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PROCEEDINGS:

COAL GASIFICATION RESEARCH AND DEVELOPMENT -

A TECHNICAL SEMINAR DESCRIBING CANMET SPONSORED CONTRACTED RESEARCH AND DEVELOPMENT PROGRAMS

OCTOBER 2 and 3, 1980.

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October 2 and 3, 1980

EDITED BY: G. V. Sirianni

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FOREWORD

The Energy Research Laboratories has since 1976 funded on a contract basin, research and development in coal gasification and related fields. The objectives of this program are (1) to stimulate and develop Canadian interest and expertise in coal gasification technology, (2) to solve specific problems as related to Canadian application of coal gasification, (3) to undertake background research and development studies okn the potential uses of different Canadian coals.

This seminar was held to give the researcher associated with the various projects funded under the coal gasification program, an opportunity to present the results of their work and to meet others interested in coal gasification and related fields.

COAL GASIFICATION IN A SPOUTED BED
I. Pilot Plant and Experimentation

by

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ABSTRACT

A 0.30-m diameter spouted bed pilot process development unit capable of treating 1.4 tonne/day of coal has been designed, constructed and commissioned. Features and operation of the major components of this unit are described. Coal gasification results are reported in Part II of this report.

INTRODUCTION

This project was undertaken as a part of the Canadian Coal Conversion program-Experimental Research and Development Studies with the general object to investigate the spouted bed as a novel coal gasifier for the treatment of Western Canadian coals, particularly those of a caking variety. The proposal to do this work was based on development work with a 0.15-m sputed bed gasifier under an E.M.R. Research Agreement during 1976 and 1977.

The specific objectives of the present program were to:

(i) build a spouted bed gasifier unit 0.30-m diameter
(ii) determine optimum operating conditions for gasification
(iii) demonstrate technical feasibility by extended experimental runs
(iv) gather data on effects of operating variables necessary for scale up

The first phase of the work had as its objectives (1) the design and construction of the pilot plant, and (2) commissioning, de-bugging and preliminary gasification tests. After these tests, modifications to the pilot plant were made both to improve its general operation, and to achieve other specific objectives such as comparison of top vs bottom feed, comparison of steam injection in spout and in annulus, and recycling of cyclone product to the gasifier.

COAL GASIFIER SYSTEM

The gasifier system and its main auxiliaries are shown in Figure 1. These units are located in a laboratory of the Chemical Engineering Department at U.B.C. Not shown in this figure is the off-gas incinerator which is located two floors above on the roof of the building. The major items of equipment and their specifications are listed in Tables 1-6. The basic design features of the main units are discussed below.

Reactor and Feed System

Table 1 and Figure 2 give details of the spouted bed reactor. The gasifier is refractory lined to an inside diameter of 0.30-m to tolerate the high gasification temperatures. The cone angle of the spouted bed was selected after visual tests in an available lucite column of the same diameter showed an angle of 60° was necessary to minimize dead space and allow solids to move freely into the spout region. A variable diameter orifice based on a design by J. Lim, K.B. Mather and J. Baranowski is employed. This device permits a continuous adjustment of spout orifice diameters from 0.50 mm. Above the cylindrical section of the column a disengaging section of larger diameter was provided to reduce particulates going to the cyclones. A sight glass on top of the disengaging section allows visual examination of the spouting bed. A spring-loaded sample valve permits sampling of the bed from the side of the cone section during operation.

An external start-up burner was designed and contructed. It consists (Figure 3) of a refractory-lined combustion chamber and is fired by propane which is stored in cylinders outside the building. Natural gas which would have been more convenient is piped to the building at too low a pressure to fire the system when the spouted bed is in place. The particular burner selected has the capability of operating with a large excess air (1000%), and a high turn-down ratio. Since the main spouting air goes through the burner, the air was originally preheated by partial combustion. In a full scale plant, gains in thermal efficiency could be made by using the waste heat in the off-gas to preheat the spouting air. This operation was simulated by employing a steam heated shell and tube heat exchanger coupled with a 6kW electrical heater to preheat the air. The design of the heat exchanger is shown in Figure 4. Steam at mains pressure of about 75 psig was used in the shell side. Air at the outlet of the heat exchanger reached a temperature of about 120°C and then was boosted by the electrical heater to temperatures of 150-400°C. The desired temperature was achieved by manipulating a temperature controller. Layout of this unit relative to the existing plant is shown in Figure 1.

Coal is fed from an available Vibra-screw feeder described in Table 2. The hopper on this unit holds sufficient coal for about 1-h operation. A coal-handling system based on a previous description was devised in order to minimize dust and spillage into the laboratory, and to allow both top and bottom feeding. A cone (Figure 5) is fitted to the top of a standard 45-gallon drum. A hydraulic loader lifts the drum of coal, which is then rotated 180° to bring the cone into position over the hopper of the Vibra-screw feeder. The loader remains in position during the gasification run until the drum is empty. This takes typically 3-4 h. The empty drum is then replaced by a full one while the small Vibra-screw hopper continues to hold coal. A sight glass on the side of the hopper permits visual inspection of the coal level. From the Vibra-screw feeder coal is conveyed pneumatically to either the bottom of the spouted bed or to the top. a glass pipe section in the conveying line provides a visual check that coal is being transported to the reactor. Velocities of 3 m/s in the 25 mm dia. feed line result in smooth conveying of the coal. Where top feeding is used the coal is separated from the conveying air in a cyclone, moves through a rotary valve and drops into the reactor. The air does not enter the reactor.

The reactor and feed system are equipped with rotameters to measure propane, air and steam flow. Air is available at 35 psig, and steam at 75 psig from the building services. Coal feed ratio is measured by level in the Vibra-screw hopper on an instantaneous basis, and by weight and volume measurements on the 45-gallon drum containers. Recently a load cell system has also been installed. Temperature is measured in the hot air entering the bed, in the spouting bed from the cone and in 8 axial positions up the column, and in the free board. Pressure is measured below the orifice, in the cone and at the top of the reactor. The safety control system, and the alarms that operate from these readings are described in section 3.

Two alternate methods of introducing steam are employed. The steam is either introduced into the central orifice at the apex of the cone, or into the annulus through a manifold leading to the cone section (Figure 6). Eight 8-mm I.D. tubes equally spaced around the cone feed steam directly into the annulus some 150 mm above the orifice.

Gas cleaning and cooling

Specifications of the gas conditioning system are given in Table 3. Hot, dusty gas leaves the reactor and passes through two cyclones in series (Figure 7). These were designed to remove particles larger than 30 MM at 95% efficiency, and those below 30 MM at 85%. Hoppers equipped with valves are connected to the bottom of the cyclones. Gases leaving the cyclones are cooled in a counter currently operated plate-coil heat exchanger designed to condense the tars (Figure 8). Commercially-available plates rather than tubes were selected for ease of cleaning. The rectangular shell of this exchanger was designed with a glass tar receiver and overflow mounted on the bottom. The glass receiver allows a volumetric estimate of tars or water condensed as a function of time. Downstream from the tar condenser the cooled gas flow rate is measured by means of a calibrated orifice plate which is connected to a pressure transducer. The output from the transducer is recorded on a strip chart recorder mounted on the control panel.

The gas issuing from the cyclone 2 was found to be excessively hot. A water jacket made of a 100 mm diameter x 1.2 m long copper pipe was installed over the pipe section linking cyclone 2 and the tar condenser. The cooling water flowed counter current to the hot gas. This allowed cooling the gas from about 400°C to about 250°C, before it entered the tar condenser.

The gas leaving the tar condenser passes through a scrubber (Table 3) where it contacts water in a counter current manner. The scrubber "packing" is a set of 21 vertical copper pipes. Vertical pipes were selected over rings or saddles for ease of cleaning. The cooled gas then passes to the off-gas incinerator located on the roof of the building.

A dust filter was later added to improve gas/solids separation, and prevent particulates reaching the turbo-blower. The design of this unit is given in Figure 9. The gas enters tangentially at the mid-section of the drum and filters through a 305 mm diameter x 762 mm long chamber wrapped with 12.5 mm thick fibre glass-sheet.

Improvements have also been made to the scrubber system. The original scrubber column was made in part with a 229 mm I.D. x 1016 mm long glass section. The glass column was susceptible to damage and was substituted with a stainless steel column. The original copper pipe bundle was also replaced with a packing of metal shavings, which are available at no cost and give adequate gas-water contacting. The metal shavings are discarded as they become dirty.

Temperatures and pressures are measured throughout the gas-conditioning equipment. Water flow to the tar condenser and scrubber is metered by rotameters.

Off-gas incineration

The off-gas is burned in an incinerator described in Table 4 and Figure 10. The incinerator is equipped with a continuous methane pilot flame, a flame detector, and a special burner selected to burn the low calorific value gas being produced. The gas from the scrubber is pulled through the pipe by the off-gas exhaust fan and contacts air from the combustion air blower in the burner. Provision is made for gas bypassing the incinerator through a spring-loaded valve attached to a fan relief line in the event of excessive pressure due to blockage in the burner. This also protects the large off-gas exhaust fan from excessive pressure.

Control panel

All units are controlled and monitored from the central panel, except the water flow to the scrubber. Metering equipment is described in Table 5, and other instrumentation in Table 6. Safety devices and controls are discussed in section 3.

Gas sampling

Gas is sampled through a line by a continuous bleed upstream of the scrubber. A condenser system described in Section 5 used to quantify tar content, and the cooled gas is analyzed intermittently by batch gas chromatography. A continuous carbon monoxide indicator located on the control panel senses CO in the same gas stream.

Fail-safe circuit

A fail-safe circuit was designed to sense the following danger conditions:

(1) loss of air supply,
(2) loss of propane supply,
(3) over-temperature in the bed,
(4) over-pressure in the bed and,
(5) leakage of gas into the laboratory.

Conditions (1), (2) and (4) are sensed by three mercury switches, condition (3) by an alarm which accepts Ch-Al signal and condition (5) by a combustible gas alarm and a carbon monoxide alarm.

A schematic wiring diagram is shown in Figure 11.

In the event any of the above conditions occur, the plant would automatically shut down and in all cases except condition (4) nitrogen would be purged into the plant, extinguishing the heat and cooling down the whole unit. The nitrogen-switching circuit is powered by a 12v battery and charger and would operate even in the event of power failure. To further increase the safety of the plant, the circuit is interlocked with the off-gas exhaust fan. The gasification plant can not be started without turning on the exhaust fan. Failure in the exhaust fan would also trip the circuit and shut the plant down subsequently.

Carbon monoxide and combustible gas detectors are located near the reactor in the laboratory to sense escaping gases and provide an audible alarm.

Fire protection service is provided through a direct alarm located outside the laboratory.

Start-Up And Shut-Down Procedures

Start-up procedures

The procedures followed in starting-up the plant are:

(1) Purge system with air or nitrogen
(2) Switch on combustion air blower
(3) Energize incinerator pilot burner
(4) Start plant exhaust fan
(5) Energize fail-safe circuit by turning on:

   (a) the high temperature alarm
   (b) the combustive gas alarm
   (c) the CO alarm
   (d) the nitrogen and
   (e) the audible alarm.

(6) Turn on main spouting air

(7) Charge spouted bed with a predetermined amount of inert while maintaining a sufficient air flow to prevent the bed being packed tight

(8) Turn on water to off-gas washer and tar condenser

(9) Energize preheater burner

(10) Allow preheater and spouted bed temperature to rise gradually to about 500°C, then start top feeding coal

(11) Adjust main spouting air flow to bring the bed into a spouting stage

(12) Watch bed temperature closely and reduce propane gradually

(13) When bed temperature rises to about 650°C divert coal feed to bed bottom

(14) Turn on steam and set at predetermined level

(15) Sample product gas and analyze with G.C. at suitable intervals

(16) Sample bed material at suitable intervals

<u>Normal shut-down procedures</u>

(1) Stop coal feed and solids transport air

(2) Turn off preheater burner

(3) Allow system to cool

(4) Turn off steam

(5) Reduce main spouting air

(6) When bed becomes cool enough, empty it through sample valve on bottom, or shut air and bed supporting shutter simultaneously

(7) De-energize fail-safe circuit

(8) Turn off exhaust fan

(9) Shut down incinerator

Emergency shut-down

Emergency shut-down is achieved simply by tripping the safety control circuit if hazard conditions were (1), (2), (3) or (5). To trip the circuit, switch off high temperature alarm.

When an over-pressure is developing in the bed:

(1) Stop coal feed
(2) Turn off preheater burner
(3) Turn off all gas flow to the bed
(4) Close bed supporting shutter

Analytical Method

Tar determination

On selected runs, the approximate tar content of the gas produced was determined by the following method. After the reactor had reached steady operation, a gas sample stream was pumped through a set of impingers for about 1/2-1 hour. Here, the tar condensed in the cold water and on the walls of the impingers. The tar was then removed by dissolving it with acetone. The acetone was then evaporated under vacuum and the residual tar weighed. The tar content of the gas was calculated by dividing the total tar thus obtained over the total gas volume through the impingers during the tar sampling period.

Gas analysis

The carbon monoxide content of the gas was continuously monitored by an infrared analyser while the remaining gases were analysed from samples taken at convenient time intervals (15-30 min) in a gas chromatograph. Both instruments were connected to a Watanabe chart recorder.

(i) Continuous CO monitor

A Beckman continuous infrared analyser, model No. 864-13-4, which operated in the range of 0.25% CO was employed. The instrument gave a continuous signal on the recorder chart. Carbon monoxide content of the gas was obtained from a factory provided calibration chart. The instrument was calibrated for every run by adjusting the readings of two points of the calibration curve. The zero reading was adjusted by passing through the instrument a standard gas stream containing 10% CO_2 and 90% nitrogen. The second calibration point was obtained by adjusting the instrument's reading on a standard sample containing 10% CO_2, 21.3% CO and nitrogen as balance, to the calibration curve reading (91% deflection).

The continuous CO analyzer provide an excellent means of monitoring the gasifier performance as well as providing an indication of whether the reactor was operating under steady conditions.

(ii) Gas chromatograph

At convenient time intervals, 5 cm^3 gas samples were injected into a Hewlett-Packard gas chromatograph model 5710 A. The gas chromatograph was equipped with a molecular sieve column, a poro-pack column and a thermal conductivity detector which resolved hydrogen, carbon dioxide, nitrogen, oxygen, methane and carbon monoxide. Typical gas chromatograph tracings are shown as Figure 12. The percentage content of each gas in the sample was determined from the peak height and the corresponding calibration curves. In order to avoid possible variations of the calibration curves, these were obtained for each run by analysing dilutions in air of a standard gas sample.

(iii) Gas calorific value

The calorific value of the gas was calculated from the gross (high) heat of combustion of the three combustible components of the gas produced (hydrogen, carbon monoxide and methane), and the dry gas composition. In order to enable comparison with values reported in the literature, the gross calorific value of a unit volume of gas (h_{cg}) was calculated at the North-American standard conditions (288.6 K = 60 F and 101.6 kPa = 30 in Hg dry). Then:

$$h_{cg} = h_{H_2} x\ (\%\ H_2\ v/v) + h_{CO}\ (\%\ C\ v/v) + h_{CH_4} (\%\ CH_4\ v/v\ \ /100$$

where:

$h_{H_2} = 12.109\ (MJ/m^3)$
$H_{CO} = 11.997\ (MJ/m^3)$
$h_{CH_4} = 37.743\ (MJ/m^3)$

Bed and cyclone material

The bed is analysed for the relative amounts of inert gravel, ash and carbonaceous material. Ultimate and proximate analyses are carried out on some selected cyclone catches to determine the extent of reaction of the carryover. The bed material and cyclone catch are sieved for size distribution.

CONCLUSION

A 0.30-m diameter spouted bed coal gasification plant has been designed and constructed based on the development work with a 0.15-m spouted bed. The unit is capable of treating 1.4 tonne/day of coal and has features that allow demonstration of technical feasibility and collection of data for process development. Operating results are presented in Part II.

ACKNOWLEDGEMENT

E. Szabo contributed to the design of the process monitoring and fail-safe circuit.

REFERENCE

1. Moss, G. Proc 2nd Eng Foundation Conference on Fluidization; J.F. Davidson and D.L. Kearns (Ed.) Cambridge Press; p. 215; 1978.

Table 1 - Design features of spouted bed reactor

Column	Carbon steel	10 ga.	O.D. = 0.50 m	Height = 1.37 m
Disengager	Carbon steel	10 ga.	O.D. = 0.76 m	Height = 0.61 m
Cone	316 Stainless steel		3.175 mm wall	Height = 0.46 m
	Angle 60°		Orifice Diameter	0-50 mm
Lining	Plisulate Insulating Block No. 201			38 mm
	Plisulate Ceramic Paper			6.4 mm
	Plisulate Plicast 40			64 mm
Instrumentation	Bed Thermocouples			12
	Cone Pressure Indicators			1
	Cone Thermocouples			1
	Orifice Pressure Indicator			1
Feed	Top or Bottom Feed			
Rupture Disk Relief	50 mm dia.			
Bed Viewing Sight Glass	75 mm dia.			
Bed Sample Valve and Discharge	25.4 mm dia.			
Propane Fired Start-up Burner	17.6 - 176 KW			
Inert Gas Flushing Points	2			
Bed Temperature Alarm				
Bed Pressure Alarm				

Table 2 - Coal feeder and start-up burner

Coal Feeder - Vibratory Screw Feeder	Vibra Screw Inc.
Coal Feed Rate	0-225 kg/h
Coal Bin Volume	0.085 m^3
Coal Drum Hydraulic Stacker	Capacity 680 kg
Start-up Burner - Eclipse Fuel Engineering	Propane Burner
Capacity	17.6 - 176 KW
Air Flow	0.28 m^3/min. - 2.83 m^3/min.
Burner Housing	0.48 m O.D. x 0.91 m shell 0.05 Plicast Tufflite Lining
Flame Safeguard	Honeywell Ltd.

Table 3 - Gas conditioning equipment

Gas/Solids Cyclones	
Material	304 Stainless Steel
1st Stage	206 mm dia x 273 mm cylinder height x 337 mm cone height
2nd Stage	194 mm dia x 267 mm cylinder height x 483 mm cone height
Solids Receivers (2)	
Material	Carbon Steel
Volume	0.08m^3
Tar Condenser	
Four Carbon Steel Serpentine Plate coils:	Tranter Canada Ltd.
Platecoil dimension	0.46 m wide x 1.19 m high
Total Area	5.17 m^2
Condenser Shell	0.483 m wide x 1.73 m high x 0.25 m deep
Material	4.8 mm Mild Steel
Cooling Water	0 - 0.55 m^3/h
Gas Scrubber	
Glass Column	.23 m ID x 1.07 m high
Packing	21 19 mm OD x 0.91 m high vertical copper tubes
Total Area	1.81 m^2
Water Flow	0.079 m^3/h

Table 4 - Off-gas incinerator

Manufacturer	Lockhead-Haggerty Engineering & Manufacturing
Combustion Chamber	0.69 m ID x 1.6 m long
Lining	0.1114 m Plicast Tufflite
Stack	0.66 m wide x 1.22 m high x 0.23 m deep
Combustion Air Blower	Lau Industries Model HPR10 Capacity 4.35 m^3/min.
Off-Gas Exhaust Fan	Eclipse Fuel Engineering SM-4628-5-3 Capacity 5.66 m^3/min. Discharge Pressure 13.79 KPa Drive 3.73 KW motor

Table 5 - Flowmetering equipment

Rotameter	Range
Main Spouting Air	0 - 3.1 m^3/min
Solids Transport Air	0 - 0.5 m^3/min
Steam	0 - 0.23 kg steam/min
Propane	0 - 0.15 m^3/min
Solids Recycling Air	0 - 0.22 m^3/min
Tar Condenser Water	0 0 0.55 m^3/h
Scrubbing Water	0 - 0.79 m^3/h

Table 6 - Instrumentation and controls

Instrument	Range
Pressure Gauges	0 - 103.4 Kpa
Marsh Master Gauges (5)	Vac. 0 - 101.3 kPa
Marsh Compound Gauges (3)	Pressure 0 - 103.4 kPa
Pressure Switches	
Air and Propane: Honeywell (2)	0 - 35 kPa
Bed : Mercoid	13.8 - 413.9 kPa
Nitrogen : Honeywell	0 - 1034 kPa
Burner Controls : Honeywell (2)	
Bed High Temperature Alarm: Telmar	130 - 1125°C
Detectors and Recorders	
CO Analyser : Beckman	0 - 25%; 0 - 50%
Off-Gas Orifice Pressure Transducer : Stratham	17.2 kPa
DC Power Supply for Transducer: Endevco Dynamic	1 - 15 VDC
Off-Gas Millivolt Chart Recorder : Linear Inst. Co.	
Scanner - 10 Channel: Hampshire Controls	
Temperature Millivolt Panel Meter : New Port Lab.	0 - 1400°C
Temperature Millivolt Recorder: Texas Inst. 24 Points	0 - 1200°C
Thermocouples : Omega	
Combustile Gas Alarm & Sensor (Lab.): M.S.A.	
CO Monitor (Lab.): M.S.A.	

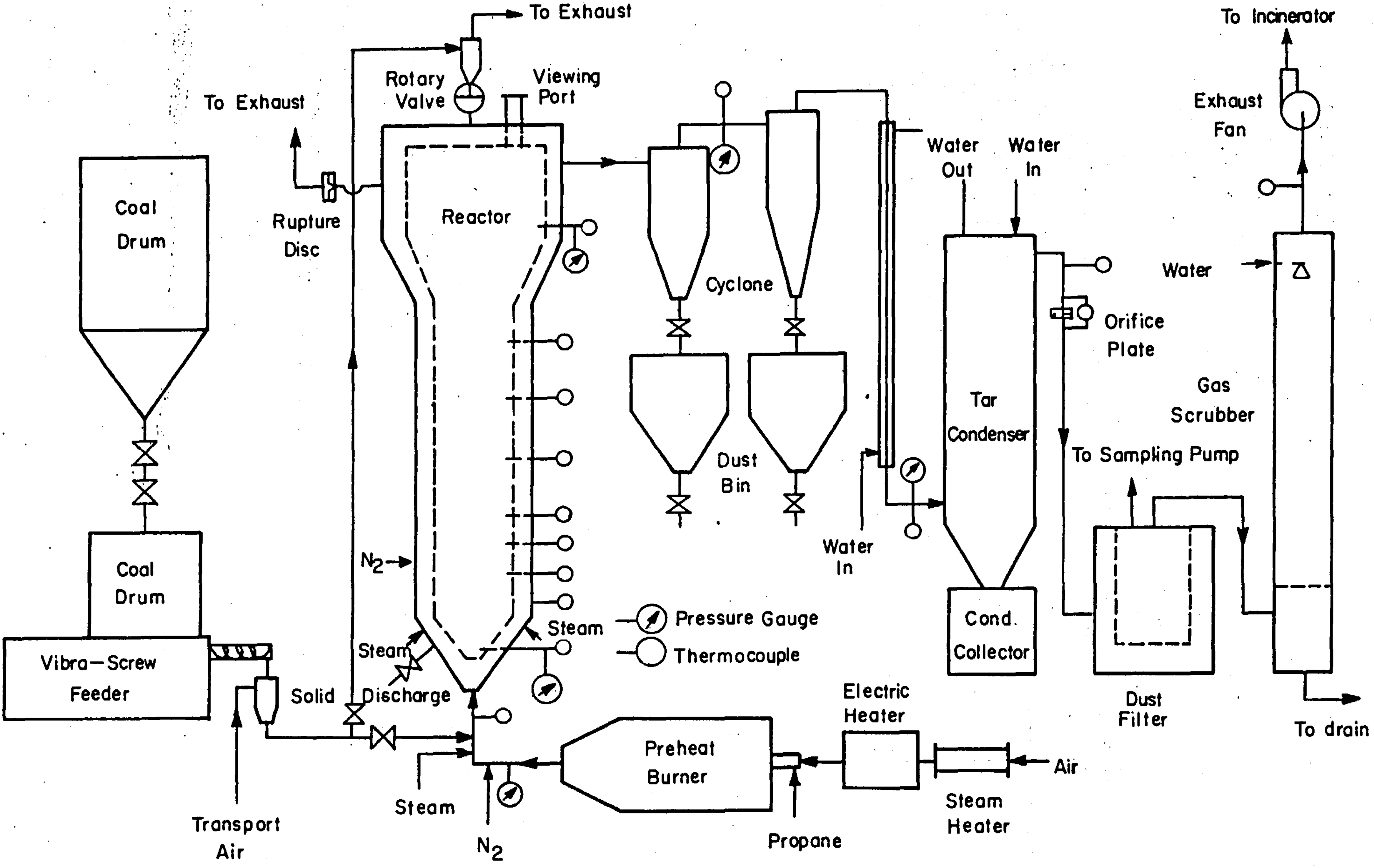


FIG. 1 SCHEMATIC DIAGRAM OF SPOUTED BED COAL GASIFIER

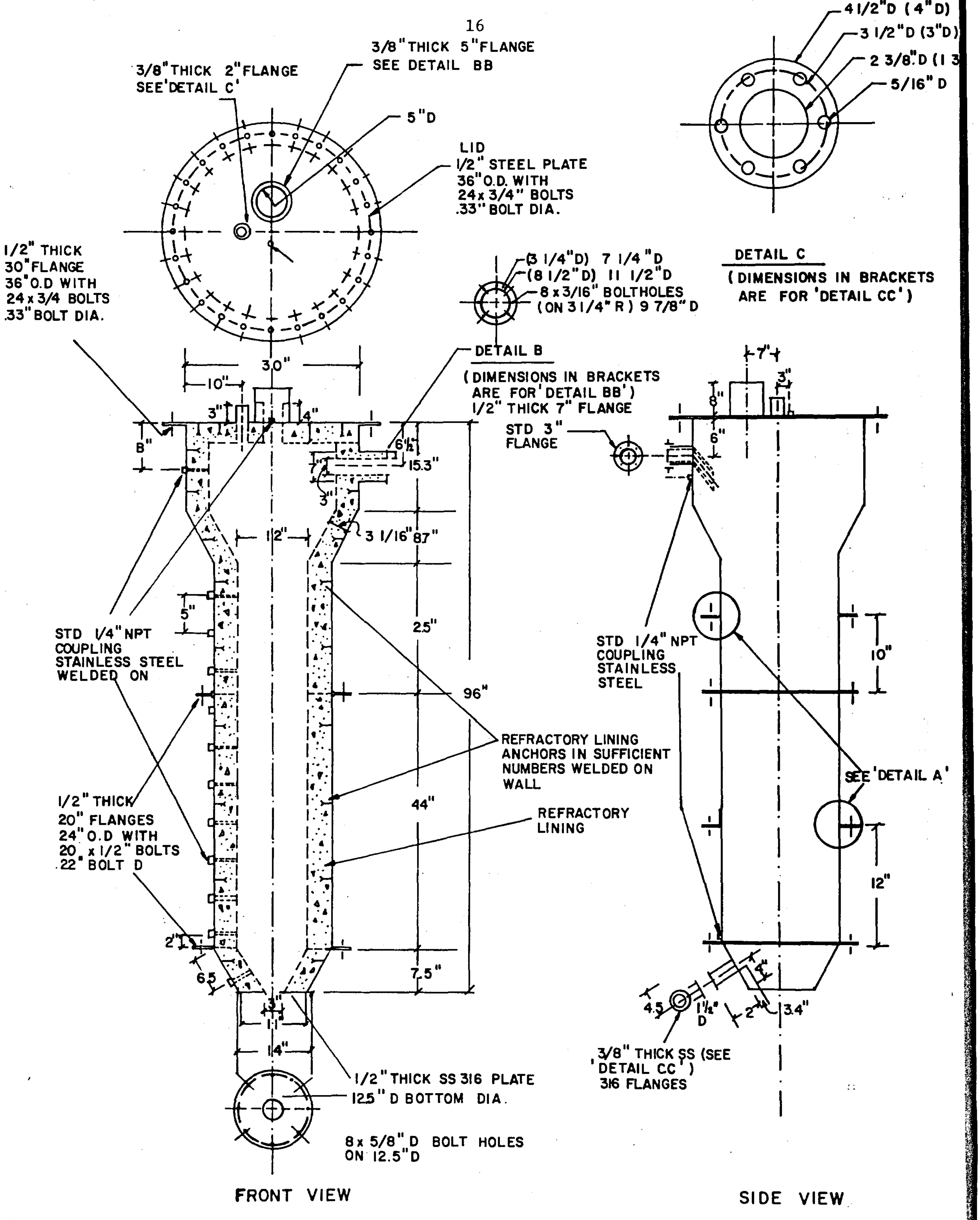


FIG. 2. SPOUTED BED COAL GASIFIER

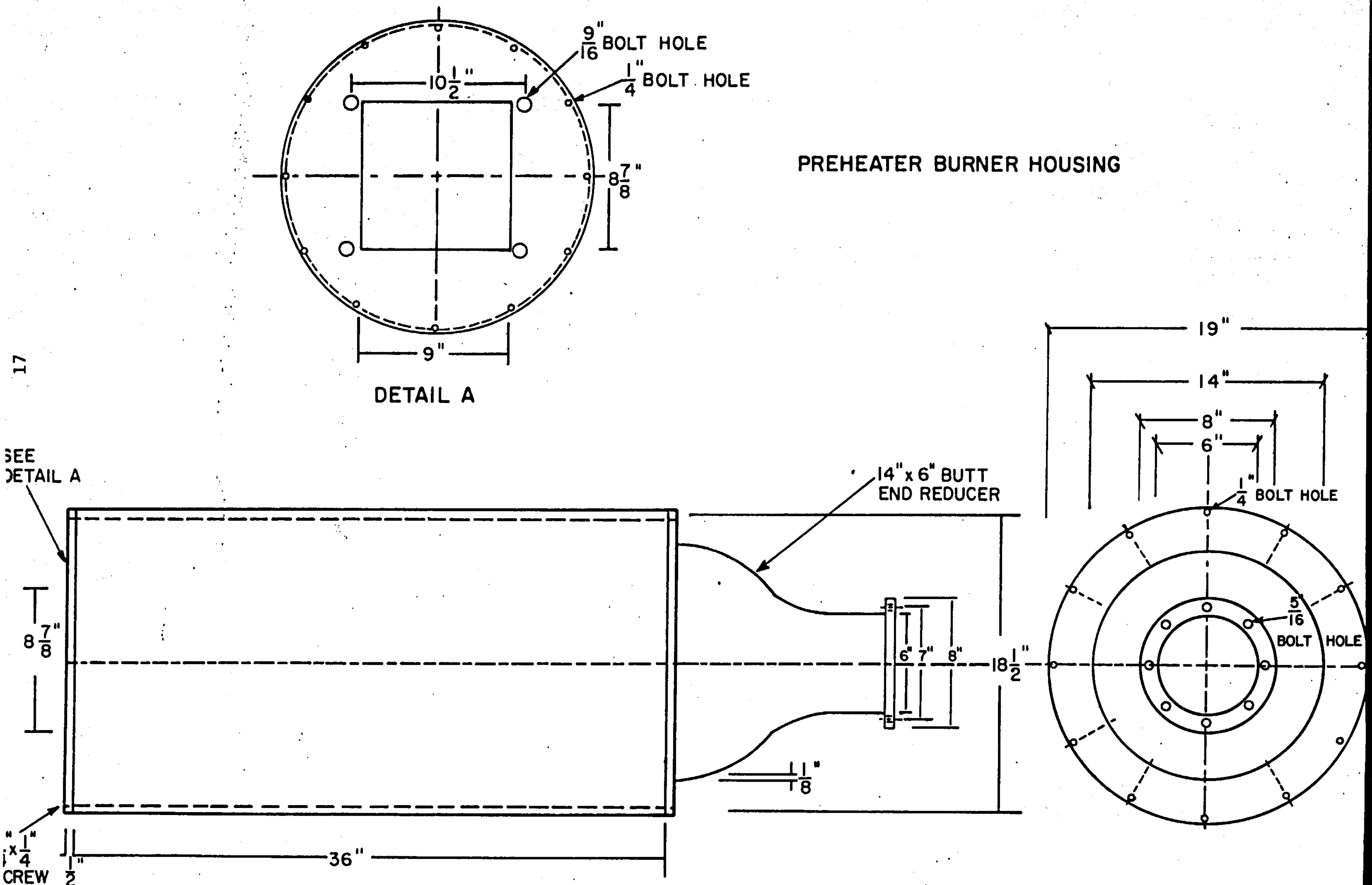


FIG. 3 START-UP BURNER

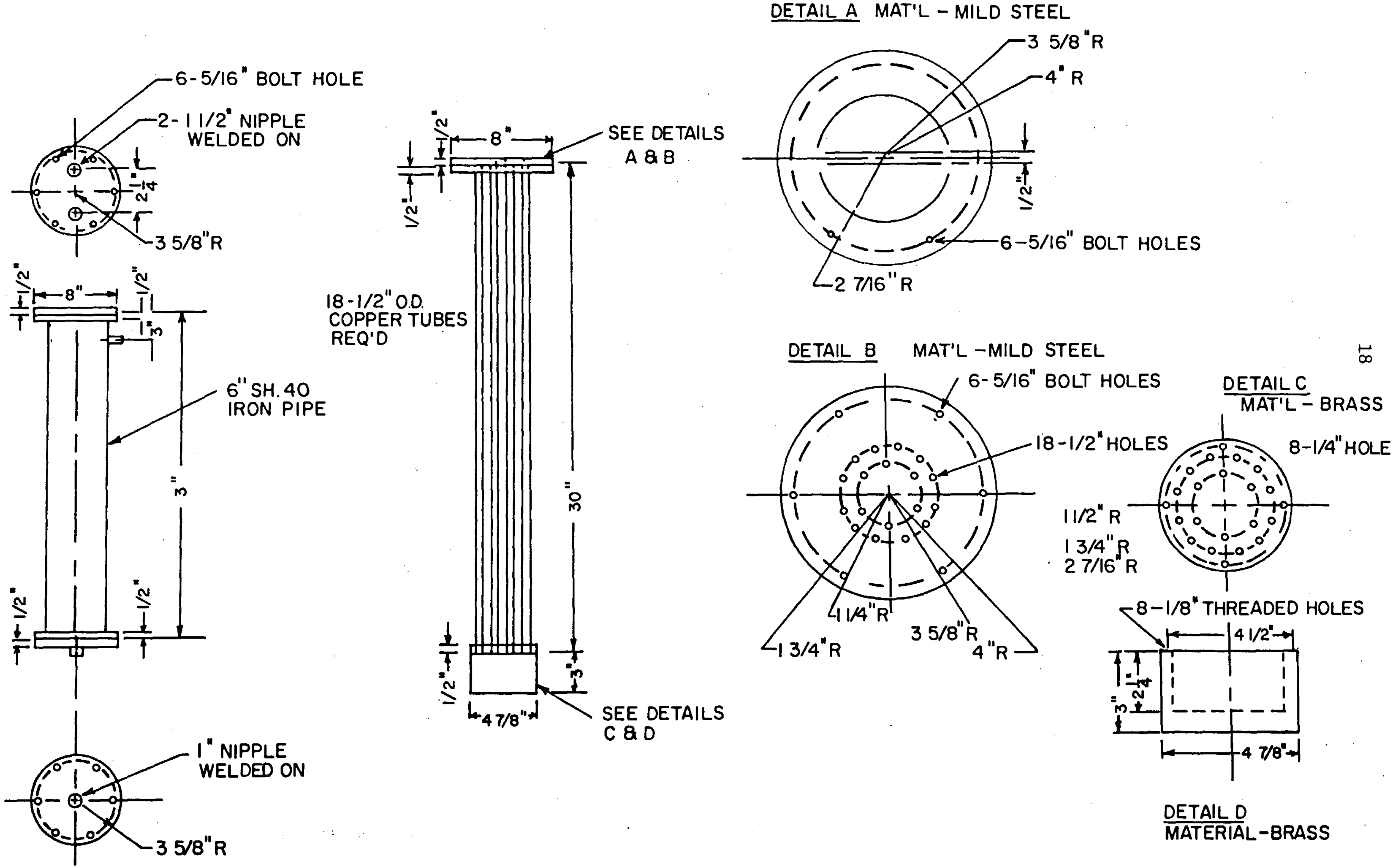
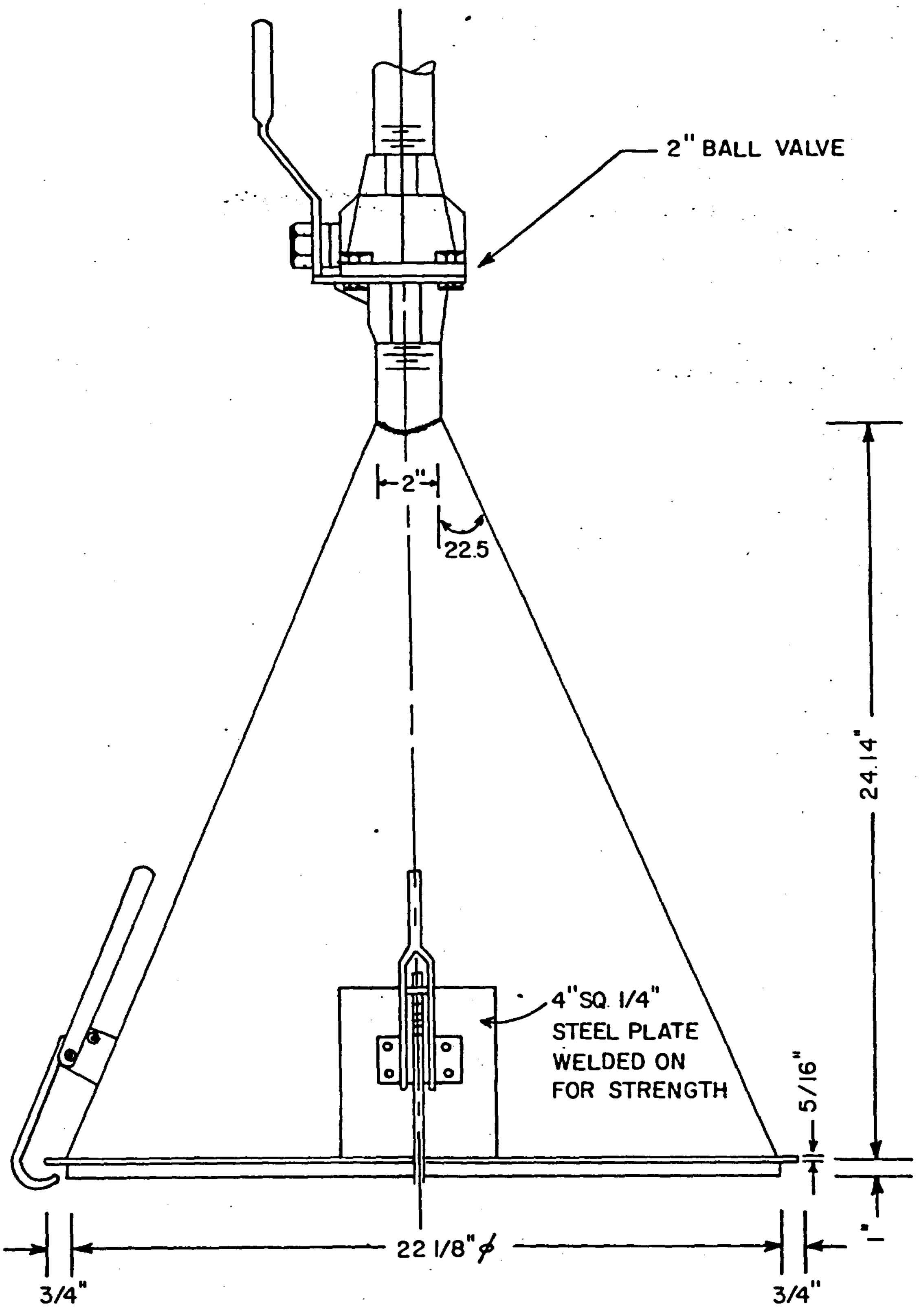


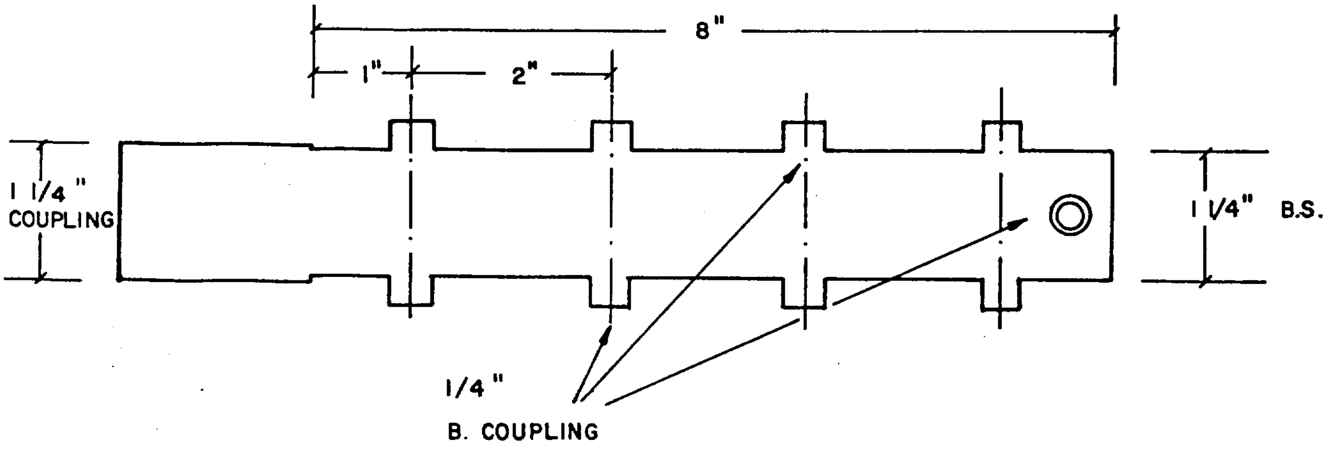
FIG. 4 SHELL AND TUBE HEAT EXCHANGER



NOTES:
(1) MATERIAL: 1/16" THICK STEEL
(2) CONE COLLER TO FIT INSIDE
DIAMETER OF STD. 44 GAL.
DRUM.

FIG. 5 FAST ENGAGING-DISENGAGING CONE

1. STEAM MANIFOLD



2. CONE SECTION MODIFICATION

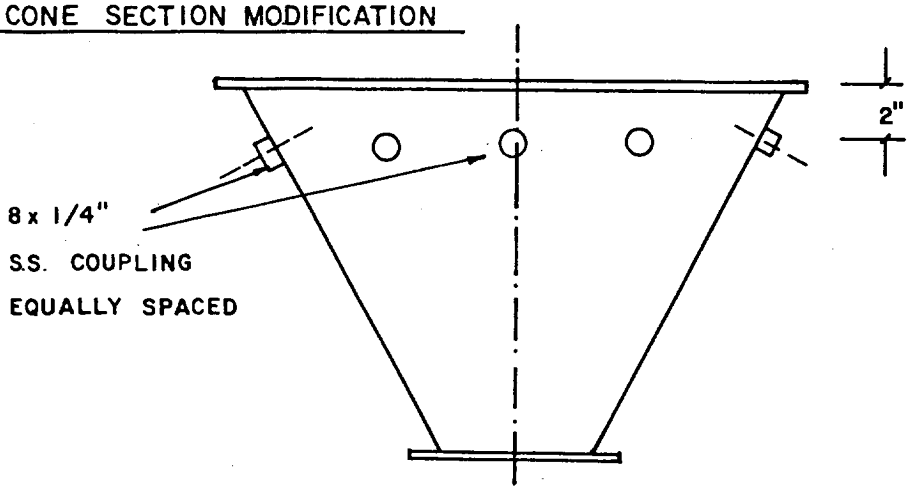
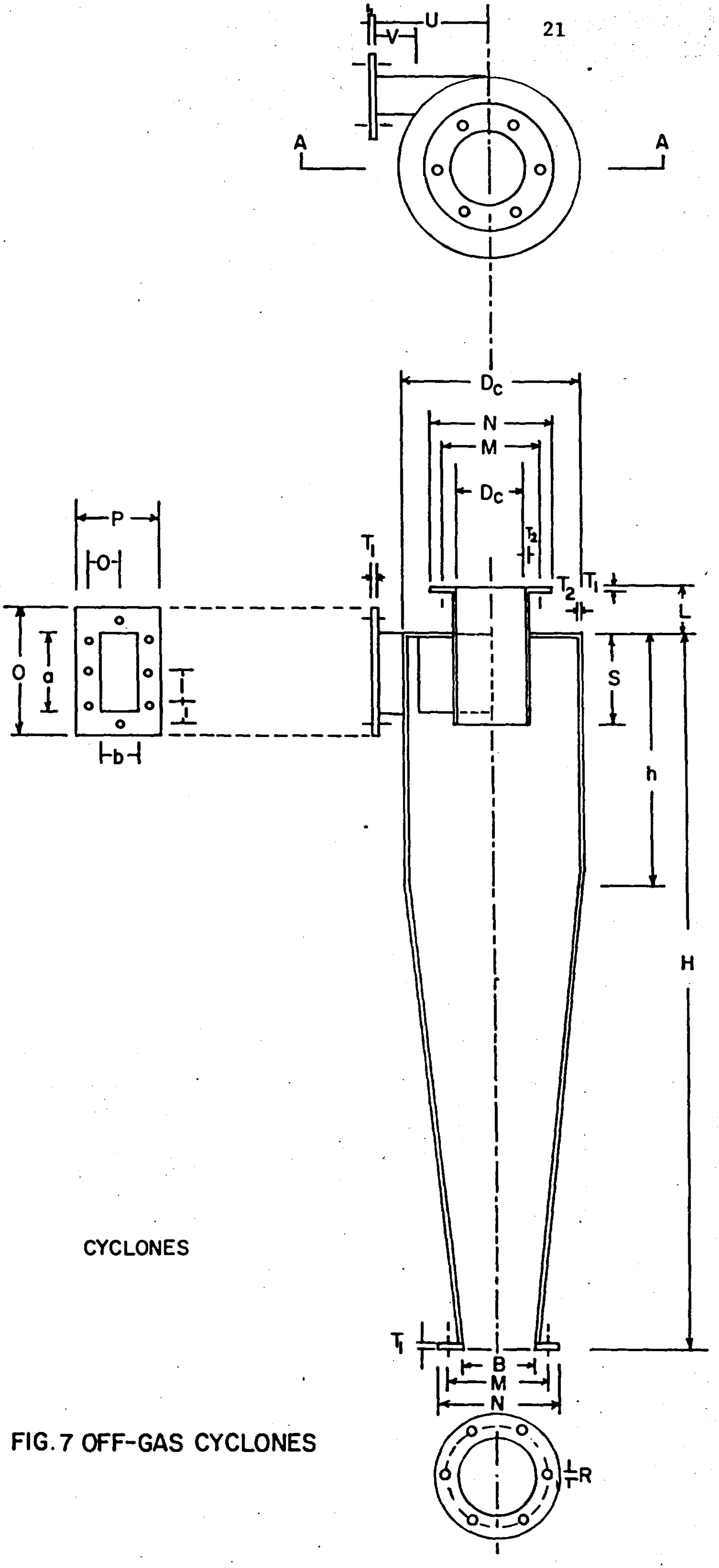


FIG. 6 STEAM INJECTION ARRANGEMENT



CYCLONES

	1	2	3
a	3.3	4 3/4	1.16
b	1.6	1 3/4	1.1
B	3	3	1 1/2
Dc	7 5/8	8 1/8	3 7/8
Dc	3	3	1 1/2
h	10 1/2	10 3/4	5 1/4
H	29 1/2	24	15
I	1.5	2 1/4	3/4
J	5/8	5/8	5/8
L	2	2	2
M	4 1/4	4 1/4	3 1/2
N	6 1/2	6 1/2	4 1/2
O	5.3	6 3/4	3.6
P	2.6	3 3/4	21
Q	1.3	1 3/8	1 1/16
R	3/8	3/8	5/16
S	3 3/4	4 3/4	1 7/8
T_1	1/2	1/2	3/8
T_2	1/16	1/16	1/16
U	5 13/16	6	3 3/4
V	2.71	2.66	2

FIG. 7 OFF-GAS CYCLONES

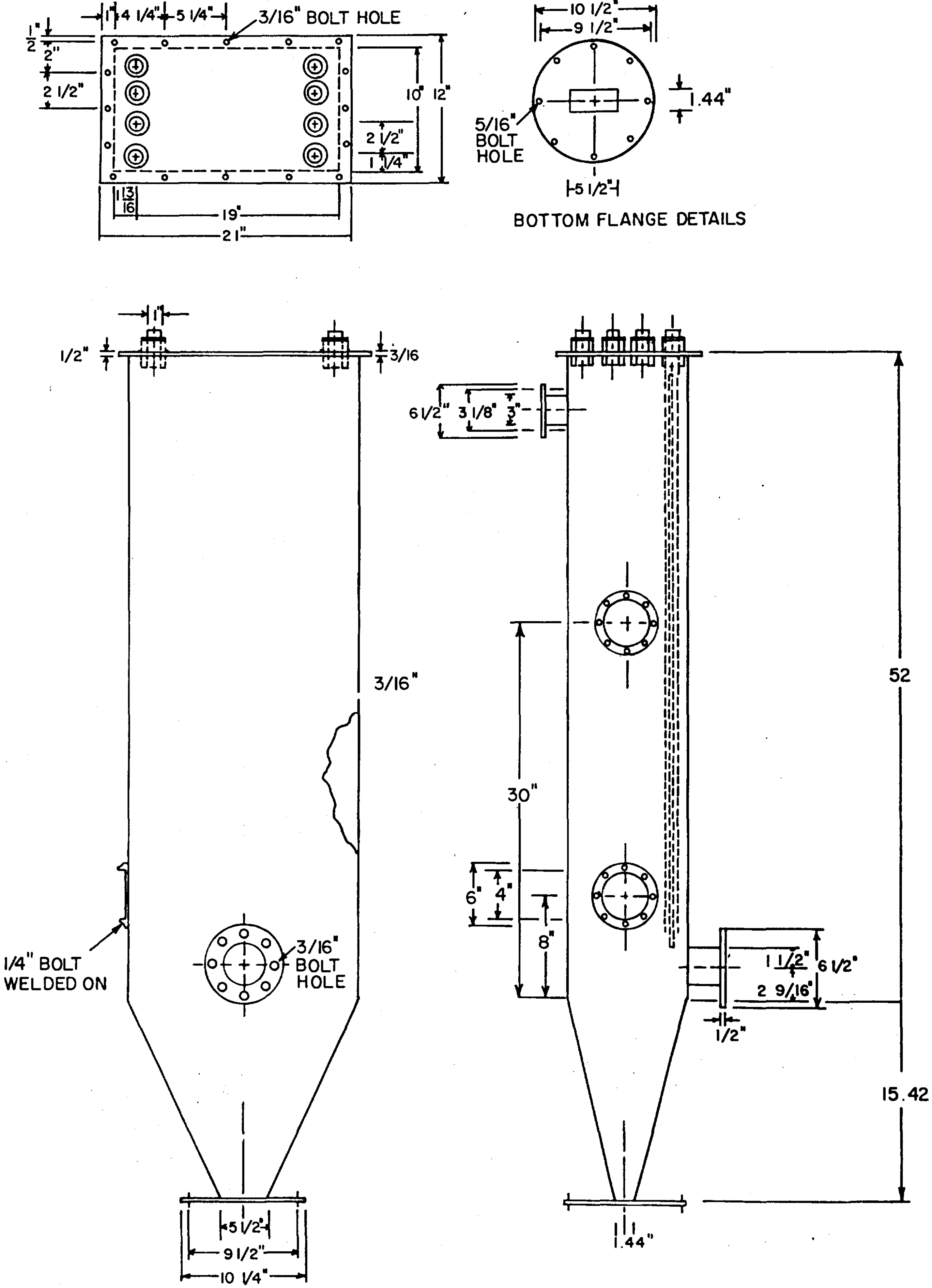
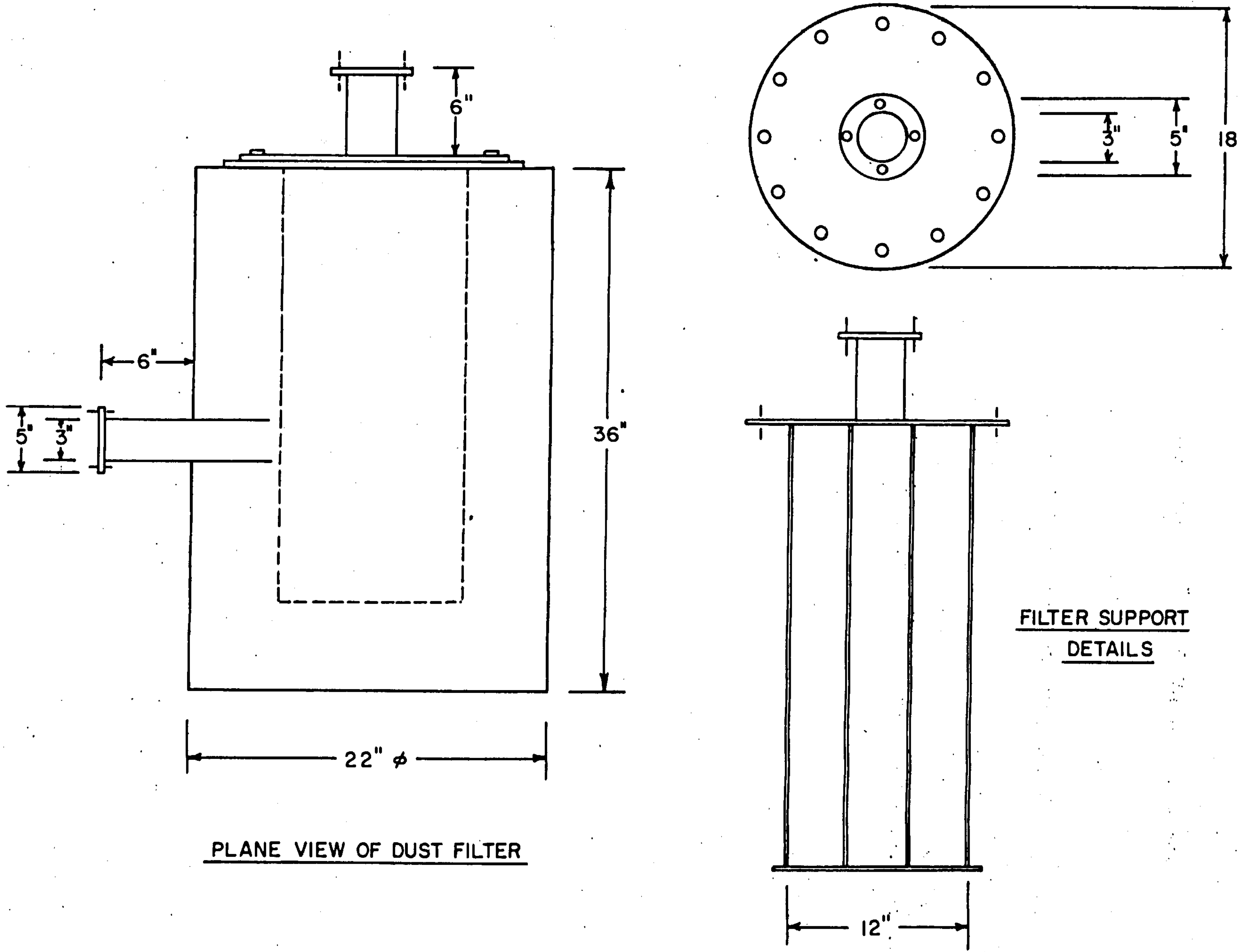


Fig. 8 Tar Condenser



PLANE VIEW OF DUST FILTER

FILTER SUPPORT DETAILS

FIG. 9 DUST FILTER DESIGN

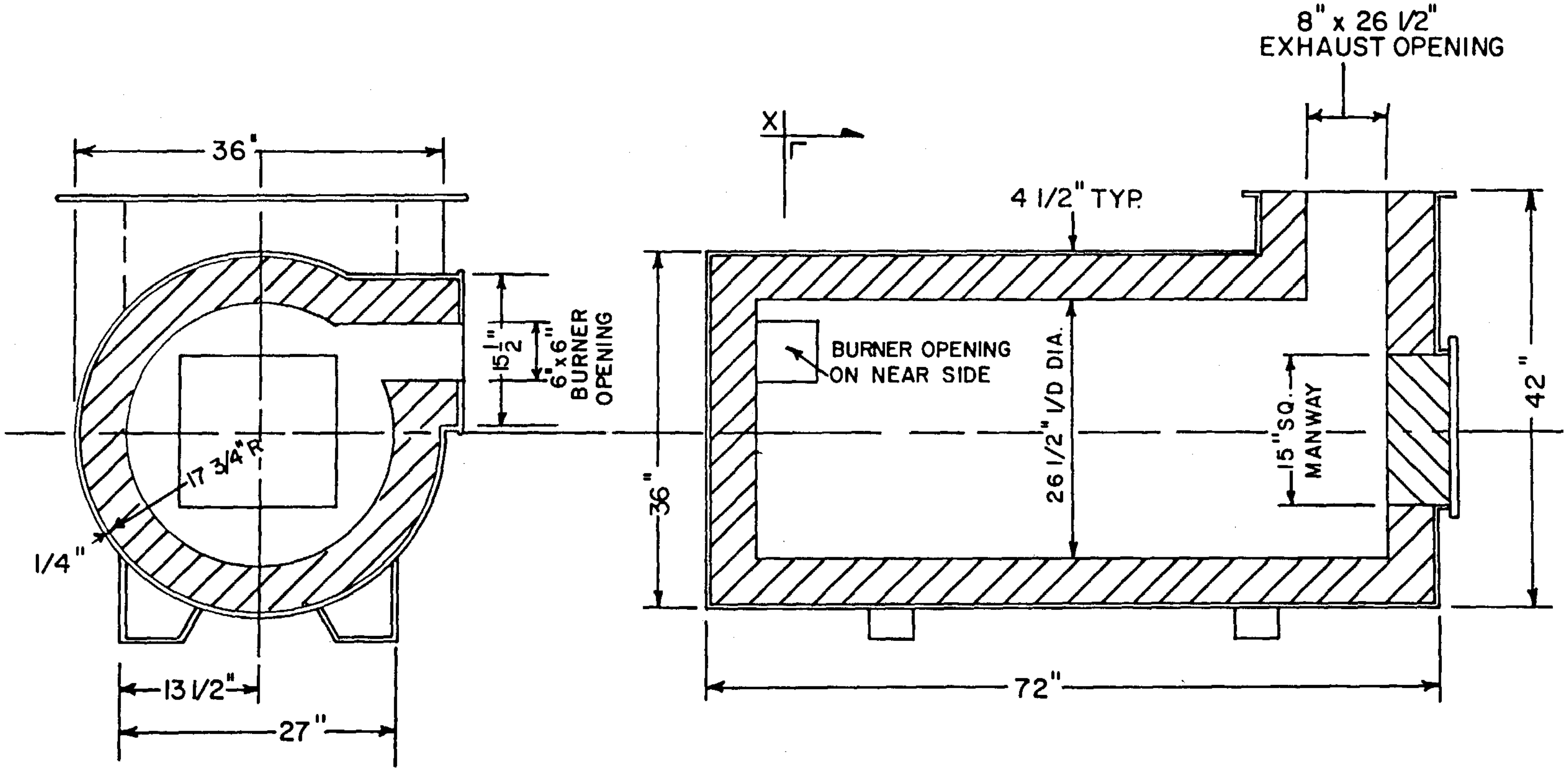


FIG. 10 INCINERATOR HOUSING

UNIVERSITY OF BRITISH COLUMBIA
DEPARTMENT OF CHEMICAL ENGINEERING
VANCOUVER, B.C.
FAIL-SAFE ELECTRICAL CIRCUIT FOR THE SPOUTED BED COAL GASIFICATION PLANT
MARCH/79

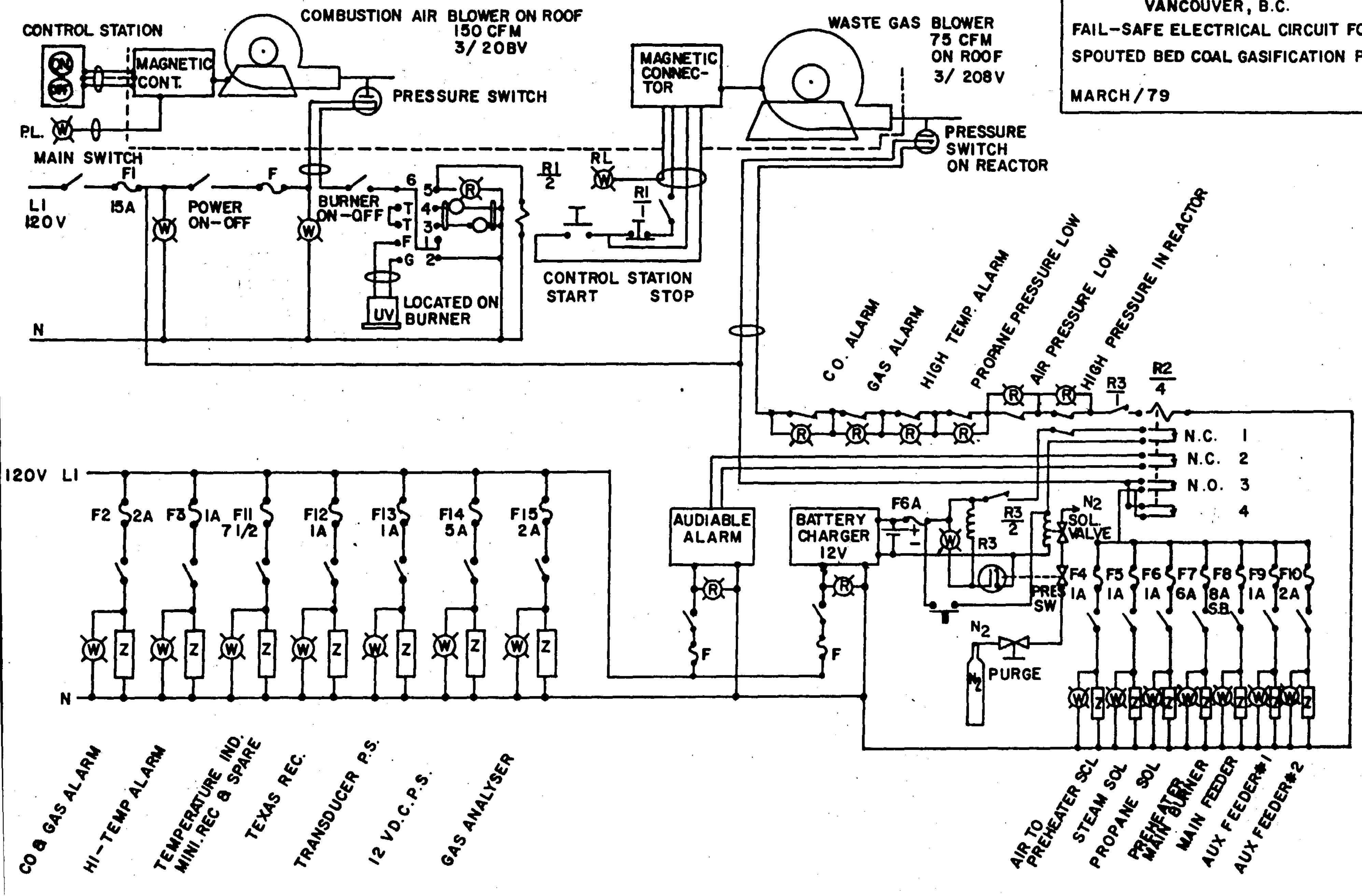


FIG. 11 FAIL-SAFE CIRCUIT

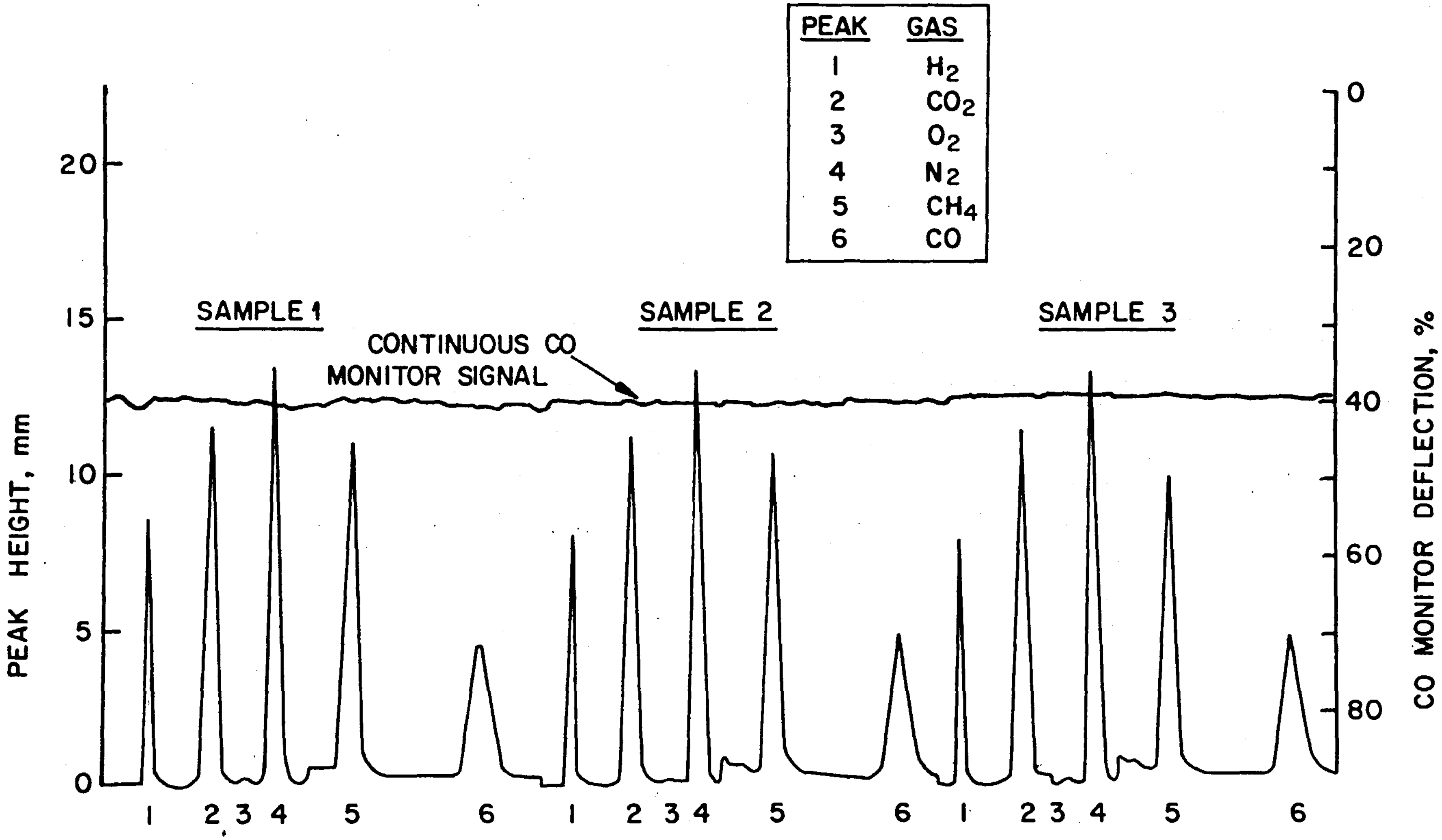


FIG. 12 TYPICAL GAS CHROMATOGRAPH TRACINGS

COAL GASIFICATION IN A SPOUTED BED

II. Results and Interpretation

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ABSTRACT

Western Canadian coals of free swelling index 0, 3 and 7 were gasified in a 0.31 m I.D. 1.4 tonne/day spouted bed process development unit described in Part I. Gasification results are compared with previously reported data in a 0.15 m I.D. reactor. Gases of heating values to 4.7 MJ/m^3 at 0.19 kg/m^2-s in the smaller unit using the same coal. The effects of coal feed rate, particle size, char recycle and top versus bottom coal feeding and other operational characteristics are reported.

INTRODUCTION

Experimental results from the 0.305 m dia. spouted bed gasifier described in Part I are presented in this paper. The objectives of the experimental program were to determine suitable operating conditions for efficient gasification of Western Canadian coals particularly those of a caking variety. In addition to general questions such as the effect of coal feed rate on gas quality, the experiments have been designed to answer specific questions such as whether it is better to feed coal into the spout with the spouting gas, or onto the top of the bed.

Three types of coal have been tested. Properties are listed in Table 1. Forestburg coal supplied by Luscar Ltd. is a highly reactive sub-bituminous coal with zero free swelling index, 36% volatiles (maf) and 67% fixed carbon. Coleman coal is a medium volatile bituminous coal of free swelling index 4, 28% volatiles and 63% fixed carbon. Sukunka coal from Chamberlain seam supplied by B.P. Canada is a highly caking coal of free swelling index 7, 25% volatiles and 75% fixed carbon (maf). These coals were supplied as 5 x 0 mm in size and were crushed and screened prior to use.

RESULTS

Tests on all three coals have been successful, yielding a gas of calorific value comparable to fluidized bed gasifiers. Caking coals were gasified without agglomeration problems, however with such coals operation is less flexible in terms of feed rate and more care is required to maintain smooth operation. Typical performance data averaged over 2-4 runs for each coal is shown in Table 2. Forestburg coal with 36% volatiles is most reactive. At throughputs of 0.22 kg/m^2-s gases with heating values up to 4.7 MJ/m^3 were produced at temperatures of 850°C and below. The best results were obtained at H_2O/coal mass ratio of 0.2, air/coal ratio of 1.8 and using 1.3-3.36 mm coal. Steam was not added to the gasifier since the coal as fed contained 20% moisture. Typical gas composition is also given in Table 2. As found in an earlier study (1) the feed rate of the less reactive caking coals was lower for a given gas quality than that for Forestburg coal. With Sukunka coal, typical feed rates were about 72% of those of the more reactive Forestburg coal, the bed temperature for gasification was about 75°C higher and slightly weaker gases of 3.3-3.6 MJ/m^3 were produced. Tar yields were generally highest with this coal. With the Coleman coal gases of 3.8 MJ/m^3 were produced at coal throughputs of 0.18 kg/m^2-s which was slightly above that for Sukunka. Bed temperatures required were about 985°C. These latter results were substantially better than those reported previously (1) with this coal in the 0.15 m dia. gasifier which was limited to lower bed temperatures, and operated at a lower throughput.

The percentage steam decomposition was calculated as % Steam Decomposition = 100 (H_2 in Gas)/(H_2 in Steam + H_2 in Coal). Under typical conditions it followed the trend of gas calorific values increasing from 43% with Sukunka, to 49% with Coleman and to 55% with Forestburg.

Most gasification runs lasted about 3 hours at steady conditions after a heat up period of several hours. Mass and heat balances for typical runs using Forestburg coal are shown in Figure 1. In this case no steam is added since there is sufficient water in the coal. The mass of gas leaving the reactor is calculated from the measured volumetric flowrate, wet gas composition, temperature and pressure. The dust in the gas is calculated by difference since measurements were not made. In recent work a gas sampling rain equipped with an Anderson classifier has been used to quantify the fine particulates. The unaccounted for carbon in the balance shown in Figure 1 includes both particulates lost in the gas and material that has deposited on the walls throughout the system. Mass balances on carbon generally closed within 10%, and were better on the longer runs. The heat balances shown in Figure 1 assumed the loss through the reactor shell could be calculated by difference. Shell thermocouples allow a direct computation in current runs. The heat of combustion of the tar also had to be assumed since no measurements have been made for the tars produced from the present apparatus. In most runs the major inefficiency is the loss of heat associated with the carbonaceous char leaving the cyclone. The two runs shown in Figure 1 allow a comparison of operation with and without char recycle. The hot raw gas efficiency

$$\eta = 100 \text{ (Heat of hot, raw gas including tar)/(Combustion Heat of Coal Feed)}$$

is about 58% with no char recycle, and 87% with recycle.

Table 2 shows that for typical conditiond, the cold gas thermal efficiency (100 x Combustion Heat of the Clean Gas at 288.6 K/Combustion Heat of the Coal Feed) also follows an increasing trend with gas calorific value and varies from 30 to 36% for the caking coals and up to 60% with the non-caking reactive coal. Recycle of the carryover product is discussed below.

Results of some tests designed to delineate preferred operating conditions are discussed first, followed by comments on the effects of process variables on gas quality. In the previous work using the 0.15 m dia. reactor coal was fed to the top of the spouted bed. It was thought that feeding coal at the apex of the cone with the air and steam would yield a better gas with lower tar content as a result of longer residence time of volatiles in the bed. Tests with two coal proved inconclusive as bottom feeding gave higher gas heating values with Forestburg, and lower heating values with Sukunka, as shown in Table 3. Tar production appeared to be unaffected. Figure 2 shows that the bed axial temperatures profile is affected by feed location, with a more uniform temperature in the upper part of the bed being observed with bottom feeding. When coal is fed to the top of the bed, the temperature near the bed surface falls yielding a profile more like that of a moving bed gasifier. Bottom feeding appears to result in better mixing which also reflected in easier control. At throughputs above 0.16 kg/m^2-s agglomeration was experienced with Sukunka coal when fed from the top. Bottom feeding of coal is therefore the preferred mode of operation, but not because of improved gas quality.

A test of the importance of steam feed location was also made. Steam was normally fed into the spout, but tests were made of steam addition into the annulus at eight peripheral locations around the cone 150 mm above the orifice. No significant effect of steam feed location was observed.

The beneficial effects of larger coal feed size reported for the 0.15 m dia. gasifier were confirmed in the present study as shown in Table 4 for both Forestburg and Sukunka coals. Gas calorific value shows a steady increase with average coal feed size. A marked effect on the carbon content of the bed is also evident. With finer coal feed at a given velocity more carbon is elutriated from the bed as expected, and the gasifier efficiency drops. Addition of 20% 1 mm coal to a feed of average size 2.18 mm resulted in a decrease in calorific value from 4.36 to 3.47 MJ/m^3 and an increase in bed carbon from 15 to 45%. The less reactive Sukunka coal builds up to greater concentrations in the bed, and hence for a given size results in much higher carryover of carbon to the cyclones.

At the substantial carryover rates shown in Tables 2 and 4 recycling of the cyclone catch to the gasifier would be required to achieve reasonable thermal efficiencies in a single stage gasification plant producing fuel gas. With no recycle, fresh coal feed of 0.20 kg/m^2-s of Forestburg coal yielded gas of 4.0 MJ/m^3 at a cold gas thermal efficiency of 54%. The loss in efficiency due to carryover was 13%, and due to sensible heat of the gases was 18%. With recycle of 8.8 kg/h of cyclone dust, gas of the same calorific value was produced with a reduction in fresh coal feed to 0.17 kg/m^2-s and a gain in cold gas efficiency to 68%. Carbon carryover not recycled represented an 8% efficiency loss, and the gas sensible heat 18%. Further work on char recycle is underway.

At a given air and steam rate, the coal feed rate has been demonstrated (1) to be the most important operating parameter as it effects bed temperature, composition, and height as well as gas quality. In Figure 3 steep increases in carbon content of the bed are observed for Forestburg and Sukunka coal as the coal feed rate is raised. An increase of 22% in the feed rate of Forestburg coal raises the bed carbon content from 10% to about 45%. High concentrations of carbon in the bed (40%) corresponding to feed rates > 43 kg/h resulted in agglomeration with Sukunka coal, particularly with top feeding. Generally up to 37% carbon in the bed could be tolerated. This was significantly higher than the value of 10% carbon reported as a maximum in the smaller spouted bed reactor. Calorific value and composition of the gas are plotted versus feed rate in Figure 4. As shown previously, increasing the feed rate up to some maximum point improves gas calorific value. This effect is due to increases in H_2 and CO in the gas with a corresponding drop in CO_2. There is virtually no change in CH_4 content.

The effect of the bed temperature at constant feed rate is shown in Figure 5. Results for each coal are taken from a given run during a slow temperature rise of the bed. For the reactive Forestburg coal, a minor improvement in calorific value is noted while a strong effect on the less reactive coal is evident. Changes in calorific value reflect increases in H_2 and to a lesser extent CO content with temperature. At still higher temperatures the curve for Sukunka coal would be expected to level out.

The lower limit of air flow to the gasifier is determined by the minimum spouting velocity which for this system is about 0.6 m/s. The bed was operated at roughly 1.2 times this velocity. In Figure 6 the effects of the air/coal and Steam/coal mass ratios are shown. The best air/coal ratio for Forestburg coal was about 1.8 while for Sukunka it was in the range 2-2.5. Attempts to lower this ratio by increassing the coal feed rate resulted in decreased bed temperatures and agglomeration problems with Sukunka coal. Preheating of the inlet air may allow higher bed temperatures to be reached and a lower air/coal ratio to be used. Steam/coal ratios are roughly one tenth the air/coal ratio and their optimal value appears to lie around 0.2 for both coals. A plot of percent steam decomposition versus steam/coal ratio shows a similar trend to the calorific value plot in Figure 6.

From the viewpoint of scale-up it is of interest to compare results from the previous study in the 0.15 m dia. reactor with those of the present work in the 0.305 m dia. reactor. This comparison is shown in Table 5. For both coals 20-33% increase in coal feed rate per unit cross section was achieved on scale-up coupled with a 10-20% improvement in gas calorific value, and a 6-16% increase in gas yield. Steam decomposition and thermal efficiency also improved with the increase in size. While improvements were expected on the basis of the simple mathematical model presented previously (1), the smaller relative heat loss in the larger system may be as important as the improved fluid dynamics characteristic of the larger system. There is also a definite improvement in thermal stability which resulted in smoother operation on the larger scale. In addition, for caking coals larger carbon content in the bed could be tolerated in the larger reactor as mentioned above.

Comparison of the data of the present work with results previously summarized (1) for fully developed large scale gasifiers gives further encouragement to continue the development of the spouted bed process.

CONCLUSIONS

Gasification of three Western Canadian coals in a 0.305 m dia. spouted bed gasifier has been demonstrated successfully in short term trials at feed rates corresponding to 1.4 tonnes/day. Tests showed that bottom feeding of coal to the orifice of the spouted bed was preferable to feeding to the top of the bed. Although larger size feed particles give stronger gases, recycle of partially gasified cyclone dust improves the cold gas thermal efficiency. A comparison of results with those of a previous study in a 0.15 m dia. reactor shows improvements in throughput, gas quality, gas yield, and thermal efficiency with increasing reactor size.

ACKNOWLEDGEMENTS

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Table 1 - Coal analysis

1.	Ultimate Analysis	Forestburg	Sukunka	Coleman
%	C	66.79	80.3	78.05
	H	4.25	4.45	4.49
	S	0.6	0.5	0.61
	Ash	10.48	10.81	9.83
	O	16.28	2.56	5.75
	N	1.60	1.37	1.27
2.	<u>Proximate Analysis</u>			
	% moisture	23.70	0.82	3.04
	% volatiles	27.38	22.32	24.47
	% fixed carbon	40.92	66.14	62.66
3.	Free Swelling Index	0	7	3.5

Table 2 - Typical performance data *of 0.305 m spouted bed gasifier

Coal Type	Forestburg	Sukunka	Coleman
Coal feed rate (kg/h)	60	43	47
Coal throughput (kg/m^2-s)	0.23	0.16	0.18
Air/Coal (kg/kg)	2.1	2.7	2.8
Steam/Coal (kg/kg)	0.24	0.23	0.26
Av. Bed Temp. (°C)	805-850	920-965	985
Gas Yield (m^3/kg coal)	3.3	2.8-3.5	2.9-3.2
Tar yield (kg/kg coal)	0.004-0.02	0.006-.017	0.002-0.003
Cyclone catch (kg/kg coal)	0.11-0.20	0.29-0.42	0.28
Cold gas efficiency (%)	52-60	30	36
Total steam decomposition (%)	55	43	39
Gross heating value of gas (MJ/m^3)	4.4-4.7	3.3-3.6	3.7-3.8
Gas composition (% Vol)			
H_2	15.5	13.0	14.3
CO	14.5-20.	9.1-12.7	13.2
CO_2	9.8-12.3	10.7-13.9	11.9
CH_4	1.3-1.6	1.6-2.1	1.7
N_2	54.5	59-66	60.4

*Average of 2-4 runs for each coal

Table 3 - Effect of coal feed method

Coal Type	Coal Feed Method	Gas Heating value MJ/m^3	Tar Yield kg/kg coal	Solids Carryover kg/kg coal
Forestburg	Bottom	4.36	0.003	0.20
	Top	3.93	0.004	0.17
Sukunka	Bottom	3.14	0.008	0.41
	Top	3.30	0.006	0.39
Coleman	Bottom	3.80	0.002	0.36

Table 4 - Effect of particle size

Coal Size Range US Sieve No.	d_p,mm	Coal Type	Feed Rate kg/h	Temp °C	Bed Carbon Content wt %	Gas Heating Value MJ/m^3
-18+45	0.736	Forestburg	54.9	870	<1	2.84
80%-6+18 20%-18+0	1.389	Forestburg	59.4	850	15	3.47
-6+18	2.057	Forestburg	61.4	810	45	4.36
-4+8	3.54	Forestburg	58.5	855	26	4.41
-18+50	0.45	Sukunka	35	1070	<1	2.50
-6+0	0.418	Sukunka	46.7	930	19	2.96
-6+18	2.057	Sukunka	42.7	940	37	3.15

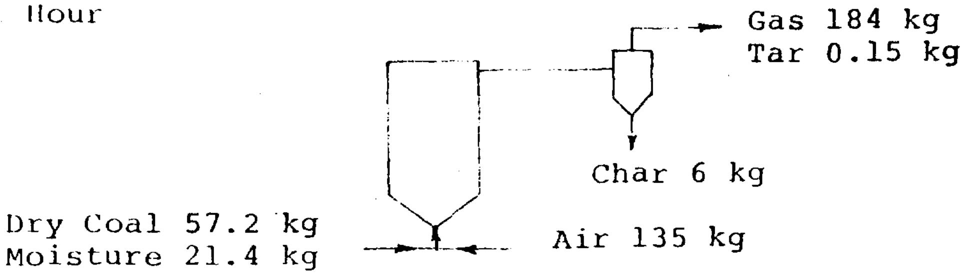
Table 5 - Comparison of gasification in 0.15 and 0.305 m diameter spouted beds

Coal	Forestburg		Sukunka	
Reactor diameter (m)	0.15	0.305	0.15	0.305
Bed depth (m)	0.40	1.0	0.40	0.9
Coal feed rate (kg/h)	12	61.3	7.6	43.7
Coal throughput (kg/m^2-s)	0.19	0.23	0.12	0.16
Gas yield (m^3/kg coal)	3.05	3.22	2.44	2.84
Gas calorific value (MJ/m^3)	3.60	4.36	2.82	3.14
Thermal* efficiency %	32	53	25	30
Steam utilization %	40	53	27	41
Bed temperature °C	860	810	860	915

*Cold gas efficiency = 100 (Combustion Heat of Product Gas at 288.6K)/(Combustion Heat of Coal Feed)

Fig. 1 Mass and Heat Balances

BASIS: 1 Hour



Mass Balance

Coal + Air = 78.6 + 135 = 213.6 kg

Gas + Tar + Char = 184 + 0.15 + 6 = 190.2 kg

Loss = 23.4 kg

Heat Balance

Coal = 1148 MJ

Gas (Combustion Heat) = 641 MJ

Gas (Sensible Heat) = 220 MJ

Tar = 19.4 MJ

Char = 92 MJ

972.4 MJ

Unaccounted For Carbon 123 MJ

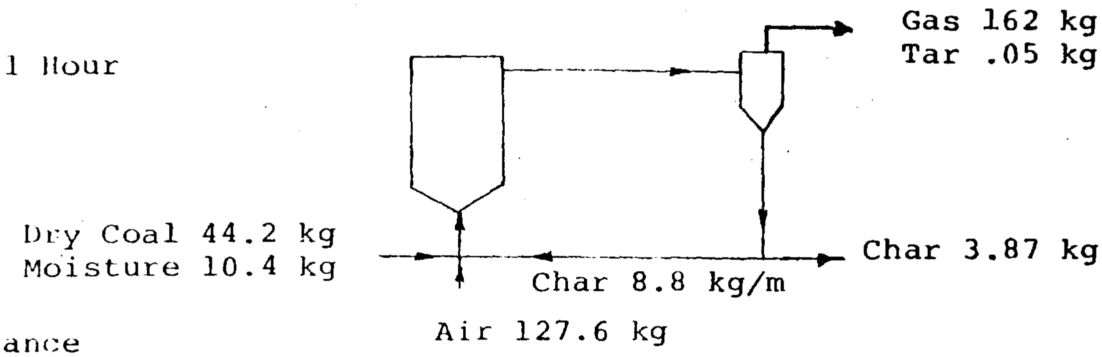
1095.4 MJ

$\eta_{Raw} = (641+220+19.4)/1148$

$= 0.767$

Losses = 1148 - 1095.4 = 52.6 MJ

BASIS: 1 Hour



Mass Balance

Coal + Air = 54.6 + 127.6 = 182.2 kg

Gas + Tar + Char = 162 + 0.05 + 3.87 = 165.92 kg

Loss = 16.28 kg

Heat Balance

Coal = 885 MJ

Gas (Combustion Heat) = 602 MJ

Gas (Sensible Heat) = 149 MJ

Tar = 18.8 MJ

Char = 93.4 MJ

863.2 MJ

Unaccounted For Carbon 0.

$\eta_{Raw} = (602+149+18.8)/885$

$= 0.870$

Losses = 885 - 861 = 24 MJ

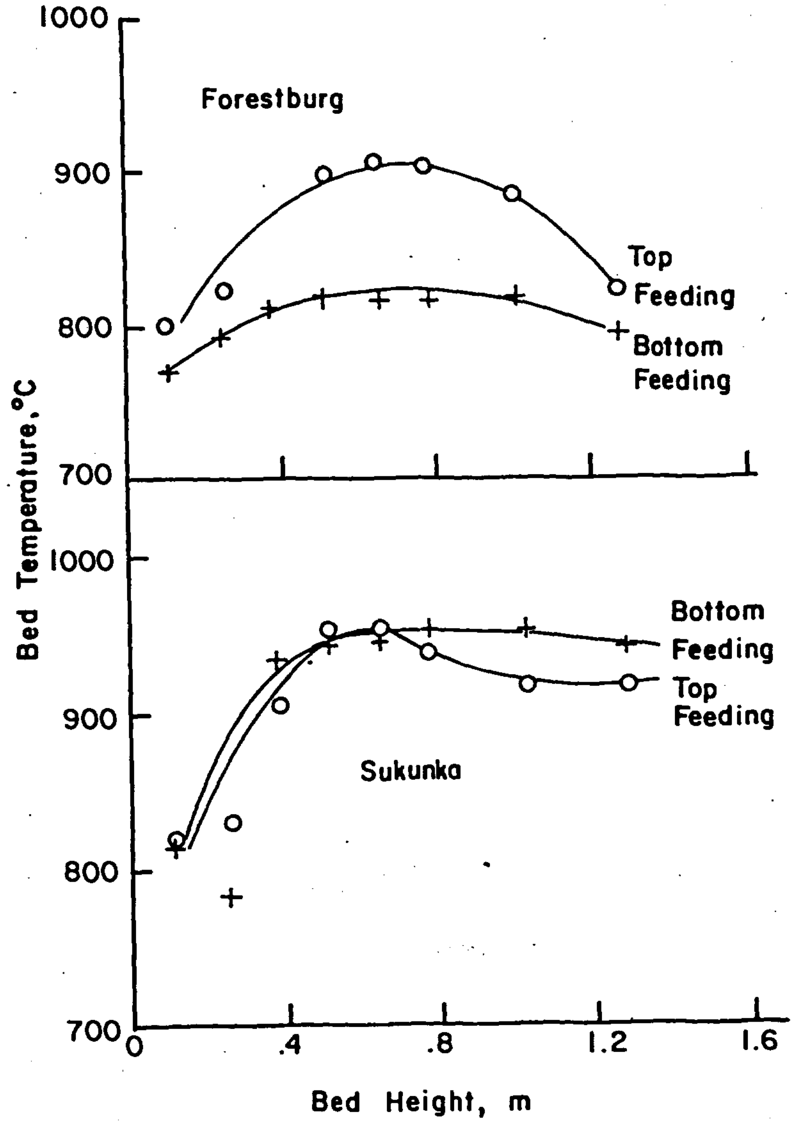


Fig. 2 Typical Axial Temperature Profile Measured 50 mm from Wall.

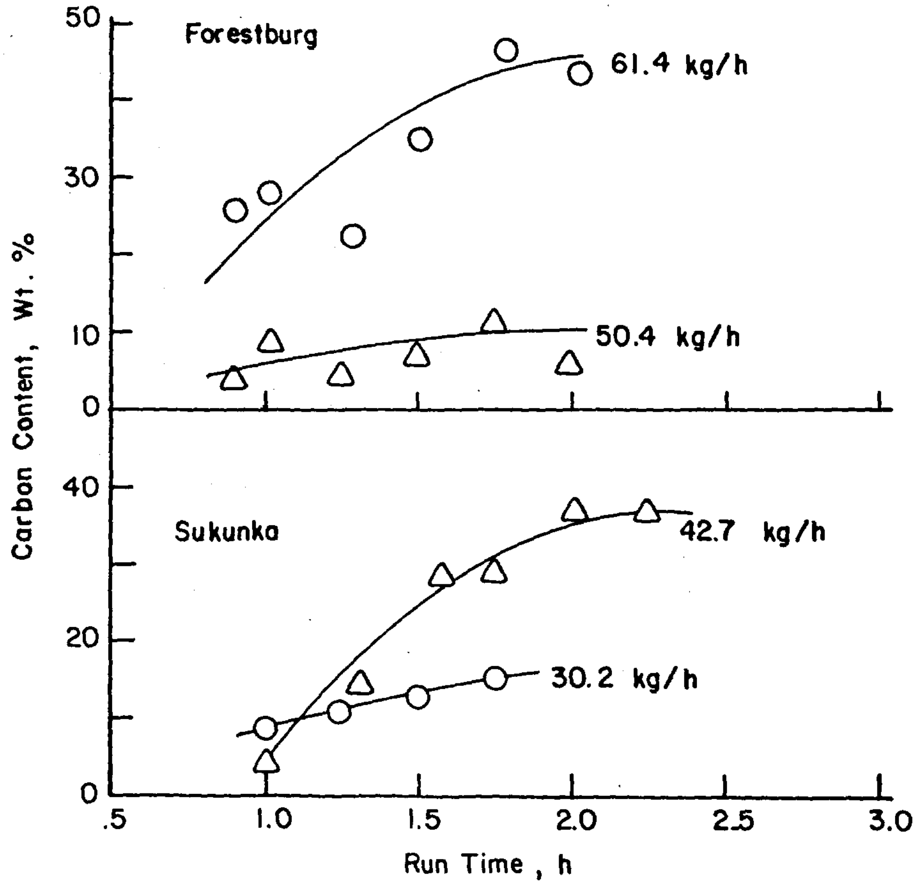


Fig. 3 Effect of Coal Feed Rate on Bed Composition
(Bottom Feeding, Coal Size -6 +18 mesh)

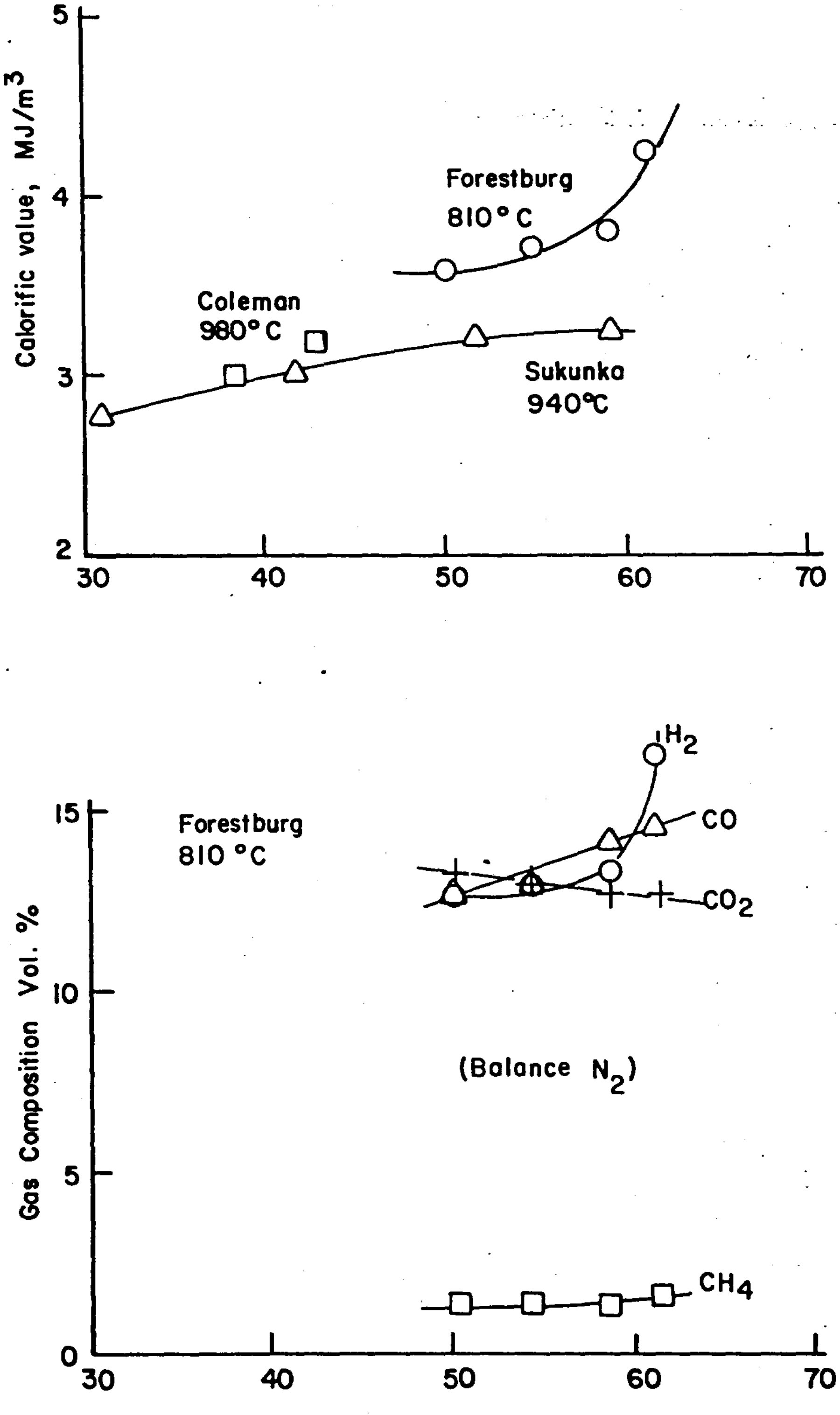


Fig. 4 Effect of Coal Feed Rate on Gas Quality (Bottom Feeding, Coal Size -6 + 18 mesh)

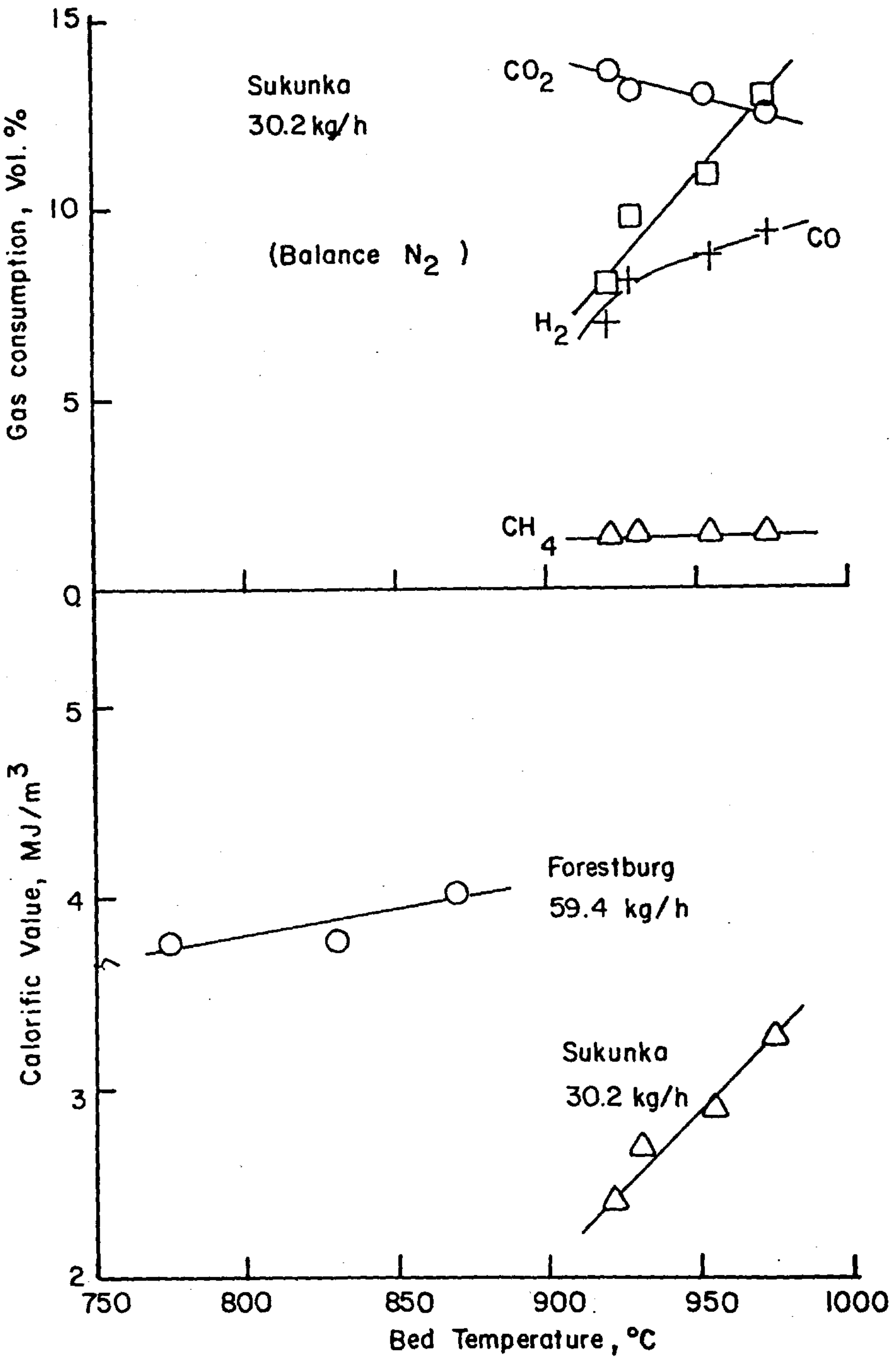


Fig. 5 Effect of Bed Temperature on Gas Quality

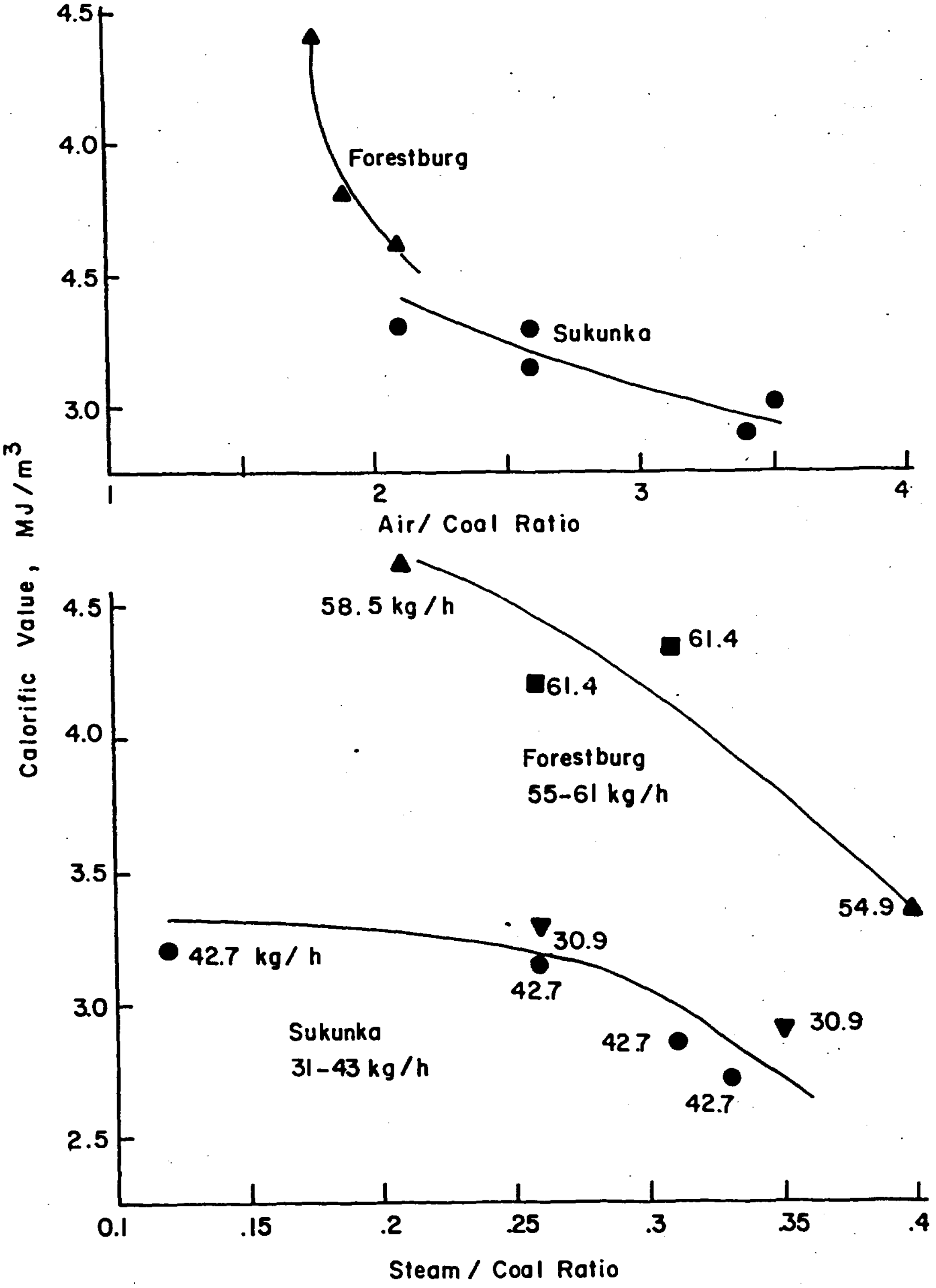


Fig. 6 Effect of Air/Coal and Steam/Coal Mass Ratios on Gas Quality

MOLTEN SALT COAL GASIFICATION PROGRESS FOR CLEAN FUEL GAS PRODUCTION

by

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SUMMARY

Most coals have a high sulphur content (1-5%) and suitable means must be found to utilize this untapped fossil reserve. An economic process to convert them to a fuel gas and meeting the present emission standards is required.

"Molten Salt Coal Gasification Processes" which accomplish simultaneous gasification and partial desulphurization have been developed independently by M.W. Kellogg Co. and Atomics International for the production of fuel gas from coal. In these processes, coal is gasified in turbulent bed of molten sodium carbonate with oxygen or air to produce medium and low-Btu gas respectively.

The unique feature of these processes is that sulphur of the coal reacts with the molten salt (Na_2CO_3) to form sodium sulphide (Na_2S) and remains in the melt. However, these processes can retain only part of the sulphur, consequently, an expensive gas purification step is required to remove this partial impurity from the product gas.

A detailed thermodynamic investigation was carried out to determine the conditions under which more sulphur would be retained in the melt. The results indicate that relatively complete desulphurization could be effected in a molten carbonate bath, if a heavy metal carbonate ($FeCO_3$ or $CaCO_3$) were added to the molten salt bath.

We believe that these results point toward a significant improvement in coal gasification technology and should be confirmed by experimental work. A preliminary design of a bench-scale unit and an experimental program to verify these results is presented.

1. INTRODUCTION

Coal can be converted to various types of gaseous products ranging from low-Btu gas to high-Btu gas. However, irrespective of the gasification route adopted, the final product must be essentially free of particulate matter (mainly fly ash) and gaseous impurities (NO_x, SO_x, H_2S).

In general, the problems associated with these undesirable elements have been approached in many ways:

- removal of fly ash by using wet scrubbers, dry filters, or by electrostatic precipitations.
- suppressing the formation of NO_x by injecting steam, recirculating the flue-gas, or carrying out the reaction in two stages.
- removal of sulphur prior to gasification (beneficiation and washing techniques) or employing tail-end purification schemes using suitable solvents.

These "add-on purification" methods are inherently expensive and are sometimes ineffective for lowering the impurities to a tolerable level.

To alleviate some of these problems, a few process developers proposed "in-situ purification" schemes where suitable materials were employed to retain the undesirable elements in the gasifier. For instance:

- M.W. Kellogg Co. (1-4) carried out the reaction in a bath of molten sodium carbonate to gasify coal. Pulverized coal along with oxygen and steam was fed in the reactor operating at 1700°F and 80 atm, to produce a synthesis gas which was subsequently upgraded to a high-Btu fuel gas. Ash and part of the sulphur were retained in the reactor.
- Atomics International (5-7) utilized the similar technique, but aimed at the production of low-Btu gas. Air instead of oxygen was used and the gasification reactions were carried out in absence of steam at 1800°F and 20 atm. Ash and most of the sulphur were retained in the reactor.

Basic characteristics and other advantages of these "Molten Salt Coal Gasification" processes are summarized in Table 1.

However, a closer examination of the Kellogg's process revealed that a portion of the sulphur escaped in the gas stream as H_2S, because the less stable sulphide (Na_2S) is partially decomposed in presence of carbon dioxide and steam at elevated temperature.

$$Na_2S_{(1)} + CO_{2(g)} + H_2O_{(g)} \text{-------} Na_2CO_{3(1)} + H_2S_{(g)}$$

Consequently, an additional purification step was necessary to remove this partial sulphur impurity from the product gas.

In the case of Atomics International, most of the sulphur was indeed captured as Na_2S, because the reverse reaction (decomposition of sulphides) does not take place to an appreciable extent in absence of steam, and especially when lower pressures are employed. However, the disadvantages are that the rate of gasification and the amount of gas produced is relatively low. In fact, bench-scale experiments conducted by Atomics International (7) confirmed that some coals cannot be completely gasified without the addition of steam.

It is evident from above, that the existing Molten Salt Coal Gasification processes have significant merit and offer a promising alternative to the conventional pollution abatement methods. However, these processes have certain limitations as mentioned above, and this study was directed towards investigating these areas and to explore the conditions under which more sulphur would be retained in the melt.

Table 1 - Basic characteristics and advantages of molten salt coal gasification process

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COAL FEED

- Accepts all types of coals (including caking coal or other carbonaceous materials)
- Close sizing and pulverizing not required.
- Pretreatment of coal not required.

PROCESS AND EQUIPMENT

- Single-stage gasification with high carbon utilization.
- The molten salt catalyzes the gasification reaction.
- Relatively smaller reactor (gasifier) required.
- High turn-down capability.

PRODUCT GAS

- Low or medium Btu gas ($\simeq$ 150 or 300 Btu/scf).
- Neglible tar, phenol or NH_3.
- Low H_2S content.

BY PRODUCTS

- No char.
- Sulphur recovered in the elemental form.
- Ash, as disposable wet cake.

2. THERMODYNAMIC INVESTIGATIONS TO DETERMINE THE CONDITIONS FOR SIMULTANEOUS GASIFICATION AND DESULPHURIZATION OF COAL

A detailed review of the existing molten salt coal gasification processes revealed that the temperature, pressure, steam/oxygen ratio, and the type of carbonate (molten salt) used are all important factors influencing the retention of sulphur in the melt, and thus the final composition of the product gas. For this reason, the investigation was directed towards two specific questions:

1. Is it possible to retain more sulphur by manipulating the process variables, when molten sodium carbonate is used?
2. Do other carbonates (such as K_2CO_3, $CaCO_3$, $FeCO_3$) retain more sulphur than sodium carbonate under similar operating conditions?

To answer these questions, we made an extensive use of an interactive computerized program F*A*C*T (8-10) (Facility for the Analysis of Chemical Thermodynamics) to thermodynamically predict the equilibrium composition of the product gas. Calculations were done for both low and medium Btu gas production, by varying the operating variables as follows:

- carbonates used: Na_2CO_3, K_2CO_3, $CaCO_3$, and $FeCO_3$.
- effect temperature:
  - pressure used: 1 atm
  - temperature varied: 1073 - 1273°K
  - steam/oxygen ratio (kg/kg) varied: 0-0.42
- effect pressSure:
  - temperature used: 1273°K
  - pressure varied: 20 - 40 atm
  - steam/oxygen ratio (kg/kg) varied: 0-0.42

2.1 Basis and Assumption

To establish the material balance and obtain coal gasification data for a molten salt gasifier, the following simplifying assumptions were made:

- the mass balance calculations are conducted on the basis of 100 kg of coal being gasified. For calculation purposes, a reference coal was used, the ultimate analysis of which is given in Table 2.

- the temperature is uniform throughout the three-phase molten salt bath.
- the operating pressure is the pressure of the product gas above the molten salt bath.
- the thermodynamic equilibrium conditions are assumed to prevail at the temperature and pressure of the gasifier.
- equilibrium calculations are performed considering that eleven reactions are occurring in the gasifier (see Table 3).
- for these equations the equilibrium constants were evaluated from the Gibbs Free Energy Data generated from the F*A*C*T program. Plots of equilibrium constants (lnk) versus reciprocal of absolute temperature are linear, and this is shown in Figure 1.
- all phases are ideal, and the activity coefficients of the reacting species are unity.
- the reactions of silica and alumina (present in ash) with molten carbonates have been neglected.

2.2 Calculation Procedure

As a first step, the data base* for all species involved in the calculation were generated using the "INSPECT" and "REACTION" sub-routines of the F*A*C*T program. Basically the procedure involves specifying the stoichiometry of the reactants, the temperature and pressure at which the reaction is to be carried out, and the possible product species which are likely to be formed. The solution is iterative and the program converges if the Gibbs Free Energy of the system is minimized and the material balance equations are satisfied.

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*Data base includes standard heat of formation (ΔH_f^o), standard entropy (s^o), heat of phase transition (ΔH_t), heat capacity (C_p) and values of thermodynamic properties change such as enthalpy (ΔH), Gibbs energy (ΔG), internal energy change (ΔU) etc.

This data is not presented here, but is included in the final report(11).

2.3 Results

Some of the results showing the variation in the composition of the medium Btu gas as a function of temperature and pressure are presented in Tables 4-7. In these tables the effect of steam/oxygen ratio and the performance of various carbonates on the quality of the product gas are also included. Similar calculations were done for the low Btu gas (using air instead of oxygen) and are presented in the final report (11).

It is seen from Tables 4 and 5 that when Na_2CO_3 is used and no steam is introduced into the gasifier, the total sulphur impurities do not exceed 180 ppm. This is in agreement with the observation made by Atomics International. However, when 0.28 kg of steam/kg of coal is added, the sulphur impurities in the gas stream increase to over 3800 ppm (even when atmospheric pressure is utilized).

Furthermore, if the pressure is increased to 20 atm, then the sulphur impurities increase to 1200 ppm (when no steam is used) and to over 10,000 ppm when 0.28 kg of steam/kg of coal is introduced. The performance of Na_2CO_3 is even worse (over 15,000 pp, or 1.5% by volume) when a higher pressure of 40 atm is used.

On the other hand, the metal carbonates ($CaCO_3$, $FeCO_3$) form a very stable sulphide, and the sulphur impurities in the product gas does not exceed 70 ppm. In other words, most of the sulphur of the coal (over 99.5%) would be retained in the melt if these carbonates are present.

2.4 Discussion and Conclusions

The following points are presented below for the integration of discussion and conclusions.

(a) Basis and Assumptions:

Calculations were done on the basis of a certain amount (100 kg) of the coal being gasified. The choice may appear somewhat arbitrary but is convenient because it makes the calculations almost completely independent of the capacity and design features of the gasifier.

Certain other assumptions were also made in order to simplify the calculations. For instance, the reaction of ash content with the molten salt was neglected. We do recognize that the silica and alimina of the ash completely react with carbonates to form complex silicates, but the effect of this reaction is really secondary in nature.

(b) Calculation Procedure:

The equilibrium calculations were done using the "EQUILIB" sub-routine of the F*A*C*T program which employs a general mathematical technique to estimate the product gas composition by minimizing the Gibb's free energy of the system.

The product gas composition predicted by this method was cross-checked against the equilibrium constants, and the difference was found to be neglible (less than 3%).

(c) Product Gas Composition:

The composition of the product gas predicted using F*A*C*T program was compared with the experimental data reported by Atomics International (7,14). Table 8 is the comparison of our calculated values against the experimental results obtained under almost similar operating conditions.

It is seen that, in spite of several simplifying assumptions made in our calculations, the values obtained are close to the experimental data.

Temperatures in the range of 1073 to 1273°K have a little effect on the overall product gas composition. The principal factors affecting gas composition are steam/oxygen ratio, pressure, and the type of carbonate used.

(e) Composition of the Alkaline Heavy-Metal Carbonate Mixture

As much as it is desirable to use a heavy-metal carbonate for the retention of sulphur, its direct use as a molten salt is restricted because most of the heavy-metal carbonates decompose without melting at ordinary pressures. For example, $CaCO_3$ normally decomposes into CaO and CO_2 if heated above 1025°K. However, if it is added to $(Na_2, K_2)\ CO_3$, it forms a eutectic mixture (15-16) and remains in the molten form even at 1250°K. Therefore, the molten alkaline carbonates $(Na_2, K_2)\ CO_3$ could be used as suitable solvents.

On this basis, we estimated the optimum composition of the alkaline-heavy-metal carbonate mixture that may be used for simultaneous gasification and desulphurization of coal. Calculations were done to determine the theo-

retical amount of $CaCO_3$ or $FeCO_3$ which must be present to react with all the sulphur of the coal*. Table 9 gives the final composition of the mixture, when the required amount of heavy-metal
carbonate is added to the $(Na_2,K_2)\ CO_3$ mixture**.

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* Reference coal (see Table 2)

** Eutectic composition of $(Na_2,K_2)\ CO_3$ mixture is given

Table 2 - Ultimate analysis of the reference coal used in the calculation

	Weight per cent
Carbon	67.3
Hydrogen	3.2
Oxygen*	7.0
Moisture	4.8
Nitrogen	1.0
Sulphur	4.3
Ash	12.4
	100.0

* By difference

TABLE 3

EQUILIBRIUM CONSTANTS* FOR THE MOLTEN SALT COAL GASIFICATION REACTIONS

No	REACTIONS	EQUILIBRIUM CONSTANTS AT			
		973°K	1073°K	1173°K	1273°K
1	$C + 0.5\ O_2 = CO$	4.22×10^{10}	1.16×10^{10}	3.96×10^{9}	1.59×10^{9}
2	$CO + 0.5\ O_2 = CO_2$	4.26×10^{10}	1.64×10^{9}	1.10×10^{8}	1.13×10^{7}
3	$C + O_2 = CO_2$	1.80×10^{21}	1.90×10^{19}	4.36×10^{17}	1.80×10^{16}
4	$C + H_2O = CO + H_2$	1.49	7.13	26.09	77.81
5	$CO + H_2O = CO_2 + H_2$	1.50	1.0	0.727	0.557
6	$CO + 3\ H_2 = CH_4 + H_2O$	8.9×10^{-2}	6.0×10^{-3}	7.0×10^{-3}	1.0×10^{-4}
7	$C + CO_2 = 2CO$	0.991	7.08	35.88	139.7
8	$Na_2CO_3 + H_2O = 2NaOH + CO_2$	4.61×10^{-5}	2.03×10^{-4}	6.88×10^{-4}	1.91×10^{-4}
9	$Na_2S + 2CO_2 = Na_2CO_3 + COS$	0.162	5.68×10^{-2}	2.50×10^{-2}	1.29×10^{-2}
10	$Na_2CO_3 + H_2S = Na_2S + CO_2 + H_2O$	0.193	0.791	2.43	6.0
11	$Na_2CO_3 + CO = 2Na + 2CO_2$	1.19×10^{-16}	1.26×10^{-13}	3.8×10^{-11}	4.41×10^{-9}

* Equilibrium constants have been calculated from Gibbs Free Energy Data (ΔG),

TABLE 4

CALCULATED COAL GASIFICATION DATA FOR MEDIUM BTU GAS

EFFECT OF TEMPERATURE

(Pressure used = 1 atm)

COAL, Kg	100		100		100		100	
OXYGEN, Kg	76		76		76		76	
STEAM, Kg	-		-		-		-	
steam to oxygen weight ratio, Kg/Kg	0.0		0.0		0.0		0.0	
Steam to oxygen mole ratio	0.0		0.0		0.0		0.0	
Carbonate used	Na_2CO_3		K_2CO_3		Ca CO_3		$FeCO_3$	
Temperature, K	1073	1273	1073	1273	1073	1273	1073	1273
Prod. gas composition (wet basis) mole % or Volume %								
CO	73.98	74.20	73.97	74.17	74.36	74.40	73.71	69.13
H_2	22.49	23.73	22.51	23.41	22.63	23.00	16.06	20.99
CH_4	0.94	0.02	0.92	0.01	0.94	0.10	5.39	-
CO_2	1.97	1.32	1.99	1.53	2.00	1.05	4.80	3.91
H_2O	0.60	0.75	0.60	0.86	0.10	0.66	0.04	5.96
N_2	-	-	-	-	0.0	0.0	-	-
H_2S ppm	147	16	2	1	1	1	<1	<1
COS ppm	22	2	1	<0.5	<0.5	<0.5	<0.5	
S_2 ppm	2	1	-	-	-	-		<1
Total sulfur inpurities in product gas, ppm	171	19	3	<2	<2	<2	<2	<2
Product gas								
- amount, Kg-mole	7.47	7.63	7.47	7.64	7.43	7.61	6.94	8.11
Kg/Kg coal	1.67	1.68	1.67	1.69	1.66	1.67	1.66	1.83
SCF/lb coal	28.79	29.40	28.30	29.44	28.65	29.34	26.75	31.26
Gross heating value								
Kcal/Kg-mol	67396	66418	67374	66188	67746	66668	72298	61100
BTU/SCF	314.7	310.1	314.6	309.0	316.3	311.3	337.6	285.3

TABLE 5

CALCULATED COAL GASIFICATION DATA FOR MEDIUM BTU GAS EFFECT OF TEMPERATURE

(pressure used = 1 atm)

COAL, Kg	100		100		100		100	
OXYGEN, Kg	66		66		66		66	
STEAM, Kg	28		28		28		28	
Steam to oxygen weight ratio, Kg/Kg	0.42		0.42		0.42		0.42	
Steam to oxygen mole ratio	0.75		0.75		0.75		0.75	
Carbonate used	Na_2CO_3		K_2CO_3		$Ca\ CO_3$		$FeCO_3$	
Temperature, K	1073	1273	1073	1273	1073	1273	1073	1273
Prod gas composition (wet basis) Mole % or volume %								
CO	55.64	57.01	55.38	57.08	55.36	56.00	53.38	56.12
H_2	32.66	31.62	32.63	31.41	32.62	31.62	32.64	30.05
CH_4	0.32	-	0.30	-	0.30	0.10	0.29	-
CO_2	6.95	5.67	7.37	5.79	7.38	5.67	7.38	7.05
H_2O	4.05	5.64	4.31	5.71	4.34	5.77	4.32	6.78
N_2	-	-	-	-	-	-	-	-
H_2S ppm	3561	533	61	13	6	4	<1	<1
COS ppm	275	42	5	2	<1	<1	<1	<1
S_2	4	2	2	1	-	-	-	-
Total sulfur impurities in product gas, ppm	3840	577	68	16	7	5	<2	<2
Product gas								
- amount, Kg-mole/hr	9.07	9.16	9.11	9.17	9.11	9.18	9.11	9.29
Kg/Kg coal	1.83	1.85	1.84	1.85	1.84	1.85	1.84	1.92
SCF/lb coal	34.97	35.32	35.12	35.36	35.13	35.39	35.12	35.80
- gross heating value								
Kcal/Kg-mole	60617	60163	60378	60073	60362	60159	60357	58485
BTU/SCF	283.0	280.9	281.9	280.5	282.8	280.9	281.8	273.0

TABLE 6

CALCULATED COAL GASIFICATION DATA FOR MEDIUM BTU GAS

EFFECT OF PRESSURE

(Temperature used = 1273°K)

Coal, Kg	100		100		100		100	
Oxygen, Kg	76		76		76		76	
Steam, Kg	-		-		-		-	
Steam to oxygen weight ratio, Kg/Kg	0.0		0.0		0.0		0.0	
Steam to oxygen mole ratio	0.0		0.0		0.0		0.0	
Carbonate used	Na_2CO_3		K_2CO_3		$CaCO_3$		$FeCO_3$	
Pressure (atm)	20	40	20	40	20	40	20	40
Prod Gas composition (wet basis) Mole % or volume %								
CO	74.06	73.77	73.97	73.44	73.90	73.43	73.93	75.17
H_2	19.78	16.36	19.90	16.77	19.88	16.78	19.91	15.78
CH_4	2.29	4.26	2.22	4.02	2.22	4.01	2.20	4.27
CO_2	2.54	3.70	2.64	4.08	2.64	4.09	2.67	3.47
H_2O	1.22	1.47	1.27	1.67	1.37	1.68	1.29	1.31
N_2	-	-	-	-	-	-	-	-
H_2S ppm	1029	3624	28	112	2	9	<1	<1
COS ppm	167	711	4	21	<1	2	<0.5	<0.5
S_2 ppm	-	-	-	-	-	-	-	-
Total sulfur impurities in product gas, ppm	1196	4335	32	133	<3	11	<2	<2
Product gas								
- amount, Kg-mole	7.27	6.99	7.29	7.04	7.29	7.04	7.29	6.93
Kg/Kg coal	1.66	1.66	1.67	1.67	1.67	1.67	1.67	1.65
SCF/lb coal	28.02	26.93	28.09	27.15	28.12	27.16	28.11	26.70
Gross heating value								
Kcal/Kg-mole	68465	70141	68334	68681	68273	69670	68278	70715
BTU/SCF	319.7	327.5	319.1	325.4	318.8	325.3	318.8	330.2

TABLE 7

CALCULATED COAL GASIFICATION DATA FOR MEDIUM BTU GAS

EFFECT OF PRESSURE

(Temperature used = $1273^{\circ}K$)

Coal, Kg	100		100		100		100	
Oxygen, Kg	66		66		66		66	
Steam, Kg	28		28		28		28	
Steam to oxygen weight ration Kg/Kg	0.42		0.42		0.42		0.42	
Steam to oxygen mole ratio	0.75		0.75		0.75		0.75	
Carbonate used	Na_2CO_3		K_2CO_3		$CaCO_3$		$FeCO_3$	
Pressure (atm)	20	40	20	40	20	40	20	40
Prod. Gas composition (wet basis) Mole % or volume %								
CO	56.77	55.89	52.98	52.18	56.41	55.42	52.93	58.34
H_2	29.82	26.46	27.41	25.49	29.68	26.69	27.40	23.93
CH_4	1.38	3.48	0.57	1.76	1.16	2.95	0.57	2.05
CO_2	5.64	6.80	9.83	10.86	6.56	8.01	9.90	9.03
H_2O	5.32	5.78	9.13	9.52	6.19	6.93	9.20	6.65
N_2	-	-	-	-	-	-	-	-
H_2S ppm	9984	14490	744	1716	26	71	<1	<1
COS ppm	825	1323	61	152	2	6	<0.5	<0.5
S_2 ppm	1	1	1	2	1	2	-	-
Total sulfur impurities in product gas, ppm	10810	15814	807	1870	29	79	<2	<2
Product gas								
- amount, Kg-mole	8.85	8.46	9.05	8.84	8.96	8.65	9.06	8.27
Kg/Kg coal	1.82	1.81	1.94	1.94	1.84	1.84	1.95	1.85
SCF/lb coal	34.10	32.62	34.89	34.06	34.5	33.4	34.93	31.90
gross heating value								
- Kcal/Kg mole	61695	63286	55776	56450	60893	61993	55733	60165
BTU/SCF	288.1	295.5	260.4	263.6	284.3	289.4	260.2	280.9

Table 8 - Comparison of calculated product gas composition against experimental data

MODE OF OPERATION	MEDIUM BTU GAS		LOW BTU GAS	
Temperature (°K)	1273	1252	1273	1258
Pressure (atm)	1	1	1	1
Product Gas Composition (volume %)	Calculated	A.I. Data(7)*	Calculated	A.I. Data(10)
CO	57.01	50.20	34.18	28.42
H_2	31.62	31.52	10.98	12.84
CH_4	-	2.66	0.01	1.20
CO_2	5.67	10.19	0.61	0.42
H_2O	5.64	4.76	0.34	1.0
N_2	-	0.60	53.94	56.34
H_2S (ppm)	533	250	15	(not reported)
Heating Value Btu/scf	281	280	142.9	138

* Atomics International Data converted to wet basis

Table 9 - Composition of the alkaline - heavy metal carbonate mixture

CARBONATES	COMPOSITION OF THE MIXTURE			
	For M* = 0.75		For M* = 1.0	
	mole %	weight %	mole %	weight %
Na_2CO_3	42.3	36.0	45.6	39.2
K_2CO_3	33.1	36.9	35.9	40.2
$CaCO_3$	24.6	27.1	18.5	20.6
Na_2CO_3	42.6	37.9	46.1	40.9
K_2CO_3	33.6	38.9	36.3	42.0
$FeCO_3$	23.8	23.2	17.6	17.1

$$^*M = \frac{\text{Amount of carbonate mixture to the gasifier (Kg/hr)}}{\text{Coal feed rates (Kg/hr)}}$$

3. PROPOSED BENCH-SCALE EXPERIMENTAL PROGRAM

On the basis of thermodynamic considerations, it was concluded that if a eutectic mixture of alkaline-heavy-metal carbonate is used then more sulphur would be retained in the melt. In fact, the results indicate that a product gas with sulphur impurities less than 70 ppm may be produced from a high-sulphur content (4.3% S) coal.

This finding must be verified experimentally and other vital information related to gasification rates, yields of product etc., must be obtained for Canadian coals.

3.1 Experimental Apparatus - Concept and Operation

Figure 3 is a flow diagram of a proposed molten salt coal gasification bench-scale apparatus (semi-continuous operation). The unit has been conceived to meet the design criteria as shown in Table 10, and comprises the following main sections:

- coal preparation/feeder system
- carbonate(s) feeder system
- air or oxygen supply
- steam generation and supply
- gasifier/quench system
- product gas separation and analysis
- ash removal/melt separation system

Coal to be used in the experiment will be ground to 200 microns or less in a rotary disc pulverizer and stored. All grinding and storage will be done in an inert atmosphere to minimize oxidation and degradation of coal. Before an experimental run, the desired amount of coal will be loaded in the hopper for pneumatic transport to the gasifier.

Readily available compressed gas cylinders containing air or oxygen are to be used to supply the oxidant for the reaction. a regulated amount of air or oxygen will be preheated (if necessary) and premixed with steam before entering the reactor. A stream of fresh molten salt (eutectic mixture of alkaline-heavy-metal carbonate) will be metered into the gasifier to maintain the desired concentration of carbonates in the reactor.

In the gasifier, coal will be partially oxidized and completely gasified by the reaction with air or oxygen and steam in a turbulent pool of molten salts maintained at the reaction temperature. The salt bath not only catalyzes the reaction, but also supplies part of the heat required for endothermic reaction. Additional heat (if required) will be supplied by an electrical heater surrounding the gasifier.

Ash and sulphur present in the coal will be retained in the melt, and a non-polluting gaseous product formed. Entrained coal particles and carbonate fumes will be removed by cooling the gas to 800°C (1073°K) in a water-cooled baffle-type solid separator. Traces of heavy-oil/tar present in the gas stream will then be condensed and collected. The remaining product gas essentially comprising CO, H_2, CH_4, CO_2, H_2O, and N_2 will be analyzed using an on-line gas chromatograph.

The carbonate melt containing ash , sulphides, and unconverted char will be continuously removed from the gasifier by an overflow system into the quench tank. At the end of an experimental run (typically 2 hours of operation) water will be added to the quench tank to form an aqueous slurry. The aqueous slurry will then be partially carbonated to aid in the precipitation and removal of ash by conventional settling and filtration. The clarified liquor will be analyzed and discarded. However, in a commercial plant, the liquor would be further treated in a regeneration unit and the carbonates recycled to the gasifier.

Table 10 - Design criteria for the molten salt bench-scale apparatus

---

- Maximum duration of run: 4 hours
- Maximum feed rate
  - coal: 2 kg/hr
  - carbonate(s): 2 kg/hr
  - air: 10 kg/hr
  - oxygen: 8 kg/hr
  - steam: 5 kg/hr

- Design temperatures
  - coal from hopper: ambient = 20°C
  - air/oxygen for transport: ambient = 20°C
  - carbonate(s) from melter: 500°C
  - preheated air/oxygen: 400°C
  - steam: 400°C
  - gasifier vessel: 1100°C

- Maximum design pressure
  - flow system: 40 atm (600 psi)

- Environment
  - corrosive acid gas and melt

- Coal and carbonate hopper capacities; air/oxygen supply; quench tank capacity; solid separator and filter capacities:
  - sufficient for one run

- Safety
  - helium and freon system for pre-pressurization and leak detection

---

Figure 1.

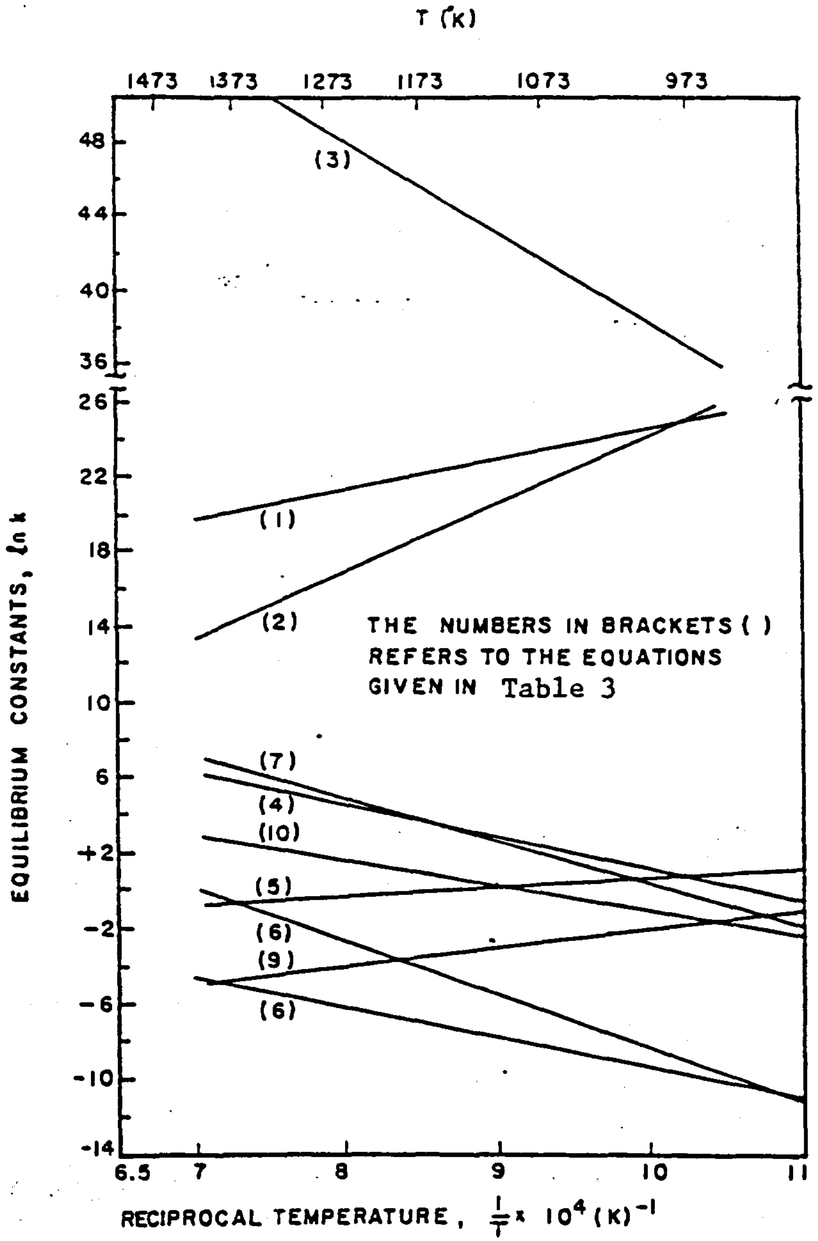

FIG. 1 - MOLTEN SALT COAL GASIFICATION EQUILIBRIUM CONSTANTS

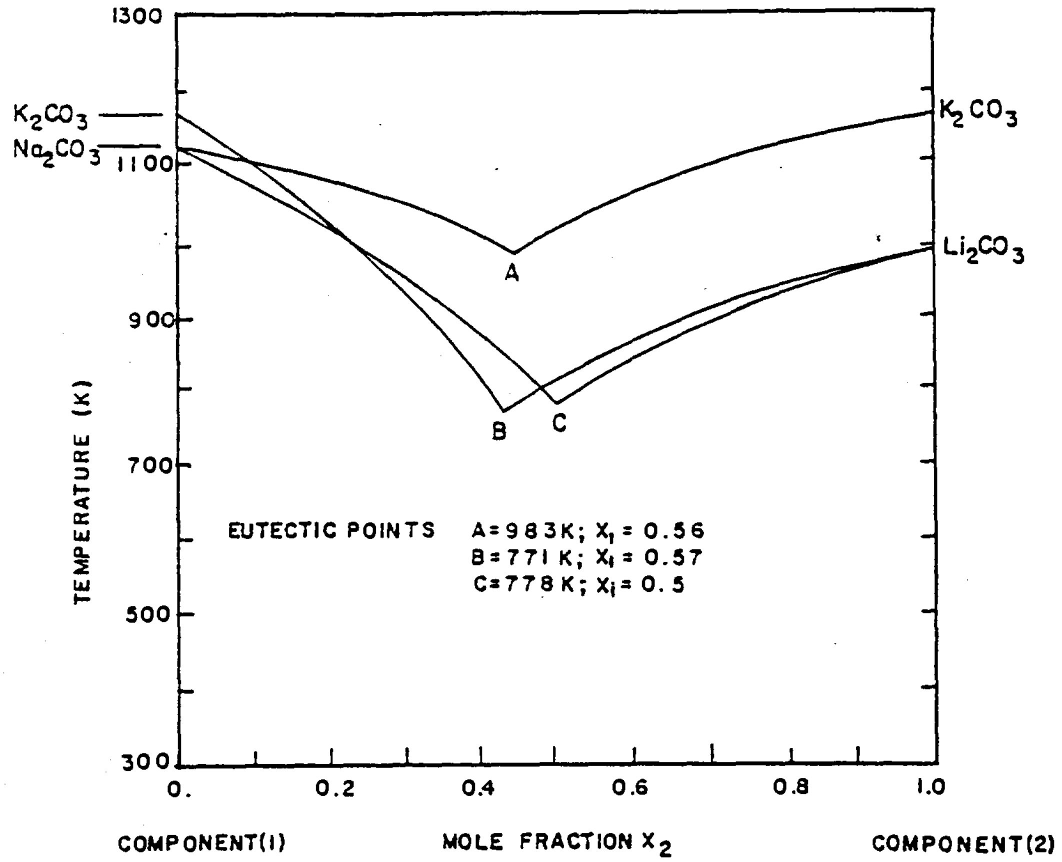

FIG. 2 PHASE DIAGRAM OF Na_2CO_3-K_2CO_3, K_2CO_3-Li_2CO_3, AND Na_2CO_3-Li_2CO_3 SYSTEM

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DEVELOPMENT OF A BENCH SCALE FLUIDIZED BED COAL GASIFIER AND GAS BURNER SYSTEM

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1.0 INTRODUCTION

The use of fluidized bed reactors to bring a fluid and solid into contact with good mixing promote reaction has been developing and expanding in scope during the past forty years, although the phenomena was first observed some seventy years ago. the concepts upon which the facility described in this paper were based are, therefore, not new or unique. They were applied for the purpose of studying reactions with Canadian coals for future needs in this country.

The facility that was designed and built was intended to be used with a variety of Canadian coals, ranging from lignite to bituminous. The intent was to gasify the coal in a fluidized bed, clean the gas of fly ash and subsequently burn the product gases in a gas-turbine type combustor. In this paper, the design considerations are described; in the sequel, some results of operating the facility are presented.

2.0 FLUIDIZED BED REACTOR DESIGN

Fluid-particulate technology enjoys wide applications in many industries under various processing conditions and will be applied more extensively with the evolution in energy generation and conversion. Fluidized bed gasifiers can, in principle, handle coals which have a wide range of size distributions and have the added advantages of temperature uniformity, high heat transfer rates, and continuous operation, coupled with the possibility of removal of sulphur at the gasification stage to minimize emissions of sulphur dioxide.

The design of a fluidized bed coal gasifier is complicated by the requirement of operation at high temperatures, with particles tending to stick and agglomerate, with feed streams having wide distributions of particle size and composition and by fines generation in situ.

In this paper, the design of a simple laboratory scale, fluidized bed gasifier is described. Some of the experimental results which are found in the literature have been employed in this design.

2.1 Design Parameters and Fundamentals

The design of a fluidized bed coal gasifier requires specification of a number of critical parameters. These parameters must be selected with the aid of available data.

2.1.1. Bed material

For the purpose of the design, one coal was chosen. This was a Canadian sub-bituminous coal which has a calorific value between 8,300 and 10,000 Btu per pound, (4.600 - 5,550 cal/gm), does not agglomerate and has a specific gravity of 1.29 on average. Any non-agglomerating coal should be useable; agglomerating coals will be studied but may cause problems.

(a) Particles mean diameter ($\bar{d}_p$)

For best fluidization results, to minimize the required fluidication velocity, the average particle size should be small. It cannot be too small or close packing of particles due to electrostatic and other attractive forces will result in flow channelling. The agglomerating tendency of powdered coal (d<100 μm) increases chances of defluidization and entrainment of the bed material. Using coarse particles requires high fluidizing velocities, and results in rough operating and correspondingly less reaction surface per unit volume. For these reasons, the particles have been selected to have diameters between 100 to 300 μm for the reactor being designed.

For a sample selected, the particles mean diameter was found to be 190 μm.

(b) Sphericity of the particle (ϕ_s)

The sphericity ϕ_s of the particle is defined as:

$$\phi_s \left(\frac{\text{surface of sphere}}{\text{surface of particle}}\right), \text{ both of same volume.}$$

For nonspherical particles, $0 < \phi_s < 1$ and for sphere $\phi_s = 1$.

From the data collected from different references:

Material	Sphericity (ϕ_s)	Ref
Bituminous coal	.625	1
Pulverized coal	.695	2 (4)
Anthracite coal	.63	3

Since the sphericity varies by less than 1% as given here, for various coals, the sphericity of our particular coal was taken to be 0.625.

(c) Void fraction of the bed (ε_m)

The void fraction in the normal packed bed is the ratio of unoccupied volume among the particles to the total volume of the bed. The void fraction in a packed bed depends on particle size, shape of the size distribution curve and sphericity of the particles. Also, for vessels of small diameter, the effect of the vessel's wall becomes important.

Reliable voidage predictions are scarce and there is no general equation with which to predict voidage. It is suggested that ε_m be found experimentally (4). The void fraction of our particular bed was found experimentally, to be: $\varepsilon_m = 0.486$.

(d) Voidage at minimum fluidizing conditions (ε_{mf})

Void fraction at minimum fluidization (ε_{mf}) is the bed void fraction at onset fluidization. The value of ε_{mf}) can be measured experimentally or estimated from random packing data.

According to data given in reference (5), the void frction of anthracite coal ($\phi_s = 0.63$) at minimum fluidizing conditions is 0.6 at $d_p = 100$ µm, 0.56 at 200 µm and 0.53 at 300 µm. Since our particular coal has been assumed to have approximately the same sphericity ϕ_s, as anthracite we can assume that $\varepsilon_{mf} = 0.56$ at $d_p = 190\ \mu m$ without significant expected error.

2.1.2. Average bed temperature

The gasification reactor should be maintained at as high a temperature as possible in order to achieve the highest reaction rates, but temperatures are avoided that promote excessive agglomeration of fluid bed particles due to softening, becoming sticky, and thereby agglomerating with others as a result. Such temperatures vary depending on composition of coal particles and the ash material, but are expected to be approximately 1100°C (∿2000°F) and higher.

Also, higher temperatures lead to produce gas oxidation as well as higher heat losses. Another disadvantage of using higher temperatures is that the methane yield may be lower at high temperatures than at somewhat lower operating temperatures.

If temperatures are below about 760°C, carbon conversion decreases markedly as a result of low reaction rates. Forney et al[6] concluded that the optimum gasification temperature is between 900 and 950°C. Matthews (7) found the best gasification temperature to be in the range from 760°C to 1093°C (2000°F) and typically may be about 927°C (1700°F). In order to maximize the production of methane in the gasifier the preferred operating temperature should be about 900°C (8).

After review of literature and experiments which had been done on this topic of gasification, the initial choice of the gasifier design temperature as equal to 900°C is considered to be a reasonable selection.

2.1.3. Gasifier pressure

The gasification of coal at superatmospheric pressure is desirable for these reasons:

(a) As the pressure increases the methane content of products gradually increases (6,9).

(b) The gas mass flow through a vessel's internal crossectional area for a given temperature and velocity, increases with increased pressure.

(c) Heat loss per unit mass of gas under pressure operation is less.

But gasification under multi-atmospheric pressure has these disadvantages:

(a) As the pressure increases the hydrogen and carbon monoxide (H_2 + CO) percentage decreases.

(b) Operation at superatmospheric pressure requires several additional safety considerations.

Because of the safety considerations and because the available steam is at low pressure, atmospheric pressure was selected as the initial design and initial operation pressure. But this reactor can be modified to work under superatmospheric pressure. Most of these modifications have now been incorporated into the system.

2.1.4. Reactant ratios

In order to determine the bed area and heat requirements for a given coal input, it is required to carry out detailed mass and heat balances on the vessel using the reactor model (5). Simulation of a fluidized bed gasifier requires using a reaction kinetic model combined with a reactor flow model (10). Although it is the best model applied to date, the suggested reaction model (unreacted-core shrinking model) does not fit the experimental data exactly (10) possibly due to different reactivity and composition from one kind of coal to another, different particle residence times and because the kinetics of the simultaneous gasification reactions are not well understood.

Some experiments have been done successfully by various investigators. One of these experiments was done Forney (11), using a reactor similar to that of our requirements. In his experiments the average reactant ratios were:

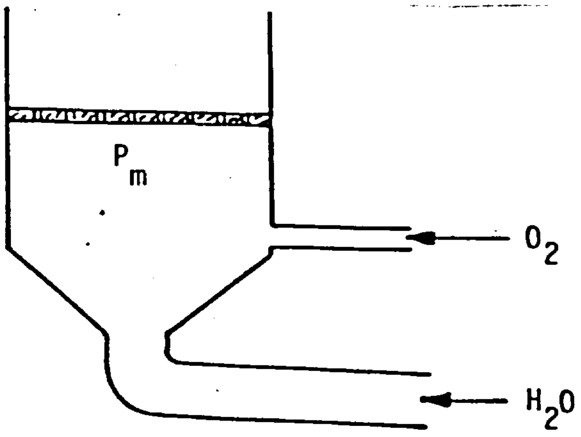
(a) Oxygen to coal ratio was $\sim$0.45 std. m^3/kg coal.
(7 std. ft^3/lb coal)

(b) Steam to coal ratio was $\sim$2.5 std. m^3/kg coal.
(40 std. ft^3/lb coal)

These ratios were taken for the initial design, rather than relying on imprecise and lengthy calculation of heat and mass balances.

2.1.5. Fluidizing gas stream

The fluidizing stream is a gas mixture of oxygen and steam. This mixture is introduced to the bottom of the gasifier with a pressure just enough to overcome the flow resistance inside the vessels. For design purposes, it was assumed that the pressure of the mixture, just before the perforated plate, was 15.7 psia (as an initial estimate based on some calculations). (1.07 atm).



(a) Steam properties

The steam used is superheated steam at a temperature of 150°C. Since the steam velocity in the steam tube after the metering valve will be very low, we can assume that the steam back pressure is equal to the mixture pressure. At these condiditons of pressure and temperature, the enthalpy and density of the steam are:

$$h = 664.7 \text{ Kcal/kg}$$
$$p = 0.544 \text{ kg/m}^3$$

(b) Oxygen properties

The oxygen temperature is equal to the room temperature = 25°C. The oxygen pressure in the oxygen tube should be controlled to equal the mixture pressure = 15.7 psia. (1.07 atm).

(c) Properties of the fluidizing stream

Since the steam in the mixture is still superheated steam at low pressure, we can consider it as an ideal gas, approximately. Then, at these conditions, the volume flowrate of the steam in the fluidizing stream is obtained from the following:

Under standard conditions,

V_{steam} = 2.5 standard m^3/kg coal

Also at these conditions, the volume flowrate of the oxygen in the fluidizing stream is:

V_{O_2} = 0.45 standard m^3/kg coal

According to Amagat's Law (law of additive volume), the volumetric stream flowrate in terms of standard conditions is

$$V_s = (V_{O_2})_s + (V_{steam})_s = 2.95 \text{ standard } m^3/\text{kg coal}$$

The corresponding actual flow rate, assuming a perfect gas mixture is

$$V_{actual} = V_s \left(\frac{Ps}{Pa} \cdot \frac{Ta}{Ts}\right)$$

$$= 4.07 \ m^3/\text{kg coal (at the mixed conditions)}$$

(iii) The density of the fluidizing stream

The mixture density is

$$\rho_m = \frac{M_{(O_2+ H_2O)}}{V_m}$$

$$= 0.59 \ kg/m^3$$

where, M is the mass flowrate of (oxygen + steam)/kg of coal.

Mixtures of nonpolar gases and steam, which have a high ratio of water vapour, have a viscosity close to that of water vapour (12). Also, it is concluded in the same reference, that there is no significant variation in the viscosity with pressure up to 1550 psia. Therefore the viscosity coefficient of the fluidizing stream can be taken equal to the viscosity coefficient of the steam at the same conditions of pressure and temperature, which is

μ = 1.14 x 10^{-4} poise

or it can be taken equal to the viscosity of a mixture of steam and nitrogen at the same conditions, (12) which is equal to

μ (steam + nitrogen) = 1.6 x 10^{-4} poise

The second value is considered to be more accurate and is taken as the viscosity of the mixture to be used in this work.

μm = 1.6 x 10^{-4} poise

If temperature rise and pressure drop through the perforated plate are neglected, the properties of the fluidizing stream just above the plate will remain the same. (The change due to pressure drop would be small).

2.1.6. Superficial velocity

The superficial velocity of the fluidizing gas should be somewhere between the minimum fluidizing velocity and the terminal velocity of the bed particles.

A. Minimum fluidizing velocity (u_{mf})

The minimum fluidizing velocity is the fluid velocity at which bed particles are all just suspended in the upward flowing gas. At this point, the drag force by upward moving gas equals weight of the particles and the bed is called an "incipiently fluidized" bed or a bed at minimum fluidization.

In calculating the minimum fluidizing velocity, two points must be considered:

(a) The minimum velocity occurs at the bottom of the bed.

(b) The mean diameter ($\bar{d}_p$) for the size distribution actually present in the bed must be used.

There are many proposed equations by different investigators to calculate minimum fluidizing velocity. Unfortunately, there is a wide variation in the value of u_{mf} obtained by these equations. Some particular cases are considered.

(i) If voidage at minimum fluidization and sphericity of particles (ϵ_{mf}, ϕ_s) are not known:

A generalized correlation of minimum fluidizing velocity based on a comprehensive study by Wen and Yu (13) is given as

$$\frac{\bar{d}_p \; u_{mf} \, \rho g}{\mu} = \left[(33.7)^2 + .0408 \, \frac{\bar{d}_p^3 \, \rho_g \, (\rho_s - \rho_g) g}{\mu^2} \right]^{\frac{1}{2}} - 33.7 \qquad (1)$$

where,

ρ_g = density of the fluidizing gas stream in gram/cm^3

(6.5×10^{-4} gm/cm^3 assumed)

μ = viscosity of the fluidizing gas in poise (gram/cm.sec) (1.6×10^{-4} poise assumed)

ρ_s = solid (coal) density in gram/cm^3 (1.29 gm/cm^3 assumed)

$\bar{d}_p$ = mean particles diameter in cm (1.90×10^{-2} cm assumed)

u_{mf} = minimum fluidizing velocity in cm/sec.

By substituting in equation (1) with the values of ρ_g, μ, ρ_s and $\bar{d}_p$, which are previously calculated, a value of $u_{mf} = 1.72$ cm/sec is obtained.

Another empirical correlation proposed by Davidson and Harrison (14) is reduced to

$$u_{mf} = 0.00114\, gd^2 \,\frac{(\rho_s - \rho_g)}{\mu} \qquad (2)$$

From this equation, using the same values for the ρ_g, μ, ρ_s and $\bar{d}_p$ as used in eqn 1, a value of $u_{mf} = 3.25$ cm/sec is obtained.

Other correlations allowing for similar property dependence but with slightly different fixed constants are used by others. The minimum fluidizing velocity is expected, therefore, to fall somewhere between about 1.5 and 4 cm/sec for the conditions cited.

ii) If voidage at minimum fluidization and sphericity of particles (ε_{mf}, ϕ_s) are known:

Kunii and Levenspiel (4) proposed the following equation:

- For small particles of small specific weight:

$$u_{mf} = \frac{(\phi_s \bar{d}_p)^2}{150}\,\frac{\rho_s - \rho_g}{\mu}\,g\left(\frac{\varepsilon^3_{mf}}{1-\varepsilon_{mf}}\right), \quad Re_p < 20 \qquad (3)$$

- For large particles:

$$u_{mf}^2 = \frac{\phi_s \; d_p}{1.75} \quad \frac{\rho_s - \rho_g}{\rho_g} \qquad g \cdot \epsilon_{mf}^3 , \quad Re_p > 1000 \qquad (4)$$

In our case, assuming $Re_p < 20$, and substituting into equation (3) yields

$$u_{mf} = 2.95 \text{ cm/sec.}$$

$$Re_p = \frac{u_{mf} \; \rho_g \; \bar{d}_p}{\mu} = 0.24 < 20$$

The assumption that Re_p > is correct.

This equation should be used if information on ϵ_{mf} and ϕ_s (4) is available, because it gives more reliable predictions of u_{mf}.

B. <u>Terminal velocity</u> (u_t)

The terminal velocity of particles is the maximum allowable velocity which the superficial velocity should not exceed to avoid carry over of solids from a bed.

In calculating the terminal velocity of particles, the following must be considered.

(a) The terminal velocity always occurs at the top of the bed for a bed of uniform voidage.

(b) In calculating (u_t) the smallest size of solid particles must be used, which is 0.01 cm. diameter in our case.

(c) The properties of the fluidizing gas should be calculated at the top of the bed (at T = 900°C).

Here,

$\rho = 2.3 \times 10^{-4}$ gram /cm^3

$\mu = 4.2 \times 10^{-4}$ poise

The terminal velocity of the particles can be estimated by using this equation (4):

$$u_t = {\frac{4 g\, d_p (\rho_s - \rho_g)}{3 \rho_g C_d}}^{1/2} \qquad (5)$$

where, C_d is an experimentally determined drag coefficient. Since the experimental value was not available, the analytic expression for the drag coefficient of spherical particles was used, which for various ranges of Reynolds numbers is:

$$C_d = \frac{24}{Re_p} \qquad \text{for} \quad Re_p < 0.4$$

$$C_d = \frac{10}{(Re_p)^{1/2}} \qquad \text{for} \quad .4 < Re_p < 500$$

Replacing these values of C_d in equation (5) yields

$$u_t = \frac{g\,(\rho_s - \rho_g)\, d_p^2}{18\mu} \qquad \text{for } Re_p < .4 \qquad (6)$$

$$u_t = \frac{40g}{225} \; \frac{2(\rho_s - \rho_g)^2}{\rho_g \mu} \; d_p \qquad \text{for } .4 < Re_p < 500 \qquad (7)$$

In our case, assume $Re_p < .4$ and substituting in equation (6) with the values of ρ_s, μ, ρ_g and d_p used previously to obtain u_{mf} except taking $d_p = 100$ µm, yields for the terminal velocity;

$$u_t = 16.7 \quad \text{cm/sec}$$

$Re_p = .09 < .4$, so the assumption is correct and the terminal velocity of the particles is 16.7 cm/sec.

C. The superficial velocity (u)

The superficial gas velocity for fluidized bed operation is limited on one by u_{mf} and on the other by entrainment of solids by the gas. Therefore, the gas velocity should be kept somewhere between u_{mf} and u_t.

In choosing the gas velocity, these two points must be considered:

a) In order to assure good solid mixing and good heat transfer, the gas velocities corresponding to (u/u_{mf}) of at least 2 should be selected (10).

b) To avoid channelling and slugging (u/u_{mf}) should not exceed a value of 4. (15)

After reviewing these considerations, the superficial gas velocity ratio is selected to be $u/u_{mf} \approx 2.4$. Therefore,

$$u = 7 \text{ cm/sec/}$$

2.2 Design of the bed

2.2.1. The bed diameter (D)

At a constant coal feed rate of 250 gram/hr, the required volumetric flow rate of the mixture (at the bottom of the gasifier)

$$= 0.25 \frac{\text{kg}}{\text{hr}} \times 4.07 \frac{\text{m}^3}{\text{kg}}$$

$$= 1.02 \text{ m}^3/\text{hr}$$

$$= \frac{\pi}{4} D^2 u$$

Therefore, the bed diameter (D) $\approx$ 7 cm.

2.2.2. The bed height (L_m)

The height of the fixed bed is an important parameter in the operation. Smooth operation at usual velocities can be obtained in shallow beds with L_m/D of about one and a maximum L_m/D of two to three (15).

Slugging starts at about a L_m/D of 2.0 (15). Therefore for smooth operation and to avoid slugging conditions, the suggested L_m/D ratio is taken 1.5. Therefore,

$$L_m = 10.5 \text{ cm.}$$

2.2.3. The bed weight (W)

Bed weight is the weight of the bed material which should be maintained at almost fixed value during operation. It can be calculated from this equation (4):

$$W = \frac{\pi}{4} D^2 L_m (1 - \varepsilon_m)\rho_s \qquad (8)$$

From the previous calculation: $L_m = 10.5$ cm

$\varepsilon_m = .486$

$\rho_s = 1.29$ gram/cm^3

By substituting in equation (8), the bed weight:

$W = 268$ gram

2.2.4. The bed height at minimum fluidization (L_{mf})

Bed height at minimum fluidizing conditions is the bed height at onset of fluidization. It is a useful parameter in calculating the pressure drop across the bed. If the gas density is neglected in comparison with the solid density, the value of L_{mf} may be calculated by using this equation:

$$L_{mf} = \left(\frac{1 - \varepsilon_m}{1 - \varepsilon_{mf}}\right) L_m \qquad (9)$$

Substituting in this equation with ε_m (0.48), ε_{mf} (0.56)$^{(3)}$ and L_m, (10.5) gives:

$L_{mf} = 12.2$ cm.

2.2.5. Pressure drop of the bed

When visual observation is not possible, as in this situation, the pressure drop across the bed is useful as a rough indication of the quality of fluidization.

For relatively low flow rates in a packed bed the pressure drop is proportional to gas velocity until ΔP becomes slightly higher than the static pressure of the bed. With a further increase in gas velocity, the voidage increases from ε_m to ε_{mf} resulting in a decrease in pressure drop to the static pressure drop of the bed, then the bed is called an incipiently fluidized bed. With gas velocity beyond minimum fluidization, the pressure drop essentially remains unchanged, Fig. (1).

Pressure drop in the fluidized bed is given by equation (10) at the minimum fluidization condition:[4]

$$\Delta P = L_{mf}\ (1 - \varepsilon_{mf})\ (\rho_s - \rho_g)\ \frac{g}{g_c} \qquad (10)$$

$$= 12.2\ (1 - .56)\ \ (1.29 - 2.3 \times 10^{-4})\ \frac{980}{980}$$

$$= 6.9\ \ \text{gram.wt/cm}^2\ \ (\text{cm } H_2O)$$

2.2.6. Transport disengaging height, TDH

The transport disengaging height is the height of exit above the top of the bed where entrainment becomes approximately constant.

There is a wide disagreement between investigators in calculating the TDH because of the wide disagreement between them in prediction of the expanded bed height.

According to reference (10), the smaller overall reactor height may be determined by applying a multiplication factor of 2.6 to the calculated expanded bed height.

From reference (4) $L_f/L_{mf} \simeq 1.5$ where, L_f is the expanded bed height. Therefore, the minimum overall reactor height should be

$$= 12.2 \times 1.5 \times 2.6$$

$$= 47.5 \text{ cm.}$$

Another approach is to add the expected transport disengaging height to the bed height at the fluidization condition. On the basis of graphs shown in reference (4) the TDH should be about seven times the reactor diameter at velocities of the order of 31 cm/sec for a 7 cm dia. vessel. Since there is no curve provided for lower velocities, if we choose this value, it corresponds to 49 cm overall for the TDH. Since our velocity will be less than 10 cm/sec. a TDH of five times the diameter seems justified. Therefore, the 47.5 cm calculated above seems appropriate.

2.3. Design of the gas distributor

The quality of fluidization in a fluidized bed is influenced by the type of gas distributor. Therefore, the choice of distributor type and its design are important in the successful application of fluidized bed techniques. Many designs of gas distributors have been proposed, as discussed in the literature (4). Both ceramic and metallic materials have been used in this application. Porous ceramic distributors give better fluidization quality, less fluctuation in density and less channelling but they have little mechanical strength against thermal shock, are very easily clogged by dust and cause a very large pressure drop in the gas flow. Perforated metallic distributors have higher strength and are able to resist corrosive atmospheres and high bed temperatures.

In the design of a gas distributor, there are some requirements that must be met concerning the apertures. Their number, size and shape must be such that the leakage of solids through the distributor is prevented, the particle-attraction is minimized and the fluidization is uniform and stable.

To achieve uniform and stable fluidization, the distributor should have a sufficient pressure drop to maintain equal flow through the openings. Agarwal (16) recommended that the pressure drop across the distributor plate should be roughly 10% of the pressure drop across the bed with a minimum in all cases of about 35 cm H_2O.

In the design of the required gas distributor, the following procedure was used:

A. Pressure drop:

According to Agarwal's recommendation (16), since the pressure drop in the bed was estimated to be 6.9 cm H_2O, the minimum required pressure drop should be found as the following:

$$10\%\ \Delta P_{bed} = 0.69 \text{ cm } H_2O$$

Since $10\%\ \Delta P_{bed} < 35$ cm H_2O, therefore, according to the minimum set by Agarwal, ΔP_d should be selected to be equal to 35 cm H_2O, at least. Therefore, the minimum recommended pressure drop through the distributor is

$$\Delta P_d = 35 \text{ cm water}$$

B. Reynolds number:

Reynolds number of the flow approaching the plate is:

$$Re = \frac{D\ p_g\ u}{\mu_g}$$

$$= \frac{7 \times 6.77 \times 10^{-4} \times 7}{1.6 \times 10^{-4}}$$

$$= 207$$

C. The orifice coefficient:

From Fig. 2 at Re = 207, the orifice coefficient is

$$C'_d = 0.7$$

D. Gas velocity through the orifices:

By definition of the orifice coefficient shown in Fig. 2 the gas velocity is given by:

$$u_{or} = C'_d \left(\frac{2g_c\ \Delta P_d}{\rho_g}\right)^{1/2} \qquad (11)$$

$$= 7046 \text{ cm/sec.}$$

for the conditions specified.

In this case, the dynamic force of the orifice jets is very high in comparison with the bed resistance. So, the jet will punch right through the bed causing severe gas by-passing.

Kunii and Levenspiel (4) used an alternate approach to a distributor design to avoid the above mentioned problem. Based on experience, they found that if the kinetic energy of the orifice jet is equal to 0.5 to 0.75 of the resistance of the bed, satisfactory operation results. This interprets to

$$u_{or} \simeq 0.7 \left(\frac{2g_c\ \Delta P_{bed}}{\rho_g}\right)^{1/2}$$

$$= 3128 \text{ cm/sec}$$

That is, the total pressure of the flow is less than 0.8 of the hydraulic head of the bed.

E. Fraction open area

The fraction open area is a simple ratio of the fluidization velocity to the velocity in the orifice, or

$$\text{Fraction open area} = \frac{u}{u_{or}} = \frac{7}{3128} = .22\%$$

F. The relation between number and size of the orifices:

Since the total of all the flow through the orifices must equal flow in the fluidizing stream, the number-size relationship is expressed as follows

$$N_{or} = 4u/(\pi d_{or}^2 u_{or}) \qquad (12)$$

where, N_{or} = number of orifices per unit area (number/cm^2)

d_{or} = orifice dia. cm

u_{or} = gas velocity through the orifice (cm/sec)

By using this equation, we can have these combinations between N_{or} and d_{or}:

d_{or} (cm)	.02	.05	.01
N_{or} (number/cm^2)	7.1	1.14	.28

G. Decision on number of orifices:

Orifices that are too small are likely to become clogged, whereas those that are too large may cause an uneven distribution of gas.

Select: d_{or} = .05 cm

N_{or} = 1.14 number/cm^2

$$\text{The total number of orifices} = \frac{\pi}{4} D^2 \times N_{or}$$

$$\simeq 44$$

These orifices have been distributed approximately uniformly on the plate as indicated in Fig. 3. The plate thickness is 1/8".

Exit Design

Using the conservation of species principle and equilibrium relations, the calculation of products made for an exit temperature of 1200°K predicted 1.04 stdm^3/hr. Therefore, the actual volumetric flow is

$$\frac{1200°K}{273°K} \times 1.04 = 4.57\ m^3/hr$$

If an exit diameter of 5 cm is chosen, the exit velocity will be

$$U_{exit} = 4.57\ \frac{m^3}{hr}\ \frac{hr}{3600\ sec}\ \frac{4}{\pi(.05)\ m^{2\ 2}} = 0.65\ \frac{m}{sec}$$

$$= 65\ cm\ sec$$

Steam and oxygen inlet design

The steam flow rate was chosen to be 1.83 kg steam per kg coal. At a col feed rate of 0.25 kg/hr, this corresponds to 0.4575 kg steam/hr. The density of the steam should be approximately 0.5 kg/m^3 at the inlet condition, so if an inlet diameter of 4 cm is chosen, the steam volocity would be about 20 cm/sec.

The oxygen flow rate selected to be 0.58 kg/kg coal, or 0.145 kg O_2 per hour at a coal feed rate 0.25 kg/hr. At 300°K, 15.7 psia, the density of oxygen should be 1.388 kg/m^3. Therefore, for an inlet diameter of 1.5 cm, the inlet would be about 16.5 cm/sec which is acceptable.

General gasifier dimensions:

In summary, the dimensions chosen for the laboratory-scale fluidized-bed gasifier are as follows: Fig. 4

D_1 = 7 cm
D_2 = 14 cm
H_1 = 70 cm
H_2 = 28 cm
L = 7 cm
D_3 = 5 cm

$D_4 = 4$ cm
$D_5 = 1.5$ cm
$D_6 = 4$ cm

3.0. GASIFIER AUXILIARIES

3.1. Cyclone Ash Separator

High operational efficiency, simple construction, and low cost make the cyclone the most extensively used type of collector for removing solids from a gas stream. Cyclones have been operated at temperatures as high as 1000°C and pressures as high as 500 atm. Cyclones for removing solids from gases are generally applicable when particles of over 5 µm diameter are involved. In special cases where dust, which shows a high degree of agglomeration, is involved, like in the case of coal ash, cyclones will remove dusts having a much smaller particle size than 5 µm (.1 to 2 µm).

Fig. 5 gives dimensions of the cyclone built for this application. The fly ash-laden gas enters the upper, cylindrical, portion of the cyclone chamber through a tangential inlet duct. The flow spirals downward toward the axis with increasing spin velocity and is discharged through the axial outlet duct. Suspended ash particles are thrown outward by the centrifugal force arising from the rotation, and find their way down into the ash hopper at the lower end of the cone.

Cyclone design

The principal design parameters of the cyclone are illustrated in Fig. 5. The recommended general cyclone proportions are (16):

$B_C = D_c/4$
$D_e = D_c/2$
$H_c = D_c/2$
$L_c = 2D_c$
$S_c = D_c/8$
$Z_c = 2D_c$
J_c arbitrary dimension, usually $= D_c/4$

The gas flow rate

From previous calculations, the product gas flow rate is 4.16 std. m^3/kg coal. For a coal feed rate of 0.25 kg/hr, the total gas yield is expected to be 1.04 std. m^3/hr.

At gasifier exit conditions ($t = 1200°K$, $p \simeq 1$ atm.), the volumetric product gas flow rate = 4.57 m^3/hr.

Gas velocity

Although efficiency is normally increased by increasing the gas through-put, in some cases the reverse may be true because of the deflocculating effect of high velocities. The cyclone inlet velocity is usually in the range of 1.5 → 7.5 m/sec. Also, the immediate entrance to the cyclone is recommended to rectangular ($B_c \times H_c$).

From previous proportions, $H_c = 2\ B_c$; select $B_c = .95$ cm (3/8").

H_c = 1.9 cm (3/4")

then, $A_c = 1.82\ cm^2$

and the cyclone inlet gas volocity (u_c) $= \frac{V}{A_c}$

$= 6.97$ m/sec

$\simeq 23$ ft/sec

Pressure Drop

Shepherd and Lapple (16) investigated a cyclone of this general type. For the cyclone friction losses, they obtained the following empirical expression:

$$F_c = \frac{K\ B_c\ H_c}{D_e^2}$$

where, F_c = Cyclone friction loss, expressed as number of cyclone inlet volocity heads, based on area A_c

K = Empirical constant for cyclone pressure drop.

For the specific proportions shown here, $F_c = 8$, and the pressure drop is given by $\Delta P = 4.06 \times 10^{-2}\ \rho\ u_c^2$ cm H_2O

where, u_c = cyclone inlet velocity, m/sec

ρ = gas density in kg/m^3

then the expected pressure drop across our cyclone should be

$\Delta P \simeq 0.4$ cm water

So, this selected entrance dimension of the cyclone gives reasonable cyclone inlet velocity and allowable pressure drop.

In summary, the design specified dimensions are:

B_c = .95 cm, H_c = 1.9 cm, D_e = 1.9 cm, D_c = 3.8 cm, L_c = 7.6 cm, Z_c = 7.6 cm, S_c = .47 cm, J_c = .95.

4.0. DEVELOPMENTAL TYPE COMBUSTOR

In the first phase of this work, the intent was to use a developmental type combustor in conjunction with the coal gasifier to study the burning characteristics of the gas produced. Figure 6 shows schematically the test facility with the combustor and fuel and air lines represented. Figure 7 shows the dimensions of the combustor.

Initially, simulated coal gas mixtures were used to check out the system. Mixtures of CO, CO_2 and H_2 were burned with appropriate amounts of air. The products of combustion were analyzed in the chromatograph.

5.0. SUMMARY

This paper has outlined the design of a bench scale fluidized bed coal gasifier and gas burner system. This system has been built and some results of operation obtained. Preliminary results are described in the sequel to this paper.

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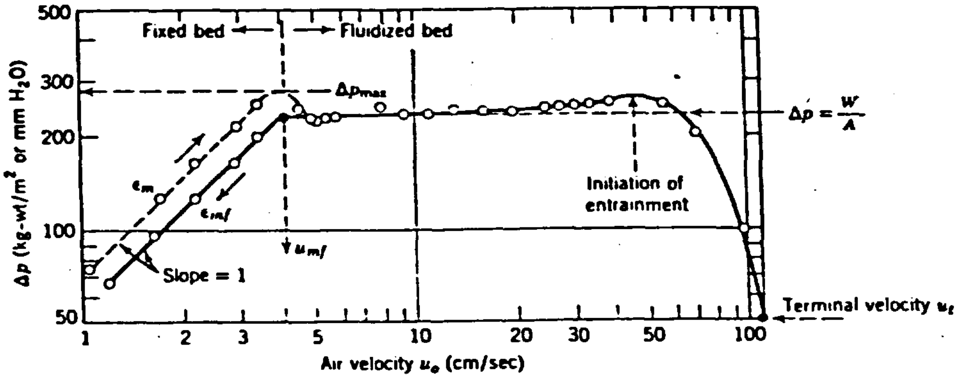
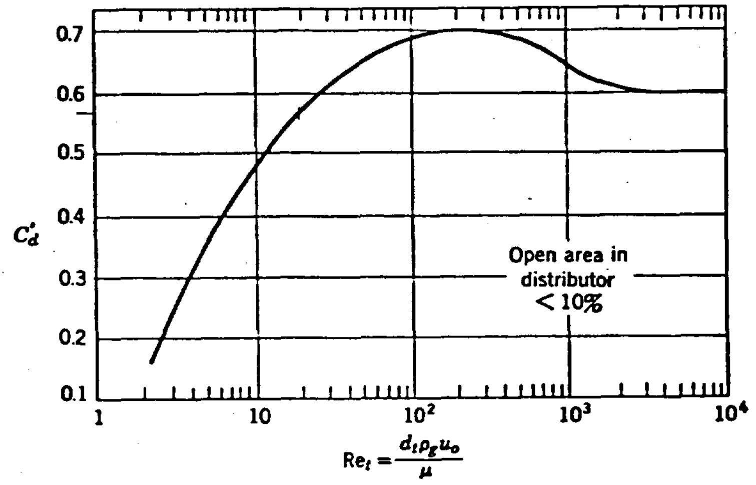


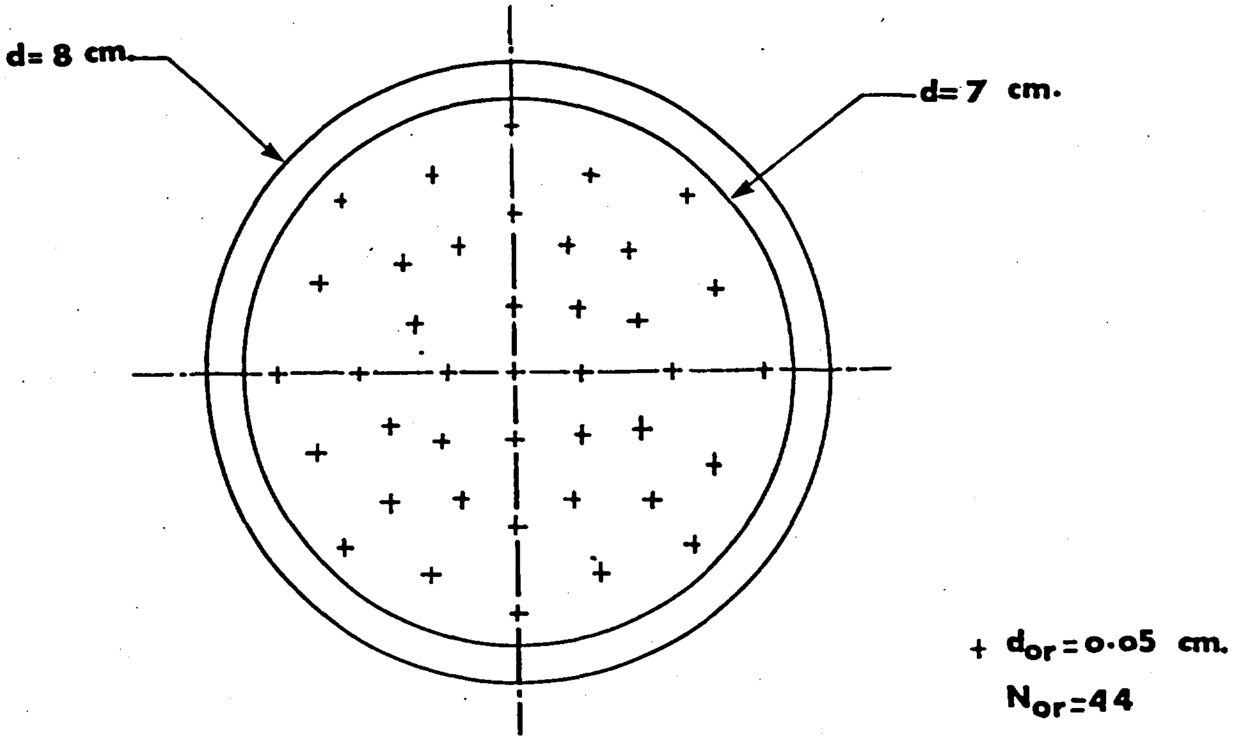
Pressure Drop Versus Gas Velocity for a Bed of Uniformly Sized Particles

Fig. (1) reference (4)



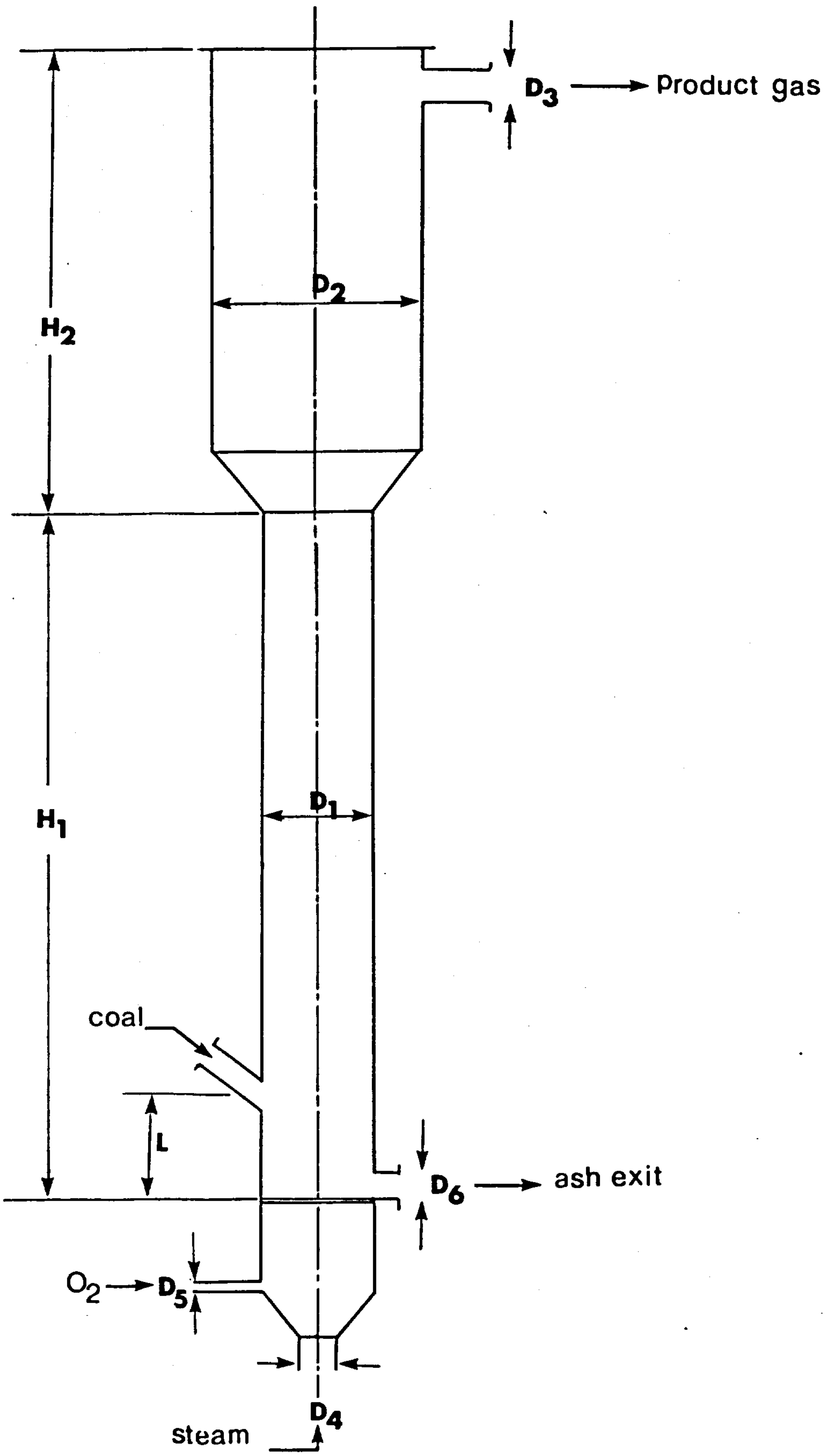
Orifice Coefficient Versus Reynolds Number
Based on Diameter of Approach Chamber

Fig. (2) reference (4)



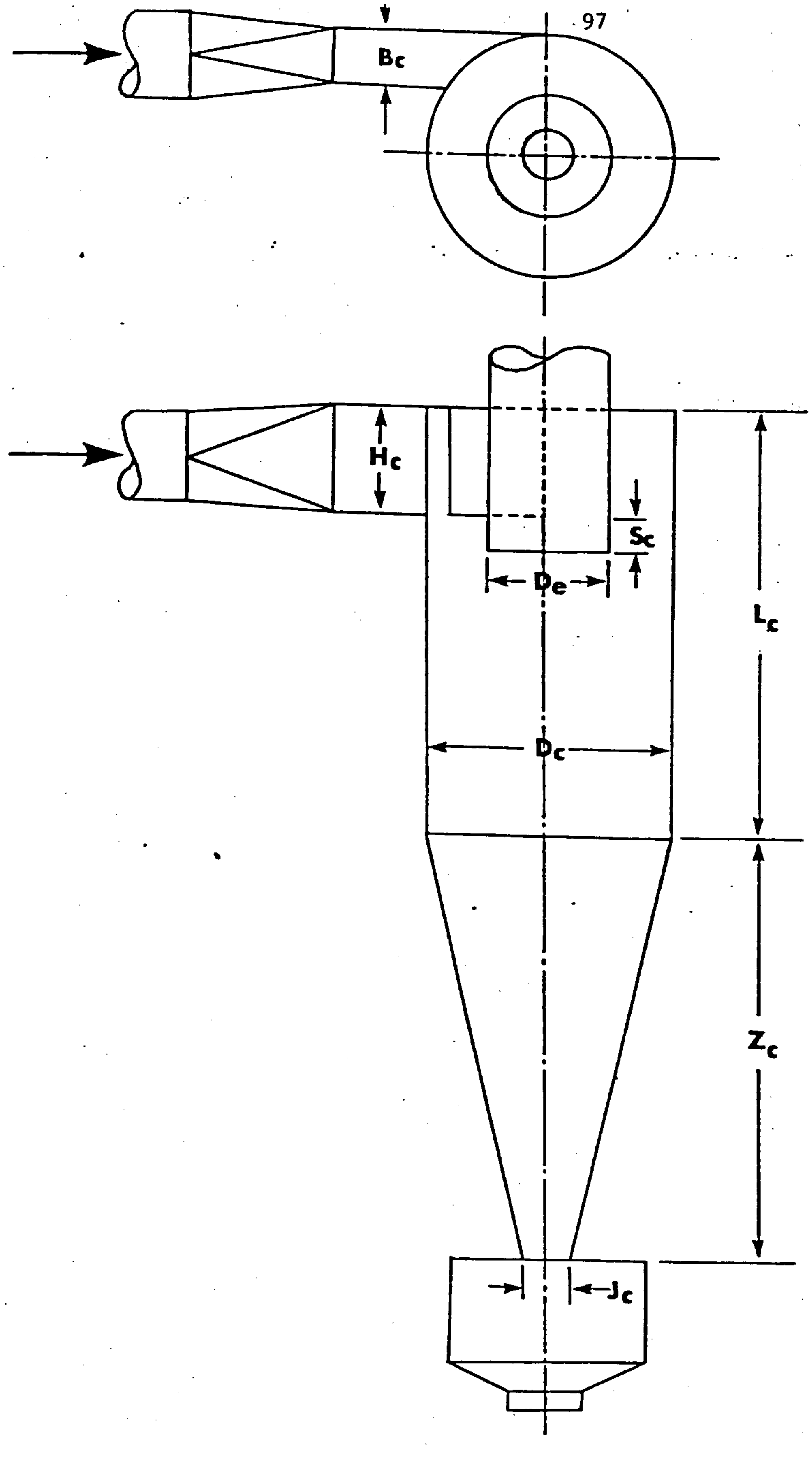
Perforated Plate Gas Distributor

Fig.(3)



General Dimensions of The Gasifier

Fig.(4)



Schematic Diagram of Cyclone Showing Important Dimensions

Fig. (5)

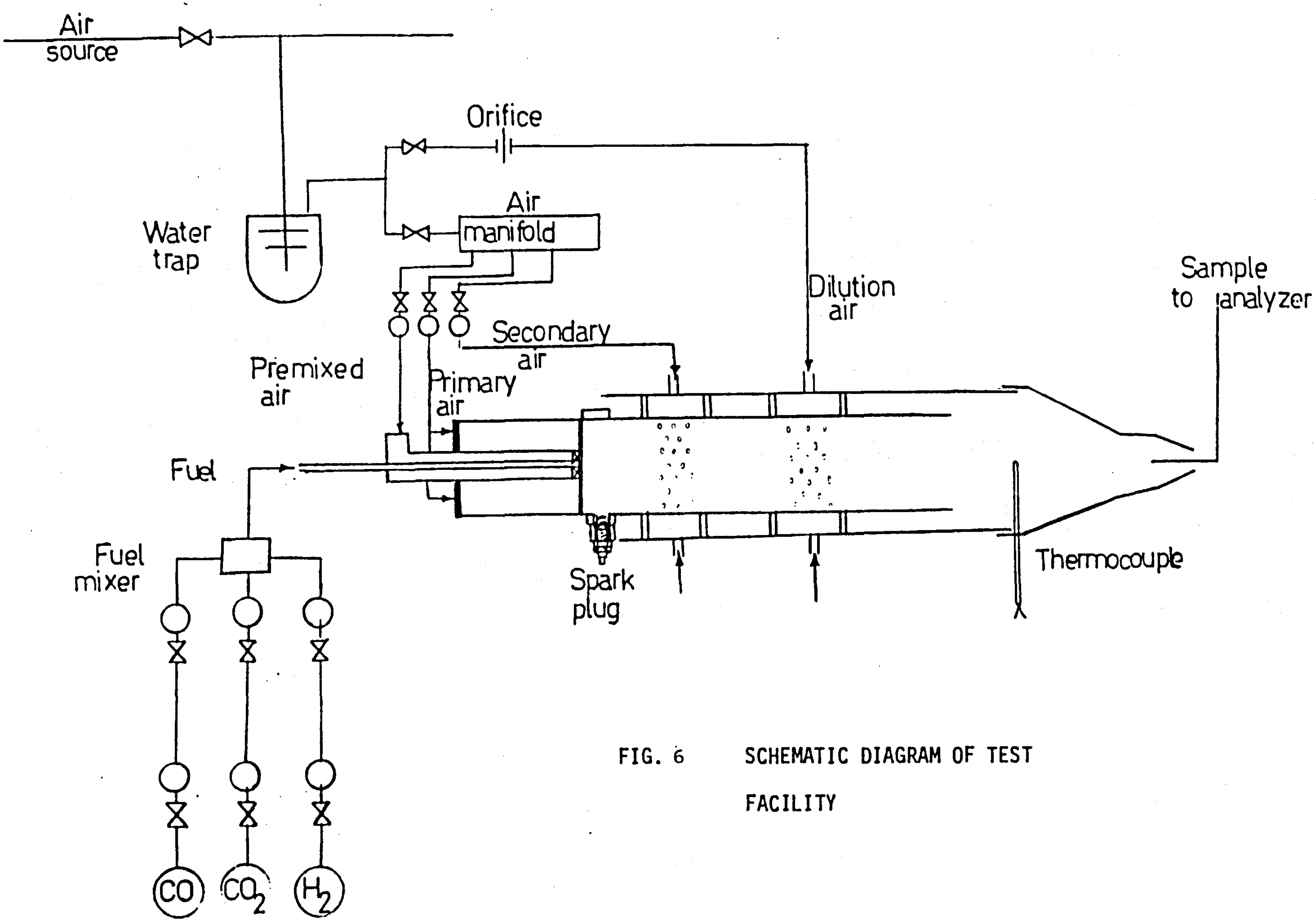
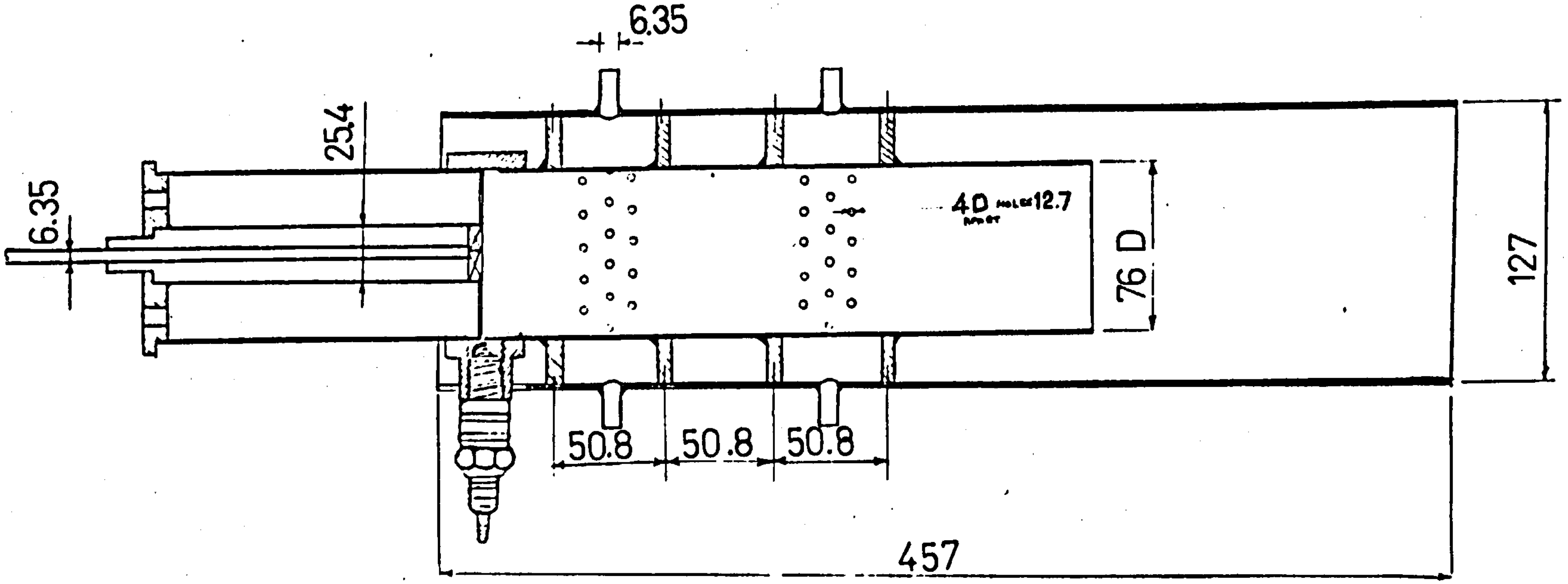


FIG. 6 SCHEMATIC DIAGRAM OF TEST FACILITY



DIMS. IN MM.

FIG. 7 LONGITUDINAL SECTION OF THE DEVELOPMENT TYPE COMBUSTOR

PERFORMANCE OF THE FLUIDIZED BED GASIFIER AND GAS BURNER SYSTEM*

by

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1.0. INTRODUCTION

The development of the fluidized bed coal gasifier and gas burner system has been described in previous reports (1,2). Preliminary results obtained with sub-bituminous coals in the system were reported in ref. 2, some of which are included in this paper. Some recent results obtained with lignite coal are also included. Attempts to gasify bituminous coals have not been successful to date, although it could probably be accomplished successfully with a modified gasifier design.

2.0. Test Procedure

Sub-bituminous coal, (from the Sundance Mine, Alberta), was tested in the gasifier to determine whether or not the gasifier would perform as intended by the design described earlier (1).

The flow diagram of the gasifier system is shown in Fig. 1. The coal is fed into the gasifier through a helix feeder. The fluidizing gas is steam plus oxygen. The product gas and coal fines pass from the top of the gasifier to a cyclone to remove dust, then through a heat exchanger to condense water and tar, and finally to a gas meter and a gas analyzer.

The gasifier was operated at atmospheric pressure with a coal feed rate of up to 0.3 kg/hr. The proximate and ultimate analysis of the coal under investigation were obtained.

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*Based on work sponsored by the Department of Energy, Mines and Resources of Canada under contracts OSU77-00413, OSU78-00368 and OST80-00006.

2.1. Ignition

Oxygen and propane (later carbon monoxide was substituted for propane) were introduced through the ports of the ignition system to provide a flame for coal ignition. At the beginning of the ignition, when the average bed temperature was still low (100°C), a sample of the combustion product gas was intorduced to the gas analyzer. The result of the analysis is given Fig. 2. From this analysis, it appears that some oxygen and probably some unburned propane appear between the combustion product constituents on the chromatogram.

The temperature of the bed continued to increase; when it arrived at about (200°C), another sample was analyzed. This analysis indicates that oxygen almost disappears in the combustion products, Fig. 3, as a result of the bed temperature rise.

2.2. Gasification - sub-bituminous coal

When the average bed temperature arrived at about 250°C, the main oxygen gasifying stream was set to 0.085 std. m^3/hr (3 std. ft^3/hr) and the ignition system was shut off. When the bed temperature reached 400°C, steam was introduced at the bottom of the gasifier (with feed rate = 0.425 std. m^3/hr). At a temperature of 450°C the feed rate of each reactant was as given below:

Coal = 0.25 kg/hr
Oxygen = 0.085 std. m^3/hr (SCMH) (3 std. ft^3/hr)
Steam = 0.425 std. m^3/hr (SCMH) (15 std. ft^3/hr)

A product gas sample was introduced to the chromatograph at this condition to give the analysis which is shown in Fig. 4.

The coal feed rate was increased to 0.3 kg/hr, while the steam and oxygen feed rates were maintained the same as before. At this condition and when the bed temperature attained 500°C, the product gas analysis shows the following: Fig. 5.

(a) An increase in CO/CO_2 ratio due to an increase in coal feed rate.

(b) A small increase in the fraction of H_2 in product gas.

(c) Fig. 6 gives the product gas analysis obtained at a bed temperature of 570°C and at the same reactants ratio as used to obtain Fig. 5. This analysis shows a further increase in CO/CO_2 ratio due to altered reactions at increased temperatures, although the reactants ratio is the same as before.

(d) To allow the bed temperature to continue to rise, the oxygen/coal ratio is increased while maintaining a constant steam/coal ratio. The reactants feed rate producing a bed temperature of 650°C was:

Coal = 0.3 kg/hr
Oxygen = 0.113 std. m^3/hr (4 std. ft^3/hr)
Steam = 0.425 std. m^3/hr (15 std. ft^3/hr)

As shown in Fig. 7, this increase in oxidizer/coal ratio causes a large increase in CO_2/CO.

(e) At the same oxygen/coal ratio as used for Fig. 7, but with a higher steam/coal ratio (steam = 0.56 std. m^3/hr, coal = 0.3 kg/hr), the product gas analysis (bed temperature = 700°C) indicates: Fig. 8

(i) The carbon monoxide to carbon dioxide (CO/CO_2) ratio is increased again due to a larger increase in temperature.

(ii) A slight increase in the hydrogen (H_2) concentration in the products.

(f) Fig. 9 represents a sample of product gas at the same reactants feed rate as in Fig. 8, but at a higher bed temperature (745°C). This analysis indicates a higher ratio of CO/CO_2) from the previous; although the carbon monoxide (CO) has the same concentration as before, the carbon dioxide (CO_2) fraction is decreased. Also, higher temperature increased the steam reactivity producing more hydrogen, even though the steam/coal ratio is the same as before.

(g) Here, the reactants ratio is increased (O_2/Coal = 0.42 std. m^3/kg coal, H_2O/Coal = 2.4 std. m^3/kg coal). The analysis of the products, when the bed temprature attains 800°C, shows: Fig. 10.

- (i) A larger increase in the hydrogen (H_2) fraction more than before.
- (ii) the concentration of the carbon dioxide in the products has dropped.
- (iii) the fraction of the carbon monoxide has continued to increase.

- Should be noted that the hydrogen fraction in the products is calculated by difference.
- The nitrogen in the product gases originated from the coal fed.

3.0. RESULTS

A. Sub-Bituminous Coal

When the average bed temperature arrived at 800°C, these performance data were measured:

(a) Input

- i. Coal: 0.3 kg/hr
- ii. Oxygen: 0.126 std. m^3/hr (4.5 std. ft^3/hr)
- iii. Steam: 0.72 std. m^3/hr (26 std. ft^3/hr)

(b) Reactants ratio

- i. Oxygen: Coal = 0.42 std. m^3/kg coal
- ii. Steam: Coal = 2.4 std. m^3/kg coal

(c) Product gas

The average product flow rate = 0.5 std. m^3/hr.

(d) Product gas analysis

Product gas analysis, which is given by the chromatograph is shown in Fig. 10. Comparison between this figure and the calibration figures, gives these results:

Approximate product	gas %
CO	24
CO_2	37
CH_4	3.2
N_2	1

By difference, the hydrogen fraction is 34.8%.

(e) Thermal efficiency

$$\text{Cold gas efficiency} = \frac{\text{Gross heating value of gas product}}{\text{Gross heating value of coal fired}}$$

Calorific value of the coal under investigation

= 23.96 MJ/kg dry coal

= 19.8 MJ/kg coal

= 4729.5 kcal/kg coal

Gross heating value of coal fed (at 0.3 kg/hr)

= 1418.8 kcal/hr

Product Gas	Percent Ratio	Flow Rate (gram.mole/hr)	Heating Value (kcal/gram.mole)	Total Heating Value (kcal/hr)
CO	24	$\frac{0.5\times10^3}{22.4}\times 0.24 = 5.3$	67.4	361
CO_2	37	-		
H_2	34.8	7.7	58	450.5
N_2	1	-		
CH_4	3.2	0.71	210	150
				961

If the energy content of the incoming steam and the thermal energy of the product gases are neglected, the cold gas efficiency is 67.7%. If the energy of the incoming steam is considered but the sensible enthalpy of the product gas neglected, the cold gas efficiency is 57%. If the energy of the steam and the sensible energy of the products are considered, the hot gas efficiency is about 63.8%.

B. Lignite coal

Some tests were carried out using Estevan lignite of -200 mesh size, 1.69 specific gravity. The values for minimum fluidization velocity and terminal velocity were to be recalculated because of the different fuel properties. These values were found to be 1.32 cm/s for u_{mf} and 13.02 cm/s for u_t using the equations mentioned in the previous report (1,2). A simple arrangement was put together in the laboratory to find these values experimentally. Experimentally obtained values were found to be about 1.5 and 15.46 cm/s for u_{mf} and u_t respectively. Fluidization was tried using O_2 or air or both without adding any steam. The steam needed for the water shift reaction was introduced through a separate pipe above the distributor plate.

During all the tests with the lignite coal, it was not possible to turn the ignition gases off, while maintaining a stable temperature in the bed. It seems that the heat loss fraction is too great to operate without auxiliary heating, with this type of lignite, in our small bed.

Tables 1, 2 and 3 show results and a summary of the operating conditions used for some tests with the Estevan lignite. It is interesting to note the high concentrations of CO_2 in the results shown in Table 1. In these tests, CO was used as the igniter gas, which may account for an imbalance toward CO_2. a mixture of hydrogen and helium was used as the carrier gas for the chromatographic analysis of these gases, so the sensitivity to hydrogen was very poor. Therefore, rather than attributing the balance to hydrogen, it is not specified here. The nitrogen appearing in Table 1 comes in from the air used in the igniter.

Table 2 results are obtained with similar conditions except that air is used in place of oxygen as the major fluidizing gas. Here again, hydrogen is not specifically shown.

Table 3 results are also obtained using air as the main fluidizing stream, but now argon is used as the carrier gas in the chromatograph allowing some indication of hydrogen. These results appear confusing, at best, and can only be termed preliminary. The wide range of gas compositions measured seem to emphasize that the gasification process is very sensitive to variables which are not adequately controlled in these experiments.

4.0. SUMMARY AND CONCLUSIONS

The gasifier described in the earlier report has given some encouraging results with sub-bituminous coals for which the original design was intended. When lignite coals were introduced, although their properties suggest that the apparatus should be appropriate, considerable difficulty was experienced in obtaining results. The results obtained for the lignite are of a preliminary nature only.

The problems encountered in connection with the lignite tests are now being analyzed to decide on the best approach to follow in improving results with such coals.

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Table 1 - Results obtained in the fluidized bed gasifier using oxygen and steam with Estevan Lignite coal

	Gas Analysis					T_{bed}	Fluidizing		Coal Fed	Steam
	CO_2	O_2	N_2	CO	Bal*	°C	O_2 SCMH	Air SCMH	rate kg/h	rate kg/h
1	49	-	9.9	36.8	4.3	300	0.264		0.275	0.48
2	45.6	1.3	11	41.1	1	316	0.264		0.33	0.48
3	47.	1.0	10.2	41.1	0.7	346	0.198		0.33	0.36
4	63.3	-	-	23.7	13.	422	0.108		0.33	0.18
3	51.7	0.4	20.6	15.3	12.	675	0.198		0.22	0
4	40.7	3.	27.2	2.1	27.	677	0.156		0.22	0.21
6	44.5	0.2	28.2	6.3	20.8	646	0.18		0.275	0.24
7	45.	0.2	9.6	43.4	1.8	619	0.228		0.165	0
8	54.2	-	12.2	24.7	9.9	765	0.198		0.165	0.3
9	56.2	-	14.8	15.3	15.7	700	0.198		0.22	0.3
10	56.7	2.	13.1	-	28.2	586	0.198		0.275	0.3
11	58.3	2.5	13.1	1.6	24.5	580	0.144		0.275	0.36
13	51.	0.1	8.2	39.2	1.5	400	0.198		0.33	0.48
14	36.7	0.2	24.6	21.1	17.4	645	0.264		0.33	0.3
15	49.2	0.2	13.1	35.5	2.	656	0.198		0.22	0.3
16	58.3	0.2	13.5	18.9	9.1	473	0.276		0.22	0.24
17	58.	0.8	14.9	4.5	21.8	587	0.144		0.22	0.24

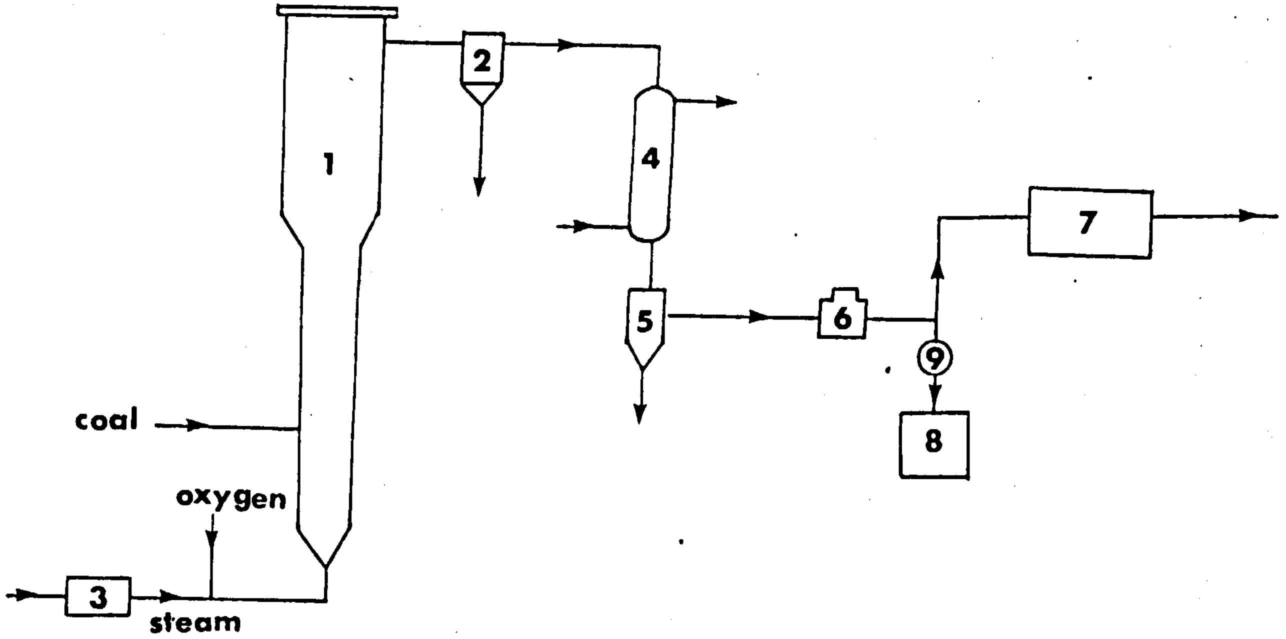
* The carrier gas used in the chromatograph for these runs was a mixture of H_2 and H_e which gave poor sensitivity to H_2 in the analysis. The balance may have been mostly H_2.

Table 2 - Results obtained using air and steam in the fluidized bed gasifier with Estevan Lignite coal

	Gas Analysis %						Fluidizing			
	CO_2	O_2	N_2	CO	Bal	T_{bed} °C	O_2 SCMH	Air SCMH	Coal rate kg/h	Steam rate kg/h
11	22.3	0.9	31.1	9.5	36.2	414		0.311	0.275	0.2
12	22.7	1.1	32.9	5.3	38.	543		0.481	0.165	0.2
14	24.	0.5	31.1	12.1	33.	480		0.481	0.22	0.2
16	22.	1.1	25.6	26.3	25.	470		0.481	0.22	0.2

Table 3 - Results obtained in the fluidized bed gasifier using air and steam with Estevan Lignite coal

	Gas Analysis %							Fluidizing			
	CO_2	O_2	N_2	CO	H_2	CH_4	T_{bed} °C	O_2 SCMH	Air SCMH	Coal kg/h	Steam kg/h
3	21.6	5.3	53.7	-	1.1	-	383		0.566	0.22	0.12
4	21.6	5.2	56.6	-	1.7	-	450		0.566	0.22	0.12
5	24.3	-	56.6	-	2.7	-	444		0.566	0.22	0.12
6	22.3	1.3	56.5	2.9	2.7	-	392		0.283	0.22	0.12
8	23.	2.	53.4	2.9	2.3	-	340		0.566	0.11	0.12
9	24.	2.	55.4	2.9	2.7	-	513		0.566	0.22	0.12
10	24.3	1.6	52.8	3.3	4.1	-	548		0.566	0.22	0.12
13	28.4	2.	52.4	3.3	3.5	-	530		0.566	0.22	0.18
17	26.4	5.5	53.	1.4	3.5	0.2	492		0.425	0.22	0.18



1- Fluidized bed gasifier
2- Ash cyclone separator
3- Steam preheater

4- Gas cooler
5- Gas liquid separator
6- Back presure regulator

7- Gas Chromatograph
8- Gas holder
9- Gas meter

Flow diagram of the bench-scal gasification unit

Fig. (1)

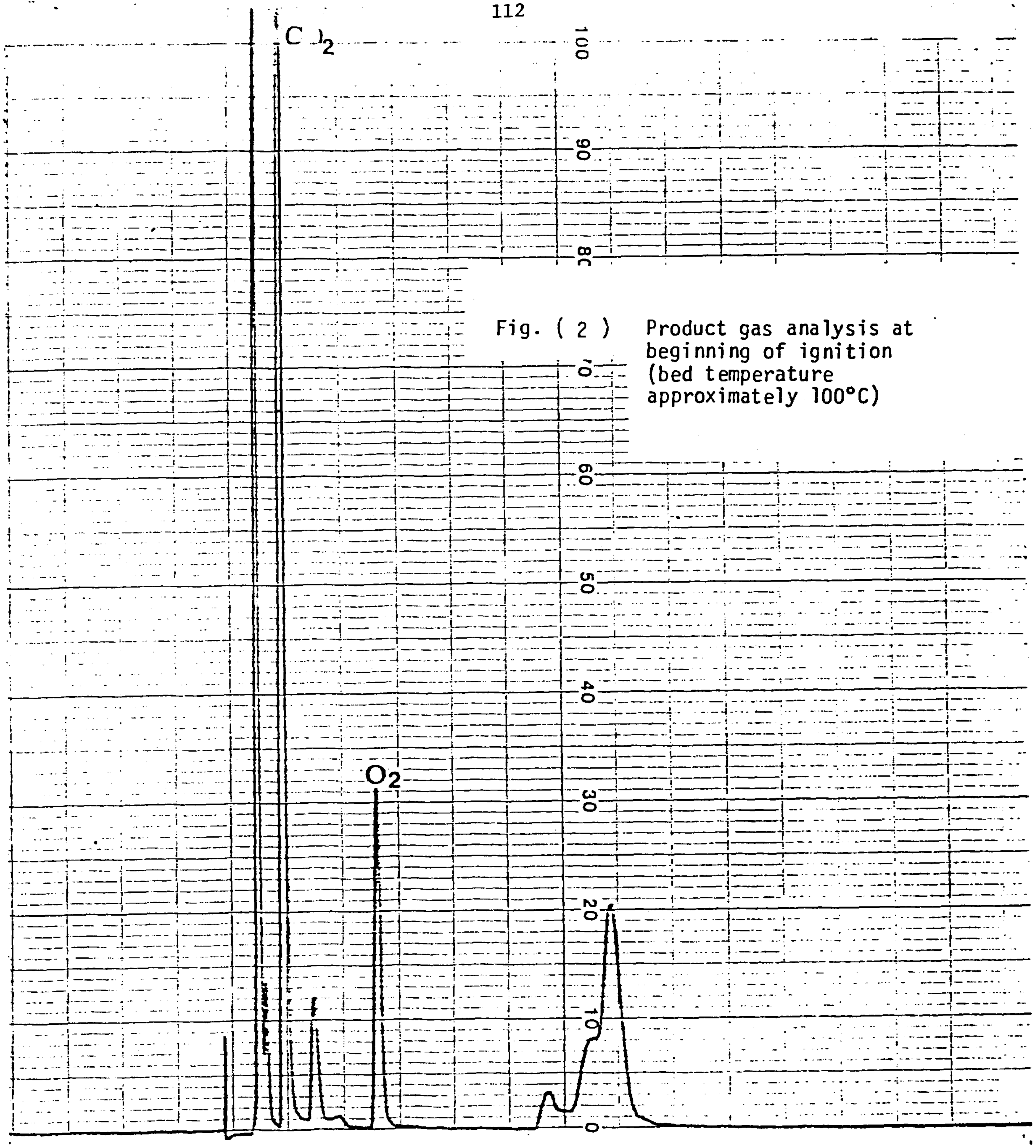


Fig. (2) Product gas analysis at beginning of ignition (bed temperature approximately 100°C)

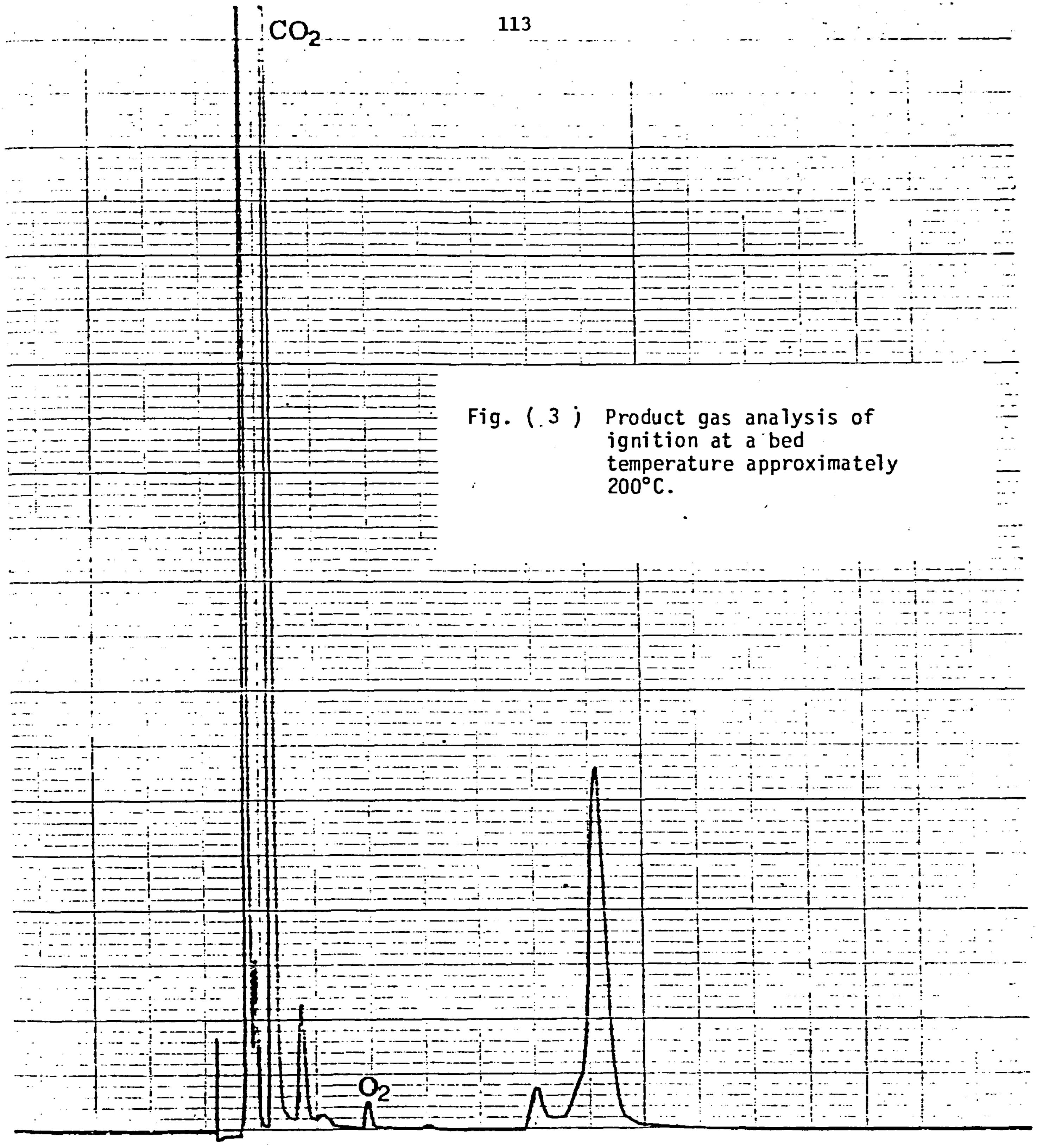


Fig. (3) Product gas analysis of ignition at a bed temperature approximately 200°C.

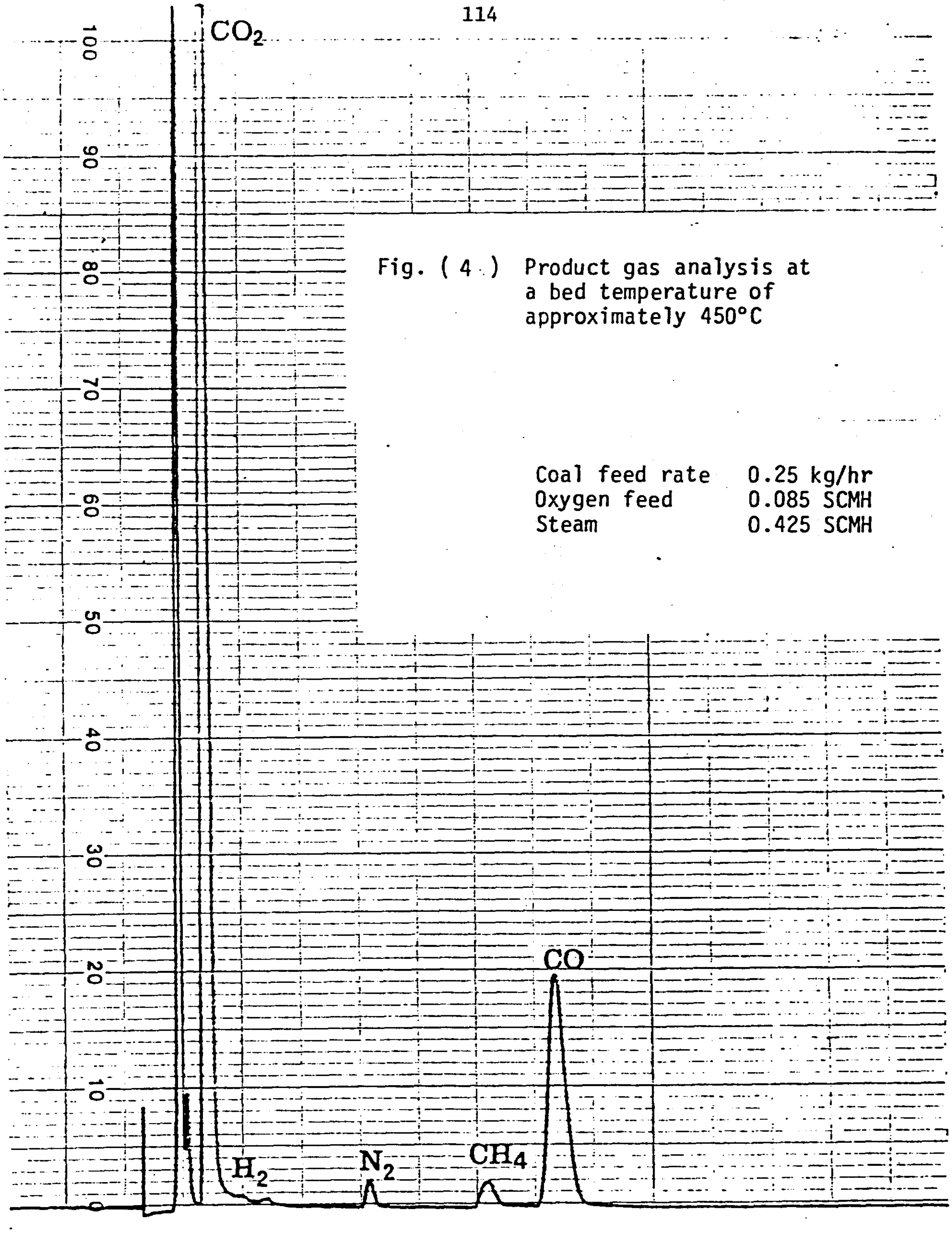


Fig. (4) Product gas analysis at a bed temperature of approximately 450°C

Coal feed rate	0.25 kg/hr
Oxygen feed	0.085 SCMH
Steam	0.425 SCMH

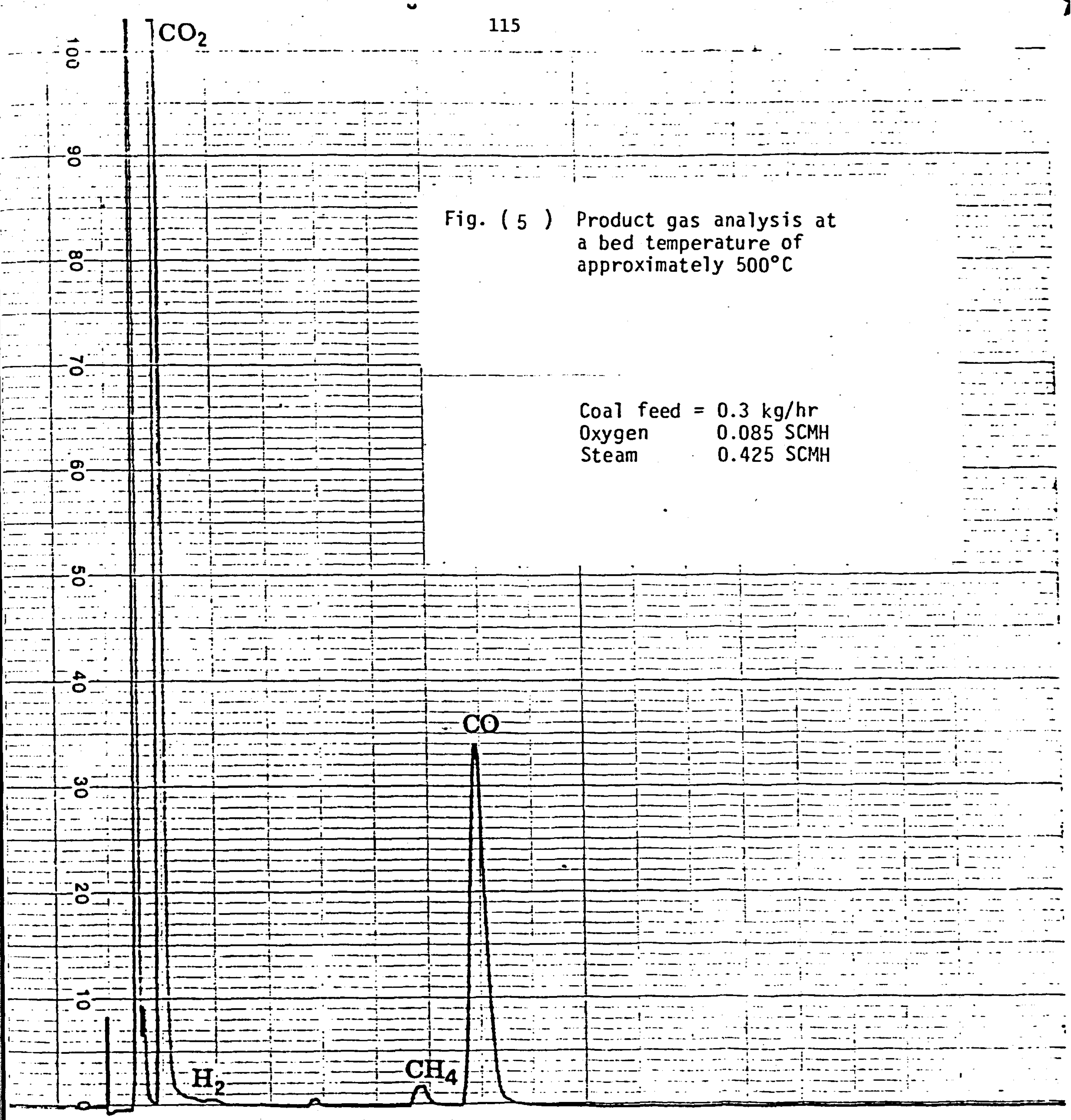


Fig. (5) Product gas analysis at a bed temperature of approximately 500°C

Coal feed = 0.3 kg/hr
Oxygen 0.085 SCMH
Steam 0.425 SCMH

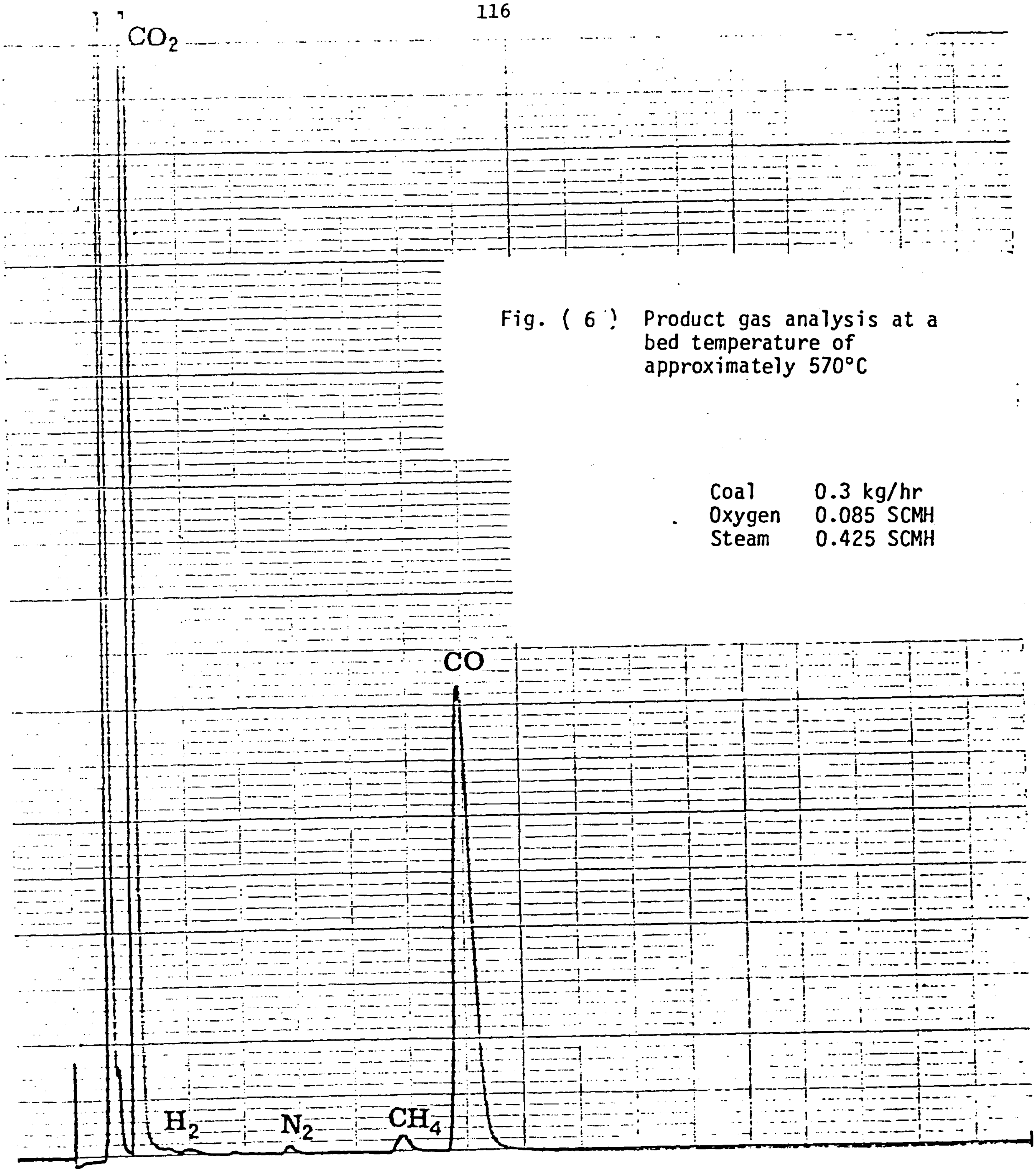


Fig. (6) Product gas analysis at a bed temperature of approximately 570°C

Coal	0.3 kg/hr
Oxygen	0.085 SCMH
Steam	0.425 SCMH

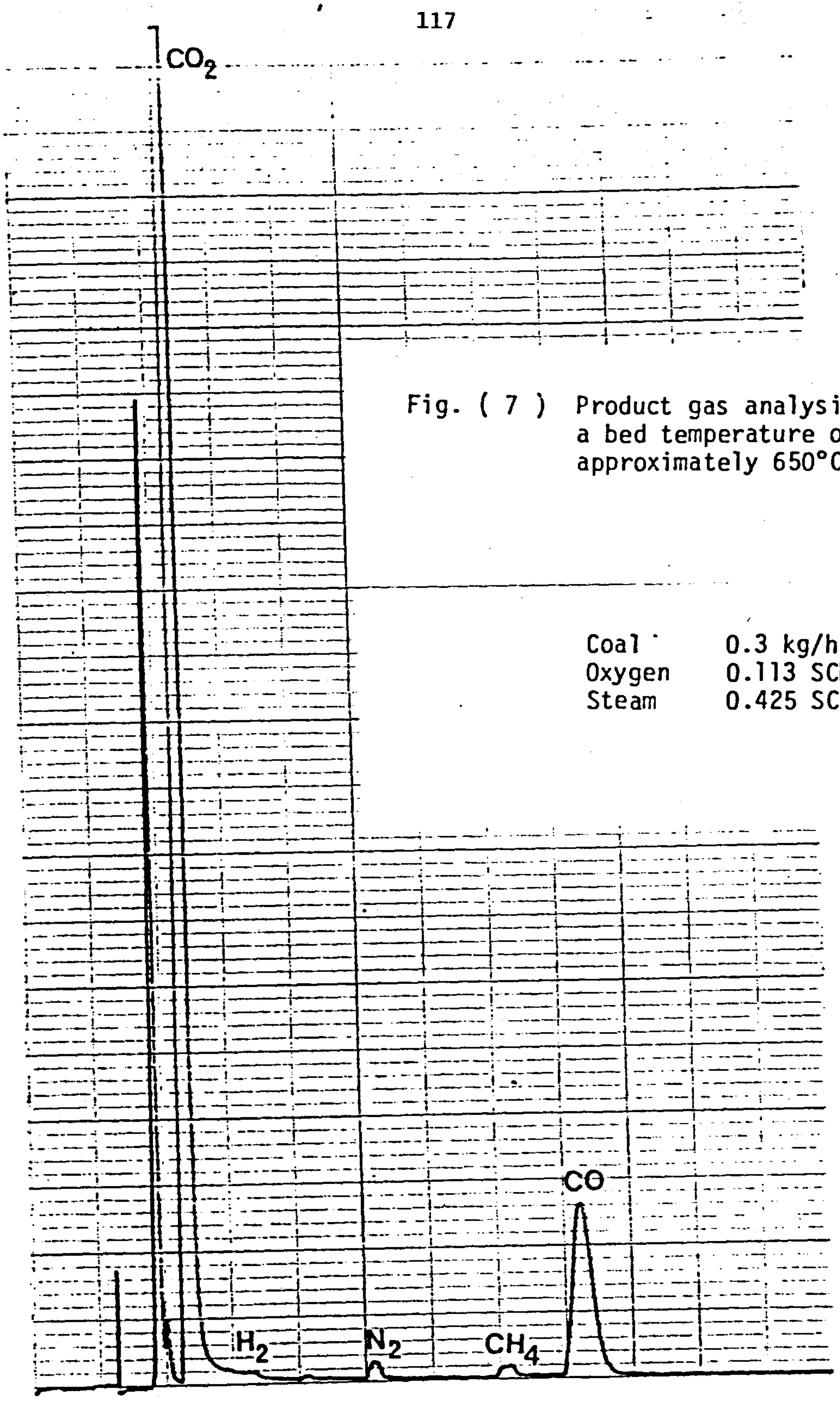


Fig. (7) Product gas analysis at a bed temperature of approximately 650°C

Coal	0.3 kg/hr
Oxygen	0.113 SCMH
Steam	0.425 SCMH

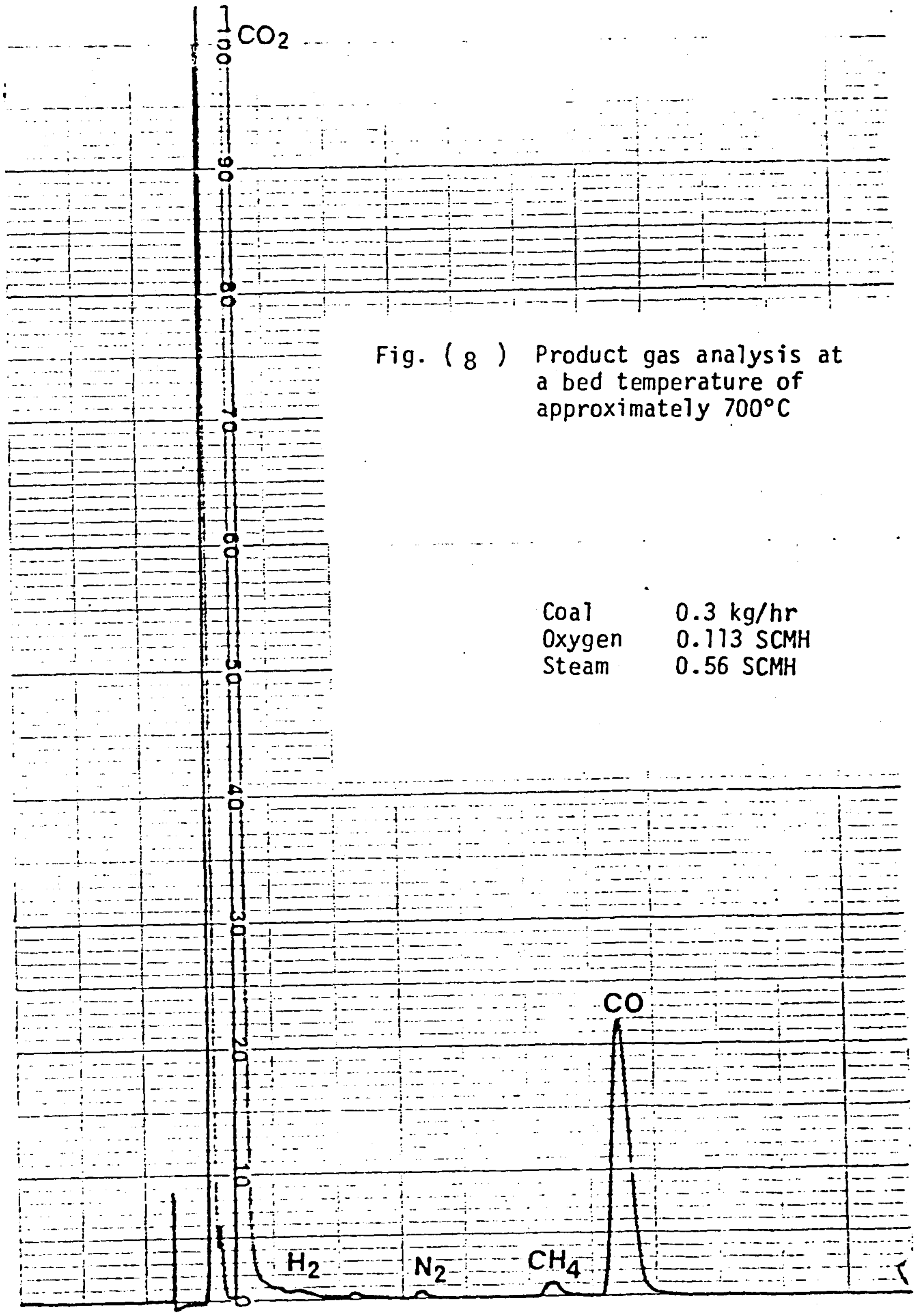


Fig. (8) Product gas analysis at a bed temperature of approximately 700°C

Coal	0.3 kg/hr
Oxygen	0.113 SCMH
Steam	0.56 SCMH

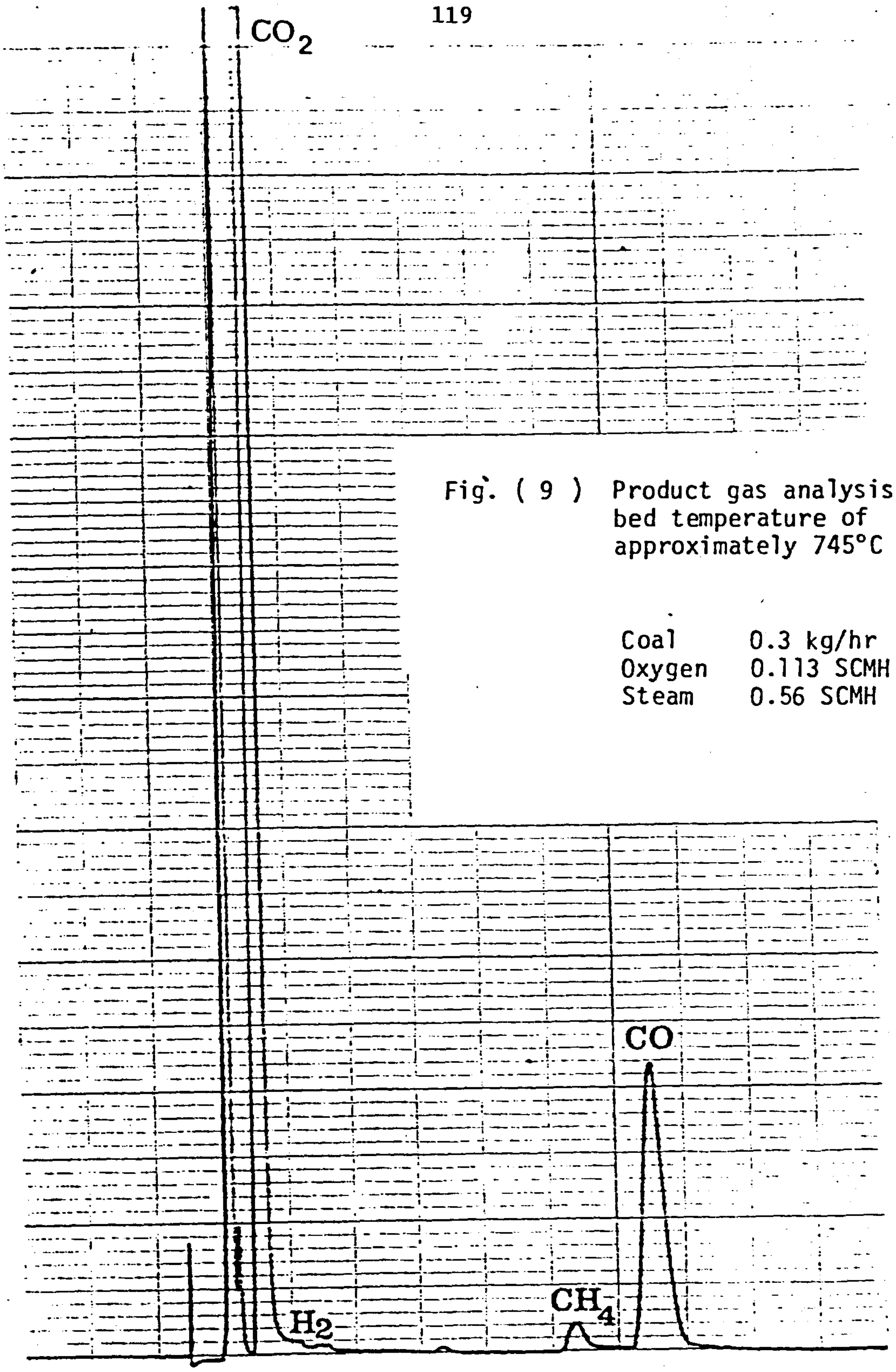


Fig. (9) Product gas analysis at a bed temperature of approximately 745°C

Coal	0.3 kg/hr
Oxygen	0.113 SCMH
Steam	0.56 SCMH

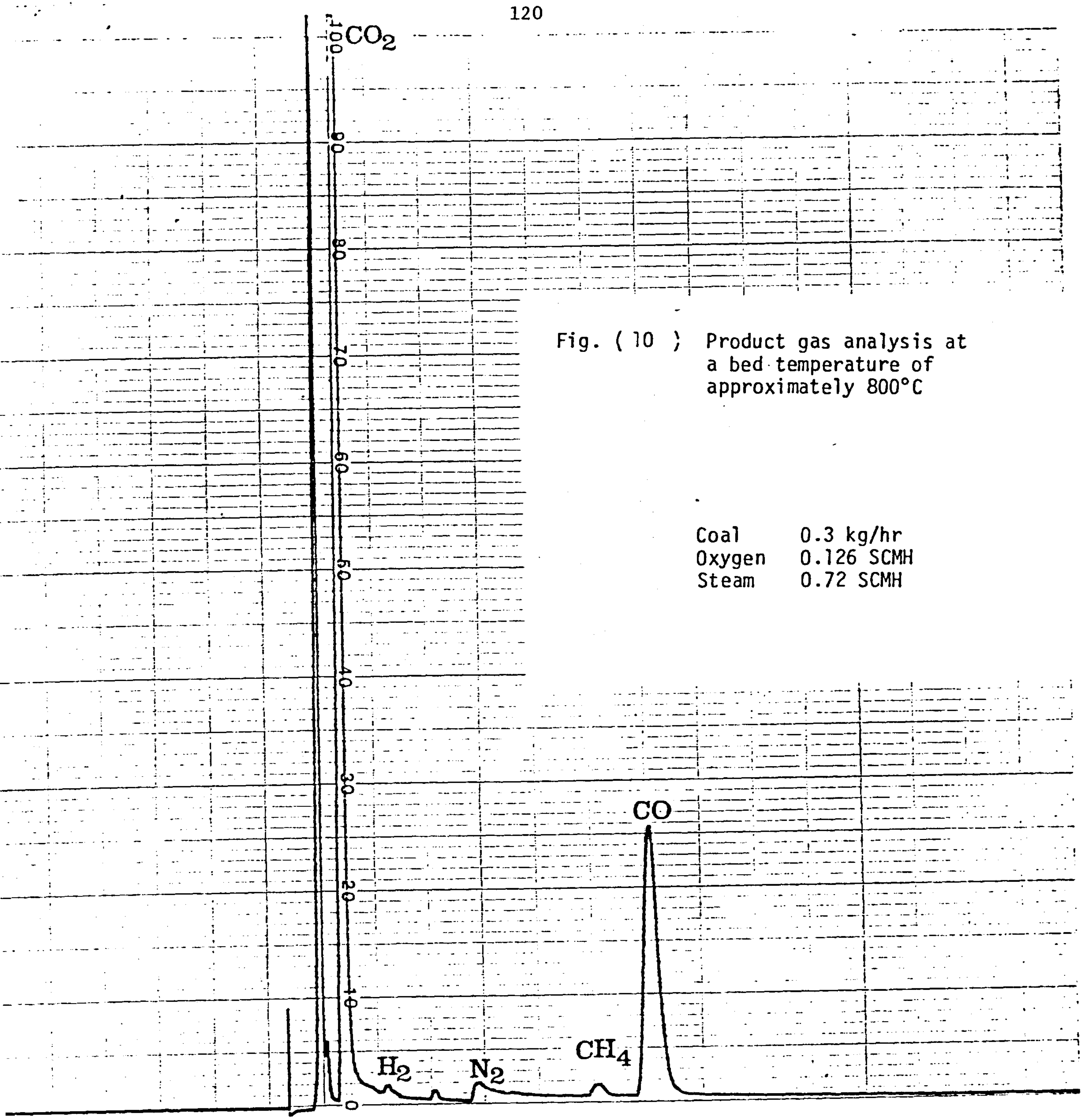


Fig. (10) Product gas analysis at a bed temperature of approximately 800°C

Coal	0.3 kg/hr
Oxygen	0.126 SCMH
Steam	0.72 SCMH

GASIFICATION OF LIGNITE AND CHEMICAL REACTIVITY EXPERIMENTS

by

B.D. Kybett and R.N. Neufeld

Microscopy is a useful tool for monitoring the chemical reactivity of a coal during gasification, but a large number of samples obtained by reaction under varied and carefully conrolled conditions are necessary to form the information bank needed to establish predictive rules. Pilot plant and bench scale coal gasification experiments are expensive, and their number limited. In addition the products have often been exposed, on the particle level, to (unknown) varying conditions within the reactor, even though the average conditions are well characterised.

A laboratory bench scale coal gasification apparatus was constructed, to provide samples for microscopic examination. It consisted of a reactor in which between 10 g. and 50 g. samples of fuel could be quickly heated to temperatures up to 1100°C. The reactor was continuously rotated to agitate the sample, mimicking a fluidized bed. The atmosphere, at 1 atm pressure, was steam and air or oxygen.

The small scale of apparatus meant that the reaction conditions could be uniform over the whole sample, and carefully controlled. It's size also made it versatile.

The apparatus was designed, to reduce costs, to operate at 1 atm pressure only. This, as well as its small size, meant that it could not provide engineering data for scale-up. Its operation was, however, compared to the pilot plant at the Research and Development Laboratory, Saskatchewan Power Corporation. Identical feedstocks were used, with the same temperatures and atmospheres, but at different pressures. Product yields were different, as these vary with pressure, but the pattern was similar. Microscopic examination of the products also showed similar behaviour, this is discussed in the paper by J. Potter.

Apparatus

The reactor system can be broken down into three sections. The inlet mainifold, for creating the appropriate atmosphere, the reactor proper, and the outlet manifold for analysing the gaseous products.

1) Inlet manifold

Air (or oxygen) was filtered, regulated, and saturated with water by bubbling through tandem water baths. The air/steam ratio could be adjusted by changing the temperature of the water baths.

2) Reactor

The reactor consisted of a 1 m length of Vycor tube, 2 cm in diameter. It was fitted at both ends with Teflon sleeved ball joints so that it could be rotated, at speeds up to 10 sec^{-1}. The tube passed through two tube furnaces. The reaction chamber was formed inside the tube by pads of asbestos wool.

The air/steam mixture was preheated to 300°C in the inlet manifold, to 500°C in the first part (furnace) of the reactor tube, and to the reaction temprature in the second part (furnace) of the reactor tube. The temperature of the gases exiting the reaction tube was measured with a bare thermocouple mounted axially in the reaction tube.

3) Outlet manifold

The exiting gases were passed through a cold trap to condense water, and their flow measured. Samples were collected for chemical analyses in Teflon sampling bags. Analyses were done using an Orsat type macro absorptiometric gas analyses system.

Procedure

A set of typical results is presented in Table 1, and is self explanatory. The mass balance is not particularly good. The major part of the unaccounted mass was water, which condensed in the tubing and could not be completely removed. Some gas was also lost, by leakage (approx. 3%) through the reactor tube bearings, and also because the very large gas flow that occurred during the initial rapid heating of the sample could not be monitored accurately. The poor mass balance was not considered significant since measurement of product yield was not the purpose of these experiments.

Results

A direct comparison with a Saskatchewan Power Corporation gasification experiment (paper by S. Van der Heijden) was difficult to make, even though the basic operating parameters (except pressure) were matched. The sample sizes were different (150 g and 11 g), and the larger sample size in the SPC tests meant that the temperature of the reacting sample was not the same as that of the reaction furnace.

Our experiment 400/R can be compared with the SPC 1600 experiment, Table 2. These experiments were done with the same feedstock, air/steam ratio and flow, and temerature. Experiment 400 R was terminated, as normal, after 15 minutes in order to provide a partially reacted char for petrographic analysis. Experiment SPC 1600 was terminated after 180 minutes.

The yields of gaseous products are compared in Figures 1 to 4. They are similar.

A total of 21 successful experiments were performed on 5 different feedstocks, at three temperatures (600°C, 800°C and 1,000°C) and with two types of atmosphere (air/steam and oxygen/steam). The products of 15 of these experiments were investigated petrographically. The results of these experiments are not presented here, because of space limitations, but are given in our report to EMR Canada.

Table 1 - Gasification of lignite, sample Bien 2B, experiment No. 400/R, in an air/steam atmosphere at 700°C

Reactants

Feedstock, Bien 2B, weight 11.28 g., moisture content 11.3%

Air flow, 100 ml. min^{-1} at 24°C and 860 Torr

Water bath temperature, 91.1°C

Air flow rate = 0.13 g. $min.^{-1}$

Water flow rate = 0.26 g. min^{-1}

Reaction conditions

Pre-heat furnace 500°C

Reaction furnace 700°C

Products

GAS SAMPLES

Sample No.	Time (min.)	Reaction Temp. °C	Product Gases Flow Rate (ml. $min.^{-1}$)	Product Gases Pressure (Torr)
403	3	608	220	712
405	5	671	480	712
410	10	695	220	713
415	15	709	155	713

COMPOSITION OF PRODUCT GASES

Sample No.	Gas Composition, Mole % CO_2	CO	H_2	CH_4	C_2+	O_2	N_2	Heating Value MJm^{-3}
403	34.3	7.3	1.2	6.6	3.9	0.8	45.9	6.9
405	21.7	13.5	24.7	6.7	3.8	1.6	28.0	7.0
410	1.6	22.7	27.3	6.1	0.0	2.0	40.3	7.0
415	1.3	21.2	9.8	3.4	0.0	3.8	60.5	4.4

Table 1 - Cont'd

YIELD OF PRODUCT GASES

	Mol.	g.
CO_2	0.022	0.98
CO	0.023	0.37
H_2	0.029	0.059
CH_4	0.015	0.25
C_2+	0.003	0.014
O_2	0.004	0.14
N_2	0.052	1.46

YIELD OF PRODUCT

Char = 5.78 g. (58% of feedstock, dry weight basis)
Gas = 3.40 g. (34% of feedstock, dry weight basis)
Tar = 1.94 g. (19% of feedstock, dry wieght basis)
Water = 1.3 g. (incomplete recovery)

Total 12.42 g.

Mass Balance

Mass of reactants	17.13 g.
Mass of products	12.42 g.
Mass recovered	73% (mass unaccounted for mainly water)

Table 2 - Comparison of experiment 400/R with S.P.C. 1600

REACTANTS	400/R	S.P.C. 1600
Feedstock	Bien 2B	Bien 2B
Measure content, feedstock, %	11.3	11
Air flow, g. min.$^{-1}$ per g. feedstock	0.011	0.0098
Steam flow, g. min.$^{-1}$ per g. feedstock	0.022	0.019
REACTION CONDITIONS		
Pressure, atmosphere	1	20
Initial temperature, °C	700	700
Temperature after 3 min. °C	608	636
Temperature after 5 min. °C	671	762
Temperature after 10 min. °C	695	765
Temperature after 15 min. °C	709	763
Total reaction time, min.	15	180

FIGURE 1

VARIATION OF MOLE % CARBON DIOXIDE, PRODUCED DURING GASIFICATION OF LIGNITE FEEDSTOCK BIEN 2B, WITH TIME

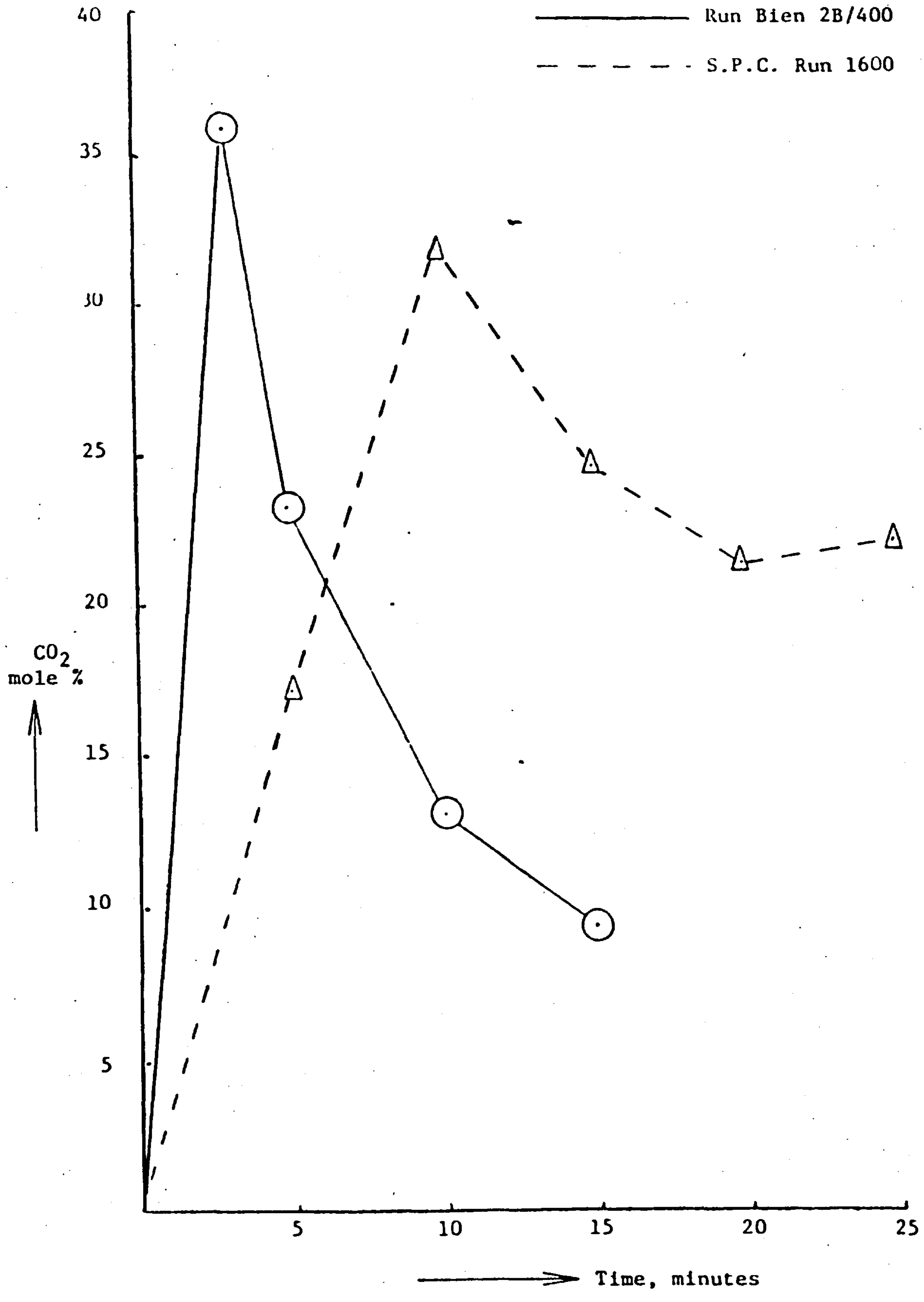


FIGURE 2

VARIATION OF MOLE % CARBON MONOXIDE, PRODUCED DURING GASIFICATION OF LIGNITE FEEDSTOCK BIEN 2B, WITH TIME

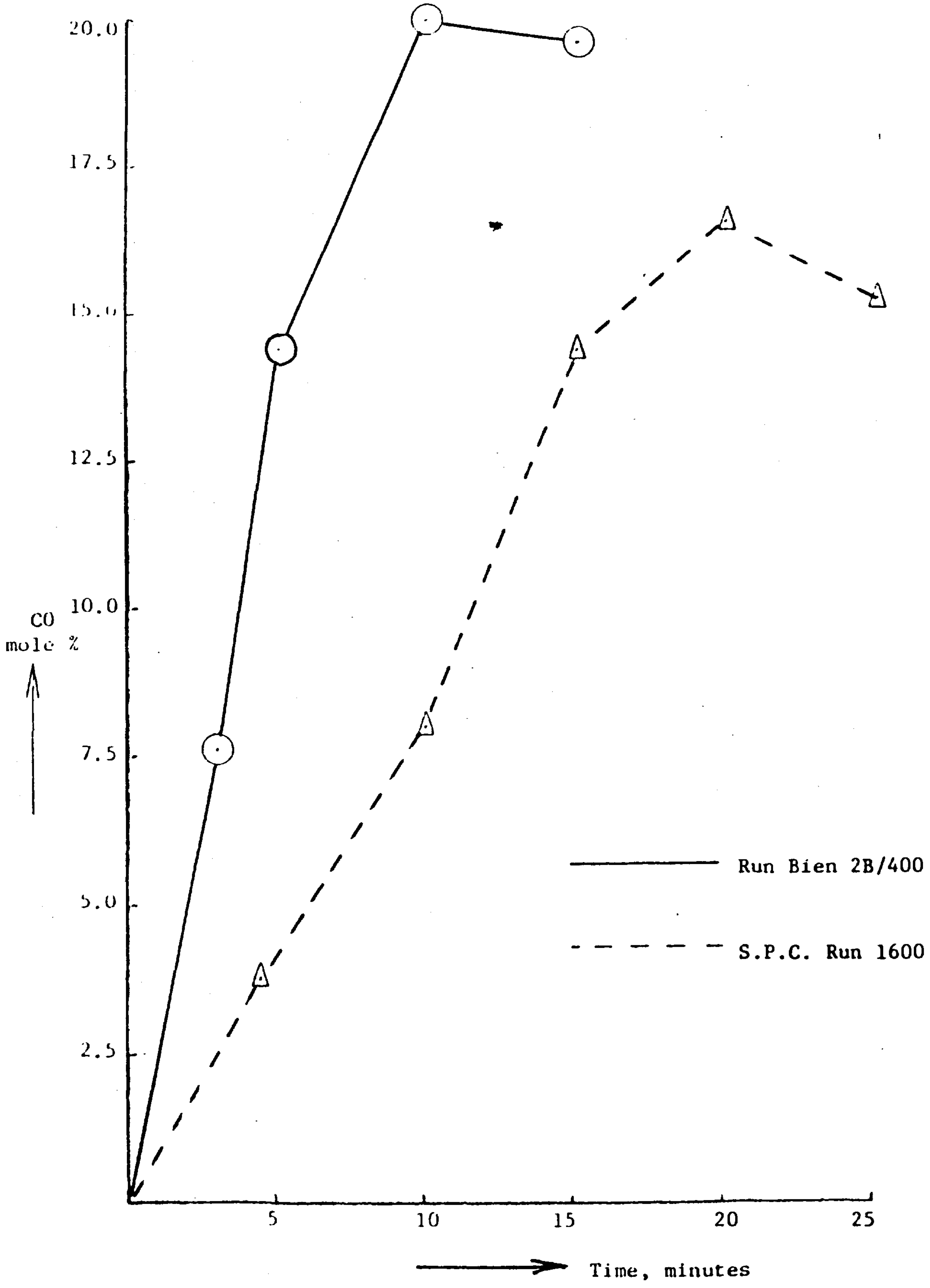


FIGURE 3

VARIATION OF MOLE % HYDROGEN PRODUCED, DURING GASIFICATION OF LIGNITE FEEDSTOCK BIEN 2B, WITH TIME

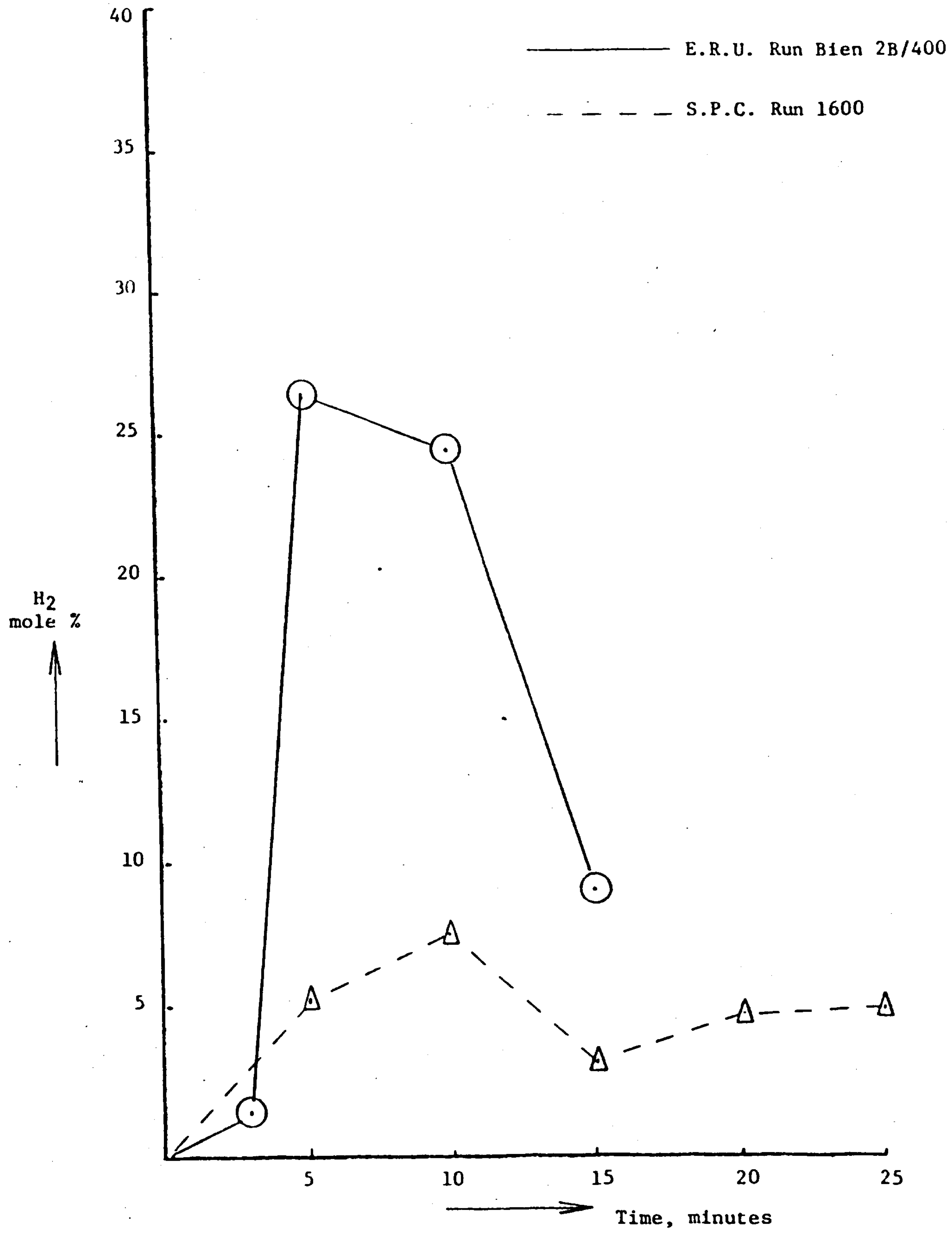
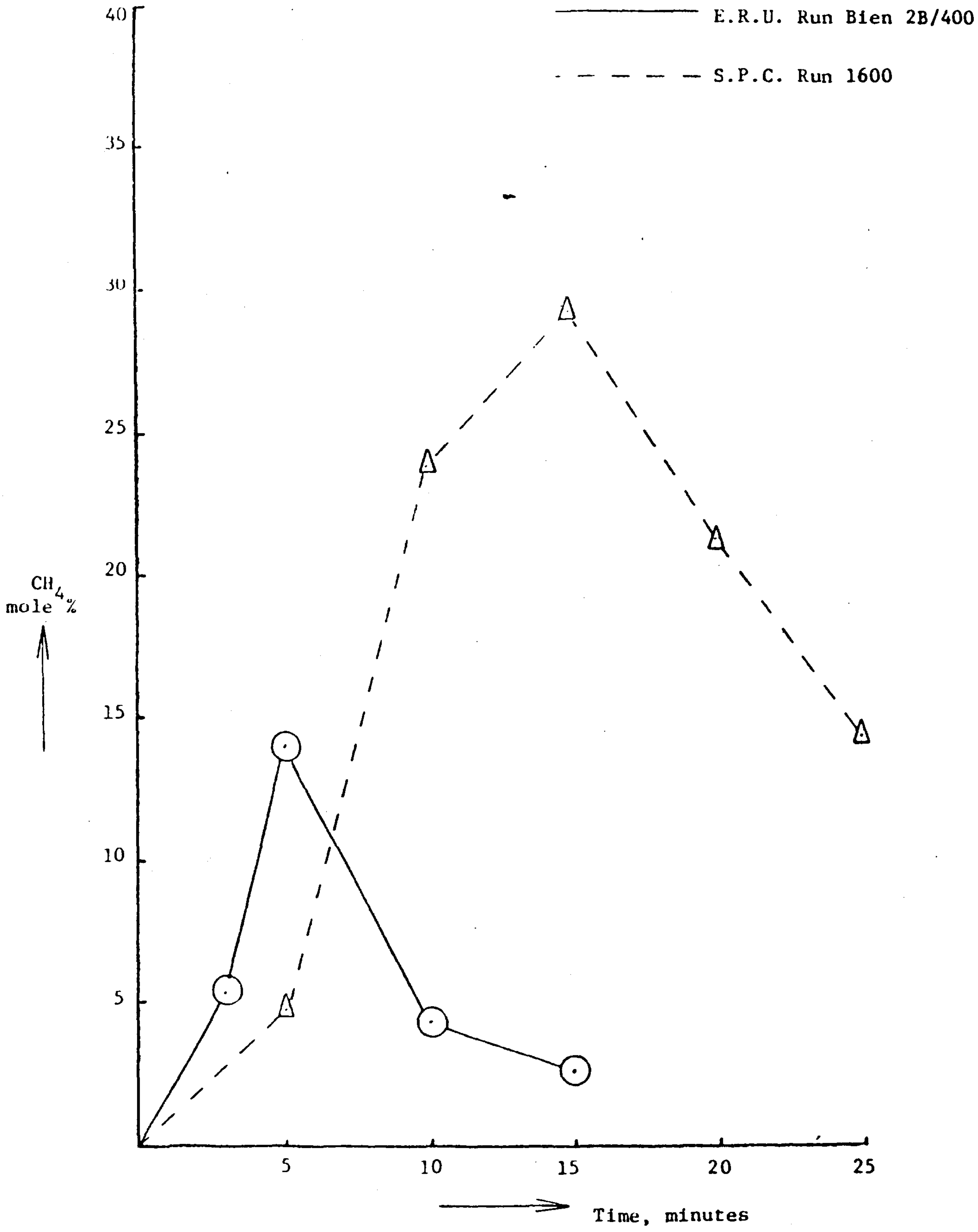


FIGURE 4

VARIATION OF MOLE % METHANE, PRODUCED DURING GASIFICATION OF LIGNITE FEEDSTOCK BIEN 2B, WITH TIME



THE ROLE OF PETROGRAPHY IN LIGNITE GASIFICATION:
The Effects of Coal Composition

by

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ABSTRACT

Petrological studies of feedstocks and residues from gasification experiments using Saskatchewan lignites have shown that composition of the feed coal affects the gaseous yields and residence times required. Petrologically dissimilar lignites were gasified under similar conditions using air/steam at 700°C and 2mPa in an engineering scale reactor; those with high reactive: inerts ratios produced greater yields at shorter residence times, particularly those in which a large proportion of the reactive macerals were structured huminites and/or liptinite. Lignite with a high inertinite content produced considerably lower gaseous yields despite longer residence times.

INTRODUCTION

In considering the influence of coal composition on conversion, it is necessary to review some of the basic concepts and nomenclature used in lignite petrology. It is a long established fact that coal is a very heterogeneous substance derived primarily from plant remains which accumulated in coal forming environments as peat, which were subsequently altered biochemically and geochemically to form coal. Complex organic polymers such as lignins, colluloses, lipids, waxes and other plant derived materials such as spores, pollen and cuticles are eventually transformed into three groups of coal constituents (commonly called 'macerals') which can be identified using morphological land reflectance characteristics when viewed in polished section, under the petrographic microscope, using reflected light in the visible and blue-violet parts of the spectrum. Table 1 shows the standard nomenclature used to describe the constituents of brown coals and lignites recommended by the International Committee for Coal Petrology (1975). A brief explanation of the terminology, and the genetic, physical and chemical properties of the macerals and maceral groups is given as follows:

i) <u>The Huminite Group</u>: It is generally agreed that the huminite macerals are the products of biochemical alteration of easily hydrolysed lignin, starches and cellulose in the peat stage; the remains are reconstituted by polymerisation and generally oxidized (at the peat surface) to form humic acids. Some of these plant meterials are more resistant to bacterial attack than others; lignified wood, tannin or resin impregnated conifer wood, for example, may be well preserved in coal. Cellulose rich tissues and certain woods like angiosperm wood of Tertiary brown coals largely decompose to a humic detritus of cell wall remains. When cellular structure is retained the macerals Textinite and Eu-Ultimate are formed; when they collectively form a detrital groundmass for other macerals, Densinite and Attrinite are formed which give rise to macerals such as corpohuminite and gelinite. Huminites are generally the most abundant macerals in brown coals, particularly in Saskatchewan lignites. Visually they are dark grey to medium grey in colour and their reflectance, in oil, is in the order to 0.2-0.3%; this is directly related to chemistry, particularly carbon content. Huminite macerals are considered to be reactive during conversion.

ii) The Liptinite Group: These are biochemically unaltered plant remains such as leaf cuticles, resins, spores, cork and algal remains which survive chemical breakdown in the peatification stage and are incorporated into a groundmass of detritial huminite. They are hydrogen-rich macerals whose names reflect their biological origins; they appear dark grey to brown in visible light, exhibiting low reflectance but because they are hydrogen-rich they fluoresce strongly in utltra-violet light. They are the most reactive of the coal macerals, readily degassing when heated (Fig. 1).

iii) The Inertinite Group: Comprises inert and semi-inert macerals, often referred to as 'fossil charcoal'. They are either introduced into the peat as charred remains of forest fires or result from dehydration and oxidation of cellulosic materials at the peat surface. The carbon content is considerably higher than the other macerals consequently they are highly reflecting, appearing white under the petrological microscope. Cellular structure is often retained in the case of fusinite or semi-fusinite and sclerotinite, (which is of fungal origin); macrinite and semi-macrinite are derived from gelified tissue. The semi-inert macerals are partially reactive whereas the totally-inert macerals do not change chemically during conversion.

Figure 2 illustrates comparative H/C : O/C ratios and Figure 3, the resulting differences in reflectance of bituminous coal macerals based on work by Van Krevelen (1961); no such work has been published on brown coals and lignites but if one produces the graphs in the direction of decreasing carbon content, this will give some indication of the chemical (and reflectance) difference between lignite macerals. The proportions in which the macerals occur in a particular coal has therefore a considerable influence upon the chemistry.

This paper is concerned with the effect of variation in coal composition upon gaseous yields based on the results from four experiments in which petrographically dissimilar lignites from Southern Saskatchewan were gasified under the same conditions.

Experimental

Samples:

Nine lignites from four mines in Southern Saskatchewan were analyzed petrographically; two samples from the Bienfait Mine, near Estevan and two from the Poplar River Mine, near Coronach were chosen for gasification because they were compositionally different. The petrographic compositions are represented graphically in Figure 4; the huminite group is sub-divided according to Cameron and Birmingham (1971). Details of the analyses are given in Table 2 and Table 3 shows the ratios of reactive : inert macerals.

Gasification:

Each of the four selected feedstocks; Coronach .1, Coronach .2, Bien 1B and Bien 2B were gasified in an engineering scale plant under the following conditions:

Temperature : 680°/700°C to 850°C
Pressure : 2mPa
Atmosphere : Air/Steam
Wt. of sample : 500 g.
Residence time : 1 hr. 50 mins. - 3 hrs.

While the conditions were the same for each experiment the residence times varied. Residence times for each process were as follows:

Sample No.	Experiment No.	Residence Time
Coronach No. 3	1400	2 hrs. 20 mins.
Bien 1B	1500	2 hrs.
Bien 2B	1600	3 hrs.
Coronach No. 1	1800	1 hr. 50 mins.

Results and Discussion

The composition of the gaseous mixture was in all four gasification experiments, a mixture of carbon monoxide, carbon dioxide, hydrogen and methane. The general pattern of gas production (Figure 5) was similar in all cases; carbon monoxide was given off in the middle stages while carbon dioxide and hydrogen production peaked at the beginning and at the end producing similar curves which are almost mirror images of the carbon monoxide curve. Methane production peaked at the onset of gasification and fell steadily throughout. The total yields of gas (and heating values) are shown in Table 4. The feedstock which produced the most hydrogen, and methane was the Coronach No. 1 sample, despite the fact that gasification was stopped after 1 hour 50 minutes. The Bien 1B sample produced the most carbon monoxide, (residence time: 2 hours). The greatest overall yield and heating value of the gaseous product came from the Bien 2B sample which was gasified for 3 hours. The Coronach No. 3 sample produced gas with a considerably lower heating value than the others despite a relatively long residence time of 2 hours 20 minutes.

It is interesting to compare the results of the gasification experiments with the petrographic composition of the feedstocks (Tables 2 and 4). The Coronach No. 3 lignite which contained almost 46% inertinite produced gas with a considerably lower heating value than the other products. The Coronach No. 1 sample which contained considerably more structured huminite (75%) than the other feedstocks produced the lowest volume of gas which nonetheless had a high cumulative heating value, in a considerably shorter time. The Bien 2B feed, which produced the largest volume of gas with the highest heating value contained approximately as much total huminite as the Coronach No. 1 but more reactive, hydrogen rich liptinites. It appears therefore that petrographic composition does indeed influence the composition of the gaseous products, yields and heating values; a lignite with a high structured huminite content requires a shorter time in the reactor to produce a gaseous mixture which may not be voluminous but has a high heating value. On the other hand a lignite with a high inertinite content produces gas of less heating value despite long residence times.

Analyses of the solid residues from gasification tend to support the inference that structured huminite is an important component of a gasification feedstock. The residues were all isotropic, vesicular chars composed of partially reacted, totally reacted and unreacted coal*, mineral matter and inertinite. Figure 6 shows the relative proportions of these components based on petrographic analyses; these values may vary according to the position of the coalin the reactor tube or with residence time hence they may not be significant; what is perhaps more important is the origin of the reacted, partially reacted and unreacted coal. On examining the individual char components it was found that the reacted component was dominantly derived from structured huminite and the partially reacted coal was dominantly derived from groundmass huminite (Table 5). This suggests that the more reactive huminite is indeed the structured portion (that part which retains cellular structure). The residue from gasification of the Coronach No. 1 lignite was particularly interesting. Unlike the others, this experiment was halted after 1 hour 50 minutes due to technical difficulties and a considerable portion of the residue was only partially reacted; some slightly altered, some reacted and some unaltered coal was present. All of the highly reacted coal was derived from structured huminite; only this portion showed any signs of vesiculation due to gas release. Furthermore, the partially reacted structured huminite contained highly reflecting* charred structures which were almost certainly cell walls suggesting that the macerals, textinite and textoulminite are among the earliest to respond and most reactive of the huminite group. In the partially reacted groundmass huminite-derived residue, charred spore-like and suberin-like (cork) structures were observed suggesting that liptinites are other maceral which react early on during gasification.

Similar features were observed on examination of residues from bench scale experiments, using the same feedstocks, which were stopped after two and four minutes and during which hydrogen, methane and carbon dioxide were evolved. This lends support to the suggestion that structured huminites and liptinites are among the first lignite components to react during gasification while gas of high heating value is being produced.

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*response to gasification is expressed as a degree of reactivity which corresponds to an increase in carbon content and loss of volatiles and is measured by the relative increase in reflectivity of the coal macerals (Blayden (1969), McCartney & Teichmuller (1972).

CONCLUSION

It can be concluded from the petrographic-gasification studies of Saskatchewan lignites that the composition of lignite has a considerable effect on its behaviour during gasification. Careful selection of a feedstock can increase the efficiency of the gasification process effecting an increase in the yield and heating value of the gas produced and a reduction in the residence time required.

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Table 1 - Summary of the macerals of brown coals

Group Maceral	Maceral Subgroup	Maceral	Submaceral*
Huminite	Humotelinite (Structural)	Textinite	
		Ulminite	Texto-Ulminite Eu-Ulminite
	Humodetrinite (Groundmass)	Attrinite	
		Densinite	
	Humocollinite (Unsaturated)	Gelinite	Porigelinite Levigelinite
		Corpohuminte	Phlobaphinite Pseudophlobaphinite
Liptinite		Sporinite	
		Cutinite	
		Resinite	
		Suberinite	
		Alginite	
		Liptodetrinite	
		Chlorophyllinite	
Inertinite		Fusinite	
		Semifusinite	
		Macrinite	
		Schlerotinite	
		Inertodetrinite	

* ICCP 1975

Terms in italics are alternative subgroup terms introduced by Cameron and Birmingham (1971)

Table 2 - Petrographic analysis of gasification feedstocks

Sample No.		Coronach 3	Bien 1B	Bien 2B	Coronach 1	Coronach 1
Maceral Analysis		(1400)	(1500)	(1600)	(1700)	(1800)
	Levigelinite	1.05	0.56	2.09	1.79	0.92
Huminite	Textinite	--	--	0.70	--	--
	Eu-Ulminite	30.55	45.88	46.05	58.93	70.05
	Texto-ulminite	1.05	11.54	10.82	3.13	5.07
	Densinite	14.74	12.10	25.47	18.75	11.98
	Corpohuminite	--	5.63	2.44	5.8	5.07
	Porigelinite	--	--	--	--	--
Liptinite	Sporinite	1.00	3.05	2.97	1.34	1.38
	Cutinite	--	0.68	0.33	--	--
	Resinite	--	1.69	0.33	0.45	0.46
	Suberinite	00	--	--	1.34	0.92
	Liptodetrinite	1.00	4.75	4.27	1.79	1.84
	Alginite	--	0.34	--	--	--
Inertinite	Fusinite	28.79	6.76	2.79	0.45	0.46
	Semifusinite	5.79	0.84	0.70	--	--
	Macrinite	1.05	--	--	--	--
	Schlerotinite	--	--	--	--	--
	Inertodetrinite	10.02	3.37	0.35	0.45	--
Mineral Matter		4.74	2.81	0.70	5.8	1.84
<u>Summary</u>						
Huminite	Structured	31.6	57.4	57.56	62.06	75.12
	Unstructured	1.05	6.19	4.53	7.59	5.99
	Groundmass	14.74	12.1	25.47	18.75	11.98
Liptinite		2.0	10.51	7.9	4.92	4.60
Inertinite		45.83	10.97	3.84	0.9	0.46
Mineral Matter		4.74	2.81	0.7	5.8	1.84

Table 3 - Selected feedstocks: Reactives : Inerts ratios

Feedstock	Total Reactives	Total Inerts	Reactives:* Inerts Ratio
Coronach No. 1	97.69	2.3	42.1 : 1
Coronach No. 3	50.75	48.67	1.0 : 1
Bien 1B	86.48	13.5	6.4 : 1
Bien 2B	95.69	4.3	22.3 : 1

* where reactives = total huminite + total liptinite + 1/3 semifusinite and inerts = total inerts - 1/3 semifusinite + mineral matter

Table 4 - Total gas and heating values obtained from gasification runs 1400-1800

Feedstock/Run (Residence time)	Cumulative Wt. of Gas (g)	Cumulative Heating Value (MJ)	Cumulative Heating Value after 1 hr 50 mins (MJ)	Time at which max. heating value attained
CORONACH NO. 3 Run 1400 (2 hr. 20 min.)	787.92	2.708	2.0	25 mins.
BIEN 1B Run 1500 (2 hr.)	942.48	4.17	3.9	15 mins.
BIEN 2B Run 1600 (3 hr.)	1667.14	5.963	4.2	15 mins.
CORONACH NO. 1 Run 1800 (1 hr. 50 min.)	506.44	3.90	3.9	20 mins.

TABLE 5 -- ENERGY RESEARCH SELECTED FEEDSTOCKS -- Gasified at Pilot Plant Level

Sample & char constituent	% reacted (recorded)	% partially reacted (recorded)	Total A + B	Reacted as % total $\frac{A}{A+B} \times 100$	Partially reacted as % total $\frac{B}{A+B} \times 100$	Ratio reacted structured/ reacted groundmass	Ratio Partially structured/partiall reacted groundmass
CORONACH #3/Run 1400							
structured huminite	1.5	17.8	19.3	7.8	92.2	3.9	0.94
groundmass huminite	0.5	24.8	25.3	2.0	98.0		
BIEN 1B/Run 1500							
structured huminite	62.6	6.9	69.5	90.1	9.9	1.3	0.32
groundmass huminite	13.2	5.9	19.1	69.1	30.9		
BIEN 2B/Run 1600							
structured huminite	28.4	22.9	51.3	55.4	44.6	1.7	0.65
groundmass huminite	9.6	20.6	30.2	31.8	68.2		
CORONACH #1/Run 1800							
structured huminite	26.6	40.9	67.5	39.4	60.6	11.9	0.63
groundmass huminite	0.8	23.6	24.4	3.3	96.7		

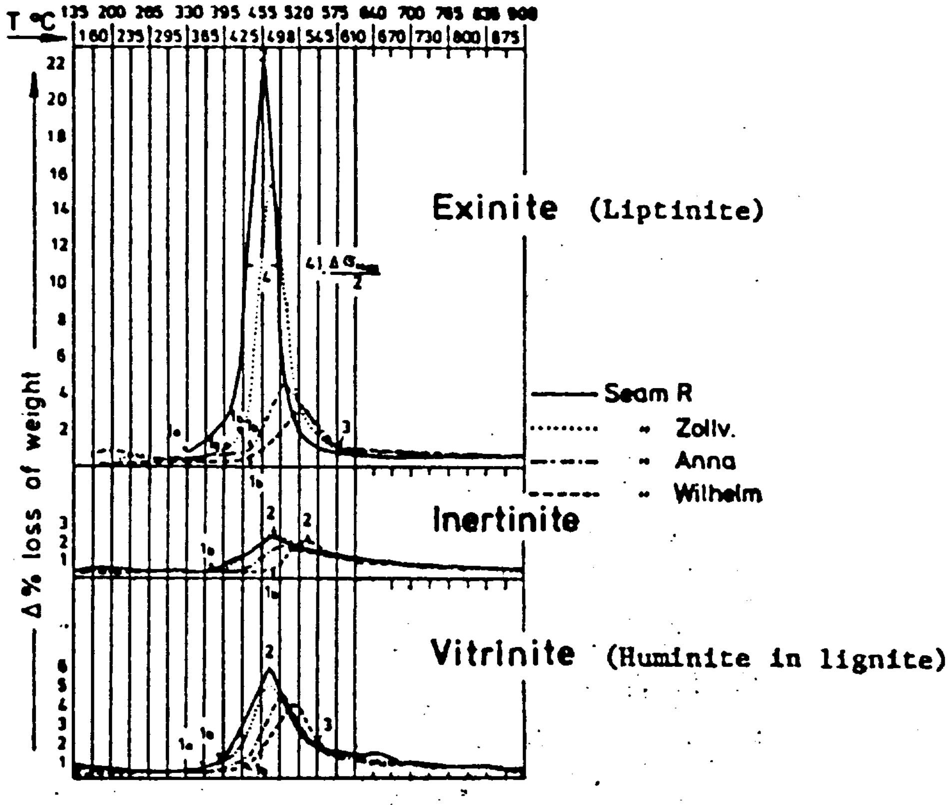


Figure 1. Degasification velocity of the three macerals as a function of their rank, from Kröger, 1958, modified. The maximum rate of decomposition is shifted towards higher temperatures with increasing rank, and the height of the maximum decreases with rising rank.

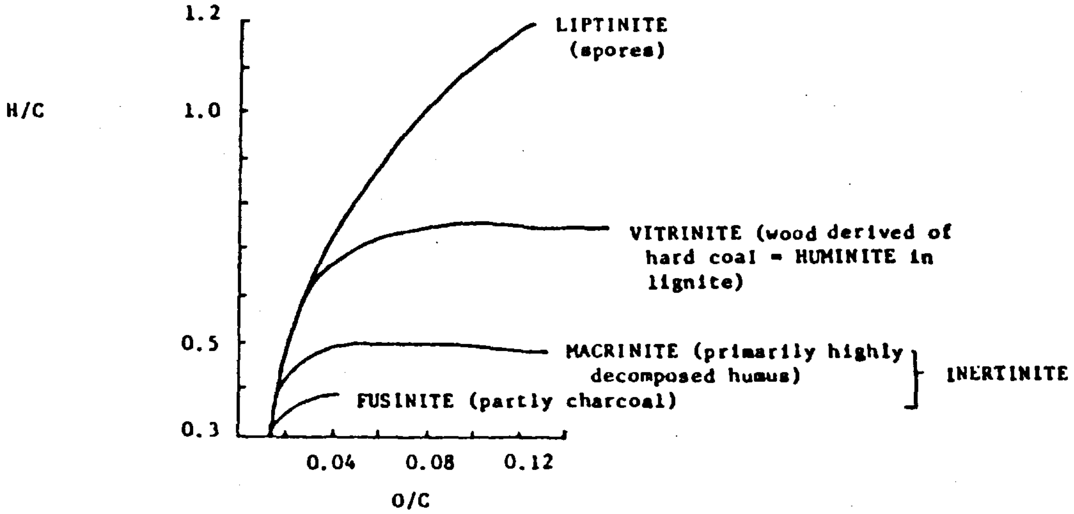


Figure 2: Coalification tracks of different macerals based on H/C : O/C atomic ratios (after Van Krevelen, 1961, modified).

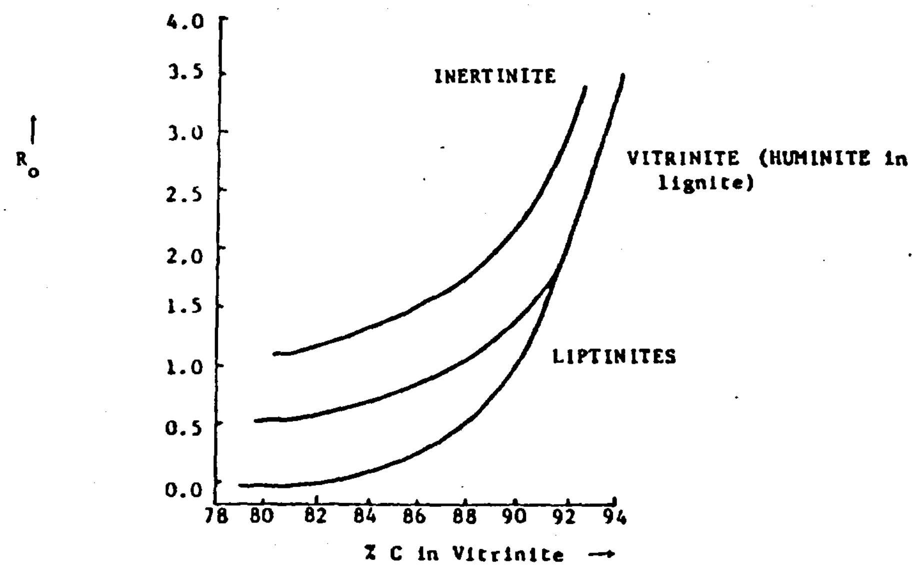


Figure 3: Reflectance of macerals (oil immersion) (after Van Krevelen, 1961, modified).

FIGURE 4 ANALYSIS OF FEEDSTOCKS

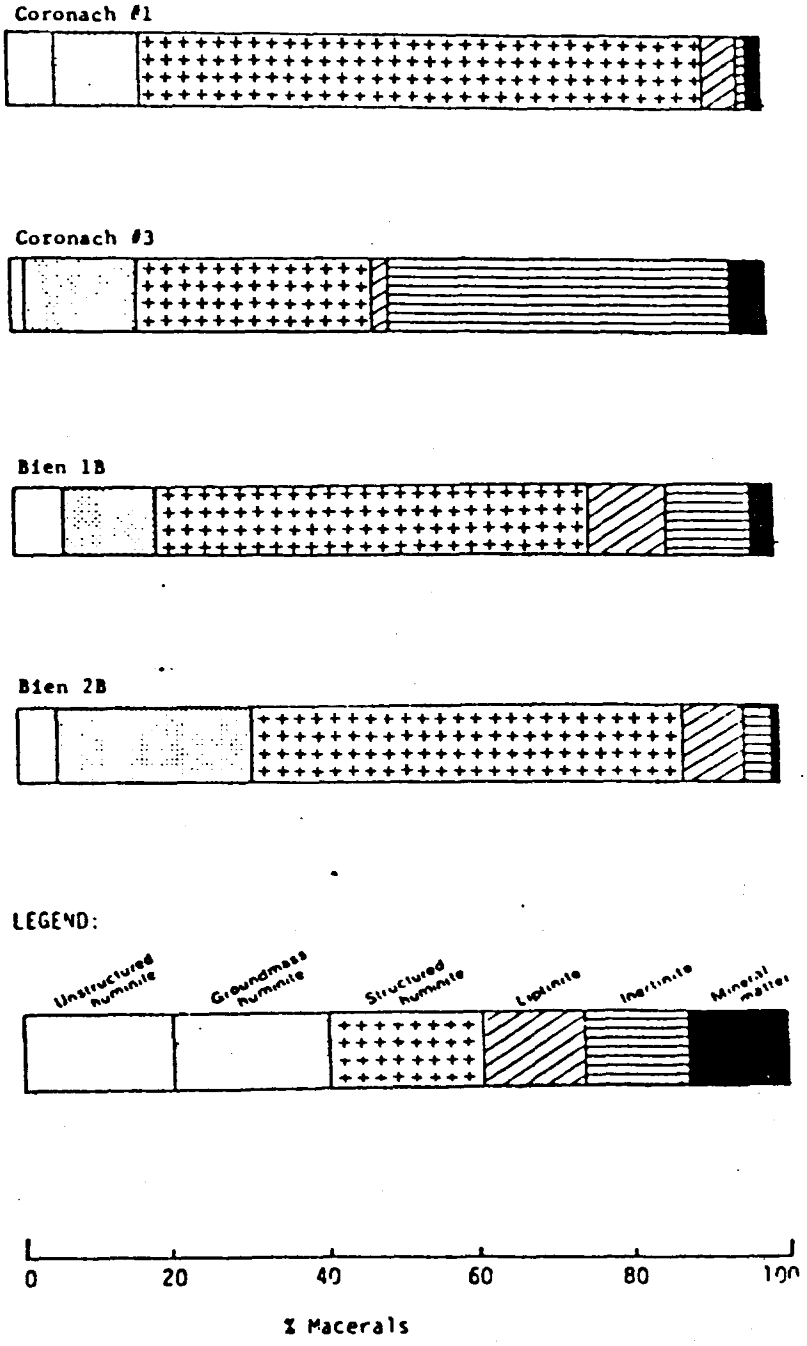


Figure 5: Plots of gases realeased during gasification of selected lignite feedstocks

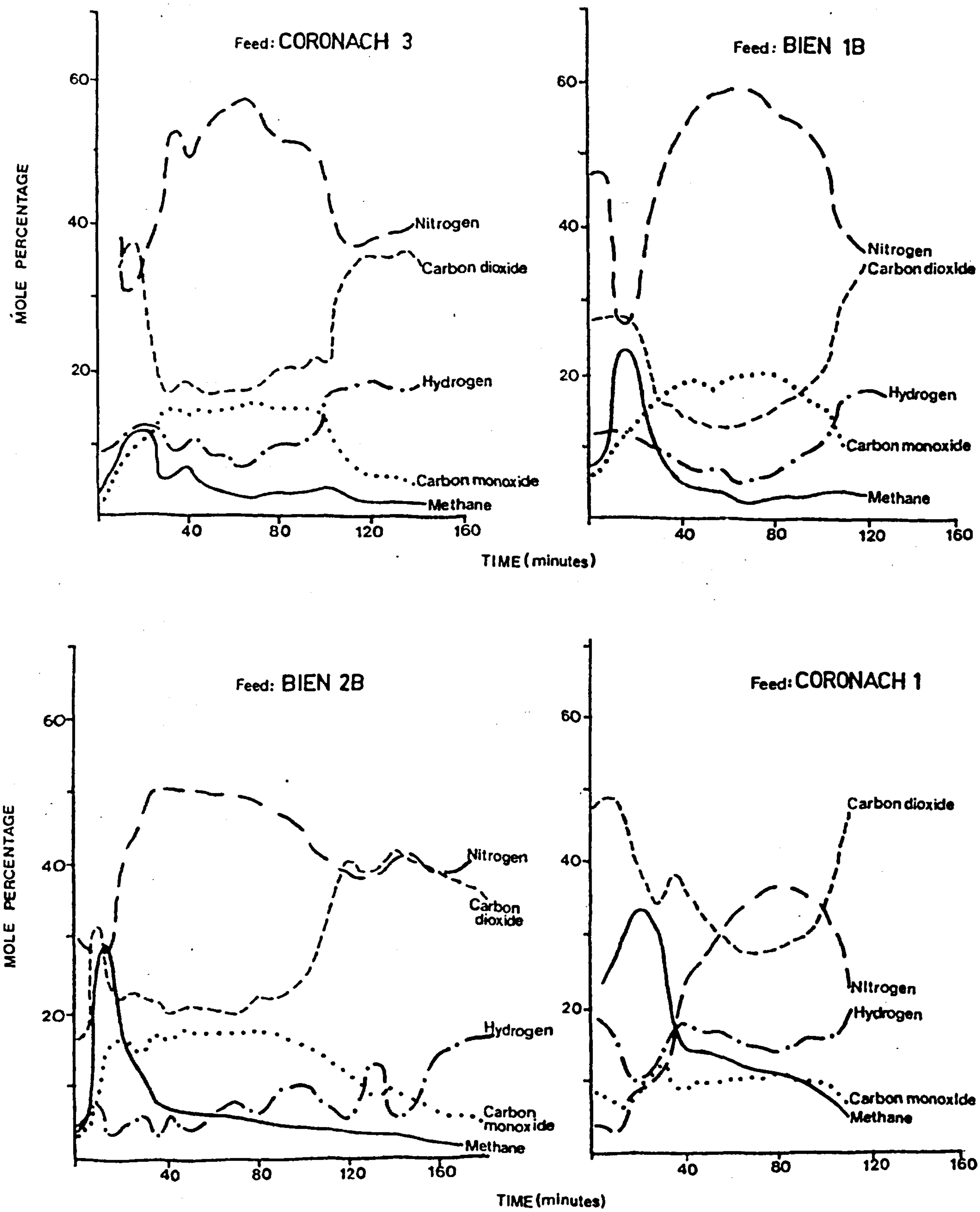
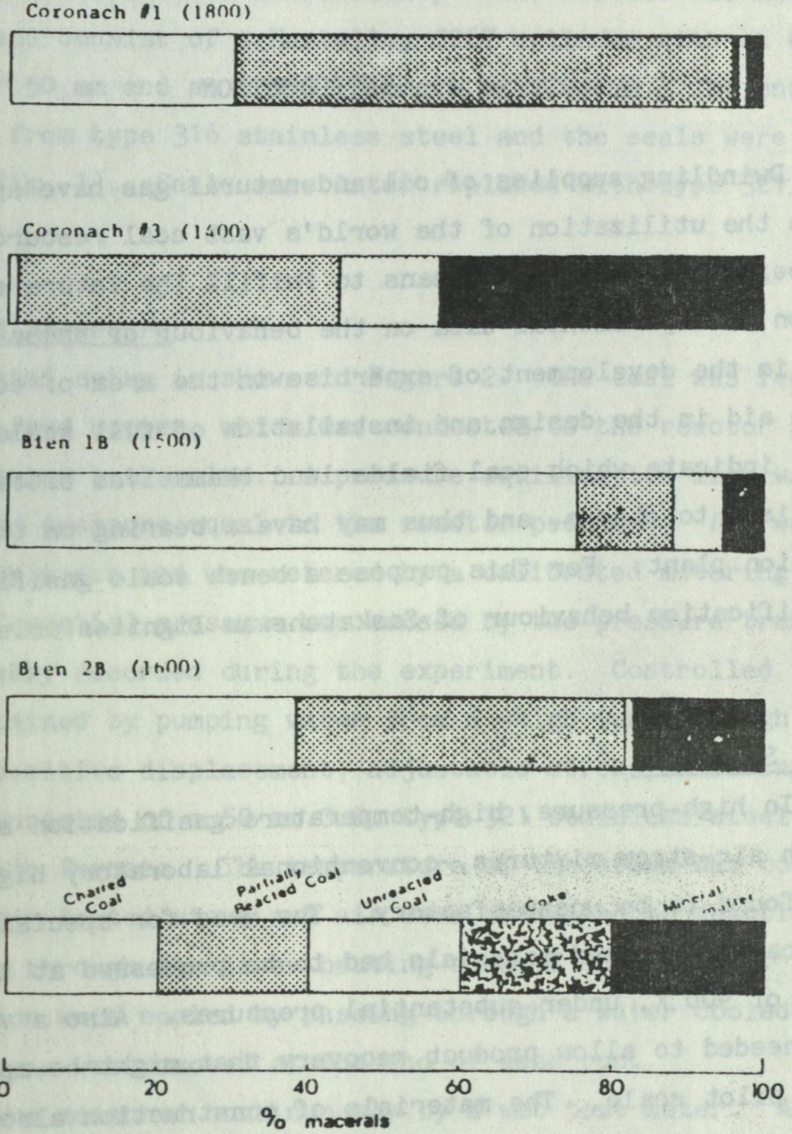


FIGURE 6

ANALYSES OF CHARS



Lignite Reactivities: Experimental Equipment

by

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INTRODUCTION

Dwindling supplies of oil and natural gas have spawned renewed interest in the utilization of the world's vast coal resources. Coal conversion is being looked at as a means to fulfill the future energy needs. The collection of experimental data on the behaviour of these coals not only will aid in the development of expertise in the area of coal conversion but also will aid in the design and installation of full scale gasifiers. The data will indicate which coal fields lend themselves better to gasification in comparison to others, and thus may have a bearing on the location of a gasification plant. For this purpose a bench scale gasifier was built to study gasification behaviour of Saskatchewan lignites using air-steam mixtures.

Equipment Selection

In high-pressure, high-temperature gasification studies of lignite coals with air-steam mixtures, conventional laboratory high pressure equipment was found to be unsatisfactory. The need for special techniques arose mainly because the feed materials had to be processed at temperatures much in excess of 900 K, under substantial pressures. Also a vessel of reasonable size was needed to allow product recovery that might be approached by bench and small pilot scale. The materials of construction also had to be strong enough at high temperature to withstand several mega Pascals of pressure. They also had to resist scaling and hydrogen attack. End closures had to be of a type that would maintain a seal at higher temperatures, but yet could be opened and closed with ease for frequent use.

Many commercial alloys are available which meet the principal criterion of adequate strength at high temperatures, however, when machineability and availability are being considered the suitable alloys become quite limited. Although molybdenum alloys are generally the strongest, they lack oxidation resistance. The nickel-base and cobalt-base alloys show sufficient strength but the machinability is generally difficult. Although Multimet N-155 alloy and Haynes Stellite alloy 25 were chosen initially, Hastelloy C267 was finally selected because of availability. The reactor was manufactured by PEMCO, Utah and consist of a Hastelloy C267 cylinder with an O.D. of 100 mm, and I.D. of 50 mm and an overall length of 1700 mm. The end closures were constructed from type 316 stainless steel and the seals were type 304 stainless steel (Fig. 1). Seals were later replaced with type 321 stainless steel.

Experimental setup and procedure

The experimental setup is shown in Figure 2. The coal was fed in single batches from a feed hopper, which was connected to the reactor by an air-operated quick opening ball valve. A pressure equilization line was used to keep the hopper pressure equal to the reactor pressure. Air was fed from a high pressure cylinder and was metered by a calibrated metering valve. The valve orifice differential pressure was sensed by two pressure transducers and was automatically recorded during the experiment. Controlled rates of steam feed were obtained by pumping water from a weigh tank through a steam generator by a positive displacement, adjustable-stroke, metering pump. The steam generator consisted of a 50 mm O.D. type 321 stainless steel tube contained in an electric furnace. The temperature of the steam was controlled manually. Super heated steam and air were preheated to reaction temperature in passing through the lower heating zone of the reactor. Gases exiting from the reactor were cooled by passing through a water cooled vessel. The condensed steam was removed at the end of each run.

Exit gases were measured continuously by a wet test meter. A small portion of the exit gas stream was stored in gas sampling bottles to be used for analysis. To avoid distortion because of backmixing and gas holdup, which would occur if the gas samples weere taken after gas metering, the gas sampling manifold was installed in the exit line immediately after the reactor. The gas samples were filtered and cooled in an inline condensor before being reduced in pressure via a normal pressure regulator.

The reactor is shown in Figure 1. The reactor tube was electrically heated by a three zone furnace. The reactor temperatures were controlled by three proportional indicating temperature controllers and three thermocouples located within the centres of the heating elements. The temperature within the reactor was measured by a single, chromel-alumel, 304 sheated thermocouple inserted in a type 316 stainless steel thermo-well.

Gas analysis were performed using a Hewlett Packard 5710 A gas chromatograph equipped with a thermo conductivity detector. The standard natural gas package was used which consists of a DC 200 silicon rubber column, a Porapack Q lcolumn and a molsieve 519 column. Part of the gas analysis were performed using a Fisher model 1200 gas partitioner.

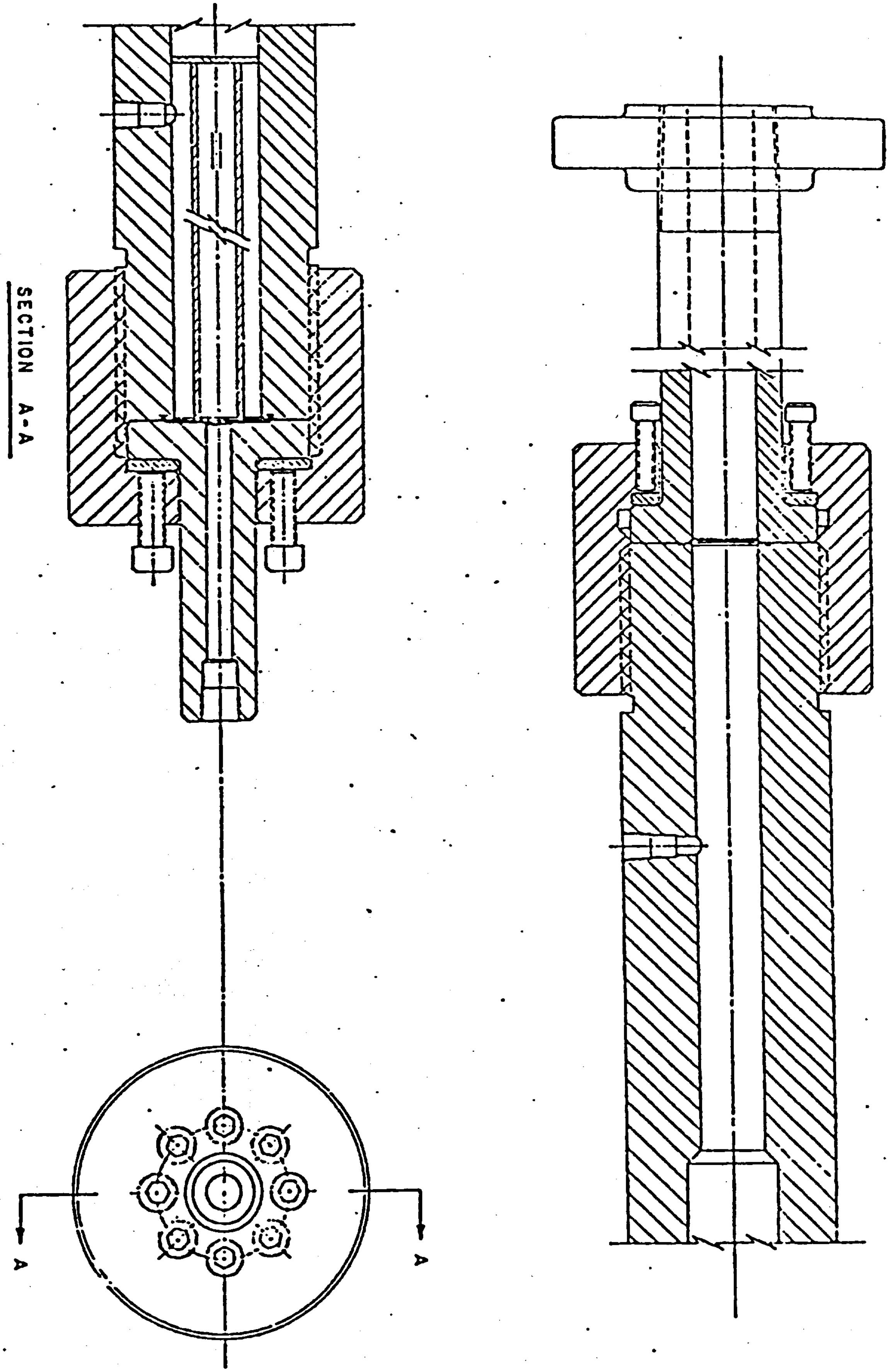
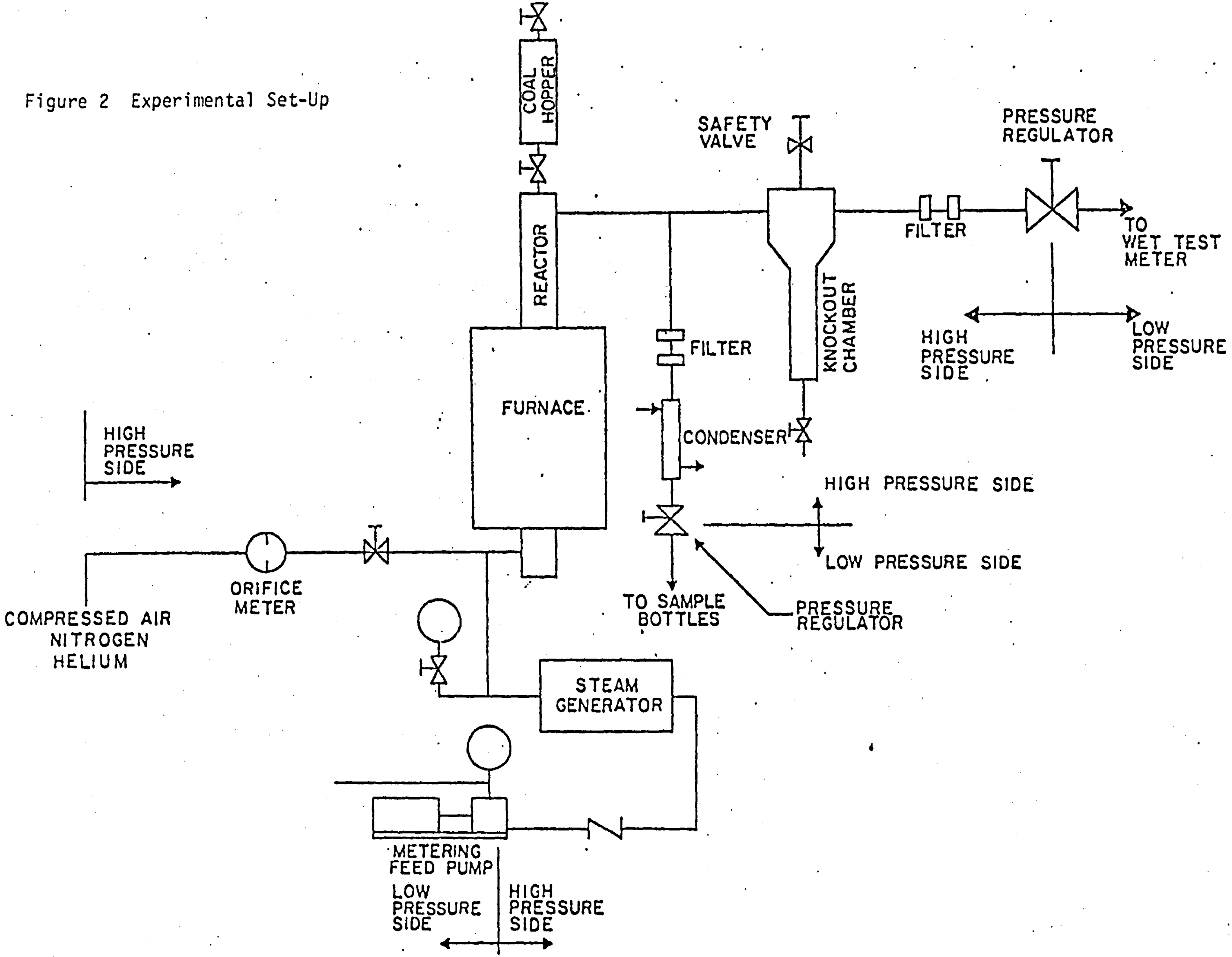


FIGURE 1. High Pressure Reactor; End Closures

Figure 2 Experimental Set-Up



Lignite Reactivities: Results and Interpretation

by

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INTRODUCTION

There are substantial coal reserves in the southern part of Saskatchewan and in the future these coal resources will play an increasingly important role in meeting the energy requirements of the province. Conversion of Saskatchewan's coal to other form of energy, such as industrial utilization of medium thermal content gas and substitute natural gas is one of the areas being closely examined by the Saskatchewan Power Corporation. Advanced electric power generation techniques in the form of combined cycle generating plants using coal gasification, are being studied with a view to reducing water requirements. Not enough ground or surface water is available in the vicinity of some of the coal fields and this combined cycle process can lessen the thermal impact on air and water surrounding the electrical generating station.

Gasification is a method to convert solid carbonaceous materials, such as coal or lignite, into a combustible gas. The operation of a particular gasifier will depend on its design relative to the fuels used, and depends in particular on the fuel density, moisture content, ash fusing temperature, etc. The gasification process with steam is highly endothermic and the heat requirements can be supplied in various ways. The method of supplying heat for the performance of reactions is an important item and has an influence on the process and its economics. There are several means of supplying heat both directly and indirectly. The most common direct method is by partial combustion of the fuel with oxygen or air. The temperature is generally controlled by regulating the steam to oxygen/air molar feed ratio.

Saskatchewan lignite coals from several coal fields were used to determine the gasification characteristics under high pressure using varying steam/air ratio's in an externally heated reactor.

Procedure

In all tests, a semiflow technique employing a flowing gas and a single, static coal charge was used. Test periods varied from 1 to 3 hours. The reactor was heated to operating temperature before pressurizing it. When the reactor had reached the desired run temperature, the reactor pressure was brought to the desired level with helium. The design and operation of the reactor have been fully described (1).

The tests were started by opening the quick opening ball valve between the hopper and the reactor, and starting the steam feed pump and air supply. Single charges of 500 g of coal were fed at reactant gas flow rates of 4 - 15 L/min for air and 2.2 - 12.6 g/min for steam. The coal charge was supported on top of a perforated carbon steel disc. The temperature of the coal charge 10 cm above the perforated disc, as well as the centres of the top, central and bottom heating zones were recorded. Exit gas samples were taken at regular intervals. After approximately 1 m^3 of gas were produced the run was stopped, the reactor heaters turned off and the unit was depressurized to minimize further reaction of the coal charge after the run. The gas analysis were performed by gas chromatograph.

Results

The assumption was made that nitrogen behaves as an inert gas and does not react with coal, and that the amount of nitrogen contributed by the coal is negligible. The total number of moles of nitrogen introduced into the reactor over a given time interval, therefore, must equal the number of moles of nitrogen in the product gas over the same time period. The nitrogen thus acts as a "tracer" gas. The total number of moles for each gaseous component in the product gas was calculated by taking the average gas composition during a certain time interval equal to the midpoint between the concentrations of each component as determined at the beginning and end of that time interval. The time intervals were chosen to be the time durations between sampling points.

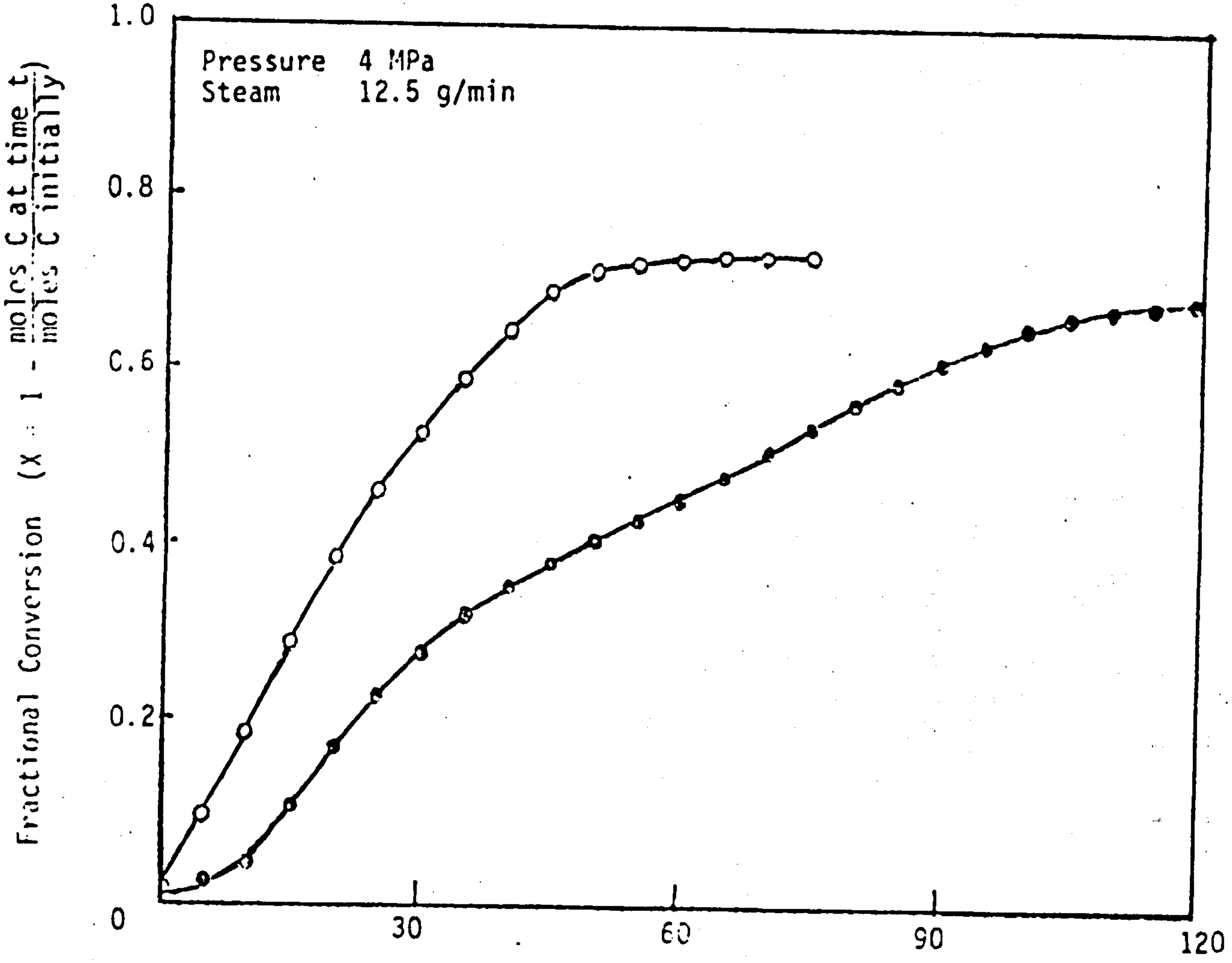
A typical curve for the fractional conversion of carbon as a function of time is shown in Figure 1. The fractional conversion is defined as $X = 1 - (\text{moles C at time t/moles C initially})$. The curve shows high initial reaction rates as the coal is pyrolysed, followed by a much slower reaction regime where the rate slowly diminishes as the remaining char is gasified.

During the inital stages of the reaction, the main components of the gases are CO_2 and CH_4. While the CO and H_2 concentrations shown only a minor change after the initial devolatilization the gas composition remains almost constant until the char is exhausted and this is shown by a rapid increase in the O_2 concentration (Figures 2 and 3). The average CH_4 concentration increases with the increasing pressure (Figure 4).

Failure to reach 100 percent conversion can be mainly attributed to entrainment of the char/ash in the reactor gas stream. These losses were verified by ashing the residual char and ash left after the experiment. The yield of ash obtained was typical 60 - 80 percent of the amount which should have been recovered on the basis of proximate analysis of the original coal sample. "Losses" tended to be lower for identical air flow rates at higher pressures as a result of reduced axial velocity in the reactor (Figure 5).

The average heating value calculated from the gas composition over the duration of the run shows an increase in pressure. This is mainly due to the increase in the formation of methane (Figure 6). When converted to a nitrogenfree basis the methane composition seems to increase while the H_2 decreases two fold suggesting that part of the hydrogen was used to form the methane (Figure 4). Modification of the equipment are being made to allow for several hours of operating using continuous coal feed.

Figure 1 Fractional Conversion of Carbon Using Different Air Flow Rates



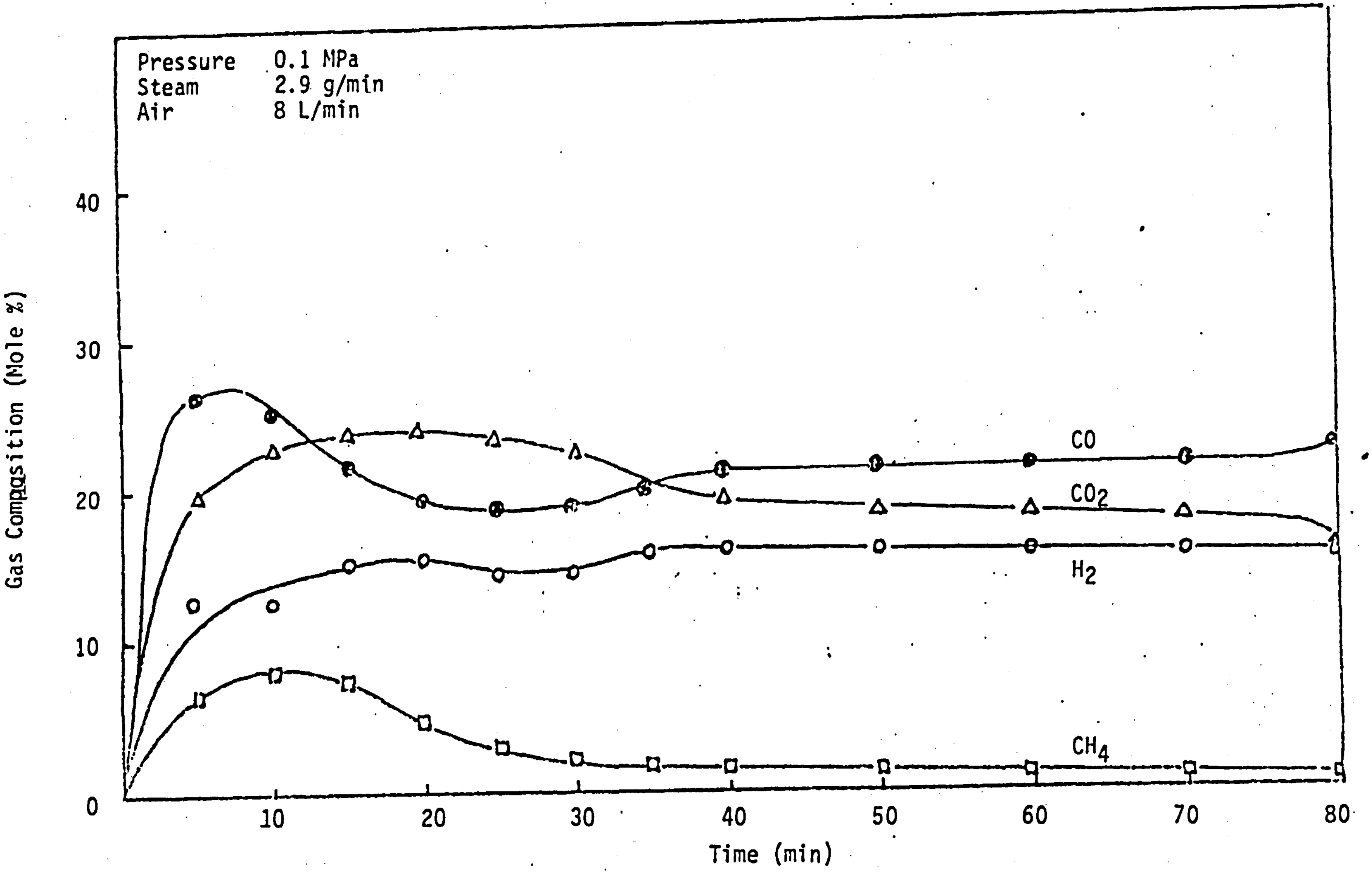


Figure 2 Typical Gas Composition Variation During a Run

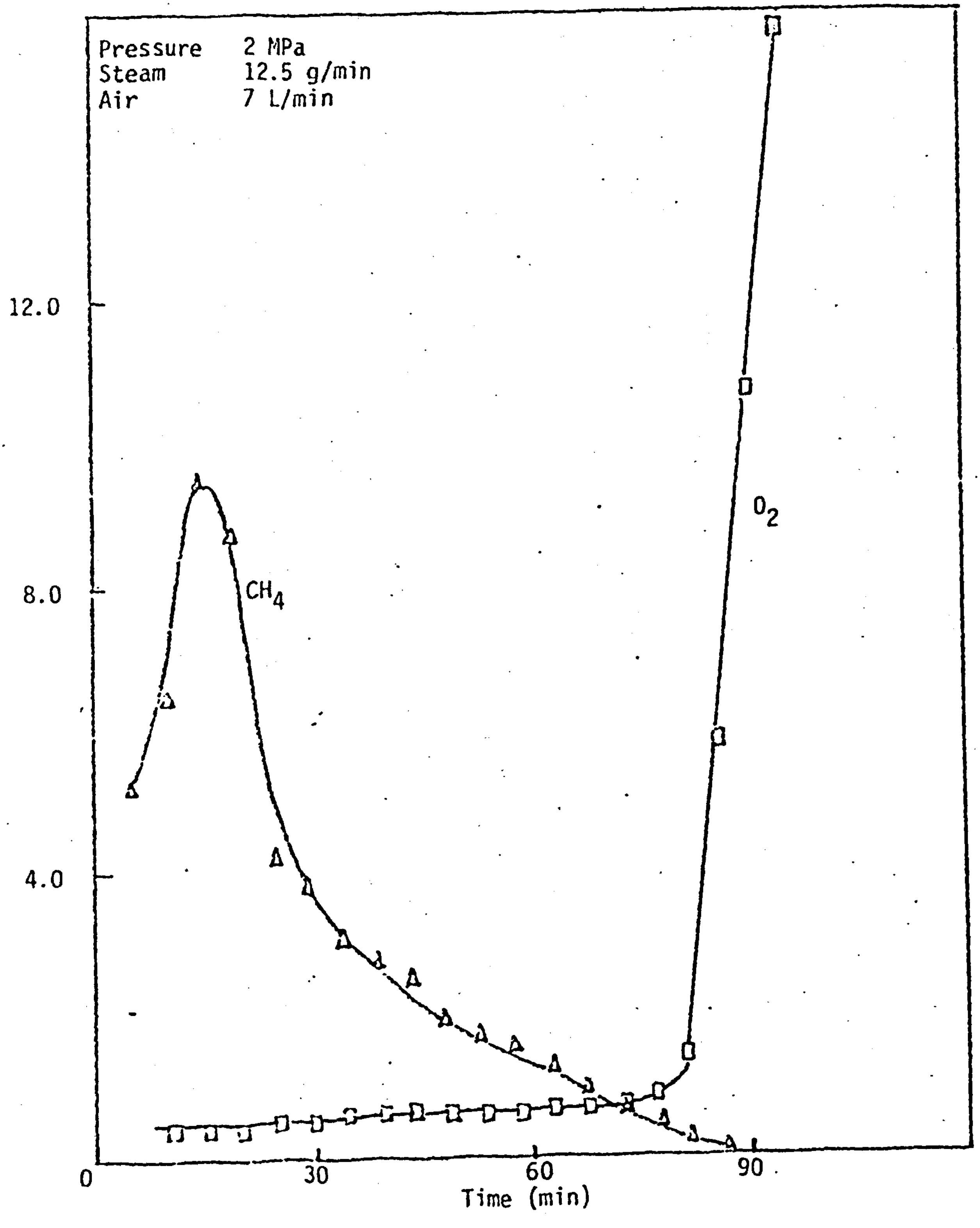
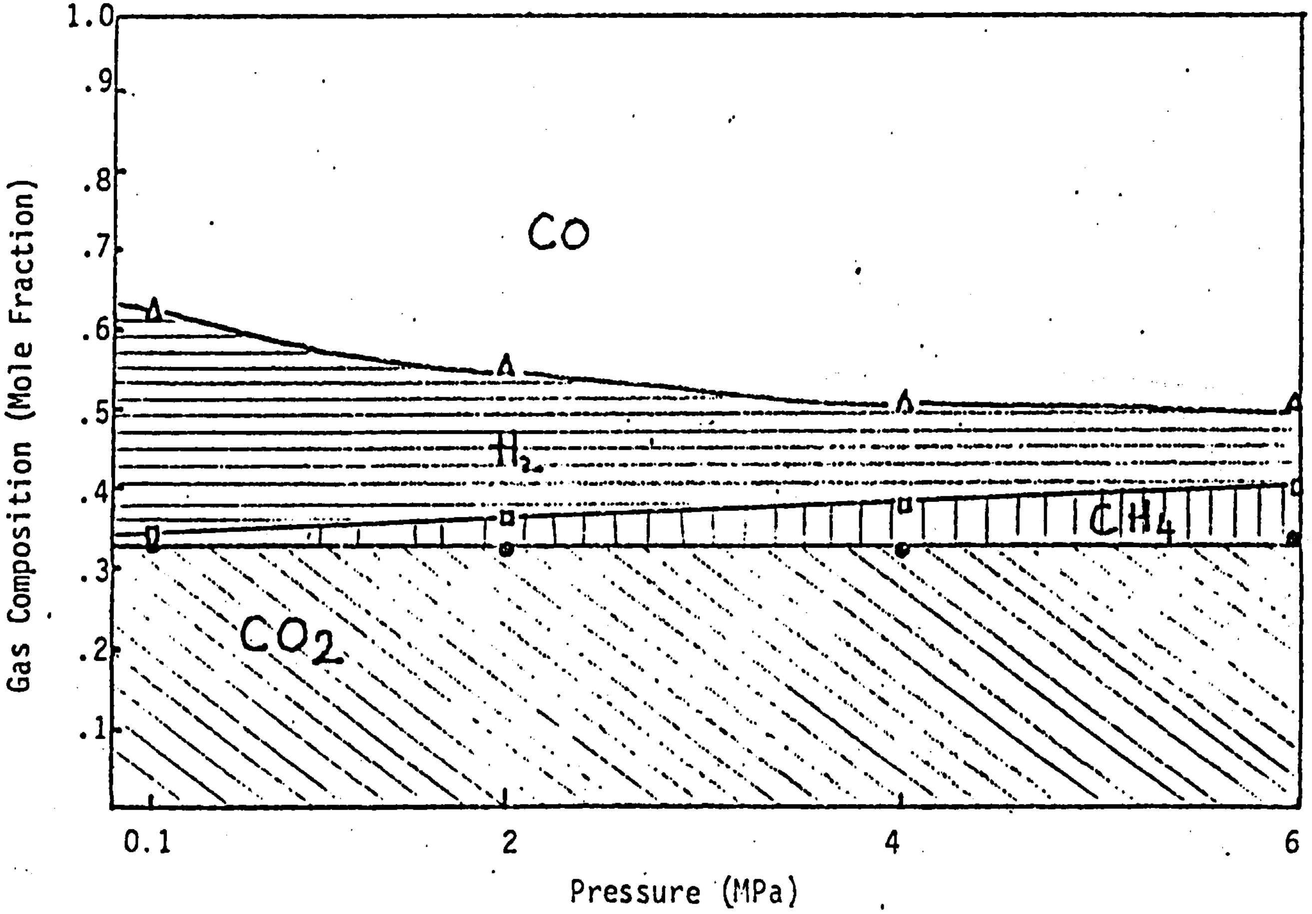


Figure 3 Methane and Oxygen Concentration in Product Gas

Figure 4. Dependence of Gas Composition on Pressure (Nitrogen Free Basis)



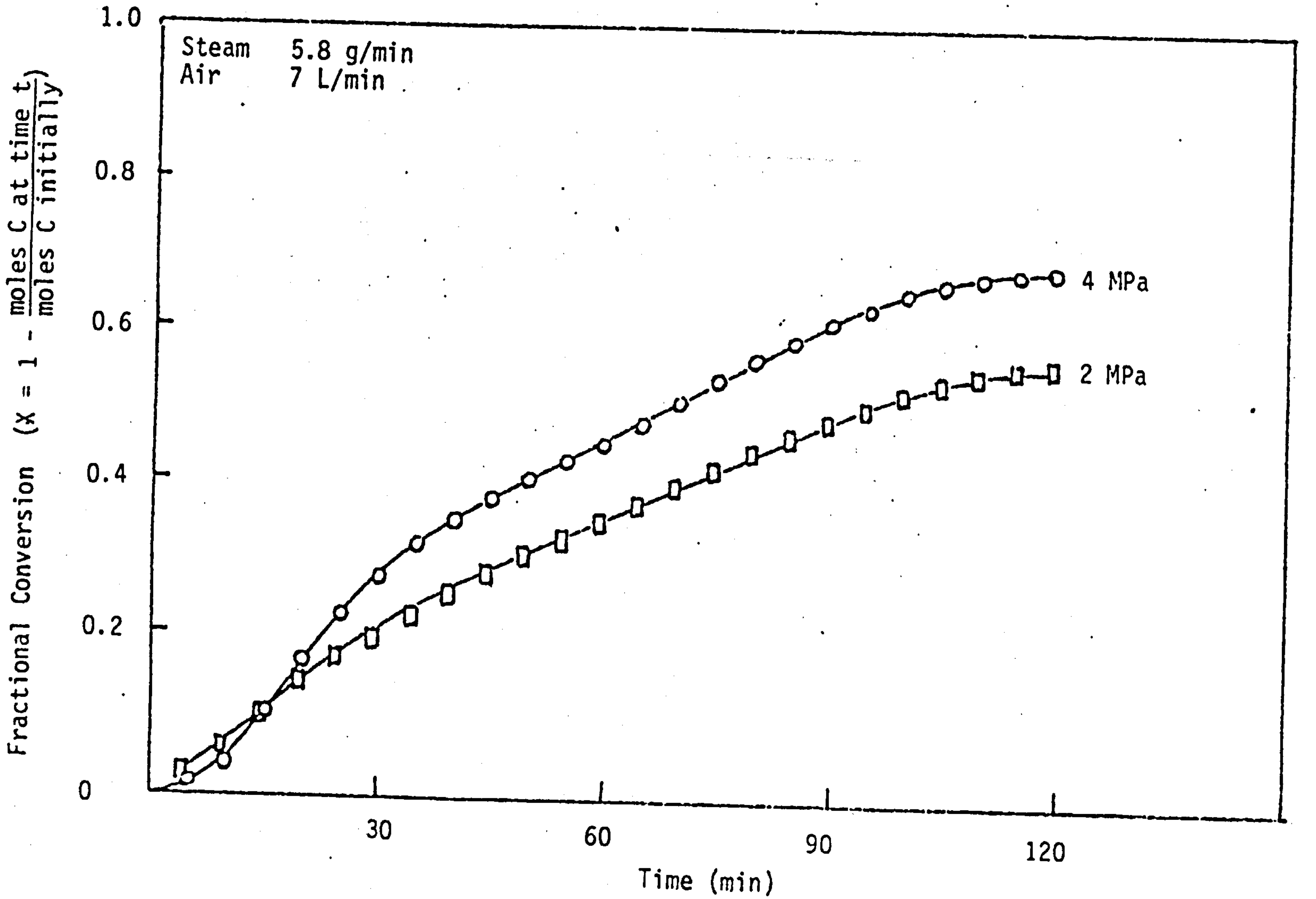


Figure 5 Fractional Conversion of Carbon at Different Pressures

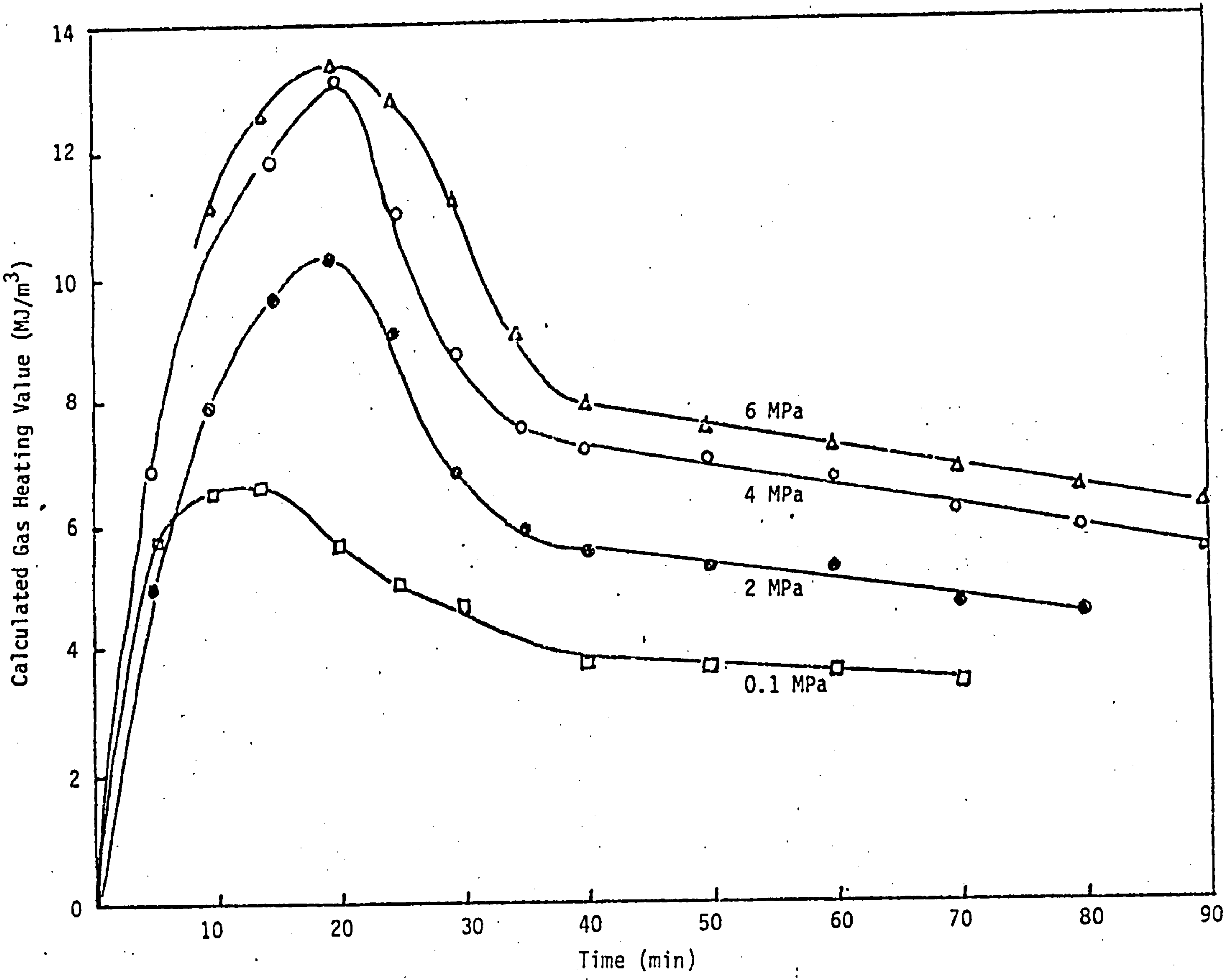


Figure 6 Average Gas Heating Values at Various Pressures