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Review

Advancements and Environmental Implications in Oil Shale Exploration and Processing

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Abstract: This comprehensive review presents a holistic examination of oil shale as a significant energy resource, focusing on its global reserves, extraction technologies, chemical characteristics, economic considerations, and environmental implications. Oil shale, boasting reserves equivalent to approximately 6 trillion barrels of shale oil worldwide, holds substantial potential to augment the global energy supply. Key extraction methods analyzed include surface mining, modified in situ, and true in situ conversion processes, each exhibiting distinct operational parameters and efficiencies. The review further delves into the chemical aspects of oil shale retorting and pyrolysis, highlighting the critical role of variables such as retorting temperature, residence time, particle size, and heating rate in determining the yield and composition of shale oil and byproducts. Economic analyses reveal that capital and operating costs, which vary according to the specific extraction and processing technologies implemented, are crucial in appraising the economic feasibility of oil shale projects. Lastly, the review acknowledges the potential environmental hazards linked with oil shale development, such as groundwater contamination and harmful emissions. It emphasizes the importance of rigorous monitoring programs, environmental impact assessments, sustainable technologies, and innovative strategies like co-combustion and comprehensive utilization systems in mitigating such impacts. The review underlines the need for a balanced approach that harmonizes technological advancement, economic viability, and environmental sustainability in oil shale exploitation.

Keywords: oil shale; pyrolysis; environmental concern; pollution; sustainability

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1. Introduction

Oil shale, an organic-rich sedimentary rock, holds significant potential as an unconventional energy source due to its abundant kerogen content, which can be converted into shale oil and gas through various extraction processes such as pyrolysis, retorting, and in situ conversion. With the world's increasing demand for energy [1], the exploration and exploitation of oil shale resources have gained considerable attention in recent years. However, the development of oil shale reserves presents several challenges, including complex geological structures, variable quality, and environmental concerns. Therefore, a comprehensive understanding of oil shale characteristics, formation processes, and extraction technologies is crucial for the sustainable and efficient utilization of this resource.

Previous studies have focused on the geological and geochemical aspects of oil shale, examining its occurrence, depositional environment, organic matter content, and hydrocarbon potential [2–4]. In addition, several reviews have addressed the challenges and advancements in oil shale extraction techniques, including retorting, pyrolysis, and in situ conversion, along with their environmental implications.

Despite the wealth of existing knowledge, gaps remain in our understanding of oil shale's multifaceted nature, particularly concerning the interplay between its geological, geochemical, and technological aspects. Furthermore, there is a need to integrate the latest findings and advancements in oil shale research to provide a comprehensive and up-to-date overview of the field.

This review aims to bridge these gaps by thoroughly analyzing oil shale's geology, geochemistry, and extraction technologies, drawing from recent studies, and incorporating novel insights. The review will also discuss the environmental impacts of oil shale exploitation, including effects on geological environments, water resources, and air quality, and will explore strategies and technologies implemented to mitigate these adverse consequences. Lastly, the review will highlight the areas where previous reviews have fallen short and the motivation behind the present study (Figure 1).

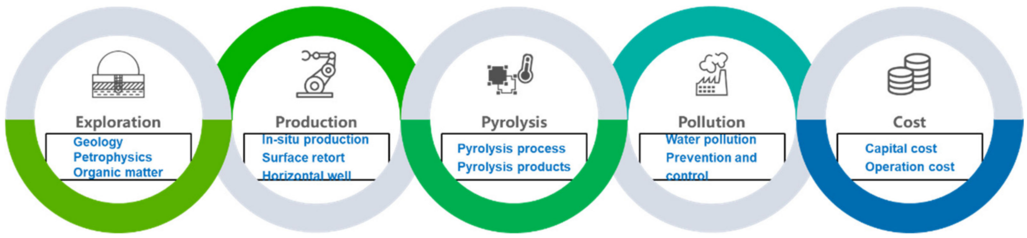

Figure 1. Scope of the oil shale review in this study.

By providing a comprehensive and current synthesis of oil shale research, this review seeks to facilitate the sustainable development of oil shale resources, foster innovation in extraction technologies, and contribute to addressing the global energy challenge. The findings and insights presented herein will be valuable for researchers, policymakers, and industry stakeholders engaged in the exploration, exploitation, and management of oil shale resources.

2. Oil Shale Properties

2.1. Oil Shale Geology

Oil shale is a type of sedimentary rock that contains organic matter known as kerogen, which can be converted into liquid hydrocarbons through thermal processing. It is considered an unconventional source of oil due to its low energy density and high production costs. According to the U.S. Energy Information Administration, global oil shale resources are estimated to be around 6 trillion barrels, with the largest deposits located in the United States, Russia, and China [5,6].

Oil shale is formed through the accumulation of organic matter in fine-grained sedimentary rocks over millions of years. The organic matter, primarily composed of algae and other aquatic organisms, undergoes chemical and physical changes under high pressure and temperature, resulting in the formation of kerogen. According to a study by Dyni (2010), the organic content of oil shale can range from 3% to 60%, with higher organic content leading to greater potential for hydrocarbon production [7].

Deposits of oil-shale can be found across numerous global locations. These deposits span from the Cambrian to Tertiary periods and originated in diverse marine, terrestrial, and lake-related settings for deposition. China's major oil shale deposits are located in Fushun, Maoming, and Xinjiang, with estimated reserves of approximately 354 billion barrels [8]. These oil shale deposits are typically found in lacustrine or marine sedimentary environments and are characterized by their low permeability and fine-grained matrix. In the United States, the Green River Formation is one of the largest oil shale deposits, with an estimated 4.28 trillion barrels of in-place oil shale resources, according to a report by the U.S. Department of Energy (DOE) [9].

During the early to middle Eocene epoch, extensive lakes formed the Green River Formation's lacustrine deposits, spanning 65,000 km^2 throughout Colorado, Wyoming, and Utah. Closures and fluctuations in these lakes led to high salinity and varying water levels, which caused diverse mineral precipitation and sulfate depletion by sulfate-reducing microorganisms [10].

The warm, alkaline conditions promoted the flourishing of blue-green algae, the main contributor to the oil shale's organic content. Home to various aquatic life and surrounded by rich land ecosystems, the Green River lake system thrived for over 10 million years in a warm temperate to subtropical climate, playing a significant role in the formation of oil shale deposits.

Situated in northeast China's Liaoning Province, the Fushun basin is a modest-sized, investigated Cenozoic fault basin recognized for its coal and oil shale reserves. The basin is primarily composed of Eocene swamp to lacustrine deposits belonging to the Guchengzi and Xilutian Formations and hosts Asia's most expansive opencast oil shale mine. The Eocene climate in the Fushun basin transformed from a warm temperate environment to a north subtropical one, generally transitioning from warm, humid conditions to subhumid-semiarid [10,11].

When comparing the geology of Fushun oil shale with the Green River Formation, both basins contain Eocene lacustrine deposits. However, while the Green River Formation was marked by significant salinity fluctuations and water level variations, the Fushun basin experienced a warm, humid climate with substantial precipitation in the Jijuntun Formation before shifting to a sub-humid to semiarid climate during the Xilutian Formation.

In the Fushun basin, the warm and humid climate during the Jijuntun Formation led to high initial productivity in lake water and a subsequent stable stratification. This resulted in oxygen deficiency at the bottom of the lake, facilitating the preservation of organic matter and the formation of the Jijuntun Formation's thick oil shale deposits. Conversely, the warm alkaline lake waters in the Green River Formation primarily supported the extensive growth of blue-green algae, which served as the main contributor to the organic content in the oil shale deposits [7].

Through geochemical assessments and microscopic studies, Jin et al., found that the primary source of organic matter in the oil shale was aquatic organisms, substantial deposits of dark mudstone and oil shale in the Sangonghe area situated at the northern foothills of the Bogeda Mountains within the Junggar Basin. These deposits were formed under anoxic conditions in a deep lake setting [12].

The lake productivity had a more significant impact on the oil shale quality than the preservation state of the organisms. Multiple factors, including the ancient climate, source rock chemical alterations, and lake conditions, contributed to the formation of high-quality oil shale. Warm and moist conditions led to the erosion of rocks and the release of nutrient elements into the prehistoric lake. Elevated paleotemperatures, alkaline waters, strong anoxic conditions, and particular hydrodynamic circumstances further aided in the release of nutrients from sediments at the lake's bottom, thereby enhancing lake productivity and enabling the development of high-quality oil shale [12].

Xu et al. (2022) describe a study on the Qingshankou Formation and Nenjiang Formation in the Songliao Basin, which aimed to reveal differences in hydrocarbon generation processes and controlling factors of oil shales of different qualities. They showed that oil shales abundant in algae possess significant hydrocarbon potential, making the investigation of hydrocarbon generation processes in oil shales of varying quality crucial for shale oil resource discovery. They found that oil yields (O.Y.s) possess a linear relationship with TOC concentration at varying depths. A threshold thermal maturity of 1.05% is found beyond which oil shale starts to produce illite due to the enhanced catalytic activity of smectite with the presence of bulk telalginite, which is beneficial for hydrocarbon generation [13].

2.2. Porosity/Permeability and Saturation Evolution

Permeability of oil shale changes as it undergoes heating and compression [14,15]. The permeability-time curves for columns of confined fragments prepared from different grades of Green River oil shales show that as compressive strain increases with time, the columns' pore geometry, porosity, and permeability undergo concurrent changes. Burnham (2017) continues studying the permeability and porosity evolution of Green River oil shale during retorting [16]. The permeability is calculated from porosity using a modified Kozeny–Carman relation, and it increases as the fraction of oil shale retorted increases. The porosity decreases as the grade of oil shale increases and as kerogen conversion occurs during in situ retorting. Compaction during diagenesis can also affect porosity [14].

Using numerical simulations, Kibodeaux (2014) described porosity permeability and saturation distributions during the ICP process [17]. Porosity, permeability, and saturation undergo significant changes driven by converting solid kerogen (and any reactive minerals) into fluids. Results show that the ICP process increases the porosity and permeability above their original values by opening up pore space through kerogen conversion. This effect is somewhat counteracted by competing factors such as compaction and coking. The degree of compaction is influenced by stress history, including zero lateral strain and a vertical load that increases with pattern depth and decreases overburden stiffness. Lack of vapor space can retard compaction. The triggers for compaction during ICP are kerogen softening, grain loss through pyrolytic conversion, and fluid pressure decrease. The evolution of porosity results in the development of permeable regions within ICP well patterns.

As the high temperatures from the ICP process propagate away from the heaters, a kerogen-depletion front leads the product-generation front, resulting in the development of permeable pathways that connect the producer well to the pyrolyzing regions. This permeable network is established relatively early in the ICP, suggesting that pre-ICP measures to enhance oil-shale permeability may not be necessary.

During ICP, there is a window of increased oil saturation, which results in a period of enhanced heavy-oil flow towards producer wells, leading to a period of greater heavy fractions in the produced oil. Conditions that may enhance this in situ peak of oil saturation include lesser initial porosity, lesser initial gas saturation, lesser initial water saturation, greater hydrocarbon richness (both kerogen and native bitumen), lesser reactive mineral content, and greater magnitude of compaction [17].

The changes in porosity, permeability, and saturation observed during the ICP process have significant implications for the efficiency and output of oil production from oil shale reservoirs.

Zhao and Kang (2021) experiment measuring The permeability of Fushun oil shale, which exhibits distinct patterns as temperature increases [18]. Below 200 °C, the oil shale is nearly impermeable. From 200 °C to 350 °C, the permeability gradually increases but then sharply decreases at 350 °C. Between 350 °C and 400 °C, the permeability shows mixed behavior among the samples. From 400 °C to 500 °C, the permeability varies with different trends for each sample. Finally, from 500 °C to 600 °C, the permeability of all samples increases, reaching a maximum at 600 °C. Results indicate that permeability variations are influenced by factors such as thermal cracking, organic matter pyrolysis, and the development of fractures and pores within the oil shale [18].

In addition to the general permeability and porosity, Yang et al. (2016) provided pore size distributions in oil shale under different conditions. With increasing temperature, the distribution of pore sizes in oil shale undergoes changes. At temperatures below 300 °C, oil shale mainly consists of large pores, small pores, and micropores. The volume of mesopores remains relatively low and is primarily concentrated in the temperature range of 20–300 °C. As the temperature rises from 300 to 600 °C, the volume of large pores significantly decreases, while the volume of mesopores increases significantly. The volume of small pores decreases, and the volume of micropores experiences a sharp decline. Particularly in the temperature range of 400–500 °C, extensive pyrolysis of organic matter leads to the aggregation and connection of micropores, resulting in an increase in mesopores and a decrease in micropores [19].

The permeability of oil shale gradually increases with temperature. At 600 °C, the permeability reaches 3.0×10^{-8} m^2, which is nearly 600 times higher than the permeability at the initial stage of pyrolysis. As the temperature increases, the generation of new pores and the connection of existing pores in oil shale contribute to its transformation from a tight rock into a porous material with high permeability, facilitating the seepage of oil and gas. Therefore, it has been proved experimentally that raising the temperature of oil shale induces changes in pore size distribution, leading to an increase in permeability. This transformation allows for enhanced oil and gas flow through the rock.

2.3. Organic Matter Type in Oil Shale

Oil shale comprises diverse organic components, such as kerogen, bitumen, and other hydrocarbons [20–22]. The composition and quantity of organic matter can differ based on the unique properties of each oil shale deposit. The kerogen-abundant portion can be employed to produce shale oil from the oil shale, while the kerogen-scarce portion can be used for generating power.

Kerogen is a complex mixture of organic compounds derived from ancient marine, terrestrial, and lacustrine (lake) sediments. It primarily consists of carbon, hydrogen, oxygen, nitrogen, and smaller amounts of sulfur. The exact composition can vary depending on the source and maturity of the kerogen. Kerogen is formed from the remains of ancient plants and microorganisms that lived millions of years ago. Over time, these organic materials were buried and subjected to high pressure and temperature, leading to the transformation of the organic matter into kerogen. Type I kerogen has a high hydrogen-to-carbon ratio and excellent oil-generating potential, derived mostly from algae. Type II kerogen is derived mostly from terrestrial and marine plants and has moderate oil-generating potential. Type III kerogen is mostly derived from terrestrial plants and has relatively low oil-generating potential.

Hutton, in 1987 classified oil shale based on existing petroleum and coal science technologies. The classification is mainly based on algal minerals. Based on different compositions of the algal minerals, oil shale could be classified into cannel coal, torbanite, lamosite, marinite, tasmanite, and kuckersite. Based on different algae forms, oil shale can be classified into charophyte macerals and lamalginite, primarily derived from unique algae and limited algal minerals.

Zhang et al. (2016) performed float and sink analyses on unprocessed oil shale samples from Longkou, China. They discovered that the predominant inorganic constituents were quartz, kaolinite, montmorillonite, calcite, analcime, dolomite, and pyrite. Additionally, they found that lower-density fractions contained a greater concentration of kerogen [23].

Siskin et al. (1995) classified the Green River oil shale based on different criteria. The shale can be classified into Mahogany Zone and Parachute Creek Zone based on origin content. The Mahogany Zone contains a higher amount of organic carbon, averaging around 30%. It is considered an oil-prone zone, as it has a higher potential for liquid oil production during thermal processing. On the other hand, the Parachute Creek Zone has a lower organic carbon content, averaging around 15%, making it a gas-prone zone. It predominantly yields natural gas upon thermal processing.

Based on thermal maturity, which refers to the degree of organic matter transformation due to heat and pressure [24–26]. Green River oil shale can be classified into the following three thermal maturity levels: immature, mature, and over-mature. The maturity level affects the type and quantity of hydrocarbons that can be extracted from the shale. Immature shale has a higher potential for oil generation, while over-mature shale tends to produce more gas.

In addition, the classification could also be based on the functional groups in the organic compounds present in oil shale, such as carboxylic acids and esters, nitrogen-containing compounds like amines and pyrroles, as well as sulfur-containing compounds such as thiophenes [27].

Hakimi et al., analyzed oil shale samples from the Lajjun area in Central Jordan. They applied comprehensive organic geochemical and petrological studies to determine the nature of these oil shales. The methods of classification and characterization involved the use of biomarkers, thermal maturity indicators, and microscopic observation of organic matter [28].

Based on the organic geochemical analysis, a series of methods, including analyzing total organic carbon (TOC), total sulfur content (T.S.), and potential yield (P.Y.) values, were applied. The TOC values in the studied samples were found to be greater than 10 wt%, indicating a high organic richness. The total sulfur content was also high (T.S. > 2 wt%), indicating a marine environmental setting with low oxygen conditions.

Based on the Biomarker, the ratios of different compounds such as C31-22R-hopane/C30-hopane, Gammacerane/C30 hopane, pristane/phytane, and steranes were measured. The biomarker distributions suggest high contributions of marine-derived organic matter that were preserved under highly hypersaline-reducing conditions.

Based on the thermal index, the thermal maturity of the organic matter was evaluated using pyrolysis data (Tmax) and specific biomarker maturity parameters. Tmax, in pyrolysis, refers to the highest temperature reached during the thermal decomposition process. It represents the peak temperature at which the pyrolysis reactions occur most rapidly. The majority of oil shale samples had Tmax values lower than 430 °C, indicating they are in the immature stage of the oil generation window.

Based on a petrological index, microscopic observations of organic matter were performed. The presence of alginite, amorphous organic matter, and planktonic foraminifera assemblages suggested a marine origin for the organic matter input. Based on the different evaluations, Hakimi et al. determined the Jordanian oil shales were classified as being organically rich, deposited in a marine environment under highly hypersaline reducing conditions, and in the immature stage for oil generation, requiring artificial heating for commercial oil hydrocarbon generation.

This categorization approaches offer valuable insights into evaluating the energy capacity, yield of hydrocarbons, and economic feasibility of distinct segments within oil shale. They aid in identifying the most effective extraction and refining methodologies to optimize the retrieval of valuable assets from this significant geological deposit.

2.4. Oil Shale Rating

To evaluate oil shale resources, it is essential to consider the correlation between the ratio of oil shale components and calorific value. Calorific value can be determined with sufficient precision based on the ratio of combustible and non-combustible parts if the mineral content of the non-combustible part is constant. When calculating the heating value of oil shale mixtures, the calorific value of the non-combustible part must be considered negative.

Energy rating and weighted average calorific value of the oil shale bed are the principal indicators for oil shale reserves. The calorific value of the bed is calculated as the weighted average value of oil shale and limestone layers, considering their thickness and volumetric weight. The energy rating considers the energy, thickness, and mass of all the layers.

However, the official method for calculating oil shale bed quality may not be accurate enough, as it does not account for differences in the component ratios of the non-combustible part A/CO2. To improve accuracy, a more precise method should be used that considers the different mineral content of the non-combustible part and the energy absorption required to decompose lime minerals.

3. Exploration and Production

3.1. Exploration Method

Advanced geological exploration techniques are essential for identifying and locating oil shale deposits [1,29]. Traditional oil shale exploration techniques involve drilling wells and analyzing core samples to determine the organic content and quality of the rock.

A program must be developed to measure various oil shale characteristics closely related to its ability to produce oil. The program needs to be designed to establish a correlation between the quality of the oil that can be extracted and the properties that can be determined from the logs. This allows for directly determining the oil shale's yield potential using logging data alone [30].

This method is time-consuming and expensive, and it can be difficult to obtain representative samples due to the heterogeneity of oil shale deposits [30].

In recent years, seismic technology has been increasingly used in oil shale exploration [31]. Seismic surveys can provide detailed information on the subsurface structure and properties of oil shale deposits, allowing for more accurate mapping and evaluation. Seismic data can also be combined with well log data to create 3D models of the subsurface, which can aid in identifying potential drilling locations [32–34].

In the Songliao basin, Jia et al. (2012) used a technique called log-seismic multi-attribute construction to create a set of quantitative attributes that could be used to identify and evaluate oil shale deposits. These attributes included natural gamma, resistivity, acoustic travel time, neutron porosity, and density [35].

By combining these attributes with seismic data, the authors were able to create 3D models of the subsurface that accurately identified and characterized oil shale deposits. They also used logging-constrained seismic inversion to further improve the accuracy of their interpretation [35].

3.2. Production Technique

In situ and surface retort technologies are used to extract oil from oil shale. These methods vary in their processes and environmental impacts. Table 1 summarizes common in situ and surface retort technologies [36].

Table 1. Description of in situ and surface retort of oil shale resources.

	Name	Description	Advantage	Disadvantage
In situ	Shell's In situ Conversion Process (ICP) [37–39]	This method involves underground heating of oil shale, transforming kerogen into shale oil. It employs a freeze wall for groundwater preservation and utilizes electrical power for shale heating.	Environmentally friendly due to the freeze wall that prevents groundwater contamination, also does not require surface mining, reducing environmental disruption.	Relies heavily on electricity for heating, which could be a disadvantage if the energy is not sourced sustainably.
	ExxonMobil's Electrofrac Process [2,40]	This technique involves kerogen conversion into oil and gas through electric heating. By fracturing the oil shale, it allows for more efficient heat distribution and oil extraction.	The fracturing of oil shale improves heat distribution and oil extraction, potentially leading to higher yields.	Like ICP, the use of electric heating could be an energy-intensive process, which might not be sustainable or cost-effective in some circumstances.
	Chevron CRUSH [2]	This approach incorporates hydraulic fracturing, heat application, and solvent injection for oil extraction from oil shale. For improved efficiency, it uses a staged procedure.	Staged process of heating and solvent injection can enhance efficiency and yield.	The process is complex and may require substantial capital investment.

Table 1. *Cont.*

	Name	Description	Advantage	Disadvantage
Surface retort	Paraho Indirect Retort [41,42]	In this process, oil shale is mined, crushed, and then heated in a retort vessel. It comes in the following two versions: Paraho I and Paraho II, with the latter focused on enhancing energy efficiency and lessening environmental harm.	The Paraho II process has been designed with a focus on increased energy efficiency and reduced environmental impact.	The requirement for mining and crushing oil shale can be environmentally disruptive and energy-intensive.
	Kiviter Vertical Retort [43–45]	This method uses a vertical vessel design and solid heat carriers for extracting shale oil. The technique, having undergone several improvements, is in use in several countries including Estonia, Russia, and China.	A proven, widely-used technology that has undergone several improvements over time.	The vertical design may limit scalability or efficiency in certain contexts.
	Alberta Taciuk Process (ATP) [46–48]	A process that utilizes a high-efficiency horizontal rotating kiln to vaporize and reclaim organic constituents in diverse feedstock materials. The method has found applications in oil shale and oil sands treatment, as well as contaminated waste management.	The process is highly thermally efficient and versatile, with applications extending beyond oil shale and oil sands to waste treatment.	The process may be complex and require substantial investment for setup and operation.
	Red Leaf's EcoShale In-Capsule Process [49,50]	This innovative method combines surface mining with low-temperature roasting in an impoundment created by the mining excavation. It is a green process that minimizes CO_2 emissions and safeguards groundwater.	This process is environmentally friendly, with rapid reclamation mining and limited CO_2 emissions. It also does not require water for extraction.	The slower, lower-temperature process might limit the rate of production.

These technologies have been developed to enhance the extraction of oil from oil shale while simultaneously addressing issues related to environmental sustainability and energy efficiency. They continue to evolve, responding to new global challenges, such as climate change, in a quest to create more sustainable and economically viable solutions. While all these methods have their distinct advantages and disadvantages, the choice of method would depend on several factors, such as the specific geology of the oil shale deposit, local environmental regulations, availability of resources, and capital investment capabilities.

3.3. Well Types and Fracturing Fluids

In addition, horizontal well drilling has been used extensively in shale oil and gas reservoirs. Shell applied horizontal drilling technology for developing oil shale resources in Colorado's Piceance Basin Shell demonstrated and commercialized shale oil recovery via a second-generation ICP (in situ conversion process) with the assistance of horizontal wells. The ICP involves leaching saline minerals with hot water to develop permeability first. Lab pyrolysis experiments and reservoir simulation studies were completed alongside with baseline hydrological monitoring program in place.

Song et al., proposed a novel method for oil shale in situ conversion using multilateral wells, which improve heat transfer and production rate in thin oil shale formations by increasing contact with the reservoir. A 3D transient model is developed to analyze the temperature field, chemical processes, production data, and energy performance. Sensitivity analyses of oil shale properties, injection management, and multilateral well structures are conducted, revealing the strong dependency of hydrocarbon yield on oil shale temperature and the sensitivity of production time to specific heat capacity, injection fluid temperature, and mass flow rate. The research suggests that optimal performance can be achieved with 5-branch wells, 60-degree branch angles, and 40 m branch lengths. The study provides valuable insights and recommendations for improving oil shale recovery and utilization through the application of multilateral wells in in situ conversion processes [51].

Lee et al., conducted numerical simulations of kerogen pyrolysis by the in situ upgrading process of Steamfrac, which entails the steam or hot-water injection into multistage transverse fractured horizontal well systems. The study analyzed the sensitivity of the temperature distribution of the reservoir to the positions of horizontal wells and productivity by applying two different irreducible saturation of the aqueous phase in the rock matrix. The research shows that recovery and utilization of oil shale could be improved through the use of multistage transverse fractured horizontal well systems [52].

Yang et al. (2017) combined the fracturing fluid with iron oxide nanoparticles, which is used to facilitate the extraction of oil from oil shale. This process integrates microwave technology and hydraulic fracturing, wherein the hydraulic fracturing fluid enhances permeation channels in the shale matrix, ensuring effective oil production [53].

Microwave irradiation is used for the thermal decomposition of kerogen, the main organic component in oil shale, into oil. The iron oxide nanoparticles act as microwave absorbers, converting the microwave irradiation to energy, thus enabling the high temperatures necessary for the transformation of oil shale. Oil extraction offers several benefits over traditional methods. Compared to conventional heating, microwave irradiation results in a faster process, lower energy usage, and increased oil production. Additionally, the quality of the extracted oil is improved, with the method both breaking down the chemical bonds in larger molecules to convert them into lighter hydrocarbons and reducing the contents of nitrogen and sulfur. As a result, the oil is potentially upgraded into petrochemical products with high saturation and aromatic compositions. The most effective results, in terms of yield and quality of oil, were found in the sample group exposed to a microwave output power of 800 W, a reaction temperature of 950 °C, and 0.1 wt% iron oxide nanoparticles. However, this method's efficiency is still subject to further investigation and potential technological advancements, particularly in material technology, to withstand higher in situ reaction temperatures.

4. Oil Shale Chemical Characteristic

4.1. Oil Shale Retorting Chemistry Characteristic

Utilizing oil shale comprehensively is a cutting-edge technique that maximizes both the chemical and energy potential while reducing environmental consequences. To achieve a high yield of shale oil and process shale char effectively, several aspects must be taken into account, including retorting temperature, residence time, particle size, and heating rate.

Chemically speaking, the retorting temperature has the most substantial impact on shale oil yield. Increased temperatures lead to improved yields; however, it also results in higher input heat, heat loss, and costs associated with retorting. Furthermore, elevated temperatures can cause the cracking and coking of shale oil. Thus, it is vital to maintain an optimal temperature range (preferably between 460 and 490 °C) for the retorting process.

The duration of residence time also affects oil yield, with an increase in time resulting in improved yield. However, excessively extending the residence time may lead to increased cracking and coking of residual organic matter inside shale char particles, which is not desired. As a result, it is crucial to balance both retorting temperature and residence time for the best outcome.

Particle size is another critical element since both larger and smaller sizes can cause secondary decomposition of shale oil, reducing the yield. A medium particle size is best for enhancing the shale oil yield.

Finally, the rate of heating influences the yield of shale oil. A higher average heating rate of 10 °C/min is generally advantageous and consistent with most previous studies.

In summary, refining the retorting factors, particularly focusing on retorting temperature, is crucial for optimizing the yield of shale oil from a chemical perspective.

4.2. Oil Shale Pyrolysis

4.2.1. Oil Shale Pyrolysis Process

Pyrolysis of oil shale is a thermochemical decomposition conducted under oxygen-free conditions, which induces complex reactions and transforms kerogen in shale into hydrocarbons. Shale constitutes diverse organic composites, including high C/H ratio components and heteroatomic species (i.e., sulfur, nitrogen, oxygen).

Heteroatomic compounds, notably those with sulfur and nitrogen, can present challenges, leading to diminished fuel stability, increased nitrogen oxide emissions, and storage complications.

The pyrolysis procedure typically necessitates oil shale heating in a tube furnace to approximately 480 °C. This thermal cracking process, also referred to as "viscosity breaking," improves oil quality by lowering the pour point. The oil then undergoes coking and rectification.

Subsequent treatment of the resultant oil can enhance gasoline and other distillate yields. A notable approach involves recycling, which reintroduces rectification column residues into the system.

Advancements in pyrolysis processes and the development of new oil shale conversion techniques demand comprehensive study. This includes exploring rapid heating conditions for shale feedstock and secondary pyrolysis techniques, primarily focusing on catalytic hydrofining for heteroatomic compound elimination, specifically sulfur and nitrogen [54].

According to data from 2020, the oil shale market was valued at an estimated USD 2.8 billion, and projections suggest it could approximately double by the year 2030. Nonetheless, it is essential to recognize the inherent limitations of oil shale as a lower-tier energy commodity, which only becomes economically viable subsequent to undergoing rigorous mining and retorting processes. Take the Green River Formation in Colorado as an example, where roughly a ton of oil shale is needed to yield a mere 0.83 US barrels of oil. The actual utilizable organic matter encapsulated within the shale accounts for just one-fifth of its total weight. The remaining four-fifths, once extracted, poses formidable environmental challenges. As such, addressing these environmental implications is a vital aspect that warrants due consideration (source: https://www.alliedmarketresearch.com/oil-shale-market (accessed on 26 June 2023)).

Current technological advancements have certainly fostered growth in mining and surface mining operations. However, the pathway toward large-scale expansion remains fraught with obstacles. In terms of cost, figures from 2010 suggest a range of USD 70–USD

95 per barrel. In situ methods have indeed demonstrated potential via small-scale tests in certain locales, yet environmental considerations might present additional concerns. Even though the costs associated with in situ methods are predicted to undercut those of mining and surface mining, the dearth of publicly available data imposes difficulty in drawing definitive conclusions (source: https://scholar.law.colorado.edu/cgi/viewcontent.cgi?article=1003&context=promise-and-peril-of-oil-shale-development (accessed on 26 June 2023)).

4.2.2. Pyrolysis Products

The extraction of oil from oil shale can be performed using two major methods, in situ conversion and surface mining followed by retorting. In situ conversion is a method where the oil shale is heated underground to extract the oil. In this process, heating elements are placed deep into the oil shale formation, and heat is applied over an extended period, usually a couple of years. The heat prompts a chemical process that converts kerogen, the organic material in the shale, into oil and gas. The oil and gas are then pumped to the surface. On the other hand, surface mining followed by retorting, involves digging the oil shale from the ground, bringing it to the surface, and then heating it in a process called retorting. The retorting process involves heating the shale in a vessel or retort to a high temperature, typically around 500 °C, causing the kerogen to decompose.

The process of pyrolysis conducted on oil shale sourced from the Green River Formation, as documented by Hiller et al., resulted in the extraction of highly valuable hydrocarbons, colloquially termed shale oil, from the kerogen contained within.

In the course of oil shale pyrolysis, the primary yield comprises light gases, char, and a tar-like substance. The light gas fraction primarily consists of butane, propane, ethane, and methane. The tar-like substance is a complex hydrocarbon amalgamation, which has the potential for further refinement and subsequent application in diverse sectors, notably as fuels for transportation. The char, which is the solid remnant post-pyrolysis, is chiefly constituted of residual carbon and mineral matter.

An in-depth analysis of the kerogen under study revealed that the carbon content was predominantly aromatic and aliphatic, accounting for approximately 79.8% of the total. Smaller proportions were found to be carbonyl, carbides, and carboxyl groups. This insight is crucial since the carbon type present within kerogen directly influences the nature of the hydrocarbons that can be generated during the pyrolysis process. Additionally, the study determined that the release of hydrogen and carbon during pyrolysis outpaced that of sulfur and nitrogen. This may indicate that sulfur and nitrogen are likely situated within the kerogen's more thermally stable aromatic clusters.

Through gas chromatography/mass spectrometry (GC/MS) analysis, an abundance of 1-alkenes and alkanes was detected, signifying that linear hydrocarbons form a substantial product of the pyrolysis operation. This substantiates the proposition that in the kerogen's chemical structure model, sulfur and nitrogen heteroatoms should be located in the aromatic region, representing the kerogen's portions that exhibit stability at moderate temperatures.

This kind of investigation underscores that oil shale pyrolysis is a multifaceted process yielding diverse products, predominantly light gases, tar (or shale oil), and char. The product composition hinges on multiple factors, most notably the kerogen composition within the oil shale and the pyrolysis conditions employed [55].

The primary environmental factor that affects pyrolysis products is temperature; other factors also play important roles. Based on the study by Pan et al., on Jimsar oil shale, varying the thermal input from 400 to 520 °C substantially influences the product stream from oil shale pyrolysis, including the increase in yields of shale oil, non-condensable gases, and aqueous products. Moreover, the constituent distribution within shale oil experiences changes, with n-paraffins diminishing and aromatic hydrocarbons augmenting as the retort temperature escalates from 400 to 550 °C. Additionally, a rise in retort temperature enhances the yield of C6-C19 hydrocarbons while reducing the output of C20+ hydrocarbons.

Wang et al. (2022) conducted a temperature-dependent oil shale pyrolysis study of Fushun oil shale. Figure 2a–d present a clear correlation between the ascending temperature regime and the increased rate of transformation during the pyrolysis of oil shale, verifying the influence of the rate of heat application. The rate of change for FSOL1 and FSOL2 is quite similar, but a noticeable extension in the transformation duration for FSOL1 underscores the role of silicates in curtailing the time required for organic matter transformation. The transformation rates heighten further for FSOL3 (Figure 2c) and reach a peak for FSOL4 (Figure 2d), where the enhanced organic matter proportion significantly contributes to the boost in transformation rates [56].

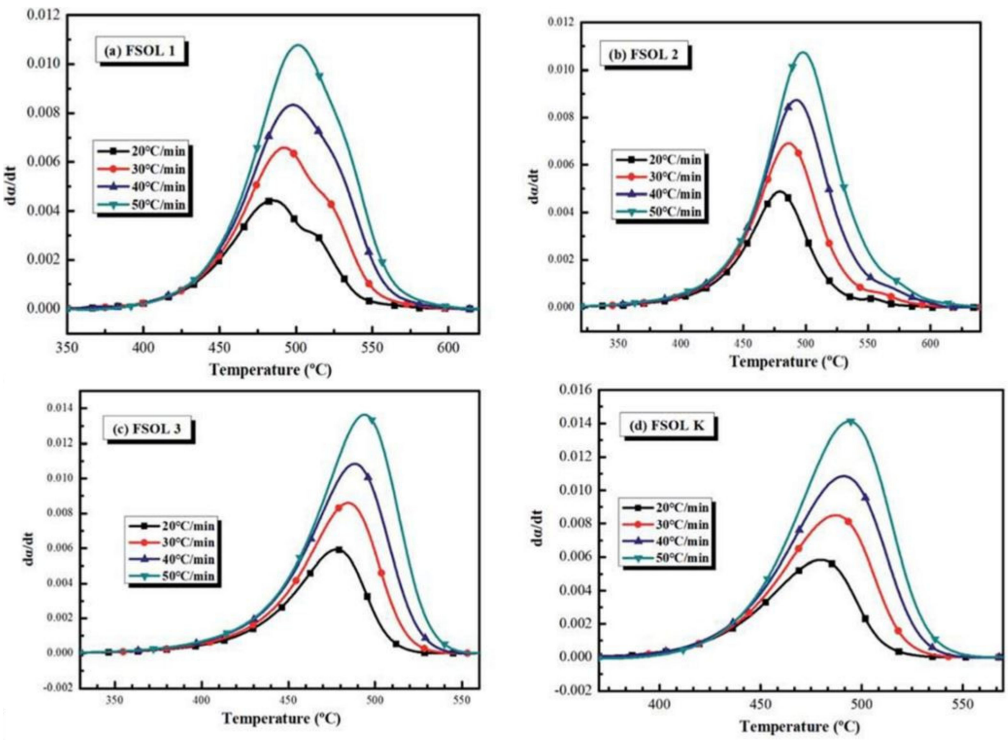

Figure 2. Oil shale of different samples conversion at different temperature. Reproduced with permission from ref [56]. Copyright 2022 Royal Society of Chemistry.

Also, the physical dimensions of the oil shale granules impact the concentration ratios of different constituents within the produced oil and gas. An upsurge in the size of oil shale granules leads to a reduction in 1-olefins and an augmentation in aromatic hydrocarbons. The proportion of n-paraffins and heteroatom-incorporated compounds also exhibit variations with granule size changes. Nevertheless, the aggregate yield of shale oil and non-condensable gases does not exhibit a significant correlation with granule size.

The retort temperature and the size of oil shale granules influence the extent of the secondary reactions happening during the pyrolysis process. These reactions, taking place within the confines of the oil shale granule and the packed bed, substantially modify the chemical composition of the end-product gases and shale oil. In addition, the spatial heterogeneity of kerogen and minerals in the oil shale matrix can also intervene in the pyrolysis reaction, thereby modifying the composition of the resulting products.

Therefore, the inherent properties of oil shale, the conditions inside the retort, and the specific characteristics of the pyrolysis process collectively govern the yield and the constituent makeup of the products obtained from oil shale pyrolysis.

Empirical models have been extensively used in the modeling of kerogen and heavy oil pyrolysis, such as the Braun and Burnham model and the Wellington model. A series of decomposition reactions are involved in these models, which are important for determining product distributions. However, it is important to note that these kinds of models have limitations and do not capture all the intricacies of the pyrolysis process. The models' predictive accuracy can vary depending on the specific oil shale composition and pyrolysis conditions. The frequency factor and reaction enthalpy may be adjusted based on more realistic scenarios.

5. Cost Analysis

5.1. Cost Composition

Seldomly successful oil shale production projects have been reported. Based on a news article by The Salt Lake Tribune (https://www.sltrib.com/news/environment/2018/10/02/major-utah-oil-shale/ accessed on 26 June 2023) on the development of oil shale in Utah, it is suggested that one of the recommendations for a successful oil shale development is the inclusion of contemporary case studies. By incorporating specific case studies showcasing successful implementations of oil shale extraction and processing, practical insights into the real-world application of the discussed technologies and methodologies can be provided.

The oil shale project in Utah has obtained authorization from the federal government to construct a 14-mile corridor across public land, which will serve a proposed strip mine and processing plant. This project is considered the first commercial oil shale operation in North America and aims to produce 50,000 barrels of crude oil per day. It has achieved significant milestones, including obtaining federal authorization for the construction of oil shale extraction and processing facilities on public land and showcasing practical application case studies of oil shale extraction technologies and methods. However, no further specific details or evaluations on oil shale development were provided. Therefore, we can conclude that oil shale development in Utah has achieved some level of success, but further information and assessments are needed for a comprehensive evaluation of its progress.

Over the past 60 years, numerous technologies have been developed to recover shale oil from oil shale and process it into fuels and byproducts. As mentioned earlier, technologies were chosen based on their development stage and availability of economic data, providing a broad range of options for surface, near-surface, and deeper oil shale deposits [57].

The costs associated with oil shale development are classified into the following two main categories: capital costs and operating costs. Capital costs encompass the expenses related to the extraction, retorting, and other necessary equipment for oil shale production. These costs are specific to the chosen technology, whether it be surface mining and surface retorting, underground mining, and surface retorting. Operating costs include daily expenses and maintenance for surface and subsurface equipment. The components of operating costs differ depending on the specific technology used, which includes mining, retorting, and upgrading facilities requirements.

Surface mining and surface retorting involve extracting oil shale near the surface and heating it to convert the kerogen into shale oil and combustible gases. This technology is applicable to deposits with a low overburden-to-pay ratio. Underground mining and surface retorting are suitable for resources that outcrop along steep erosions or are accessed via shafts, using room and pillar mining techniques.

The modified in situ (MIS) conversion process involves mining some of the target shales before heating. The mined shale is rubblized to create void spaces, and combustion is initiated on the rubblized shale, converting kerogen to shale oil and gases, which are captured and returned to the surface. The true in situ (TIS) conversion process requires no mining and is applicable to deeper oil shale deposits. Similar to Shell's in situ conversion process (ICP), this technology uses electric heaters in closely spaced vertical wells to heat the shale slowly, creating micro-fractures in the rock to facilitate fluid flow to production wells.

Mining and drilling cost data can be derived from related industries, such as coal mining in the U.S. and oil sands mining in Alberta, Canada. Upgrading cost data are available from technologies used in Alberta's oil sands industry [58]. Retorting costs, however, are highly dependent on the selected technology, and publicly available data are limited [59,60].

In conclusion, oil shale development offers a promising alternative energy source with various technologies available to extract and process shale oil. These technologies have different capital and operating costs, depending on the specific mining, retorting, and upgrading facilities required. Understanding each technology's cost classification and implications is crucial for decision-makers to evaluate the feasibility and economic viability of oil shale development projects.

5.2. Cost Status of Oil Shale

In 2020, the estimated value of the oil shale market was USD 2.8 billion, and it is projected to approximately double by 2030. However, it is important to acknowledge that oil shale is a low-grade energy commodity that becomes profitable only after mining and retorting processes. For instance, in the Green River Formation in Colorado, it takes about one ton of oil shale to produce 0.83 US barrels of oil. Only approximately one-fifth of the weight contains organic matter that can be utilized, while the remaining four-fifths must be mined and dealt with, posing environmental challenges. Addressing these environmental concerns is a significant aspect to consider (source: https://www.alliedmarketresearch.com/oil-shale-market accessed on 26 June 2023). Current technologies can support the development of mining and surface mining, but large-scale expansion still faces challenges. The cost in 2010 ranged from USD 70 to USD 95 per barrel. In situ methods have shown promise through small-scale tests in some areas, but environmental concerns may also be a cause for concern. Although the costs are expected to be lower than mining and surface mining, it is challenging to find relevant data in the public domain (source: https://scholar.law.colorado.edu/cgi/viewcontent.cgi?article=1003&context=promise-and-peril-of-oil-shale-development accessed on 26 June 2023).

6. Oil Shale Pollution Status and Preventive Measures

Oil shale development has gained significant attention due to its potential as an energy resource. However, the extraction and utilization of oil shale can have detrimental environmental impacts, particularly in terms of pollution [61,62]. Gharaibeh examined the oil shale pollution status in Jordan comprehensively. The primary concern is the potential contamination of groundwater due to the leaching of trace elements and heavy metals from spent shale [63]. The sequential extraction procedure conducted revealed that elements like Ti, V, Cr, Co, Zn, As, Zr, Cd, Pb, and U may pose a risk. Although their concentrations were below the maximum acceptable limits, precautionary measures are necessary to prevent any future contamination. The mobilization and bioavailability of trace elements in spent shale can also lead to soil pollution. Immobilization methods using kaolin as a soil amendment showed reduced mobility for certain elements but not for Ti, V, and Cr. Further research should explore alternative amendments and evaluate their efficacy in reducing the mobility and bioavailability of these elements.

To prevent water pollution, it is crucial to establish rigorous monitoring programs for groundwater quality. Regular monitoring of key pollutants should be conducted, and regulations should be implemented to ensure compliance with maximum acceptable limits. Implementing advanced oil shale extraction technologies that prioritize environmental sustainability is essential. Companies should adhere to international best practices and minimize the environmental impact of oil shale extraction. Also, prior to commencing oil shale operations, comprehensive environmental impact assessments should be conducted. These assessments should evaluate potential pollution risks and outline appropriate preventive measures to be implemented throughout the project's lifecycle.

The combustion of oil shale in China, particularly notable in regions like Huadian, contributes significantly to environmental pollution through the generation of harmful emissions such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), and carbon dioxide (CO_2).

This is primarily due to the unique characteristics of oil shale, including its high ash content, which can reach up to 50%, and the volatile matter released during combustion.

Several innovative technologies and strategies have been implemented to manage these environmental impacts. One such strategy is the use of circulating fluidized bed (CFB) combustion technology, which has been successfully deployed in a project with a handling capacity of 2.6 million tons per year of Huadian oil shale. The technology offers several advantages, including high efficiency and low emission of pollutants [64,65].

Another adopted measure is the co-combustion of oil shale with high sulfur fuel for in-furnace desulfurization, which leverages the high molar Ca/S ratio in some oil shale types. This can reduce sulfur dioxide emissions by up to 90%.

Additionally, comprehensive utilization projects have been implemented, transforming waste products from one process into inputs for another. In Huadian, China, a comprehensive utilization system has been established, including oil shale retorting, refining shale oil, burning oil shale semicoke in CFB boilers for electricity generation and heat supply, and producing building materials with oil shale ash. This integrated system significantly reduces waste and pollution, optimizing the use of resources and improving the environmental footprint of oil shale exploitation.

These strategies, when used collectively, aim to create a balance between meeting China's growing energy demands and mitigating the environmental impact of oil shale combustion [66].

For in situ oil shale development, it is crucial to implement these preventive measures effectively and adhere to strict environmental regulations to minimize the potential impact on groundwater resources. One frequently employed method is the use of freezing walls, also known as groundwater cutoff walls or ice walls, which are commonly used to isolate contaminated areas from the surrounding groundwater. This technique involves circulating a refrigerant through a network of boreholes surrounding the oil shale extraction zone. The refrigerant extracts heat from the ground, causing the groundwater to freeze and form a barrier, preventing the migration of contaminants. By employing methods like freezing walls and implementing appropriate treatment techniques, the industry can responsibly manage the extraction process and protect precious groundwater supplies from pollution.

7. Conclusions

Oil shale represents a significant, yet largely untapped, energy resource that holds considerable potential to meet global energy needs. This review has sought to provide a comprehensive overview of the current state of oil shale technology, including exploration and extraction methods, chemical characteristics, economic considerations, and environmental impacts. The multifaceted nature of oil shale development necessitates a holistic approach that balances technological advancements, economic viability, and environmental sustainability.

- Reserves and distribution: Oil shale is a widespread resource, with major deposits found globally. Estimates suggest that the world's total resources of oil shale are equivalent to 6 trillion barrels of shale oil, with the largest reserves situated in the United States, China, and Russia. The vastness of these resources underscores oil shale's potential to contribute significantly to the global energy supply;
- Extraction technologies: The extraction technologies employed for oil shale have evolved over time, reflecting the industry's progress and adaptability. Conventional mining, modified in situ conversion, and true in situ conversion represent key methods, each with their unique operational parameters, strengths, and limitations. However, these techniques are continually subject to improvement and innovation, with future advancements expected to further enhance efficiency;
- Chemical characteristics: Oil shale retorting and pyrolysis are central to the conversion of kerogen into usable hydrocarbons. Key parameters influencing these processes include retorting temperature, residence time, particle size, and heating rate. These factors, in turn, determine the yield and composition of shale oil and other products.

Future research can further optimize these parameters for improved yield and product quality;
- Economic analysis: The costs associated with oil shale development are primarily divided into capital and operating costs, which vary depending on the specific extraction and processing technology employed. Understanding these costs and their implications is vital for assessing the economic viability of oil shale projects;
- Environmental impacts and mitigation: The extraction and utilization of oil shale can lead to significant environmental pollution, particularly through potential groundwater contamination and harmful emissions. Implementing rigorous monitoring programs, environmental impact assessments, and sustainable technologies can help mitigate these impacts. Innovative strategies, like the co-combustion of oil shale with high sulfur fuel and comprehensive utilization systems, have also shown promise in reducing pollution.

In summary, oil shale offers promising prospects for energy production. However, its development necessitates careful balancing of technological advancements, economic considerations, and environmental sustainability. With continued research and innovation, oil shale has the potential to become a significant contributor to the global energy landscape. Nevertheless, achieving this potential will require a commitment to best practices, responsible stewardship, and a focus on sustainability.

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