Int. J. Therm. Sci. (2001) 40, 858–863

S1290-0729(01)01272-8/FLA

Free-moisture removal from oil shales

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(Received 25 September 2000, accepted 5 December 2000)

Abstract — In this paper, the drying behavior of two Jordanian oil shales was investigated using three different techniques: a thermogravimetric analyser (TGA), direct solar insolation, and a convection drying furnace. The weight losses and drying rates of the samples were measured gravimetrically. It was noticed that the drying rate falls off at a specific temperature of approximately 120 °C, and becomes zero beyond this temperature. For both types of examined oil shales, the surface-moisture loss is proportional to the drying temperature: the higher the temperature the more of free-water can be driven-off. © 2001 Éditions scientifiques et médicales Elsevier SAS

oil shale / drying / TGA / Jordan

1. INTRODUCTION

Jordan, who lies in the western part of Asia, possesses small known reserves of conventional recoverable energy resources. The domestic natural gas resource, which used to fuel 4·30 MW gas turbines for electricity generation, satisfies less than 4% of the current annual energy demand [1, 2]. Oil shale deposits in Jordan are vast, i.e., proven reserves of about $5 \cdot 10^{10}$ tonnes, and these are not exploited yet. Without the development and utilisation of local oil shales, the corresponding fuel imports will cost about $1.2(\pm 0.2) \cdot 10^9$ US$ per annum, depending on the international unit crude-oil price, at the end of this decade, when the annual rate of fuel consumption in Jordan will probably, if existing trends continue, be approximately twice the present rate [3].

There is little information available concerning the processing of Jordanian oil shales because there has been only little interest, shown from concerned governmental agencies, in developing this resource. This is due to the prevailed low crude oil unit prices compared with those for the final energy produced from oil shale during the last 15 years. Nevertheless, it is likely that, within the next few years this situation will be reversed because of the rising crude oil prices, which are currently more than 30 US$ per barrel [4], i.e., more than twice the price that prevailed two years earlier.

Any industrial complex for the utilisation of oil shale would consist of the following: mining facilities, material-handling equipment, processing plant (for the recovery or conversion of the shale's organic matter) and support systems [5]. The latter include raw-water and waste-water treatment plants, spent-ash cooling and disposal system as well as storage and general facilities. The oil shale, as mined, will require crushing and sizing prior to being fed 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 material-transportation costs, the primary crushers would be located close to the mine, whereas the secondary crushers should be near by the processing site. To avoid shutdowns of the down stream processing units, sufficient quantities of the coarse ore and fine shale should be maintained in strategic stores. This may be achieved by diverting the excess quantity excavated during periods when the ore production exceeds the feed requirements of the plant. For each patch of oil shale, the storage may last for days or weeks, during this period, significant changes may occur, the most important being the loss of carbon dust and the reduction of moisture con-

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tent. This natural drying, i.e. removal of water from the oil shale, is a vital process because the moisture content of the shale may assume some importance due to the associated energy and the longer time required for the utilisation of oil shale deposits. In practice, two classes of water association, with shale, can be distinguished: surface water and interlayer or retort water. Surface water is that free-moisture that can be removed from the sample when heated to a temperature of about 105 °C, while the bound water is that portion driven off only at significantly higher temperatures, as those encountered in the retorting process [6, 7]. Jordanian oil shales, as mined, contain 3–5% of moisture by weight [8, 9], the exact value being dependent on composition, characteristics and structure of the oil shale deposit, and hence upon the location of the mine. In any process aiming to utilise oil shale, its moisture content may cause sticking problems either during preparation and storage or transport. Hence an understanding of drying characteristics is desirable in order to enable to choose or design the required drying system effectively.

Generally, for Jordanian oil shales, the bound moisture content exceeds the free moisture content, i.e., as with Colorado, USA, oil shales. However, there is a dearth of published information concerning the retention of moisture in shales, and even less is known about the kinetics of moisture removal, especially for Jordanian shales [10–12]. Thus prime aim of the present experimental study is to dry various oil shale samples, from the Ellujjun and Sultani deposits in the central part of Jordan, using a convective furnace, direct solar radiation, and thermogravimetric analyser. The weight losses, i.e., the free-moisture content as percentage of the original mass of the sample, as a result of heating under various conditions were determined. This information facilitates the design and operation of an efficient oil shale handling, storage and processing systems.

2. EXPERIMENTAL METHOD

2.1. Oil shale samples

In this study, the employed representative samples, from the Ellujjun and Sultani deposits about 120 and 150 km, respectively, south of the capital city, Amman, were provided by the Natural Resources Authority. Each of the as received samples was crushed, and without further treatment, then sieved into two categories, i.e., grain size $\leq$ 0.85 mm, which can be considered as granular-powder, and 0.85–3.36 mm.

2.2. TGA analysis

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

For each drying test, which was carried out non-isothermally, a small sample of about $15{\cdot}10^{-6}$ kg, was 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 pre-programmed control unit regulates all the automatic functions of the recorder, e.g., the continuous change in the mass of the sample is measured, as well as the temperature programming of the furnace. Finally, and after the furnace temperature had achieved its set value, i.e., 150 °C, the sample was allowed to cool to normal room temperature.

2.3. Convection drying oven

The laboratory-scale convective 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 was controlled by a programmable temperature controller. This enabled final temperatures up to a maximum of about 150 °C to be achieved, and the sample held at the desired final-temperature for a predetermined period in order to remove the free-water content from the shale. The pre-weighed oil shale sample, of $\sim 150{\cdot}10^{-6}$ kg, was placed in a ceramic crucible, and which was then inserted into 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 of the sample.

Additional set of experiments was carried out to determine the effectiveness of employing direct solar radiation for drying the samples—a desirable alternative as the required drying energy is readily available and free in Jordan. Three ceramic boats, containing similar oil shale samples, were used to simulate different possible storage techniques, as follows: first sample was covered with a ventilated greenhouse made of glass, the second one was placed in a chamber made of cardboard, with two slots in the top to allow for free air movement, in order to simulate storage in silos and the last sample was left uncovered in the ambient environment to represent external-stockpiling of crushed oil shale. These were placed together in a very large open-top cardboard box, and left for one week on the roof of the Institute of Land, Water and Environment, at the Hashemite University, during July 2000, where the dry-bulb temperature in the shade exceeded 40 °C, to minimise effects of weather and surrounding environment. This was a comparative experiment; but it is worth noting that in the oven and direct solar drying experiments, the influences of water content in the ambient air on the drying rate being ignored. In this investigation, an attempt has been made to determine the effects of different variables, such as temperature and particle size on the drying profile of two Jordanian oil shales.

In order to reduce the margin of error and so produce more reliable data, each individual test was repeated three times, at least, and then the average result was taken. However, this study was carried out in a laboratory-scale drying furnace and 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 an actual commercial oil shale storage and processing facility. Nevertheless, the presently identified qualitative trends, can serve as reliable guidelines. But more detailed research concerning the properties and behaviours of Jordanian oil shales is still needed.

3. RESULTS AND DISCUSSION

The association of water with its host mineral matrix is due to several mechanisms, e.g., the weak physical-interactions arising from surface tension to the much stronger stable chemical bounds which occurs in the hydrates. Drying consists of the simultaneous heat- and mass transfers 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 particles, then by conduction into the core of each oil shale particle. Simultaneously, mass is transferred from the solid surface by evaporation. For the initial phase of the drying process, almost all the heat transferred is stored in the particle as sensible heat, so its temperature rises. Once the solid-particle's surface temperature reaches about 70–80 °C, part of the transferred heat will provide the heat capacity, i.e., latent heat, required for vaporisation of the free-water content. Hence the particle starts losing some of its affiliated moisture.

Figures 1 and *2* show the weight loss thermogravimetry, i.e., TGA curves, for Ellujjun and Sultani oil shales respectively. The rate of weight loss due to the evaporating moisture is directly related to the drying temperature:

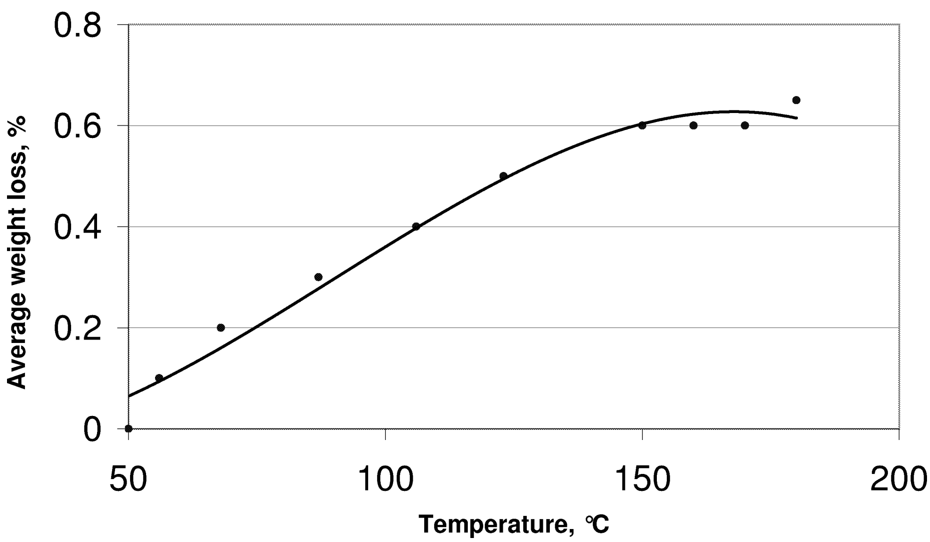

Figure 1. TG profile of the Ellujjun oil shale with respect to drying temperature.

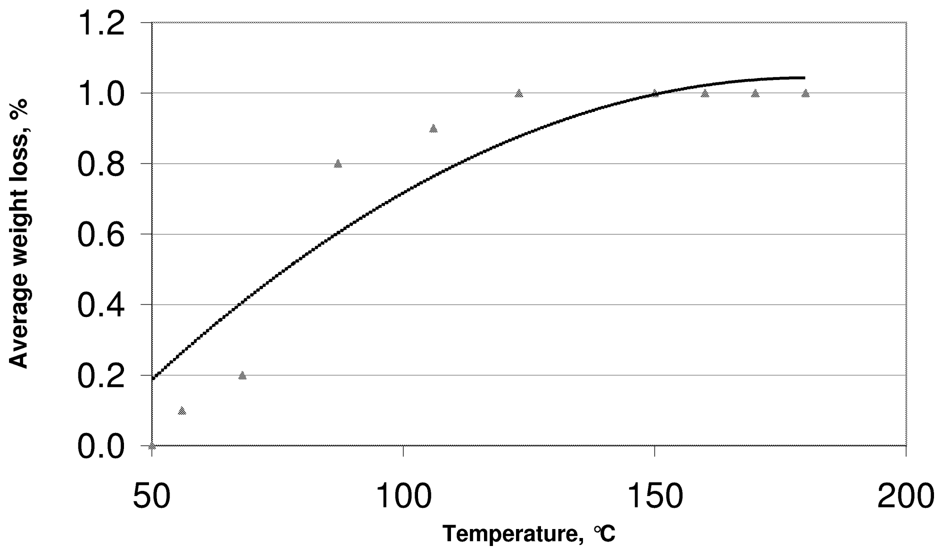

Figure 2. TG profile of the Sultani oil shale with respect to drying temperature.

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 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 movement of moisture within the particle. As the final drying temperature increased from 70 to about 150 °C, while keeping the other experimental parameters constant, the overall yield of water, i.e., sample's weight loss, increased by 3 to 5 times as that obtained at 70 °C 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.

From the experimental data, by regression and best-fit line approach, a polynomial model can be assumed to relate the lost moisture to the drying temperature or time. The weight loss, i.e., free moisture, curve follows the second-order profile—see *figure 3*.

Figure 4 presents weight-loss data, obtained by employing a laboratory-scale drying oven, for both shale specimens as a function of temperature and for the same particle size: increasing drying temperature resulted in higher moisture loss. As can been seen that there is a slight jump at a temperature of 120–130 °C, this is most probably due to the mass transfer from core, i.e., under surface layer, of oil shale particles to their surface. But examination of these curves reveals that there is a slight difference in samples' lost portion, as with TGA results, between the two shales. But, the important feature of such figure is that the total surface moisture contents in the Ellujjun and Sultani oil shales is approximately 0.8% of the initial samples weight. This result is in agreement with that obtained from the TGA experiments, which showed that the free water content was slightly higher for the Sultani shale. This indicates clearly that a little more time should be allowed for drying the Sultani oil shale for most of the moisture content to be driven out. Increasing residence time at final drying temperature, from half-hour to one hour, ended up with a positive increase, i.e., about 4 to 7% for the Ellujjun and Sultani shales respectively, in the extracted free moisture. The opposite occurred when larger particle grain size, i.e., 0.85–2.36 mm, was used: a net reduction in lost water of about 35 and 40% for the Ellujjun and Sultani shales respectively, compared with those for fine particles of less than 0.85 mm in size. This occurs because the small particles have a greater surface area per unit mass and so more free water would be available on the surface of the shale. Equally

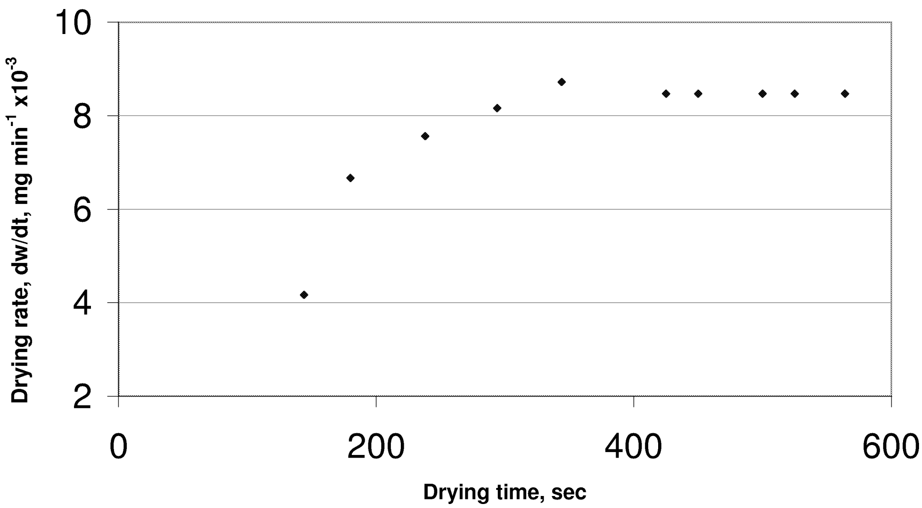

Figure 3. Drying-rate curve for the Ellujjun oil shale with respect to drying time.

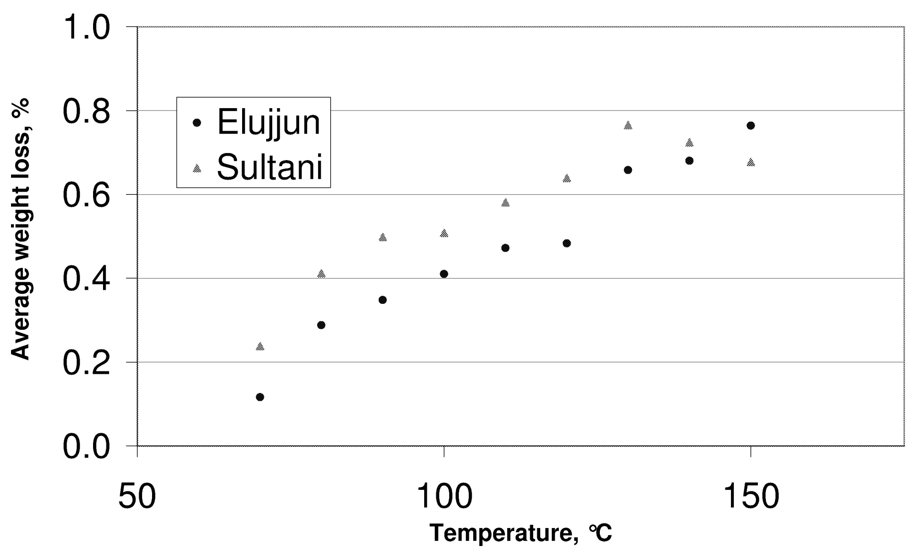

Figure 4. Average loss of free-moisture from the Ellujjun and Sultani shales under isothermal conditions and constant residence time.

important is that changing the particle size, and 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 other non-Jordanian shales by different researchers [10–12].

In the case of oil shale drying under direct normal solar insolation, and due to the dark color, i.e., black, of Jordanian oil shales, which enhances the absorption of solar radiation, its temperature was within the range of between 60 and 70 °C, or higher during mid of the day. At such a relatively high temperatures oil shale samples did lose a fraction of its moisture content—see *table I*.

TABLE I
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

As can be seen from *table I*, 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 because samples heated indirectly and not as those placed in the glass-greenhouse, which allows most of solar radiation to pass through the glass walls. Results from samples left uncovered are a bit high and can be considered far from reality. Because such result represents not only evaporated moisture to ambient air, but also some of fine dust from oil shale samples may be lost due to the effect of wind blowing.

Previously, it has been observed from the fixed-bed pyrolysis tests, that water starts emerging from the oil shale sample within the temperature range 120–160 °C, and the devolatilization of organic matter in the oil shale occurred at temperatures as low as 250 °C and up to $\sim$ 500 °C [13]. However, TGA and drying furnace experiments showed that weight loss due to surface water evaporation from the two oil shale samples occurred at relatively lower range of temperatures of between 70 and 130 °C [14]. The difference in these conclusions can be attributed to dissimilarity of what is happening in the fixed bed retort and the TGA analyser as well as the drying furnace. For example, in the fixed bed retort, by placing thermojunction in the bed, i.e., being in direct contact with sample, enabled a good temperature control and measurement. However, in the case of the TGA apparatus the thermojunction was placed close to the cell containing oil shale sample; hence the measured temperature is 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 and noises in the experimental system due to particle cracking and condensation, may affect the final conclusions. With limitation of experimental error, there is a full agreement between results obtained from three independent experiments conducted in this investigation.

4. CONCLUSION

TGA experiments implied that the Sultani shale contains higher free moisture than that for the Ellujjun shale. This was confirmed by examining the total weight loss in the drying furnace tests. However, the main conclusion of this investigation proved for both types of oil shale, within the limits of experimental error, that increasing drying temperature, and residence time, will increase the ratio of free-moisture that can be driven out from oil shale samples, till a critical value, after which the evaporation rate approaches zero. It was found also that particle size, under the studied conditions, is inversely related to the loss of free-moisture, from oil shale particles, during drying process. Finally, there is a full agreement in the results attained from different experiments carried out in this investigation.

Acknowledgement

The authors wish to thank the Natural Resources Authority, for providing required oil shale samples and Eng. N. Morshid, from the Applied Science University, Amman, Jordan, for technical support during this study.

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