FINAL TECHNICAL REPORT
September 1, 1986 through August 31, 1987

Project Title: Reactivity and Combustion Properties of Coal and Coal-Derived Solid Fuels: Part B - Fluidized-Bed Combustion Testing

ICCI Project Number: 86-87/2.3A-5
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ABSTRACT

The combustion characteristics of an untreated Illinois coal and three partially devolatilized batches of the same coal were determined in fluidized-bed combustion tests at Argonne National Laboratory (ANL). The primary objective of the fluid-bed experiments was to obtain data on combustion efficiency (i.e., carbon burn-out) which could be used to compare the "reactivity" of the Illinois coal at the three levels of devolatilization to the baseline untreated coal, and with the thermogravimetric (TGA) measurements of combustion reactivity made by the Illinois State Geological Survey (ISGS) (see Part A).

While the combustion and emission characteristics of fluidized-bed combustors are influenced by a broad range of process parameters, the parameters for investigation in this study were limited to measuring the effects of the volatile content of the fuel (11.4% to 35.7%), combustion temperature (750, 825, and 900°C), and excess air (10 and 30%) on combustion efficiency.

Each of the four fuels was tested at four test conditions covering the above ranges of bed temperature and excess air. Combustion efficiencies (conversion of fuel C to CO_2) ranged from 66 to 92% with combustion efficiency improving with fuel volatile content, bed temperature and excess air. The results are consistent with the TGA reactivity tests of the same fuels by the ISGS.

EXECUTIVE SUMMARY

The national concern with the issues of air pollution and acid rain has had a serious negative impact on the Illinois coal industry due to the generally high sulfur content of Illinois high volatile bituminous coals. One approach to increasing the use of Illinois coals would be to pyrolyze the high volatile coals to produce a low-grade liquid fuel that could be upgraded to an acceptable refinery feedstock and a devolatilized solid fuel that could still be used in industrial and utility boilers.

The purpose of this project, therefore, was to investigate the combustion characteristics of a high volatile Illinois coal and three samples of the coal at different levels of devolatilization in a fluidized-bed combustor operated at atmospheric pressure, with the data obtained on combustion efficiency for each fuel compared with the results of laboratory thermogravimetric (TGA) measurements of reactivity obtained in Part A of this Project.

A total of four tests were conducted; one test with the untreated coal, and one with each of the three different samples of the devolatilized coal. During each test, the bed temperature was varied from 750 to 900°C. Excess combustion air was varied between 10 and 30%. Bed height, gas residence time, and Ca/S were held nominally constant for all test conditions.

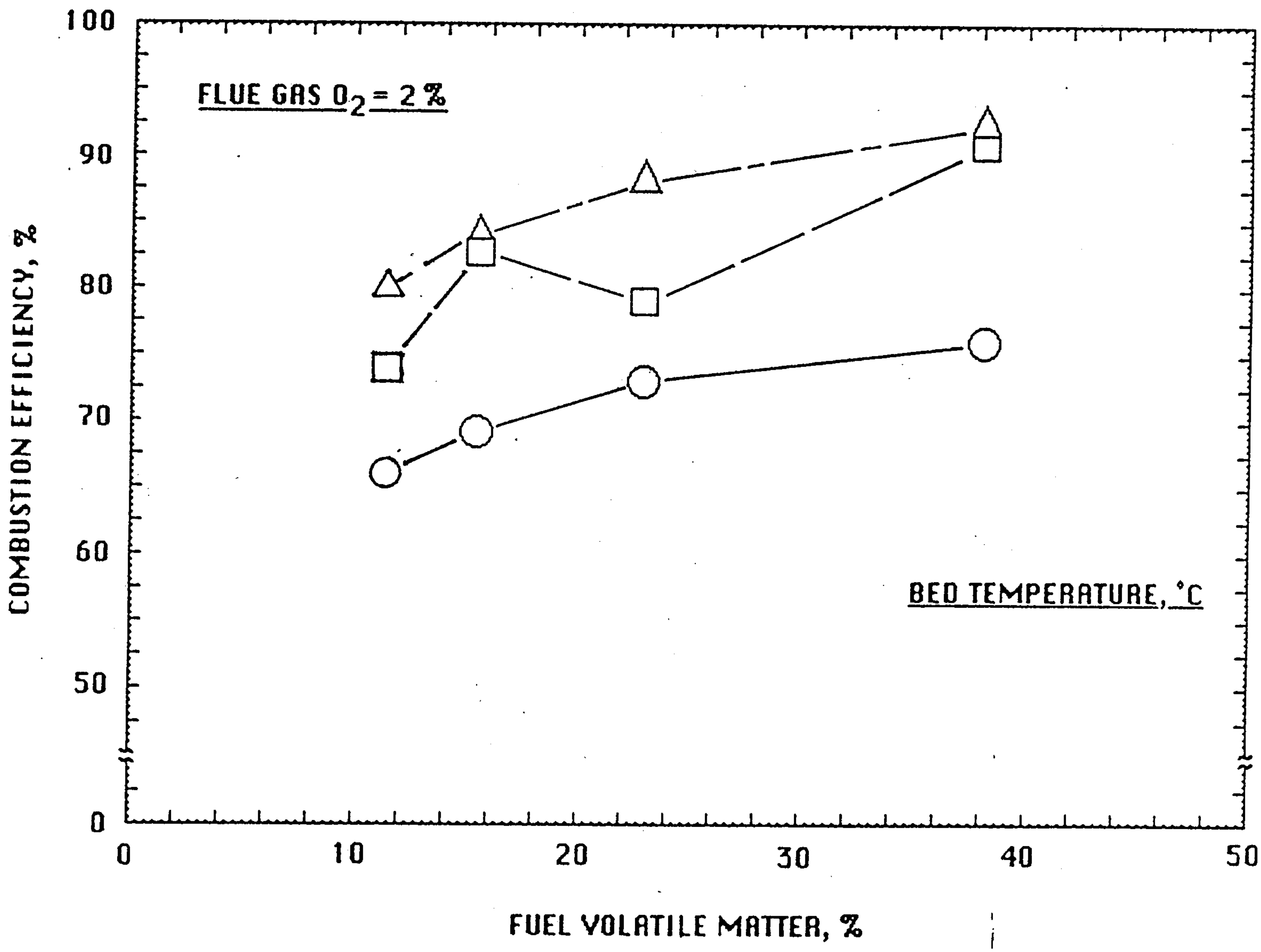
The tests were performed in the ANL 0.15-m-dia (6-in.-dia) fluidized-bed combustor (FBC) that has been used in a number of fluidized-bed combustion test programs sponsored by the U. S. Department of Energy. The facility has all the necessary ancillary equipment and instrumentation to adequately perform the tests and obtain the required test data.

The coal used in the test program was obtained from the Illinois Basin Coal Sample Program (IBCSP Coal #3). The coal containing 35.7% volatiles was partially devolatilized at the United Coal Company Research Corporation's mild gasification unit to produce separate batches of partially devolatilized coals (PD Coals) containing 23.0, 15.4 and 11.4% volatiles (PD Coal #1, 2, and 3 respectively).

The combustion tests went very well, with the carbon material balances for all but two of the sixteen test conditions being with ± 10% of 100%. In 12 of the 16 tests, the carbon balances were 100 ± 4%. Over the range of test conditions, combustion efficiencies ranged from 66 to 92%. It should be noted, that these combustion efficiencies are considerably lower than those achievable in larger diameter FBC units at optimized conditions of bed temperature, gas residence time and excess air.

The combustion efficiency data correlated very well with bed temperature, fuel volatile matter, and the O_2 level in the combustion gas. Figure 1 clearly illustrates the improvement in combustion efficiency with increasing volatile content at each of the three bed temperatures investigated (750, 825, and 900°C). The results also indicate the expected improvements in combustion efficiency for each of the four fuels tested with increasing bed temperature and O_2 level in the flue gas.

The observed effect of fuel volatile matter on combustion efficiency is also consistent with the results of TGA reactivity testing of the four test fuels by the ISGS. Those results clearly indicate that the coal was the most



FLUE GAS O_2 = 2 %
BED TEMPERATURE, °C
COMBUSTION EFFICIENCY, %
100
90
80
70
60
50
0
0
10
20
30
40
50
FUEL VOLATILE MATTER, %

FIGURE 1

readily combustible fuel followed by the PD Coals in order of decreasing volatile content.

It is recommended that the combustion characteristics of a low-volatile fuel ($\leq$15%) be determined in a larger-scale (2-3 MWt) pulverized coal burner to determine the suitability of partially devolatilized coals as industrial and/or utility boiler fuels.

OBJECTIVES

The primary objective of "Part B: Fluidized-Bed Combustion Testing," was to determine the relative combustion efficiencies of an untreated Illinois coal and three samples of partially devolatilized coals (PD Coals) prepared from the untreated coal in a 6-in.-dia atmospheric fluidized-bed combustor. Secondary objectives of the combustion tests were to obtain data on sulfur retention and NO_x emissions for each of the four fuels tested.

INTRODUCTION AND BACKGROUND

Much of the coal mined in Illinois is high volatile bituminous coal with a high sulfur content. Concerns over air pollution and acid rain currently discourage the continued use of this resource in both industrial and utility steam boilers. It would be highly beneficial to the Illinois Coal Industry, therefore, if processes were developed to convert the high-sulfur coal into environmentally more acceptable feedstocks.

One approach to achieving this objective would involve the pyrolysis of Illinois high volatile bituminous coals to produce a low-grade liquid fuel that could be further upgraded using conventional technology to an acceptable refinery feedstock. During pyrolysis, about 55 to 65 weight percent of the coal pyrolyzed would be converted to a low volatile solid fuel (char) with essentially the same calorific value as the initial coal. For this concept to be economically feasible, the acceptability of the solid fuel as a boiler fuel must be demonstrated.

The development of a char combustion data base is essential to demonstrating that devolatilized solid fuels (chars) can be seriously considered for utility and industrial boilers.

The purpose of this task, therefore, was to investigate the combustion characteristics of an Illinois coal and three samples of the coal after undergoing different levels of devolatilization. The combustion tests were performed in a 6-in.-dia atmospheric pressure fluidized-bed combustor (FBC). The results of these tests have been compared with the combustion reactivity of the fuels as determined by thermogravimetric analysis (TGA) in Part A of this project.

EXPERIMENTAL PROCEDURES

APPROACH

A total of four fluidized-bed combustion experiments were performed: one test with the untreated coal and one test with each of the three samples of the coal at different levels of devolatilization. During each test, data was collected at four different operating conditions of bed temperature and excess air.

Following startup, the bed temperature was held at 750°C for up to two hours at an excess air level of 30%. The excess air was then decreased to a level of 10% and the bed temperature held at 750°C for a second nominal two hour data collection period. Holding the excess air constant at 10%, the bed temperature was then raised to first 825°C, and finally 900°C to complete the third and fourth test conditions. Bed height, gas residence time, and Ca/S ratio were held nominally constant at all test conditions. Pressure, nominally at atmospheric pressure, was adjusted slightly to maintain a constant superficial fluidizing velocity at the different bed temperature and excess air test conditions.

Limiting the test periods for two hours reduces the fuel requirements for the fluidized-bed tests to between 4 to 6 kg per test condition. Since the carbon levels in fluidized-bed combustors are generally quite low, the performance characteristics of combustion efficiency and NO_x emissions tend to respond quickly to changes in operating conditions. Hence, the constant operating periods of two hours were of sufficient duration for these parameters to stabilize at their steady-state values. While this is not as true for sulfur retention, which is affected by the large concentration of limestone in the fluidized-bed, the reactivity of the sorbent bed is primarily controlled by a small fraction of the bed that has a residence time well below the average solids residence time. This follows from the fact that sorbent reactivity tends to decrease rapidly as the limestone becomes partially sulfated. Hence, even though the chemistry of the limestone bed was not expected to stabilize during the short two-hour test periods, this was not expected to seriously affect the measured values of sulfur retention.

For each experiment, the fuel being tested (the untreated coal or one of the three partially devolatilized fuels) was sampled and submitted for standard proximate and ultimate analysis. Due to the limited supply of the test fuels, an available coal from another test program was used in all four tests for startup and bringing the unit to the first set of test conditions. Combustion of the selected fuel for each test was then carried out in four nominal two-hour test periods. Samples of the bed material and fly ash/sorbent elutriated from the combustor were collected periodically and analyzed for total carbon and sulfur. Selected samples were also analyzed to determine the amount of carbon present in the sample as CO_2. The flue gas analyzed continuously for SO_2, CO, CO_2, O_2, and NO/NO_x.

The test data was used to calculate a steady-state carbon balance at each condition, assuming no accumulation of carbon in the fluidized-bed. Combustion efficiency was determined as the ratio of the rate of carbon leaving the fluidized bed as CO_2 to the rate of carbon fed to the bed in the fuel. At the high temperature conditions of 825 and 900°C, the carbon as CO_2 in the flue gas was corrected for the rate of carbon fed to the bed as CO_2 in the dolomite sorbent. At the low bed temperature of 750°C, the correction was limited to the amount of carbon entering the combustion as CO_2 in $MgCO_3$, since the dolomite only half-calcines at 750°C.

FACILITIES AND EQUIPMENT

The experimental equipment and instrumentation consists of a 6-in.-dia fluidized-bed combustor which can be operated at pressures ranging from atmospheric to 10 atm, a compressor to provide fluidizing-combustion air, a preheater for the fluidizing-combustion air, peripheral-sealed rotary valve feeders for metering coal and sorbent into a pneumatic feed line, cyclone separators and a filter in series for solids removal from the flue gas, associated heating and cooling arrangements and controls, and temperature and pressure sensing and display devices. A simplified schematic flowsheet of the combustor is shown in Fig. 1.

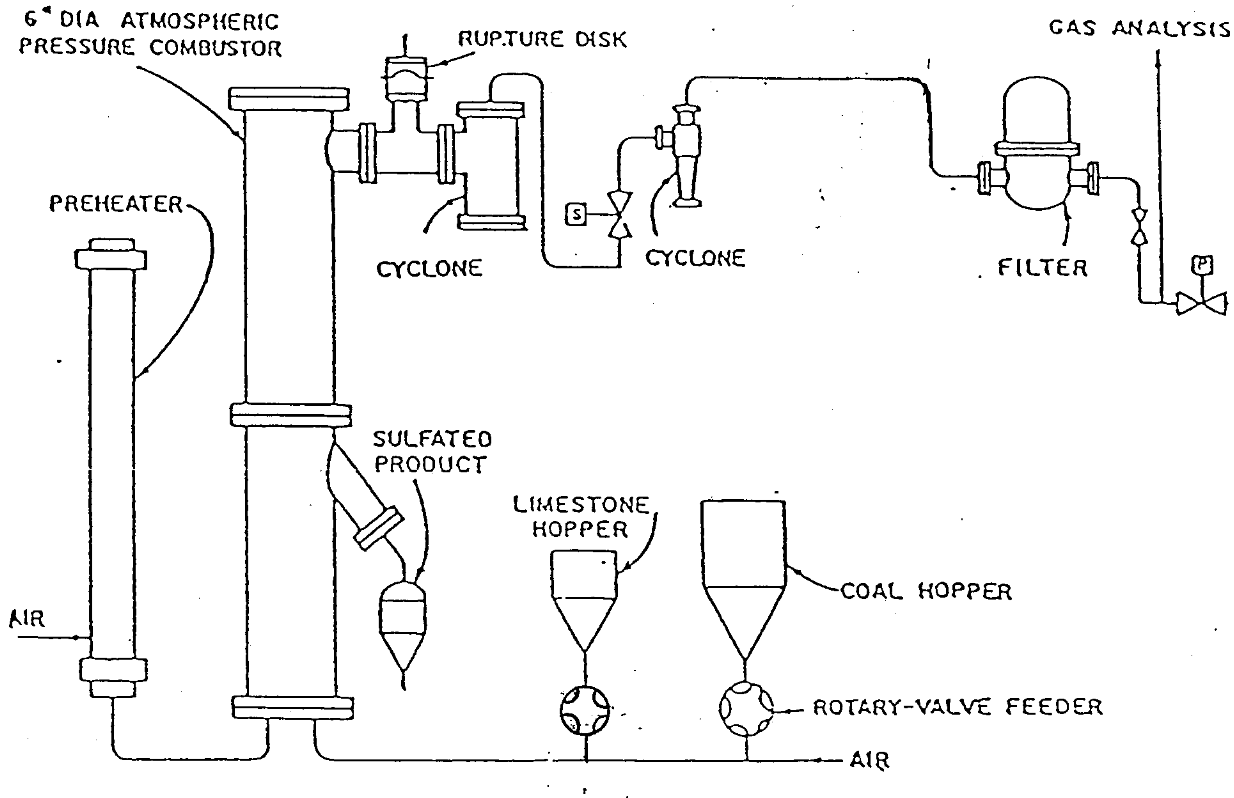


Fig. 1. Simplified Schematic of ANL Fluidized-Bed Combustion Facility

The fluidizing combustion air is supplied by a 56 kW (75-hp), screw-type compressor capable of delivering 0.12 standard m^3/s at 1.7 MPa (250 scfm at 250 psig). The air can be preheated to approximately 540°C (1000°F) in an electrically heated preheater.

Coal and limestone additives are pneumatically fed from hoppers to the combustor. Rotary valves are used to regulate the feed rate of both the coal and the limestone. The feeder-hopper sets are mounted on platform-type load cells for monitoring feed rates.

The flue-gas was sampled and analyzed continuously for the components of primary importance. Continuous determinations of CO_2, CH_4, and CO are made using infrared analyzers. Oxygen is monitored continuously using a paramagnetic analyzer. Sulfur dioxide is monitored by a pulsed fluorescent analyzer. A chemiluminescent analyzer measures NO and NO_x levels. Prior to and during each experiment, the response of each instrument is checked using standard gas mixtures of flue-gas components in N_2.

The fluidized-bed combustor was designed for service up to 1.7 MPa (250 psig) with a maximum internal design temperature of the refractory lined unit of 1150°C (2100°F). The level of the fluidized-bed can be maintained at either 0.4 or 0.8 m by means of solids overflowing through side arms attached to the unit. Fluidizing-combustion air enters the combustor through a bubble-cap type gas distributor that also accommodates a solids feed line and a bed drain line.

Bed temperature in the unit is controlled by five hairpin-shaped internal cooling circuits. Air is used as the coolant during atmospheric pressure operation, whereas water is the coolant at high-pressure operation.

The flue gas passes through two cyclone separators and a positive-type filter before being vented. The solids collected by the cyclones (and the bed overflow) can be sampled periodically during the course of a test via lockhopper systems.

MATERIALS

Fuels. The fuels used in the test program were obtained from the Illinois State Geological Survey (ISGS). The untreated coal was the Illinois Basin Coal Sample Program (IBCSP) #3 coal containing 35.7% volatile matter (dry basis). The three PD coals supplied by the ISGS were prepared by the partial pyrolysis of the IBCSP #3 coal in the mild gasification unit at the United Coal Company Research Corporation in Bristol, Virginia.

The four fuels to be tested were received from the ISGS in sealed plastic bags in 55-gallon drums (under a nitrogen blanket). The fuels were stored under a continuous nitrogen purge at ANL until tested. The analyses of the fuels supplied by the ISGS (and confirmed by independent analyses obtained from a commercial laboratory) are given in Table 1.

Sorbent. The sorbent used during the combustion tests was Tymochtee dolomite, a material containing 51.8 wt % $CaCO_3$ and 43.3% $MgCO_3$. The impurities in the sorbent are primarily SiO_2 (3.6%) and Al_2O_3 (1.5%).

Table 1. Proximate and Ultimate Analyses (Dry Basis) of Four Fuels Tested

	ANL Test Number and Fuel Designation			
Component	183-139 IBCSP #3	183-145 PD COAL #2	183-147 PD COAL #1	183-150 PD COAL #3
Moisture	5.3	6.3	1.0	3.5
Proximate, wt %				
Volatile Matter	35.7	15.4	23.0	11.4
Fixed Carbon	55.9	73.1	66.8	76.2
H.T Ash	8.5	11.4	10.1	12.4
Ultimate, wt %				
Hydrogen	5.5	2.9	3.8	2.2
Carbon	77.6	79.9	78.1	80.9
Nitrogen	2.0	1.9	1.8	1.9
Oxygen	4.2	2.1	4.2	0.85
Sulfur	2.2	1.8	2.0	1.8
Chlorine	--	0.14	--	0.10

RESULTS

OPERATIONS

The tests with the IBCSP #3 coal and the three PD Coals went very well, achieving steady operating conditions at each of the four planned test conditions. The first attempt, however, to run with PD Coal #1 (ANL Test No. (183-143) encountered difficulties in feeding the fuel to the combustor due to the relatively high moisture content of the fuel as received (10.1 wt % H_2O). As a result, PD Coal # 1 was placed in a 55-gallon drum and purged with nitrogen for several days, reducing the moisture to 1 wt %. A second attempt was then made to run with PD Coal #1 (ANL Test No. 183-147), and the experiment proceeded without any major problems.

The nominal test conditions for each of the four fuels are given in Table 2.

Table 2. Operating Conditions for the Fluidized-Bed Combustion Experiments

Coal	Bed Temp. (°C)	Flue Gas O_2(%)	Fuel Feed Rate (kg/h)	Air Flow Rate (m^3/h)	Pressure (kPa)	Fluidizing Velocity (m/s)	Gas Residence Time(s)
IBCSP#3	750	5	2.9	24.3	145	0.91	0.89
	750	2	3.1	21.3	122	0.95	0.86
	825	2	2.7	21.9	131	0.98	0.83
	900	2	2.8	22.2	140	0.99	0.82
PD #1	750	5	2.8	24.1	145	0.90	0.90
	750	2	2.9	19.9	122	0.89	0.91
	825	2	2.8	20.2	131	0.90	0.90
	900	2	2.7	20.7	140	0.92	0.88
PD #2	750	5	3.2	24.2	145	0.91	0.89
	750	2	3.3	19.2	122	0.86	0.95
	825	2	2.8	20.0	131	0.89	0.91
	900	2	2.9	20.3	140	0.90	0.90
PD #3	750	5	2.4	17.6	101	0.95	0.86
	750	2	2.7[a]	16.7	101	0.90	0.90
	825	2	2.6	16.7	101	0.97	0.84
	900	2	2.2[a]	15.8	101	0.97	0.84

[a]Adjusted value of the measured fuel feed rate to obtain ratio of carbon out to carbon in of 1.0

In each test, the air flow rate was set and the fuel feed adjusted to give a nominal O_2 concentration in the combustion gas of either 5 or 2%. These O_2 levels correspond to approximately 30 and 7-10% excess air, respectively. It must be noted, however, that these excess air levels are based on the fuel burned and not on the air to fuel ratios (since not all the fuel was combusted at each test condition, the air to fuel feed ratios were always less than the nominal values of 1.3 and 1.1).

During the test with PD Coal #3, the load cells used to determine the coal feed rate behaved in an unusual manner, occasionally exhibiting large decreases in weight over relatively short intervals. As a result, the feed rates of the fuel during that test were adjusted for two of the test conditions to obtain a ratio of carbon out to carbon in of 1.0.

CARBON BALANCES AND COMBUSTION EFFICIENCIES

The carbon balances and combustion efficiencies for the four fuels at each of the four test conditions are tabulated in Table 3. In all but two tests, the

Table 3. **Measured Carbon Balances and Combustion Efficiencies for the Four Test Coals**

Coal	Volatile Matter (%)	Bed Temperature (°C)	Flue Gas O_2 (%)	Carbon[a] Balance (%)	Combustion[b] Efficiency (%)
IBCSP#3	35.7	750	5	96	83
		750	2	90	76
		825	2	103	91
		900	2	102	92
PD #1	23.0	750	5	103	83
		750	2	100	73
		825	2	100	79
		900	2	102	88
PD #2	15.4	750	5	100	78
		750	2	97	68
		825	2	109	83
		900	2	101	83
PD #3	11.4	750	5	101	70
		750	2	123[c]	66
		825	2	97	74
		900	2	115[c]	80

[a] Ratio of rate of carbon leaving combustor to rate of carbon entering combustor x 100%

[b] Percent of carbon in fuel leaving combustor as CO_2

[c] To determine combustion efficiency in these tests, feed rate was adjusted to give an adjusted carbon balance of 100%

carbon material balances were 100 ± 10%. In 12 of the 16 tests the carbon balances were within ± 4%. As already described above, in two of the test conditions with PD #3, erratic behavior of the coal feed hopper load cells resulted in two relatively poor carbon balances of 123 and 115% (see Table 3). For the purpose of calculating the combustion efficiencies at those two test conditions, the fuel feed rate was adjusted to give a carbon balance of 100%.

Carbon efficiencies for the tests varied from a low of 66% with the lowest volatile fuel (PD Coal #3, VM = 11.4%) at 750°C bed temperature at 2% O_2 in the combustion gas to a high of 92% with the highest volatile fuel (IBCSP Coal #3, VM = 35.7) at 900°C bed temperature at 2% O_2 in the combustion gas.

It must be emphasized that the test conditions were selected to highlight the differences in the combustion characteristics of the high to low volatile fuels. Hence, the combustion efficiencies obtained during these tests are considerably lower than those achievable in larger diameter fluidized-bed combustors and optimized conditions of bed temperature, excess O_2 and gas residence time.

The combustion efficiency data are correlated with bed temperature, fuel volatile matter, and the O_2 level in the combustion gas in Figures 2, 3, and 4, respectively. Figure 2 clearly illustrates the improvement in combustion efficiency with increasing fuel volatile content at each of the three bed temperatures investigated. Figures 3 and 4 also illustrate the expected improvement in combustion efficiency for each of the four fuels tested with increasing bed temperature and O_2 level in the combustion gas.

The observed effect of fuel volatile matter on combustion efficiency is consistent with the results of the TGA reactivity testing of the same four fuels in Part A of this project. Burn-off curves obtained during the TGA tests indicated that fuels with higher volatile matter content burned more rapidly. For example, while heating at a constant rate the amount of material burned off at 500°C was 70% for the raw coal, 55% for PD Coal #1, 40% for PD Coal #2 and only 20% for PD Coal #3, clearly indicating that the coal (IBCSP Coal #3) was the most readily combusted fuel followed by the PD coals in order of decreasing volatile content.

SULFUR RETENTION AND NO_x EMISSIONS

Secondary objectives of the program were to obtain data during the combustion tests on sulfur retention and NO_x emissions. A malfunction of the NO_x analyzer early in the test program prevented obtaining any data on NO_x emissions being obtained.

Post test analysis of the sulfur retention data also indicates that there may have been a problem in the SO_2 analysis. Measured sulfur retentions (based on sulfur in the flue gas and sulfur in coal) ranged from a low of 76% to a high of 98% at Ca/S ratios ranging from 2.3 to 3.6. The results, however, exhibited no consistent correlation with bed temperature, Ca/S ratio, fuel, or flue gas O_2, with most of the measured SO_2 retentions in the range of 94 to 96%. The SO_2 analyzer is not suspected of malfunctioning, as it responded well to the zero and span gases used to calibrate the instrument before and during each test. It is possible that the gas conditioner used to condition the sample gas (dry and filter) may have affected the SO_2 gas analysis (by removing SO_2), although this has not been a problem encountered previously in the FBC facility at ANL.

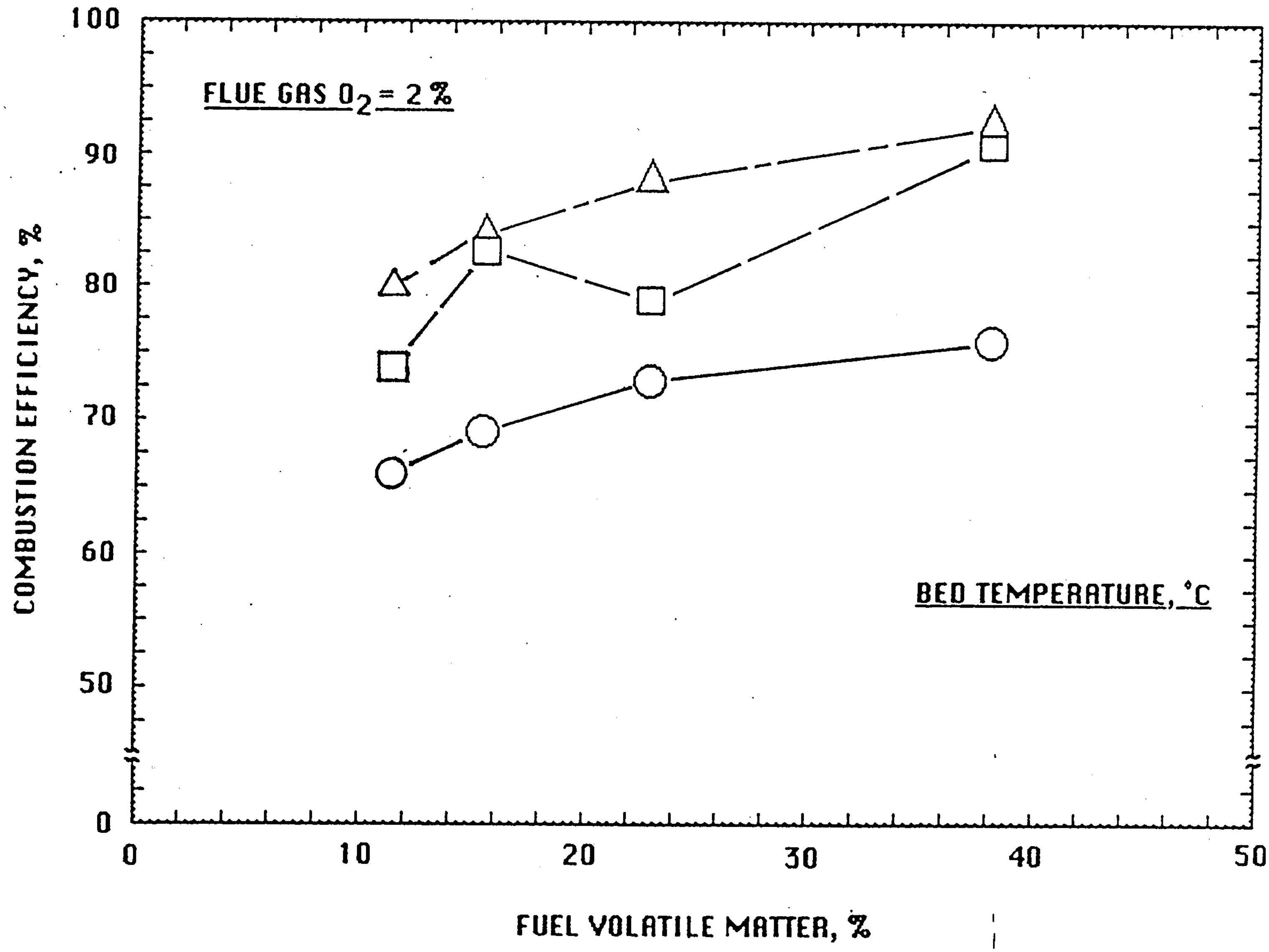

FLUE GAS O_2 = 2 %
BED TEMPERATURE, °C
COMBUSTION EFFICIENCY, %
100
90
80
70
60
50
0
0
10
20
30
40
50
FUEL VOLATILE MATTER, %

FIGURE 2

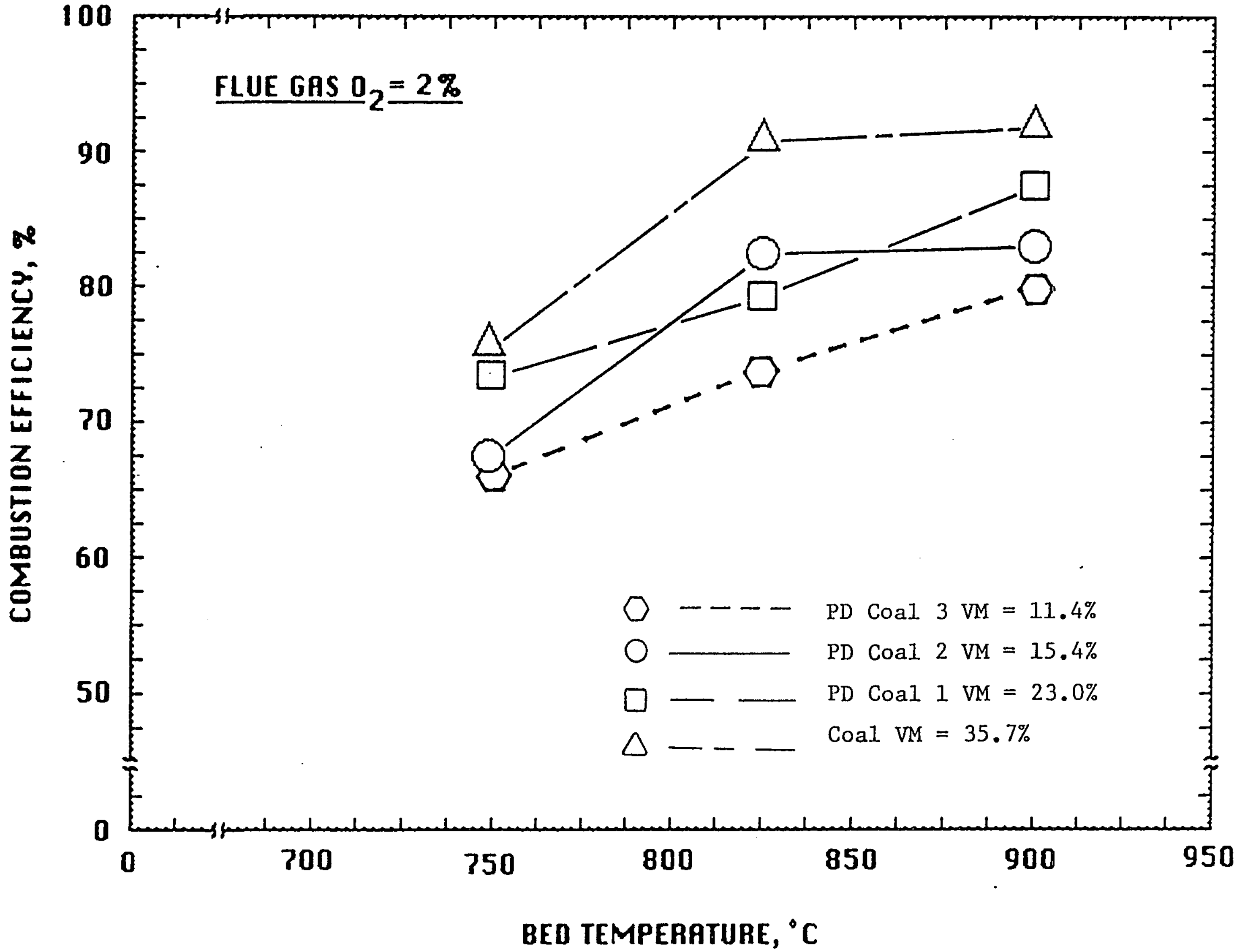
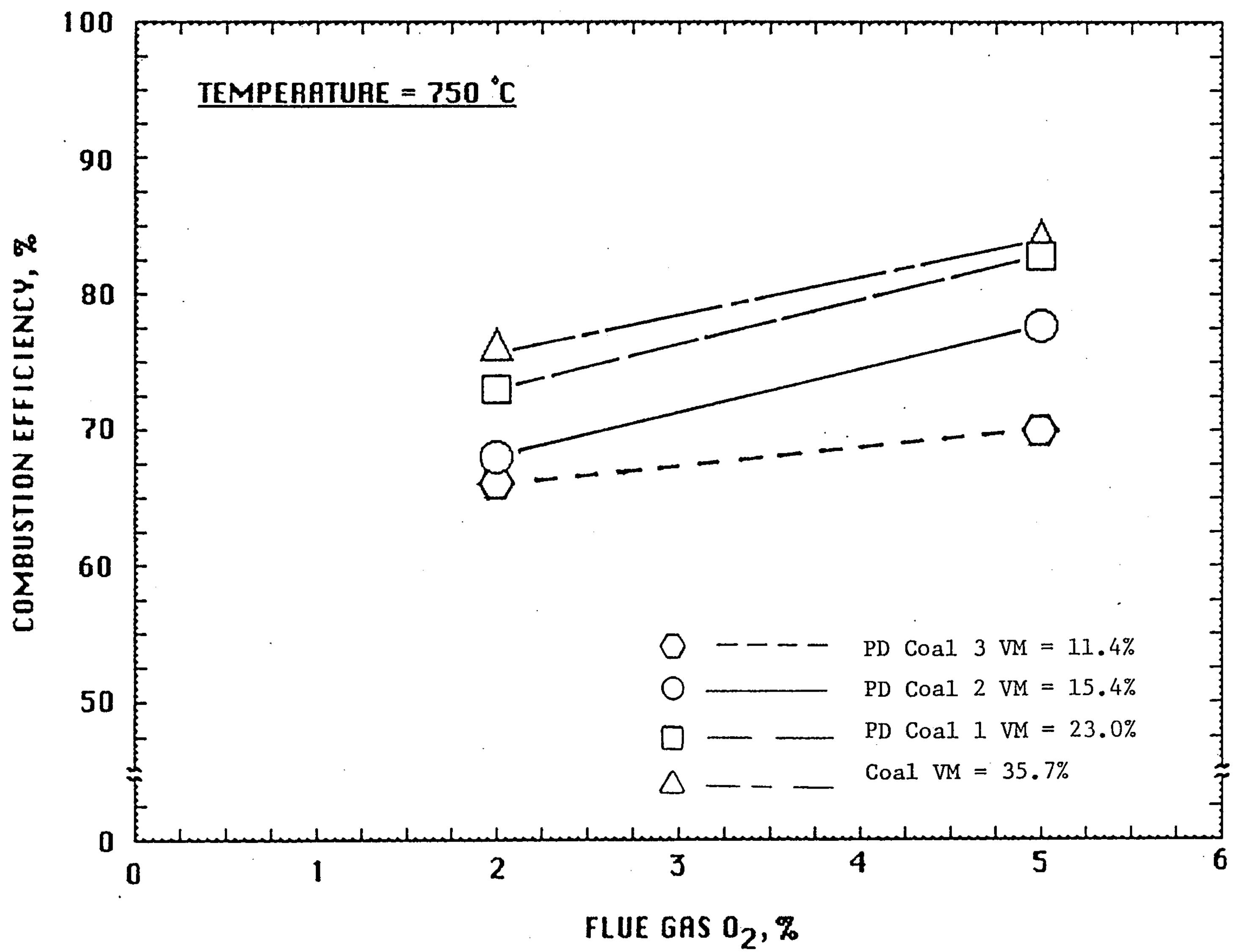

FLUE GAS O2 = 2%
PD Coal 3 VM = 11.4%
PD Coal 2 VM = 15.4%
PD Coal 1 VM = 23.0%
Coal VM = 35.7%
COMBUSTION EFFICIENCY, %
100
90
80
70
60
50
0
0
700
750
800
850
900
950
BED TEMPERATURE, °C

FIGURE 3



100
90
80
70
60
50
0
COMBUSTION EFFICIENCY, %
TEMPERATURE = 750 °C
PD Coal 3 VM = 11.4%
PD Coal 2 VM = 15.4%
PD Coal 1 VM = 23.0%
Coal VM = 35.7%
0
1
2
3
4
5
6
FLUE GAS O2, %

FIGURE 4

CONCLUSIONS AND RECOMMENDATIONS

Tests were successfully completed to determine the combustion characteristics of an Illinois coal (IBCSP Coal #3) and three partially devolatilized fuels prepared from the parent coal. Specific conclusions based on the results of tests in the ANL 6-in.-dia atmospheric fluidized-bed combustor can be summarized as follows:

(1) The combustion of solid fuels in a fluidized-bed combustor is influenced by the volatiles content of the fuel with carbon conversion increasing with increasing volatiles

(2) Combustion efficiency improves with increasing volatile content, excess air and bed temperature

(3) The fluidized-bed combustion results are consistent with TGA reactivity testing of the four test fuels

(4) The measured combustion efficiencies (between 66 and 92%) should not be considered representative of those achievable in larger diameter FBC units at optimized conditions of bed temperature, excess air and gas residence time.

It is recommended that the combustion characteristics of a low-volatile fuel (<15%) be determined in a pulverized coal burner to determine the suitability of partially devolatilized fuels as industrial and/or utility boiler fuels.