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by

Larry L. Gasner
A. O. Denloye
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RESEARCH REPORT

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Larry L. Gasner
A.O. Denloye
Thomas M. Regan

A CONSTITUENT LABORATORY OF
THE SYSTEMS RESEARCH CENTER

THE UNIVERSITY OF MARYLAND
COLLEGE PARK, MARYLAND 20742

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MICROWAVE AND CONVENTIONAL PYROLYSIS OF A BITUMINOUS COAL

LARRY L. GASNER, A.O. DENLOYE† and THOMAS M. REGAN

Department of Chemical and Nuclear Engineering
University of Maryland
College Park, Maryland 20742

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A high volatile bituminous coal was pyrolyzed by use of microwave energy and by traditional convective heating. In both cases the pyrolysis was conducted under vacuum to facilitate product recovery. The gas and liquid fractions produced on pyrolysis were analyzed by means of gas chromatograph.

Processing by microwave energy was not usually successful unless a plasma initiation procedure was employed. This study found the addition of copper wires within the coal mass to be satisfactory for the purpose. Processing by microwave heating resulted in a higher yield of coal tar liquids compared to convective coal heating. The solid char product produced by the microwave process was highly porous and friable, an indication of a suitable feedstock for gasification.
KEYWORDS Pyrolysis Microwave Coal Plasma Gas chromatography

INTRODUCTION

Coal still remains the most abundant fossil fuel in the United States and the world, but its use has been limited mainly by the difficulties of handling solid fuel and of burning coal without pollution. Efforts aimed at converting coal to cleaner gaseous or liquid fuels have recently been intensified. One way of achieving this is the rapid pyrolysis of coal. The rapid heating of finely ground coal to a temperature of about 500 to 600°C leads to an increase in the production of tar fractions, suggesting a possibility of converting coal to liquid hydrocarbons (Probstein and Hicks, 1982). The pyrolysis of coal at temperatures greater than 600°C favor the production of gaseous products. Recent investigations of the pyrolysis of coals in plasmas using plasma jets (Nicholson and Littlewood, 1972; Mueller and Peuckert, 1981), laser beams (Hanson, 1978; Karn *et al.*, 1972, Joy *et al.*, 1970) and high intensity D.C. arcs (Gannon *et al.* 1970) have shown that very rapid pyrolysis of coals in high temperature plasmas produces high yields of acetylene. The use of microwave energy offers the possibility of pyrolyzing coal in a cold plasma where temperature of the gas is much lower than the temperature of the electrons, unlike the situation in high temperature plasmas.

In this paper, the preliminary results of the pyrolysis of a high volatile Kentucky coal by the use of microwave energy are presented and compared to

† Current Address: Department of Chemical Engineering, University of Lagos, Lagos, NIGERIA.

those obtained by traditional convective heating. The experiments were carried out in an evacuated system to increase yields of volatiles and to reduce gas temperature in the microwave reactor.

EXPERIMENTAL

Microwave Oven

The microwave energy was produced by a Sharp home microwave (Model R-7810) oven. The oven had a rated ouptut of 650 W and operates at a frequency of 2450 mHz. The reactor vessel consisted of a pyrex vacuum desiccator, 2.8*L*, containing a 100 ml beaker filled with 40 g of coal. The gas output of the desiccator was passed through a port in the side of the oven into three cold traps maintained at 27°C, 0°C, and −18°C, Figure 1. A vacuum pump connected to the cold traps initially evacuated the chamber to an absolute pressure of about 2 torr in the ractor. The noncondensable gases that passed through the cold traps were collected in a 9.4*L* pyrex desicator. The pressure increase during a run was a primary indicator of noncondensable gas production.

Convection Oven

A Hoskin type FD 2040 muffle furnace was used for the convective heating experiments. A high temperature, high pressure bomb was used as the reaction vessel. The arrangement of the downstream equipment was similar to that for microwave heating. The only difference was that the three cold traps were replaced by one condenser made up of copper coils and maintained at 0°C.

Procedure

A coal sample of approximately 40 gm was chosen to give an aspect ratio, L/D, close to unity in the 100 ml beaker. The beaker was then placed in the vacuum desiccator which was sealed and then connected to the downstream equipment. The system was evacuated to a pressure of about 2 torr, then isolated from the vacuum pump and the microwave oven was activated. The progress of the

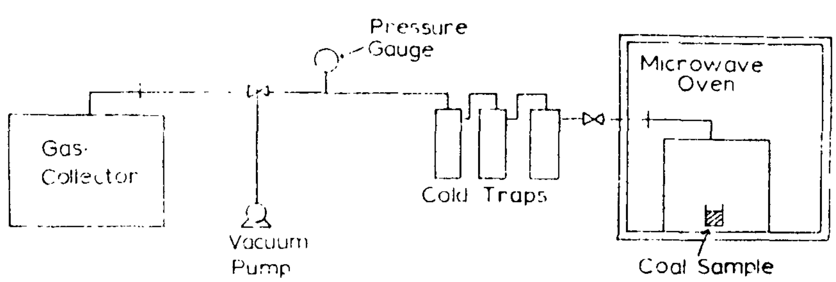

FIGURE 1 Schematic diagram of apparatus for microwave processing

reaction was monitored by the indication of the vacuum gage. A run was terminated when the change in pressure in one minute was negligible, or at a total time of 15 min. The microwave oven was then turned off, and the desiccator disconnected from the system. The remaining char was rapidly quenched with benzene and the benzene fraction collected for analysis. The benzene wash from the cold traps was collected and also analyzed. Liquids and gases were analyzed on a Hewlett Packard 5700 gas chromatograph with a flame ionization detector. A column with a packing consisting of Durapak stationary phase and *n*-octane—porasil C as the solid phase was used for the gas analysis, while a column of Dexsil 300, 3% on Anakrome A was used for the liquid analysis.

The experimental procedure for the experimental runs using the muffle furnace is similar to that described above for microwave heating. The muffle furnace was operated at 760°C and the coal was pyrolyzed for 25 min.

RESULTS AND DISCUSSION

The proximate and ultimate analysis of the Kentucky high volatile bituminous coal used for the studies is given in Table I. The pressure in the reactors increased with time as the coal is pyrolyzed and as gases and tar are evolved, Figure 2. Processing by microwave energy was not usually successful unless a plasma initiation procedure was employed. It was necessary to initiate pyrolysis for the 6 mm to 12 mm size coal particles by inserting two copper wires into the coal mass and parallel to the bottom of the containing vessel. One wire was placed about 10 mm from the bottom and the other positioned about 10 mm from the surface of the coal particles. There was very little gas generation or pyrolysis in the absence of these wires, Figure 2. Introduction of the wires led to the generation of a plasma in the desiccator and to a rapid production of volatiles. There was an initial lag period of about 1 min. during which the plasma developed. This was followed by a period of rapid reaction, and most of the volatiles are evolved from the coal mass within ten minutes. On occasions, arcing occurred instead of plasma generation and this resulted in a slower generation of volatiles from the coal.

TABLE I

Proximate and ultimate analysis of Kentucky high volatile bituminous coal

Ultimate analysis		Proximate analysis	
(Dry basis)	Wt. percent	(Dry basis)	Wt. percent
Carbon	79.35	Ash	6.37
Hydrogen	5.27	Volatile	35.57
Nitrogen	1.59	Fixed carbon	58.06
Chlorine	0.19		
Sulfur	0.55		
Oxygen	6.68		
Ash	6.37		

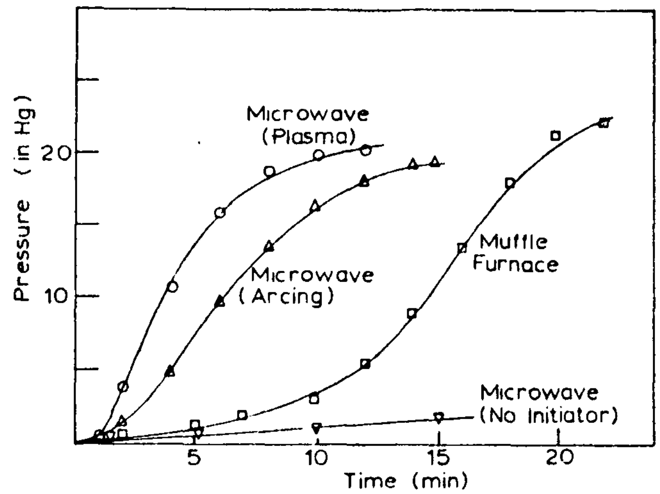

FIGURE 2 Variation of reactor pressure with time of reaction: typical data.

When pulverized coal (less than 100 mesh) was pyrolyzed with microwave energy, pyrolysis without initiation occurred, but was characterized by a lag time of approximately 8 min. The presence of the two parallel copper wires led to a concentrtion of microwave energy within the coal mass and the wires in effect acted as antennae for microwaves.

The gases evolved during the pyrolysis of coal in a microwave plasma consisted of H_2, CO, CO_2 and light hydrocarbons. The hydrocarbon gases consisted mainly of methane, acetylene, ethylene and ethane (Table II). The composition of the gases produced by the convective heating of coal is similar. There was, however, no trace of acetylene in the gas stream in this case. The liquids produced by the microwave pyrolysis consist mainly of heavy hydrocarbons with carbon numbers greater than 20, Figure 3. The liquids darken rapidly indicating the probable presence of free radicals and unsaturation.

The total conversion to gas and liquids produced by microwave pyrolysis and by convective heating was about equal at 30 percent (Table III). About half of the volatiles produced by microwave heating are liquids, while very little liquid is

TABLE II

Typical noncondensable hydrocarbon product gas composition

Component	Microwave Mole %	Convection Mole %
C_1	51.02	51.80
C_2	30 82	35.03
C_3	6.53	3.89
C_4	6.73	5.39
C_5	3.06	2.99
C_6	1.84	0.90

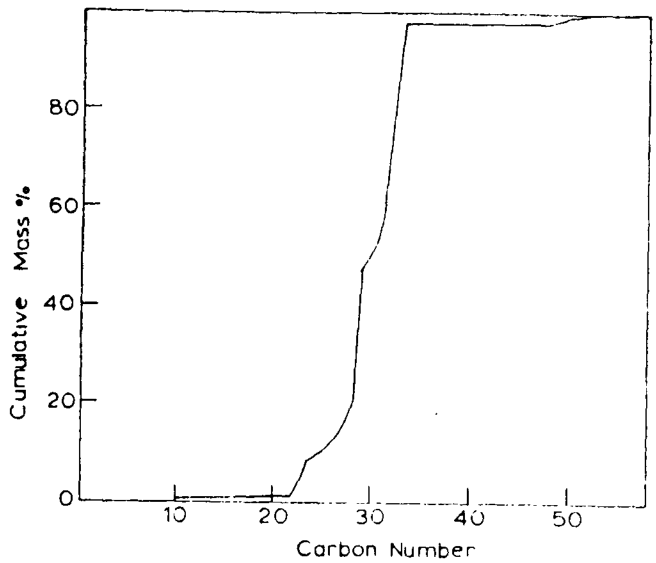

FIGURE 3 Product distribution of liquid product from microwave processing.

produced by convective heating. The char produced by microwave heating was posous and friable and showed no evidence of deformation or having passed through a plastic phase. This is an indication that this char would be a suitable feedstock for either combustion in a fluidized bed or gasification. Alternately, the char could be processed to generate the electricity that would be required for the microwave generation. A preliminary energy balance calculation indicated that coal pyrolysis by microwave energy will be more efficient in the use of energy than pyrolysis in convectional furnaces because of the much lower residence time anticipated in a microwave reactor. Further experimentation is necessary to confirm this.

CONCLUSIONS

Coal of 6 to 12 mm size can be pyrolyzed using microwave energy at fairly low power levels by introducing copper wires into the coal mass to initiate a plasma

TABLE III

Comparison of product yields obtained by microwave and convective heating processing

	Product yield, mass %	
	Microwave	Convection
Gas	17	30
Liquid	14	1
Char	69	69

which promotes the pyrolysis reaction. The total conversion to gases and liquids is comparable to that produced by he pyrolysis of coal in a muffle furnace, but the yields are achieved at much shorter residence times by using microwave energy. The proportion of liquid products produced by microwave energy is, however, much greater than when convective heating was used. Further experiments are necessary to determine thermal efficiency and assess the economics of using microwave energy to pyrolyze coal.

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