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Project Summary

Fluidized-Bed Gasification of Peat, Lignite, Subbituminous, and Pretreated Bituminous Coal

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This report summarizes and compares results of gasifying four different feedstocks in a pilot-scale fluidized-bed gasifier. Effects of operating variables (e.g., temperature, steam/carbon feed ratios, bed height, and feed rate upon carbon conversion and gas production) are described. Gas compositions, expressed as mole percentages on a dry N_2-free basis, showed only small differences except for sulfur gases. No correlation of wastewater species production rates with reactor operating conditions was observed. Tar production rate was greatest for a New Mexico subbituminous coal. A coastal peat and a Texas lignite gave similar tar production rates.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

As a part of continuing research on the environmental aspects of fuel conversion, the U.S. Environmental Protection Agency has sponsored a research project on coal gasification at North Carolina State University's Department of Chemical Engineering. The facility used for this research is a small coal-gasification/gas-cleaning pilot plant. Overall objectives of the project are to characterize the gaseous- and condensed-phase emissions from the gasification/gas-cleaning process, and to determine how emission rates of various pollutants depend on adjustable process parameters. While the overall project objectives include both gasification and gas cleaning, the research described in this report is concerned primarily with gasification.

The following paragraphs describe the pilot plant and discuss results of gasifying four feed materials: a devolatilized Kentucky bituminous coal, a New Mexico subbituminous coal, a North Carolina peat, and a Texas Lignite.

Pilot Plant

The pilot plant consists of three major subsystems: the gasifier, the raw gas cleaning system, and the acid gas removal system.

The fluidized-bed reactor is a 15.2 cm (6 in.) I.D. Schedule 40 pipe (316 SS) enclosed in several layers of insulation and contained in a 61 cm (24 in.) I.D. Schedule 80 carbon steel pipe. The overall height of the unit is about 3.7 m (12 ft). The gas feed is introduced into the reactor through three feed nozzles, spaced triangularly near the bottom of the reaction chamber. Coal is fed at the top of the reactor from a pressurized feed hopper and removed at the bottom by nitrogen-purged screw conveyors.

The temperature profile in the bed is monitored by six thermocouples in a central thermowell in the reactor. The thermocouples are at positions 13, 25, 64, 89, 114, and 140 cm (5, 10, 25, 35, 45, 55 in.) above the feed nozzles. The 25 cm thermocouple is used for reactor temperature control; the oxygen feed rate is adjusted to maintain the selected temperature. A reactor pressure tap is at the top of the reactor. Differential pressure taps—on the gas feed line below the feed nozzles, at 38 and 89 cm above the feed nozzles, and at the top of the reactor — are used to monitor the pressure drop across the feed cones and two pressure drops in the bed. The level of the fluidized bed is monitored with a nuclear level gauge, and is controlled by adjusting the char removal screw rotation rate. The height of the bed above the feed nozzles is normally 97 cm (38 in.), although occasional runs with a 132 cm (52 in.) bed have been made.

The PCS (particulates, condensables, and solubles) removal system is shown in Figure 1. Gas exiting the gasifier passes through a cyclone separator, where most of the elutriated particles are removed and collected in a fines collection vessel. After the cyclone, the gas line expands to 7.6 cm (3 in.), and the gases pass through a scrubber system. The scrubber system consists of a set of three spray nozzles to quench the gas and a reduction of the pipe diameter to 2.5 cm (1 in.) to promote gas/liquid contacting.

The condensate accumulates in the PCS tank, where cooling coils maintain the condensate temperature well below its boiling point. The emerging gas passes through a single-pass shell-and-tube heat exchanger (city water on the shell side, and gas on the tube side). The exchanger is situated vertically above the PCS tank so that condensing water runs back into the tank. A demister and coalescing filter remove any remaining water. The gas leaving the filter is either burned in a shielded flare or fed to the acid gas removal system (AGRS).

Major components of the AGRS are a packed-tower absorption column, a flash tank, and a packed-tower stripping column. The major purpose of the system is to remove CO_2 and H_2S from the gasifier make gas; however, it also usually removes most of the other sulfur gases and many of the hydrocarbons of molecular weight higher than methane. The system is modular, so that alternative absorption processes can be evaluated.

Approximately 100 process sensors are used to obtain information during pilot plant operation. The plant is controlled by a Honeywell TDC-2000 process control computer which can regulate 16 process control loops throughout the pilot plant.

The 100 process sensors (temperatures, pressures, differential pressures, flow rates, and levels) are monitored through a data acquisition system linked to a PDP 11/23 minicomputer. Each sensor is interpreted every 5 seconds for display on a video terminal. The information is averaged and stored every 5 minutes on magnetic disk for future data reduction.

Results and Discussion

A devolatilized Kentucky bituminous coal, a New Mexico subbituminous coal, a North Carolina peat, and a Texas lignite were gasified with steam and oxygen in a pilot-scale fluidized-bed reactor. The reactor was operated at pressures of 570 - 840 kPa (80-120 psia), molar steam-to-carbon feed ratios of 0.6 - 1.9, and average bed temperatures of 795-1010 °C (1460-1850°F). The coal feed rate ranged from 14 to 33 kg/hr (30 to 73 lb/hr). All reactor effluent streams were measured and analyzed, enabling performance of mass and energy balances and identification of potentially hazardous species.

Proximate and ultimate feed analyses were performed for all gasifier runs. Typical results for the four feed materials are given in Table 1.

Carbon conversions were calculated as the percent of the carbon in the feed coal, converted to a gaseous form. The calculated conversions ranged from 16% (a Kentucky char run at a low temperature) to 91% (a high temperature New Mexico coal run). The Kentucky char was the least reactive of the materials studied, and the North Carolina peat and the Texas lignite were the most reactive.

The effect of the reactor operating variables on carbon conversion and gas production was determined from runs which effectively isolated particular variables. Examples for the New Mexico subbituminous coal are given in Figures 2 and 3. Conversion and gas production increased with both increasing temperature and steam/carbon feed ratios. Increasing the reactor temperature increases the extent of devolatilization and the reaction rates in the fluidized bed. Increases in the steam/carbon ratio promote the steam/carbon reaction, which is the primary gasification reaction. Increases in the solid-phase space time increased both conversion and gas production when the increase was achieved by increasing the bed height. The solid-phase space time can also be increased by lowering the coal feed rate. Increases achieved in this manner give a decreased gas production rate and a higher carbon conversion. Insufficient experimental data were obtained to determine the effect of pressure on reactor performance. For the small pressure range covered (65-105 psig), no significant effects were observed.

Gasification of the devolatilized Kentucky coal yielded a gas composed primarily of H_2O, CO, CO_2, H_2, CH_4, N_2, H_2S, and COS. The raw coal feedstocks yielded the same gaseous species, plus a wide range of hydrocarbon and sulfur gases. The hydrocarbon species measured were aliphatics (CH_4 to C_4H_{10}) and aromatics (benzene, toluene, xylenes, and ethylben-

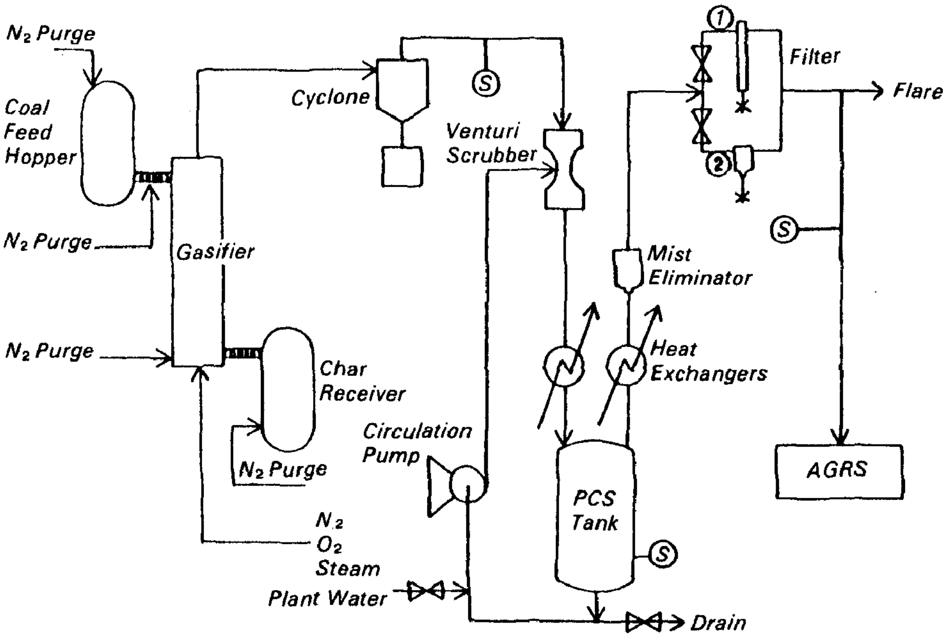

Figure 1. *Particulates, condensables, and solubles (PCS) removal system.*

Table 1. *Gasifier Feed Analyses*

	KY Char	*NM Coal*	*NC Peat*	*TX Lignite*
Proximate, wt %				
Moisture	0.9	10.5	22.8	24.3
Vol. Matter	2.4	31.7	46.3	29.1
Ash	10.7	22.6	4.6	23.5
Fixed Carbon	86.0	35.2	26.3	23.1
Ultimate, wt %				
Carbon	83.8	52.5	45.9	39.2
Hydrogen	0.6	4.8	4.3	4.2
Oxygen	2.2	18.1	44.1	32.1
Nitrogen	0.1	1.2	0.9	0.5
Sulfur	2.6	0.8	0.2	0.5
Ash	10.7	22.6	4.6	23.5

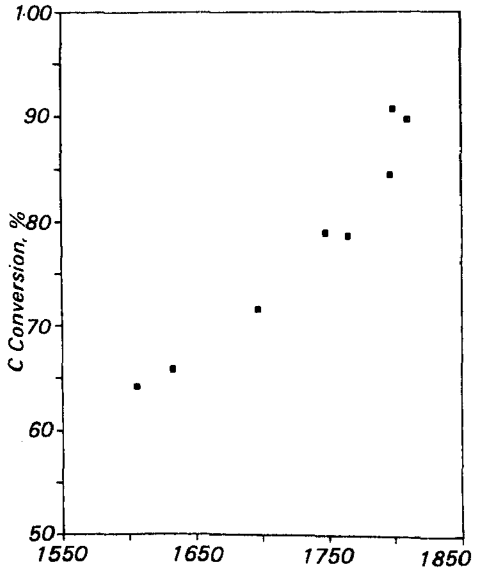

Figure 2. *Effect of average bed temperature on carbon conversion for New Mexico coal.*

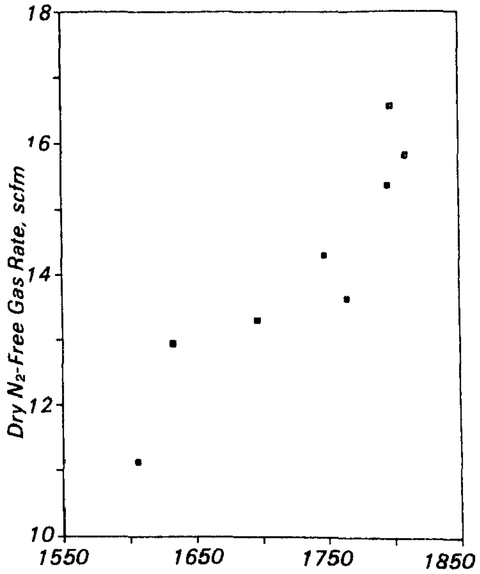

Figure 3. *Effect of average bed temperature on gas production for New Mexico coal.*

zene). In addition to H_2S and COS, the sulfur species CS_2, CH_3SH, C_2H_5SH, and thiophene were detected in the effluent gases for the raw coal feedstocks. The gas compositions, expressed as mole percents on a dry N_2-free basis, showed small differences (except for the sulfur gases). The differences in sulfur gas concentrations were generally proportional to the sulfur contents of the feed coals. Table 2 gives the average dry nitrogen-free make gas compositions.

It is likely that, during steam/oxygen gasification, the gas/solid reactions formed mainly hydrogen sulfide. Gas-phase reactions then tended to produce COS. The two gas-phase reactions of most importance, involving H_2S and COS, are:

$$COS + H_2O = H_2S + CO_2 \quad (1)$$

$$COS + H_2 = H_2S + CO \quad (2)$$

The equilibrium constants for these two reactions are:

$$K_1 = [H_2S]\,[CO_2]/[COS]\,[H_2O] \quad (3)$$

$$K_2 = [H_2S]\,[CO]/[COS]\,[H_2] \quad (4)$$

where the brackets indicate any convenient concentration units. Ideal gas behavior is assumed.

Figures 4 and 5 show plots of the experimental values of the constants K_1 and K_2, defined earlier. Also shown on the graphs are lines representing equilibrium data given in a 1979 publication. The data for K_2 generally lie below the equilibrium curve; therefore, it appears that reaction 1 is at equilibrium, but reaction 2 is not.

Wastewater samples, from the reactor product gas condensate, were subjected to several standard analyses. The wastewater from gasification of the Kentucky char was relatively clean, a consequence of pretreating the coal. The raw coal feedstocks yielded a much filthier wastewater. The concentrations and production rates of the species in the wastewater from gasification of the raw coals were nearly the same, except for dissolved carbon (carbon, COD, TOC, and TVC). Peat yielded a wastewater with much higher levels of dissolved carbon than either the subbituminous coal or lignite, for which the levels were roughly the same. No correlation of wastewater species production rates with reactor operating conditions was observed. Ranges of results for the various coals are given in Table 3.

Tar production rates were calculated from cold trap measurements. The devolatilized Kentucky coal yielded no measureable tar, but the other feedstocks gave tar production rates of 0.05 to 0.14 kg/kg dry ash-free coal feed. The tar production rate was greatest for the New Mexico subbituminous coal, for which the tar rate ranged from 1.5 to 2.2 kg/hr, or 0.10 to 0.14 kg/kg of the dry ash-free feed. The tar production rates for the North Carolina peat and the Texas lignite were similar, ranging from 0.9 to 1.5 kg/hr (0.05 to 0.10 kg/kg of the dry ash-free feed). The tar contained a significant fraction of the carbon in the feed coal. Based on estimated tar compositions, the carbon lost in the tar ranged from 7 to 16% of the feed carbon. This value can be reduced significantly by intrabed feeding of the coal.

Particulates entrained with the reactor exit gas were collected in a cyclone separator, and particulates escaping the cyclone were trapped in the sample train cold trap. Cyclone efficiencies were calculated to be as great as 96%. However, for several runs the cyclone did not function, probably due to tar condensation. The total particulate collection rates ranged from 1.0 to 3.7 kg/hr, or 4.7 to 16.4% of the coal feed rate. The carbon in the particulates represented from 3 to 17% of the feed carbon, indicating the desirability of recycle of particulates to the reactor. The elutriation rates, expressed as percent of the coal feed rate, were greatest for the Texas lignite. The feed particle size distributions were similar for all of the feeds; the greater elutriation rates observed for lignite thus suggest that it is a more friable material than the other feeds.

Samples of the spent char were taken from two locations following completion of a run — the char receiver and the bed. Analyses of char samples from the two locations yielded nearly identical results, indicating that the solids in the fluidized bed may be assumed to be well-mixed. As

Table 2. *Comparison of Make Gas Compositions Mole %, dry N2-free basis*

	KY Char	NM Coal	NC Peat	TX Lignite
CO	26.0	21.4	21.5	18.8
H_2	40.9	40.0	37.3	41.6
CH_4	2.4	8.3	8.6	6.2
CO_2	29.7	28.3	30.2	32.1
H_2S	0.9700	0.3330	0.0670	0.2480
COS	0.0420	0.0100	0.0035	0.0060
CS_2	—	0.0003	0.0003	0.0001
CH_3SH	—	0.0025	0.0040	0.0035
C_2H_5SH	—	0.0003	—	0.0001
Thiophene	—	0.0030	0.0020	0.0025
Ethylene	—	0.5260	0.7200	0.3240
Ethane	—	0.6340	0.8960	0.4760
Propylene	—	0.1760	0.3880	0.1140
Propane	—	0.0530	0.1120	0.0460
Butylene	—	0.0620	—	0.0500
Butane	—	0.0280	0.0500	0.0160
Benzene	—	0.1020	0.1180	0.0670
Toluene	—	0.0600	0.0790	0.0200
Ethylbenzene	—	0.0015	—	0.0020
p-Xylene	—	—	—	0.0008
m-Xylene	—	0.0040	—	0.0035
o-Xylene	—	0.0020	—	0.0020

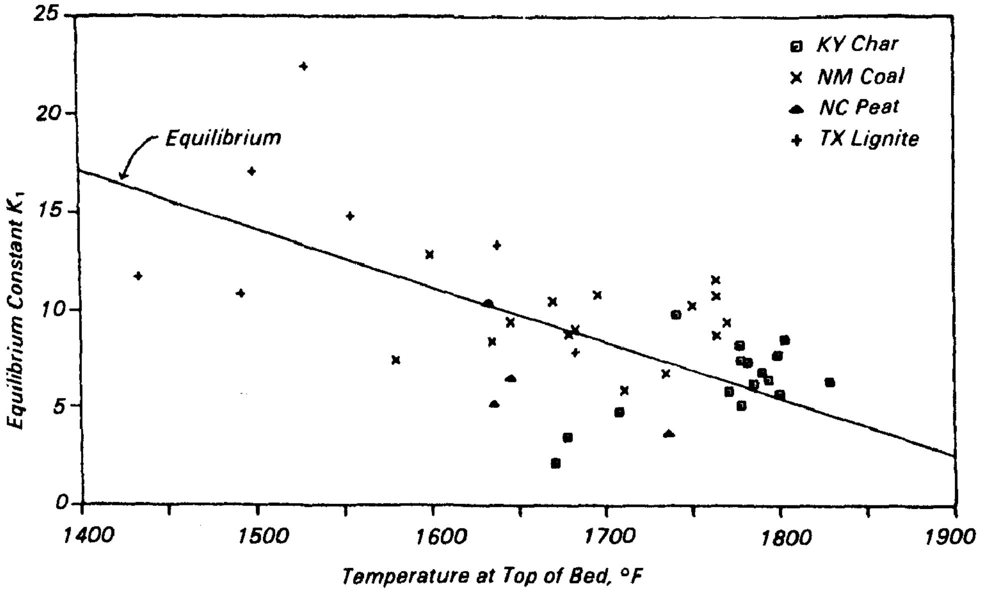

Figure 4. ***Experimental values of K_1 versus temperature.***

would be expected, the char samples generally showed a greater ash content and a lower carbon content than the feed materials. The ash content was as high as 91% for a Texas lignite run.

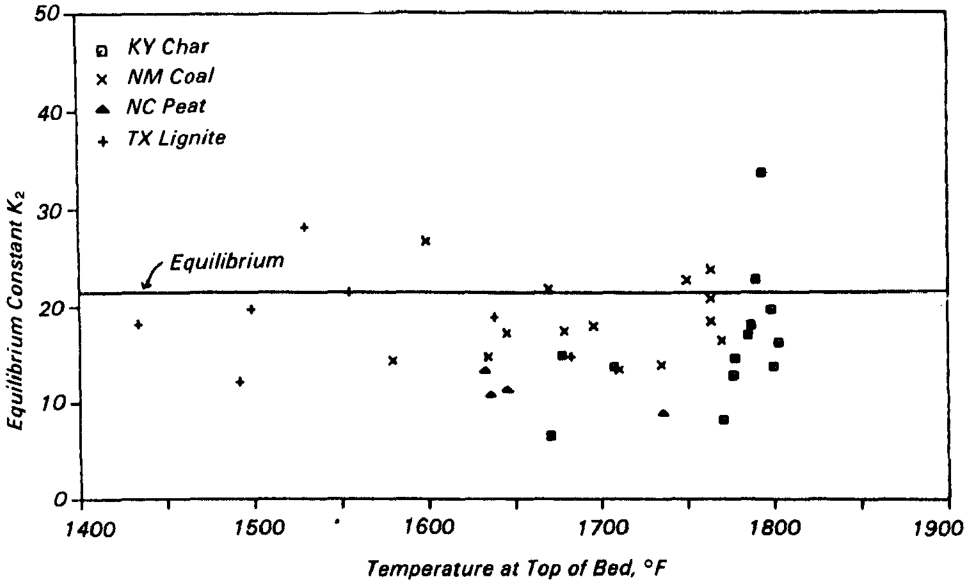

Figure 5. Experimental values of K_2 versus temperature.

Table 3. Sample Train Trap Water Species Concentrations

	Range of Results, mg/l except pH			
	KY Char	NC Peat	NM Coal	TX Lignite
pH	6.3-7.0	8.2-8.9	8.0-8.7	8.1-8.4
Nitrogen	470-650	4700-9800	5400-8200	4500-8500
Ammonia	900-1300	3900-8000	3900-7400	5500-6100
Cyanide		37-75	10-270	50-180
Cyanate		600-1770	360-7200	3200-6000
Thiocyanate		230-430	100-700	254
Sulfate	6-40	2-20	10-300	3-7
Sulfite	6-40	6-60	10-70	9-18
Sulfide			830-840	30-60
Chloride	5-50	50-180	10-130	8-22
Fluoride	8-60	12-120	6-30	4-15
Carbon		10400-18000	2500-7500	3200-4400
COD		30500-39600	950-10300	10700-13300
TOC		9300-13600	1200-6800	2400-3700
TVC		2500-11000	900-2400	1000-2400

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