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RESEARCH AND DEVELOPMENT IN OIL SHALE COMBUSTION AND PROCESSING IN ISRAEL

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Methods of the combusting and processing of Israeli oil shale (higher heating value 1150 kcal/kg, oil yield 5-6 %), and shale oil (sulphur content 5-7 %) are described. Oil shale is the only source of energy and only organic natural resource in Israel. Its reserves are of about 12 billion tons.

Introduction

Oil shale is the only source of energy and only organic natural resource in Israel. Large deposits of high-sulphur oil shales are widely distributed throughout Israel (Fig. 1). The main deposits are concentrated in the northern Negev desert, where nearly half of the total reserves (12 billion tons) are located. It is equivalent to 600 million tons of oil. At the present level of the liquid fuel consumption (ca. 8 mln.t/year) it will be enough to meet the requirements of the country for more than 80 years. The Rotem Yamin deposit is the largest and the most investigated. The shale beds overlie the phosphate-rich rocks. The thickness of the shale bed ranges from 35 to 80 m.

The average organic content for the Rotem-Yamin deposit is 14-18 % and oil yield 60-71 l/t, based on Fischer assay. Economically, the relatively low grade of Israeli shale may be compensated by the easy accessibility of the deposits in the phosphate mining areas, where low-cost, large-scale, open-pit mining techniques may be applied. Oil shale price is low - about $4 per ton.

The characteristics of the Rotem Yamin shale as compared to the shales of some other important deposits of the world are shown in Table 1. As seen from these data, the oil yield and heating value of Israeli oil shale is lower than that of other shales. Kerogen and shale oil contain a considerable amount of sulphur. These features of Israeli oil shale require special methods for its processing and combustion. Many countries, such as the USA, Austria, Russia, Morocco, Jordan, Syria, Uzbekistan, etc., have deposits of high-sulphur oil shales. Technology of processing of high-sulphur oil shales and utilization of their chemical potential is a very important problem [1].

Since 1981, Israeli companies with the Government support have been carrying out a multi-year program of using oil shale.

Table 1. **Characteristics of Oil Shales**

Oil shale	LHV, kcal/kg	Organic matter content, %	Fischer assay, oil yield, %	Sulphur content of oil, %
Israel	700	14.3	6.2	7.3
Estonia	2600	31	20	0.7
USA	800	15	10.4	0.7

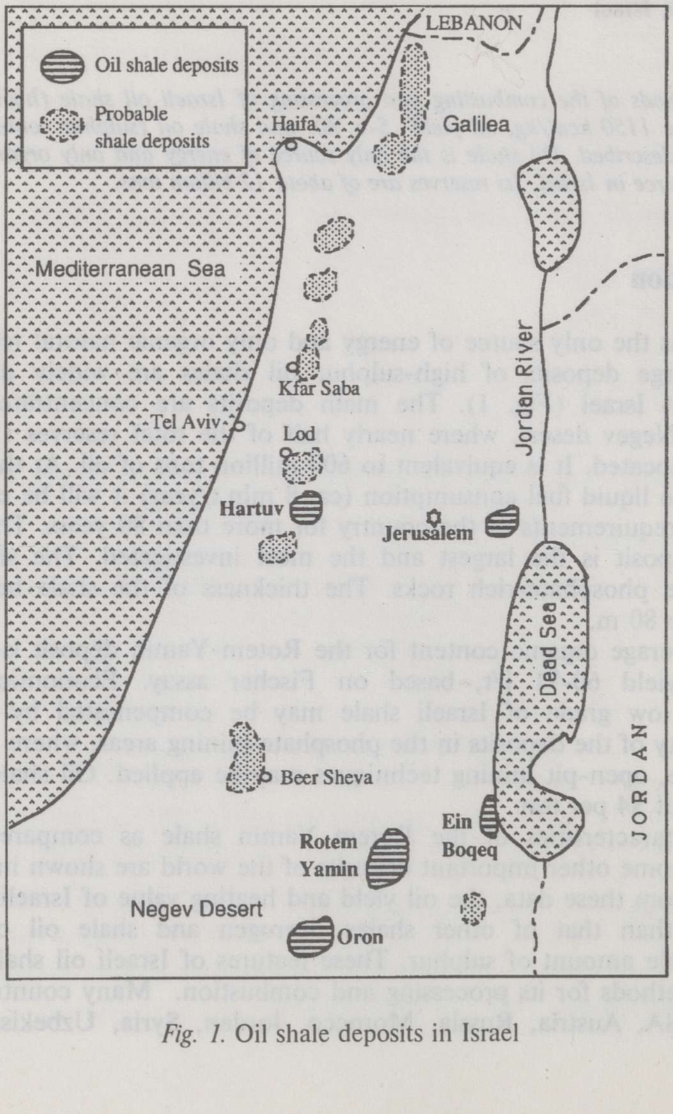

Fig. 1. Oil shale deposits in Israel

Oil Shale Energetics

Oil shale can be an effective source of power production. High-capacity oil shale power plants operate many years in Estonia, Russia, and Germany. Since 1989, a 25 MW demonstration plant for power generation has been operating successfully also in Israel. The boiler for this plant was constructed by the Ahlström Oy (Finland). Oil shale is combusted in a bubbling fluidized bed at a temperature of 800-850 °C. The boiler is characterized by the following data [2]:

Oil shale consumption	55 tph
Steam (44 bar, 480 °C) production	50 tph

Oil shale, crushed to a particle size of 6-12 mm, is conveyed to the feed bin designed to provide three hours of the full load work. From the bin, oil shale is fed by four mechanical feeders to the boiler combustion chamber. Each feeder is designed for a 50-% load of the boiler. Oil shale quantity is controlled by the boiler load. The boiler is also equipped with two fuel oil burners to provide stability of the shale combustion at low load values. They are situated above the fluidized bed. There are also four lances situated within the bed.

Feed water goes from the deaerator to heat exchangers in the ash cooler, then passes through an economizer into the upper drum of the boiler. Steam production takes place both in tubes within the combustion chamber and in the back pass (boiler bank).

Streams of primary and secondary air are heated by flue gases. Flue gases pass through two parallel cyclones. Solid particles are recycled to the combustion chamber, and gas exits from the cyclones to the back pass which is similar to those of conventional boilers. Volatile ash is retained mainly by the bag filters. Heavy solid particles of the ash are removed from the combustion chamber and cooled in the ash coolers by the air. Ash is directed by transporters to the bunker, and then removed by dump-cars to the place of disposal.

The steam from the boiler is directed to a turbine with a nominal capacity of 6.3 MW. The steam leaving the turbine is used in industry.

Experiments carried out on the demonstration plant proved that a stable combustion may be achieved even if oil shale with a heating value of 500 kcal was used. The combustion efficiency of the fuel is about 99 %. The efficiency of the boiler is about 84 % which is quite satisfactory as large quantities of ash and flue gases are taken into account. 65-75 % of the oil shale carbonates are decomposed into carbon dioxide and calcium oxide during the combustion. Calcium oxide represents a good absorbent for sulphur oxides formed during combustion. The concentration of SO_2 in the flue gases at the exit of the boiler is about 50 ppm. Quantities of NO_x and ash particles in the flue gases are also very low.

Direct combustion of fuel of such a poor quality as Israeli oil shale involves many problems. Some undesirable phenomena were observed in the work of the boiler, e.g., formation of the "hard" and "soft" deposits on the boiler surfaces. Some of these problems have been successfully solved.

Several projects involving large-scale oil shale power plants with capacity of 75-450 MW are now being developed by the government and private companies. The construction of these plants is planned for the late 1990-s. Negotiations with U.S.-based multinational Mid-Atlantic Co. for the establishment of a private 150 MW electric power plant in the Negev desert are now under way. The estimated investment costs of the plant are $300 million. Israel Electric Corporation has already signed agreements to buy electricity from two private producers: Nordan Co. (10 MW plant), and Escogen (27 MW plant). The IEC is in advanced negotiations with Ormat Co. for an additional private power generation unit.

Oil Shale Processing

Several new oil shale processing technologies are being studied in Israel. A prospective and most advanced process for oil shale retorting was developed by the PAMA Co [3]. The process is based on heating fine-grained oil shale by mixing it with circulating hot ash. The mixture of the spent shale with the ash is combusted in a separate apparatus. The major part of the hot ash is recycled to the retorting reactor. The excess of the ash is cooled with air and discharged. Hot air is used for the spent shale combustion and raw shale drying. Volatile products are cooled and condensed. Retort water containing organic components is combusted together with the spent shale. Retort gas is proposed to utilize for hydrogen production. The following characteristics of the planned commercial unit are:

Capacity, ton oil shale per hour	243
Power consumption, kWh	28.7
Commercial oil yield, %	4.6
Personnel, men	163
Investments, millions dollars	244
Cost of production, $/ton of oil	126.7

PAMA's process provides: good heat utilization of the ash and spent shale carbon; high capacity; simple and reliable construction of the reactor; and satisfactory solution of environment problems. Construction of a pilot plant using this technology is planned for 1996-1997.

Since oil yield of Israeli oil shale is low, attempts have been made to increase it. One of these attempts - a Hom-Tov ("Good Heat") process, was developed by a private company. The main feature of the process is pyrolysis of oil shale mixed with some quantity of liquid fuel. The details of the process are not reported. The experiments are still being carried out on laboratory scale.

Another attempt of this kind - extraction of oil shales under super-critical conditions - was made at the Weizmann Institute [4]. This laboratory investigation was not aimed at commercial results. Oil shale liquefaction, the process investigated by PAMA Ltd. since 1992, appears to be more practical [5]. This process is based on thermal extraction of the kerogen with a H-donor solvent. The following preliminary block-scheme of the process is proposed (Fig. 2):

1. Oil shale preparation and mixing with the solvent to form a slurry
2. Heating of the slurry and thermal extraction of oil shale organic matter in a reactor
3. Separation of products
4. Distillation of liquid products
5. Solvent regeneration

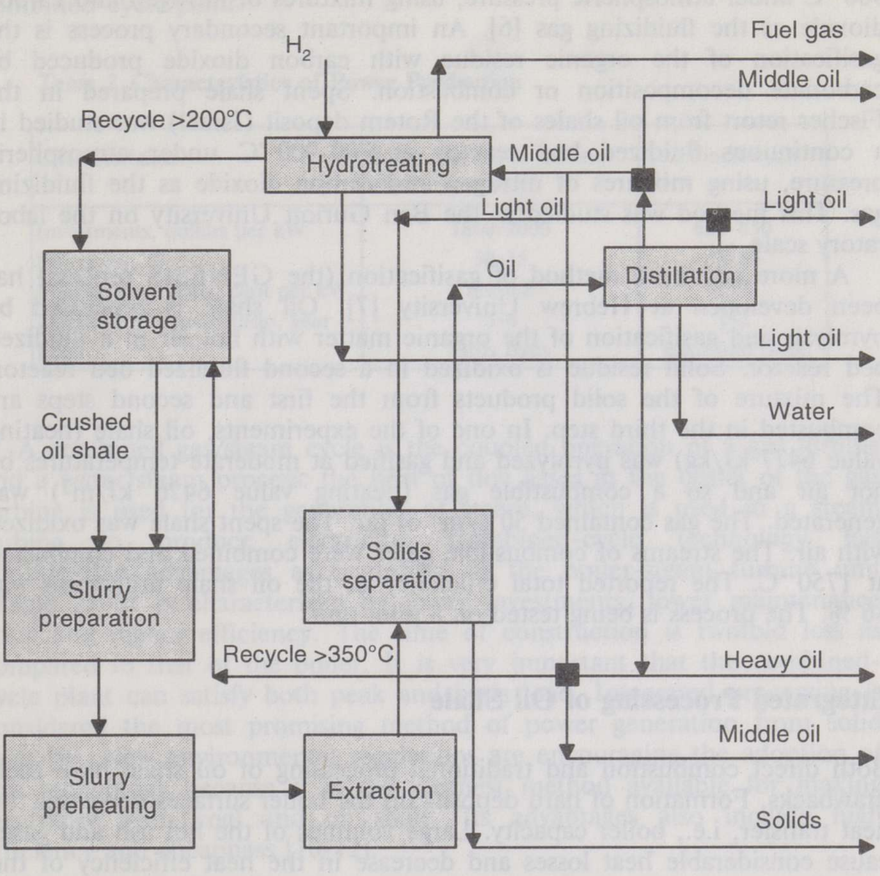

Fig. 2. Block diagram of oil shale thermal extraction

This scheme is based on experiments at a laboratory batch unit (a part of the experiments were carried out in a continuous operation unit in England). An autoclave was used for oil extraction. The shale/solvent ratio was 1/1.3-1.05, and the residence time in the reactor 0-30 min. High pressure and temperature were used. The expected maximal commercial oil yield is 7.1 %, which is 56 % more than by traditional retorting. However, this process has some disadvantages: problems of separation of mineral matter and extracted oil, a rather complicated scheme of solvent regeneration, the necessity of hydrogen production, etc. There are many engineering problems which are still to be solved.

Gasification may turn out to be a more attractive process for Israeli oil shales, as compared to retorting or solvent extraction, because the high-sulphur shale oil requires expensive hydrotreating, whereas the gas may be easily cleaned from sulphur and other impurities. Therefore several methods of oil shale gasification are proposed.

Spent shale was gasified in a continuous fluidized bed reactor at 700-900 °C under atmospheric pressure, using mixtures of nitrogen and carbon dioxide as the fluidizing gas [6]. An important secondary process is the gasification of the organic residue with carbon dioxide produced by carbonate decomposition or combustion. Spent shale prepared in the Fischer retort from oil shales of the Rotem deposit (Israel) was studied in a continuous fluidized bed reactor at 700-900 °C under atmospheric pressure, using mixtures of nitrogen and carbon dioxide as the fluidizing gas. This method was studied at the Ben Gurion University on the laboratory scale.

A more advanced method of gasification (the GENESIS process) has been developed at Hebrew University [7]. Oil shale is processed by pyrolysis and gasification of the organic matter with hot air in a fluidized bed reactor. Solid residue is oxidized in a second fluidized bed reactor. The mixture of the solid products from the first and second steps are combusted in the third step. In one of the experiments, oil shale (heating value 6477 kJ/kg) was pyrolyzed and gasified at moderate temperatures by hot air and so a combustible gas (heating value 6426 kJ/m^3) was generated. The gas contained 50 g/m^3 of tar. The spent shale was oxidized with air. The streams of combustible gases were combined and combusted at 1750 °C. The reported total efficiency of the oil shale utilization was 86 %. The process is being tested on a pilot unit.

Integrated Processing of Oil Shale

Both direct combustion and traditional processing of oil shale have their drawbacks. Formation of hard deposits on the boiler surfaces decreases the heat transfer, i.e., boiler capacity. Large volumes of the hot ash and gases cause considerable heat losses and decrease in the heat efficiency of the boiler.

As to retorting, it produces oil of low quality, since the oil has a high pour point, low content of light fractions, and high content of nitrogen, sulphur and oxygen. In addition, such an oil contains no extractable valuable individual components. An increase in the processing temperature increases gas yield, but heating and treating the ballast mineral matter of oil shale causes considerable heat losses and are accompanied by the decomposition of carbonates, present in the mineral matter of the shale, what also requires additional heat consumption for the process and decreases the heating value of the gas. Mineral matter may melt at such high temperatures, and this hampers oil shale processing.

For this reason, we, at the Technion, have developed a new method of oil shale integrated processing, providing an exhaustive utilization of its energy and chemical potential [8]. The principal features of this method are two-stage processing of oil shale and power co-generation by means of a combined cycle turbine plant. As a result of the two-stage processing, two principal products - liquids and gas are obtained. Owing to the two-stage processing, the liquid products have a simpler composition, as compared to the traditional shale oil, and represent an excellent source for the production of chemicals. The gas is used for the power production in a combined-cycle plant.

Table 2. **Characteristics of Power Production**

Characteristics	Boiler and steam turbine (fuel - oil shale)	Combined-cycle plant
Investments, dollars per kW	1800-2000	600-650
Efficiency, %	30-35	48-53
Maintenance costs, cent per kW	0.6-0.8	0.35
Duration of construction, year	3-4	1-2
Load	Only basis	Basis and peak

A combined gas/steam cycle is the coupled operation of a gas turbine and a water/steam process: the heat of flue gases at the outlet of the gas turbine is used for the generation of steam, which is used in a steam turbine to produce electricity. Combined-cycle technology has considerable advantages as compared to the boiler-steam turbine unit (Table 2). It is characterized by lower investments, lower maintenance costs and higher efficiency. The time of construction is twofold less as compared to that of the boiler. It is very important that the combined-cycle plant can satisfy both peak and basis load. Integrated processing is considered the most promising method of power generation from solid fuels [9]. New environmental regulations are encouraging the adoption of this technology because it is the cleanest method available for making electricity from coal and oil shale. Its advantages also include high efficiency and cheapness [10-12].

Table 3. **Products Yield of Oil Shale Integrated Processing (Dry Basis), wt.%**

Product	Conversion regime			
	-	I	II	III
Cleaned gas	1.03	2.84	3.32	4.01
Gasoline fraction	0.34	0.45	0.49	0.71
Oil	4.71	1.64	1.50	1.07

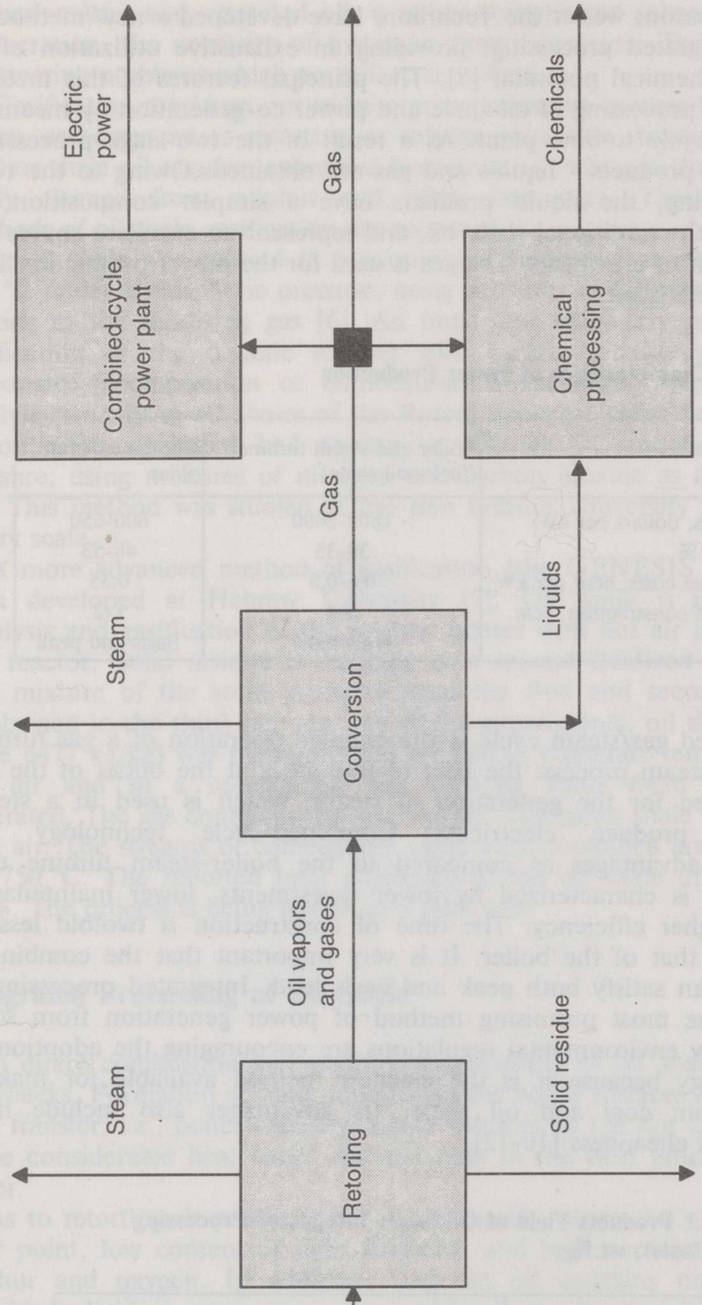

Fig. 3. General sheme of integrated utilization of oil shale

Table 4. **Composition of Gasoline Fraction, wt.%**

Components	Without conversion	With conversion
Total identified aliphatic	11.9	0.55
Benzene	2.2	33.8
Thiophene	1.7	7.4
Toluene	5.2	20.1
2-Methyl thiophene	7.8	9.1
3-Methyl thiophene	1.3	3.2
Total identified aromatics and sulphur-containing heterocyclic compounds	28.1	84.5
Total unidentified	59.9	14.9

Technology of integrated processing of coal [13] coupled with the combined-cycle power plant has been developed to a high extent. Meanwhile, this method can give especially good results when applied to such low-quality fuels as oil shales.

The general scheme of integrated oil shale processing is presented in Fig. 3. In the first stage, oil shale is processed at a temperature, at which the maximum yield of products of kerogen thermal decomposition is observed, i.e. 500-530 °C. The second stage is the conversion of the oil vapours and gases obtained at the first stage. The products of decomposition make up only 15-25 % of the initial oil shale; they do not contain ballast components or solid substances. Besides, they are directed to thermal treating immediately from the outlet of the retort, in the form of a vapour-gas mixture having a temperature of 500 °C. Therefore, their processing can be accomplished without significant expenditures.

The products of the conversion are cooled, and the liquid is condensed and separated from the gas stream. Due to the secondary treatment, the liquid products have a simpler composition, as compared to the initial shale oil, and can be an excellent source for production of chemicals such as benzene, naphthalene, anthracene, thiophene, and other products.

The cleaned and compressed gas is used for energy production in a combined-cycle power plant. It may also be used for chemical syntheses. Hydrogen sulphide obtained as a result of the gas cleaning is used for the production of sulphuric acid and steam.

As a whole, the developed process contains within itself the co-generation of energy and chemicals and provides exhaustive utilization of the energy and chemical potential of the oil shale.

Experiments on the two-stage processing of oil shale were carried out at a batch bench-scale unit. The product yields are presented in Table 3. The yield of the liquid decreased as compared with the traditional retorting, but the gas yield increased by three times.

The composition of the gasoline fraction ("crude benzine") is presented in Table 4. Eighty per cent of this product (in the process with the conversion) consist of only five components - benzene, toluene, thiophene, 2-methyl thiophene, and 3-methyl thiophene. All these compounds can be easily separated from each other, and all of them can be used for chemical syntheses so having a stable market value.

The oil obtained from integrated processing of shale is similar to coal tar and can be processed and used in a similar way. It consists mainly of naphthalene and other aromatic compounds, such as methyl naphthalenes, fluorene, phenanthrene, anthracene, etc.

The liquid products of oil shale two-stage processing differ substantially from the traditional (primary) shale oil. The primary oil consists of a very large number (a few hundred) of compounds, and it is almost impossible to identify (see, e.g., Table 4) and separate them from each other. They cannot be used in chemical syntheses. The converted products consist of a relatively small number of valuable aromatic and sulphur-aromatic individual components that can be easily separated by the traditional methods.

The plant using integrated processing of oil shale should consist of three main blocks: (1) Two-stage retorting of oil shale; (2) Recovery and processing of the liquid products of the conversion; (3) Power production by means of combined-cycle technology. The process of combined production of energy and chemicals has better economical characteristics than the direct combustion of shale or coal. The costs of the power production in the integrated process are 2.53 cents per kWh, as compared to 4.9 cents per kWh for the power production by the direct combustion of oil shale. The integrated process requires a lower investment and has lower operating costs. The high economic effectiveness of the integrated process is due to the three main factors:

1. Using a high-quality ashless, moistureless and sulphurless gas fuel, which can be combusted in turbines of a combined-cycle power plant.
2. Production and efficient utilization of chemical by-products.
3. Integration of the power production and chemical utilization of oil shale in one process.

This new method for oil shale processing has a number of advantages as compared to the traditional ones:

1. Co-generation of energy and chemicals and using combined-cycle technology has better economical characteristics than direct combustion of shale. It requires lower investments and lower operating costs.
2. Better characteristics from the point of view of the environment protection. The preservation of the natural environment will be provided by using high-quality ashless and sulphurless gas fuel instead of oil shale (or sulphur-rich shale oil).
3. Feasibility: the process can easily be controlled in a way enabling to change the quantity and assortment of chemicals and power/chemicals ratio.

Shale Oil Research and Processing

The specific feature of Israeli shale oil is a high content of hetero-atoms - about 10 %, mainly of sulphur. Both natural petroleum and shale oils usually do not contain more than 1-2 and in some cases up to 3 % of sulphur, whereas the sulphur content of Israeli shale oil is as high as 7 %.

The high content of heteroatomic, especially sulphur compounds, requires the development of special methods for processing Israeli shale oil.

Investigations of the composition of Israeli shale oil were started years ago, but carried out only sporadically. Lately, compounds belonging to different chemical groups were identified in shale oil by Yurum and Levi in the Weizmann Institute of Science [14]. The determining role of sulphur in the shale oil composition is confirmed by Z. Aizenshtat (at the Hebrew University), who found that thiophene alkylderivatives constitute a dominant part of the shale oil sulphur compounds.

We also have investigated the composition and properties of Israeli shale oil [15]. The content of phenols in the total shale oil was found to be 5.4%, and that of pyridine bases 3.2 %. In the products of the separation of shale oil, more than 20 individual compounds - phenol and pyridine derivatives - were identified. Aldehydes, alcohols, ethers Ar-O-R and R-O-R, and esters Ar-COO-R and R-COO-R are not present in the shale oil.

The main trends in shale oil utilization are fuel production and processing for obtaining chemicals. The composition and properties of Israeli shale oil (high content of hetero-elements, low content of light fractions, high pour point, low chemical and thermal stability, etc.) are unfavorable for production of motor fuels. Before its processing into fuel, shale oil must be subjected to hydrotreating. Research on the hydrotreating of shale oil is being carried out at Ben Gurion University, Beer Sheeva.

Production of chemicals from shale oil may be much more profitable than that of fuel, but it entails some difficulties. The oil does not contain significant amounts of any individual components which could be used for syntheses. Traditional methods of petrochemistry (reforming, etc.) for the production of individual chemicals (e.g., benzene, toluene, etc.) also cannot be applied to shale oil. Nevertheless, obtaining some chemical products may turn out to be feasible. Shale oil contains a significant amount of biologically active sulphur compounds. Therefore, the principal possibility exists of using shale oil for the production of human and veterinary medicines. In the Technion, a pharmaceutical product called *Ichthyol* is obtained from the shale oil [16].

The composition and properties of shale oil heavy residues as a source for road bitumens were investigated [17]. Both principal methods of asphalt production - vacuum distillation and oxidizing with air - were studied. The straight-run bitumen had satisfactory characteristics. As to the oxidized bitumen, the values of its penetration, ductility and softening point are also satisfactory. The drawback of shale oil bitumens is their high viscosity after the thin film oven test, which means a high sensitivity to aging. The composition of shale oil changes significantly during distillation and oxidation. The content of asphaltenes sharply increases from 3.5 to 20-22 % in the straight-run bitumen and to 30 % in oxidized bitumen, explained by the condensation and polymerization of the most unstable polar components during heating and oxidation. Unlike petroleum, shale oil is a product of pyrogenetic origin, i.e., it is formed as a result of thermal decomposition of large molecules of kerogen, and a part of this cracked material consists of unstable fragments of these molecules, which have a tendency to polymerize.

Utilization of Oil Shale Mineral Matter

As a result of the combustion or processing of oil shale, about 640 kg of ash is produced from every metric ton of oil shale. Rational utilization of the huge quantity of solid material is necessary both from the viewpoint of environmental consideration, and of the improving the profitableness of oil shale processing and combustion. The main target of the investigations in this field is the finding of methods for production of high-grade cement from shale ash. A project is proposed in Israel for the utilization of shale ash as a material for the construction of artificial islands and lagoons.

Conclusion

Israeli companies and universities are carrying out wide-scale research aimed at investigation and commercial use of oil shale. Oil shale reserves have been evaluated, a demonstration plant for power generation has been constructed, first steps in shale processing at experimental units have been made, and a few commercial projects for oil shale utilization are under consideration. In spite of the low quality of local shales, there are good prospects for their use in the profitable production of electric power and chemical products.

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ADDITIONS AND CORRECTIONS

OIL SHALE **1995, Vol. 12, No. 4, p. 364**

"75 YEARS SINCE THE BIRTH OF ACADEMICIAN OLAF EISEN"

...O. Eisen remained a scientific adviser at The Institute of Chemistry until his death on March 14, 1989.

OIL SHALE **1996, Vol. 13, No. 1, pp. 43-64**

V. LIBLIK, H. KUNDEĻ **"POLLUTION SOURCES AND FORMATION OF AIR CONTAMINATION MULTICOMPONENTIAL CONCENTRATION FIELDS OF ORGANIC SUBSTANCES IN NORTH-EASTERN ESTONIA"**

The concentrations of pollutants in the air must be given in **μg m^{-3}**, not in mg m^{-3} (in the text and in the heads of Tables 3, 4, and 6). We apologize for printer errors.

Editors