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OIL SHALE INTEGRATED TRI-GENERATION SYSTEM: THE TECHNOLOGY AND PREDICTED PERFORMANCE

J. O. JABER*
S. D. PROBERT

Department of Applied Energy
Cranfield University
Bedford MK 43 0AL, UK

M. A. TAHAT

Department of Mechanical Engineering
University of Science and Technology
Irbid, Jordan

This paper presents the general layout and main features of a new method for utilising oil shale: integrated tri-generation system. This new scheme is expected to be more efficient and environment-friendly, as well as a less-costly method, for producing electric power and synthetic (i.e. liquid and gaseous) fuels from oil shale, compared with traditional utilisation methods. By integrating the gasifier, circulating fluidised-bed combustor (CFBC), retort and combined-cycle turbine system in one plant, higher efficiencies can be achieved as well as lower costs of the final products.

Introduction

Low-grade alternative fossil fuels, such as oil shale, tar sands and biomass as energy sources are receiving serious attention because such materials are the most abundant sources of organic matter on Earth and found in many technically-developed as well as less-developed countries. Recent estimates of oil shale, which is a low-grade solid fuel, suggest that its remaining reserves world-wide are about $1.3 (\pm 0.2) \times 10^{13}$ tonnes [1, 2]. This is equivalent to approximately 2.2×10^{13} barrels of shale oil, and the estimated energy that has accumulated in oil shale deposits world-wide is ~2.5 times that of coal and at least 30 times that of the proven reserves of crude oil. However, only about 20-25 % of oil shale reserves are easily recoverable using current technologies [3].

* e-mail: J.O.A.Jaber@cranfield.ac.uk

Interest in oil shale technologies reached a peak in most developed countries just after the Second World War [4, 5]. However, oil shale operations in Estonia, China and the former USSR countries are still flourishing [6, 7]. After the crude oil unit-price hikes, during the 1970s and mid-1980s, extensive research-and-development projects were undertaken concerning the harnessing of renewable resources of energy as well as oil shale, especially in the USA, Canada and European countries. At present, oil shale utilisation technologies are limited to either destructive distillation (i.e. retorting) processes to produce shale oil and synthetic gases, or direct combustion for electric power generation and other industrial purposes. However, new technologies such as supercritical solvent extraction or bio-leaching to recover shale oil are promising but still in their early stages of development [8].

The conversion of solid fuels (e.g. coal, biomass or oil shale) to cleaner-burning and more user-friendly synthetic liquid or gaseous fuels for the transportation and industrial sectors is becoming more popular [9]. For example, integrated coal gasification combined cycle (ICGCC) is being employed in several countries because it offers the potential for higher-efficient electricity generation with less adverse environmental impacts [10, 11]. Synfuels (i.e. synthetic liquids and gases made from oil shale and other types of fossil fuel) have been used as supplements to petroleum products and natural gas supplies. Several processes (e.g. oil shale retorting and/or gasification) for making these fuels as well as for the direct generation of electricity from oil shale have been employed successfully on a commercial scale in various countries (e.g. Estonia, Russia and China). Other processes, with a similar purpose, have been developed and are approaching commercial readiness (e.g. in Australia, Japan and the USA). So, the utilisation of oil shale, as a fuel, is well established.

Oil shale could prove to be an alternate route in supplementing traditional sources of fossil fuels, such as crude oil and natural gas. But its utilisation involves several different technologies. These include mining and crushing, which are similar to those used in coal processing technology [12]. Retorting, gasification or direct combustion to generate electric power are the core of oil shale processing. However, the disposal of spent shale and other wastes in an environmentally acceptable manner and final upgrading the liquid and/or gaseous products into commercial forms are equally important. An understanding of these processes can help in selecting the most effective system design and operational procedures that lead to minimising the total (i.e. lifetime operating and capital) cost, including that associated with meeting environmental regulations.

The main aim of this article is to assess the integration of the gasification, retorting and combustion of oil shale to produce shale oil and fuel gas as well as electric power.

Oil Shale Utilisation Systems

There are two principal streams for developing oil-shale-fired power systems. The first is based on the steam turbine (using the Rankine-cycle), and the second on the gas turbine combination using a combined cycle. The efficiency of the first option would be approximately similar to that achievable with coal-fired systems: based on limited experience with a FBC, which burns oil shale, it would be about 36 (± 2) % and it may reach 40 % for the best scenario [13]. The efficiency increases dramatically (i.e. from ~40 to 47 % compared with 52 to 58 % for natural gas fuelled combined cycles) when a combined cycle arrangement is used as in a Pressurised Fluidised-Bed Combustor (PFBC) [14].

The alternative way of achieving direct firing is to operate a CFBC and a gasifier, which feeds fuel gas to a combined-cycle system and the char would be circulated to the CFBC for combustion, in order to exploit the available energy potential more fully. Hence the final efficiency will be raised. Improving the efficiency of a system reduces the fuel consumption as well as the production of pollutants which are released for a specific power output. It also has the advantage of minimising adverse environmental impacts along the whole fuel-supply train (i.e. mining, handling and crushing as well as storage and transportation).

Advanced oil-shale-based electric-power generation systems would offer the potential for the immediate future to meet the increasing energy demands by using the world's most-abundant fossil fuel. Such an approach has the advantage of preserving premium fuels, natural gas and mineral oil, for applications (e.g. in chemical industries) in which their natural advantages can be exploited more appropriately.

The future financial viability of oil shale as an energy source is uncertain because it is influenced by international crude-oil unit prices and security of supply. Indeed, the higher the unit prices and the tighter the supplies of crude oil and/or natural gas, the greater the interest in oil shale and other non-conventional energy resources.

Retorting and Gasification Processes

Oil shale retorting and gasification processes are quite similar. However, retorting aims primarily at producing the highest possible quantities of shale oil. Retorting is a thermal pyrolysis process, which takes place in a closed vessel (i.e. a retort), where the oil shale is heated, directly or indirectly, at a temperature of between 400 and 600 °C in order to extract its organic content as shale oil and fuel gas [15, 16]. The shale oil, which is usually highly viscous and has high contents of sulphur and nitrogen, depending on the quality of the feed stock, may be upgraded in a refinery to produce synthetic fuels or further processed to yield

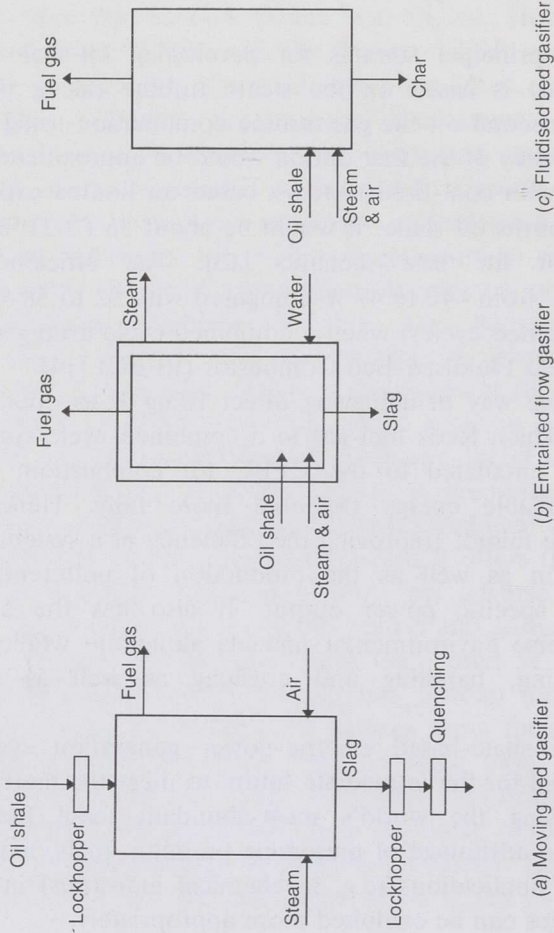


Fig. 1. Different types of gasifiers

chemical products. It can also be consumed directly, without any extra treatment, as a fuel for electric-power generation and industrial applications. Oil shale retorting is a well-developed technology: it has been used for decades in many countries (e.g. Estonia and China on a commercial scale and semi- or near-commercial scales in the USA) to yield shale oil [17, 18]. However, these methods tend to be inefficient with respect to liberating the organic content of oil shale. State-of-the-art technology can extract, on an average, between 70 and 80 % of the organic matter; the remainder being locked in the spent shale as a residual char [19, 20].

There are many indirectly-heated retorts, such as Lurgi [21, 22], Petrosix [23], Union [24, 25] and Fushun [26, 27], available in the international market. Any one of these could be employed in the proposed oil shale integrated scheme.

Oil shale was first gasified, in the early 1940s, in the former USSR in order to supply the city of Leningrad with about one million cubic metres (at atmospheric pressure) of gaseous fuel per day: it was required primarily for military industries [28]. Gasification of oil shale can be achieved in one of two ways: with the heat supplied directly (i.e. by partial oxidation of the feed stock) or indirectly (i.e. through an external heat-source and/or heat-exchanger). Direct heating is the basic process applied in pressurised coal-gasifiers, and oxygen is used to achieve the high temperatures required for efficient gasification. But such an operation is relatively costly, especially for commercial-scale plants. So, a promising opportunity for such an application is the fluidised-bed gasifier, which uses air instead of oxygen to produce a fuel gas with a relatively low energy content.

An important key feature of the indirectly-heated gasifiers, which operate usually at relatively low temperatures (i.e. 700 to 850 °C), is that they can produce a high-calorific gaseous fuel, without the use of oxygen, which is costly. However, oil shale gasification is still a complex task, because the desirable theoretical and experimental background is missing. Consequently, the oil shale gasification process is presumed (as a first approximation) to be similar to coal gasification, which has been studied for almost a century. Several coal gasification processes (i.e. moving-bed, entrained flow or fluidised-bed gasifiers) - see Fig. 1 - have been developed and reached industrial maturity, especially in the electric-power generation industry.

Direct Combustion

This offers an opportunity to generate electricity in commercial electric-power plants. In Estonia, the combustion of pulverised oil shale, has been used for electricity generation since 1924, when the Tallinn Thermal Power Plant (with a capacity of 22 MW) was modified to burn this fuel.

During the 1960s and 1970s, the Baltic Power Plant (1624 MW), and Estonian Thermal Power Plant (1610 MW) were commissioned [29].

The power plant required for the combustion of pulverised oil shale, in principle, is similar to that for pulverised coal, with slight modifications in feeding and ash-handling equipment. However, the combustion behaviour of oil shale is different from that of coal, because oil shale contains much more oxygen and sulphur than coal does. Also its mineral content is several times higher than that of coal and more ash is formed when oil shale is burnt. Experience in Estonia has indicated that pulverised oil shale burns faster than pulverised coal, because the heat generated during the combustion of the volatile matter of the oil shale is greater than that for the fixed carbon [30].

In employing pulverised oil shale for combustion, there are serious operational problems including the low availability of the boilers as a result of fouling and slagging as well as water and air pollution problems [31-35]. The average thermal efficiency of commercial pulverised oil shale plants is about 30 % (without flue gas clean-up) and they have an availability of only 50 % [36, 37]. In Estonia, the public and environmental groups have pressed the Government not to expand the electric power generation capacity until the present method of pulverised oil shale combustion is improved [38]. The common problems of fouling and corrosion of the heat-exchange surfaces by ash and slag deposits are enhanced in the presence of alkali metals, sulphates and chlorides in the raw shale. Usually the alkali metals and chlorides have higher corrosion activities compared with those of the sulphates [39]. Corrosion causes a sharp drop in the system's thermal-efficiency as well as its availability. This problem could not be mitigated by intense regular cleaning: whatever happens, the boiler's output will always be lower than its design value [40]. In general, such technology for firing solid fuels including oil shale has proved to be technically inefficient, economically unacceptable and environmentally disastrous [29, 41, 42]. Thus, it is hardly surprising that this technology has achieved a slow market-penetration only.

More stringent emission control requirements and the need to generate electric power and/or heat for industrial processes from fossil fuels (with higher efficiencies and lower costs) have led to new (to the power industry) technologies being introduced (e.g. fluidised-bed combustion). In 1960, Ruhrgas-Lurgi built two fluidised-bed units for the combustion of oil shale to generate steam and electricity. The unit capacity was 360 tonnes of oil shale per day, with the spent shale being used for cement production [43].

CFBC is an energy-efficient and environment-friendly technique for burning low-grade solid fuels, with high sulphur contents and low calorific values, including: tars; industrial, agricultural and municipal wastes; poor-quality coals [44, 45], and oil shale [46]. CFBCs are already

employed on a commercial scale, with a nominal system capacity up to ~400 MW_e (i.e. ~700 MW_{th}) [47]. However, at present CFBCs of approximately 300 MW_e, for steam production and electric power generation, can be found in the market with full commercial guarantees. There are approximately 25 countries using this technology for electricity generation, with additional countries focusing on this technique to solve environmental, waste and fuel problems. World-wide, there are more than 40 FBC boiler suppliers and more than 250 (representing over 50,000 tonnes/hour of high-pressure steam capacity or about 8500 MW of electric-power generating capacity) operating large units for electric utilities, burning different low-grade fuels including oil shale, and the demand is growing rapidly due to its good technical and environmental performances [48]. Other commercial applications of FBC are numerous, especially using refuse-derived fuels, biomass, paper-mill sludge and industrial wastes. For example, in Japan, there is extensive commercial experience in using FBCs for the incineration of municipal refuse and industrial wastes; there are already more than 113 small-size FBC refuse facilities [49].

In this system (i.e. a CFBC), oil shale is combusted in a bed of solids fluidised by high-velocity primary air. The off-gases and the entrained solids (e.g. fly ash) are separated in cyclones and the collected solids returned to the bed. Heat is extracted from the combustor (i.e. the furnace) and from the flue gases (before being cleaned up and released to the atmosphere via the stack), and used to produce superheated steam which will drive a conventional (i.e. condensing or back-pressure) steam turbine to generate electricity [50-56].

Tests performed indicate that a CFBC is capable of burning high ash as well as high sulphur content oil shales successfully, with high combustion (i.e. > 98 %) and boiler (i.e. > 80 %) efficiencies [23, 57, 58]. It is concluded that emission rates from CFBCs are low and there is no need for flue gas clean-up downstream except for particulates, which could be collected easily by employing approved systems such as bag filters or electrostatic precipitators [59, 60]. In reality, there are only two CFBCs (i.e. a demonstration co-generation plant in Israel, with an installed capacity of 50 tonnes of steam per hour [1] and a full commercially operating unit in China, with a steam flow of 1750 tonne per hour [48]) fuelled by oil shale. Experience gained in these countries that concerns electricity generation by using oil shale, which contains carbonates (i.e. CaCO₃ and MgCO₃) in its inorganic part, showed that a CFBC is suitable for burning sulphur-rich (e.g. Jordanian shales) and low-grade oil shale [60]. Due to the encouraging results and experience gained from a demonstration plant, the board of directors of the Israel Electric Corporation Ltd. approved a project to build, during the period 1997 to 1999, a commercial oil-shale-fired electric-power station with a

nominal installed capacity of 75 MW [13]. Gasification is also an acceptable technical solution for processing sulphur-rich oil shales: it is even more economic than the direct combustion of the shales [58]. Thus, due to its relatively high sulphur content, the gasification of Jordanian oil shale tends to be cheaper and technically more viable compared with oil shale retorting.

The Proposed Oil Shale Integrated Tri-Generation System

This system (i.e. OSITGS) is similar in some respects to coal integrated-gasifier gas turbine (e.g. the British Coal Topping-Cycle) systems, which incorporates coal gasification and combustion. But the entrained solids and/or the bottom ash from the CFBC are used as a heat carrier either to the gasifier or the retort (to produce synthetic gaseous and liquid fuels), which are added to the OSITGS process in order to achieve a high

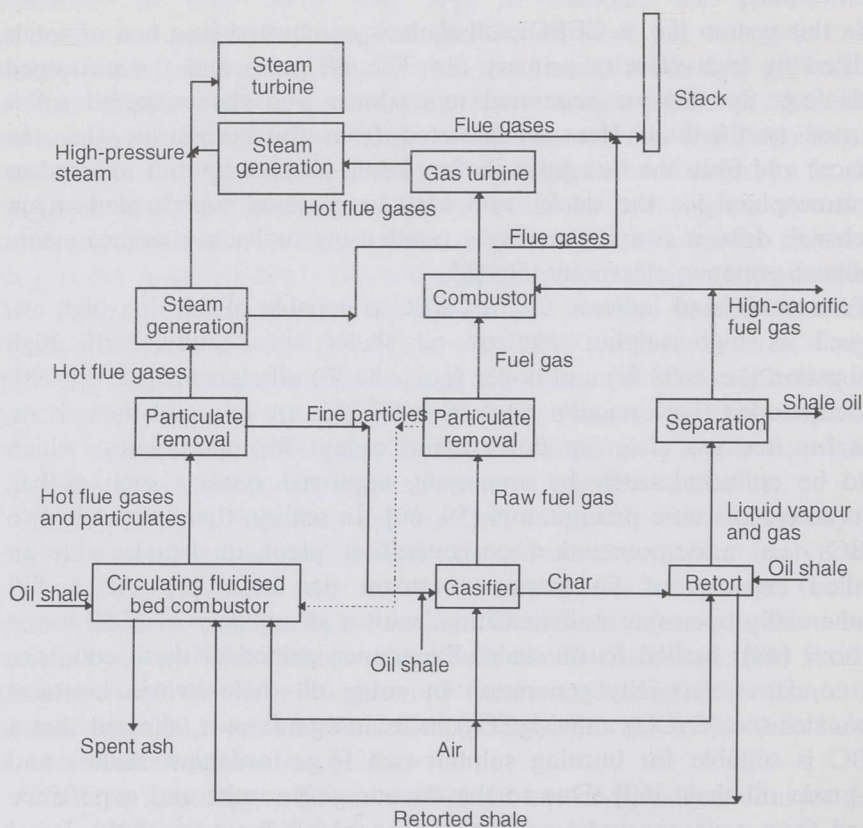


Fig. 2. The tri-generation scheme (with an indirectly-heated gasifier)

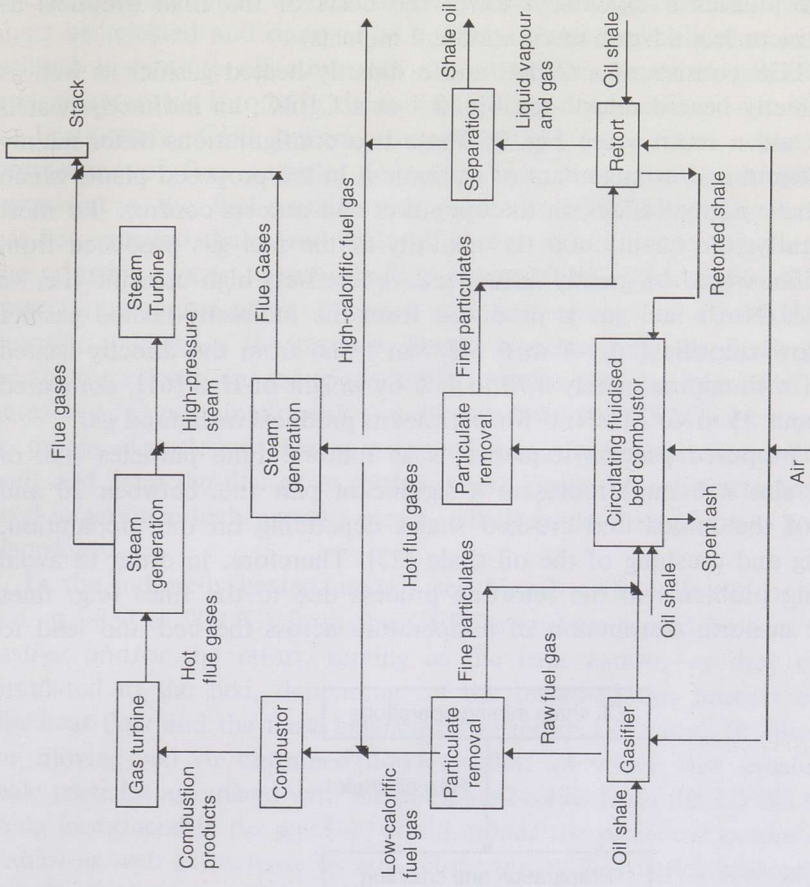


Fig. 3. The tri-generation scheme (with a directly-heated gasifier)

resource-utilisation efficiency, lower the costs of the final products as well as incur less adverse environmental impacts.

OSITGS consists of a CFBC and a directly-heated gasifier as well as an indirectly-heated retort -see Fig. 2 - or a CFBC, an indirectly-heated gasifier and a retort - see Fig. 3. These two configurations differ mainly in the layout and arrangement of equipment in the proposed plant, which would have a great effect on the operation and process control. But most importantly, the quality and the quantity of the fuel gas produced from the gasifier would be greatly influenced. A relatively high-calorific (i.e. ~ 8 to 10 MJ/Nm^3) fuel gas is produced from the indirectly-heated gasifier and a low-calorific (i.e. ~ 4 to 6 MJ/Nm^3) one from the directly-heated gasifier, with approximately 0.75 to 1 % by weight of H_2S [61], compared with about 35 to 40 MJ/Nm^3 for petroleum products or natural gas.

The proposed process sequence is as follows. Fine particles (i.e. of average size $< 6 \text{ mm}$) represent a significant part (i.e. between 20 and 30 %) of the mined and crushed shales depending on the preparation, handling and crushing of the oil shale [23]. Therefore, in order to avoid operating problems in the retorting process due to the fines (e.g. fines prevent uniform distribution of temperature across the bed and lead to

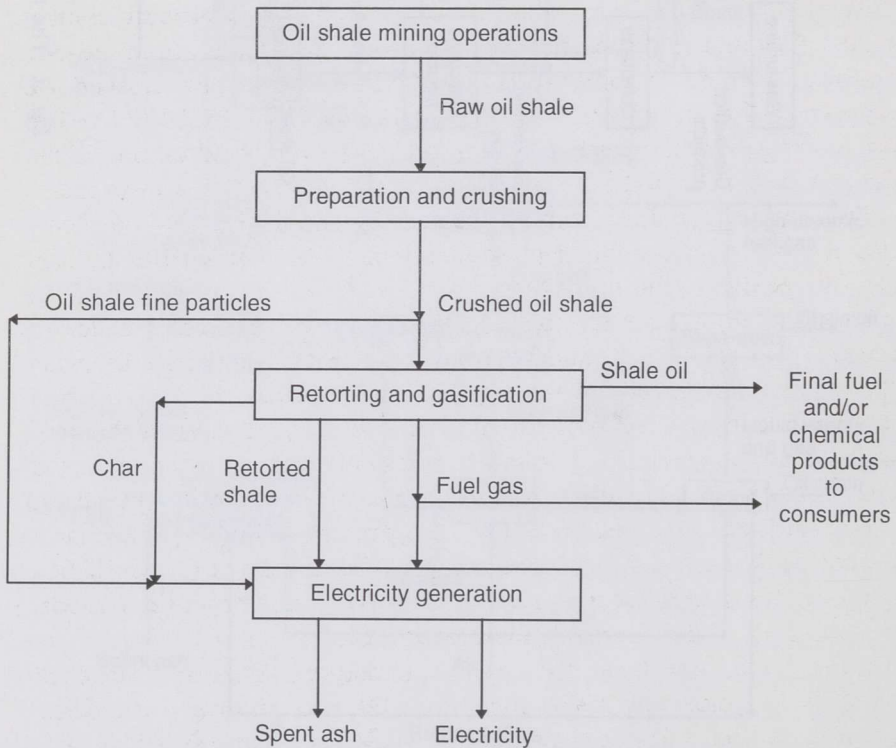


Fig. 4. Flow chart of the proposed process

excessive entrainment of dust in the product fuel gas stream) [62], they must be rejected and dumped as a waste. Instead of this, they could be utilised in a more effective way. An attractive means to harness their energy potentials is to burn them directly in a fluidised bed - see Fig. 4.

Fortunately, CFB boilers are capable of firing fuels with a high percentage of fine particles [63, 64], and the combustion efficiency increases as the fuel-particle size decreases [65]. Also char from the gasifier, retorted shales and shale oil sludge, which are by-products from the retorting process, can be burned in the CFBC. This will increase the resource usage and plant efficiency as well as reduce the environmentally negative impacts of the process. These, together with the finely-ground oil shale particles, are fed to the CFBC, where they are burned. The flue gases and the entrained solids (i.e. fly ash from the CFBC) are separated by means of cyclones. Heat is extracted from the furnace (i.e. a fluidised bed) and from the flue gases (before being cleaned up downstream), and used to produce high-pressure steam, which drives the turbine to generate electricity.

In the indirectly-heated mode - see Fig. 2 - the collected hot solids (i.e. at ~850 to 900 °C) from the CFBC's cyclone would be sent to the gasifier and/or the retort, serving as the heat carrier, or they can be circulated to the bed, depending on the instantaneous process control (for heat flow and the mass balance). The indirectly-heated fluidised-bed (or moving-bed or entrained-flow) gasifier, in which raw crushed oil shale particles are mixed with the circulated solids from the CFBC before being introduced to the gasifier, would reduce the projected output of the gasifier as well as increase its size. However, with a better arrangement, the fluidised-bed gasifier can be heated by employing in-bed heat-exchanger tubes, while the heat source is still the hot CFBC fly-ash. Shale oil, produced in the retort, has a high density as well as high nitrogen and sulphur contents, could be fed to the gasifier in order to produce a fuel gas of high quality. After gasification reactions take place in the reactor, a relatively high-energy gas will be produced.

In the directly-heated mode operation - see Fig. 3 - an air-blown, pressurised or atmospheric, fluidised-bed gasifier would be used as in the ICGCC. Raw crushed oil shale is fed to the gasifier, where it is pyrolysed (i.e. devolatilised) at a relatively low temperature, thereby producing a low-calorific fuel gas and char. The latter will be circulated to the CFBC. This mode has a more logical lay-out than the indirectly-heated mode. Hence the flow of the process is more acceptable.

The raw fuel gas from the gasifier undergoes an initial stage of cleaning in a cyclone or high-temperature filter to remove particulates. Then, it is cooled, via a heat exchanger, where the heat is used to raise more steam for feeding the steam cycle, between 400 and 600 °C, in order to reduce its alkali salts (i.e. potassium and sodium compounds)

content to meet the gas turbine requirements. Finally, the clean fuel gas is burnt in an advanced gas turbine combustor. The resulting hot combustion products are passed to the turbine expander, which drives both the compressor and an electric-power generator. The heat from the high-temperature exhaust gases (i.e. at >500 °C) is recovered by a waste-heat boiler, which will produce steam that can be used to drive a steam turbine.

Residual char from the base of the gasifier, together with the fines collected from the fuel-gas stream, are taken to the CFBC, where they are burnt to release heat. The retorting and/or gasification residues have low volatile-matter contents, high sulphur content in various forms such as unreacted sulphur and calcium sulfide and unreacted calcium oxide, and hence low reactivities as well as high ignition temperatures. These residues can be burnt in the CFBC with high combustion efficiencies of at least $95 (\pm 3)$ %. This rate can be improved to reach 99 % when the residues are mixed with raw oil shale fine particles [66]. Similarly, coal-gasifier residues can be burned successfully in a CFBC with only a low rate of generation of environmentally contentious species, giving a combustion efficiency of 99 % and a sulphur-retention effectiveness of 96 % [67].

In the retort, the crushed raw oil shale is mixed with the hot ash, which carries the required heat for the pyrolysis process, from the CFBC or the indirectly-heated gasifier, and introduced to the retort. Such a configuration has been investigated in China. However, the fine particles elutriated and entrained by the flue gases from the CFBC were collected and returned to an auxiliary bed for burning [68]. The produced liquid-vapour and hydrocarbon gases would be collected in the upper part of the retort, cooled and separated. The bottom ash (i.e. the spent shale) will be conveyed to the CFBC in order to recover the energy content from the residual char in the retorted shale. Finally, the CFBC bottom ash would be either disposed of, after being cooled, as solid waste, or processed to recover some of the trace metals because they have become concentrated in the spent shale: the eventually-produced ash can be used as a raw material in the building and construction industries. Such an approach will enhance the economic feasibility of oil shale harnessing.

The design of different components of OSITGS will be based on proven technologies especially for the retort, gas turbine, waste-heat recovery boilers, ash cooler and CFBC. But, for the gasifier, more experimental information is required. However, in the mean time, experience from coal and biomass gasification processes is used due to the lack of information about oil shale gasification. Nevertheless, experimental work concerning different gasification processes, hot fuel gas clean-up and the gasifier's char combustion tests in a CFBC should be undertaken to assess the key parameters which influence the

availability and efficiency of the cycle. Moreover, it will be necessary to ensure that the gas turbine has the ability to burn the produced fuel gas from the gasifier with an acceptable efficiency and a low emission rate. Preliminary experimental work regarding retorted shale combustion tests in a FBC has been undertaken in a testing facility at the Japan National Oil Corporation: there is a *prima facie* case that it can be burnt with relatively high efficiencies and low emission rates [66].

Oil shale integrated tri-generation system will offer the advantage of a higher efficiency over systems employing the standard steam cycle. The projected cycle efficiency would be higher than 45 % compared with ~36 % for conventional CFBC and about 30 % for the pulverised oil shale combustion system. Consequently, this will lower the cost of electricity generation compared with what can be achieved with conventional technologies. In addition, lower rates of emissions per unit of electricity generated would be released to the environment. The high efficiency of the OSITGS is mainly due to the low energy-loss rate in the cycle as a direct result of the integration of different processes leading to greater thermodynamic efficiencies and the relatively high efficiency of the combined cycle.

Performance of the Integrated Tri-Generation System

The design of this newly proposed integrated system will be based on proven technologies, such as those associated with oil shale mining and retorting, gas turbines, waste-heat recovery boilers, CFBCs and ash cooling and disposal. However, in the case of gasification, more experimental information is required because it is believed that the gasifier would play a major role in the integrated cycle. In the mean time, experience gained from the employment of coal and/or biomass gasification processes could be used. It is predicted that the directly-heated mode of this scheme will have a high cycle-efficiency and a low cost for the generated electricity.

Expected Benefits

The proposed tri-generation scheme would possess advantages compared with employing either the direct combustion or retorting of oil shale alone. The most important are:

1. The plant could be located near oil shale deposits and in remote areas, because final products (i.e. electricity as well as gas and liquid fuels) can be readily transported.
2. Gaseous and liquid fuels with high calorific values can be produced: these could be substituted for petroleum products and/or natural gas in different sectors.

3. The resulting low-calorific fuel gas can be cleaned easily and burnt in a combined-cycle gas turbine to generate electricity at relatively high thermal efficiencies (e.g. ~45 %). This would eliminate the need for expensive treatment facilities, and consequently the cost of electricity generation would be reduced.
4. Total water consumption is expected to be less, because wastewater from one process can be used as the feed water for another (e.g. boiler blow-down water can be used for cooling and moisturising spent ash and/or in oil shale mining operations to reduce dust generation).
5. Waste heat at different points of the cycle will be recovered: this permits the generation of more electricity with little additional equipment.
6. The cycle is expected to be more energy-efficient, and so the associated adverse environmental impacts are expected to be less.
7. It is financially attractive, because it allows high recovery ratios for the organic content of the oil shale. Hence the cost of the final products would be less, as well as the cost of the adverse environmental impact mitigation measures.

Such an integrated scheme for utilising oil shale resources appears to be promising for electric power generation from this solid fuel. However, it will require a high capital investment. But, in the long-term, it should achieve better technical and economic performances compared with those for the direct combustion or retorting processes of the oil shale when employed on an individual basis.

Expected Operational and Technical Problems

Aside from the choice of the gasification and/or the retorting technology, some of the key issues, which influence the performance of the proposed system, are: fuel gas clean-up, the integration between the fuel's production processes, the choice of the electric-power plants and the suitability of commercial gas turbines for burning the relatively low-calorific fuel gas derived from oil shale.

Retorting and Gasification

Oil shale retorting technologies, unlike gasification, have been used in Estonia, China and the former USSR for many decades. But the employed methods are relatively inefficient with respect to the oil-recovery ratio (i.e. ~75 (± 5) % of the organic content of oil shale). This would increase the cost of the final products as well as the related environmental protection measures, when employed as stand-alone processes. There are many retorting methods (e.g. Lurgi, Petrosix and Union-B) available in the international market that use hot solids as the heat carrier, and could be employed in the proposed oil shale integrated

system. However, careful behavioural analysis should be carried out before selecting any of these methods, because the technical-economic performance of any retorting process depends on the chemical and physical characteristics of the oil shale deposit.

Pressurised gasification is preferred to atmospheric gasification from the thermodynamic losses point-of-view. These losses, associated with compressing the fuel gas before it is injected into the gas turbine combustor are higher than those associated with compressing the fluidising air. But this benefit would involve operational problems, such as feeding the raw oil shale to the pressurised reactor, and losses of inert gas if a lockhopper mechanism is used as the sealing device [69]. The commercial-scale pressurised fluidised gasifier is now available and being used in ICGCC systems. These could be employed in the proposed cycle, but assessments of several fundamental issues, such as oil shale feeding, gasification kinetics and fluidisation characteristics are needed.

Fuel Gas Cleaning

The use of a combined-cycle system, which employs a combination of gas and steam turbines to generate electricity, will impose constraints on the permitted levels of particulates, alkali metals, sulphur and tars in the fuel gas delivered to the gas turbine combustor. Otherwise, excessive corrosion and erosion of the gas turbine blades will occur: the turbine efficiency will be decreased, blade durability will be reduced and the operation of the turbine would eventually become unsafe [70]. Also, the heat-recovery boiler for the hot exhaust-gases exiting from the gas turbine, which is used to generate steam, would suffer from severe corrosion, due to the formation of sulphuric or hydrochloric acids, as a result of condensation in the heat-recovery system. The levels of such contaminants that can be tolerated by a gas turbine or the combined-cycle combination are not well established because no such oil-shale-fired plant exists as yet.

Manufacturers of gas turbines have specified strict limits for contaminants in the fuel gas: in addition to those previously mentioned contaminants, these include nitrogen compounds derived from nitrogen in the oil shale. Particulates, even in relatively small amounts, can cause turbine blade erosion. Thus, stringent limits for particulates are imposed by manufacturers (e.g. General Electric, GE, specifications for its turbines, both industrial and aero-derivative, require a total concentration to be below 600 ppmw for particulates less than 10 μm maximum diameter, 6 ppbw for particulates between 10 and 13 μm and 0.6 ppbw for those larger than 13 μm). In other words, GE requires a total concentration below 1 ppmw at the turbine inlet, with 99 % of the particles less than 10 μm diameter [71]. This corresponds to a particulate concentration in the fuel gas of about 5 ppmw. However, particulate

concentrations in the raw fuel gas from most gasifiers will range approximately from 5000 ppmw to 10,000 ppmw or more in the case of biomass [72]. But for oil shale, due to its high content of inorganic matter (i.e. ~70 % ash), it is predicted that such a level is much higher and may be more than twice these rates.

In order to meet the requirements of the manufacturers and to protect the gas turbine from damage as well as the environment, it is necessary to remove almost all the particles from the fuel gas stream before being introduced to the gas turbine combustor. This can be done by employing proven systems such as high-efficiency cyclones, ceramic-candle filters or rigid ceramic-filter elements, ceramic fibre blankets and advanced particle filters [73, 74]. These are used for similar purposes in coal and biomass gasification processes as well as pressurised fluidised-bed combustors [75-77]. Gas clean-up of particles is expected to enhance the economics of the power plant. Savings are anticipated to be derived from: higher cycle-efficiency and increased electric power generation, elimination of the need for conventional filters downstream before the flue gases are released to the atmosphere, increased gas turbine availability as well as reduced maintenance especially of the turbine blades, and the reduced size of the waste-heat recovery boiler and auxiliary systems [78]. Flue gases emerging from a CFBC do require the use of particulate capturing or cleaning equipment such as bag filters, as in other combustion systems in order to achieve a low stack-opacity (i.e. < 10 %). The collected particulates could be conveyed to the bed in order to exploit their energy content or mixed with ash for disposal [79].

During combustion and/or the gasification of oil shale, the alkali metals such as sodium and potassium are vapourised, released and may condense on heat-transfer surfaces and other parts of the system. Alkali metals corrode these surfaces as well as the gas turbine blades, when such a fuel gas is used, as the prime fuel, in a combined cycle system [80]. Gas turbine manufacturers specify the maximum allowable concentrations of such materials should not exceed 8 to 12 ppbw in the combustion products: this corresponds to approximately 40 to 60 ppbw in the fuel gas [71].

For the efficient removal of these metals, the key issue is cooling (to about 350 to 400 °C before particulate removal may be effective) the fuel gas or flue gases in the presence of liquids or solids (e.g. ceramic filters) on which the condensed vapours can be deposited and removed from the gas stream. A fixed granular-bed of activated bauxite sorbent, which is used to control alkali vapours in the PFBC's flue gas could be employed here for the same purpose [81]. Wet scrubbing may be used: this would result in the complete removal of alkali salts, but this consumes a lot of fresh water. The latter technique is considered to be an expensive solution because of the shortage of freshwater supplies locally. Jordanian

oil shales are free from Cl and have, on average, relatively low alkali contents (i.e. < 0.4 % of Na₂O and 0.6 % of K₂O by weight) in their inorganic component [82, 83]. However, the P₂O₅ content is relatively high (i.e. 1.5 to 3.5 % by weight) [84]. But, in spite of that, the ash composition does not lead to the phenomenon of ash softening at temperatures used in the gasifier or CFBC [1].

Tars, which are usually formed during the gasification process of oil shale may account, on average, and depending on the temperature, for between 0.5 and 1.5 % by weight of the produced gaseous fuel from a typical fluidised-bed gasifier [85]. When tars condense on cool surfaces, severe operational problems may occur, such as clogged fitters, pipes and valves. Thus, a separate reactor with a catalyst should be placed immediately after the gasifier [86]. This will lead to the production of a gas with relatively low levels of tar.

Sulphur content, on average, of the Jordanian oil shales is approximately 3 % by weight (i.e. 7 to 9 % of the organic matter content) [84]. When such an oil shale is processed (i.e. retorted or gasified), the produced shale oil or hydrocarbon gas will have a high sulphur content (e.g. ~10 % by weight of the produced shale oil) and so must be treated before being used as a commercial fuel. This would increase the upgrading and utilisation costs. Nevertheless, such a high concentration of sulphur might make its recovery, as a by-product commodity, financially viable. However, in the case of direct combustion, when a CFBC is employed, and gasification of oil shale due to its high Ca/S molar ratio (i.e. ~4.5 for Jordanian shales) would make in-bed effective sulphur retention (i.e. >95 %) occur during combustion: this rate is equivalent to a SO₂ concentration of less than 30 ppm or 260 mg/MJ [87], and most of the sulphur would otherwise be released to the fuel gas. It is reported that shale is more effective than typical limestone (which is added to coal-fired and/or gasification systems) as a sorbent, and the oil shale's own sulphur is captured more readily because of its inherent dispersion in the mineral matrix which contains the calcium-based compounds [61, 88]. Field experience has shown that the bed temperature has the dominant influence on emissions performance and it should be kept below ~900 °C in order to optimise the general performance, unit efficiency and control of both SO₂ and NO_x emissions [89].

The sulphur content of the fuel gas has little impact on the corrosion rate of the turbine, so it is not limited by turbine protection standards. However, sulphur oxides (SO_x), which are produced, will affect downstream heat-recovery equipment. Hydrogen sulphide (H₂S) and carbonyl sulphide (COS) can be removed from the fuel gas efficiently by using sulphur sorbents (e.g. zinc ferrite and zinc titanate), but these are expensive and must be regenerated. The sorbent regenerator produces a

gaseous sulphur stream that is converted either to elemental sulphur or sulphuric acid (H_2SO_4). The latter is easier and cheaper to produce, but more difficult to store [90]. Such by-products could be sold to the chemical industries (e.g. to produce ammonium sulfate fertiliser), thereby improving the plant's financial performance. Alternatively, dry desulphurisation by using the available spent ash (i.e. from the CFBC and/or retort) or pulverised limestone (i.e. CaCO_3), which would be injected to the fuel gas piping system, could be used to capture sulphur. In this step, which is required to achieve the desired sulphur-emissions limit, the use of spent ash as a sorbent would enhance the economics of the proposed integrated oil shale plant [91]. Similarly, pulverised alkali-sorbent particles (e.g. emathlite and bauxite) may be injected into the fuel gas to remove alkali vapours within a filter vessel.

Nitrogen oxides can be produced in both the CFBC and the gas turbine combustor from nitrogen in the combustion air (i.e. thermal NO_x) and from the compounds produced during gasification or retorting from nitrogen in the raw oil shale (i.e. fuel NO_x). In a CFBC, three nitrogen oxide species are emitted in significant quantities. These are NO (nitric oxide), NO_2 (nitrogen dioxide) and N_2O (nitrous oxide). The first two are classified under the general label of NO_x ($= \text{NO} + \text{NO}_2$). These are formed during the burning of oil shale particles, by either

- (i) thermal oxidation at high flame temperatures of the nitrogen in the combustion air (i.e. thermal NO_x , which depends strongly on the temperature and can be controlled by reducing the combustion temperature) and/or
- (ii) the oxidation of the organic nitrogen in the fuel (i.e. fuel NO_x , which depends on the amount of oxygen available in the flame), which can take place at lower temperatures.

Thermal NO_x formation as a result of the combustion of relatively low-calorific fuel gas in a gas turbine combustor is likely to be very low due to the lower flame temperatures: this is particularly so for gas produced from the directly-heated gasifiers [92]. When burning synthetic fuels (i.e. shale oil and/or synthetic gas) or raw oil shale, NO_x emissions are produced principally as a result of the high nitrogen content of the initial shale used. Thus, fuel NO_x predominates in fluidised-bed and gas turbine combustors [93]. NO_x emissions from a CFBC can be controlled by injecting ammonia at the furnace exit [61]. The low-calorific fuel gas generated by the directly-heated gasifier should give low thermal NO_x concentrations when burnt.

Nitrogen content in oil shale may be as much as 3 % (by weight) of the organic matter [84]; when oil shale devolatilises, in the gasifier or CFBC, this nitrogen is distributed between the char and the volatiles. Combustion of char nitrogen proceeds proportionally at approximately the same rate as the carbon, resulting in the formation of nitric oxide

(NO). Volatile nitrogen is released or decomposes in the gas from ammonia (NH_3) and hydrogen cyanide (HCN) [94]. However, destruction of nitrogen oxides is a complex process involving various reactions, and it is difficult to describe and/or estimate the NO_x emissions by simple expressions. Wet scrubbing, directly after the gasifier, can remove ammonia completely from the fuel gas, but there is a thermodynamic penalty as well as the requirements for additional quantities of freshwater and wastewater treatment. Also catalytic oxidation (e.g. selective catalytic reduction, SCR) of NH_3 at elevated temperatures could be used for the same purpose [93].

N_2O emissions have been received more attention recently, because this gas contributes significantly to the greenhouse effect as well as destroying the ozone layer in the stratosphere [95]. Such a gas is many times more powerful than CO_2 as an absorber of infrared radiation. Hence there is an increasing interest in its presence in the atmosphere. Low N_2O emissions are achieved by raising the temperatures and decreasing the excess air percentage; high temperatures lead to the thermal decomposition of N_2O . Thus, the same factors that promote low NO_x emissions promote high N_2O emissions, so there is a trade-off between these two pollutants. However, air staging (i.e. part of the combustion air is introduced in the form of secondary air at later stages in the combustion process, while the primary air flow is reduced to obtain sub-stoichiometric conditions in order to create reducing conditions in the bed which increases NO reduction, and simultaneously maintains the fluidising velocity), which is used in the CFBC, can reduce the rate of N_2O production without increasing the rates of emissions of other pollutants (i.e. NO_x and SO_x) [96].

Suitability of Commercial Gas Turbines for the Produced Fuel Gas

Oil-shale-derived fuel gases possess relatively low energy contents of approximately one tenth of the calorific value of natural gas or petroleum products, for which most gas turbine combustors have been designed. So, with gasified oil shale, combustors must accommodate larger volumetric flows of gas, which are injected into the combustor through a nozzle originally designed for a higher-quality fuel with a much higher energy content, in order to achieve an equivalent output. However, several different industrial gas turbines have operated successfully on low-calorific fuel gases (e.g. off-gas from blast furnaces with $\sim 3 \text{ MJ/Nm}^3$). Moreover, based on the experience gained from coal- and biomass-derived fuel gases, the development of the integrated combined cycle and the relevant modifications to conventional gas turbine combustion technology have allowed such low-calorific gases to be burnt successfully [11, 71, 92, 97]. GE carried out successful combustion studies of low-

calorific fuel gases using its gas turbine combustor designs. These indicated that a gas, having a calorific value as low as ~ 3.7 MJ/Nm³, could be burnt with relatively good results, providing there is some hydrogen in the fuel gas [98]. A fuel gas derived from the gasification of Jordanian oil shale is expected to have a calorific value of ~ 5 (± 1) MJ/Nm³ and a hydrogen content in the range of 15 to 20 % by volume, but a relatively high content of H₂S [28]. This should be removed from the fuel gas before it is burnt in the combustor. Thus, there is unlikely to be any problem with the combustibility of such a gas in gas turbine combustors.

Another serious issue is related to the increase in mass flow through the turbine expander, when such a low-calorific fuel gas is used rather than natural gas. All gas turbines operate under shock flow conditions at the expander inlet: the large mass-flow rate can be accommodated only by increasing the turbine's inlet pressure or decreasing the temperature. Such actions would reduce the plant's efficiency. Higher turbine inlet pressures result in increases in the compressor's pressure ratio and move the compressor towards its stall limit. In order to avoid such harsh operating conditions, the compressor's outlet can be bled to provide the air required for fluidisation. Because the mass flow of air needed for the gasifier almost equals the fuel gas flow, the mass flows through both the turbine and the compressor would be almost the same (with a small difference resulting in only a marginal increase in the pressure ratio), so there is a little concern about the compressor stall. However, in the case of atmospheric fluidised gasification systems, there is no need for high-pressure air and such an issue is still more critical.

Lastly, the use of low-calorific fuel gas, as the prime fuel for a gas turbine, will require some modification to the turbine. Nevertheless, there are many plants operating around the world using coal- or biomass-derived gaseous fuels with relatively low energy contents [99]. A similar approach and technologies may be used to fire fuel gases derived from oil shale.

Integration of Gasifier, Retort, Combustor and the Combined Cycle

Many important opportunities arise when integrating gasification, combustion, retorting and the combined cycle in one plant (i.e. the proposed OSITGC system). Fuel gas produced by the gasifier must be cooled to about 350 (± 50) °C to permit the effective removal of particulate and alkali compounds: this allows the production of more high-pressure steam, which would be used either for the process itself or for generating more electricity. Further cooling of the fuel gas can be carried out by exchanging heat with the raw oil shale, before the latter is introduced to the retort. The fluidised-bed combustor and gasifier require

a substantial amount of high-pressure air to achieve bed fluidisation. This could be provided from the gas turbine compressor (which is more efficient than conventional industrial compressors); by bleeding fluidisation air from the compressor, stall can be avoided.

In a gas-turbine-based combined-cycle power plant, the air compressor, the gas and steam turbines, various pumps, valves and other components are standardised equipment, which are available in various standard sizes. A waste heat-recovery boiler is the only key component. It should be noted that the hot-gas piping and control valve technical feasibility are issues of concern. Piping systems for high temperatures exceeding 900°C are feasible, though close integration of the plant's components is required to reduce both the capital and operating costs. Also, hot valves for temperatures up to ~700 °C are currently available. However, for higher temperatures, such valves are still under development [90].

Residual carbon from the gasification process, as well as the fines collected from the fuel gas stream, tar, shale oil sludge and the retorted shale from the bottom of the retort together will be taken to the CFBC, where they are burnt to raise heat. Consequently, more steam and electricity are generated, while simultaneously less pollutants are released into the environment. Also, spent ash from the bottom of the CFBC and/or retorted ash, which would be injected to the fuel gas piping system, could be used to capture the sulphur. The use of spent ash as a sorbent would improve the environmental performance as well as the economics of the proposed integrated plant.

In summary, the OSITGS is somewhat similar to that for coal-fired ICGCC: proper CFBC, gasifier and retort integration is the key to achieving high overall efficiencies.

Conclusions

Oil shale together with coal are predicted to be major sources of primary energy during a significant part of the twenty-first century, because other fuels will become scarcer and their unit costs will increase. The dominant uses of oil shale are likely to be for electricity generation and/or synfuel production. Thus scientifically novel technologies for utilising oil shale should be developed to meet the growing requirements for energy and in particular electricity.

An oil shale integrated tri-generation system comprises of a gasifier, which produces a fuel gas and char residue, a CFBC and a retort. The produced fuel gas or part of it, after cooling and cleaning, is fired in a combined cycle, whilst the char is burned in a CFBC to raise steam for the steam cycle. Also, the synthetic gases and/or liquid fuels produced

would have relatively high calorific values, so this will make it financially feasible to store and transport these fuels over long distances.

Significant developments are still required for both the system and its components. Thus a development strategy should be produced which will identify the need for a component development phase leading to a pilot or prototype integrated oil shale plant before moving to the commercial scale. Both the gasifier and hot fuel gas clean-up, as well as the process control of the proposed cycle would represent the main areas requiring technical improvements. Assuming the successful development of the main components of the proposed system, the commercial plant based on the multi-purpose production approach can offer significant advantages in efficiency and cost over conventional oil-shale-fired power plants, as well as lower rates of emissions to the environment. Developing an oil shale integrated tri-generation system would ensure that Jordan will have cost-effective and environmentally acceptable options for supplying its fuel and electricity needs for many decades to come.

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