conditions, the performance of ICP technology is not that good.

2.2.2. Electrofrac technology

ExxonMobil's Electrofrac process uses an innovative hydraulic fracturing technology specifically designed for oil shale treatment. The technology first employs horizontal well technology to create cracks in oil shale, which are then injected with proppants, composed mainly of calcined petroleum coke. These proppants not only keep the cracks open but also form an electric heating body. When current passes through the conductive proppants, heat is efficiently transmitted to the oil shale (Fig. 2).

Unlike ICP technology, which uses vertical electrode heaters, Electrofrac employs a flat fracture heater with a large heating area. This significantly increases the contact area with the reservoir, resulting in higher heating efficiency. As the kerogen in the oil shale is heated, it converts into oil and natural gas, which can then be extracted using conventional mining methods [27].

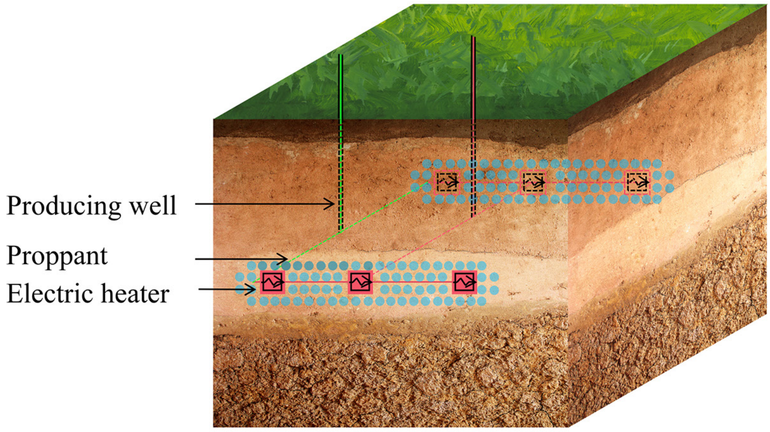

Fig. 2. Schematic diagram of Electrofrac technology.

Similar to Shell's ICP technology, the Electrofrac process transfers heat to the shale through thermal diffusion. However, a significant advantage of the Electrofrac technology is its use of a flat fracture heater with a larger surface area. This allows for fewer heaters to be used while still providing sufficient heat to the reservoir, thus increasing heating efficiency. In addition, the use of flat heaters reduces surface disturbance, making the process more environmentally friendly and efficient than ICP technology. The core of Electrofrac technology lies in its specialized well completion process and the conductive agent used in the cracks. Combining these factors helps achieve the goal of efficient heating. This technology can increase the permeability of the formation by using high voltage, but it involves high upfront investment costs due to equipment construction and formation modification. Moreover, the high electricity costs in the later stage remains a problem that cannot be underestimated.

2.2.3. GFC in situ conversion technology

Independent Energy Partners (IEP) has proposed an innovative underground fuel cell heating technology, known as geothermic fuel cells process (GFC), which cleverly uses the heat generated by solid oxide fuel cells to generate electricity and heat the oil shale.

In practice, these fuel cells can be deployed directly within the shafts of oil shale reservoirs. Through the efficient energy conversion of the fuel cells, only a small amount of electricity is released, while a large amount of heat is transferred to the surrounding area, raising the temperature of the oil shale to 400 °C (Fig. 3). At this temperature, extensive pyrolysis occurs, producing

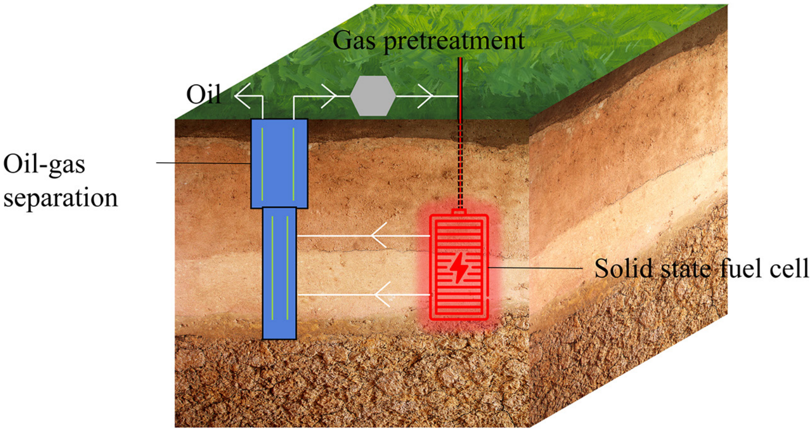

Fig. 3. Schematic diagram of GFC in situ conversion technology.

valuable resources such as shale oil, hydrocarbon gas, and carbon. Compared with other in situ mining technologies, GFC demonstrates greater cleanliness and sustainability. In particular, the technology does not rely on traditional, potentially polluting coal-fired power sources, but instead achieves efficient use of energy.

According to IEP estimates, GFC technology can generate 24 kilowatt-hours of electricity per ton of oil shale, while producing minimal toxic waste. Furthermore, in a simulation study conducted by Anyenya [28], the technology demonstrated up to 43% heating efficiency and cogeneration efficiency of 79%, further showcasing its excellent performance and potential.

In general, GFC also offers the advantage of more uniform heating, particularly in deeper reservoirs. However, the high intensity and elevated temperatures during heating result in frequent battery charging and discharging, which considerably reduces battery lifespan. Therefore, the equipment construction in the early stage and the battery loss and energy supply in the later stage will lead to high usage costs.

2.2.4. HVF in situ conversion technology

High-voltage power frequency electric heating (HVF) technology is an innovative mining method developed by Jilin University in China. This technology is cleverly divided into three main stages. First, the technical team carefully arranges electrodes within the shale layer and applies high voltage. Under certain conditions, oil shale transitions from an insulator to a conductor. When high voltage is applied to the transformed oil shale, irreversible breakdown of solids occurs, resulting in the formation of conductive channels and plasma through melting or combustion – a process known as insulation failure (Fig. 4). It has been found that when the electric field intensity is maintained in the range of 100–180 V/cm, the higher the intensity, the more likely thermal breakdown is to occur, thus reducing overall energy consumption. With large electrode spacing, hydraulic fracturing is also employed. This not only effectively creates cracks within oil shale but also significantly reduces the breakdown voltage required to suppress water seepage cracks [29].

Subsequently, power frequency is applied, and the surrounding oil shale is heated using the inner surface of created by the prior breakdown and the conductive channels. Heat is transferred within the oil shale through the plasma in the conductive channels, and after a period of time, some of the oil shale reaches the temperature required for pyrolysis and begins to produce shale oil and combustible gas. As the pyrolysis reaction progresses, the kerogen undergoes fragmentation, resulting in increased porosity of the oil shale. The evaporated shale oil and combustible gas produced during this process continue to heat the oil shale through convection, facilitating the flow of heat-conducting fluids and the expansion of conductive channels. Eventually, these products are expelled through the production well [30].

This new field mining method offers significant advantages over ICP technology and hydraulic fracturing technology in terms of environmental protection. It does not involve the injection of materials into the ground that could affect the environment or groundwater. In addition, the underground equipment required by this technology is relatively simple, and the geological requirements of the oil shale layer are minimal, making it more adaptable and

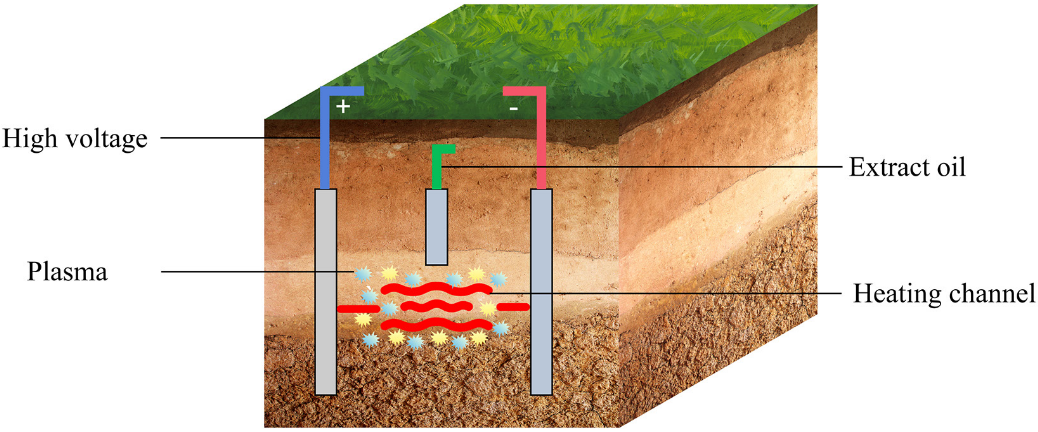

Fig. 4. Schematic diagram of HVF in situ conversion technology.

feasible in practical applications. Research shows that the advantages of GFC and HVF technologies are obvious. They not only improve heating efficiency based on mature applications but also solve known deficiencies. For example, HVF technology gradually overcomes the high energy consumption associated with Electrofrac by improving both heating efficiency and cogeneration efficiency.

2.3. Fluid heating technology

Fluid heating technology involves injecting high-temperature fluids into shale oil reservoirs – such as water, steam, carbon dioxide (CO_2), and other media – and transferring heat to the shale oil layer through fluid convection and conduction. Although fluid heating technology has high heating efficiency, high energy utilization, and low environmental impact, it presents challenges in temperature control. In some cases, it is difficult to maintain a specific temperature range. Representative technologies include Chevron's CRUSH technology and U.S. Shale's CCR technology, which have already been tested, as well as Taiyuan University of Technology's MTI technology and supercritical water heating technology, which are still in the theoretical stage.

Wang et al. [31] used CO_2 and steam as fluid media, conducting experiments by injecting them separately and in combination. The experimental temperature was set to 100–550 °C and pressure to 6, 10, and 15 MPa, with the heating time set to four hours. By comparing the results of different conditions, they found that the pore structure of shale would undergo significant changes at around 400 °C, and the content of organic matter would decrease markedly at around 400–500 °C. They concluded that combining CO_2 and high-temperature steam as fluid media improves the yield and efficiency of shale oil extraction.

Supercritical water refers to water whose temperature and pressure exceed its critical point (critical temperature: 374.3 °C, critical pressure: 22.1 MPa). Under these conditions, the properties of water change significantly, exhibiting dispersibility of gas and solubility of liquid, while also having high reactivity. These unique properties enable supercritical water to penetrate shale pores more effectively, strengthening the heat transfer effect and improving the heating speed and spread range. Therefore, adjusting the temperature and pressure of critical water helps regulate oil shale production.

Sun et al. [32] set the temperature to 350 °C, the pressure to 16–18 MPa, and the heating time to 0–100 hours to keep the water in a subcritical state. They found that the maximum shale oil extraction rate from the test samples from different regions was 18.1%, which occurred about 70 hours after heating. With the decrease of heating time, the extraction rate also gradually decreases.

Xie et al. [33] set the temperature to 380, 400, and 430 °C, maintaining water in a supercritical state throughout the whole process, with the pressure set to 25 MPa and the heating time to 120–720 minutes. By analyzing the content of gas components, the reaction kinetics model was established, and the conclusions were similar: CO_2 and other gas components decreased with the increase of heating time and temperature, proving the effectiveness of that method on the production of light oil. Supercritical water enabled shale to directly generate a large amount of stable saturated hydrocarbons, with the reaction path favoring saturated hydrocarbons rather than asphalt intermediates.

Lu et al. [34] set the temperature to 300, 360, 450, and 500 °C, the pressure to 10, 20, and 27 MPa, the heating time to 48 hours, and the heating rate to 1 °C/min. The experiment included both subcritical and supercritical water conditions. It was found that the oil discharge rates at 360 and 450 °C were 5.44 and 14.33 times higher, respectively, than those under anhydrous pyrolysis. In the process of subcritical and supercritical water pyrolysis, the yields of hydrogen, carbon monoxide, and hydrocarbons were significantly higher than those of anhydrous pyrolysis, due to the synergistic effect of chemical hydrogen supply and acid-base catalysis. This boost was mainly attributed to the occurrence of the water-gas conversion reaction, which promotes the production of hydrogen and carbon monoxide. In addition, subcritical and supercritical water enhanced the side-chain cleavage in aliphatic and aromatic hydrocarbons in kerogen, further increasing the production of hydrocarbon gases.

Fluid heating offers high energy utilization efficiency and heating effect, but as the experiments above show, the combined use of CO_2 and high-temperature steam as fluid media yields better results than using them separately. Therefore, when other substances – such as subcritical or supercritical water – are used as reservoir heating media, the addition of appropriate additives or reactants may help the entire fluid medium reach the target heating temperature faster while consuming less energy.

In summary, taking supercritical water in situ conversion technology as an example, this method not only features high thermal efficiency but also stands out for its ability to use oilfield waste liquid or water as heating carriers. This reduces both energy consumption and the environmental pollution risks associated with other methods. However, as a high-temperature and high-pressure in situ conversion method, it requires a continuous energy supply to maintain the necessary. Therefore, the energy consumption involved must be carefully considered.

2.4. Radiation heating technology

In situ conversion of oil shale through radiation heating is a technology that uses radiation energy to directly transfer heat to shale, promoting the pyrolysis and cracking of organic matter to generate into liquid petroleum products. It is mainly divided into microwave heating and radio frequency heating.

Microwave heating involves placing a microwave emitter inside the reservoir to heat it to the pyrolysis temperature of kerogen. This method was proposed by Raytheon in the United States, which introduced the concept of placing radio frequency transmitters in the reservoir to heat the bottom layer using radio frequency energy. This technology has the advantages of high efficiency and environmental friendliness, but it also has its disadvantages – namely heavy reliance on electric energy and high costs. Representative technologies include the electromagnetic wave radiation heating system developed by the Chinese Academy of Sciences and the radio frequency heating technology (RF/CF) pioneered by Raytheon.

As a new in situ conversion technology using electromagnetic waves as the heat transfer medium, thermal radiation heating offers a significant advantage: it does not require drastic transformation of the strata to meet the geological conditions, which improves its cost-effectiveness and offers high flexibility. However, because electromagnetic waves penetrate complex formations, the associated power consumption is also quite high, which is why the overall economic efficiency of this technology remains significantly lower than that of fluid heating technology.

2.5. Combustion heating technology

Combustion heating in situ conversion technology refers to the conversion of target substances by injecting a high-temperature heat source into the target reservoir to trigger combustion reactions. It has the characteristics of high heating efficiency and high energy utilization rate, and can further reduce the pyrolysis temperature of kerogen by artificial means such as adding water and catalyst, but the disadvantage is that its control technology is relatively complicated. A representative technology is the TS-A technology developed by Jilin University. The advantage of combustion heating is that, in the combustion process, the underground low-calorific-value exhaust gas can be used to participate in the entire combustion heating process, reducing external energy consumption. However, the harmful gases generated during the combustion process are difficult to ignore, and it is known that two dry distillation demonstrations conducted by the U.S. Bureau of Land Management failed to pass environmental requirements and were halted.

2.6. Summary

In summary, each in situ conversion technology for oil shale has its own technical characteristics and inherent advantages. However, since these technologies are ultimately intended for commercial application, if they cannot generate corresponding commercial value, then no matter how advanced they are, they will remain underwhelming. Therefore, Shell’s ICP technology is still the most suitable for the current production environment. It has undergone multiple rounds of experimental demonstration and is more mature than other original conversion technologies. If appropriate methods for formation transformation and energy supply can be introduced on the basis of this technology, its level of maturity could be further improved.

Looking ahead, Jilin University’s HVF technology may offer a better solution, as compared with the currently most mature ICP technology and the similarly principled Electrofrac technology, its dependence on electric energy is lower. In the case of China, for example, various eastern cities still face electricity consumption restrictions, which could be a serious obstacle for certain industrial operations. At the same time, HVF technology includes a mechanism for independent stratum transformation, making it adaptable to a wider range of production environments. Compared with other in situ conversion technologies, its environmental impact is relatively small, as it does not require the injection of substances such as hot fluids into the ground. This also suggests that the early-stage equipment costs of this technology could be further reduced.

3. Pyrolysis of oil shale

3.1. Pyrolysis mechanism of oil shale

The pyrolysis of oil shale is a highly complex physical and chemical process. From a physical perspective, Brownian motion plays a vital role in accelerating the thermal decomposition of oil shale. These reactions can be broadly divided into two categories [35, 36].

The first category is the decomposition reactions, which are mainly characterized by the breaking of covalent bonds. Specifically, it includes the depolymerization of kerogen, cleavage of various carbon bonds, removal of oxygen-containing functional groups, and dehydrogenation reactions. These decomposition reactions are entropy-increasing and endothermic, and they mainly produce oil and gas with relatively low molecular weight [37].

The second category involves condensation reactions, characterized by the re-formation of covalent bonds. During the pyrolysis process of oil shale, small and medium-sized molecules undergo reactions such as overlap, addition, cyclization, aromatization, thickening cyclization, and condensation. These polycondensation reactions often lead to a reduction in entropy, and most of them are exothermic [38].

The exploration of the kinetic characteristics of the oil shale pyrolysis process helps to reveal the essential principles governing the conversion process, enabling more efficient control and optimization [39]. This control is reflected not only in improving the overall conversion efficiency of oil shale – ensuring that more raw material is converted into valuable products – but also in enhancing product quality, striving to obtain purer and more efficient energy outputs [40]. Moreover, a deeper understanding of energy conversion and material flow during oil shale pyrolysis allows for the design of more efficient, low-energy distillation processes. This helps reduce unnecessary energy loss and the emission of harmful substances, thereby supporting the environmentally sustainable use of oil shale resources [41].

By using advanced technologies such as thermogravimetric analysis, iso-conversion method, and differential scanning calorimetry, we can obtain a large number of kinetic parameters and characteristics data about the pyrolysis and combustion of oil shale.

Research into the reaction dynamics of oil shale is not only directly related to efficient resource utilization and improved economic value, but also plays an irreplaceable role in promoting the sustainable development of the energy industry and advancing energy diversification strategies. It provides an important way and a powerful tool for exploring the potential of oil shale resources and addressing global energy shortages [42].

The focus of kinetic research is to obtain kinetic parameters. According to previous studies, the methods for determining these parameters include the curve method and the model method. The core of the curve method lies in experimental measurement – that is, recording the concentration or properties of each substance in the reaction system at different time points, and plotting a kinetic curve based on these data. This curve visually reflects the changes in substance concentrations over time, providing an intuitive perspective on the dynamic behavior of the reaction system. Furthermore, researchers can use these curves to draw tangents at specific time points to calculate the instantaneous reaction rate, thereby gaining a deeper understanding of the reaction's kinetic characteristics. The model method, by contrast, is based on the known reaction mechanism and involves establishing a corresponding kinetic model to accurately describe the system's kinetic behavior.

At present, the Friedman method is widely used. Its essence lies in analyzing thermogravimetric data to reduce experimental errors. In this method, the conversion rate (α) is assumed constant. To determine the activation energy (E_a), reaction rate constants (k) are measured at multiple temperatures. The natural logarithm of k (ln k) is plotted against the reciprocal of temperature (1/T), yielding a linear relationship. The slope of this line ($-E_a/R$) is then used to calculate the activation energy [43]. As temperature increases, the conversion rate of different particle sizes also increases gradually. In experiments conducted by Zhu et al. [44], it was observed that the conversion rate changed little between 150–350 °C and 620–800 °C. That is, the conversion rate steadily increased before reaching 520 °C, but changed sharply between 520 °C and 650 °C. Once the temperature reached 750 °C, the conversion rate returned to a steady state [45].

By analyzing the combustion reaction of oil shale using the iso-conversion method, it has been found that the activation energy gradually decreases during the initial stage of combustion, but rises rapidly in the later stages [46]. This phenomenon shows that oil shale exhibits high reactivity at the beginning of combustion, allowing for rapid ignition and sustained combustion; however, complete burnout becomes relatively more difficult as the process continues [47]. In addition, the thermal decomposition of oil shale under aerobic conditions can be approximated as a first-order reaction, with a reaction order close to 1. The standard error of the straight line obtained through data fitting is very small (less than 0.01), indicating a good fit [48]. Furthermore, it has been found that the apparent activation energy of oil shale redistillation in an aerobic environment is lower than that in a pure nitrogen atmosphere. This finding provides an important rationale for the development of aerobic autothermal redistillation technologies, suggesting that redistillation may be more efficient under aerobic conditions [49].

Building a reasonable chemical reaction model is also a key component in the study of oil shale pyrolysis. Moreover, different heating methods involve different mechanisms of heat and mass transfer, which requires the construction of tailored models for different reaction processes [50]. It is also important to note that the hydrocarbons generated during oil shale pyrolysis originate not only from kerogen but also from soluble organic matter. In short, oil shale pyrolysis is a complex reaction process, and only by knowing the specific generation and decomposition pathways of each target product can a correct reaction model be constructed [51].

Research on hydrocarbon generation dynamics began in the 1920s, initially focusing on the redistillation of coal and oil shale. By the 1960s, research in this field was extended to include kerogen pyrolysis, leading to significant advances in the field. Over the course of this development, scientists proposed a variety of hydrocarbon kinetic models, including the total package reaction model, continuous first-order reaction model, parallel first-order reaction model, and tandem reaction model [52]. These models are widely used to predict and calculate hydrocarbon generation rates and yields in sedimentary basins, and have become crucial tools in basin simulation and hydrocarbon generation evaluation.

Since the 1970s, with the deepening of research, more complex kinetic models of oil shale pyrolysis have been developed, which include multiple chemical reaction equations to more accurately describe and predict the yield of different target products over time. Such models are widely used in the field of in situ heating modification of oil shale, including the BB model [53] and the W model [54], two of the more common approaches.

Using the heterogeneous reaction characteristics of oil shale pyrolysis, the conversion rate α is used as an index of the reaction degree to replace product concentration in a homogeneous system. Based on the law of conservation of mass and Arrhenius equation, the kinetic equation of the pyrolysis reaction is derived as $d\alpha/dt = k(T)f(\alpha)$, where $k(T)$ is the temperature-dependent reaction rate constant and $f(\alpha)$ is the reaction order function [55]. According to the Arrhenius equation, the reaction rate constant $k(T)$ is expressed as $A \exp(-E/RT)$, where A is the frequency factor, E is the activation energy, and R is the gas constant. Combined with a given heating rate β (dT/dt), the kinetic equation is further transformed into the form of $d\alpha/f(\alpha) = (A/\beta) \exp(-E/RT)\, dT$. By testing different reaction orders n (0.5, 1.0, 1.5, 2.0), it was found that the best linear fit occurs at $n = 1$ [56].

Finally, key kinetic parameters such as activation energy E_a and frequency factor A were successfully determined, leading to the construction of the kinetic model for Jimusar oil shale [57]. According to the reaction model, the pyrolysis process of Jimusar oil shale can be divided into three stages:

- In the low-temperature stage (room temperature to 350 °C), a large amount of water release occurs and the reaction rate is relatively slow;
- In the medium-temperature stage (350–550 °C), the reaction rate increases rapidly;
- In the high-temperature stage (550–900 °C), the pyrolysis reaction enters a stable phase where the reaction rate proceeds at a fast rate but steadily [58].

Based on experimental data, the kinetic model of Jimusar oil shale was successfully established and confirmed to belong to the first-order reaction type. Using the Arrhenius equation, the apparent activation energy E of the pyrolysis reaction was calculated to range between 148 and 222 kJ/mol, while the frequency factor A ranged between 8.67×10^{11} and 1.67×10^{17}. These kinetic parameters provide an important scientific basis for a deeper understanding of the pyrolysis properties of Jimusar oil shale [59].

The most critical factors affecting oil shale pyrolysis are the pyrolysis efficiency of kerogen and the controllability of the pyrolysis process for engineering purposes. Studies also show that pyrolysis efficiency varies under different response atmospheres [60]. For example, the appropriate addition of hydrogen and CO_2 [61] can promote pyrolysis, while also reducing heat generation and the production of semi-coke. Overall, the pyrolysis process of kerogen is a complex reaction sequence that varies with temperature, and the dominant reaction paths and product distribution differ across temperature intervals [62].

3.2. Influencing factors of oil shale pyrolysis process

3.2.1. Heating temperature

Temperature has an important influence on oil shale pyrolysis, as it determines the amount, composition, and rate of pyrolysis products. In practice, it is necessary to choose an appropriate pyrolysis temperature range based on the specific situation to obtain the best pyrolysis effect. The influence of temperature on the pyrolysis process can be roughly divided into the following stages.

Approximately 180 °C is considered the threshold temperature for oil shale pyrolysis – that is, the point at which oil shale begins to convert into a large amount of petroleum hydrocarbons. Below this temperature, the primary process is the evaporation of water within the oil shale. Li et al. [63] found through experiments that oil shale undergoes heating, drying, and dehydration over a certain temperature range. This stage is mainly characterized by the reduction of internal moisture. From about 200 °C onward, the main products released are heavy oil, a small amount of light oil, and some hydrocarbon gases, due to the thermal decomposition of salts such as $NaHCO_3$.

When the temperature reaches about 350 °C, Huang et al. [64] found through experiments that the yield of heavy oil reaches its highest value. In order to explore the cause of this phenomenon, Jiang et al. [65] conducted further experiments and concluded that this is because at this temperature, the main deoxygenation reactions of alkanes take place, and carbon-hydrogen bonds begin to break. As a result, hydrogen content begins to rise, while the concentrations of other gases and components remain low. Beyond about 350 °C, the reaction becomes more intense, characterized by a significant increase in light oil and gas composition, which is attributed to the continuous pyrolysis of heavy oil [65].

Through observation, Li et al. [63] also found that in this stage, inorganic matter and fixed carbon begin to combine and gradually produce semi-coke. As a proppant and a key substance in certain in situ conversion technologies, the emergence of semi-coke indicates that the main oil production phase in these technologies begins after 350 °C. After the temperature exceeds about 400 °C, the production of light oil reaches its peak. In addition, Huang et al. [64] measured a steady increase in hydrocarbon gas production, indicating that hydrocarbon gas becomes the main product in this temperature range.

When the temperature rises above 500 °C, heavy oil undergoes complete pyrolysis to produce shale oil and gas. Based on this, Huang et al. [64] speculated that the CO_2 detected during earlier combustion stages likely originated from residual carbon components, which continue to decompose at higher temperatures. In view of the continuous retention of CO_2, Jiang et al. [66] suggested that this is mainly due to the condensation of free radicals and the decomposition and reorganization of carboxyl and aromatic functional groups. Within the temperature range of about 500–700 °C, further decomposition of heavy oil occurs, enabling the measurement of reactants [67]. The reaction characteristics change, leading to not only the formation of hot asphalt [68] and other high-density substances but also the continued production of shale oil and shale gas. The content of hydrocarbon gases continues to rise. At the same time, the concentration of hydrogen gas increases markedly due to the dehydrogenation reaction of aromatic hydrocarbons and the cracking of macromolecular alkanes. Moreover, small amounts of CO_2 are also formed [69].

When the high-temperature range (about 700 °C) is reached, the pyrolysis reactions are basically complete. While the overall composition of the gaseous products is similar to that of the previous stage, the contents of the components change. The further decomposition of heavy oil components causes a significant increase in other hydrocarbon gases, which become the main products. At the same time, the proportion of hydrogen gas also increases, likely due to further dehydrogenation of aromatic hydrocarbons [70] and continued pyrolysis of macromolecular alkanes. Moreover, some low-bond energy [71] benzene ring bonds break at high temperatures and combine with oxygen atoms, producing a small amount of CO_2.

3.2.2. Heating time

Oil shale is a special type of sedimentary rock that contains organic material that can be converted into oil or gas. Heating time (also referred to as constant temperature time) is a crucial operational parameter in the pyrolysis of oil shale, as it directly influences the production of hydrocarbons (such as oil or gas) and other by-products [72]. Research shows that with the extension of the heating time, the small pores within the oil shale gradually expand into relatively large pores, and this change of the pore structure has a positive effect on improving the physical properties of oil shale [73].

Changes in the physical structure not only influence the shale's physical properties but may also profoundly affect its chemical properties, thereby affecting both the yield and composition of the final product. Therefore, the precise control of heating time and heating rate is critical to optimizing the pyrolysis process. This enables to improve the production and quality of hydrocarbon products, while also contributing to greater economic efficiency and environmental friendliness in oil shale exploitation and processing [74].

In a study by Zhu et al. [75], heating durations of about 10, 20, and 30 minutes were tested. Results showed that with increased heating time, the number of pores and cracks in the oil shale samples rose, leading to increased porosity. In contrast, Song et al. [76] conducted experiments with heating times ranging from about 3 to 4 hours and found that such extended durations could significantly improve the efficiency of low-temperature distillation.

3.2.3. Pressure

Pressure has a significant blocking effect on the pyrolysis process, which is reflected in several key aspects. First, as pressure increases, the pyrolysis of oil shale is clearly impeded, resulting in a significant decrease in the yield of the pyrolysis products. Specifically, the yield of pyrolysis oil is reduced, while the total yield of oil and gas also shows a downward trend, which indicates that under high-pressure conditions, the organic matter in oil shale is not fully converted into the desired oil and gas products [77].

Second, pressure has an important influence on the kinetics of oil shale pyrolysis. Analysis of the total package first-order reaction kinetic model reveals that the introduction of pressure changes the kinetic parameters of the pyrolysis reaction. These changes affect both the rate and the reaction path of the whole pyrolysis process. This effect facilitates a deeper understanding of the role pressure plays in oil shale pyrolysis [78].

In addition, pressure significantly affects the distribution of oil shale pyrolysis products. Under high pressure, not only does the yield of oil and gas decrease, but the amount of residual carbon in the semi-coke increases significantly. This may be due to high pressure inhibiting the full decomposition of organic matter in oil shale, resulting in more carbon being retained as residual material instead of being converted into valuable oil and gas products [79].

Therefore, pressure is an important parameter that cannot be ignored in oil shale pyrolysis. It has a significant effect on product yield, distribution, and reaction kinetics. To optimize the pyrolysis process and improve resource utilization, the influence of pressure factors must be fully considered in order to enable a more efficient and economical oil shale pyrolysis process [80].

Baruah and Tiwari [81] found that high pressure has significant effects on oil shale and its product yield. Specifically, under high pressure, the volatilization temperature of hydrocarbons in oil shale increases, and the average apparent activation energy rises with increasing pressure – although it decreases again at very high pressure, such as 300 MPa. Moreover, as pyrolysis pressure increases, the pyrolysis process in oil shale follows different reaction models: from the D3 diffusion model with lower conversion rates between 0.05 and 0.5 and the F2 secondary reaction model between 0.4 and 0.6 to the A3 Avrami–Erofeev model with higher conversion rates between 0.62 and 0.95. These changes directly affect product yields: oil production tends to decrease due to enhanced pyrolysis and secondary reactions, while gas production increases accordingly [82]. In particular, the concentration of C_2–C_4 gases increases significantly under high pressure. Further analysis showed that at 300 MPa, the proportion of light (gasoline) and intermediate (kerosene) fractions increases significantly. At the same time, the concentrations of aldehydes, ketones, carboxylic acids, and aromatic compounds decrease markedly, while the concentration of alkanes increases [83]. In conclusion, high pressure has a complex and far-reaching effect on the non-isothermal pyrolysis process and the resulting product distribution of oil shale [84].

3.2.4. Heating rate

Different heating rates can have a significant impact on the pyrolysis process of oil shale. A slower heating rate facilitates the development of organic pores within the oil shale, which may increase the space for hydrocarbon formation; a faster warming rate is more likely to promote the formation of microcracks, which may act as a channel in the pyrolysis process and promote the flow of gases and liquids.

According to Li et al. [85], experiments have shown that as the heating rate increases, a more significant thermal lag appears in the pyrolysis process of oil shale. At the same time, the kinetic characteristics of the chemical reactions involved lead to an increase in the initial pyrolysis temperature, the temperature corresponding to the maximum mass loss, and the final pyrolysis temperature. This is shown by the peak on the differential thermogravimetric analysis (DTG) curve moving toward higher temperatures. The pyrolysis reaction of oil shale is considered to be a first-order reaction, meaning that the reaction rate is proportional to the primary square of the reactant concentration. Although the heating rate has less influence on the activation energy of the pyrolysis reaction, there is a mutually compensating effect between the activation energy and the frequency factor, with the two parameters showing a numerical correlation [86].

3.3. Pore evolution law

During the pyrolysis of oil shale, the pore structure undergoes significant changes, which can influence both the pyrolysis behavior and the properties of the resulting products. Specifically, the evolution of pores, including their formation, expansion, and enhanced connectivity, not only promotes the smooth flow of fluids within the oil shale but also accelerates the generation and effective release of pyrolysis products (Fig. 5).

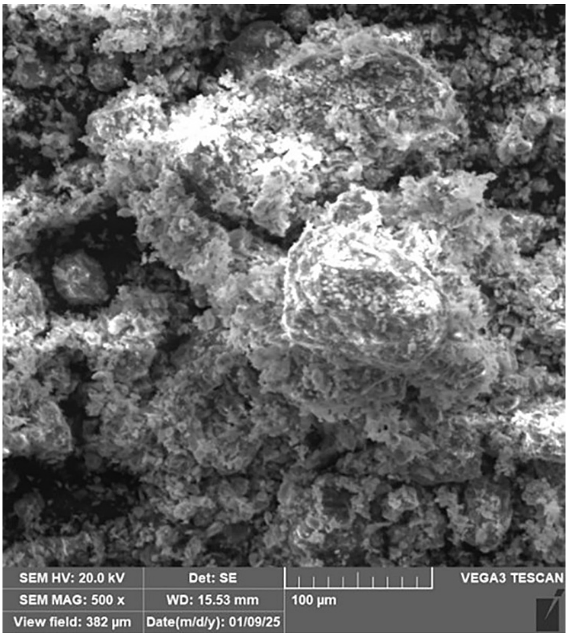

Fig. 5. Pore evolution law.

The evolution of pores also affects the kinetic and thermodynamic behavior of oil shale pyrolysis. Under high-pressure conditions, oil and gas products may undergo secondary reactions within the pores, leading to coke formation and pore blockage, thus increasing the activation energy of the pyrolysis reaction and making the process more difficult. In addition, changes in porosity can influence the pyrolysis characteristics of oil shale, thereby affecting both pyrolysis efficiency and product properties [87].

Different heating modes lead to changes in the fracture structure and seepage paths of oil shale pores. In convective heating, superheated water vapor can react with the semi-coke around the pores to generate higher-quality liquid pyrolysis products. These products can then be carried out of the oil shale by airflow, further increasing shale oil yield and resource recovery.

Overall, the pore evolution law plays a vital role in the pyrolysis process of oil shale. It provides an important scientific basis for optimizing oil shale development and utilization by affecting fluid flow, promoting product formation and release, changing reaction kinetics and thermodynamics, and influencing the evolution of seepage paths [88].

Thus, the evolution of pores within oil shale is strongly correlated with the temperature at any given moment. Kang et al. [89] conducted an experiment using φ 0.82 mm × 7 mm oil shale samples and found that at about 300 °C, water present within the oil shale and between the clay mineral layers underwent thermal evaporation. However, they did not observe any changes in the internal structure of the samples at this temperature – that is, parameters such as surface area, porosity, inner diameter, and pore structure remained largely unchanged. This indicates that at around 300 °C, oil shale remains structurally stable.

When the heating starts at about 300 °C, the porosity, pore number, and pore surface area increase sharply until about 500 °C. This is attributed to the transition of organic matter from a solid, cheese-like state at ~300 °C to a phase where chemical bonds begin to break, producing shale oil and gas. The rapid volumetric expansion of these pyrolysis products generates internal pressure that forcibly expels them, significantly enhancing porosity. The continuous escape of shale oil and gas further contributes to pore expansion. Within this temperature range the changes in porosity are largely driven by chemical reactions. Near 500 °C, as the chemical reactions approach completion and reactants are nearly exhausted, little to no additional product is available to further expand the pore system. Instead, existing pores extend along their original development pathways without the formation of new ones [90]. Additionally, between about 500 and 600 °C, a small amount of residual carbon within the oil shale undergoes carbonization, generating considerable crystalline structures that can block pores and inhibit further increases in porosity [91].

Oil shale is mainly composed of aqueous clay minerals and a small amount of fixed carbon (usually about 6%). When the temperature is further increased to 500–600 °C, part of the crystalline water in clay minerals is lost, and the fixed carbon undergoes pronounced carbonization and coking. Together, these processes lead to the loosening and disintegration of the matrix rock particles and the collapse of some pore walls, which blocks part of the pores and reduces their total volume. This is the main reason for the slight decline in pore structure parameters during this stage.

3.4. Inorganic mineral evolution

Inorganic minerals, as important components in oil shale, affect the pyrolysis reaction by either promoting or inhibiting catalytic pyrolysis reactions. Understanding their role significant for exploring the pyrolysis process of oil shale. Jin et al. [92], using hydrochloric acid-d1 (HCl-D) as the experimental object, proved that carbonates facilitate kerogen pyrolysis, whereas silicates inhibits kerogen pyrolysis. Moreover, although the self-heating peak temperature of HCl-D is lower than that of nitric acid (HNO_3), its oil yield efficiency is higher. This is because the lower peak temperature in HCl samples is still sufficient to initiate full kerogen cracking, and the reduced temperature also implies diminished air reactivity during pyrolysis [93].

Notably, the self-heating retorting (SHR) process used in this study differs from all previously reported distillation methods in that the heat is generated internally from the oil shale itself. While acid treatment has the above effects on the SHR process, it does not change the composition of the generated shale oil. After acid treatment, the composition of shale oil is still mainly hydrocarbons, just like in the non-demineralized (non-D) samples. However, the boiling point distribution of the shale oil generated by different processes varies. The shale oil generated by hydrofluoric acid (HF-D) has the highest light component content, while the shale oil generated by HCl-D has the highest recombination content. This is due to the higher autothermal temperature of HF-D, which allows for recombination splitting and leads to the production of more light components [94]. This is related to the presence of silicate, which has a significant impact on chemical processes. Silicate is able to promote the cleavage of long-chain aliphatic hydrocarbons into shorter hydrocarbon chains. At the same time, it promotes the binding of hydrogen radicals, which together accelerate the formation of short-chain gases such as methane and ethane. However, in contrast, the effect of silicate reduces the rate of hydrogen production. In short, silicate promotes the pyrolysis of complex hydrocarbons and the generation of specific gases but slows down hydrogen production [95].

Shale pyrolysis is a complex physicochemical process, which involves the decomposition of organic matter when heated to high temperature in the absence of air, along with recombination of products and secondary reactions. In this process, three kinetic factors together determine the rate and efficiency of the pyrolysis reaction [96].

Specifically, the activation energy E indicates how easily the pyrolysis reaction occurs, reflecting the energy required for reactant molecules to reach the activated state. The lower the activation energy is, the more easily the reaction occurs, and the higher the pyrolysis efficiency. The pre-reference factor A is related to the reaction rate constant and reflects the rate at which reactant molecules are converted to products per unit time at a given temperature. The higher the A value, the faster the reaction rate, and the more hydrocarbons are produced by pyrolysis [97].

The reaction mechanism function $f(\alpha)$ describes the relationship between the untransformed shale and the reaction rate, reflecting the comprehensive action of various reaction mechanisms in the pyrolysis process. Since shale pyrolysis is a complex process in which multiple reaction mechanisms coexist, the choice of $f(\alpha)$ is crucial for accurately describing the pyrolysis process [98].

In summary, these three kinetic factors are interrelated and act together in the shale pyrolysis process. Their changes directly affect the rate and efficiency of the pyrolysis reaction. Further study of the relationships among these factors and shale pyrolysis is of great significance for optimizing the pyrolysis process and increasing hydrocarbon production [99, 100].

4. Conclusion and outlook

As a sedimentary rock rich in organic matter, oil shale contains abundant energy resources, which are of great importance for the diversification and sustainable development of the global energy structure. In the development of oil shale resources, electric heating in situ conversion, fluid heating, radiation heating, and combustion heating technologies – as the main in situ conversion methods – have shown their unique advantages and limitations.

In situ conversion technology by electric heating directly heats the oil shale layer with electric energy to realize the in situ pyrolysis of organic matter and transform it into recoverable oil and gas resources. This technology has the advantages of high control precision and uniform heating, but the problems of high energy consumption and cost still need to be further solved. Fluid heating technology uses high-temperature fluids (such as steam or supercritical water) to circulate through the oil shale layer to transfer heat and promote transformation. Its advantages are high thermal efficiency and flexible adjustment of the heating range, but the fluid injection and recovery system is more complex.

Radiation heating technology uses electromagnetic waves (such as microwave or infrared) to directly radiate the oil shale layer to achieve non-contact heating. It is characterized by fast heating speed and small heat loss. However, this technology has certain requirements for the physical properties of the oil shale, and the efficiency and safety of radiation sources are also areas of concern. Combustion heating technology introduces combustion reactions into the oil shale layer, using the heat generated by combustion for heating. This method is simple and direct, but the combustion process may produce harmful gases, posing environmental risks.

At present, researchers are conducting in-depth studies on the advantages and disadvantages of these technologies, aiming to improve conversion efficiency, reduce costs, and minimize environmental impact through technological innovation and process optimization. For example, efforts are made to develop more efficient power conversion devices, optimize fluid circulation systems, improve the precision control of radiation heating technologies, and explore cleaner combustion methods. At the same time, interdisciplinary cooperation is key to promoting the development of oil shale resources – by integrating knowledge from geology, materials science, chemical engineering, environmental science, and other fields – to jointly solve technical bottlenecks.

Looking ahead, with the continuous progress of technology and the gradual reduction of costs, electric heating in situ conversion, fluid heating, radiation heating, and combustion heating technologies are expected to play increasingly important roles in the development of oil shale resources. At the same time, with the global pursuit of low-carbon development, these technologies will also pay more attention to energy conservation, emission reduction, and environmental protection – contributing to the harmonious coexistence of energy development and ecological protection.

Data availability statement

All data used in this article are publicly available. No new data were created or analyzed in this study.

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