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BEHAVIOUR OF TWO JORDANIAN OIL SHALES AT DRYING

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The drying kinetics of two oil shales from different deposits was investigated over a temperature range of 70–150 °C in thermogravimetric analyser (TGA) and under direct insolation. A series of experiments in a convection-drying oven was carried out with particles of similar size and at the final temperature applied in TGA. The weight loss and drying rates of the samples were determined gravimetrically. It has been observed that drying rate falls off at a critical temperature of about 120 °C and approaches zero beyond this temperature. For both types of oil shale, there is a slight increase in the surface-water yield upon increasing the final drying temperature.

Introduction

Jordan differs from neighbouring Arab countries: it is not an oil-producing country having a relatively small population of about 4.5 million. The kingdom possesses small known reserves of traditional commercial energy sources such as crude oil. The indigenous natural gas resource is satisfying less than 4 % of the current annual energy demand, and it is used directly for power generation, i.e. to fuel 4 × 30 MW gas turbines [1]. So the country is almost totally dependent on imported crude oil and petroleum products.

On the other hand, there are vast oil shale deposits, i.e. ~5×10^{10} tonnes, occurring in Jordan, not exploited yet. Based on the current energy consumption these would satisfy local energy demand for hundreds of years if developed and utilized wisely. Thus, such a domestic resource has a potentially major role to play in reducing Jordan's dependence on imported crude oil and/or petroleum products: the annual imported crude oil cost for 1999 was ~5.5×10^8 US$, which represented about 7 % of Jordan's gross domestic product [2]. Without exploitation of indigenous oil shale, the corresponding imports will cost about $1.2(\pm 0.2) \times 10^9$ US$ per annum depending on the international unit crude-oil price, at the end of this decade, when the annual

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rate of fuel consumption in Jordan will be approximately twice as mach as the present rate [3].

There is not much information available about processing Jordanian oil shales. This may be attributed to the prevailing low crude oil unit prices as compared with the price of the energy produced from oil shale during the last 15 years. Therefore, concerned governmental agencies have shown only little interest in developing the exploitation of this resource. Nevertheless, most likely this situation will change within the next few years because of rising crude oil price, which is currently around 30 US$ per barrel [4], i.e. more than twice the price prevailing two years earlier.

Any basic or advanced above ground, i.e. ex situ, plant for the utilization of oil shale would consist of the following: mining operations, material-handling facilities, processing plant as the core of oil shale utilization chain in which actual conversion of organic matter occurs, and support systems [5]. The latter includes raw-water treatment unit, waste-water treatment plant, spent ash cooling and disposal system as well as storage and general facilities.

The mined oil shale requires crushing and sizing prior to feeding it to the processing facilities. Sizing operations include secondary crushing and screening. The required size of the crushed oil shale depends on the specific operating conditions of the process being employed. In order to reduce the cost of raw shale transportation, the primary crushers are located close to mining operations, whereas the secondary crushers are located at the processing site, i.e. outside the mine.

In order to avoid a shutdown of the processing units, a sufficient quantity of the coarse ore and fine shale should be maintained in strategic stores to facilitate uninterrupted operation of the downstream processing facilities. This may be achieved by diverting the excess quantity excavated during periods when the ore production exceeds the feed requirements of the plant.

During oil shale storage, which may last for days or weeks, significant changes may occur, the most important are the loss of carbon dust and some of the moisture. So, drying, which is a separation or removal of water from oil shale, is a vital process because the moisture present in shale may be of some importance due to the associated energy and quite long times required for the utilization of oil shale deposits. In practice, there are two broad classes of water associated with shale: surface water and interlayer or retort water. Surface water is the moisture that can be removed from the sample when heated to a temperature of about 105 °C while the bound water is the portion evaporated only at higher temperatures like those in the retorting process [6, 7].

Jordanian oil shales contain small percentage of moisture – about 3–5 %, by weight [8, 9]. This may vary widely from one site to another, depending on composition, characteristics and structure of the oil shale deposit. In any process aiming at utilization of oil shale, when moisture may cause sticking problems either during preparation and storage or transport, the understanding and determination of drying characteristics can be considered very important in enabling to choose or design required drying system effectively.

It has been reported that generally the bound moisture content exceeds that of the free moisture in Jordanian oil shales, which is typical of Colorado (USA) oil shales. However, there is very little information published in the open literature concerning the retention of moisture in shales, and even less information is known about the kinetics of moisture removal, especially concerning Jordanian shales [10–12].

The prime aim of this experimental study was to study drying of different oil shale samples from the Ellujjun and Sultani deposits in the central part of Jordan, using a drying furnace, direct solar radiation and thermogravimetric analyser. The weight loss (free-moisture content) as a result of heating under various conditions was determined. Such information is highly needed as the basis to aid in the design and operation of an efficient oil shale handling, storage and processing systems.

Experimental Method

Types of Shales Used

The oil shale samples investigated (from the Ellujjun and Sultani deposits, 120 and 150 km south of the capital city Amman, respectively) were provided by the Natural Resources Authority. However, the details concerning the sampling method used were not provided. These two samples were crushed as received, separately and without further treatment by crusher, then sieved into two categories – grain size ≤ 0.85 mm, which can be considered granular powder, and 0.85–3.36 mm.

TGA Analysis

Kinetic data were obtained using a Shimadzu Model-50 Series TG Analyser, with nitrogen (at a constant rate of $\sim 5 \times 10^{-5}$ m^3 min^{-1}) being employed as the purge gas. The TGA apparatus provides for the continuous measurement of sample weight as a function of temperature and provision is made for an electronic differentiation of the weight signal to give the rate of weight loss. In this study, TGAs were used to determine the effects of final temperature and particle grain size on the weight loss of the oil shale sample.

Drying was carried out non-isothermally using a small sample of about 15 mg, placed in the alumina crucible, which was then put on the sample pan hanging down in the reaction tube, where the atmosphere could be controlled. The furnace tube was raised to close the system, and the start button depressed. The pre-programmed control unit regulates all the automatic functions of the recorder, e.g. the continuous measuring of the sample mass, as well as the temperature programming of the furnace. Finally, after the furnace temperature had achieved its set value, i.e. 150 °C, the sample was allowed to cool down to the normal room temperature.

Drying Furnace

The laboratory-scale convection-drying furnace was used to heat the oil shale sample, in order to determine its surface-moisture content. The furnace is heated externally by an electric-heating element and its behaviour is controlled by a programmable temperature controller. This enabled to vary the temperature up to a maximum of about 150°C, and to hold the sample at the desired final temperature for a certain period in order to remove the free water from the shale. The pre-weighed oil shale sample, of ~150 mg, was placed in a ceramic crucible and then put inside the pre-heated furnace – drying was carried out isothermally. Upon completion of each experiment, the crucible was removed and weighed again in order to determine the difference, i.e. weight loss.

Another set of experiments, under direct solar radiation, was conducted to confirm the results obtained from other tests. Three ceramic boats containing relatively equal oil shale samples were used to simulate different possible storage techniques: the first sample was covered with ventilated greenhouse made of glass, the second one was placed in a chamber made of cardboard with two slots in the top to allow free air movement, in order to simulate the storage in silos, and the last sample was left uncovered in the ambient air to represent external stockpiling of crushed oil shale. These samples were placed together in a very large cardboard box and left for one week on the roof of the Institute of Land, Water and Environment at the Hashemite University, during July where the dry-bulb temperature in shade exceeded 40 °C, to minimise the effects of weather and surrounding environment. It is worth noting that in the oven and direct solar drying experiments, the influence of water content of the ambient air on the drying rate was ignored.

The thermal analysis of oil shale helps in understanding as well as in evaluating the environment and mechanisms of processing, so that the behaviour of the shale could be predicted with a high degree of certainty for a wide range of operating conditions. In this study, an attempt has been made to determine the effect of various variables, such as temperature and particle size, on the drying profile of two different Jordanian oil shales. In order to reduce the margin of error and so to produce more reliable data, individual tests were repeated at least three times, and then the average result was taken.

However, it should be remembered that this study was carried out in a laboratory-scale drying furnace and using a non-isothermal TGA that has its own limitations. Because of the small scale of the experiments, it would be difficult to extrapolate quantitatively the results obtained so as to predict what will happen exactly in actual commercial oil shale storage and processing facility. However, the presently identified trends could serve as guidelines or indicators, but a more detailed research concerning the properties and behaviours of Jordanian oil shales is still highly needed.

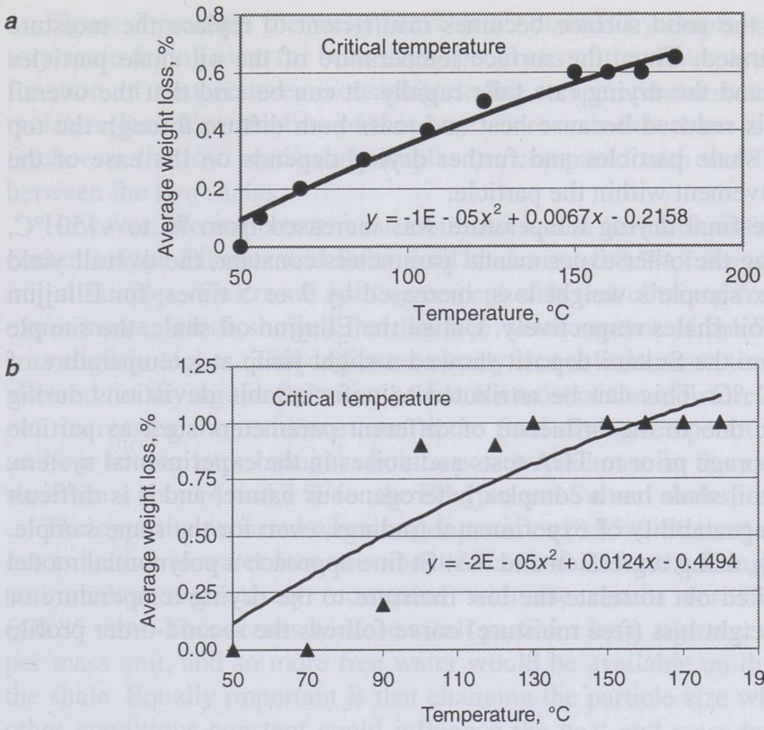

Fig. 1. TG profile of the Ellujjun (*a*) and Sultani (*b*) oil shales with respect to drying temperature

Results and Discussion

The association of water with its host mineral matrix can be expected to vary from weak physical interactions such as surface tension forces to stable chemically bound hydrates. The mechanism of drying consists of simultaneous heat- and mass-transfer processes between drying medium, e.g. hot air in the case of oven drying, and the oil shale sample. Heat is transferred by convection and radiation to the solid surface of the particles, then by conduction into the core of the oil shale particle. Simultaneously, water mass is transferred from the solid surface by evaporation. At the beginning of drying process almost all the heat transferred is stored in the particle as sensible heat; so its temperature rises. Once the solid-particle temperature reaches about 70–80 °C, part of the transferred heat will provide required latent heat for vaporisation of free water. Hence, the particle starts losing some of its inherited moisture, and the drying rate remains constant.

Figure 1 shows the weight loss thermogravimetry, i.e. TGA curves, for Ellujjun and Sultani oil shales.

The rate of weight loss due to the evaporating moisture is directly related to the drying temperature: the higher the final temperature, the higher the weight loss, till it reaches a critical value. Beyond this point, the movement of liquid to the solid surface becomes insufficient to replace the moisture being evaporated. Thus, the surface temperature of the oil shale particles rises again, and the drying rate falls rapidly. It can be said that the overall drying rate is reduced because heat and mass both diffuse through the top layer of oil shale particles and further drying depends on the ease of the moisture movement within the particle.

When the final drying temperature was increased from 70 to ~150 °C, while keeping the other experimental parameters constant, the overall yield of water, i.e. sample's weight loss, increased by 3 or 5 times, for Ellujjun and Sultani oil shales respectively. Unlike the Ellujjun oil shale, the sample obtained from the Sultani deposit showed a slight jump at a temperature of about 80–90 °C. This can be attributed to unfavourable deviations during experiments, due to the influence of different parameters such as particle geometry, storage prior to TGA tests and noises in the experimental system. In addition, oil shale has a complex heterogeneous nature, and it is difficult to achieve repeatability of experimental findings, even for the same sample. Nevertheless, using regression and best-fit line approach a polynomial model may be worked out to relate the lost moisture to the drying temperature or time. The weight loss (free moisture) curve follows the second-order profile – see Fig. 2.

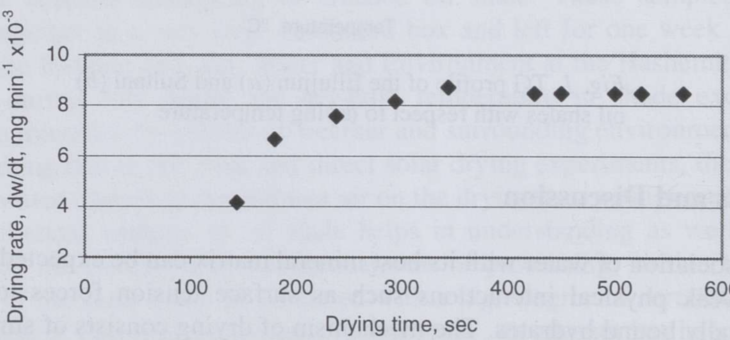

Fig. 2. Drying-rate curve for the Ellujjun oil shale with respect to drying time

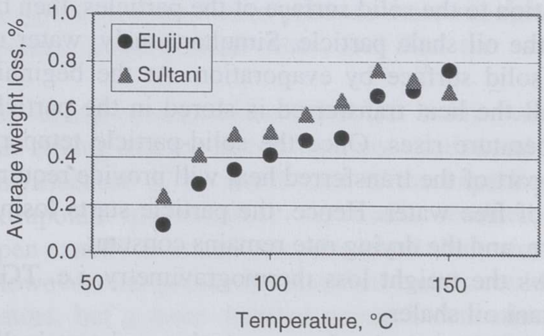

Fig. 3. Average loss of free moisture from the Ellujjun and Sultani shales under isothermal conditions and constant residence time

Figure 3 presents the weight-loss data obtained by employing a laboratory-scale drying oven for both shale specimens of the same particle size as a function of temperature: increasing drying temperature resulted in higher moisture loss. Examination of these curves reveals that there is a slight difference in the lost portion of samples (as compared with the TGA results) between the two shales.

However, the important result obtained is that the total surface moisture content of the Ellujjun and Sultani oil shales is ~0.8 % of the weight of the initial samples. This result is in agreement with that obtained from the TGA experiments, which showed that the free water content of the Sultani shale was slightly higher. This indicates clearly that a little more time should be allowed for drying the Sultani oil shale for most of the moisture to be driven out. Increasing the residence time at the final drying temperature from half an hour to one hour brought about a positive increase in the extracted free moisture – about 4 and 7 % for the Ellujjun and Sultani shales, respectively.

The opposite occurred when larger particles (0.85–2.36 mm) were used: a net reduction in water loss was about 35 and 40 % for the Ellujjun and Sultani shales, respectively, compared with the results for fine particles (≤0.85 mm). This occurs because small particles have a greater surface area per mass unit, and so more free water would be available on the surface of the shale. Equally important is that changing the particle size while keeping other conditions constant could influence the heat and mass transfer processes within oil shale particles. Such results are in full agreement with those reported for the Ellujjun and some non-Jordanian shales by various researchers [10–12].

In the case of oil shale drying under direct normal insolation, due to the dark (black) colour of Jordanian oil shales which enhances the absorption of solar radiation, shale temperature was within the range of 60 to 70 °C or higher during mid of the day. At such relatively high temperatures oil shale samples lost a fraction of its moisture – see the Table.

As can be seen from it, average weight loss was reduced when oil shale samples were covered with a naturally ventilated chamber made of cardboard, which simulate storage in silos or storehouses. This is a logical result as the samples were heated indirectly, not as those placed in the glass-greenhouse, which allows most of solar radiation to pass through the glass walls. The results from the samples left uncovered are a bit too high and can be considered far from the reality. Such a result represents not only the loss of evaporated moisture to ambient air, but accounts also for some loss of fine dust from oil shale samples due to the effect of blowing wind.

Average Weight Loss under Direct Solar Radiation

Experiment	Weight loss, %	
	Ellujjun	Sultani
Greenhouse	0.242	0.261
Cardboard	0.156	0.207
Uncovered	0.843	0.771

Previously, it has been observed at the fixed-bed pyrolysis tests that water starts emerging from oil shale sample within the temperature range 120–160 °C, and the devolatilization of organic matter in the oil shale occurs at temperatures as low as 250 °C and up to ~500 °C [13]. However, TGA and drying furnace experiments showed that the weight loss due to surface water evaporation from the two oil shale samples occurred at relatively lower range of temperatures – between 70 and 130 °C [14]. This difference can be attributed to the dissimilarity of processes happening in the fixed-bed retort and in the TGA analyser as well as in the drying furnace.

For example, in the fixed-bed retort the thermoelement placed in the bed, i.e. being in direct contact with the sample, enables a good temperature control and measurement. However, in the case of the TGA apparatus the thermoelement was placed close to the cell containing oil shale sample; hence the measured temperature is that of the environment surrounding the sample [14].

Also, in the TGA experiments a small amount of oil shale sample was tested, so any heterogeneity or non-uniformity in the distribution of moisture or organic matter will manifest itself in the final results. In addition to the complications caused by simultaneously occurring mineral decomposition reactions noises, in the experimental system due to particle cracking and condensation may affect the final conclusions. Within limitations of experimental error, there is a full agreement between the results obtained from three independent experiments conducted in this investigation.

Conclusion

Drying furnace tests demonstrate that the Sultani shale contains more free moisture than the Ellujjun shale. This result was confirmed by examining the total weight loss in the TGA experiments.

However, the principal conclusion of this study is that for both types of oil shale, within the limits of experimental error, increasing drying temperature and residence time will increase the share of free moisture that can be driven out from oil shale samples. After a certain critical point the rate of evaporation approaches zero.

It was also found that under the studied conditions the particle size has a negative effect on the moisture loss during drying process: it is inversely related to the particle size.

Finally, the results obtained from various tests carried out in this experimental study are in a good agreement.

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