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Comprehensive utilization of oil shale: a minireview

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Abstract. *Oil shale, a significant fossil energy source, has garnered global attention due to its huge reserves and potential as an alternative to conventional petroleum. This minireview evaluates the comprehensive utilization of oil shale, focusing on its entire lifecycle, from extraction to energy production, treatment of abandoned mines, and subsequent utilization to gain more economic and environmental benefits. In addition, the minireview underscores the necessity of green and high-value utilization of oil shale and its by-products (i.e., semi-coke and ash) to mitigate environmental pollution. To promote comprehensive utilization of oil shale and its by-products, we summarize current knowledge extending beyond traditional energy applications to encompass construction materials, environmental functional materials, and other high-value products. Strategies such as circulating fluidized bed combustion, in situ conversion, and co-combustion and co-pyrolysis with biomass are introduced for efficient resource use. The treatment of abandoned oil shale mines for energy storage and the recovery of trace and rare earth elements are also addressed. The minireview concludes with recommendations for improving testing strategies, assessing environmental impacts, and exploring new applications to ensure green and sustainable development in the oil shale industry.*

Keywords: *comprehensive utilization, oil shale, semi-coke, ash, energy storage, high-value materials.*

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1. Introduction

Petroleum, as a critical raw material and energy source, is in high demand across various industries, including chemical, agricultural, transportation, and other sectors. With the increasing global demand for petroleum, many countries are exploring alternative fossil energy sources beyond conventional oil and gas reservoirs, such as clastic and carbonate rocks. Among these alternatives, oil shale (OS) has emerged as a significant potential substitute due to its vast reserves, widespread distribution, and relatively easy extraction. Countries such as Australia, Brazil, China, Estonia, Jordan, Russia, Turkey, and the USA have recognized the potential of OS as a crucial resource for future energy needs [1–7].

OS is a solid, combustible organic sedimentary rock with high ash content. Its industrial standards vary by country, depending on local economic and technological conditions. For instance, in China, OS is considered industrially viable if it has an oil yield greater than 3.5% and a calorific value exceeding 4.18 MJ/kg (~1000 kcal/kg) [1, 4, 7–9]. The primary components of OS are minerals, organic matter (mainly kerogen with small amounts of bitumen), and water [3, 5, 6, 10, 11]. Kerogen, due to its macromolecular structure, cannot be extracted using common solvents but can be converted into hydrocarbons through heating [6]. The network structure of the kerogen is broken down by low-temperature (450–600 °C) retorting (pyrolysis or distillation) without air contact to produce shale oil, gas, water, and a solid residue known as semi-coke (SC) (Fig. 1) [5, 6, 12]. SC is a mixture of carbonaceous residues and minerals, and it is a by-product of the retorting process (Fig. 1) [5, 6, 12, 13]. In addition, OS and SC can generate electricity through combustion – especially OS, which is used in large quantities at OS-fired power plants –, leaving behind another by-product mainly consisting of inorganic materials known as oil shale ash (OSA) (Fig. 1) [5, 12, 14, 15].

In the process of OS retorting or combustion, large amounts of SC and OSA are yielded [6, 16–19]. OS, SC, and OSA often contain toxic substances, including organic compounds (e.g., polycyclic aromatic hydrocarbons (PAHs) and water-soluble phenols (WSP)), S, and heavy metals (e.g., As, Cd, Co, and V) [4, 6, 16–18, 20]. The massive accumulation of tailings and by-products of OS not only occupies considerable valuable land resources but also leads to spontaneous combustion due to long-term weathering and oxidation in open space, which may cause land disruption, geological hazards, and workplace accidents [16, 19, 20]. In addition, SC and OSA may cause air, soil, and water pollution as wind, rain, and groundwater transport pollutants from dumping sites to other areas, resulting in serious environmental problems and risks to human health [6, 16–18, 20, 21]. Therefore, the green and high-value utilization of OS and its by-products (i.e., SC and OSA) has become an urgent issue that needs to be addressed.

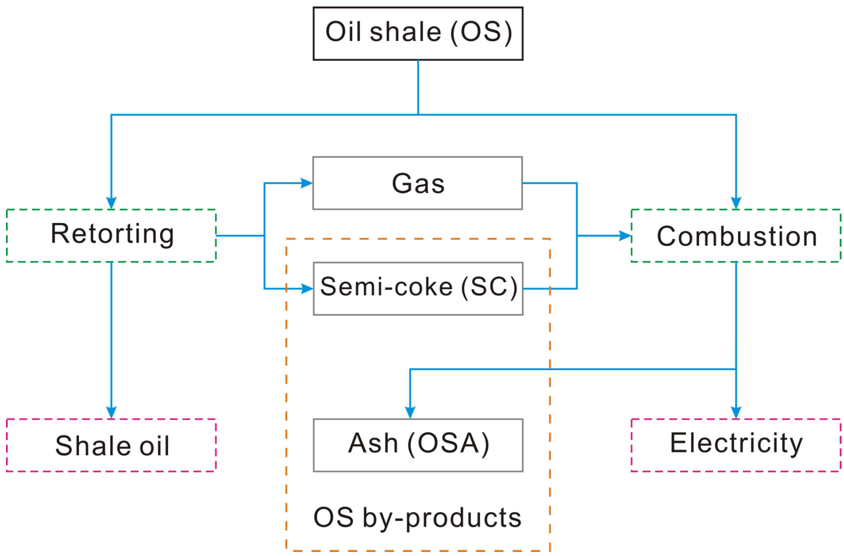

Fig. 1. Schematic diagram of conventional utilization system in OS industry.

Although factors such as low calorific value and high ash content have limited the rapid development of the OS industry in the global energy sector, OS provides more than just energy [22]. OS and its by-products also serve as valuable material resources. OS is relatively rich in Ca, Al, Si, trace elements, rare earth elements (REEs), and organic carbon. SC and OSA, in particular, tend to concentrate these elements due to their lower carbon content after retorting or combustion [6]. These materials have been utilized in various fields, including construction, environmental protection, agriculture, and the chemical industry [22–24].

The OS waste rock, SC, and OSA cannot simply be piled up as solid waste. Underutilization not only wastes resources and land but also results in adverse social, economic, and environmental impacts. Comprehensive utilization of OS transforms waste into value, significantly reducing solid waste, land use, and carbon emissions while helping to achieve carbon neutrality in the OS industry. For example, using SC and OSA as raw materials in cement production can substantially reduce greenhouse gas emissions compared to traditional cement manufacturing processes [19, 25, 26]. Additionally, comprehensive utilization of OS resources (including SC and OSA) can increase development benefits, especially by balancing the development costs of OS in the event of lower oil prices. To achieve these goals, it is crucial to adopt innovative technologies and sustainable practices that align with the principles of zero waste and green ecological development.

In recent years, there has been growing interest in exploring optimal ways to comprehensively utilize OS and its by-products. Advances in materials science and the development of unconventional energy resources, such as shale gas and coalbed methane, have provided valuable insights into the potential applications of OS resources. This minireview focuses on the entire lifecycle of the OS industry, from extraction and energy production to the treatment of abandoned mines and the subsequent utilization of by-products. It also provides recommendations for future research and development to ensure the sustainable and responsible use of OS resources.

2. Raw oil shale for energy

Due to the complex chemical characteristics of kerogen, OS can be converted into shale oil and gas only through retorting (Fig. 1) [27]. OS retorting technology are generally divided into two main categories based on the location of the retorting process relative to the OS reservoir: above-ground (ex situ) retorting and in situ conversion [13, 27, 28]. In addition to its common use for producing shale oil and gas, raw OS can also be combusted to generate electricity (Fig. 1). In some cases, OS power plants are integrated with above-ground retorting facilities.

2.1. Above-ground retorting and power generation

2.1.1. Above-ground retorting technologies

Above-ground retorting has become the most commonly used method in the OS industry due to its low cost, long history, and mature technology [6, 13, 28]. Almost all shale oil and gas produced globally today come from this category of technology. The above-ground retorting process typically includes mining (i.e., open-pit and underground methods), grinding, sieving, drying, heating, and ultimately producing shale oil and gas [13, 27]. Although this technology is suitable for large-scale mining development, it requires a large land area for operation and can only exploit shallow OS resources [13, 27, 28]. Therefore, above-ground retorting is unsuitable for deeply buried underground resources, especially those located beneath farmland and wetlands, such as in the southern Songliao Basin in Northeast China.

The most well-known above-ground retorting technologies are AOSTRA Taciuk Processing (ATP, Australia), Petrosix (Brazil), Fushun (China), Kiviter (Estonia), and Galoter (Estonia) types [5, 6, 27, 29]. Among these, two primary retorting methods are widely used: the solid heat carrier (SHC) and gaseous heat carrier (GHC) methods [6, 30, 31]. The SHC retorting method, such as the Galoter process and its derivatives (e.g., Petroter, Enefit-140, and Enefit-280), involves the use of solid materials (typically OSA) as heat carriers to transfer heat to the OS during retorting [6, 30]. The GHC retorting method, on the other hand, uses hot gases as heat carriers, which can be more complex in terms of heat transfer and control [6, 30].

To improve the efficiency of OS utilization, enhance shale oil and gas yield, and reduce the production of SC, above-ground retorting technologies are being integrated with advanced, proven technologies such as circulating fluidized bed (CFB) and SHC [6, 30]. The SHC process, particularly when integrated with CFB technology, allows for more efficient heat transfer and better control over the retorting process, leading to higher oil yields and reduced environmental impact [6, 30]. For instance, the Enefit-280 technology, a modern iteration of the Galoter process, utilizes CFB to prepare SHC for the retorting stage, significantly improving shale oil production efficiency and reducing emissions [30].

2.1.2. Power generation from OS and SC

OS has a long history of being used for electricity generation, similar to coal, with ash as the primary by-product [14, 15]. The thermal energy of the steam produced by OS combustion is converted into mechanical energy, which drives a generator to produce electricity [20]. CFB and pulverized firing (PF) are commonly used OS combustion technologies for power generation [31–34]. In recent decades, CFB technology has been replacing PF technology due to its superior economic and environmental performance [16, 32–34]. In addition to OS, fuels for power generation include SC and retorting gas produced during the retorting process (Fig. 1) [24].

Since the organic compounds remaining in SC have potential combustion heat, SC can be used as a fuel for combustion to generate electricity and provide heating to local residents [6]. Furthermore, technologies such as SHC and CFB can convert SC into OSA, a value-added multi-purpose material [6, 32–34]. This process significantly reduces emissions of harmful gases such as SO_2 and NO_x, while improving both economic and environmental benefits [6, 32–34].

2.1.3. Co-combustion and co-pyrolysis with biomass

Every year, a large amount of biomass waste is produced globally, especially crop straws (e.g., wheat, rice, and maize) [35]. Biomass is a potential green energy source with advantages such as easy storage, high combustion efficiency, and high calorific value [35]. When properly managed, biomass can be efficiently combusted in industrial applications, contributing to sustainable energy production and reduced environmental impact [36]. Studies on the co-combustion of OS and biomass in CFB boilers have demonstrated significant reductions in CO_2 emissions and OSA formation compared to conventional OS combustion [37, 38]. These findings highlight the potential of biomass as a valuable co-fuel in the context of OS utilization, especially when integrated with advanced combustion technologies.

Co-combustion of biomass with OS and SC offers several benefits, including increased combustion efficiency and reduced emissions of harmful gases such as SO_2 and NO_x [36, 39–41]. Industrial-scale applications have shown that biomass can be effectively utilized in CFB boilers, with minimal incomplete combustion when appropriate combustion conditions are maintained [37, 38]. This synergy not only enhances overall energy efficiency but also provides a sustainable solution for managing agricultural residues and other biomass wastes [35, 36]. By integrating biomass into the combustion process, these wastes can be converted into valuable energy resources, thereby reducing landfill use and minimizing the environmental footprint of waste management [36, 41].

In addition to co-combustion, the co-pyrolysis of OS and biomass offers another promising strategy for enhancing the efficiency and sustainability of energy production. The co-pyrolysis process involves heating OS and biomass together in the absence of oxygen, which can improve the yield and quality of liquid and gaseous products while reducing the formation of solid residues [35]. For example, studies have shown that co-pyrolysis of OS and biomass (e.g., wheat straw) can enhance overall energy recovery by 15–20% compared to individual pyrolysis of each feedstock, primarily due to the synergistic effects between the two feedstocks [35]. A notable example is the co-pyrolysis of OS with peanut shells, which has been shown to improve the thermal conversion efficiency by 18.5% and increase the yield of liquid products by 12.3% compared to individual pyrolysis of OS [42]. The interaction between OS and peanut shells during co-pyrolysis promotes the release of volatile compounds and enhances overall energy recovery, while reducing the formation of solid residues by 15.7% [42]. These co-combustion and co-pyrolysis characteristics provide a new approach for the co-governance of OS by-products and other combustible solid wastes, such as municipal solid waste [41].

2.1.4. Above-ground energy development strategies

The single use of OS for retorting or power generation could result in low resource utilization efficiency and considerable residual waste. To comprehensively utilize OS and reduce environmental pollution, especially solid waste pollution, researchers have proposed many prospective strategies based on different chemical and combustion characteristics of OS and SC [6, 29]. These strategies focus on the comprehensive utilization of OS and its by-products, encompassing processes such as retorting to produce oil and gas, combustion to generate electricity, and by-product utilization, which are currently or may be used in the future in China, Estonia, Jordan, and other countries or regions [2, 4, 7].

In addition to energy production, OS can be processed into chemical products, supporting the circular economy in the OS industry. For instance, VKG, an Estonian company, extracts fine chemicals, such as alkylresorcinols, from phenol water by-products of the Kiviter oil production process. These chemicals are used in rubber, plywood, petroleum, and high-end applications, such as automotive parts, cosmetics, and pharmaceuticals [43]. This not only boosts OS's economic value but also reduces waste, advancing sustainable development in the sector.

2.2. In situ conversion and carbon neutrality

In situ conversion technology for OS refers to artificially heating the underground reservoir, retorting the in-place solid kerogen into liquid oil and gas by maintaining a certain temperature for a period of time, and then extracting these products to the ground through specific processes [13, 27, 28, 44]. In situ conversion technology can be classified into four major groups according to the heating methods used in OS reservoirs: combustion heating, conduction heating, convection heating, and radiation heating (Table 1). Among these technologies, in situ conversion process (ICP), topochemical reaction (TSA), and in situ fracturing–combustion–heating have been successfully field-tested, while some others are still in the laboratory stage (Table 1) [13, 28]. Compared to above-ground retorting technology, in situ conversion can extract deep OS resources with fewer surface environmental impacts and avoid the safety risks associated with ground operations [13, 27, 28, 44–46]. However, its disadvantages include low technology maturity, potential underground environmental risks, high initial investment, and low mining efficiency (due to long heating times) [13, 27, 28, 44]. Integrating reservoir stimulation, underground sealing, in situ catalysis, and other advanced technologies may help overcome these disadvantages [28, 45].

As carbon neutrality has become a global consensus in response to global warming and climate change, carbon capture, utilization, and storage (CCUS) has emerged as a major research focus in the field of earth sciences. In situ conversion technologies, such as convection heating and radiation heating, use CO_2 either as a high-temperature fluid medium to heat reservoirs or as a carrier to displace produced hydrocarbons toward production wells, while the underground target formations provide space for CO_2 fixation [28, 45]. Therefore, in situ conversion of OS is an important application in CCUS. It is worth noting that there is a risk of CO_2 leakage during heating, displacement, and storage processes in CCUS projects, highlighting the need for robust monitoring and the adoption of new technologies.

In addition to in situ conversion, other CCUS technologies, such as capturing CO_2 from OS power plants, are essential for reducing the environmental impact of OS utilization. As demonstrated by Saia et al. (2022), retrofitting existing OS power plants with CO_2 capture technologies, including post-combustion capture (PCC) and oxy-fuel combustion (OXY), can significantly reduce CO_2 emissions [49]. However, the integration of these technologies reduces power plant efficiency and increases costs, which may exceed CO_2 emission allowance fees and environmental charges [49]. Therefore, a comprehensive evaluation of the technical and economic feasibility of CCUS technologies is necessary to achieve sustainable OS utilization.

Table 1. Classifications of in situ conversion technologies for oil shale [13, 27, 28, 44–48]

Heating method	Technology	R & D unit
Combustion	In situ combustion	U.S. Bureau of Mines
	TSA (Topochemical reaction)	Jilin University
	ATS (Autothermic pyrolysis in situ conversion process)	Jilin University
	In situ fracturing–combustion–heating	Jilin Zhongcheng Oil Shale Company
Conduction	ICP (In situ conversion process)	Shell
	HVF (High-voltage-power frequency electric heating)	Jilin University
	Electrofrac™ (Electric heating through induced fractures)	ExxonMobil
Convection	MTI (In situ steam injection technology, proposed by the Mining Technology Institute)	Taiyuan University of Technology
	SCW (Subcritical water)	Jilin University
	CCR (Conduction, convection, and reflux)	American Shale Oil LLC (AMSO)
Radiation	Borehole microwave	Phoenix Wyoming LLC
	RF/CF (Radio frequency/critical flow)	Raytheon
	RF (Radio frequency)	IIT Research Institute

3. Abandoned mines treatment

Abandoned OS mines may bring about various hazards, including geological disasters, surface damage, soil degradation, groundwater pollution, spontaneous combustion, and health risks [50–52]. However, due to their huge space and internal water resources, it is necessary to make rational use of these sites beyond simply backfilling them with waste rock, SC, or OSA.

3.1. Land reclamation and ecological restoration

Reclamation and restoration after treatment not only effectively solve the geological and ecological risks associated with abandoned mines but also significantly improve the living environment around OS sites. These efforts considerably increase the comprehensive utilization value of abandoned land and gradually restore regional ecosystem services and environmental quality.

Some abandoned mines are suitable for agriculture through reclamation, expanding arable land and supporting the local economy. In addition, the urban landscape has been dramatically improved following treatment, and several abandoned mines have become well-known tourism (e.g., industrial tourism) and recreational destinations, thus promoting the sustainable development of resource-exhausted cities. For example, after the treatment of the Maoming (China) OS open-pit mine, the area now serves as an ecological park covering about 10 km^2 for residents and tourists and functions as a reservoir with a storage capacity of 160×10^6 m^3 of water for irrigation (Fig. 2a) [53]. In another case, Seafield Law (Scotland) – a bing (hill) composed of spent OS and municipal landfill – has been reshaped and restored as a local landmark and valuable leisure and entertainment space, modeled after a local glacial "crag-and-tail" landform [54]. Similarly, a spoil tip (waste rock) in Ida-Viru County (Estonia), created by OS mining, is being transformed into an adventure park with a racetrack to enhance ecological value and promote tourism (Fig. 2b) [55].

Fig. 2. Examples of OS mines after treatment (source: Google Earth): (a) Maoming OS open-pit mine (Museum of Opencast Mining in the red-framed area); (b) MTÜ Estonia Elamuspark developed from a spoil tip in the red-framed area.

The underground spaces of abandoned mines can also be repurposed for museums, agriculture, laboratories (e.g., DUSEL in the USA), warehouses, ecological cities, data centers, waste disposals, parking garages, and more [51].

3.2. Energy storage

In recent years, the research and development of energy storage technologies have gained significant attention across various sectors – including power, transportation, and telecommunications – in many countries [56–58]. Abandoned OS mines, with their vast, well-sealed spaces and considerable vertical drop-offs, offer potential as facilities for the physical storage of electrical energy (Fig. 3). Compared to other energy storage technologies, such as chemical, electromagnetic, and phase-change systems, the physical storage of electrical energy in abandoned mines presents advantages such as low cost, large-scale storage capacity, and high technological maturity [59]. In addition, abandoned mines, particularly underground mines or depleted formations, can be utilized to store gas and heat for energy storage, peak shaving, and emergency supply purposes (Fig. 3) [51].

3.2.1. Electrical energy storage (EES)

Pumped hydro energy storage (PHES), gravity energy storage (GES), and compressed air energy storage (CAES) are the most promising technologies for storing electrical energy in abandoned OS mines (Fig. 3) [51, 52].

PHES and GES operate on similar principles: both technologies use electrical energy to elevate materials, thereby creating gravitational potential energy, which is later converted back into electrical energy through a generator [60–62]. The primary difference between the two lies in the materials used: PHES relies on water resources, while GES utilizes solid masses such as sand or gravel [60–62]. Open-pit and underground mines can be repurposed

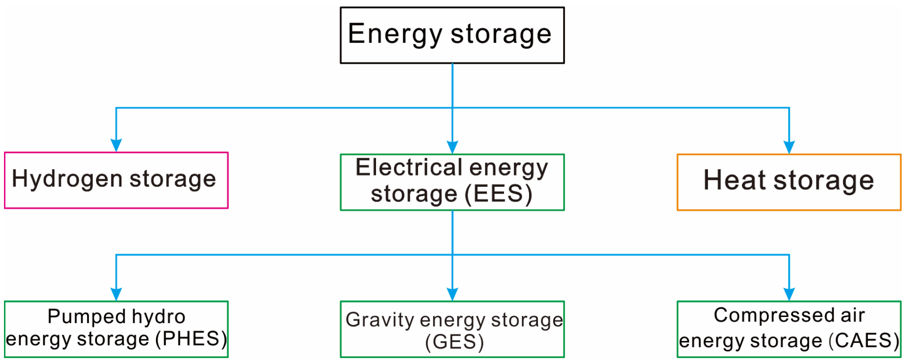

Fig. 3. Energy storage in abandoned OS mines.

for GES and PHES systems, provided sufficient drop heights exist [60, 62]. However, PHES also requires access to adequate water resources. CAES refers to pressurizing air using electricity during off-peak hours and releasing the high-pressure air through turbines to generate electricity during peak periods [56]. For example, Estonia is planning to build a PHES system with a capacity of up to 225 MW in Ida-Viru County, utilizing limestone rubble and closed mining tunnels left over from OS mining [63]. This project highlights the potential for repurposing abandoned OS mines as large-scale energy storage facilities.

In recent years, renewable energy sources, particularly solar and wind power, have experienced substantial growth in response to global climate change. Some abandoned mines have been or are being transformed into photovoltaic or wind power plants. However, the inherent volatility and intermittency of these energy sources can significantly impact power grid stability [60–62]. By integrating EES systems in abandoned mines with the power grid, especially in proximity to renewable energy power plants, it is possible to develop smart microgrids that ensure safety, stability, and strong peak regulation capabilities [62].

3.2.2. Hydrogen and heat storage

Hydrogen, with its high energy density and low mass density, has emerged as a promising solution to reduce dependence on fossil fuels, addressing environmental, climate change, and energy security concerns [64–66]. Therefore, hydrogen storage has recently gained significant prominence in green development. Compared with other hydrogen storage technologies, such as physical storage and material-based storage, underground hydrogen storage (UHS) offers the advantages of large-scale capacity and low cost (Fig. 3) [64]. UHS in depleted shale reservoirs is particularly promising due to favorable sealing properties, suitable infrastructure, and confirmed geological structures [64–66]. Depleted OS formations, especially those exploited through in situ conversion technology, could also serve as potential UHS facilities [67]. However, since in situ conversion, unlike hydraulic fracturing, significantly changes the original structure and composition of the formations, further research is necessary.

Geothermal energy, a clean and renewable source for heating and cooling, has gained attention worldwide due to its stability, environmental friendliness, low cost, and ease of operation. Mine water refers to any groundwater present in underground OS mines with a relatively stable temperature that fills after the mine is abandoned (Fig. 3) [68]. It can be a potential geothermal energy source for heat pumps and provide sustainable thermal energy solutions for local areas [68–71]. Utilizing mine water for heating in the winter period and cooling in the summer period can help reduce air pollution and greenhouse gas emissions compared to traditional fossil fuel-based systems [68–70].

3.3. Abandoned mine drainage (AMD)

AMD typically contains high concentrations of sulfates and metal ions, which can cause serious harm to the ecological environment and human health [72, 73]. Managing AMD has become a serious issue that plagues the treatment of abandoned mines. Both in situ conversion of OS and shale energy development require vast volumes of water for hydraulic fracturing and produce considerable quantities of wastewater during the flowback period [13, 28, 44, 72, 73]. If abandoned mines are located close to conversion or extraction sites, AMD can be considered a potential water source for hydraulic fracturing. The benefits of employing AMD for hydraulic fracturing include alleviating pressure on freshwater sources and mitigating AMD's adverse environmental impacts in mining regions [72, 73]. Additionally, AMD can be used as a substitute water source in operating OS mines for dust control and mineral processing [52].

4. Trace elements and REEs

OS, a fine-grained argillaceous sedimentary rock, is often rich in trace elements (e.g., Mo, U, Th, Pb, Zn, Cr, V, Ni, Co, Sr, Ba, Rb, Re) and REEs (including the lanthanides, Sc, and Y) [5, 16, 74–78]. Some trace elements, such as Mo (used in high-temperature metallurgy) and U (a nuclear fuel), hold significant economic value [79, 80]. REEs, often referred to as "industrial vitamins," are critical in military, petroleum, chemical, and advanced materials industries [81].

The average contents of trace elements (e.g., Be, B, Zn, Ga, Cs, Ta, W, Tl, Pb) and the total REEs (∑REE) in Chinese OSs are higher than crustal abundance, with average light REE (∑LREE, where LREE refers to La–Eu) levels slightly exceeding those of the North American shale composite (NASC) [78]. The graptolite argillite (also known as Dictyonema shale or Dictyonema argillite), a type of OS in northern Estonia, differs from the well-known Estonian kukersite OS used for retorting or combustion and is characterized by high concentrations of U (up to 1200 ppm), Mo (1000 ppm), V (over 1600 ppm), Zn, and other heavy metals [16, 74–77]. It is estimated that Estonian black shale contains approximately 5.666×10^6 t U, 12.762×10^6 t Mo, 47.754×10^6 t V, $0.213\text{-}0.254 \times 10^6$ t Th, and 16.533×10^6 t Zn [82].

Recent studies have shown that OSA can also be a significant source of REEs. For example, a study conducted in Jordan investigated the presence of REEs in OSA generated from different locations [83]. The study found that OSA contains various REEs extractable through acid digestion methods, with La and Ce being the most abundant elements [83]. The maximum concentration of REEs in all OSA samples reached up to 74.4 ppm, depending on the region [83]. Another study on Estonian kukersite OSA from CFB boilers reported the presence of REEs and demonstrated that the selective separation of ash fractions enables their recovery, particularly from the finer ash particles collected in electrostatic precipitators [84].

However, these elements can be converted into harmful substances if not appropriately treated and may cause pollution to the surrounding environment after OS retorting or power generation. The extraction of trace metal elements from OS and its by-products has not attracted enough attention due to grade, economic, and technological constraints. In fact, the large-scale extraction of elements from OS has a long history of nearly 80 years [16, 74–77]. Between 1948 and 1952, about 22.5 tons of elemental U were produced by processing 0.271×10^6 t of graptolite argillite from an underground mine near Sillamäe town in northeastern Estonia [16, 75–77]. In recent years, with the gradual maturation of element extraction technologies, efficient and clean extraction of trace elements and REEs from OS and its by-products has become increasingly feasible, helping to minimize environmental pollution and support the green and sustainable development of the OS industry [82].

5. Construction materials

OS and its by-products usually contain quartz, calcite, and clay minerals, making them valuable raw materials for various construction applications [19]. Among these, cement production stands out as a primary use, offering significant potential for reducing carbon emissions in the construction industry. Additionally, the by-products have historically been utilized in the production of bricks and blocks, providing a sustainable alternative to traditional clay-based materials [19]. Furthermore, the by-products can be employed as subgrade material in road construction, enhancing the physical properties of soil and contributing to the development of more durable infrastructure [85]. A notable example of this sustainable approach is the low-carbon construction materials project in Beipiao City, Liaoning Province, China, which processes 3.4 million tons of OS and its by-products annually. Integrating OS and its by-products into these construction applications helps reduce land occupation, minimize environmental pollution, and achieve a more sustainable approach to construction material production.

5.1. Cements

Cement is one of the world's most used construction materials, yet its production is one of the largest sources of global CO_2 emissions [26, 86]. Significant greenhouse gas emissions arise from calcite decarbonization and fuel consumption during clinker burning in cement production [86]. The cement industry is in urgent need of finding alternative raw materials and fuels to reduce carbon emissions. Utilizing OS and its by-products as substitutes can significantly reduce CO_2 emissions [19, 26, 86].

Among OS, SC, and OSA, OSA is the most commonly used raw material in cement production, followed by OS. OSA is recognized as a self-cementitious material, comprising two functional components: a cementitious part with high CaO content and a pozzolanic part containing SiO_2, Al_2O_3, and Fe_2O_3 [19, 86]. The use of OSA in cement production has a long history, dating back to the early 20th century [87]. For instance, the cement works at Dotternhausen in Germany have utilized OSA from burnt Toarcian Posidonia shale as an additive in blended cement since 1939 [22, 86–89]. Similarly, OSA from OS power plants is used as a supplementary cementitious material (SCM) at Kunda Nordic Cement in Estonia, replacing nearly 30% of clinker [22, 86, 90].

Calcareous OSs with low calorific value, found in many countries, are unsuitable for hydrocarbon and electricity production. However, their chemical compositions are similar to Portland cement clinker, making them suitable for partially replacing the fuel and raw materials needed in eco-friendly belite cement clinker production [86]. This substitution not only reduces CO_2 emissions but also lowers energy consumption [86]. It is worth noting that finding suitable blend ratios of OS and its by-products, along with optimizing operating temperatures, is essential to maintain cement's essential properties, such as strength, durability, and setting time [91].

Recent studies have shown that mechanical activation of OSA can significantly enhance its cementitious properties. For example, Paaver et al. (2021) demonstrated that even a short period of mechanical activation of Ca-rich circulating fluidized bed combustion (CFBC) OSA can yield a nearly tenfold improvement in the compressive strength of hydrated OSA pastes, reaching up to 60 MPa after 90 days of curing without any chemical activation or blending [92]. This suggests that mechanically activated OSA can be a viable alternative to traditional cement clinker, especially in regions where clinker production has been discontinued, such as in Estonia.

5.2. Bricks and blocks

Traditional brick and block production requires considerable clay resources, destroying large land areas and topsoil. OSA has historically been used for brick and block manufacturing [87]. Using OSA for bricks and blocks production, it is possible to reduce the waste of land resources and support a circular economy through the rational utilization of industrial solid waste.

A study by Hadi and Abdelhadi (2018) found that OSA, as a self-cementitious material, can be blended with marble and granite sludge to produce low-cost, compressed, strong, and lightweight bricks, while also minimizing their negative impacts on the environment [93]. The low thermal conductivity of these bricks makes them suitable for arid and semi-arid climates, contributing to improved indoor thermal comfort [93].

Silbet, a construction block produced from OSA, was one of the three most widely used small blocks for wall construction in Estonia, alongside Aeroc (aerated concrete block) and Fibo (compacted LECA, or lightweight expanded clay aggregate, blocks) [94]. A previous study indicates that Silbet blocks exhibited moderate density, thermal conductivity (λ_{10} and λ_n), and diffusion constant (μ) compared to the other two, making them suitable for lightweight block walls in Estonia's climate [94]. However, Silbet blocks have not been produced for about ten years. Until the end of 2016, Roclite blocks were also manufactured in Estonia from a mixture of OSA, sand, and water, offering durability and environmental benefits through the utilization of industrial by-products [95]. These practices highlight the potential of OSA and other by-products for sustainable brick and block production.

5.3. Subgrade materials

Road construction requires large quantities of aggregates and filling materials [85]. OSA, a type of solid waste, can be utilized as a subgrade material to partially substitute traditional aggregates and filling materials. Previous research has shown that using OSA as a fine aggregate in open graded friction courses (OGFC) improves pavement performance, extends service life, and enhances resistance to springtime clogging, which is beneficial for sponge city construction [96].

OSA also improves the physical properties of soil, supporting its role as a subgrade material [85, 97]. For example, OSA was used as a binder in the mass stabilization of soft peat soil and in the upper layer of the Simuna-Vaiatu Road constructed in a pristine swamp area in eastern Estonia from 2013 to 2014 [97]. A study by Wei et al. (2014) found that silty clay modified with OSA and fly ash (another solid combustion by-product) exhibited better mechanical and physical properties than the original silty clay, making it a promising "green" subgrade material [85].

In addition to subgrade applications, OS and its by-products, especially OSA, can also be used in a wide range of construction materials, including concrete, mortar, aggregates, geopolymers, glass-ceramics, soil stabilizers, asphalt binders, and even 3D printing materials, owing to their excellent properties such as self-cementitious behavior (Fig. 4) [19, 25, 98–101].

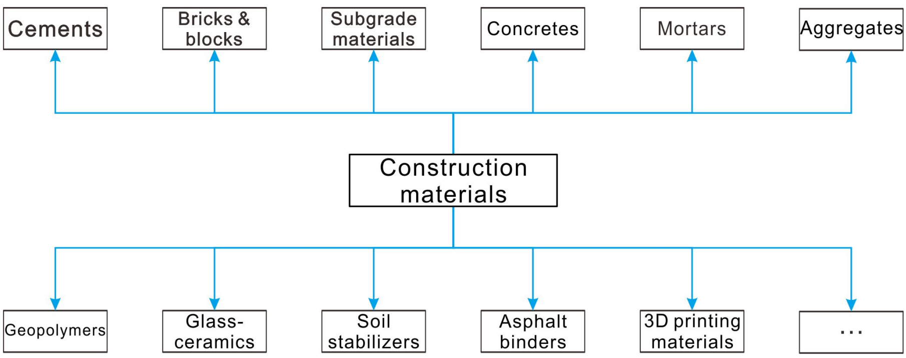

Fig. 4. Construction materials produced from OS and its by-products.

Besides OSA, waste rock from OS mining can also be utilized as aggregate in civil engineering and road construction, particularly in frost-free environments [102, 103]. This not only reduces the environmental impact of waste rock disposal but also provides a sustainable alternative to traditional construction materials, further contributing to the circular economy in the OS industry.

6. Environmental functional materials

Although OS and its by-products are usually considered potential pollution sources, they can serve as environmental functional materials to control pollution after activation and modification – especially SC and OSA. These materials are mainly used for water and air pollution control.

6.1. Water pollution control

SC has potential for use in adsorbent preparation due to its rich organic and inorganic components. However, its dense structure and complex composition result in poor adsorption performance [104]. The adsorption properties of different new functional materials synthesized after modification of SC with chemical and physical methods were significantly improved compared to the original SC, exhibiting excellent adsorption capacity for cationic and anionic dyes as well as other water pollutants [104, 105].

Similarly, OSA can be modified to synthesize effective adsorbents for dyes, heavy metals, and U, such as zeolite and hydrotalcite, providing new strategies for treating wastewater and managing solid waste from the OS industry [106–108]. Since OSA is rich in Al, Fe, and silicon oxides, modified OSA can also be used to prepare inorganic polymer coagulants for municipal sewage treatment [109]. For example, Shawabkeh et al. (2004) demonstrated that OSA can be converted into zeolite for the removal of heavy metals, such as Cd and Pb, from wastewater, showing high adsorption efficiency [106]. Additionally, OSA-based hydrotalcite has been used to remove U from contaminated water, achieving significant removal rates [108].

6.2. Air pollution control

Environmental functional materials prepared from modified SC and OSA can help reduce not only water pollution but also air pollution and carbon emissions. These materials exhibit outstanding gas adsorption characteristics – particularly for CO_2 – and good adsorption reusability, making them suitable for use in industrial processes involving physical adsorption, contributing to atmospheric pollution control [110, 111]. For instance, Reinik et al. (2011) demonstrated that alkaline-modified OSA exhibits a significant CO_2 adsorption capacity and could be utilized in industrial processes for capturing CO_2 emissions from flue gases, thereby contributing to the reduction of carbon emissions [110].

The OSA formed during the combustion of OS has a porous structure with good H_2S adsorption capacity and does not adsorb hydrocarbons, making it suitable as an adsorbent for removing H_2S from liquefied petroleum gas [112]. This application is particularly important in the petroleum industry, where H_2S is a common pollutant that needs to be removed to comply with environmental regulations. Using OSA for H_2S removal not only reduces air pollution but also enhances the safety and efficiency of industrial processes.

In addition to these materials, activated carbon (AC) and its composites – prepared from raw OS through chemical activation – exhibit strong adsorption capacity due to their high specific surface area and porous structure. These materials can effectively adsorb dyes, heavy metals, harmful gases, and other pollutants [113, 114]. For example, Hamdan et al. (2023) demonstrated that AC derived from OS effectively adsorbs volatile organic compounds (VOCs) from industrial emissions, contributing to air quality improvement [114].

7. Agriculture and forestry

OS and its by-products contain various nutrient components that support plant growth, offering potential applications in agriculture and forestry.

7.1. Agriculture

OS and SC are rich in organic matter, minerals, and trace elements, providing essential nutrients while also improving soil pH, aeration, structure, and water retention [115–118]. These enhancements increase soil fertility and support microbial activity. The use of OSA in agriculture has also shown promising results in improving crop yields and quality [23].

OSA has been widely recognized for its potential in neutralizing acid soils and improving the fertility of arable and grassland areas. Its high content of free CaO makes OSA an effective sorbent for binding acidic gaseous compounds such as SO_2 and CO_2, significantly enhancing soil pH and reducing acidity [119]. This process not only improves the soil's chemical properties but also enhances its physical structure, making it more suitable for agricultural use.

SC and OSA can be utilized to produce organic fertilizers and soil conditioners [115, 117]. Organic fertilizers supply sustainable nutrient sources, promote soil health, and stimulate plant growth. Soil conditioners further improve water and nutrient retention, bolstering plant resistance to diseases and pests. For instance, Mangrich et al. (2014) developed a slow-release K^+ fertilizer using OSA from the Brazilian OS industrialization process (Petrosix), demonstrating its potential to reduce nutrient loss in acidic soils [115]. Additionally, superabsorbent polymers (SAP) made from chitosan and SC have been explored for their excellent water absorption and retention capabilities, promoting plant growth and drought resistance [117].

7.2. Forestry

OS and its by-products are suitable for forestry, particularly in the afforestation of peatlands, the improvement of saline-alkali soils, and desertification control [118, 120]. When applied to soil, OS and SC can increase nutrient levels, crucial for the survival and growth of plants such as sea buckthorn to resist stressors like drought and salinity [118]. Alkaline OSAs, especially those rich in P and K, have also been shown to significantly enhance the growth and above- and below-ground biomass formation of species such as Silver birch and Scots pine [120].

8. High-value materials

SC and OSA, by-products from OS retorting and combustion – especially OSA – are gaining enormous attention in material science for their potential in high-value applications, offering innovative approaches for waste management and promoting environmental sustainability.

Rich in Si and Al, OSA is transformed into various valuable materials. Notably, it has been utilized to produce nano-sized α-Al_2O_3 and γ-Al_2O_3, which are applied in catalysts, electronics, and fine ceramic composites [121–123]. The production process typically involves calcination, acid leaching, and homogeneous chemical precipitation, which are crucial for controlling particle size and morphology [121]. Furthermore, OSA has been employed to synthesize shape-stabilized composite phase change materials (ss-CPCMs), which exhibit enhanced thermal properties, making them suitable for thermal energy storage in building envelopes, potentially leading to substantial energy savings [124]. The innovative use of OSA also extends to the production of silica nanoparticles and aerogels [125–127]. Silica nanoparticles derived from OSA have demonstrated high purity and specific surface area, desirable for catalysts, adsorbents, and lightweight structural materials [125]. Similarly, aerogels synthesized from OSA have shown excellent thermal insulation properties due to their low density and high porosity [126, 127].

SC has also garnered attention due to its substantial SiO_2 content and trace amounts of other metals. Extracted SiO_2 from SC can be used as a hard template for creating hierarchical porous carbon skeletons [128]. These nitrogen-doped porous carbon materials have superior performance as cathode hosts in Li–S batteries, highlighting their importance in energy storage applications [128].

9. Conclusions and recommendations

This minireview has highlighted the significance of OS as a resource that goes beyond its traditional energy applications, offering substantial potential for construction, agriculture, and other high-value uses. The comprehensive utilization of OS and its by-products, SC and OSA, presents an opportunity to address the environmental challenges associated with its exploitation while enhancing economic value.

The integration of OS into sustainable development strategies is contingent upon the adoption of innovative technologies and practices. These include using CFB, SHC, and other advanced proven technologies to improve oil yield; the application of in situ conversion technologies to reduce surface impacts; the implementation of carbon capture and storage solutions to mitigate greenhouse gas emissions; and both co-combustion with biomass to improve combustion efficiency and co-pyrolysis with biomass to enhance thermal conversion efficiency.

Equally important is the comprehensive treatment of abandoned OS mines to reduce adverse impacts on human society and the environment. With expansive and secure interiors, abandoned OS mines present an ideal setting for the physical storage of electrical, chemical, and thermal energy. The consolidation and rehabilitation of these sites for energy storage is a testament to the circular economy and a pragmatic solution to the intermittency challenges faced by renewable energy sources.

Furthermore, recovering valuable trace elements and REEs from OS and its by-products provides an additional source of revenue and contributes to reducing ecological harm. Transforming OS and its by-products into construction materials, environmental functional materials, and other high-value products is a prime example of the circular economy in action, promoting a sustainable and diversified utilization paradigm.

To ensure the sustainable and responsible development of OS resources, the following recommendations are proposed:

1. **Improve testing strategy:** To achieve optimal economic and environmental benefits, it is essential to improve the fundamental testing strategy for OS and its by-products. Varying oil content, calorific value, and chemical composition require customized development strategies. A comprehensive analysis will allow for the categorization and appropriate application of different OS, SC, and OSA, whether for energy extraction, material production, or other uses. This approach is crucial for maximizing resource value and minimizing negative environmental impacts.
2. **Assess environmental impacts:** Future research should focus on conducting comprehensive environmental impact assessments to quantify the benefits of utilizing OS by-products in various applications. Specific

attention should be given to comparing the carbon footprint of OS-derived materials with that of conventional products, such as cement and construction aggregates, to provide a clearer understanding of their environmental advantages.

3. **Explore new applications:** New technologies open up exciting possibilities for utilizing OS resources. For instance, our research team is working on using OS and its by-products to prepare a variety of novel high-value materials, such as carbon quantum dots (CQDs). Exploring the potential of OS resources in producing carbon nanotubes, graphene, and other advanced carbon-based materials could lead to the development of high-performance composites, electronic devices, and energy storage solutions. Additionally, further research is needed to investigate the potential of OS and its by-products in producing sustainable materials. By diversifying the applications of OS, it is possible to maximize resource utilization and contribute to the transition toward a more sustainable and circular economy.

Data availability statement

No new data were generated or analyzed in support of this study.

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