Gasification of Jordanian oil shale Using a Nitrogen Non-thermal Plasma

M.Al-Amayreh[1], A.Al-Salaymeh[2], V.Jovicic[1], A.Delgado[1]

[1] Institute of Fluid Mechanics, University of Erlangen- Nürnberg, Germany malik.amayreh@lstm.uni-erlangen.de, Vojislav.Jovicic@lstm.uni-erlangen.de, Antonio.Delgado@lstm.uni-erlangen.de [2] Mechanical Engineering Departments, the University of Jordan, Amman salaymeh@ju.edu.jo

Abstract:

A number of experiments on the non-thermal plasma gasification of Jordanian oil shale have been carried out using a nitrogen plasma jet. Oil shale batches containing average moisture of 2.5%, volatile 18%, fixed carbon 20%, and ash 59.5% were exposed to ion bombardment. This process converts part of the carbon and oxygen into the gas phase. This method provides a main advantage that gasification of the oil shale can be performed without water at low power of 300 W. The basic requirements for oil shale gasification include generating plasma at low temperatures (200-550°C), using a pulsating high voltage (f=20 kHz). Additional rotation of the samples during process increases the surface exposed to the plasma jet. Experimental study was conducted in order to investigate the influence of the main process parameters such as nitrogen plasma gas mass flow (in the range of 7-10 l/min), mean diameter (d) of the oil shale particles (in the range 4.5-10 mm) and distance of the plasma nozzle from the oil shale (5-15 mm). It has been found that the maximum attainable gasification percentage using a nitrogen flow rate of 10 l/min is 23.0% of the original oil shale weight.

Introduction

The gasification of oil shale is the process of converting the solid organic matter into a gas with a calorific value. Oil shale is an important energy resource in Jordan. Much work has been done in order to study the characteristics of this carbonate-rich rock (Hamarneh, 2006; Speers, 1969, Abu-Ajamieh, 1980). The oil shale contains inorganic constituents and organic matter. The inorganic constituents are carbonates, clay minerals, silica, phosphates, and sulphur. Oil shale gasification can be achieved either by heating the oil shale directly using a partial combustion, or indirectly by using an external heat source or heat exchanger. The thermal plasma or electric arc can be used as a source of heat required to gasify the organic matter (Kalinenko, *et al*., 1993). In addition, microwaves or radio waves at very high frequency can also be used in the gasification process (Balanis, 1983). Usually, temperatures in excess of 800°C are required to carry out the gasification process (Higman *et al*., 2008).

In the past few years new patents have discussed the use of non-thermal plasmas in the gasification of carbonates at low temperatures - 300 -700° C (Rabovitser *et al*., 2007). In this method the electron temperature being several times higher than the temperature of heavy particles induces a plasma. Samples of oil shale were obtained from the deposit El-Lajjun, located in the west-central part of Jordan. The kinetics of the thermal decomposition of these samples have been analysed by using a non-isothermal thermogravimetric analyser (TGA). This apparatus heats the oil shale sample at a constant rate and records the weight changes. Also, a non-thermal nitrogen plasma jet, which consumes a low power (300W), was used to convert the organic matter (kerogen) within the oil shale into a gas. The influence of some key factors such as the diameter of the particles, the plasma mass flow rate in the range 7-10 l/min and the separation distance between the nozzle head and the sample have been investigated.

Experimental Setup

A TGA was used to heat non-isothermally a 1 g oil shale sample at a heating rate of 15K/min with purge gas N_2. The results from this system provide general information about the overall reaction kinetics of oil shale. TGA provides the differentiation of the weight signal as a function of temperature. The pre-programmed control unit regulates all the automatic functions of the recorder. The tube furnace was raised up to 1000 °C, and then the sample was allowed to cool to atmospheric temperature.

An atmospheric plasma jet shown in Figure 1 was used to create a non-thermal atmospheric plasma. The high voltage AC generator used is equipped with a voltage rectifier converting the line voltage of 230 V AC at 50/60 Hz into a pulsed high voltage (up to 10 kV at frequency f=20 kHz). The plasma gas must be pressurized between 5-10 bar before it enters the plasma unit. A mass flow controller (BRONKHORST) was used to regulate the mass flow rate of nitrogen in the range 7-10 l/min. A high torque stepper motor (ASTROSYN) was used to rotate the oil shale sample at a specific rotation speed in order to expose a higher sample area to the plasma jet.

About a 10g batch of oil shale was used in the experiment. The oil shale sample was milled followed by sieving at different mean diameters in the range 4.5-10mm. The sample rotates at constant rotational velocity ω, which was defined in a pre-programmed control. The required mass flow rate is defined in the automatic mass flow rate controller box. After rotating the oil shale sample for a limited time *t*, in the plasma, the sample of the oil shale is removed again to calculate the mass percent-

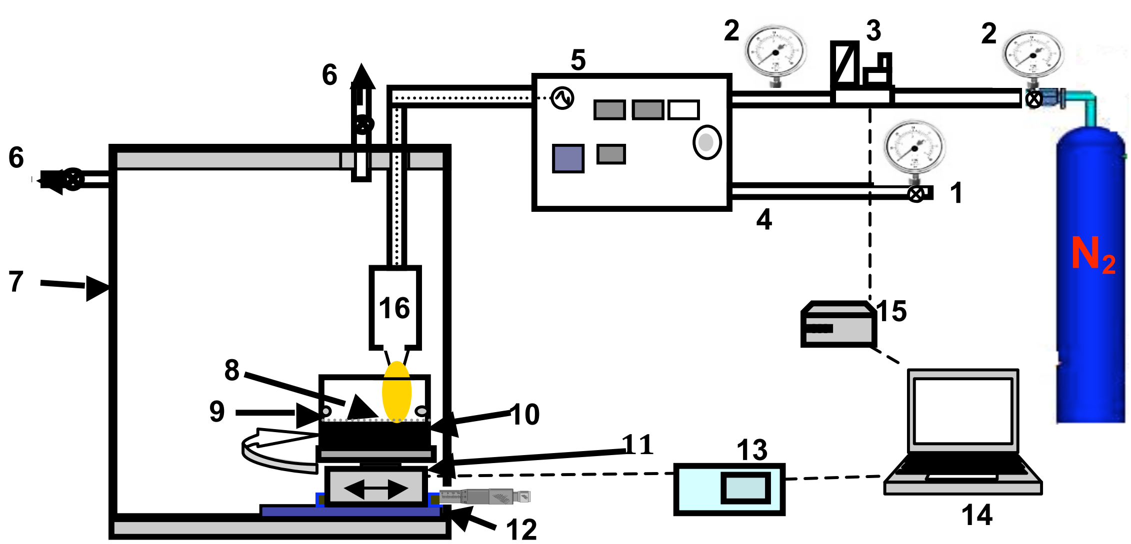

Figure 1: Setup: 1) Dry, oil and dust free, cooling (7 bar) compressed air, 2) N2 pressure regulator with manometer, 3) Mass flow controller (MFC), 4) Cooling air line, 5) High voltage generator unit, 6) Gas outlets to ventilation 7) Protective hood, 8) Wire grid or perforated plate, 9) Fixation ring, 10) Oil shale sample, 11) Stepper motor, 12) Moving base, 13) Motor controller, 14) PC with LabView for process control, 15) MFC control box, 16) Atmospheric pressure plasma jet .

age released.

Results

The results of the TGA appear in Figure 2, which shows the percentage conversion as a function of time. It is clear that due to the conversion of the kerogen, the rate of weight loss increases with increasing temperature. Figure 2 also shows three stages. The first stage is the pre-heating stage *i.e.*, temperature is less than 180°C. In this stage 2% of the mass is lost due to removal of water from the clay minerals. In the second stage (*i.e.*, between 175°C and 560°C) the kerogen is converted into bitumen (Burnham, et al., 1983; Williams and Nasir, 2000). About 18% of the total mass is lost during this stage from pyrolysis of the oil shale, and the sample color converts to black. In the third stage (i.e., temperatures exceeding 560°C) about 20% of the rock mass is lost due to carbonate decomposition. The carbonate includes calcite, dolomite and ankerite (Haddadin and Mizyed, 1974; Jaber and Probert, 1999).

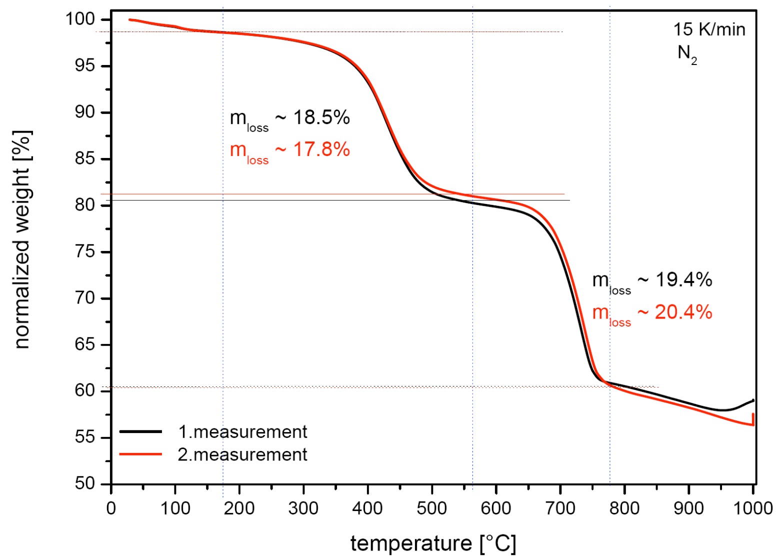

Figure 2: TGA results for two test at the same conditions.

Two parameters that are of utmost importance in the non-thermal plasma are the temperature and the frequency. A rectangular pulsating voltage at 20 kHz with amplitude 10kV has been used. Figure 3 shows the axial temperature profile. The temperature decreases away from the root of the arc plasma.

The influence of nitrogen mass flow rate on oil shale gasification is presented in Figure 4. Oil shale samples with a particle size of 5 mm were subjected to atmospheric plasma at different N_2 mass flow rates. The oil shale sample was placed 5 cm below the plasma nozzle. The motor rotating velocity is fixed at ω=3 rev/min. The rate of conversion of the organic content to gas is

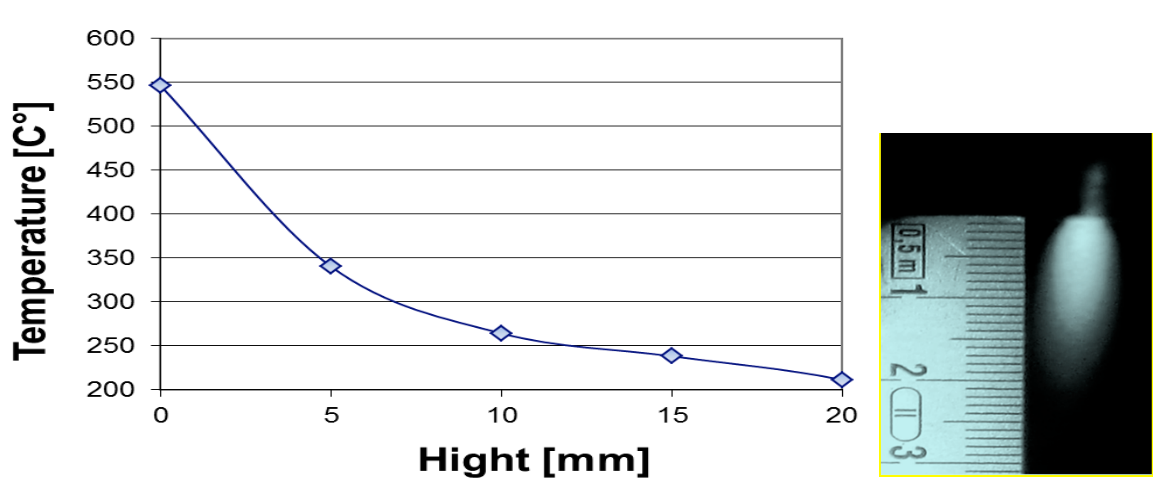

Figure 3: Axial temperature profile measured for volume flow Q=10 l/min.

high during the first six minutes. Then the degree of gasification remains constant due to the fact that no more organic content is converted into gas. As indicated in this figure the maximum value of gasification is 21%. The influence of the mass flow rate on the gasification percentage in the range 7-10 l/min is very small.

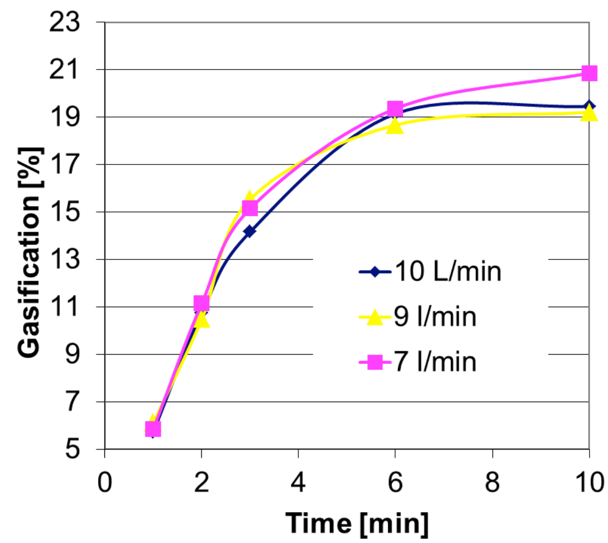

Figure 4: Influence of the plasma gas mass flow rate for h=5 mm, d=5 mm, f=20 kHz, ω=3rev/min.

The influence of the distance between head of the plasma nozzle and the oil shale sample (h) is shown in Figure 5. It can be observed that increasing the distance between head of the plasma nozzle and the oil shale sample results in lower gasification percentages. The effect of particle size on gasification appears in figure 6. As shown, the maximum percentage of gasification was obtained when d = 4.5mm; reducing the particle size increases the surface area to volume ratio. This improves the gasification process.

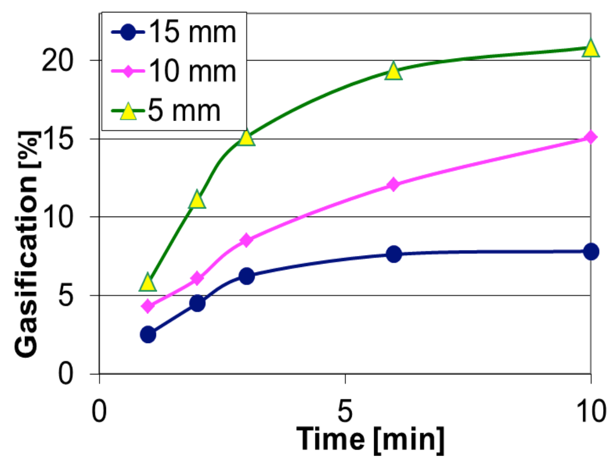

Figure 5: The influence of distance of plasma nozzle from the oil shale on evaporated mass for d=5 mm, f=40 kHz, Q=10 l/min, ω =3 rev/min.

Conclusions

Atmospheric nitrogen non-thermal (cold) plasma jet was successfully used for selective gasification of oil shale samples. Good levels of evaporation were reached using a low energy (300 W) plasma generator. Oil shale gasification using cold plasma was found to be fast, efficient and does not require water or steam. Influences of the most important system and process parameters on the quality of the gasification process were determined:

- Gasification increases with increase of the plasma temperature and (to a certain extent) treatment time.
- Increase of the plasma gas mass flow rate (in the test range of 7-10 l/min) does not affect oil shale gasification significantly.
- Best gasification results were achieved with lower mean diameter of oil shale particles.

Acknowledgments

The authors would like to gratefully acknowledge the Alexander Mayer-Stiftung for the financial support of the first author Malik Al-Amayreh. The technical assistance received from Dr. Kaschta's group at the Institute of polymer materials, Erlangen-Nurnberg University, is much appreciated.

References Cited

Abu-Ajamieh. M. (1980) An Assessment of The El-Lajjun Oil Shale Deposit. Report by the Natural Resources Authority, Amman Jordan.

Balanis, C. A. (1983) Electromagnetic techniques in the development of coal –derived energy sources – A review. The Journal of

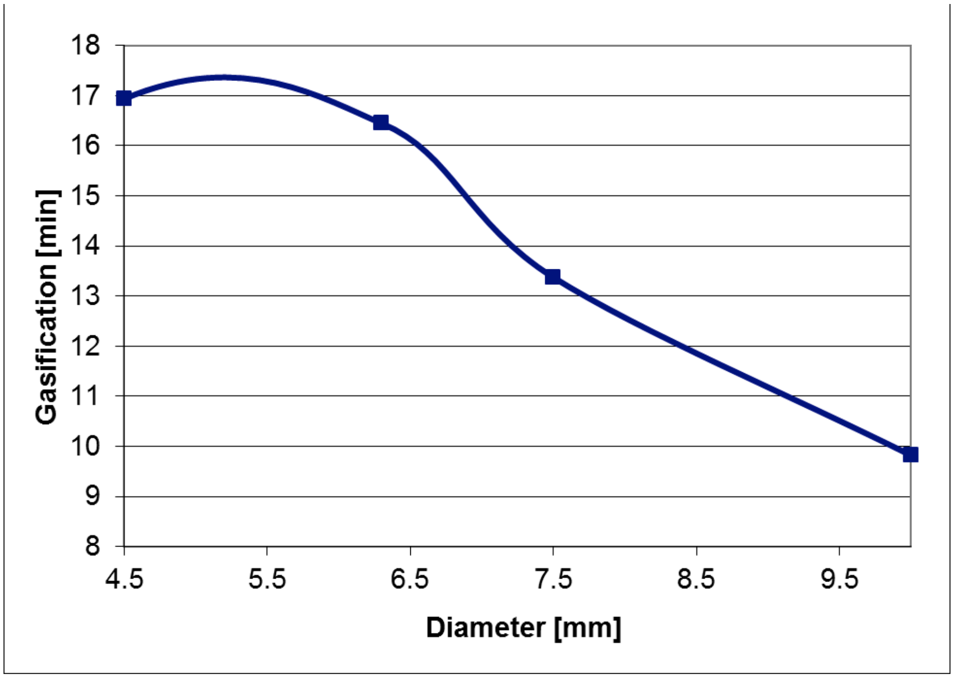

Figure 6: Influence of oil shale particle size h=5mm,ω=3 rev/min.

Microwave Power and Electromagnetic Energy (JMPEE) 1983.18(1) pp 45-53.

Burnham, A. K., Huss, B.E., Singleton, M. F. (1983) *Pyrolysis kinetics for Green River oil shale from the saline zone*. Fuel .Vol 62. pp 1199–1204.

Haddadin, R.A., and Mizyed, F.A. (1974) Thermogravimetric analysis kinetics of Jordan oil shale. Industrial Engineering Chemistry, Process, Design and Development, Vol 13. pp 332–336.

Hamarneh, Y. (2006) Oil Shale Resources Development In Jordan. Natural Resources Authority report, Amman Jordan.

Higman, C., van der Burget, Maarten J. (2008) Gasification. 2nd edition. Elsevier, Oxford ,UK. p. 11.

Jaber, J., and D. Probert (1999). Pyrolysis and gasification kinetics of Jordanian oil shales. Appl Energy Vol 63. pp 269-286.

Kalinenko, R. A., Kuznetsov, A. P., Levitsky, A. A., Messerle, V. E., Mirokhin, Yu. A., Polak, L. S., Sakipov Z. B., and Ustimenko, A. B. (1993) *Pulverized Coal Plasma Gasification*. Plasma Chemistry and Plasma Processing, Vol. 13, No. 1, pp 141-167.

Rabovitser, .L. K, Naster.,S. and Bryan, B. (2007). Plasma assisted conversion of carbonaceous materials into synthesis gas. US Patent 0186472 A1.

Speers, G. C. (1969) El-Lajjun oil shale deposits in Jordan, Unpublished report by the Natural Resources Authority, Amman Jordan.

Williams, P. T., and Nasir, A. (2000) *Investigation of oil-shale pyrolysis processing conditions using thermogravimetric analysis*, Applied Energy, Vol. 66, 2000, pp. 113-133.