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COMPARATIVE STUDY OF FIVE CANADIAN COALS IN A LABORATORY GASIFIER

D.P.C. Fung
Coal Resource and Processing Laboratory

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by

D.P.C. Fung*

ABSTRACT

Results of a gasification study of five Canadian coals in a laboratory fixed-bed gasifier are reported. The coals were three bituminous coals (Byron Creek, Prince and Devco) and two lignites (Coronach and Bienfait). When gasified in an equal mixture of nitrogen and oxygen at 850°C, the lignites reacted more rapidly and produced more liquid and gaseous products than the bituminous coals, while the bituminous coals produced more methane but less carbon monoxide. Overall rates for both the gasification and combustion processes were 3.20, 2.80, 1.84, 1.70 and 1.25 % carbon conversion per minute for Coronach, Bienfait, Prince, Byron Creek and Devco coals.

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

INTRODUCTION

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. Coal gasification and liquefaction are two main objectives in this program.

The main objective of the coal gasification program is to provide sufficient background information on the gasification characteristics of various Canadian coals for the selection of advanced gasification technologies for converting coal to electricity in different regions of Canada. For example, the gasification combined cycle process has potential of achieving high electrical conversion efficiency and meeting the gas emission standards. Other areas of importance are the utilization of low or medium calorific value gas for process heat, conversion of the product gas into liquid hydrocarbons, and possible application of gasification in heavy-oil extraction and processing.

The gasification program was implemented through two efforts: external contracts and in-house research. Outside research and development contracts are intended to provide techno-economic evaluation of gasification applications for both power generation and process industries. In-house research is geared towards providing coal reactivity data of Canadian coals which are candidates for use in various gasification processes¹.

This paper describes the in-house examination of the performance of five Canadian coals in a fixed-bed gasifier under an atmosphere of nitrogen and oxygen. This work is the first part of a series of gasification studies and its purpose is to rank the chemical reactivity of these coals towards common gasifying media under isothermal conditions at 850°C. This temperature was selected because active gasification occurred at 850°C in a preliminary study. Experimental data on the chemical composition of the gas product and the rate of carbon conversion are presented.

EXPERIMENTAL METHOD

GASIFICATION PROCEDURE

Materials

The particle size of the coal sample ranged from 3 mm to 6 mm. All

samples were air-dried, then oven-dried for two hours at 105°C before use. Chemical analyses of coal samples are reported in Table 1. Bituminous coals were Byron Creek from British Columbia, Prince and Devco from Nova Scotia. Lignite coals were Bienfait and Coronach from Saskatchewan. It was estimated in 1979 that coal reserves (mineable coal) in Canada were about 16 gigatonnes² and their occurrences in different provinces are shown in Figure 1.

For the present study only carbon dioxide, hydrogen, nitrogen, methane, propane, acetylene, ethane, ethylene and carbon monoxide were analyzed. Sulphur gases were removed prior to gas analysis so that the compositions of the clean product gas of the coals studied could be compared.

Gasification Unit

Figure 2 depicts the gasification unit used. 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 1.8 L/min for all the experiments. Inert material (Berl Saddles) was used to minimize channelling and caking of the coal sample during gasification. In the sample holder the sample mixture rested on a supporting disc that 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. Four thermocouples, 15 cm apart, were positioned in the reactor tube (Figure 2) to monitor the temperature of the gasification process.

At the desired temperature (850°C) 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, reactor temperature rebounded to 800°C in about two minutes. The temperature in the sample holder as detected by thermocouple No. 1 varied from 800 to 860°C between the period from 2 to 30 minutes.

The gaseous product from the gasifier passed through a condenser and two cold traps, one at 0° and the other at -70°C. The uncondensed tar was removed by a cellulosic paper (Balston) filter. The product gas was passed through 1000 cm³ of 25 weight % sodium carbonate solution, a filter, drierite and wet-test meter before it was separated and analysed by gas

chromatography. A minimum of two experiments were performed for each coal sample; gasification time was 30 min. The residue in the reactor was cooled with a stream of N_2 (0.9 L/min) for about two hours before it was removed and weighed.

Gas Sampling and Analysis

A total of 15 samples of the product gas were collected at two-minute intervals over a period of 30 minutes and analysed in a gas chromatograph (Perkin-Elmer Sigma 1 Analyzer). Carrier gas was a mixture of helium (91.5%) and hydrogen (8.5%) with a flow rate of 45 ml/min at column temperature programming at a rate of 15°C/min from 40 to 75°C. Porapak N and molecular sieve 5A columns were used for the separation and analysis of the product gas.

RESULTS AND DISCUSSION

The rates of production of methane, carbon monoxide and hydrogen from the three bituminous coals are presented in Figures 3, 4 and 5. The gasification reaction of Byron Creek coal proceeded rapidly at 850°C and its methane production reached a maximum after four minutes. The average chemical composition of the product gas was 11.3% hydrogen, 0.4% oxygen, 33.3% nitrogen, 14.0% methane, 0.6% propane, 0.6% ethane, 3.5% ethylene, 28.8% carbon dioxide and 7.5% carbon monoxide. The trace amount of oxygen indicates that the fed oxygen has reacted with the carbon in the coal and the gasification reaction has taken place at this stage. The other two bituminous coals, Prince and Devco, had maximum methane production at 6 minutes. The product gas from Prince coal at 6 min contained 9.4% hydrogen, 31.8% nitrogen, 15.1% methane, 1.9% propane, 0.6% acetylene, 0.6% ethane, 4.3% ethylene, 24.3% carbon dioxide and 12.0% carbon monoxide. Among the three bituminous coals studied, Devco yielded the most methane having a chemical composition in the product gas of 17.4% hydrogen, 0.4% oxygen, 23.2% nitrogen, 20.7% methane, 1.5% propane, 0.2% acetylene, 0.8% ethane, 5.2% ethylene, 16.6% carbon dioxide and 14.0% carbon monoxide. Devco produced more methane, carbon monoxide and hydrogen than the other two coals.

At 850°C the two lignite coals, Coronach and Bienfait, gasified more rapidly than the bituminous coals with a maximum production after four

min of gasification. At four min the product gas of Coronach lignite contained 12.0% hydrogen, 22.3% nitrogen, 9.6% methane, 1.7% propane, 0.3% acetylene, 0.4% ethane, 1.5% ethylene, 30.0% carbon dioxide and 22.2% carbon monoxide. The product gas of Bienfait contained 12.5% hydrogen, 0.8% oxygen, 24.9% nitrogen, 7.6% methane, 0.7% propane, 0.4% acetylene, 0.5% ethane, 2.3% ethylene, 29.7% carbon dioxide and 20.6% carbon monoxide. Figures 6 and 7 summarize the production rates of methane, carbon monoxide and hydrogen from these two lignites. The gasification characteristics of these two lignites are very similar; both of them had the same production rates of methane, carbon monoxide and hydrogen, and produced a large amount of carbon dioxide and carbon monoxide at the onset of reaction. Formation of these oxides could be due to the high oxygen contents in the Coronach lignite (19.8% O_2) and Bienfait lignite (17.3% O_2). This was also observed by Morris and Kearins in their devolatilization study of subbituminous coals³. The high carbon monoxide content of the lignite product gas also indicates that use of oxygen for gasification favours carbon monoxide formation. Bituminous coals produced more methane and higher molecular weight hydrocarbons but less carbon monoxide than the lignites in the presence of oxygen at 850°C.

Table 2 compares the volumes of the exit gases (corrected to standard temperature and pressure) from the five coal samples at different times at 850°C. A large volume of gas evolved at the beginning of the gasification but gas production began to level off at about 10 min. At 30 min, both the Bienfait and Coronach lignite generated an average of 43.0 L of nitrogen free gas from two separate experiments (A and B). Following the lignites in decreasing order were Devco, Byron Creek and Prince coal, producing 31.2, 30.9, and 26.8 L of the gas. Except for Byron Creek this order seems to be in general agreement with the chemical analyses in that coals containing more volatile matter should yield larger volumes of gas on gasification.

Rates of carbon conversion can be calculated from the chemical compositions of the coals and product gases and product gas volumes. Figure 8 summarizes the rates of total carbon conversion. The two lignites had a higher carbon conversion rate than the bituminous coals. Coronach had a similar rate as Bienfait up to 10 min but their rates began to decrease differently thereafter. Over the 30 min Coronach lignite had a higher overall conversion rate than the Bienfait lignite.

In general there are three stages in gasification reaction namely, the devolatilization process, gasification process and combustion process. The devolatilization process occurs at the beginning of the reaction and terminates when all the volatile matter has evolved or when there is no further methane formation in the product gas. Hence, for the calculation of the reaction rate, the percentage of carbon in the volatile matter can be used to determine the termination time of the devolatilization process.

For example, in the case of Byron Creek coal, the percentage of carbon in the volatile matter is the difference (15.9%) between the total carbon (74.4%) and the fixed carbon (58.5%) based on moisture free analysis (Table 1). This means that the devolatilization process ends at 9.4 min with 15.9% of the total carbon conversion (Figure 8). As for the entire period of 30 min the total carbon conversion is 43.5% (Figure 8) and the rate of fixed carbon (FC) removal, or chemical reactivity, of Byron Creek at 850°C is

$$\begin{aligned} \frac{d(\text{FC})}{dt} \text{ gasification \& } &= \frac{(43.5 - 15.9)}{(30.0 - 9.4)\text{min}} \times \frac{74.4}{58.5} \% \text{ FC conversion} \\ \text{combustion} &= 1.70\% \text{ FC conversion/}_{\text{min}} \end{aligned}$$

This rate represents an overall rate of both the gasification and the combustion process.

The duration of the gasification process is the period between the termination of the devolatilization process and the commencement of the combustion process. Further, the time for the commencement of the combustion process can be defined as the time when the oxygen concentration exceeds 1% in the product gas. This time however varies among the coals under investigation. For Byron Creek coal the gasification process ends at 22.0 min with 34.5% of the total carbon conversion (Figure 8). Therefore the rate of gasification of this coal is

$$\begin{aligned} \frac{d(\text{FC})}{dt} \text{ gasification} &= \frac{(34.5 - 15.9)}{(22.0 - 9.4) \text{ min}} \times \frac{74.4}{58.5} \% \text{ FC conversion} \\ &= 1.88\% \text{ FC conversion/min} \end{aligned}$$

Similarly, the overall rates of both the gasification and combustion processes for the other four coals can be obtained as shown in Table 3. These rates are 3.20, 2.80, 1.84, 1.70 and 1.25% FC conversion/min for Coronach, Bienfait, Prince, Byron Creek and Devco respectively. Our finding is similar to that by Mahajan et al. that reactivity of char increases with decreasing rank of the coal from which the char is produced⁴.

Table 4 summarizes the material balance from the gasification reaction of the five coals studied. This table shows the weight of the tar in the cold traps, the solid particulate in the filter and the gasification residue. 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. It was found that under same gasification conditions lignites lost more weight and generated more product gas than the bituminous coals. The carbon balance averages 89, 95, 88, 92 and 91% for Byron Creek, Prince, Devco coal and Coronach and Bienfait lignite.

CONCLUSIONS

A 100 g fixed-bed gasifier has been used as a convenient tool for the evaluation of the performance of Canadian coals. It has been found that in the presence of an equal mixture of oxygen and nitrogen, lignites react more rapidly than bituminous coals at 850°C and generate more liquid and gaseous products. Oxygen seems to enhance the gasification rate and increase the production of carbon monoxide from lignites. Bituminous coals produce more methane but less carbon monoxide than the lignites. Among the three bituminous coals, Devco yields more methane, carbon monoxide and hydrogen than Byron Creek and Prince coal.

The overall rates of the gasification and combustion processes for five Canadian coals have been determined based on the rate of carbon conversion in the product gas. These rates are 3.20, 2.80, 1.84, 1.70 and 1.25% fixed carbon conversion/min for Coronach, Bienfait, Prince, Byron Creek and Devco coal respectively.

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

Table 1 - Chemical analysis of coal samples (moisture free) gasified

	Byron Creek (B.C.)	Prince (Nova Scotia)	Devco (Nova Scotia)	Coronach (Sask.)	Bienfait (Sask.)
Proximate analysis (weight %)					
Ash	15.3	15.6	2.9	13.4	12.5
Volatile matter	26.2	34.6	35.4	43.5	41.4
Fixed carbon ^a	58.5	49.8	61.7	43.1	46.1
Ultimate analysis (weight %)					
Carbon	74.4	66.0	84.7	61.1	66.1
Hydrogen	4.3	4.5	5.6	3.6	2.2
Sulphur	0.8	4.9	1.3	1.1	0.6
Nitrogen	1.2	1.4	1.3	1.0	1.3
Ash	15.3	15.6	2.8	13.4	12.5
Oxygen ^a	4.0	7.6	4.3	19.8	17.3
Calorific value of sample (as received) (MJ/kg)					
	28.6	23.8	33.9	17.3	18.3

^a Determined by difference

Table 2 - Volume of product gas, nitrogen and oxygen from five coals at 850°C

Volume (L, STP) of product gas, nitrogen and oxygen at 850°C											
Time (min)	Byron Creek		Devco		Prince		Coronach		Bienfait		
	Experiment		Experiment		Experiment		Experiment		Experiment		
	A	B	A	B	A	B	A	B	A	B	
2	4.5	4.7	4.5	4.1	4.2	4.1	5.6	6.5	6.1	6.1	
4	8.2	9.5	9.5	9.4	9.1	7.6	12.4	13.7	12.4	12.6	
6	13.2	14.4	15.2	13.8	12.9	12.6	18.6	18.6	18.8	18.8	
8	18.0	18.7	20.7	19.6	17.7	17.8	24.2	23.8	24.4	24.6	
10	22.5	22.8	25.3	24.3	21.5	21.7	29.4	28.9	29.6	30.0	
12	26.4	26.4	29.1	28.7	25.0	25.1	34.2	33.7	34.6	35.1	
14	30.2	30.0	32.4	32.5	28.3	28.3	39.0	38.4	39.4	40.1	
16	33.9	33.6	35.7	36.2	31.6	31.7	43.7	43.0	44.2	44.9	
18	37.6	37.0	38.9	39.7	34.9	34.9	48.3	47.5	47.5	49.3	
20	41.3	40.4	42.1	42.9	38.2	38.3	52.9	51.7	51.1	53.6	
22	44.9	43.8	45.2	45.8	41.5	41.6	57.3	55.5	54.8	54.5	
24	48.5	47.0	48.5	48.8	44.7	45.1	61.5	59.1	58.5	61.0	
26	52.1	50.3	51.8	51.9	47.6	48.3	65.3	62.3	62.1	64.2	
28	55.5	53.6	55.1	54.9	50.3	51.6	68.4	65.3	65.6	67.7	
30	59.0	56.8	58.4	58.0	52.7	54.8	71.5	68.5	68.9	71.1	
Average volume, L	57.9		58.2		53.8		70.0		70.0		
Average volume, L (Nitrogen free)	30.9		31.2		26.8		43.0		43.0		

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Table 3 - Gasification rates of five Canadian coals at 850°C

Sample	Devolatiliz- ation time, t_D (min)	FC ^a removal between t_D and 30 min (% FC conversion)	Chemical reactivity, $\frac{d(FC)}{dt}$ Gasification and Combustion (% FC conversion/min)	Gasification termination time ^b , t_G min	FC removal between t_D and t_G (% FC conversion)	Chemical reactivity, $\frac{d(FC)}{dt}$ Gasification (% FC conversion/min)
Byron Creek	9.4	35.2	1.70	22.0	23.7	1.88
Prince	10.3	36.2	1.84	30.0	36.2	1.84
Devco	15.1	18.7	1.25	22.0	9.6	1.39
Coronach	5.4	78.7	3.20	30.0	78.7	3.20
Bienfait	6.0	68.8	2.80	24.0	57.4	3.19

^a Fixed carbon

^b The time when $O_2 > 1\%$ in the product gas

Table 4 - Carbon balance of the five coals studied

Sample ^a	C in sample (g)	Gasification product (g)							$\frac{\text{C in product}}{\text{C in sample}} \times 100\%$
		tar	C in tar ^b	Solid	C in solid	Residue	C in residue	C in gas	
Byron Creek A	37.4	5.8	4.6	0.6	0.6	17.9	10.4	17.1	87
Byron Creek B	37.5	6.0	4.8	0.2	0.2	21.4	13.3	15.6	91
Prince A	33.1	5.8	4.6	0.3	0.3	18.6	11.3	14.1	92
Prince B	33.2	6.6	5.3	0.2	0.2	17.1	12.3	14.8	98
Devco A	42.4	5.8	4.6	0.2	0.2	18.4	16.7	16.0	88
Devco B	43.0	5.7	4.6	0.2	0.2	19.3	17.2	16.0	88
Coronach A	31.0	6.6	5.3	0.3	0.3	5.3	0	23.5	94
Coronach B	30.6	6.7	5.4	0.2	0.2	4.8	0	22.0	90
Bienfait A	33.3	8.4	6.7	0.4	0.4	6.6	0	23.7	93
Bienfait B	33.9	10.3	8.2	0.3	0.3	7.8	0	21.8	89

^a Duplicate experiments A and B

^b The carbon (c) content in tar was not determined but estimated at 80% for the calculation

Captions

- Figure 1 Occurrences of coal in Canada by rank
- Figure 2 ERL gasification unit
- Figure 3 Production of CH_4 , CO and H_2 at various times from Byron Creek at 850°C
- Figure 4 Production of CH_4 , CO and H_2 at various times from Prince coal at 850°C
- Figure 5 Production of CH_4 , CO and H_2 at various times from Devco coal at 850°C
- Figure 6 Production of CH_4 , CO and H_2 at various times from Coronach lignite at 850°C
- Figure 7 Production of CH_4 , CO and H_2 at various times from Bienfait lignite at 850°C
- Figure 8 The rate of carbon conversion of five coals at 850°C

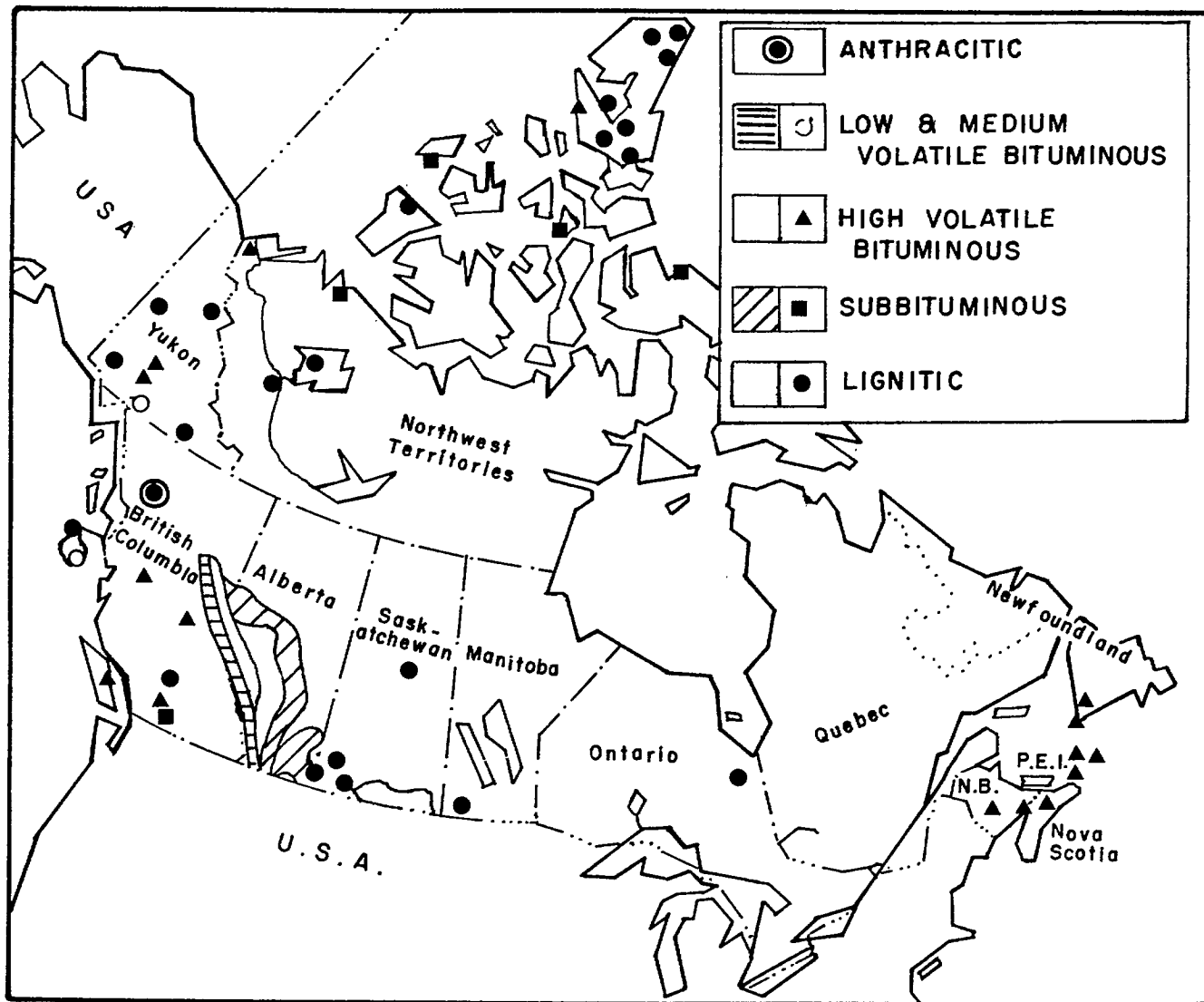


Figure.1-Occurences of coal in Canada by rank.

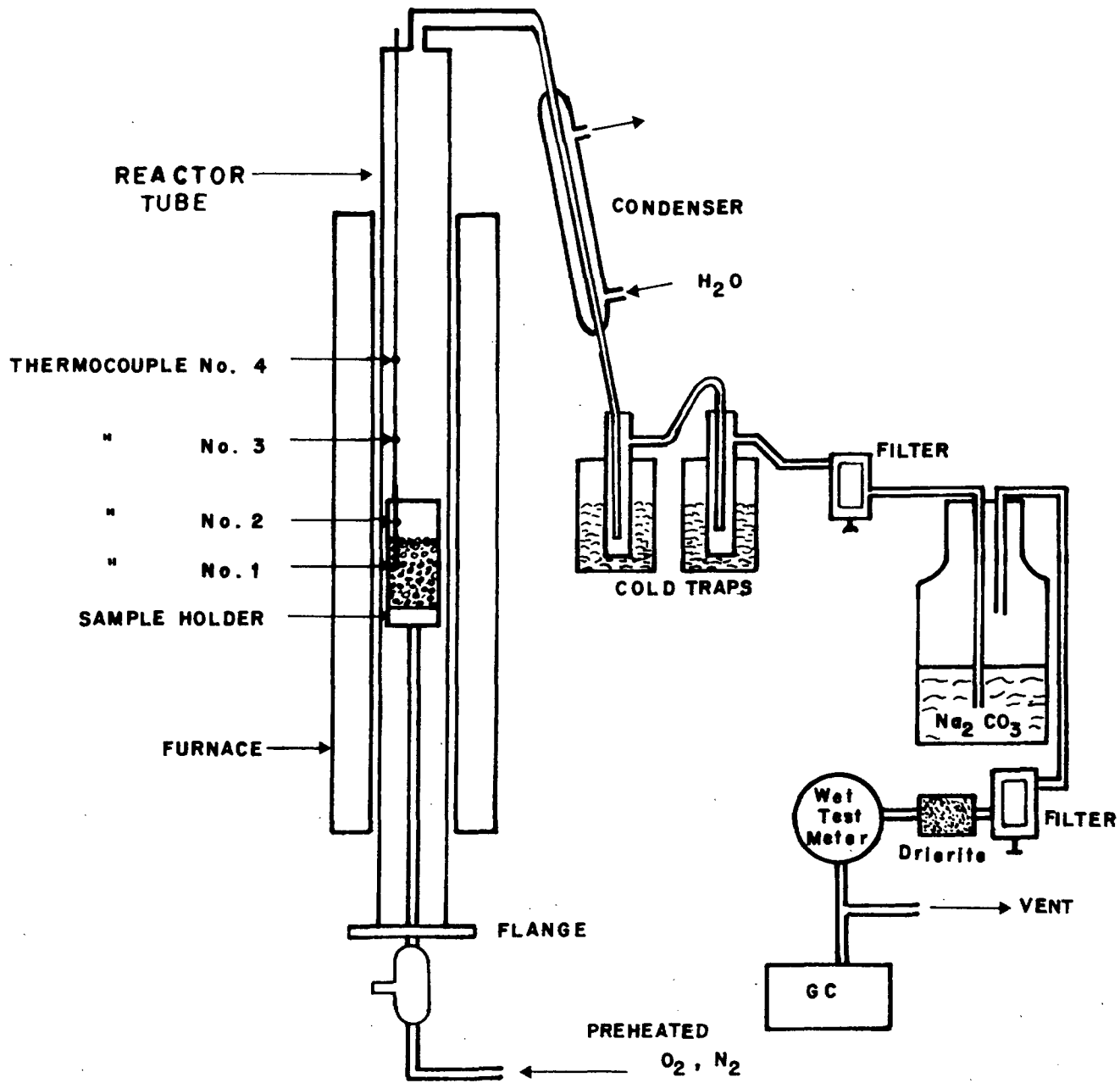
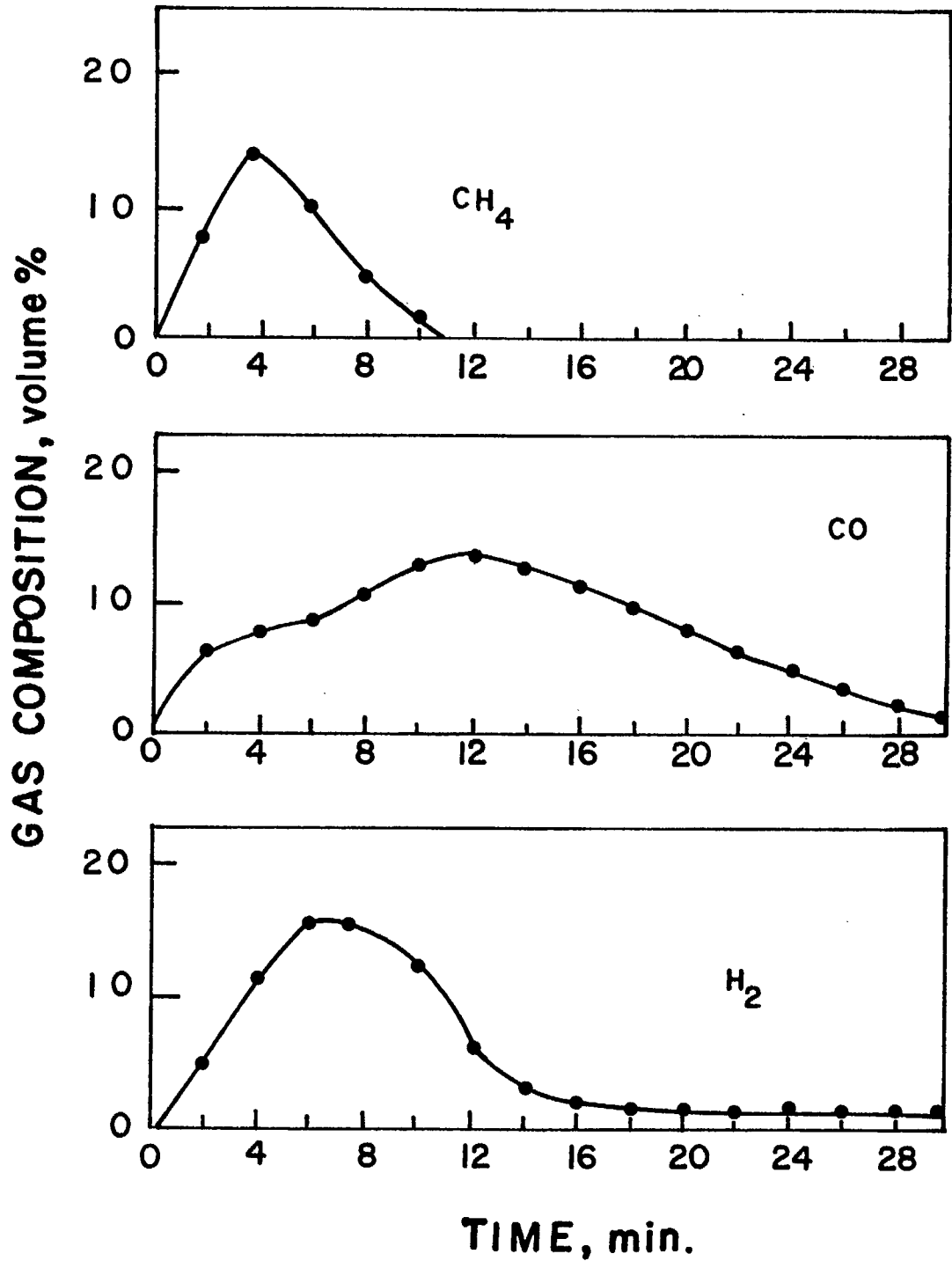
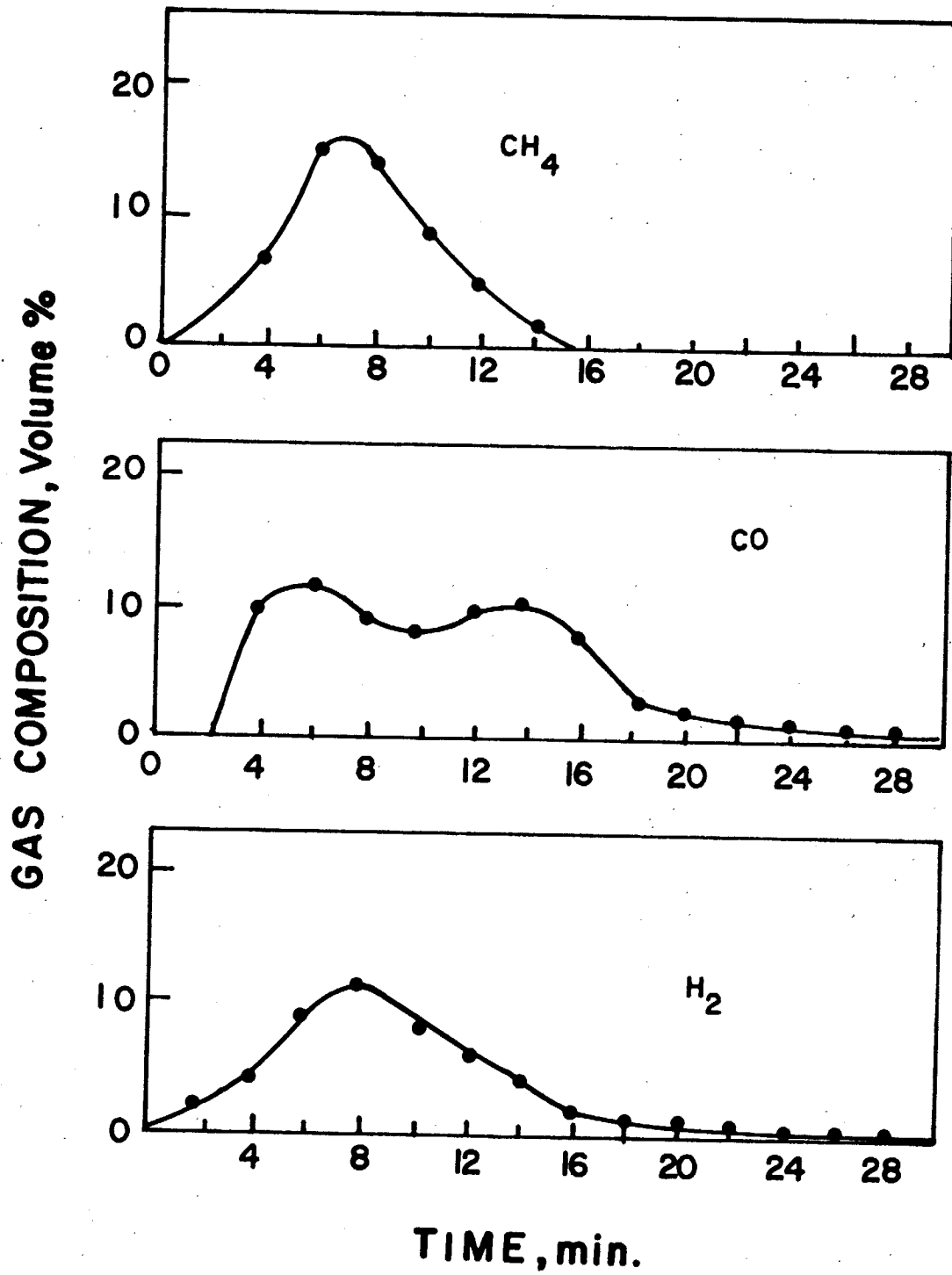


Figure 2 ERL GASIFICATION UNIT





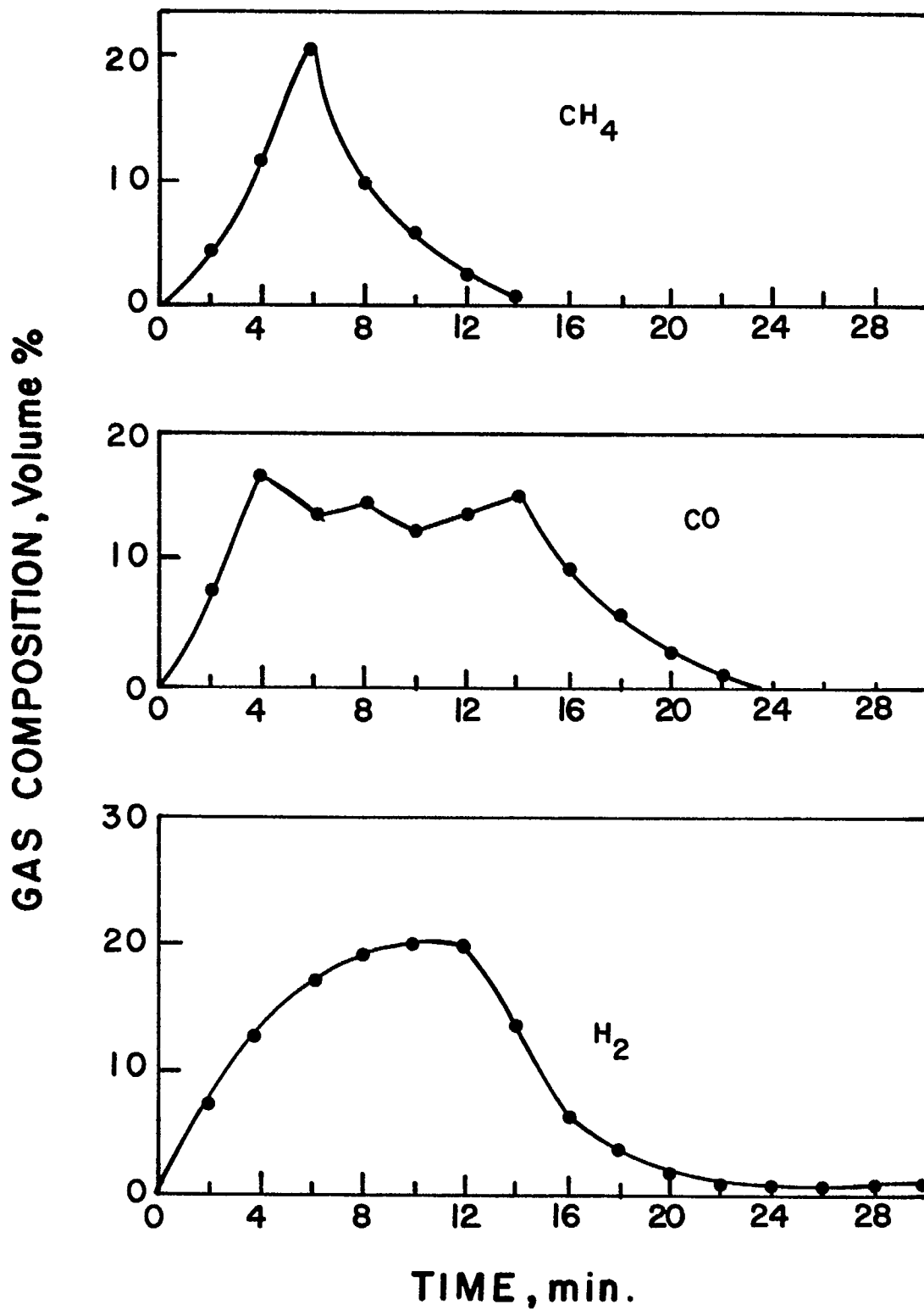
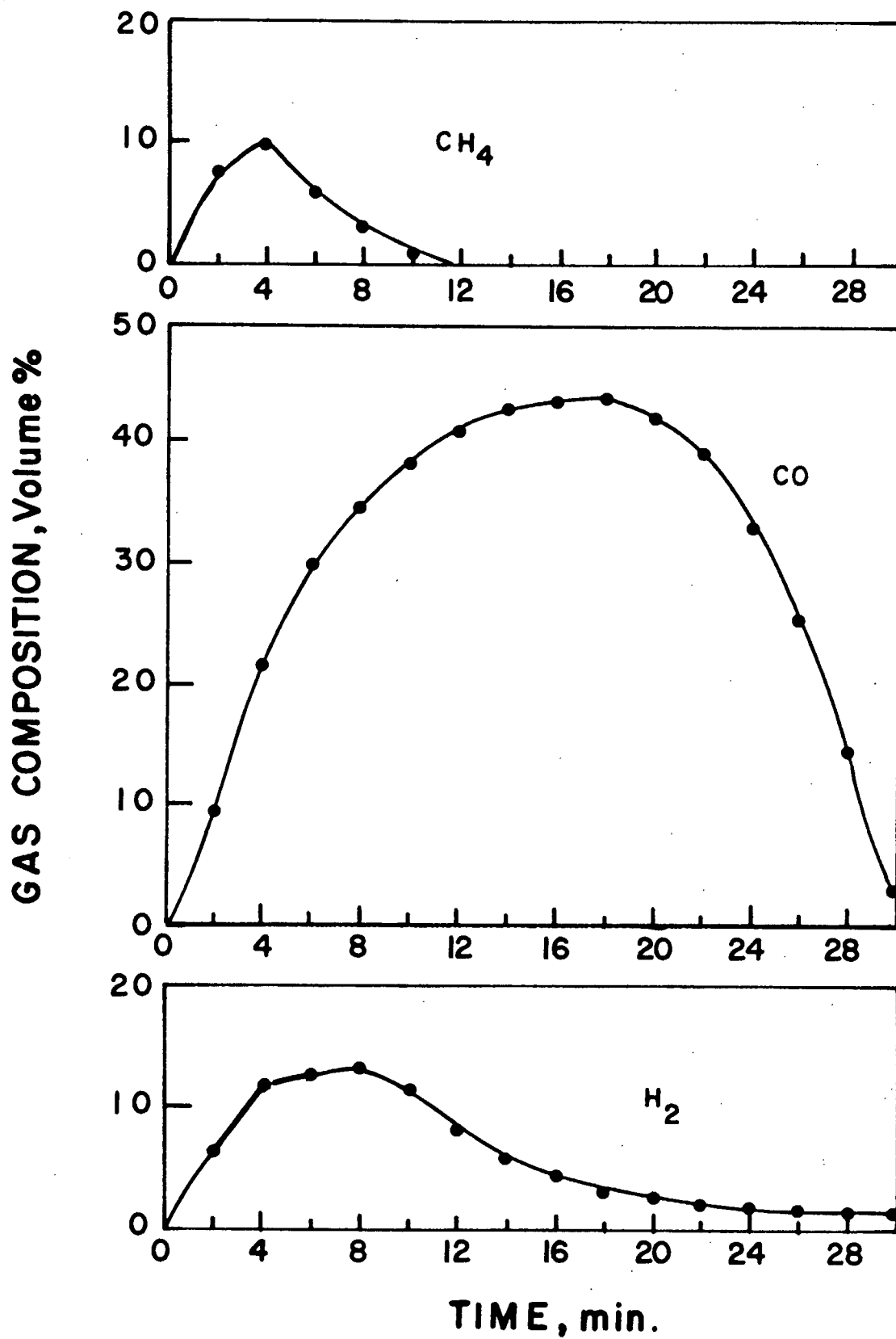


Fig. 5 D.P.C. FUNG



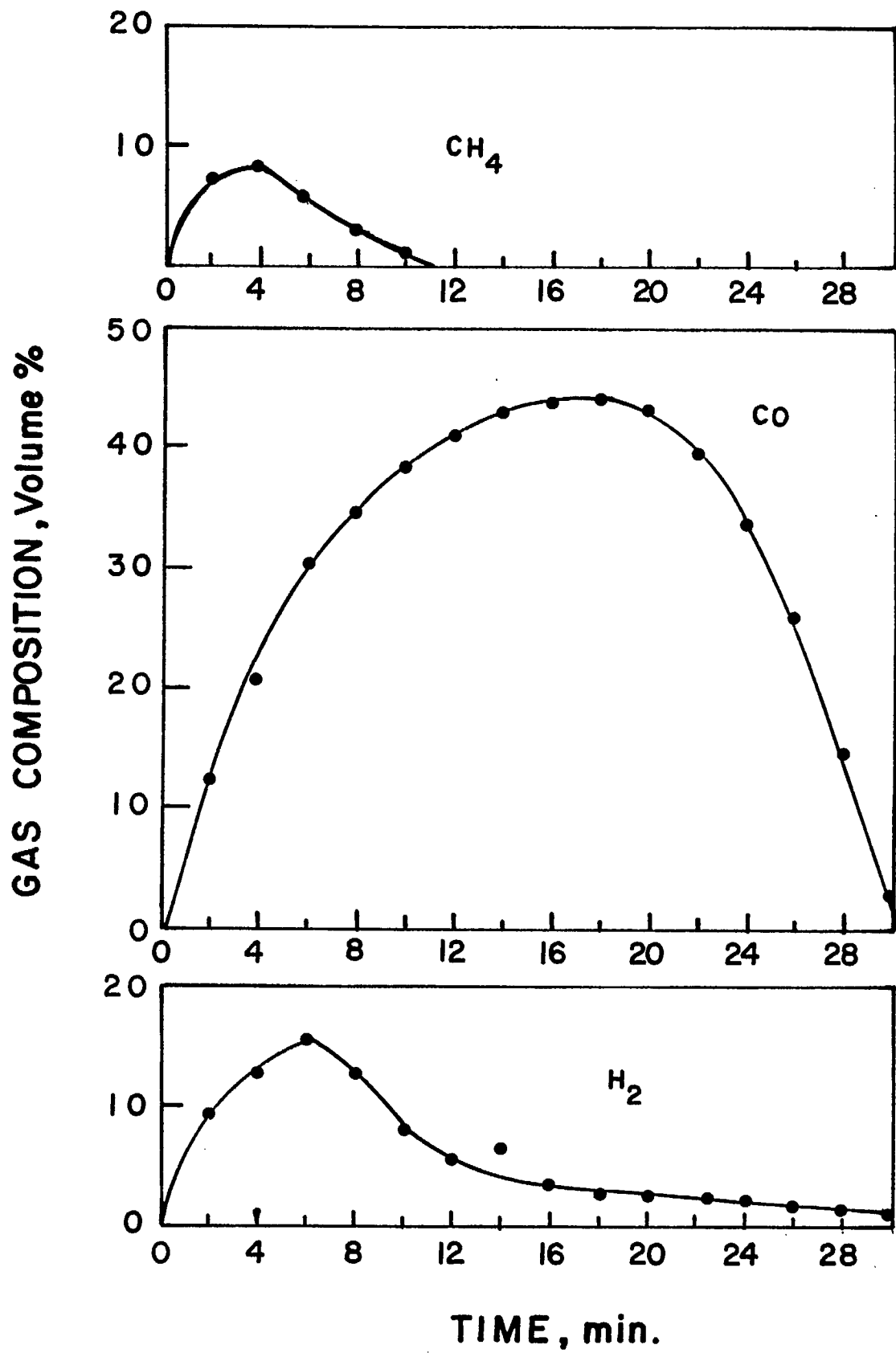


Fig 7 D.P.C. FUNG

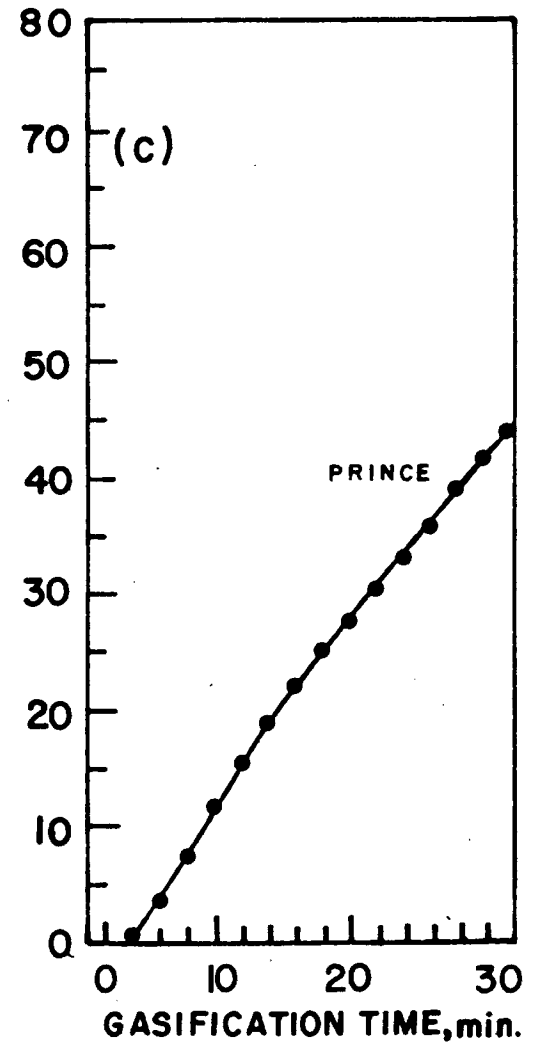
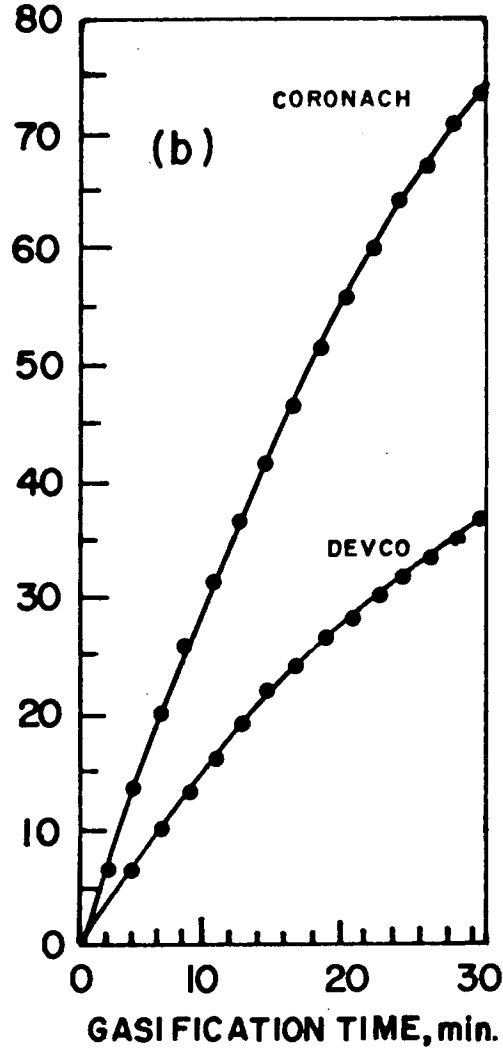
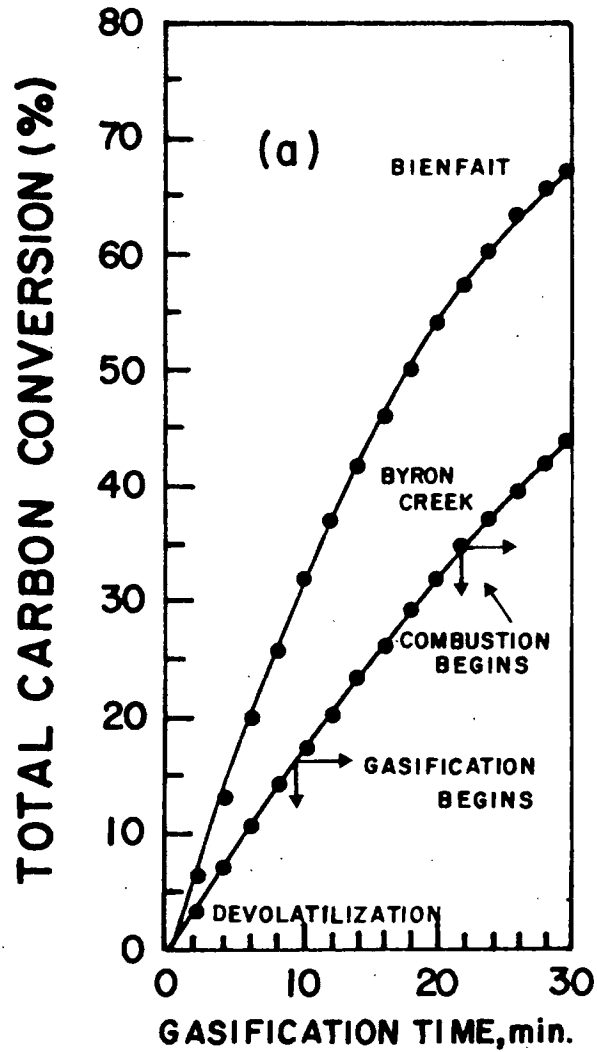


Fig. 8 D.A.C. FUNG