COAL PRETREATMENT IN FLUIDIZED BED

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INTRODUCTION

During development of the IGT coal hydrogasification process for making synthetic pipeline gas,[4,7] the most reactive part of coal for producing methane was found to correspond to its volatile matter. Therefore, research efforts were concentrated on high-volatile bituminous coals. However, when such coals were exposed to an atmosphere of hydrogen at high temperature and pressure, rapid and severe fusion of the coal particles caused caking and agglomeration in the reactor bed. To prevent this agglomeration, the coal must be pretreated to destroy its agglomerating tendency before being used as hydrogasification feed. This program was initiated to 1) establish operating conditions that would produce, with a minimum or pretreatment, a nonagglomerating material that retains maximum volatile matter, and 2) supply sufficient feed for the hydrogasification test program.

BACKGROUND

Much work has been reported on the destruction of caking properties of coal. Low-temperature carbonization, if carried far enough, drives off enough volatile matter so that the remaining char does not agglomerate. Examples of this are Consolidation Coal Company's Montour char used in much of the early hydrogasification work, and the more recent char produced by FMC Corp. by multistage pyrolysis.[6] However, the Montour char contains only about 17% volatile matter, while the original Montour bituminous coal contained 31%.

Several investigations[2,3,5] concluded that the agglomerating tendency is destroyed by oxidation of the surface of the coal particles. Treatment in small fluidized-bed reactors with nitrogen, steam, carbon dioxide, or helium[2,3] all failed to destroy the caking properties. But adding a small amount of oxygen to these inert gases produced nonagglomerating coal. Minimizing pretreatment, therefore, requires minimizing the extent of such oxidation.

Investigators agree that since oxidation is involved, a fluidized bed would be necessary to dissipate the heat released and to maintain a uniform bed temperature without hot spots. However, much of the reported work was conducted in small batch reactors.[2,3] In a large continuous unit, the uniformity of pretreatment and the residence time required would be quite different than with a batch system. In addition, batch reactors, after being charged, require a time to heat the charge up to the reaction temperature. This makes the residence time at reaction temperature uncertain. A continuous operation avoids this problem, but the backmixing in a fluidized bed leads to short-circuiting of some untreated coal. For this program, a continuous unit capable of fairly high throughput was designed and built.

EQUIPMENT AND PROCEDURE

The pretreater system is shown schematically in Figure 1. Figures 2 and 3 give actual views of some sections. The pretreater is a 10-in. Schedule 40 pipe, 15 ft long, made of A-335 alloy. The upper portion is enlarged for solids disengagement. Heat is supplied to the reactor by external electrical heaters. The bottom 76 in. of the reactor is heated by a 12-kw Huppert furnace. The next 56 in. is surrounded by four zones of heaters, each with four Hevi-Duty heating elements, providing 2.7 kw per zone. The enlarged portion of the reactor is heated by six 1-kw strip heaters. Variable voltage transformers control the power input to the heaters, which are needed primarily at start-up. Once the reaction is initiated, little external heat is required.

A 1/8-in.-thick sintered stainless steel plate serves as the gas distributor. Preheated inlet gas enters a plenum chamber and then passes through the distributor. Offgases pass through a cyclone for fines removal and then to a knockout pot for heavy tar condensation. After the gas is cooled in a water-jacketed condenser, the oils and water are collected. A final venturi water scrubber and cartridge filter remove the oil and tar mist from the gas. The gas is then metered, sampled, and vented.

The coal is crushed in a hammer mill, dried, and then screened to −16+80 mesh. About 500 lb of coal is charged to a hopper which is connected at the bottom to the pretreater by a screw feeder. The feed enters the pretreater about 6 in. above the distributor plate. Feed rates of up to 100 lb/hr can be attained. A 3-in.-diameter overflow pipe controls the bed height. The overflow collects in a receiver and is periodically dumped into drums. Fines from the bed were originally returned to the bed by an internal cyclone with a dipleg sealed in the bed, but tar tended to build up in the cyclone and caused the reactor pressure to increase. At present, a heated external cyclone with a collector pot is installed and operates much more smoothly.

Temperature is measured and recorded by a group of thermocouples in the bed at several levels, and pressure taps at various points indicate the state of fluidization. Both gases and solids are sampled and analyzed.

In addition to chemical analyses, an agglomeration test was performed on each sample to give a quick indication of the degree of pretreatment. This test is a modified version of that reported by Forney, et al.[3] A sample in a stainless steel wire mesh boat is placed in a quartz tube heated by an electric furnace. Nitrogen is purged through the system while the temperature is brought up to 1400°F. Hydrogen is then passed through the system for 1/2 hr; nitrogen is again used during cooling. We found that the Bureau of Mines test[3] was too mild at 600°C (1112°F) and too strong at 900°C (1652°F), and did not correspond to the hydrogasifier conditions. With our conditions, materials that are free-flowing or slightly crusted would pass through the hydrogasifier without difficulty.

DISCUSSION OF RESULTS

Of the coals to be studied in this project, attention was focused on high-volatile bituminous coal. Most of the work was devoted

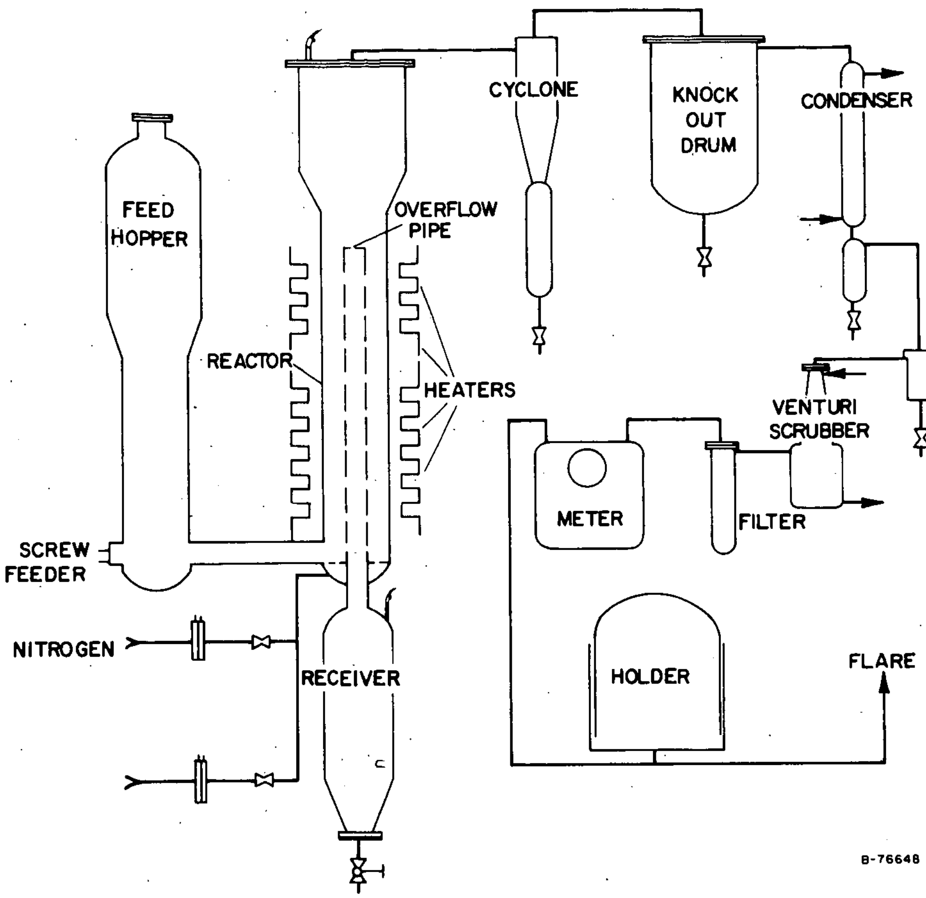

Figure 1. CONTINUOUS-FEED COAL PRETREATMENT UNIT

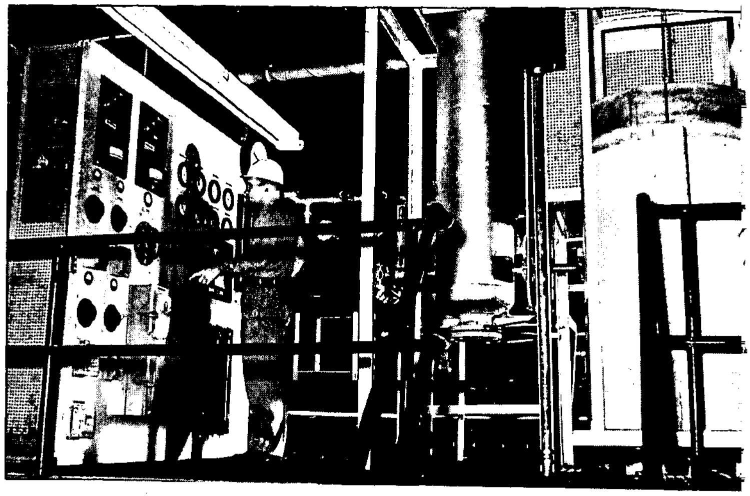

Figure 2. Fluid-Bed Pretreat
Feed Hopper, and Panel Boar

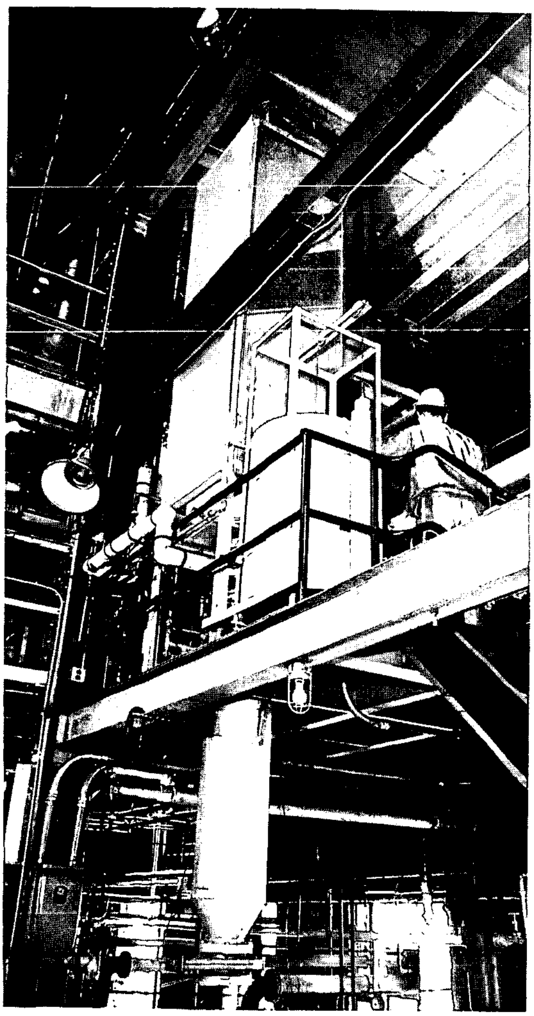

Figure 3. Fluid-Bed Pretreater
Residue Receiver.

to Pittsburgh No. 8 Seam coal from Ireland mine, with some tests on Ohio No. 6 Seam coal from Broken Arrow mine and a highly volatile West Virginia No. 5 Block coal. Proximate and ultimate analyses of these are given in Table 1. Typical run data and product analyses are shown in Table 2.

Besides the visual evaluation from the agglomeration test, the volatile-matter content was chosen as the quantitative index of pretreatment severity. Operating experience with the hydrogasifier showed that minimum coal pretreatment must reduce the volatile matter to 24-26%. Various aspects of pretreatment are discussed in more detail below.

Pretreatment Temperature

Perhaps the most critical operating variable is temperature. The coals tested have a plastic point temperature around 700°F. Below this temperature, we could not produce a free-flowing coal with a 2-hr residence time and a wide range of oxygen concentrations in the pretreatment gas. On the other hand, localized combustion begins at bed temperatures near 775°F. The heat released can not be dissipated by the bed and runaway temperature results. Similar observations were made by Agarwal, et al.[1] in operating a fluidized coal dryer. They noted that combustion occurred on the air distributor deck when stagnant pockets of coal caused by poor gas distribution reached temperatures of about 800°F. Consequently the practical pretreatment temperature range is quite narrow: between 725° and 750°F. The necessary close temperature control can be maintained with a fluidized bed.

Extent of Coal Oxidation

Since pretreatment is an oxidation process, the extent of oxidation is clearly related to the extent of pretreatment. We found that minimum pretreatment required reaction of more than 1.0 SCF of oxygen per pound of coal fed. Figure 4 shows the region of operation in terms of temperature and oxygen consumption required for successful pretreatment. Increasing the oxygen consumption would produce more and more devolatilized coal, which is contrary to our desire to preserve as much volatile matter as possible. Minimum pretreatment, therefore, requires oxygen consumption of 1.0-1.5 SCF/lb coal fed.

We investigated the effect of oxygen concentrations of from 2% to 21% (air) by volume in the pretreatment gas, by blending the correct amounts of air and nitrogen. The oxidation rate was rapid enough to consume all the oxygen fed so no oxygen breakthrough was noted in the effluent gas. Pretreatment is governed by the amount of oxygen reacted rather than by the oxygen concentration. However, at high oxygen concentration, faulty gas distribution leads to localized combustion. Moreover, with the fluidizing velocity essentially fixed by the coal particle size distribution, the coal residence time must be shortened at the higher oxygen concentration to maintain the same oxygen consumption per pound. We found 10% oxygen to be satisfactory. In large-scale operations, such a mixture can be obtained cheaply by adding air to the recycled pretreater effluent in the proper proportions. We are, however, studying the use of air (21% oxygen) in pretreatment.

Coal Residence Time

Because the rate of oxidation is very fast, there is essentially no residence time required for chemical reaction. In a plug flow reactor,

Table 1. ANALYSES OF HIGH-VOLATILE-CONTENT BITUMINOUS COALS TESTED IN PRETREATER

Coal Source	Pittsburgh No. 8 Ireland Mine	Ohio No. 6 Broken Arrow Mine	West Virginia No. 5 Block
Run No.	FP-44	FP-38	FP-48
Proximate Analysis, wt %			
Moisture	1.1	2.4	1.1
Volatile Matter	34.7	40.2	34.6
Fixed Carbon	52.5	52.5	56.5
Ash	11.7	4.9	7.8
Total	100.0	100.0	100.0
Ultimate Analysis (dry), wt %			
Carbon	71.1	75.3	77.1
Hydrogen	5.01	5.63	5.25
Sulfur	4.04	3.53	1.17
Ash	11.88	5.00	7.86
Screen Analysis, USS, wt %			
+20	3.2	4.5	4.0
+30	21.7	23.8	21.0
+40	24.6	24.3	22.7
+60	26.4	25.4	24.4
+80	12.4	11.3	12.6
–80	11.7	10.7	15.3
Total	100.0	100.0	100.0

Table 2 OPERATING DATA AND PRODUCT ANALYSES OF SELECTED PRETREATMENT RUNS

Coal Source	Pittsburgh No. 8				Ohio No. 6			W.Va. No.5
Run No.	FP-11	FP-14	FP-25	FP-31	FP-33	FP-35C	FP-39	FP-48
Temperature, °F								
Average	765	666	749	750	749	634	739	738
Maximum	775	698	757	751	752	648	746	744
Coal Feed Rate, lb/hr	49	76	37	18	28	56	47	51
Coal Residence Time, min	53	51	111	228	120	116	98	81
Feed Gas Rate, SCF/hr	603	590	760	584	630	574	663	619
Oxygen Concentration, vol %	21.0	9.8	9.8	9.9	10.5	21.0	9.3	5.9
Oxygen/Coal, SCF/lb	2.6	0.76	2.0	3.2	2.4	2.1	1.3	0.72
Agglomeration Test Results*	C	C	F	F	F	C	F	PC
Proximate Analysis, wt %								
Moisture	0.4	0.3	1.0	0.8	0.3	0.4	1.5	0.3
Volatile Matter	27.4	30.3	24.7	21.3	24.6	36.6	26.6	25.2
Fixed Carbon	58.5	55.6	65.0	63.8	68.6	58.5	66.9	67.9
Ash	13.7	13.8	9.3	14.1	6.5	4.5	5.0	6.6
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Ultimate Analysis (dry), wt %								
Carbon	69.8	69.7	72.0	69.2	75.7	74.9	74.9	77.9
Hydrogen	4.11	4.33	3.81	3.37	3.85	4.91	3.83	4.33
Sulfur	†	†	†	†	†	†	2.75	0.96
Ash	13.74	13.88	9.38	14.21	6.56	4.49	5.09	6.59

* C = caked
F = free flowing
PC = partially caked

† Not analyzed

coal feed rate would depend only on the oxygen input rate and the unit oxygen consumption. Moreover, if gas distribution is uniform, the oxidation would be uniform. In a continuous fluidized-bed reactor, however, residence time must be long enough to minimize the effect of the short-circuiting of untreated feed into the product. The solids mixing in the fluid bed results in a coal particle age distribution, and hence a distribution in the extent of oxidation. Therefore, the average residence time and unit oxygen consumption are greater for a continuous fluidized bed than for a batch unit. Also, gas of higher oxygen concentration cannot be used with a correspondingly higher coal feed rate – and hence shorter residence time – to maintain the same unit oxygen consumption, because shortened residence time leads to increased short-circuiting of untreated coal. If the residence time is not shortened, the volume of bed material required would exceed the capacity of our unit. So most of our work has been done at oxygen concentrations of 10% or less.

Figure 5 shows the region of operation for successful pretreatment in terms of coal residence time and unit oxygen consumption. The two are related in almost direct proportion. Residence times of 1/2 – 2-1/2 hr were investigated. Minimum pretreatment at reasonable feed rates requires coal residence times of between 1 and 2 hours.

Petrographic Study

To better understand the mechanism of the pretreatment reaction, samples of coal before and after pretreatment were mounted and examined petrographically.[8] A skin of high reflectance was formed on the coal particles by the surface oxidation. Variation in the skin thickness was not significant, indicating rapid surface reaction and slow penetration below the skin. Short-circuited untreated coal particles can be identified as those without this skin. A free-flowing sample had fewer "skinless" particles than a sample that caked.

SUMMARY

A 10-in.-diameter continuous fluidized-bed reactor system is being operated to produce nonagglomerating coal from high-volatile bitiminous coal by mild surface oxidation. Minimum pretreatment is defined by acceptability of the coal as a feed to the hydrogasifier. This corresponds to 24-26% volatile matter in the pretreated coal. The following operating variables, in the order of importance, contribute to satisfactory pretreatment: reaction temperature of 725°-750°F, oxygen reacted/coal feed ratio of 1.0-1.5 SCF/lb, and a coal residence time of 1-2 hours.

ACKNOWLEDGMENT

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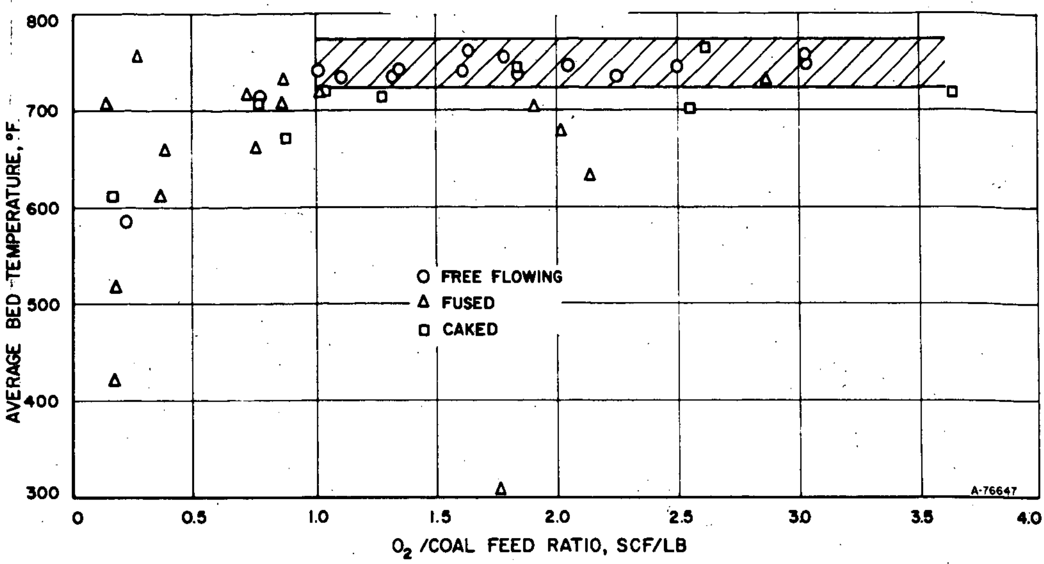

Figure 4. PRETREATMENT RESULTS AS A FUNCTION OF BED TEMPERATURE AND UNIT OXYGEN CONSUMPTION

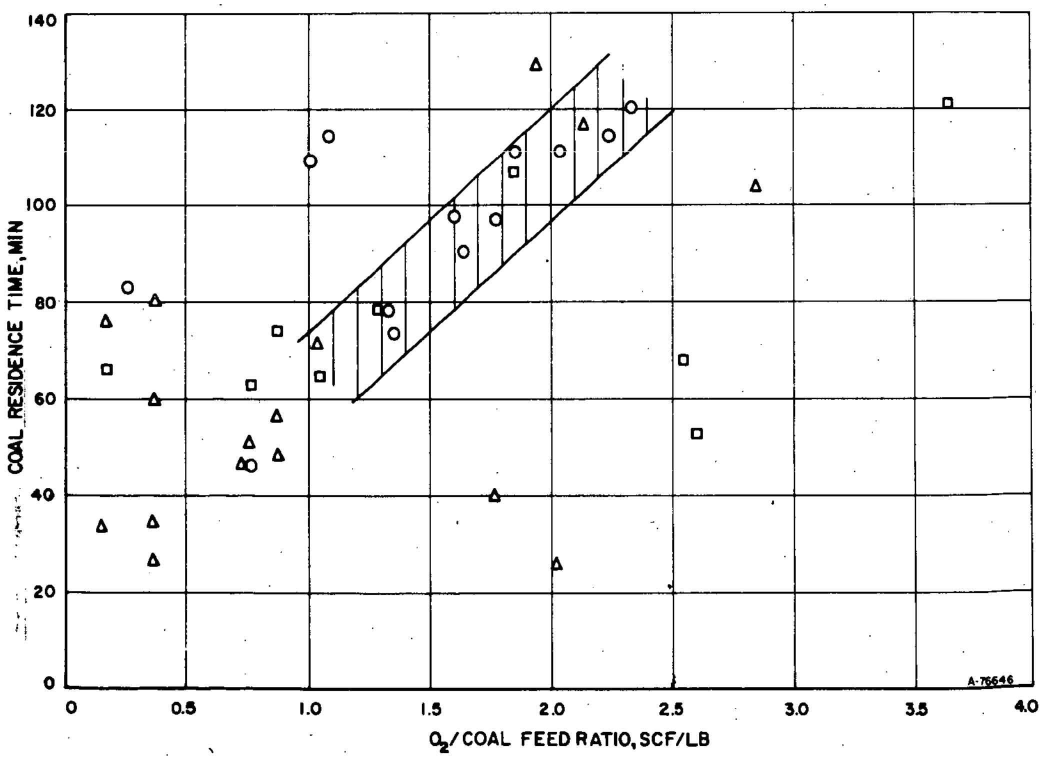

Figure 5. PRETREATMENT RESULTS AS A FUNCTION OF COAL RESIDENCE TIME AND UNIT OXYGEN CONSUMPTION

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