Fluid-Bed Gasification of Pretreated Pittsburgh-Seam Coals

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INTRODUCTION

An efficient coal gasification process operating at 800°-1,000° C and at pressures of 20-30 atmospheres is desirable for producing a synthesis gas which can be used to make a high-Btu gas. At these temperatures considerable methane is produced and less oxygen is consumed than at the higher temperatures of 1,100°-1,200° C used generally in entrained gasification systems. A fluidized bed can be operated at the lower temperature with sufficient residence time to obtain high conversion of the coal. Because most of the coals found in the East and Midwest are caking, they must be treated to destroy their caking properties before they can be used in a fluidized bed. The purpose of these tests was to investigate an integrated pretreatment and gasification system whereby the pretreated coal would be processed in a fluid-bed gasifier at conditions to produce a synthesis gas with high methane content.

Two methods of pretreating caking coal in a fluid bed to produce a free-flowing char have been developed--one at high temperature and one at low temperature. In the high temperature treatment[2, 3] coal is fluidized in steam plus air or oxygen at 430° C for 5-10 minutes. The oxygen-to-coal ratio is about 0.4 SCF per pound. In the low temperature treatment,[1] coal is fluidized in air at 240° C for 30-40 minutes. The oxygen-to-coal ratio is about 2.4 SCF per pound.

EXPERIMENTAL PROCEDURE AND RESULTS

Equipment and Method of Operation

Briefly, the technique used in the experiments was to test the treated coals in the gasifier to find if the coals were sufficiently noncaking to use in the gasifier. If the gasification were operable, then the pretreater and gasifier would be operated in series starting with a raw coal feed. When the coal was pretreated using steam plus oxygen, the gas from the pretreater, as well as the pretreated coal, would be sent to the gasifier.

The equipment used is shown in figure 1. In the usual method of operation, raw Pittsburgh-seam coal from the Bruceton mine (70 percent through 200 mesh with coarser than 35 mesh removed) is fed into a fluid-bed pretreater, 1 inch diameter by 30 inches long, operated at

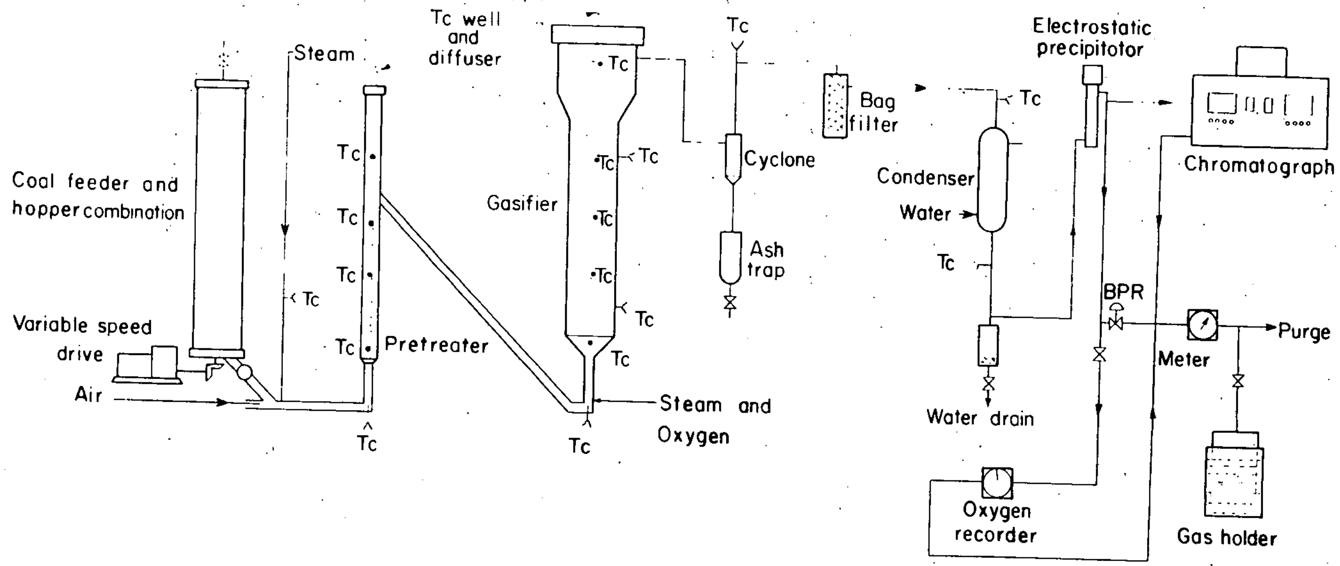

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Figure 1. Fluid-Bed Pretreater and Gasifier System.

either 450° or 260° C. The fluidizing gas is steam plus air (or oxygen). The products, gas, tars, and treated coal, pass from the top of the pretreater and flow with additional steam (or steam plus oxygen) into the fluid-bed gasifier (3 inches diameter by 30 inches long, topped by an expanded section 6 inches in diameter by 12 inches long). The product gas and coal fines pass to a cyclone and bag filter to remove the dust, then through a condenser and electrostatic precipitator to remove water plus tar, and finally to a gas meter. An inline chromatograph analyzes the gas for six components every 20 minutes. An oxygen analyzer continuously monitors the product gas. The range of process variables, such as gas fluidizing velocity and coal-feed rate, is limited. An increase in the coal feed rate decreases the residence time of the coal in both the pretreater and gasifier. The superficial gas velocity for most tests was about 0.25 foot per second.

Tests with Pretreated Coals

The first tests in the gasifier were made with coals which were pretreated in a separate unit so the free-swelling indices (FSI) and other coal properties could be determined. Pittsburgh-seam coal was used in all tests. The tests were made with high temperature treated coals (HT) having indices of 2, 1, and noncaking, respectively, and low temperature treated coals (LT) with indices of 1-1/2 and noncaking, respectively. The gasifier was operated at 2-1/2 atmospheres and 871° C with a coal feed rate of 0.4 pound per hour. Table 1 shows the analysis of the coals and the method of pretreatment.

The main difference between the LT and HT coals is that the loss of volatile matter is greater in the HT coals (35.6 percent in the raw coal and 26.0 percent in HT-440), and the oxygen content is greater in the LT (8.1 percent in the raw coal and 11.8 percent in LT-56). HT coals with an FSI of 2 and the LT coals with an FSI of 1-1/2 were caked in the reactor, while the coals with FSI of 1 or less were operable.

The main difference in gasifying LT and HT coals is that the HT coals yield more gas and are less likely to agglomerate in the gasifier. Test 61 made with HT-2 coal had a yield of 22 SCF (CO_2- and N_2-free) per pound compared to 17 in tests 58 with LT-56 coal. The temperature was held constant in both tests, but 1/2 SCFH of oxygen had to be fed to the gasifier in test 58 to prevent caking. In tests 62 and 59, oxygen was fed to the gasifier. The HT-2 coal yielded 23 SCF per pound compared to 22 for LT-56, even though the LT coal had greater carbon conversion. The quality of the product gas was about the same in all the above tests. (See Table 2.)

TABLE 1.- Analyses of pretreated coals used in the gasifier

	Raw Coal 1/ D-2	Raw Coal 1/ D-1	HT-400 2/	HT-440 3/	HT-1 4/	LT-55 5/	LT-56 6/
Moisture	1.5	1.5	0.5	0.8	0.8	0.9	0.9
Volatile matter	36.6	35.6	30.4	26.0	22.7	34.0	31.2
Fixed carbon	53.2	56.5	62.2	67.1	69.3	56.2	57.5
Ash	8.7	6.4	6.9	6.1	7.2	8.9	10.4
Hydrogen	5.1	5.2	4.8	4.5	4.1	4.7	4.2
Carbon	75.2	77.4	77.8	78.8	78.2	73.6	70.9
Nitrogen	1.5	1.5	1.4	1.5	1.7	1.5	1.4
Oxygen	8.1	8.4	8.0	8.1	7.8	9.8	11.8
Sulfur	1.4	1.1	1.1	1.0	1.0	1.5	1.3
FSI	8	8	2	1	NC 7/	1½	NC 7/

1/ Bruceton coal; 70 percent through 200 mesh with coarser than 35-mesh removed.
2/ Pretreated at 400° C; O_2/coal ratio = 0.4 SCF/lb. Residence time is 10 minutes.
3/ Pretreated at 440° C; O_2/coal ratio = 0.4 SCF/lb. Residence time is 30 minutes.
4/ Pretreated at 440° C; O_2/coal ratio = 1.1 SCF/lb. Residence time is 30 minutes.
5/ Pretreated at 240° C with air; O_2/coal ratio = 2.4 SCF/lb. Residence time is 30 minutes.
6/ Pretreated at 250° C with air; O_2/coal ratio = 2.4 SCF/lb. Residence time is 30 minutes.
7/ Noncaking.

Pretreater and Gasifier in Series

In these tests the pretreater and gasifier were operated in series; the products from the raw coal pretreatment--the char, tars, and gases--were sent directly to the gasifier. The gasifier operated satisfactorily with no agglomeration of the coals. Table 2 shows a comparison of the gasification of the two types of treated coal. The coals pretreated at 450° C yielded more product gas than those treated at 260° C. When no oxygen was fed to the gasifier, the HT coal (test 36) yielded 22 SCF of H_2+CO+CH_4 compared to 18 SCF per pound coal from the LT coal (test 60). When oxygen was fed to the gasifier the results were similar; 21 SCF per pound in test 33 compared to 19 for test 63.

TABLE 2.- Conditions of operation and results of tests using pretreated coals in the fluid-bed gasifier; pressure is 2½ atmospheres

Test No.	61	58	62	59	36	60	33	63
Coal	HT-2	LT-56	HT-2	LT-56	Raw coal D-1	Raw coal D-2	Raw coal D-1	Raw coal D-2
Input								
Steam, SCFH	14	15	15	15	15	13	14	17
Oxygen, SCFH, pretreater	0	0	0	0	1.0	1.6	1.0	1.6
Oxygen, SCFH, gasifier	0	0.5	2.5	2.5	0	0	1.4	1.1
Nitrogen, SCFH	9	10	10	10	10	10	10	10
Coal, lb/hr	0.38	0.40	0.38	0.36	0.37	0.36	0.36	0.36
Temperature, °C								
Pretreater	-	-	-	-	450	261	450	258
Gasifier, avg	868	871	872	873	860	869	857	863
Gasifier, max	875	874	877	878	874	877	873	866
Oxygen/coal, SCF/lb	0	1.3	6.6	6.3	2.7	4.5	6.7	7.5
Steam/coal, SCF/lb	37	38	41	42	41	36	39	47
Carbon conversion, pct	51	43	77	83	51	54	68	73
Steam conversion, pct	-	-	-	-	20	-	17	-
Product gas,1/ SCF/lb coal	22	17	23	22	22	18	21	19
Product gas2/ pct								
H_2	59	55	45	44	55	48	45	42
CH_4	3	3	2	2	4	3	3	2
CO	20	22	25	26	22	24	29	24
CO_2	18	20	28	28	19	25	23	32

1/ Nitrogen- and CO_2-free.
2/ Nitrogen-free.

These tests show that pretreatment at 450°-460° C with less oxygen is superior to pretreatment at 250° C, not only in increased gas production, but also because there is less likelihood of the coal caking in the gasifier. There is more CO_2 made from the coals with the 250°-260° C treatment because of oxygen added to the coal during pretreatment as shown in table 1. This evolution of CO_2 from the LT coal was also noted in earlier tests.1/ These present tests also show more gas is produced in the 2-stage process with raw coal feed than in the gasification of a coal pretreated separately considering the 15-20 percent loss of coal in pretreatment.

Table 3 shows some results in tests where oxygen, coal, and steam rates were varied to give a range of operability. At a constant coal feed rate, an increase in the oxygen rate from 2.4 to 9.1 SCF per pound coal increased the conversion from 46 to 80 percent and also the product gas (H_2+CH_4+CO) yield from 19 to 23 SCF per pound coal. The hydrogen and methane yields decreased. Increasing the steam from 26 to 56 SCF per pound (tests 31 and 35) resulted in only minor changes in carbon conversion and gas yield. The hydrogen content of the product gas increased from 42 to 48 percent. An increase in the coal rate from 0.36 to 0.91 pound per hour decreased the carbon conversion from 68 to 50 percent and the gas yield from 21 to 16 SCF per pound coal.

TABLE 3.- The effect of variables on the gasification of Bruceton coal using the pretreater and gasifier in series; pressure is 2-1/2 atmospheres

Test No.	39	37	31	35	33	30
Input						
Steam, SCFH	11	15	10	22	14	35
Oxygen, SCFH, pretreater	1.0	1.0	1.5	1.0	1.0	1.5
Oxygen, SCFH, gasifier	0	2.2	0.9	1.4	1.4	4.5
Nitrogen, SCFH	8	10	12	10	10	12
Coal, lb/hr	0.33	0.35	0.39	0.39	0.36	0.91
Temperature, °C						
Pretreater	450	450	450	450	450	450
Gasifier, avg	860	865	867	858	857	858
Gasifier, max	874	876	875	875	873	871
Oxygen/coal, SCF/lb	2.4	9.1	6.2	6.1	6.7	6.6
Steam/coal, SCF/lb	32	43	26	56	39	38
Carbon conversion, pct	46	80	61	64	68	50
Steam conversion, pct	20	13	11	11	17	10
Product gas,1/ SCF/lb coal	19	23	19	21	21	16
Product gas,2/ pct						
H_2	56	42	42	48	45	47
CH_4	4	2	2	3	3	3
CO	21	29	30	24	29	30
CO_2	19	27	26	25	23	20

1/ N_2-, H_2O, and CO_2-free.
2/ N_2- and H_2O-free.

The Effect of Pressure

The effect of pressure on the methane yield and on the caking property of the coal treated at high temperature was studied at 2-1/2, 5, and 8 atmospheres (table 4). With an increase in pressure, however, the gas flow of steam plus oxygen must be increased to maintain the same linear velocity in the fluid-bed. To maintain the desired ratios of oxygen/coal and steam/coal the coal feed must be increased, but with a fixed-bed height the residence time of the coal in the pretreater and gasifier will decrease. At 8 atmospheres pressure operation of the gasifier was difficult because of coal agglomeration due to the decreased residence time in the pretreater. At 0.4 pound per hour the residence time of the coal in the pretreater was about 40 minutes, and at 1.25 pounds per hour about 13 minutes, apparently too short a time for pretreatment at these operating conditions. When the coal rate was decreased to about 1 pound per hour the unit operated satisfactorily, with no coal agglomeration, but the carbon conversion was low, indicating insufficient residence time of the coal in the gasifier.

TABLE 4.- Effect of pressure on gasification of Bruceton coal using pretreater and gasifier in series

Test No.	39	41	45	44	46	55	54
Pressure, atm	2.5	2.5	5	5	8	8	8
Input							
Steam, SCFH	11	14	36	34	65	50	42
Oxygen, SCFH, pretreater	1.0	2.0	1.6	1.6	2.2	2.0	1.2
Oxygen, SCFH, gasifier	0	1.0	0	4.0	0.6	0	5.2
Nitrogen, SCFH	8	8	11	11	8	22	27
Coal, lb/hr	0.33	0.37	0.75	0.73	1.25	0.91	0.97
Temperature, °C							
Pretreater	451	451	455	456	456	453	452
Gasifier, avg	860	855	862	831	842	823	864
Gasifier, max	874	867	873	874	869	869	866
Oxygen/coal, SCF/lb	2.4	8.1	2.1	7.6	2.2	2.2	7.0
Steam/coal, SCF/lb	32	38	48	46	52	55	43
Carbon conversion, pct	46	66	52	66	43	41	53
Steam conversion, pct	20	18	16	13	-	14	8
Product gas,1/ SCF/lb coal	19	18	20	20	16	17	15
Product gas,2/ pct							
H_2	56	46	54	46	52	58	45
CH_4	4	3	5	3	8	6	5
CO	21	22	20	23	18	13	20
CO_2	19	29	21	28	22	23	30

1/ N_2-, H_2O-, and CO_2-free.
2/ N_2- and H_2O-free.

The methane content of the product gas increased from 4 percent at 2-1/2 atmospheres (test 39) to 8 percent at 8 atmospheres (test 46) when there was no oxygen fed to the gasifier. Less amounts of methane and hydrogen were formed when oxygen was used in the gasifier.

There was also less methane formed at 8 atmospheres when the coal-feed rate was decreased from 1.25 to 1.0 pound per hour. At this lower rate the nitrogen rate had to be raised to maintain fluidization since the steam and oxygen rates are fixed by the coal feed. This increase in nitrogen decreased the partial pressure of the reacting gases. To operate at the desired 20 atmospheres to produce the maximum yield of methane the gasifier height must be increased to increase the residence time, or possibly the product gas could be recycled so that the coal rate could be decreased. A recycle probably would not be effective if oxygen were being fed to the gasifier since present tests showed that oxygen would react with gas in preference to the coal.

Methane yields in these tests may be compared with yields from the Lurgi gasifier[4/] where 11 percent of methane is produced at a pressure of 8 atmospheres; however, the carbon conversion of the brown coal used in the Lurgi is much higher than in our tests.

The best coal-feed rate for this size gasifier at 2-1/2 atmospheres is about 0.4 pound per hour. The minimum oxygen rate to the pretreater is 1.2 SCF per pound of coal feed when oxygen is fed to the gasifier also but 2.0 SCF per pound with no oxygen to the gasifier. These figures compare with only 0.4 SCF oxygen per pound needed in the earlier tests[3/] to pretreat the same coal.

Advantages of the 2-Stage Process

The advantages of the present process using two fluidized beds in series are:

1. Raw caking coal can be fed to this system without difficulty. Coal which had been drastically pretreated did not agglomerate in the gasifier.

2. Mixing the treated coal from the pretreater with ash in the gasifier reduced the possibility of the coal caking in the gasifier.

3. Feeding treated coal into the bottom of the gasifier with oxygen and steam also reduced the possibility of the coal caking in the gasifier. This is shown in the tests where oxygen fed into the gasifier prevented agglomeration of the coal treated at 250° C in the pretreater.

Disadvantages of the 2-Stage Process

The disadvantages of the 2-stage system are:

1. Two fluid beds in series are difficult to operate. Any surge in the pretreater throws untreated coal into the gasifier.

2. The coal, gases, tars, and oxygen entering at the bottom of the gasifier, make temperature control difficult.

3. At least 1.2 SCF oxygen per pound coal is needed for pretreatment. This is almost one-third the requirement for gasification in the Lurgi[5] which requires 4 SCF per pound.

4. Production of methane is lower than the Lurgi because in this system the gas-solids flow is not countercurrent.

New Gasification System

In an attempt to overcome these difficulties, the system has been revised as shown in figure 2. Now the coal is fed into the top of a 10-foot long, 6-inch diameter pipe and falls freely through an upward flow of gas from the 3-inch diameter fluid-bed gasifier. The coal, as it drops, is carbonized and devolatilized so by the time it enters the fluid bed it should be noncaking. Preliminary tests of the revised system indicate that it is operable, but no data are available at this time. The methane yield should be higher due to countercurrent flows.

CONCLUSIONS

Low-temperature (250° C) treated coal and high-temperature (450° C) treated coal can be reacted in a fluid-bed gasifier operated at 870° C without agglomeration of the coal. The Pittsburgh-seam coal passes first to a pretreater and then to a gasifier, the two being in series. The raw coal used in the 2-stage system yielded more product gas than when the coal was pretreated separately. The minimum oxygen needed is 1.2 SCF per pound of coal. At a pressure of 8 atmospheres the methane content of the product gas is 8 percent. An increased methane yield is anticipated by use of countercurrent free-fall system. Further improvement in the gasification can result if means are found of supplying heat indirectly to decrease the oxygen requirement.

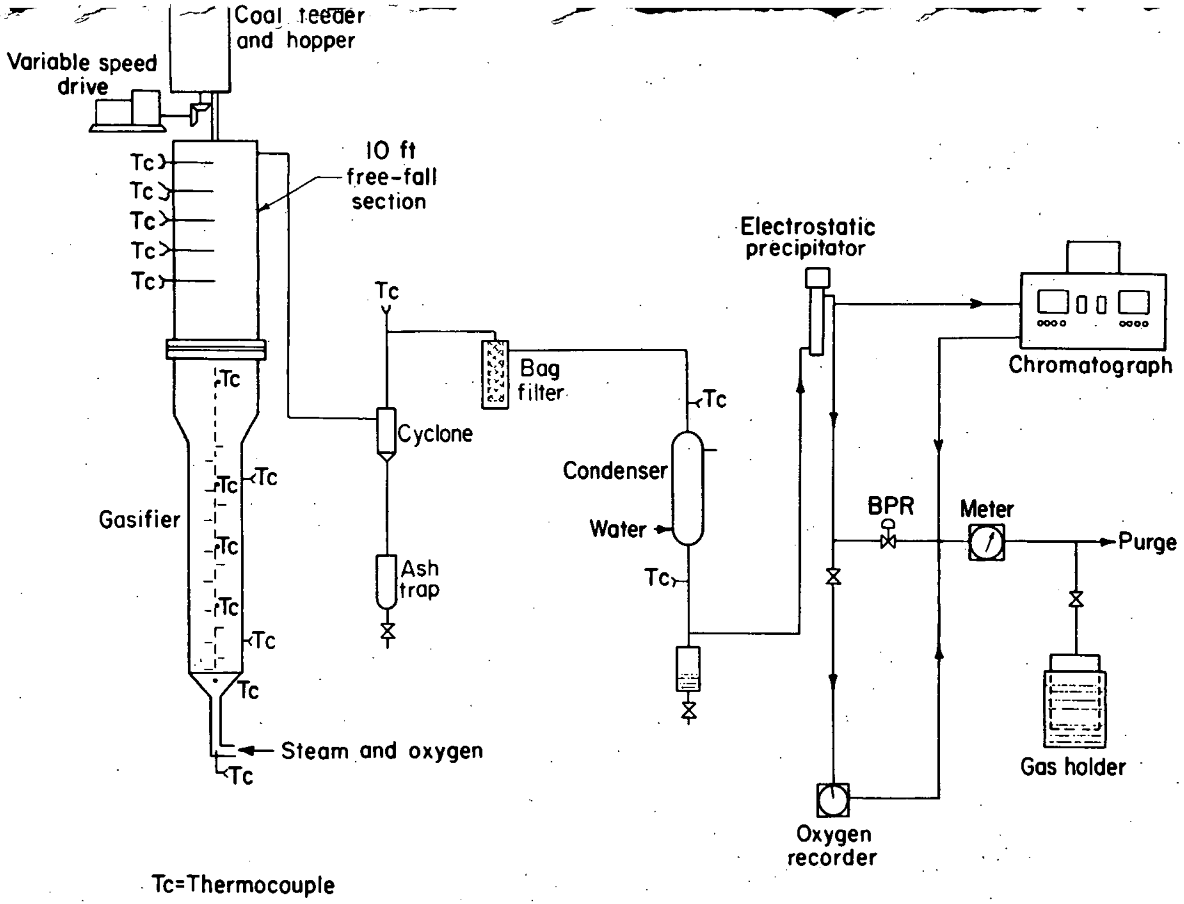

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Figure 2. Free-Fall Reactor and Fluid-Bed Gasifier System.

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