

Characterization of Shale Oil as Compared to Crude Oil and Some Refined Petroleum Products

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Shale oil prepared by pyrolysis of Jordanian oil shale was characterized and compared to petroleum. Results show that shale oil compares well to crude oil and its fractions. For example, 85% of the shale oil sample fractions fall within the range of the light and heavy gas oil fractions. The 50% boiling range temperature of shale oil is 305°C, whereas for light and heavy gas oils, they are 270°C and 425°C, respectively. This indicates that shale oil may be used as a substitute for gas oil or heavy fuel oil. Major disadvantages of shale oil are that it has high aromatics, heavy metals, and sulfur contents of more than 8.3%.

Keywords characterization, shale oil, crude oil, distillation, petroleum products, Jordan

Jordan is a nonoil-producing Middle Eastern country. It depends heavily on imported crude oil to satisfy its national energy demand. The annual consumption of heavy fuel oil, diesel fuel, and kerosene amounts to more than 70% of the entire amount of all fuel consumed, based on an energy equivalent value (Akash and Mohsen, 1999; Ministry of Energy and Mineral Resources, 2002). On the other hand, the country enjoys an abundant amount of oil shale reserves; the sole indigenous fossil fuel resource, which approximately totals 40 billion tons (Jaber and Probert, 2000; Khraisha and Shabib, 2002; Natural Resources Authority, 2001). However, oil shale is a low calorific value fuel and requires special treatment and consideration concerning some issues, such as fuel management and system design (Jaber et al., 1998a).

At present, oil shale utilization technologies are limited to either retorting to produce shale oil, which is crudelike oil, and synthetic gases, or direct combustion for electric-power generation and other industrial purposes. However, the main interest worldwide has been directed to the pyrolysis and retorting. Thus, tens of retorting processes can be found in oil shale literature. Some of these processes are currently available in the

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international market on commercial scale, such as those operating in Estonia, Russia, and China, while others are still on pilot scale, or under development in lab-testing phase (Lee, 1991; Shuyuan and Jialin, 1992; Jaber et al., 1998b). The produced shale oil can be used either as a substitute for heavy fuel oil or diesel in industrial boilers and furnaces, or further treated and processed to yield commercial products. However, there are major difficulties facing the development of the oil shale-industry, mainly, the economics of existing utilization technologies and environmental impacts of the processes involved. For example, the relatively low quality of synthetic fuels, e.g., crude shale oil, produced and high amounts of gas emission to atmosphere. Moreover, solid-waste handling in addition to land use impacts can be of much greater concern than emission (Jaber and Probert, 1999).

Pyrolysis and thermal kinetics of some Jordanian oil shales were presented in a number of studies (Jaber et al., 1999; Jaber and Probert, 2000; Jaber et al., 2001a; Khraisha and Shabib, 2002; Khraisha, 1998). It is a fact that synthetic oils can be produced at pyrolysis temperature in the range of 200–620°C (Jaber et al., 1999; Williams and Ahmad, 1999). The addition of polypropylene enhances the decomposition of organic matter in oil shale (Gersten et al., 2000). Also, hydro-treatment can reduce nitrogen and sulfur contents in shale oil significantly (Chishti and Williams, 1999). In general, fuel products from shale oil contain higher levels of nitrogen, sulfur, ash, and toxic inorganic matter when compared to other liquid fuels obtained from various sources (Schmith-Collerus et al., 1976; Rovero et al., 1983; Shao-Hui and Kuang-Zong, 1993; Okutan et al., 1994). The presence of such biologically active compounds in the derived oil shale pyrolysis oils, particularly if they are present in high concentration, may limit the use of this fuel as a direct substitute for petroleum-derived commercial fuels since the fuel would represent a health hazard. For example, polycyclic aromatic hydrocarbons containing sulfur and nitrogen are important because of their carcinogenic and/or mutagenic activity (McFall et al., 1984; Williams and Nazzal, 1997), and increased concentrations of such compounds have been shown to give increased soot and pollutant emissions in combustion systems (Guerin, 1978; Longwell, 1983; Henderson et al., 1984; Williams, 1990). Therefore, it requires more extensive refining than crude oil (Luik et al., 1995; Abu-Qudais and Al-Widyan, 2002). Separation and quantification of the slate of nitrogen compounds present in shale oil was discussed and investigated by many researchers (Mushrush et al., 1999; Williams and Chishti, 1999).

In a recent study, combustion of shale oil was carried out in a water-cooled furnace to study the effect of heat transfer and emissions (Abu-Qudais, 2002). It was found that heat transfer was improved when shale oil was used when compared to using diesel fuel. Also, there was some improvement with some exhaust emissions. In another study, shale oil was used as an alternative to diesel fuel in a single-cylinder, direct-injection diesel engine (Abu-Qudais and Al-Widyan, 2002). It was found that shale oil resulted in good engine performance having higher thermal efficiency than diesel fuel.

Understanding the chemical structure of kerogen gives information on the nature of the origin of the organic matter (Kribii et al., 2001). Kerogen is a complex substance that consists of large hydrocarbon molecules with sulfur, nitrogen, and oxygen atoms introduced into the molecule. An oil shale may have molecular weight in the order of 3000, with an approximate empirical formula $C_{200}H_{300}SN_5O_{11}$ and C/H ratio of about 1.5 (Torrente and Galan, 2001; Dick et al., 2002). Heating the oil shale in an inert atmosphere causes kerogen to decompose, leaving liquid and gaseous products though this process.

Recently, due to increase in the price of imported crude oil, there is a growing interest in developing oil shale deposits, especially those in the central part of Jordan.

For example, the Ministry of Energy and Mineral Resources has signed a memorandum of understanding with a specialized Canadian firm, SUNCOR, which aims to develop the Ellujjun oil shale deposit and produce shale oil. The proposed technology is similar to what has been used on pilot scale for processing the Sturt oil shale in Queensland, Australia (Southern Pacific Petroleum, 2001). However, this is still unproven on a commercial scale, and there are no scientific field studies concerning the associated operating and environmental difficulties. These need a lot of work; e.g., technical and financial feasibilities, environmental impact assessment, and post-treatment of produced shale oil must be implemented before being available for safe and reliable commercial applications (Jaber et al., 2001b). Hence, significant information gaps regarding the utilization of Jordanian oil shale exist to inhibit the making of accurate assessments concerning the characteristics and behavior of the raw shale and/or the produced shale oil.

The present investigation was conducted in order to gain an understanding of the possibility of utilizing synthetic oil to substitute for mineral crude oil and its refined products. It is deemed that this experimental study will provide a small portion of the missing information. This article is primarily aimed at characterization of a shale oil sample, produced from the Ellujjun oil shale deposit in order to give future researchers some introductory overview about the general physical, chemical, and thermal characteristics of Jordanian shale oil compared to crude oil, diesel fuel, and heavy fuel oil.

Materials and Methods

Fuel Samples

Shale Oil. The shale oil samples used in this investigation were extracted from raw oil shale that was mined from the Ellujjun deposit and provided by the Natural Resources Authority. The elemental and proximate analyses of the raw shale are presented in Table 1. It is clear that the raw oil shale has a high sulfur content of about 3–4%, while the C/H ratio is 8.9. The extraction process was performed using the Chinese Fushun retort. The latter is similar to the gas producer employed for the gasification of coal, and it is directly heated. It has two main sections: the pyrolysis zone with an average temperature of about 500°C, and the combustion part with a temperature of ~800 to 1000°C. Oil shale lumps

Table 1
Analysis of Jordanian (Ellujjun) oil shale

Elemental analysis (%)	
Total carbon	22.80
Organic carbon	17.93
Hydrogen	2.57
Sulfur	4.54
Nitrogen	0.40
Oxygen	—
Proximate analysis (%)	
Volatile matter	43.96
Fixed carbon	0.42
Moisture	1.11
Ash	54.51
Carbonate	40.60
Higher heating value (MJ kg ⁻¹)	6.0

are fed from the top and move downward by gravity. The pyrolysis takes place by the flow of hot recycled gases as well as those generated in the combustion zone. Products such as vapors and hot gases are removed from the top, then cooled down in order to recover condensable fraction, i.e., shale oil. The produced shale oil is pretreated by removing its water content. The spent shale is discharged through a water seal at the bottom of the retort. However, the exact technical details and procedure of the Fushun process can be found elsewhere (Zhou, 1995). The shale oil sample is kept in an air-tight container.

Crude Oil. The crude oil samples used in this study are obtained from the imported Iraqi oil, with a benchmark known as Medium Basra. These samples were provided by the Jordanian Petroleum Refinery Company (JOPETROL) and taken from fresh received shipments with no prior treatment at the refinery site. The only treatment carried out was the usual pretreatment at origin; this includes gas separation, salt removal, and dewatering.

Gas Oil. JOPETROL produces one grade of light gas oil, i.e., diesel fuel, which is mainly used in compression ignition internal combustion engines. However, the same fuel is used for heating purposes in residential and industrial furnaces and boilers. The diesel fuel sample used in this study was also provided by JOPETROL.

Heavy Fuel Oil. Heavy fuel oil, known as fuel #6, is produced as distillation by product in JOPETROL. This fuel is highly viscous because it consists of high molecular weight compounds. It is an inexpensive fuel and mostly used to supply large industries and thermal power plants in Jordan, because it requires special arrangement, including storage and firing systems. The sample used in this analysis was also provided by JOPETROL.

Procedure and Sample Characterization

All fuel samples were distilled under vacuum in order to obtain various cuts or fractions. The first distillation fraction is the light cut, i.e., gasoline fraction, which contains mostly low molecular weight hydrocarbon volatiles, followed by other heavier fractions as shown in Table 2. This table also lists the distillation or recovery temperature ranges for all normal cuts. Physical and thermal characteristics of these samples were obtained employing standard tests adopted in petroleum refineries for crude oil and petroleum products. Table 3 summarizes the experimental tests carried out, standard testing method, and the apparatus used for each test in this investigation.

In order to reduce the margin of error, a daily calibration of all the testing devices and instruments employed in this investigation was carried out, and the response of the

Table 2
Distillation temperature range of
various fractions

Fraction	Temperature range (°C)
Gasoline	0–145
Kerosene	146–245
Light gas oil	254–370
Heavy gas oil	371–550
Vacuum residue	> 550

Table 3
Experimental tests and laboratory apparatus employed

Test	Standard testing method	Name of apparatus used
Distillation	ASTM D-110/IP-24	Vacuum distillation
Initial boiling point		Gecil process
Density	ASTM D-1298	Hydrometer
Water content	ASTM D-1796	Distillation
Sediment	ASTM D-1796	Centrifuge
Flash point	ASTM D-93	P.M.C tester
Total sulfur	ASTM D-4294	X.R-F MiniPal
Kinematic viscosity	ASTM D-2170	Viscometers
Pour point	ASTM D-2170	Viscometers
Salt content	IP-77	Separation flask
Heavy metals		
Fe	CMM-82	ASS/vario-6
Na, K, V & Ni	ASTM D-5863	ASS/vario-6
Ca & Mg	ASTM D-4628	ASS/vario-6
Si	ASTM D-5184	ASS/vario-6
Heating value	ASTM D-240	IKA C-5003

equipment, when approved standard calibration procedure followed, was checked before any analyses were carried out. Also, the same experimental technique, including sample preparation procedure, analysis method, and the adopted calculation model for all tested fuel samples was employed in order to enable a fair comparison.

Results and Discussion

The results of fractionation of shale oil and crude oil by vacuum distillation are presented in Figure 1. It can be shown that crude oil results in five major fractions ranging from gasoline fraction, followed by kerosene, light gas oil was next, followed by heavy gas oil, and finally, the fraction that represents residue. These five fractions have an equal volume distribution of approximately 20% of the total sample. On the other hand, the shale oil sample produces a negligible amount of the gasoline fraction. Also, it was rich in light and heavy gas oil fractions; their combined portions add up to approximately 85% of the total sample. The absence of the light cut in the shale oil sample can be attributed to the prevailed retorting process controlling conditions, i.e., secondary coking and cracking reactions. These are direct functions of pyrolysis temperature. In this case, secondary reactions are more important because the Fushun retort is directly heated by burning the residual carbon in the retorted shale. Thus, the products in vapor phase may experience higher temperatures than desired. Cracking involves vapor phase bond fission reactions leading to the formation of distribution of molecular weight species, mostly species smaller than the original molecule, i.e. gases, plus some carbonaceous residue.

The initial boiling point for shale oil is too high, 200°C compared to only 41°C for crude oil. But it is close to that for reduced crude, i.e., atmospheric distillation unit bottom. Also, the density at 15°C for shale oil (0.982) is about that for reduced crude (0.965), and both of these are higher than that for crude oil (0.857). This suggests that the tested shale oil sample is similar to the fuel sample obtained from the atmospheric unit bottom. However, it was possible to recover 90% of the shale oil sample, up to

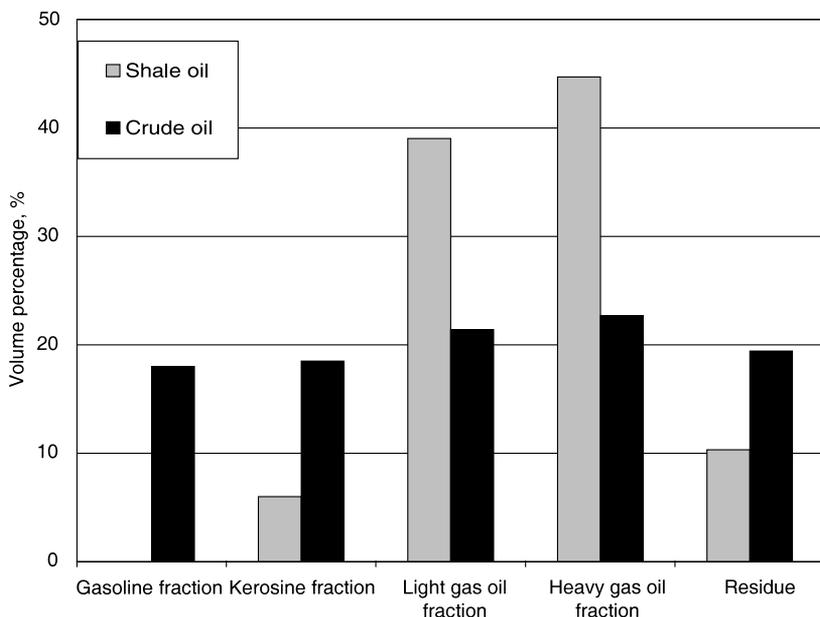


Figure 1. Volumetric distribution of various fractions obtained from shale oil and crude oil.

temperatures of 528°C, where only 52% of the reduced crude sample could be recovered under similar conditions. Similar observations related to reduced crude apply to the heavy fuel oil sample. But it should be kept in mind that the latter is a final commercial product that fuels industrial boilers and thermal power plants, whereas reduced crude is an intermediate product resulting from the atmospheric distillation unit, and used to feed vacuum distillation units in petroleum refineries.

Boiling range distributions of shale oil compared to light gas oil and heavy gas oil are presented in Figure 2. It is clearly shown that the initial 10% boiling range temperature is about 260°C for shale oil, whereas it is about 200°C for light gas oil. But, boiling range for 10% recovery for both samples are much higher than that for the tested crude oil sample; it was about 100°C. Also, at the other extreme, the 90% boiling range temperature, is about 528°C for shale oil. The 90% boiling range temperature of heavy gas oil is about 500°C. Finally, the 50% boiling range temperatures for light gas oil, shale oil, and heavy gas oil are about 270°C, 305°C, and 425°C, respectively. Therefore, it is necessary to compare each fraction of shale oil to those fractions obtained from crude oil. Figure 3 shows the boiling range for kerosene fraction obtained from shale oil and crude oil samples. It is clear that the percentages of volatiles that exist in crude oil are higher than those in the shale oil sample. Similarly, boiling ranges for light gas oil fractions obtained from shale oil and crude oil are presented in Figure 4. For both samples the results were very similar. The 50% boiling range temperature of light gas oil fraction from shale oil was about 300°C, which is similar for that of crude oil. Similar results were obtained for the heavy gas oil sample as shown in Figure 5. The 50% boiling range temperatures are about 425°C and 435°C for shale oil and crude oil, respectively.

Among most important factors influencing final users in the selection process of suitable fuel are unit cost, calorific value, and other properties such as sulfur, heavy metal contents, and pour point, etc. Table 4 summarizes key physical and thermal properties

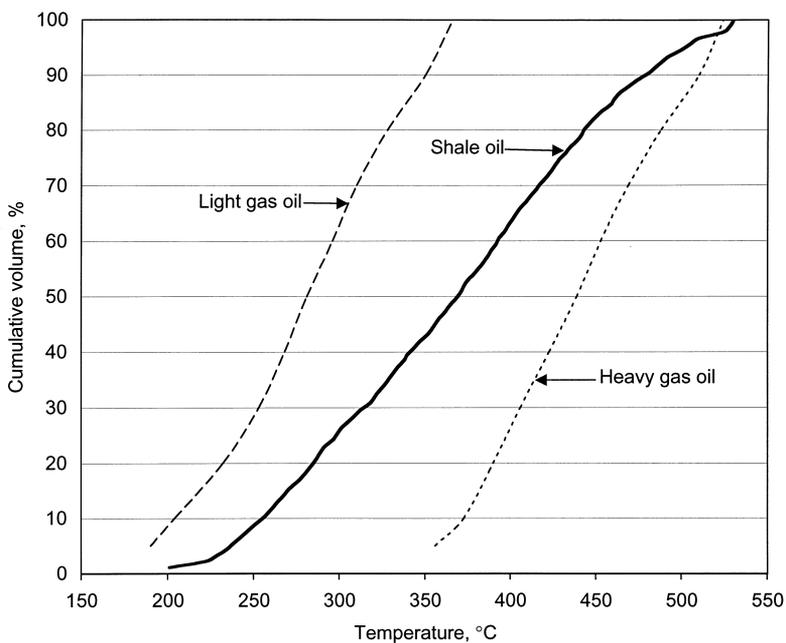


Figure 2. Boiling range distributions of shale oil as compared to light and heavy gas oil fractions obtained from crude oil.

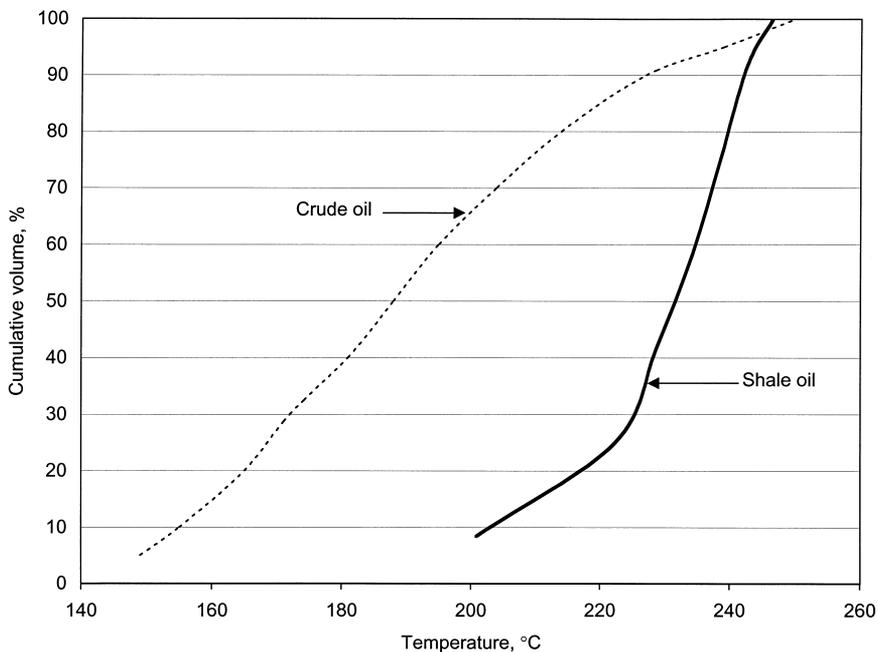


Figure 3. Boiling range distributions of kerosene-range fractions obtained from shale oil and crude oil.

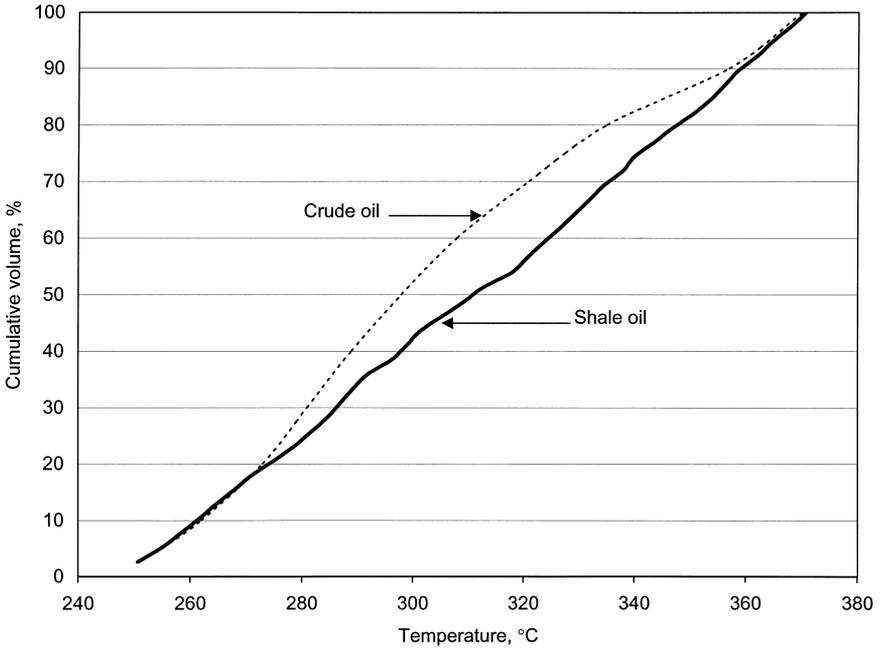


Figure 4. Boiling range distributions of light gas oil-range fractions obtained from shale oil and crude oil.

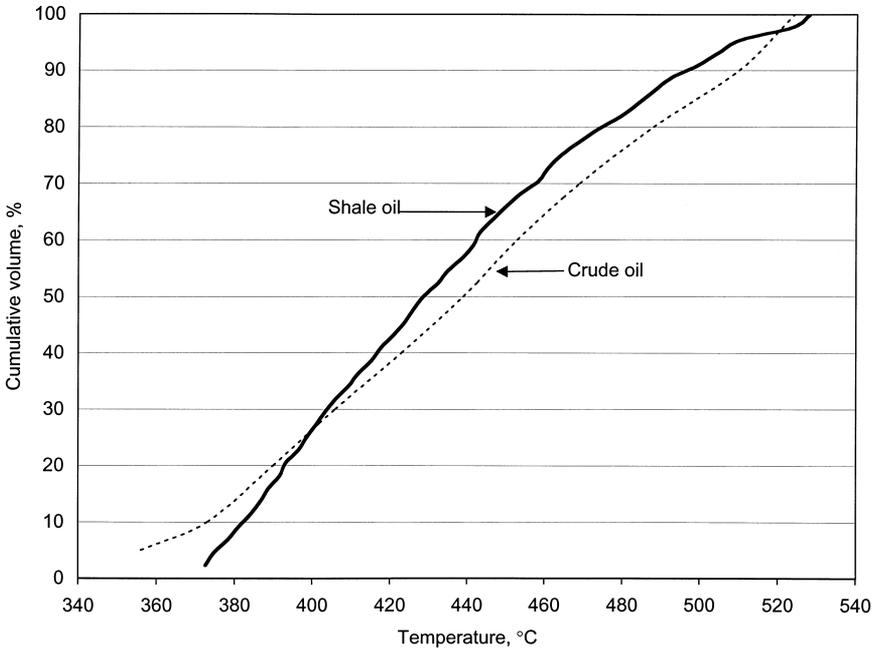


Figure 5. Boiling range distributions of heavy gas oil-range fractions obtained from shale oil and crude oil.

Table 4
Properties of shale oil and other liquid fuels

Property	Shale oil	Crude oil	Diesel fuel	Heavy fuel oil
Density (@ 15°C), kg m ⁻³	982	857	835	967
Total sulfur, wt%	8.3–9	1.7–2	0.9–1.1	3.5–4.0
Pour point, °C	21	(–30)	(–6)	3
Flash point, °C	93	—	65	80
Heating value (low), MJ kg ⁻¹	38.8	42.0	42.8	40.7

for shale oil, crude oil, and some refined products. It is clear, by all means, that shale oil is of relatively poor quality compared to crude oil, diesel, and heavy fuel oil. Its net calorific value is lower, on average, by 8% and 5% of that for crude oil and heavy fuel oil, respectively. The pour point is high due to the fact that shale oil is very paraffinic. This means that it requires preheating before being pumped in pipes for further treatment or transported to other sites. More important is the high sulfur content in the shale oil sample. This is approximately 500% and 250% of the sulfur content in the imported crude oil, i.e., Medium Basra and produced heavy fuel oil, respectively. Moreover, shale oil content of some heavy metals is several times higher than that for crude oil and heavy fuel oil as shown in Table 5, especially iron, calcium, and nickel. But the content of heavy metals, i.e., vanadium and alkali metals (Na and K) causing high temperature corrosion in shale oil is very low compared to crude and heavy fuel oil. This can be regarded as an extra advantage for shale oil over crude oil and its derived heavy fuel oil, especially if it is used to fuel heavy-duty industrial gas turbines. However, the presence of high metal and sulfur contents in shale oil is undesirable. For example, sulfur usually leads to the formation of sulfur oxides when used directly as a substitute for petroleum products, e.g., heavy fuel oil, as a result of combustion process.

After the distillation test, the final products from the shale oil sample were analyzed, and it was found that aromatics constitute more than 90% of these products, especially the light cut which is comprised mainly from gasoline and kerosene. This is in total agreement with oil shale literature, except for other types and origins of oil shale (Putun et al., 1988; Williams and Nazzal, 1995; Williams and Chishti, 1999). Many researchers reported that high levels of nitrogen, sulfur, ash, and toxic inorganic matter in the derived

Table 5
Heavy metals content in shale oil and other liquid fuels (ppm)

Heavy metal	Shale oil	Crude oil	Heavy fuel oil
Ni	110	12	20
Fe	145	12	7
Si	6	5	8
Ca	5	1	3
V	2	24	53
Na	2	3	27
K	0	0.2	0.5
Mg	0	0	0

shale oil by pyrolysis, particularly if they are present in high concentration, might limit the use of this fuel as a direct substitute for petroleum-derived commercial fuels, since the fuel would represent a health hazard (Rovero et al., 1983; Schmith-Collerus et al., 1976; Okutan et al., 1994). For example, polycyclic aromatic hydrocarbons containing sulfur and nitrogen are important because of their carcinogenic and/or mutagenic activity (McFall et al., 1984; Williams and Nazzal, 1997). Also, increased concentrations of such compounds have been shown to give increased soot and pollutant emissions in combustion systems (Williams, 1990; Longwell, 1983; Henderson et al., 1984; Guerin, 1978). Therefore, it requires more extensive refining, e.g., cleaning and hydrotreatment, than crude oil (Williams and Chishti, 2001; Abu-Qudais and Al-Widyan, 2002). A new method for separation and quantification of the slate of nitrogen and sulfur compounds present in shale oil was suggested by many researchers (Mushrush et al., 1999; Williams and Chishti, 1999).

In the oil shale literature, it was found that an actual refinery test run conducted on the shale oil produced as a result of retorting raw oil shale from Colorado in the USA, which is approximately similar to Jordanian oil shales, has shown that shale oil can be refined into commercially valuable products. These can be used to substitute for petroleum derivatives with only minor modifications and adjustments of the operating conditions being necessary in an existing petroleum refinery (Reeg et al., 1990; Harada, 1991). The light naphtha-fraction obtained from fractionation of the hydrotreated product is very paraffinic with a low octane rating, whereas the bottom fraction is more or less equivalent to heavy fuel oil, but with a low viscosity. Such differences are attributed to the fact that shale oil pyrolytically derived from oil shale is modestly rich in certain unsaturated hydrocarbons, which is one of the principal differences between crude and shale oils (Coburn et al., 1978; Putun et al., 1988). It is suggested that one way of utilizing produced shale oil is to mix it with crude oil at certain ratios to suit the existing refinery configuration. As a preliminary conclusion, it can be said that shale oil is amenable to convert into commercial fuels in existing refineries. However, the costs involved are higher than those for conventional crude oil refining processes (Cameron Engineering Inc., 1978; Sullivan, 1978). Thus, the final unit cost of commercial products derived from shale oil may be higher than those accrued for similar petroleum products.

Conclusion

More energy resources will be required to satisfy the needs of modern societies. Thus, synthetic fuels derived from oil shale, coal, and different bio-sources would have a major role in our future. Vast oil shale deposits in Jordan represent the chief fossil energy source. Hence, its development and utilization to cover part of the growing national energy demand may become a reality within next few years. On the other hand, there is little information available in the literature about processing Jordanian oil shales. The general composition of crude oil differs from shale oil in that unsaturated hydrocarbons are present in the latter, and are generated by cracking during the pyrolysis process, i.e., decomposition of the kerogen of oil shale.

Some chemical and physical properties of Jordanian shale oil are presented in this study. They are compared to crude oil, heavy fuel oil, and diesel fuel. The analysis shows that shale oil compares well to heavy fuel oil and diesel fuel. However, shale oil has a major disadvantage: its aromatic, heavy metals, and sulfur contents are very high. If it is decided to be further processed to yield commercial synfuels, then the produced shale oil should be upgraded and hydrotreated first before being refined. An alternative route is to utilize shale oil as boiler fuel as a substitute for heavy fuel oil in power plants

and industrial furnaces, after being cleaned and pretreated. However, high aromatic and sulfur contents suggest that shale oil has a good potential to be processed in order to obtain industrial products, such as adhesives, synthetic materials, and sulfur. This article is deemed to help both decision makers working in energy planning and future researchers in the field of oil shale to get information regarding this fuel and needed future studies.

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