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GASIFICATION STUDY OF NINE CANADIAN COALS

D.P.C. Fung
Hydrocarbon Processing Research Laboratory

January 1982

ENERGY RESEARCH PROGRAM
ENERGY RESEARCH LABORATORIES
DIVISION REPORT ERP/ERL 82-7 (J)

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by

D.P.C. Fung*

ABSTRACT

The chemical reactivity of nine Canadian coals was compared in a laboratory fixed-bed gasifier under two experimental conditions, one with steam at 820°C and the other without steam at 850°C. It has been found that in general the reactivity of a coal increases with decrease in rank. This trend is more pronounced when the gasification is carried out in the absence of steam. The chemical composition of the product gas of these coals from steam gasification is also reported. Each coal has its own gasification characteristics with different products and chemical reactivity. Among the coals studied, lignite coals were most reactive and producted between 30 to 45 vol% of hydrogen and 10 - 13 vol% of carbon monoxide.

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*Research Scientist, Coal Gasification Section, Hydrocarbon Processing Research Laboratory, Energy Research Laboratories, CANMET, Energy, Mines and Resources Canada, Ottawa.

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INTRODUCTION

Canada has large coal resources and this enormous reserve can be used as an alternate fuel for the substitution of the more scarce commodity such as oil and natural gas. Recent development in gasification technology allows an expanded range of uses for coal to produce a low to medium calorific value gas for industrial use, or synthesis gas for production of liquid fuels and chemicals. The Energy Research Laboratories (ERL) of the Department of Energy, Mines and Resources, Canada, participates in the national coal conversion R & D program dedicated to achieving energy self-reliance. In-house research at ERL is geared towards providing coal reactivity data of Canadian coals which are candidates for use in various gasification processes. First part of a series of gasification studies of five coals under the in-house activities has been reported (2). This report describes the gasification characteristics of three bituminous coals and two lignites in an oxygen enriched atmosphere at 850^{o}C in a laboratory fixed-bed gasifier. Oxygen was used to speed up the gasification process and lower the nitrogen content in the product gas. With this gasifier, the chemical reactivity, or the rate of gasification and combustion, of these five coals was measured in terms of the rate of fixed carbon (FC) removal under similar conditions. This rate was calculated from the chemical composition of the coal, product gas and product gas volume. It was found that the overall rates of the gasification and combustion processes for Devco, Byron Creek, Prince, Bienfait and Coronach were respectively 1.25, 1.70, 1.84, 2.80 and 3.20 wt% FC conversion per minute at 850^{o} C.

This paper presents additional chemical reactivity data of coal samples from the rank of semi-anthracite, bituminous, subbituminous and lignite under two sets of gasification conditions. One was under an atmosphere of nitrogen and oxygen at 850^{o}C and the other under an atmosphere of steam, oxygen and air at 820^{o}C. Experimental data on the chemical composition of the product gas from steam gasification are also presented.

EXPERIMENTAL METHOD

Gasification Procedure

Materials

The particle size of the coal sample ranges from 3 mm to 6 mm. All samples were air-dried, then oven-dried for two hours at 105^{o}C before use. Chemical analyses of coal samples are reported in Table 1. Canmore (semi-anthracite), McIntyre (bituminous) and Coalspur (subbituminous) were from Alberta, Prince (bituminous) and Devco (bituminous) from Nova Scotia, Bienfait (lignite) and Coronach (lignite) from Saskatchewan and Onakawana (lignite) from Ontario. Figure 1 shows the occurences of these coals in Canada.

Gasification Unit

Figure 2 depicts the laboratory gasifier used for the experiments. The gasification prodecure was described in detail in a previous communication elsewhere (2). Briefly a mixture of coal sample (50.0 g) and Berl Saddles (25.0 g) was gasified in a cylindrical sample holder (4 cm diam x 30 cm long) which had an opening (6 mm diam) for the incoming reacting gases at the bottom of the holder. Two reacting gas mixtures were used; the first one consisted of an equal mixture of nitrogen and oxygen with a total flow rate of 1.8 ℓ min^{-1} and the second one contained a mixture of air (1.5 ℓ min^{-1}), oxygen (0.5 ℓ min^{-1}) and steam (6.0 cm^{3} min^{-1}). The gasification temperature for the first set of experiments ranged from 800 to 860^{o}C with an average temperature of 850^{o}C and for the second set of experiments with steam ranges from 800 to 840^{o}C with an average temperature of 820^{o}C.

Duplicate experiments were performed for each coal sample. Gasification time was 30 min. The residue in the reactor was cooled with a stream of Nitrogen (0.9 ℓ min^{-1}) for two hours before it was removed and weighed.

Gas Sampling and Analysis

A commercial Valco 16-port valve system was used for gas sampling and storage during the 30 min period of the gasification. Operation of this system in conjunction with a gas chromatograph unit has been described elsewhere (3). With this system it was possible to collect 15 samples at two-min intervals and analyze the samples individually at the end of the gasification.

Perkin-Elmer Sigma 1 Analyzer equipped with Porapak N and molecular sieve 5A columns were used for all the analyses. Carrier gas was a mixture of helium and hydrogen (8.5 vol %) with a flow rate of 45 cm^3 min^{-1} at column temperature programming at a rate of 15^oC min^{-1} from 40 to 75^oC.

RESULTS AND DISCUSSION

Chemical Reactivity

The chemical reactions of coal gasification process are very complex but it is generally believed that four main reactions are involved as follows(4):

$$3C + 2O_2 \longrightarrow 2CO + CO_2$$
$$C + H_2O \longrightarrow CO + H_2$$
$$C + CO_2 \longrightarrow 2CO$$
$$C + 2H_2 \longrightarrow CH_4$$

The end products from carbon-oxidation reaction are mainly carbon monoxide and carbon dioxide. Carbon also reacts with steam to give hydrogen and carbon monoxide. When carbon reacts with carbon dioxide more carbon monoxide is produced. At high pressure methane forms as a result of the reaction between carbon and hydrogen.

In general, the rate of reaction and chemical composition of the product gas depend on the coal reactivity and gasification conditions. It has been found in our previous study that lignites are more reactive than bituminous coals at $850^{o}C$ in an oxygen enriched environment. Additional gasification study on semi-anthracite and subbituminous coals was performed and the results are reported in Table 2. This Table summarizes the chemical reactivities of nine coals in the presence of oxygen and nitrogen at $850^{o}C$. The chemical reactivity of a coal sample, or the rate of gasification and combustion, is the rate of FC conversion per unit time within the 30 min gasification period as defined previously.

It has been determined that the chemical reactivity for Devco, Byron Creek, Prince, Bienfait and Coronach are 1.25, 1.70, 1.84, 2.80 and 3.20 wt % carbon conversion min^{-1}. In this paper new reactivity data for Canmore, McIntyre, Coalspur and Onakawana are included so that a comparison of chemical reactivity of coals from the rank of semi-anthracite, bituminous, sub-bituminous and lignite can be made.

An example of the method of calculation for the chemical reactivity of Canmore is shown below:

$$\frac{\text{Total carbon conversion at 30 min wt\%} - \text{Carbon in volatile matter, wt\%}}{\text{Gasification time, min} - \text{Devolatilization time, min}} \times \frac{\text{Total carbon in sample, wt\%}}{\text{FC in sample, wt\%}}$$

$$= \frac{34.6 - 5.6}{30.0 - 7.0} \times \frac{78.6}{73.0}$$

$$= 1.36 \text{ wt\% FC conversion min}^{-1}$$

With this method, the chemical reactivity of the high rank coal Canmore (semi-anthracite) was determined and its rate of gasification and combustion was 1.36 wt% FC conversion per min. On the other hand, Onakawana, a low rank lignite had a rate of 3.84 wt% of FC conversion per min under similar gasification conditions. With the exception of Devco, the trend is that the chemical reactivity of coal increases with decrease in rank in the order of Devo, Canmore, McIntyre, Byron Creek, Prince, Coalspur, Bienfait, Coronach and Onakawana lignite. This trend was further investigated under gasification conditions with steam, oxygen and air.

Chemical Reactivity with Steam

Table 3 summarizes the results of chemical reacticity of nine coals at a gasification temperature of 820^{o}C in the presence of steam, oxygen and air. These results also indicate that the higher rank coals are less reactive as demonstrated in those experiments performed at 850^{o}C, without steam (Table 2). The chemical reacticity of these coals decreases in the order of semi-anthracite and bituminous, subbituminous and lignite coal. The difference in reactivity between semi-anthracite and bituminous coals was small and could not be distinguished clearly under the relatively mild gasification condition at 820^{o}C with oxygen (0.5 ℓ min^{-1}) and air (1.5 ℓ min^{-1}). Nonetheless, the data in Table 3 show that there is a relation between the chemical reactivity of coal and its rank at 820^{o}C.

Chemical Composition of Product Gas

Table 4 lists the chemical composition of the product gas from steam gasification at 820°C of the nine coal samples under investigation. The semi-anthracite coal, Canmore, reacted slowly at this temperature and the gasification reaction took about 16 min to reach a steady-state which lasted to 30 min. The product gas at steady-state had a chemical composition of hydrogen, 18; oxygen, 1; nitrogen, 44; carbon dioxide, 20; and carbon monoxide, 17 vol%. McIntyre had similar chemical reactivity and composition of the product gas as Canmore. The product gas from Devco contained more hydrogen but less carbon monoxide than Canmore and McIntyre.

Byron Creek and Prince had similar product gases which contained relatively less hydrogen and carbon monoxide than those from Canmore, McIntyre and Devco.

It is interesting to note that the subbituminous coal, Coalspur, and lignite coals, Bienfait, Coronach, and Onakawana, produced relatively more hydrogen than those from the higher ranking coals. The increased amount of hydrogen was produced at the expense of nitrogen and hydrogen can be used for hydrogen cracking and coal liquefaction processes. Onakawana lignite yielded hydrogen, 45; nitrogen, 22, methane, 1; carbon dioxide, 19; and carbon monoxide, 13 vol%, at 820°C with steam,oxygen and air as reacting gases between the period from 8 to 16 min of the gasification,

Carbon balance

Table 5 summarizes the material balance of the gasification study of nine coals with steam, oxygen and air. This Table shows the carbon distribution in the residue, tar, solid particulate and gaseous products. The carbon content in the coal residue was determined by chemical analysis. The carbon in the product gas was calculated cumulatively from the volume of the product gas and its chemical composition throughout the entire 30 min period. The tar was estimated to contain 80 wt% of carbon and the amount of tar was taken from respective gasification experiments without steam in a previous study (2). In general the carbon balance is satisfactory for all the nine coals studied, ranging from 83% of Onakawana lignite to 97% of Coronach lignite. The relatively higher carbon content in the residues of Canmore, McIntyre, Devco, Byron Creek and Prince coal confirm that these coals are less reactive when compared to Coalspur, Bienfait, Coronach, and Onakawana lignite.

Figure 3 shows the relation between the chemical reactivity and rank of the 9 coals gasified with and without steam. In general the chemical reactivity of coal decreases with its rank. However, this relation is more pronounced in the case of steam gasification in which there are some differences in reactivities among the bituminous coals (Figure 2). This kind of relation has also been observed by Hippo and Walker (5) in their reactivity study of chars prepared from coals of different ranks at 500 and 900^{0}C. These workers suggested that the higher reactivity of lignites was partly attributed to a large percentage of pore volume in the macro and transitional pores in lignites.

CONCLUSION

A relationship between the chemical reactivity of nine Canadian coals and their rank has been established under two gasification conditions. In general the reactivity of a coal increases with decreasing in rank. This trend is more pronounced when the gasificaiton is carried out in the absence of steam.

The chemical composition of the product gas of these coals from steam gasification has been determined. Each coal has its own gasification characteristics with different products and chemical reactivity. Among the coals studied, lignite coals were most reactive and produced between 39 to 45 vol% of hydrogen and 10 to 13 vol% of carbon monoxide.

ACKNOWLEDGEMENTS

The author thanks Mr. M. W. Channing for performing the experiments and Mr. G. V. Sirianni for many helpful suggestions during the course of the work.

REFERENCES

1. Bielenstein, H.U., Chrismas, L.P., Lantour, B.A. and Tibbetts, T.E. Report ER 79-9, CANMET, Energy, Mines and Resources Canada, 1979.

2. Fung, D.P.C. Fuel, 1982 (in press).

3. Fung, D.P.C. and Channing, M.W. J. Chromatgr. Sci. 1982, (in press).

4. Mahajan, O.P., Yarzab, R and Walker, Jr., P.L. Fuel, 1978, 57, 643.

5. Hippo, E., and Walker, Jr., P.L. Fuel, 1975, 54, 245.

TABLE 1 -- Chemical analysis of coal samples (moisture free)

Coal (Rank)	Proximate Analysis (wt%)			Ultimate Analysis (wt%)					
	Ash	Volatile matter	FC[a]	C	H	S	N	Ash	O[a]
Canmore (semi-anthracite)	15.1	11.9	73.0	78.6	3.6	0.8	1.5	15.1	0.4
McIntyre (low volatile bituminous)	8.2	18.1	73.7	83.1	4.3	0.6	1.2	8.2	2.6
Devco (high volatile bituminous)	2.9	35.4	61.7	84.7	5.6	1.3	1.3	2.8	4.3
Byron Creek (Med. volatile bituminous)	15.3	26.2	58.5	74.4	4.3	0.8	1.2	15.3	4.0
Prince (High volatile bituminous)	15.6	43.6	49.8	66.0	4.5	4.9	1.4	15.6	7.6
Coalspur (sub-bituminous B)	9.5	37.1	53.4	71.9	4.7	0.2	1.1	9.5	12.6
Bienfait (lignite)	12.5	41.4	46.1	66.1	2.2	0.6	1.3	12.5	17.3
Coronach (lignite)	13.4	43.5	43.1	61.1	3.6	1.1	1.0	13.4	19.8
Onakawana (lignite)	26.8	40.0	33.2	49.7	3.3	5.4	0.7	26.8	14.1

[a] Determined by difference

TABLE 2 -- Chemical reactivity of nine Canadian coals in oxygen and nitrogen atmosphere at 850°C

Coal	Carbon in volatile matter wt %	Devolatilization time, t_D min	FC removal between t_D and 30 min, wt%	FC Conversion wt % min^{-1}
Canmore	5.6	7.0	31.2	1.36
McIntyre	9.4	8.2	30.9	1.42
Devco	23.0	15.1	18.7	1.25
Byron Creek	15.9	9.4	35.2	1.70
Prince	16.2	10.3	36.2	1.84
Coalspur	18.5	12.0	46.2	2.57
Bienfait	20.0	6.0	67.2	2.80
Coronach	18.0	5.4	78.7	3.20
Onakawana	16.5	5.0	96.1	3.84

TABLE 3 -- Chemical reactivity of nine Canadian coals in steam, oxygen and air at 820°C

Sample	FC removal between t_D and 30 min, wt%	FC conversion wt% min^{-1}
Canmore	33.0	1.40
McIntyre	30.6	1.42
Devco	16.8	1.40
Byron Creek	24.7	1.37
Prince	20.3	1.45
Coalspur	28.4	1.67
Bienfait	61.7	2.80
Coronach	68.5	2.84
Onakawana	87.1	3.44

TABLE 4 -- Chemical composition of product gas from steam gasification study

Sample	Steady State duration time, min	Composition of product gas, vol %					
		H_2(SD[a])	O_2	N_2	CH_4	CO_2	CO
Canmore	16-30	18(4)	1(0)	44(5)	0(0)	20(1)	17(3)
McIntyre	16-30	17(2)	1(0)	46(4)	1(1)	20(1)	15(3)
Devco	12-20	22(4)	1(0)	41(3)	3(3)	23(3)	10(2)
Byron Creek	8-16	16(4)	1(0)	46(4)	2(3)	27(3)	8(0)
Prince	10-22	14(3)	1(0)	52(5)	3(3)	21(1)	9(2)
Coalspur	10-24	31(2)	0(0)	36(4)	2(2)	21(1)	10(1)
Bienfait	8-18	45(3)	0(0)	22(1)	1(1)	19(1)	13(2)
Coronach	8-18	39(4)	1(0)	24(3)	1(0)	25(2)	10(2)
Onakawana	8-16	45(3)	0(0)	22(1)	1(1)	19(1)	13(3)

[a] Standard deviation of 2 experiments.

TABLE 5 --Carbon balance of steam gasification study

Sample	C in Sample (g)	Gasification Product (g) C in residue	C in solid	C in tar[a]	C in gas	$\frac{\text{C in product}}{\text{C in sample}} \times 100\%$	SD[b]
Canmore	39.6	18.1	0.1	3.8	14.3	92	4
McIntyre	42.1	18.9	0.1	2.9	15.4	89	1
Devco	42.6	19.7	1.5	4.6	15.0	96	6
Byron Creek	38.4	14.2	1.0	4.6	13.6	87	0
Prince	33.3	15.0	1.4	4.8	10.5	95	0
Coalspur	36.2	11.6	1.7	4.8	14.3	90	5
Bienfait	33.3	0.6	1.8	7.5	21.0	93	1
Coronach	31.1	0.2	3.1	5.4	21.4	97	3
Onakawana	25.3	0.1	0.2	1.8	18.8	83	1

[a] The amount of tar was found in respective gasification experiments without steam and used here. This tar was estimated to contain 80 wt% for the calculation.

[b] Standard deviation of 2 experiments.

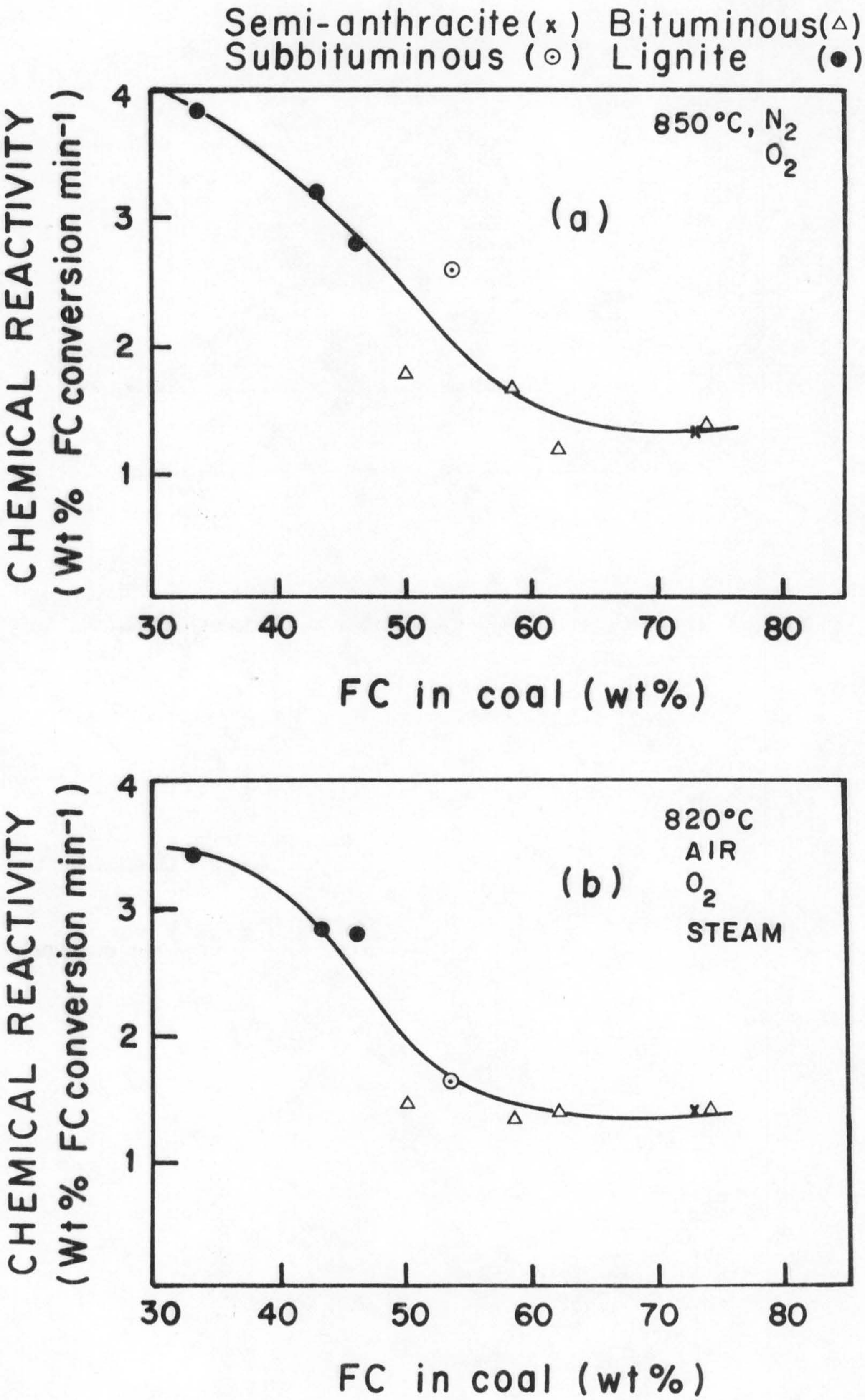

Figure 1 The relation between chemical reactivity and FC in sample without steam gasification (a) and with steam gasification (6).