GASIFICATION OF CAKING COAL IN A FREE-FALL, FLUID-BED REACTOR

A. J. Forney, R. F. Kenny, and J. H. Field

Pittsburgh Coal Research Center, U. S. Bureau of Mines,
4800 Forbes Avenue, Pittsburgh, Pennsylvania 15213

INTRODUCTION

The Bureau of Mines is investigating the gasification of caking coals in a fluid-bed reactor as part of the Bureau's overall program of converting coal to liquid or gaseous fuels. The caking coals, common to the East and Midwest, cannot be gasified without being pretreated, usually with steam and oxygen. The method of gasification discussed in this report is free-fall pretreatment combined with fluid-bed gasification.

The primary objectives of the project are: 1) To check our earlier method of pretreating caking coals before gasification;[2, 3] 2) to determine the minimum amount of oxygen needed for pretreatment; 3) to maximize the methane content in the product gas. A secondary aim, which developed from the above tests, is to study a method of substituting air for oxygen.

EXPERIMENTAL PROCEDURE

The pretreatment is achieved by dropping Pittsburgh-seam coal (70 percent through 200 mesh) through a free-fall reactor 6 inches in diameter and 10 feet high (later reduced to 7 feet). This pretreater is located above a fluid-bed reactor 3 inches in diameter and 3 feet high (later increased to 6 feet). The oxygen and steam for the gasification enter the bottom of the reactor, figures 1 and 2. The gases from the gasifier flow up through the free-fall section to pretreat and carbonize the coal as it falls. They are enriched with an oxygen-steam feed entering the side of the free-fall section to further pretreat the coal because the gases rising from the gasifier did not pretreat sufficiently to prevent agglomeration.

Pressures from 2.5 to 20 atmospheres, temperatures of 835° to 955° C, and oxygen-coal ratios from 0 to 5.9 were used.

RESULTS AND DISCUSSION

Pressure Effects

The pressure was gradually increased from 2-1/2 to 20 atmospheres as shown in table 1 and figure 3. The methane content gradually increased to 14 volume percent of the product gas. This means that more methane can be produced in this gasification than would be produced in subsequent methanation to produce high-Btu gas. For example, in experiment N-12 the 50 percent H_2+CO would yield less methane during methanation than the 14 percent methane already produced in gasification. As the pressure was increased from 2-1/2 to 20 atmospheres, figure 3, the percentage of methane in the product gas increased from 8 to 12, while the hydrogen and carbon monoxide percentages decreased. Also the carbon dioxide increased while the hydrogen and carbon monoxide decreased at the same rate, verifying Schuster's[8] claim that the methane-making reaction in gasification is $2H_2 + 2CO \rightarrow CH_4 + CO_2$. These curves, figure 3, based on average values from several experiments are similar to those of O'Dell.[4]

Table 1.- Effect of Pressure on Methane Yield

Test No.[1]	F-19	F-33	F-46	N-10	N-12	N-13	N-17
Pressure, atm	2.5	5	10	15	20	20	20
Coal[2]	D-2	D-4	D-4	D-4	D-4	D-4	D-4
Coal feed, lb/hr	0.43	0.70	1.53	1.60	1.60	1.63	3.40
Input, SCFH:							
Gasifier, steam	18	25	50	60	60	60	100
Gasifier, oxygen	1	1.5	4	4	4	4	16
Pretreater, steam	10	10	20	30	30	20	20
Pretreater, oxygen	1	1.5	3	4	4	4	4
Nitrogen	4	4	8	6	6	5	5
Temperature, °C:							
Pretreater	375	409	390	375	400	400	400
Gasifier, avg	893	891	893	880	882	876	883
Gasifier, max	900	900	900	900	900	900	900
O_2/coal, SCF/lb	4.6	4.3	4.6	5.0	5.0	4.9	5.9
Steam/coal, SCF/lb	42	36	33	37	37	37	29
Carbon conversion, pct	67	68	60	66	65	73	68
Steam conversion, pct	12	10	15	17	14	23	--
Product gas,[3] SCF/lb	19	19	16	17	15	18	16
Methane, SCF/lb	2.1	1.8	2.5	2.7	3.3	3.7	3.4
Product gas, pct:							
H_2	44	41	39	39	34	36	35
CH_4	8	7	11	11	14	14	14
CO	21	25	22	20	16	18	18
CO_2	27	27	28	30	36	32	33
Tar, pct of coal feed	10	6	3	3	4	3	5

1/ F series with 3-foot reactor; N series with 6-foot reactor.
2/ D-2: H 5.1, C 72.9, N 1.4, O 8.2, S 1.3, Ash 11.1 percent.
D-4: H 5.1, C 76.5, N 1.5, O 8.1, S 1.0, Ash 8.0, VM 34.8 percent; FSI = 7-1/2.
3/ $H_2+CO+CH_4$.

Tar Plus Oil

As the pressure increased the tar yield decreased from 10 to about 5 percent. The tars were analyzed by chromatography to find the effect of pressure on the composition of the tar. Results were inconclusive; no correlation could be made.

The coal feed rate was increased from 1.54 pounds per hour (33 lb/hr ft^2) to 6.1 pounds per hour (133 lb/hr ft^2) at 20 atmospheres pressure (table 2). The carbon conversion was lower at the higher coal rate. The methane yield was almost steady at 3 SCF/lb, although its percentage in the product gas increased from 12 to 15 percent.

Table 2.- Effect of Increased Coal Feed on Product Distribution at 20 Atmospheres Pressure

Test No.	N-11	N-15	N-17	N-19	N-21	N-24	N-22	Lurgi[1/]
Coal[2/]	D-4	D-4	D-4	D-5	D-5	D-5	D-5	
Feed, lb/hr	1.54	2.25	3.40	5.40	5.82	6.1	5.2	
Coal feed, lb/hr ft^2	33	49	74	120	127	133	113	
Coal feed, lb/hr ft^3	6	8	12	20	21	22	18	
Input, SCFH:								
Gasifier, steam	60	65	100	150	180	180	198	
Gasifier, oxygen	4	6	16	24	22	18	6	
Pretreater, steam	30	15	20	24	36	36	36	
Pretreater, oxygen	4	3	4	6	6	6	6	
Nitrogen	6	6	5	5	5	5	5	
Temperature, °C:								
Pretreater	375	400	400	400	400	400	400	
Gasifier, avg	889	885	883	907	864	890	889	
Gasifier, max	900	898	900	959	890	910	900	
O_2/coal, SCF/lb	5.2	4.0	5.9	5.6	4.8	4.0	2.3	4
Steam/coal, SCF/lb	39	29	29	28	31	30	38	19
Carbon conversion, pct	73	68	68	75	59	55	52	90[3/]
Steam conversion, pct	14	28	21	29	12	12	15	
Product gas,[4/] SCF/lb	19	19	16	16	13	12	15	19
Methane, SCF/lb	3.2	3.3	3.4	3.2	3.0	2.7	3.2	2.6
Product gas, pct:								
H_2	38	39	35	33	33	34	40	40
CH_4	12	13	14	13	15	15	17	10
CO	21	23	18	19	16	18	18	25
CO_2	29	25	33	35	36	33	25	25
Tar, pct of feed	4	4	5	9	4	3	3	

1/ Westfield plant coal: Moist. = 15.6, ash = 14.6, VM = 28.7, FC = 41.1 pct.
2/ D-4: H 5.1, C 76.5, N 1.4, O 8.1, S 1.0, Ash 8.0, VM 34.8 pct; FSI = 7-1/2.
D-5: H 5.0, C 74.9, N 1.5, O 7.6, S 1.1, Ash 9.9, VM 34.7 pct; FSI = 8-1/2.
3/ Estimated.
4/ H_2+CO+CH_4.

The results of N-23 (table 3) and N-19 (table 2) may be compared with those of the Westfield[7/] Lurgi. Our methane percentage is higher, and our methane yield slightly higher. We used more oxygen and more steam and had a lower carbon conversion; however, the Lurgi could not be operated with the caking coal used in our tests.

Effect of Low Oxygen Feed

In test N-22 the oxygen feed to the gasifier was reduced from the 4 to 5 cubic feet per pound used in the other tests to 1 cubic foot per pound. Comparing test N-22 with N-19 shows that the carbon conversion decreased, the methane yield remained the same, but the percentage of methane in the product gas increased from 13 to 17 percent (table 2). This increase is desirable, but a commercial gasifier would have to be heated externally to operate with such a low oxygen feed.

Table 3.- Effect of Temperature on Product Distribution at 20 Atmospheres Pressure

Test No.	N-26	N-25	N-24	N-27	N-23	N-28	N-30
Coal[1/]	D-6	D-6	D-5	D-6	D-6	D-6	D-6
Coal feed, lb/hr ft^2	123	136	133	140	133	140	136
Feed, lb/hr	5.65	6.25	6.1	6.5	6.0	6.4	6.3
Input, SCFH:							
Gasifier, steam	180	180	180	180	180	180	120
Gasifier, oxygen	18	18	18	18	24	18	18
Pretreater, steam	36	36	36	36	36	36	36
Pretreater, oxygen	6	6	6	6	6	6	6
Nitrogen	5	5	5	5	5	5	5
Temperature, °C:							
Pretreater	400	400	400	400	400	400	400
Gasifier, avg	835	860	890	880	900	901	912
Gasifier, max	857	880	910	910	955	934	963
Oxygen/coal, SCF/lb	4.3	3.8	4.0	3.7	5.0	3.8	3.8
Steam/coal, SCF/lb	32	29	30	28	30	28	19
Carbon conversion, pct	51	58	55	59	81	65	56
Steam conversion, pct	4	--	12	--	24	--	--
Product gas,[2/] SCF/lb	11	14	12	14	19	16	13
Methane, SCF/lb	2.5	2.9	2.7	3.5	3.6	3.2	2.9
Product gas, pct:							
H_2	31	36	34	34	36	36	33
CH_4	15	14	15	17	13	14	16
CO	20	17	18	17	20	20	18
CO_2	34	33	33	32	31	30	33
Tar, pct of feed	7	8	3	4	3	5	4

1/ D-5: H 5.0, C 74.9, N 1.5, O 7.6, S 1.1, Ash 9.9, VM 34.7 pct: FSI = 8-1/2.
D-6: H 5.1, C 74.4, N 1.5, O 8.2, S 1.1, Ash 9.7, VM 36.1 pct; FSI = 8.
2/ H_2+CO+CH_4.

Effect of Temperature

For two reasons the temperature was not varied widely in these tests. The safe limit for the reactor at 20 atmospheres is 950° C and carbon conversion decreased markedly below 850° C. The yield of product gas increased from about 11 SCF of H_2+CO+CH_4 per pound of coal at 860° C to 19 at 950° C (table 3). The methane percentage decreased but not drastically. However, the yield of methane per pound coal feed did increase from 2.5 to 3.6 SCF per pound. In test N-30 the steam rate was reduced from 30 SCF per pound of coal to 19; the temperature increased in the gasifier but the carbon conversion dropped. Some sintering of the ash occurred at the base of the reactor, indicating that the flow of steam was too low. The optimum temperature seems to be about 900°-950° C. Further tests may be necessary to determine the maximum temperature because the methane yield may decrease so much at higher temperatures that the advantage of our method of operation may be lost.

Oxygen Needed for Pretreatment

Changing the port of entry for the pretreating steam and oxygen from the middle of the free-fall section, as shown in figure 1, to the top of the free-fall section reduced the amount of oxygen necessary for pretreatment. When the pretreating gases were fed at the top they entered through a tube surrounding the coal feed inlet port. Thus the coal was entrained by the treating gases for about 2 feet before it fell 5 feet into the fluid bed. In tables 1 and 2 the results of both systems represented by tests N-12 and N-24 are compared. The amount of oxygen needed for pretreatment was decreased from 2.5 SCF per pound (N-12) to less than 1 SCF per pound (N-24). When the pretreating steam and oxygen were fed at the middle of the free-fall section, some oxygen was used in gasification because of the high temperature in this zone. When they were fed at the top of the reactor, oxygen was used only for pretreatment.

Studies in Glass Equipment of a System Using Air Instead of Oxygen

Oxygen accounts for about 10 to 14 cents per MCF of the cost of making high-Btu gas. Many different systems have been tried both in England and America in the hope of substituting air for oxygen.[1,5,6] Our approach is to feed air and steam through separate entry ports to a single fluid bed so the products of combustion can be separated from the products of steam gasification. Two designs to achieve this are shown in figures 4 and 5, and glass models based on these designs have been made. The model similar to figure 4 is constructed with a straight baffle in a 6-inch-diameter glass tube; with an L/D ratio of 2 (12-inch height/6-inch diameter), the mixing of the air and inert gas streams is only 5 to 10 percent if the baffle extends 1 to 2 inches into the bed. When the baffle is raised above the bed, the mixing is 30 to 40 percent. When the ratio is 3, the mixing is about 25 percent. The second model similar to figure 5 was constructed with a 4-inch-diameter tube inserted into a 6-inch-diameter tube. The areas of the annulus and the inner tube are about the same. This model shows more mixing of the gases than the first--21 percent when the center tube is embedded 2 inches into a 6-inch layer of coal.

After a satisfactory model has been designed for gas flow, we will use a 6-inch-diameter steel reactor to study the mixing of the solids to determine if uniform temperatures in the bed can be obtained.

CONCLUSIONS

Caking coals can be gasified in a combined free-fall, fluid-bed gasifier. The methane in the product gas is about 14 to 15 percent, which means more methane is being produced in the gasification than would be produced in the subsequent methanation. The oxygen needed for pretreatment is about 1 cubic foot per pound coal feed.

A conceptual process being investigated substitutes air for oxygen during gasification.

BIBLIOGRAPHY

1. Curran, G. P., C. H. Rice, and E. Gorin. Carbon Dioxide Acceptor Gasification Process. Division of Fuel Chemistry, A.C.S., Philadelphia, Pa., April 5-10, 1964.

2. Forney, A. J., R. F. Kenny, S. J. Gasior, and J. H. Field. Fluid-Bed Gasification of Pretreated Pittsburgh-Seam Coal. Division of Fuel Chemistry, A.C.S., New York, N. Y., Sept. 1966.

3. Forney, A. J., R. F. Kenny, S. J. Gasior, and J. H. Field. Decaking of Coals in a Fluid Bed. BuMines Rept. of Inv. 6797, 1966, 22 pp.

4. O'Dell, W. W. Gasification of Solid Fuels in Germany by the Lurgi, Winkler and Leuna Slagging Type Gas Producer Processes. BuMines Inf. Circ. 7415, 1946, 46 pp.

5. Pieroni, L. J., G. T. Skaperdas, and W. H. Heffner. Development of the Kellogg Coal Gasification Process. A.I.Ch.E. Symposium on Processing of Coal and its Byproducts, Philadelphia, Pa., Dec. 5-9, 1965.

6. Raynor, J. W. R. Gasification by the Moving Burden Technique. Gasification and Liquefaction of Coal Symposium, A.I.M.E., New York, N. Y., Feb. 20-21, 1952.

7. Ricketts, T. S. The Operation of the Westfield Lurgi Plant and the High Pressure Grid System. The Institution of Gas Engineers 100th Annual General Meeting, May 1963, London, England.

8. Schuster, Fritz. The Chemical Reaction of Methane Formation in Oxygen Pressure Gasification. Das Gas und Wasserfach <u>104</u> (49), 1963, pp. 1418-1419.

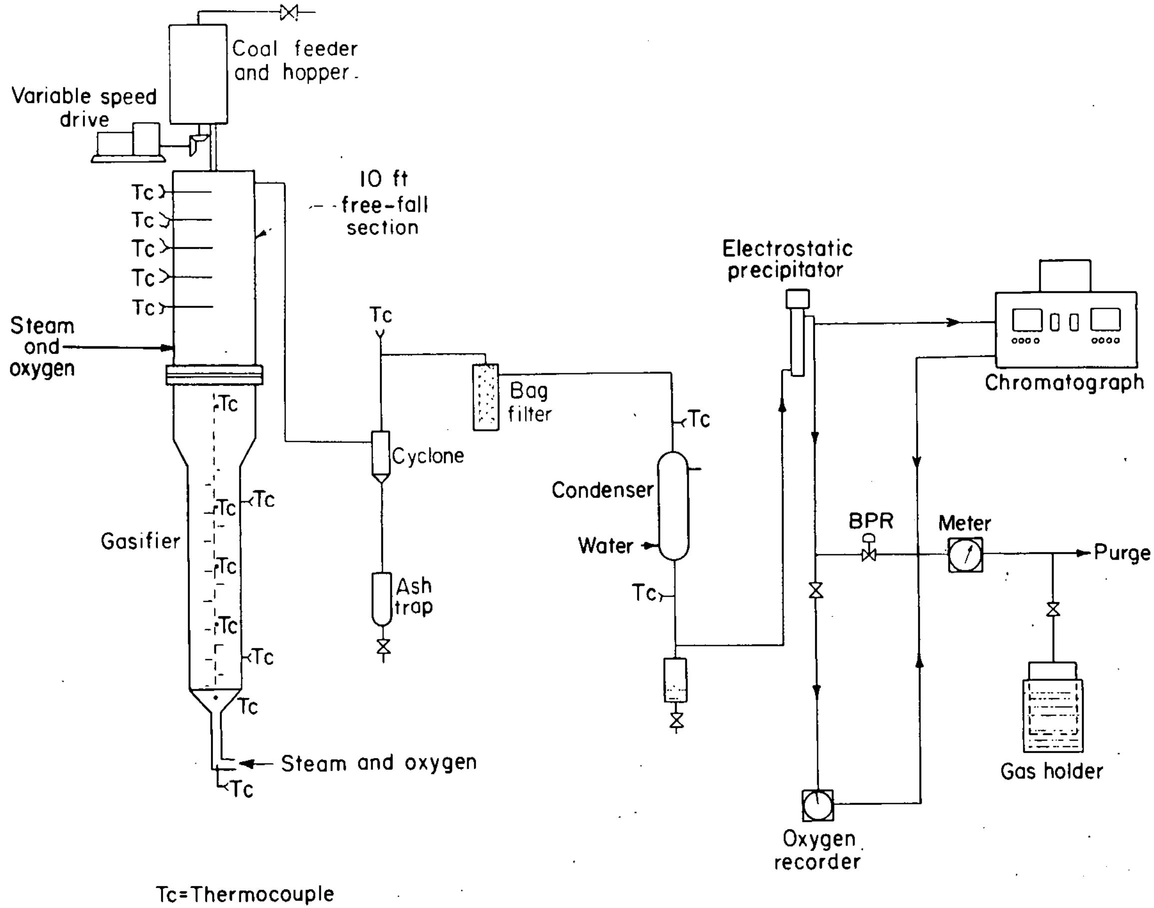

Figure 1. Flowsheet of Gasification Equipment.

9-1-65 L-9024

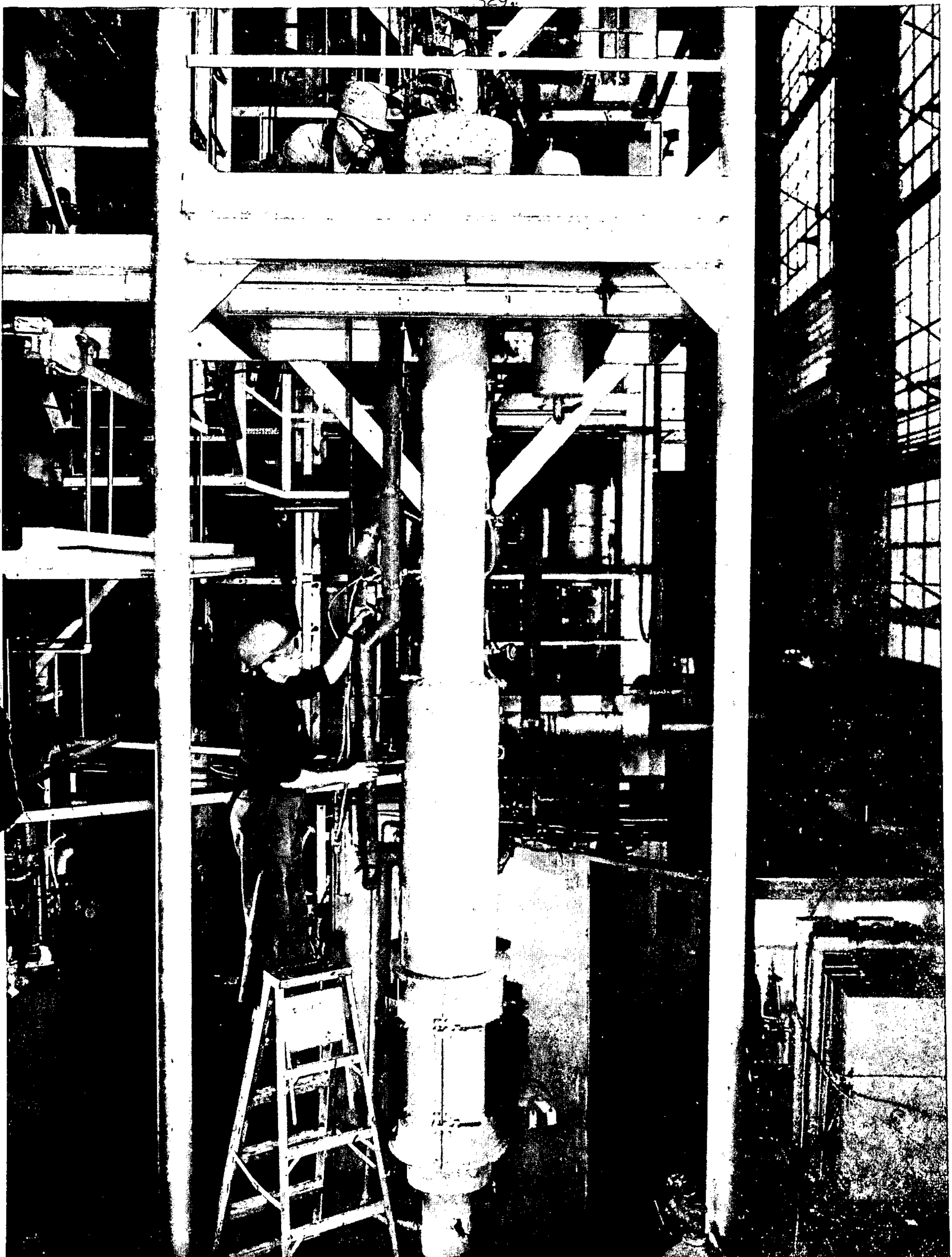

Figure 2. Gasification Equipment.

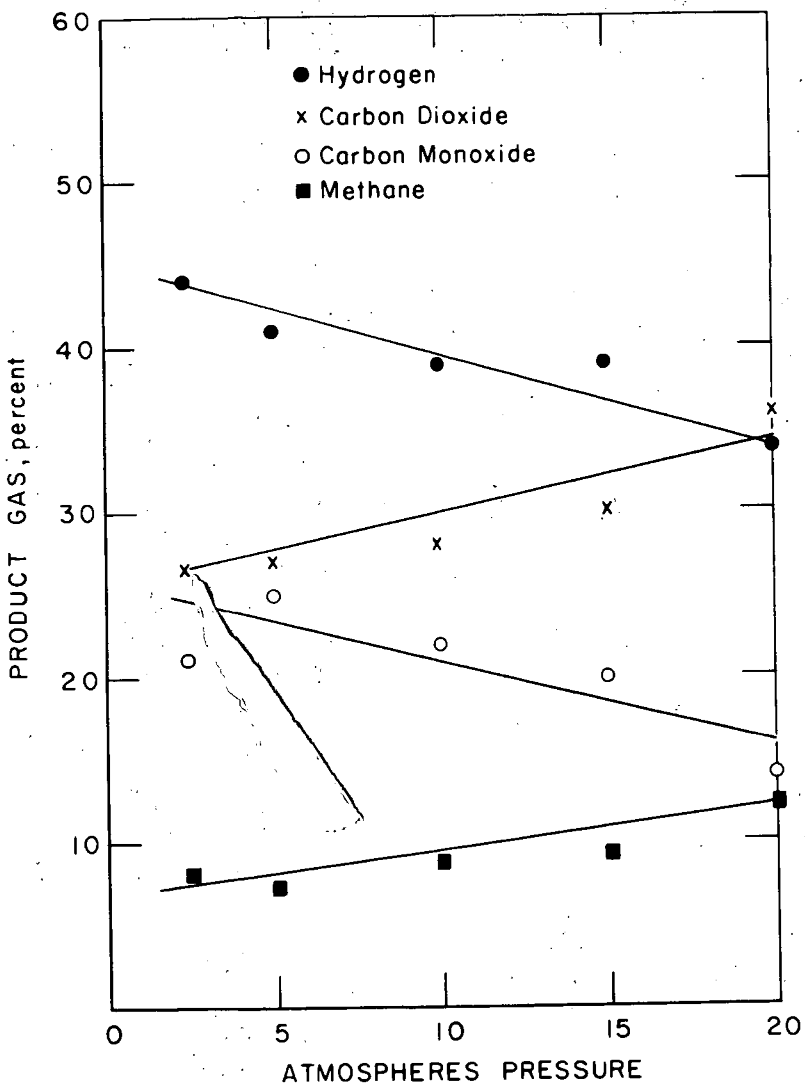

Figure 3.– Effect of pressure on the product gas.

4-13-67 L-9907

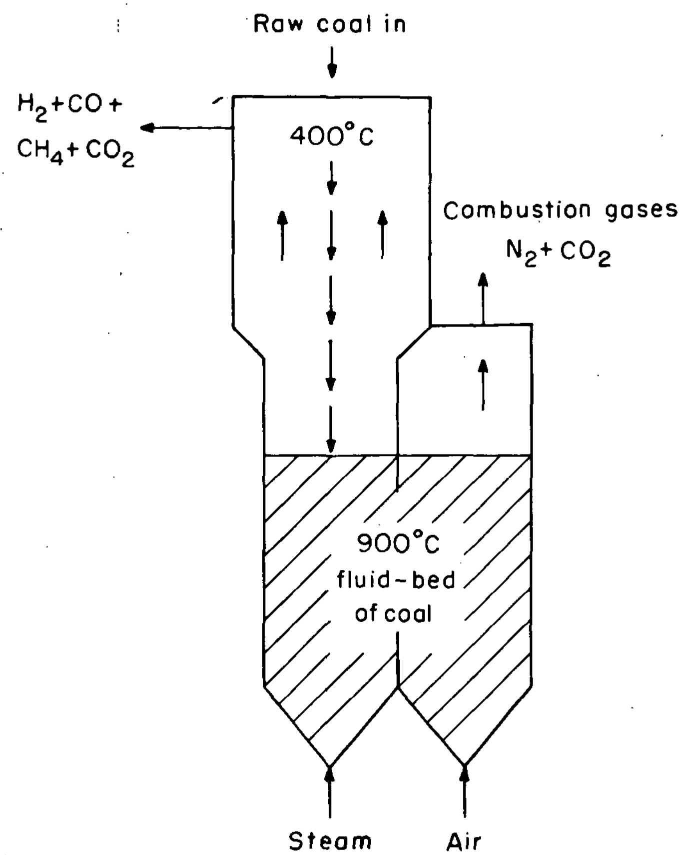

Figure 4 - Steam-coal, air-coal reaction in single fluid-bed gasifier

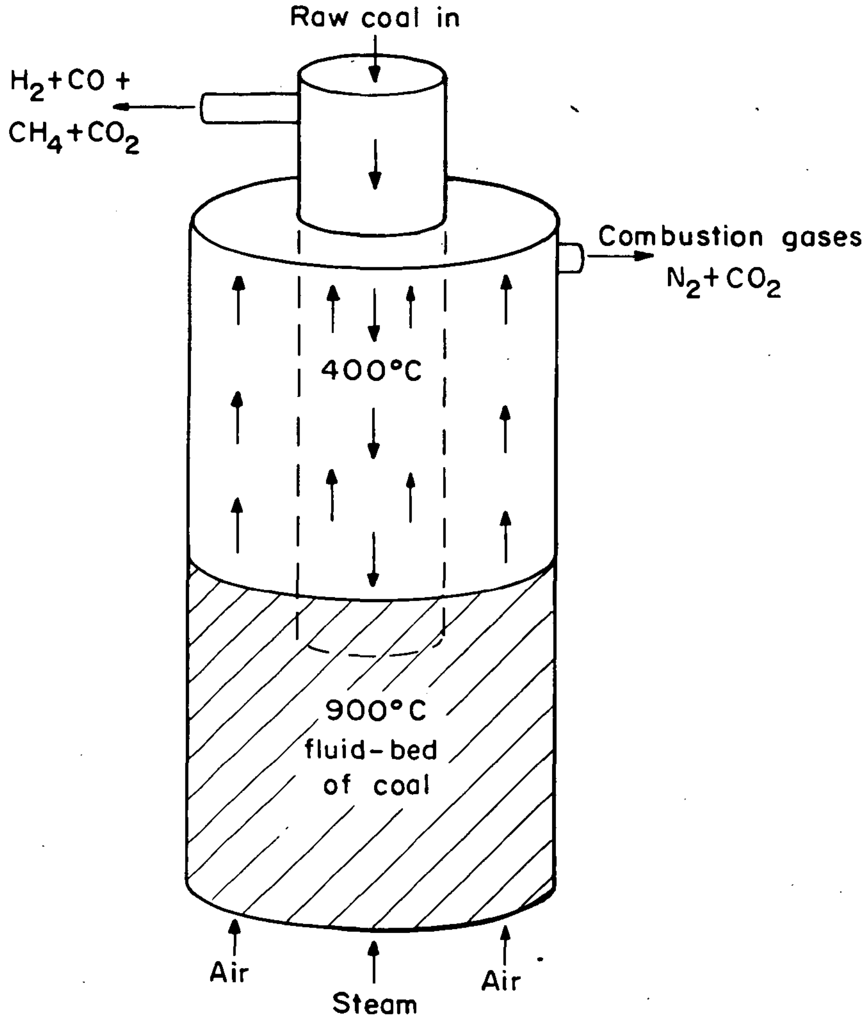

Figure 5. —Steam-coal, air-coal reaction in single fluid-bed gasifer.

4-5-67 L-9889