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GASIFICATION STUDIES OF CANADIAN COALS IN OXYGEN-ENRICHED ATMOSPHERE

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by

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ABSTRACT

A study of gasification characteristics of five Canadian coals in a fixed-bed gasifier at temperatures of 600 and 800 C is reported.

Bituminous coal samples were Devco, Byron Creek and Prince, and lignite samples were Bienfait and Coronach. When an equal mixture of nitrogen and oxygen was used as a gasifying medium lignite coals reacted more rapidly than bituminous coals at 600 C. Gasification characteristics at 800 C were similar for Devco, Byron Creek and Prince coals although they differed from one another in chemical compositions. A typical composition of the gas product from Devco coal was 36% carbon dioxide, 30% hydrogen, 22% methane and 12% carbon monoxide on nitrogen and oxygen free basis. Bienfait and Coronach lignites behaved similarly at 800 C but their gasification characteristics were different from those of the bituminous coals. A typical composition of the gas product from Bienfait was 32% carbon dioxide, 26% hydrogen, 10% methane and 32% carbon monoxide.

At 800 C lignite coals were more reactive and generated more product gas than bituminous coals in a decreasing order of Coronach, Bienfait, Devco, Byron Creek and Prince coal. Under similar gasification conditions more methane but less carbon monoxide was obtained from bituminous coals than from lignite coals.

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INTRODUCTION

In 1978 a program was initiated at the Energy Research Laboratories (ERL) of the Department of Energy, Mines and Resources of Canada to conduct research and development on gasification technology of Canadian coals. Goals were established at ERL to meet the Department's objectives of achieving energy self-reliance by the substitution of coal for domestic and imported oil.

Specific objectives of the gasification program were to provide sufficient background information on the gasification characteristics of various coals. This information is useful for the provision of advice on the selection of advanced gasification technologies for the conversion of coal to electricity in various regions of Canada. Other areas of major importance are the utilization of low or medium Btu gas for process heat, conversion of the product gas into liquid hydrocarbons, and possible application of the gasification process for the extraction and processing of heavy oils.

Implementation of the gasification program was carried out in two efforts: (i) the external contract effort by the private sector and (ii) the in-house effort by the ERL staff. Outside research and development contracts are intended to provide techno-economic evaluation of gasification applications for both power generation and process industries (1). In-house research is geared towards providing coal reactivity data of Canadian coals which are candidates for use in various gasification processes (2).

This report summarizes the progress of the in-house effort at ERL and presents the results of a gasification study of five Canadian coals. The study was designed to examine a series of Canadian coals in a fixed-bed gasifier under an atmosphere of nitrogen and oxygen and to rank their reactivity towards common gasifying media under isothermal conditions at 600 and 800 C respectively. Reactivity of coal may be defined as the rate at which different forms of carbon react with oxygen, steam or carbon dioxide under uniform conditions. Preliminary data on the chemical composition of the gas product at different stages of the gasification are presented. These data will be used for interpretation of reaction mechanisms of the gasification process and evaluation of the heat content of various coals.

EXPERIMENTAL METHOD

GASIFICATION PROCEDURE

Figure 1 depicts the gasification unit used for this study. A mixture of coal sample (50.0 g) and inert material (25.0 g) was placed in a cylindrical sample holder (4 cm diameter by 30 cm long) which had an opening (6 mm diameter) for the incoming reacting gases at the bottom of the holder. Reacting gases were an equal mixture of nitrogen and oxygen with a total flow rate of 2 liters per minute for all the experiments. Inert material (Burrell Saddles) was used to minimize channelling and caking of the coal sample during gasification. The sample mixture was placed in the sample holder in such a manner that it rested on a supporting disc which had small openings to allow the incoming gases to react with the coal and acted as a mini-grate for the gasification process. Both the sample holder and the reactor tube were made of stainless steel 304.

At the desired temperature, the sample holder was inserted into a tube reactor (5 cm diameter by 1.8 m long) which was heated by an electric furnace. When the sample holder was introduced, the temperature of the tube reactor dropped considerably, due to thermal shock. However, the temperature of the tube reactor rebounded to the original temperature in about 10 minutes for the gasification temperature of 600 C. Four thermocouples, 15 cm apart, were positioned in the reactor tube as shown in Figure 1 to monitor the temperature of the gasification process. A temperature profile of the sample holder as detected by thermocouple No. 1 at various times is shown in Table 1.

The gaseous products from the gasifier passed through a condenser and two cold traps, one at 0 C and the other at -70 C, where the liquid portion of the gasification products was trapped. The uncondensed tar was removed by a cellulosic paper filter in a Balston filter. The gaseous products were passed through a volume of 1000cc of sodium carbonate solution (25% by weight), a filter, drierite and the wet test meter before they were separated and analyzed by gas chromatography. A rubber bulb gas sampler was used to collect the gas sample at different time intervals. Each gasification experiment was 1 hour and duplicate experiments were performed for each coal sample.

Gas Analysis

All gas analyses were performed on a Perkin-Elmer Sigma 1 Analyzer. Carrier gas was a mixture of helium (91.5%) and hydrogen (8.5%) with a flow rate of 18 ml/min at column temperature of 60 C isothermally. Chromosorb 102 and molecular sieve 5A columns were used for the separation and analysis of carbon dioxide, hydrogen, oxygen, nitrogen, methane and carbon monoxide.

Materials

The particle size of the coal sample ranged from 3 mm to 6 mm. All samples were air-dried and oven-dried at 60 C before use. Chemical analyses of coal samples are reported in Table 2.

RESULTS AND DISCUSSION

In 1953 it was suggested by Batchelder and his co-workers (3) that the reaction mechanisms of coal gasification could be grouped into two general types of reactions which are the exothermic and the endothermic reactions. Exothermic reactions include the reactions of oxygen with carbon, hydrogen and carbon monoxide. Endothermic reactions involve the reactions of carbon with carbon dioxide and steam. In this report the reactions of carbon with oxygen will be discussed.

Five Canadian coals were used for this study and their proximate and ultimate analyses are shown in Table 2. Bituminous coals were Byron Creek from British Columbia, Devco and Prince from Nova Scotia. Lignite coals were Bienfait and Coronach from Saskatchewan. Table 3 shows a typical chemical composition in volume per cent of the gas product from Devco, Bienfait and Coronach, at gasification temperatures of 600 and 800 C. For the present study only carbon dioxide, hydrogen, oxygen, nitrogen, methane and carbon monoxide were analyzed. In order to have a better comparison of the gasification performance among these five coals, the composition of the gas product was computed on a nitrogen and oxygen free basis as shown in Table 3.

As it can be seen in Table 3, Devco coal reacted slowly with oxygen in the gasifying medium (50% N_2 + 50% O_2) at 600 C to give 74% carbon dioxide and 21% carbon monoxide. Only 5% of methane was produced and

hydrogen was absent in the product gas. On the other hand, Bienfait lignite reacted more readily at the same temperature to yield 50% carbon dioxide, 21% hydrogen, 7% methane and 22% carbon monoxide. Coronach lignite was gasified to a similar extent as Bienfait and gave 39% carbon dioxide, 23% hydrogen, 5% methane and 33% carbon monoxide. Both hydrogen and methane were generated from lignite coals at 600 C. These data suggest that Bienfait and Coronach lignites are more reactive than Devco bituminous coal in an oxygen-enriched environment at this temperature.

At 800 C (Table 3) the gasification reaction of Devco coal proceeded rapidly and its methane production reached a maximum after 6 minutes of the gasification. Composition of the gas product was 36% carbon dioxide, 30% hydrogen, 22% methane and 12% carbon monoxide. The small amount of oxygen in the gas product indicated that the gasification, or oxidation, was not quite completed at this stage. The other bituminous coals, Byron Creek and Prince, had similar gasification characteristics and the compositions of their gas product were similar to that of the Devco coal. Production of carbon monoxide varied from 10 to 12% among Devco, Byron Creek and Prince coals. Production of carbon dioxide, hydrogen, methane and carbon monoxide from Bienfait lignite was 32, 26, 10 and 32% respectively after 6 minutes of gasification at 800 C. Coronach lignite yielded 27% carbon dioxide, 27% hydrogen, 9% methane and 37% carbon monoxide under same conditions. Hence, under similar gasification conditions and based on the compositions of the gas product, more methane but less carbon monoxide can be obtained from bituminous coals than from lignite coals at 800 C.

Table 4 shows the composition of gas products from three coal samples at the gasification temperature of 600 C for different periods of time. For Devco coal the gasification was slow at 6 minutes but reached a steady state at about 10 minutes, yielding 50% carbon dioxide, 25% hydrogen, 15% methane and 10% carbon monoxide. At 29 minutes, combustion of coal occurred as only carbon dioxide was detected in the gas product. For Bienfait lignite the reaction was moderate during the first 6 minutes of the reaction, producing substantial amount of combustible gases such as hydrogen (23%), methane (5%) and carbon monoxide (33%). As the gasification continued, the amount of methane formation decreased while the hydrogen formation remained relatively constant from 10 to 18 minutes. Formation of

carbon monoxide increased gradually at the expense of methane and hydrogen. Coronach lignite behaved similarly as Bienfait lignite, producing both hydrogen and carbon monoxide in relatively large quantities between the 10 to 18 minute period. Combustion occurred for both lignites after 29 minutes of the gasification.

In general, the ratio of CO/CO_2 , from Devco coal is lower than that from either Bienfait or Coronach at 600 C.

The gasification reaction seemed to move faster at 800 C for both the bituminous coals and the lignite coals as shown in Table 5. Methane production from all five coals was at maximum at 6 minutes. The bituminous coals reacted much slower when compared to the lignite coals but they continued to produce hydrogen and methane beyond 13 minutes of the gasification, producing carbon monoxide and carbon dioxide at a ratio of about 0.4. Although these three bituminous coals were quite different from one another in chemical compositions (Table 2), their gasification characteristics were very similar.

The two lignite coals behaved similarly at 800 C and produced large amounts of carbon dioxide and carbon monoxide at the beginning of the gasification (Table 5). Formation of these oxides could be due to the high oxygen contents in Bienfait (17.27%) and in Coronach (17.73%) as this was also observed by Morris and Kearins in their devolatilization study of sub-bituminous and bituminous coals (4). The ratio of CO/CO_2 in the product gas was high and this suggests that the gasification condition was favorable for the formation of carbon monoxide. However, the gasification reaction slowed down at 13 minutes and produced only carbon dioxide and carbon monoxide as the reaction progressed further. After 33 minutes the combustion reaction became predominant.

Figure 2 compares the volumes of the exit gases from the five coal samples at different times at 800 C. The volume was corrected to that of standard temperature and pressure. It was found that a large amount of gas evolved at the beginning of the gasification and the gas production began to level off at about 25 minutes. At 30 minutes, Coronach generated a total of 81.4 liters of gas, 22.3 liters of which was the product gas and 59.1 liters was the gasifying medium ($\text{N}_2 + \text{O}_2$). Following Coronach in decreasing order were Bienfait, Devco, Byron Creek and Prince, producing respectively

18.8, 8.1, 7.5 and 6.3 liters of the product gas. Except for Byron Creek this order seems to be in accordance with the chemical analyses that coals containing more volatile matters yield larger volumes of gas on gasification. These results also indicate that lignite coals are more reactive since they produce relatively more gas than bituminous coals under similar gasification conditions.

It should be pointed out that a reactive coal may not necessarily be a good quality coal. A good quality coal, bituminous coal in this case, released on gasification more methane of high heating value than a lignite coal, although the latter coal was more reactive especially at the beginning of the gasification at both 600 and 800 C.

Table 6 summarizes the products from the gasification of five Canadian coals at 800 C. This table shows the weight loss, liquid product and gas product from the five coals studied after 60 minutes of the gasification. Liquid product included water and condensable tar in the traps, and uncondensable tar in the filter. The weight of the gas product was obtained by subtracting the weight of the liquid from the weight loss of the sample, assuming that there was no loss of products in the connecting lines between the exit port of the gasifier and the filter (Figure 1). From these results, it appears that lignite coals will lose relatively more weight and produce more gas product than bituminous coals under these gasification conditions.

This study is the first of a series of experiments on the evaluation of the reactivity of various Canadian coals by the gasification process. It provides important information on gasification rates, product distribution and the quality of the gas product from these five coals in an oxygen-enriched environment. Since both oxygen and steam are common gasifying media, the data collected from this work can be used in future gasification studies in which steam is also a gasifying medium. It has been demonstrated that the laboratory fixed-bed gasifier used for this study is a convenient tool for determining the volatilization, gasification and combustion characteristics of Canadian bituminous and lignite coals.

CONCLUSIONS

The ERL fixed-bed gasifier can be used as a convenient tool for the evaluation of the gasification performance of Canadian coals. When an equal mixture of nitrogen and oxygen was used as a gasifying medium, lignite coals reacted more rapidly than bituminous coals at 600 C. Gasification characteristics at 800 C were similar for Devco, Byron Creek and Prince coals although they differed from one another in chemical compositions. Bienfait and Coronach lignites behaved similarly at 800 C but their gasification characteristics were different from those of the bituminous coals.

At 800 C lignite coals were more reactive and generated more product gas than bituminous coals in a decreasing order of Coronach, Bienfait, Devco, Byron Creek and Prince coal under similar gasification conditions. More methane but less carbon monoxide was obtained from bituminous coals than from lignite coals.

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REFERENCES

1. Fung, D.P.C. "A review of ERL gasification research and development contracts"; Division Report ERP/ERL 79-65(TR); CANMET, Energy, Mines and Resources Canada; 1979.
2. Banks, G.N. "Gaseous fuels from coal"; Division Report ERP/ERL79-12(J); CANMET, Energy, Mines and Resources Canada; 1979.
3. Batchelder, H.R. and Bushe, R.M. "Kinetics of coal gasification - proposed mechanism of gasification"; Ind Eng Chem 45:1856-1878; 1953.
4. Morris, J.P. and Kearins, O.L. "Coal volatilization studies in support of the Westinghouse fluidized-bed gasification process"; Fuel 58:465-471; 1979.

Table 1 - Temperature profile of sample holder as detected by
thermocouple No. 1 at different times of gasification

Time (min)	600 C	800 C
1	407	729
6	539	783
10	601	799
13	627	807
16	643	813
19	657	818
33	716	877
46	-	869
60	659	861

Table 2 - Chemical analysis of coal samples

Proximate analysis weight %	Devco (Nova Scotia)	Byron Creek (B.C.)	Prince (Nova Scotia)	Bienfait (Sask.)	Coronach (Sask.)
Ash	2.84	15.26	15.64	12.51	9.59
Volatile	35.43	26.21	34.62	41.35	46.62
Fixed Carbon	61.73	58.53	49.74	46.14	43.79
Ultimate analysis weight %					
Carbon	84.72	74.44	65.97	66.11	70.62
Hydrogen	5.60	4.32	4.48	2.24	0.10
Sulphur	1.25	0.80	4.90	0.55	1.05
Nitrogen	1.25	1.15	1.39	1.32	0.91
Ash	2.84	15.26	15.64	12.51	9.59
Oxygen*	4.34	4.03	7.62	17.27	17.73

*Determined by difference

Table 3 - Composition of gas product after gasification for 6 minutes

Temperature (C)	Coal	% by volume						% by volume (N ₂ , O ₂ , free basis)			
		CO ₂	H ₂	O ₂	N ₂	CH ₄	CO	CO ₂	H ₂	CH ₄	CO
25		0	0	50	50	0	0	0	0	0	0
600	Devco	14	0	11	70	1	4	74	0	5	21
600	Bienfait	28	12	6	38	4	12	50	21	7	22
600	Coronach	24	14	3	36	3	20	39	23	5	33
800	Devco	22	18	6	34	13	7	36	30	22	12
800	Byron Creek	25	16	5	37	11	6	43	28	19	10
800	Prince	26	13	4	39	11	7	46	23	19	12
800	Bienfait	23	19	2	26	7	23	32	26	10	32
800	Coronach	20	20	1	25	7	27	27	27	9	37

Table 4 - Composition of gas product from various coals at 600 C
(% by volume N₂, O₂, free basis)

Time (min)	<u>Devco</u>				<u>Bienfait</u>				<u>Coronach</u>			
	CO ₂	H ₂	CH ₄	CO	CO ₂	H ₂	CH ₄	CO	CO ₂	H ₂	CH ₄	CO
6	74	0	5	21	50	21	7	22	39	23	5	33
10	50	25	15	10	34	24	7	35	42	20	7	31
13	51	24	15	10	29	22	5	44	31	23	5	41
16	59	23	11	7	31	21	1	47	32	22	2	44
18	59	22	10	9	27	17	1	55	30	20	1	49
29	100	0	0	0	58	0	0	42	47	0	0	53
43	100	0	0	0	100	0	0	0	100	0	0	0

Table 5 - Composition of gas product from various coals at 800 C
(% by volume N₂, O₂, free basis)

Time (min)	<u>Devco</u>				<u>Byron Creek</u>				<u>Prince</u>				<u>Bienfait</u>				<u>Coronach</u>			
	CO ₂	H ₂	CH ₄	CO	CO ₂	H ₂	CH ₄	CO	CO ₂	H ₂	CH ₄	CO	CO ₂	H ₂	CH ₄	CO	CO ₂	H ₂	CH ₄	CO
6	36	30	22	12	43	28	19	10	46	23	19	12	32	26	10	32	27	27	9	37
10	35	36	17	12	41	32	12	15	46	27	13	14	22	25	4	49	16	24	4	56
13	39	35	11	15	41	32	8	19	47	30	6	17	17	19	1	63	13	16	1	70
16	56	28	4	12	50	28	2	20	52	24	2	22	22	0	0	78	19	0	0	81
19	63	22	0	15	72	0	0	28	73	0	0	27	25	0	0	75	30	0	0	70
33	99	0	0	1	81	0	0	19	89	0	0	11	97	0	0	3	51	0	0	49
46	100	0	0	0	100	0	0	0	100	0	0	0	100	0	0	0	100	0	0	0

Table 6 - Products from five Canadian coals at 800 C for 1 hour

	Devco	Byron Creek	Prince	Bienfait	Coronach
Weight of starting material, g	50.0	50.0	50.0	50.0	50.0
Weight loss, g	43.0	39.0	41.0	44.0	45.0
Weight of liquid collected in 2 traps and filter, g	5.0	5.0	6.0	4.0	4.0
Coal converted into gas*, g	38.0	34.0	35.0	40.0	41.0

*Determined by difference

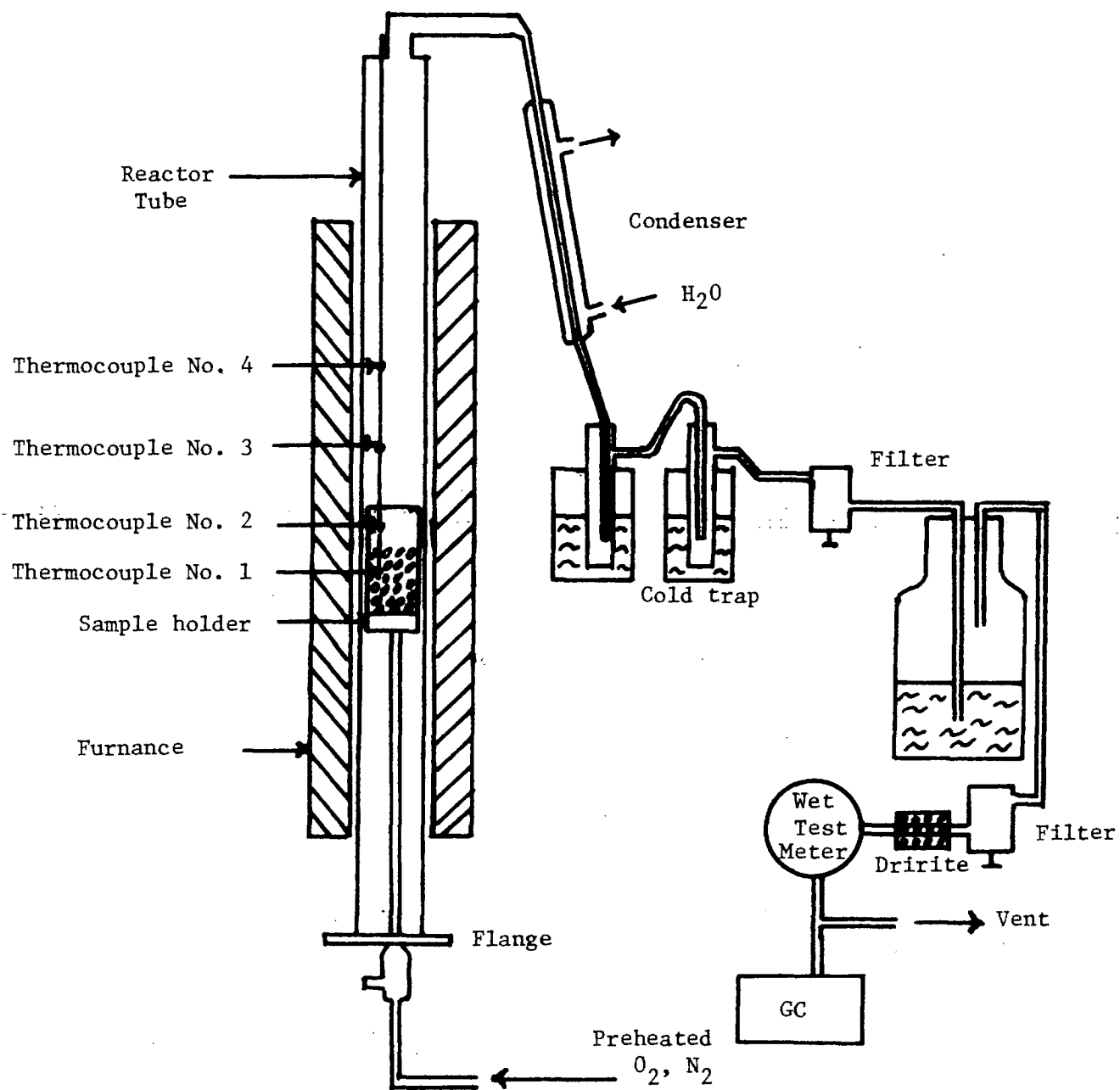


Figure 1. - ERL Gasification Unit

Figure 2. - Volume of gas product at different time intervals at 800°C

