TECHNICAL REPORT
September 1 through November 30, 1994

Project Title: **COMBUSTION OF CHAR-COAL WASTE PELLETS FOR HIGH EFFICIENCY AND LOW NO_x**

DOE Cooperative Agreement Number: DE-FC22-92PC92521 (Year 3)
ICCI Project Number: 94-1/5.2A-1M
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ABSTRACT

Illinois coals are prime candidates for use in Integrated Gasification Combined Cycle (IGCC) plants because of their high volatility and good char reactivity. In these plants, partial gasification of the coal in the presence of limestone eliminates the major portion of the sulfur species in the product gases, which are used as fuel for the topping cycle. The char produced is high in ash content, the major portion of which is calcium sulfide. It is also low in volatiles and of low density, compared to the parent coal. The economic success of the gasification route depends on the subsequent utilization of the residual char for raising steam for use in a Rankine cycle bottoming plant and/or preheating the air to the gasifier. Fluidized bed combustion of the char appears an attractive way of utilizing the char. Areas of concern in the fluidized bed combustion of the high ash, low volatility char are:

1. attainment of high carbon conversion efficiencies,
2. reduction of oxides of nitrogen emissions,
3. reduction/elimination of corrosive chlorine species,
4. reduction/elimination of sodium and other alkali species and
5. efficient usage of the calcium present in the ash to reduce sulfur compounds.

The aim of the present project is to investigate ways of improving the carbon conversion efficiency, sulfur capture efficiency and NO_x reduction during the fluidized bed combustion by pelletizing the low density char with coal and coal wastes using cornstarch or wood lignin as binder.

During this first quarter, the parent coals and the chars to be tested have been analyzed. Particle size distributions have been measured. Sample pellets have been made for evaluation of their properties.

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EXECUTIVE SUMMARY

The overall efficiency of 48-50% projected for high efficiency power plants systems (HIPPS) are realized only if the low volatile gasification char residue can be burnt efficiently. The low char density makes it difficult to secure the longer combustor residence times required for high carbon conversion, the problem being compounded by the loss of the more easily reacted volatiles. These difficulties could be mitigated by pelletizing the char with waste coals from gob piles or flotation columns.

Char produced by Foster Wheeler Development Corporation (FWDC) in their work for DOE under the HIPPS and Combustion 2000 programs is utilized in this research. To improve its combustion performance, the char is pelletized with Illinois waste coals to increase its residence time and carbon conversion efficiency. The reduction in SO_2 and NO_x emissions including nitrous oxide (N_2O) is evaluated. Chars currently being produced at Southern Illinois University at Carbondale in support of FWDC research may also be used in the experiments.

The objectives of the research are:

1. to pelletize the low volatile pyrolyzer char that has been obtained from FWDC with coal wastes using cornstarch or lignin as binder.
2. to conduct combustion experiments with the char pellets in a bench scale circulating fluidized bed combustor and measure carbon conversion efficiencies and SO_2 , NO_x , N_2O , HCl , and other emissions.
3. to conduct similar experiments with the char alone.
4. to demonstrate the increased carbon conversion efficiency and lower SO_2 , NO_x and other emissions obtainable from the char-waste coal pellets.

The combustion evaluation is conducted in a laboratory scale fluidized bed combustor. The experiments measure the carbon conversion efficiencies of these pellet fuels under typical fluidized bed operating conditions of bed temperature and excess air ratio. The influence of Ca/s mole ratios is investigated with a selected Illinois limestone. Sulfur dioxide, oxygen, carbon monoxide, carbon dioxide, oxides of nitrogen emissions are continuously monitored during the tests. Emissions levels of HCl will be measured. Baseline experiments under similar conditions of operation will be performed with a standard coal. The mineral matter content of the spent limestone and ash will be analyzed to study the distribution of any hazardous materials in the ash.

Towards achieving these goals, two chars obtained from FWDC have been analyzed, along with their parent coals. One char is produced from an Illinois No. 6 coal and the other from a Pittsburgh No. 8 coal. The chars and the parent coals have been analyzed for their elemental composition. Particle size analysis of the fuels has been made. The char has been pelletized with various percentages of the original coal using 15% cornstarch as binder. Properties of the pellets are being evaluated.

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OBJECTIVES

The objectives of the research are:

1. to pelletize the low volatile pyrolyzer char from Illinois coals obtained from FWDC with coal wastes using cornstarch or lignin as binder.
2. to conduct combustion experiments with the char pellets in a bench scale circulating fluidized bed combustor and measure carbon conversion efficiencies and SO_2 , NO_x , N_2O , HCl , and other emissions.
3. to conduct similar experiments with the char alone.
4. to demonstrate the increased carbon conversion efficiency and lower SO_2 , NO_x and other emissions obtainable from the char-waste coal pellets.
5. to investigate the effects of cornstarch and lignin as binders.
6. to analyze the ash and spent limestone residues with a view to proposing waste disposal strategies.

INTRODUCTION

Illinois coals have good potential for use in advanced High Efficiency Power Plants (HPPs) because of their good gasification properties and high reactivity. Companies such as Foster Wheeler Development Corporation and others are currently involved in developing such High Efficiency Power Plants. The approach here is to partially gasify the coal in a pyrolyzer producing a fuel gas which will power the topping cycle gas turbine. The residual char will then be burnt to raise steam for the Rankine cycle bottoming plant.

Because the char is low in volatiles and its density is lower than the original coal, it tends to elutriate from the bed during fluidized bed combustion and carbon conversion efficiencies are reduced. The work proposed here seeks to improve the char carbon conversion efficiency while also finding an end-use for waste coals from gob piles. This is accomplished by pelletizing the char with the gob pile wastes using cornstarch or wood lignin as binder. Additional limestone may be added to the pellets as necessary. The char pellets will be burnt in a 4-in. internal diameter circulating fluidized bed combustor to investigate carbon conversion efficiencies, SO_2 , NO_x and HCl emissions. The results will be correlated with other literature data. The use of char from Foster Wheeler Development Corporation, a leading boiler manufacturing contractor to DOE on these IGCC projects, provides a direct link to near term commercialization of this technology. The successful utilization of Illinois high sulfur coals via IGCC plants will provide near term economic benefits to the coal industry by overcoming the roadblocks currently placed upon it by the current stringent Environmental Protection Agency (EPA) emissions requirements. The high volatility and good reactivity of Illinois coals make it a viable coal for IGCC applications, with good opportunities for success. The enhanced char-pellet combustion, emissions and reactivity data obtained from the research in the bench scale experiments will make Illinois coals more attractive for these IGCC applications. The research will extend the database and permit high efficiency IGCC plants to be designed and fired with Illinois high sulfur, high chlorine coals.

In particular, the research will

- (a) reduce the difficulties in burning the low volatility char
- (b) ensure overall high plant efficiency which is not possible without the char utilization
- (c) promote lower emissions of SO_2 , NO_x , N_2O from char combustion

EXPERIMENTAL PROCEDURES

I. Equipment and Instrumentation

The experiments are being conducted in the 4" internal diameter circulating fluidized bed combustor shown schematically in Figure 1. The combustor is lined with a castable refractory to reduce heat losses. As shown in Figure 1, a blower supplies fluidizing air that is split into two streams. The main stream enters the fast fluidized bed section of the combustor through a distributor plate specially designed to provide even fluidization. This section of the air duct also houses a propane-fired preheat system, which is utilized to bring the bed solids up to temperatures required to ignite the main fuel. Unburnt fuel, limestone and ash entrained by the gases in the main bed column pass through a refractory-lined hot cyclone, which traps the larger particles and deposits them into an auxiliary bubbling bed attached to the bottom end of the hot cyclone. The second smaller air stream enters this bubbling bed into which the carry-over solids from the fast fluidized bed trapped by the hot cyclone are deposited. A non-mechanical seal ensures that this unburnt fuel and bed solids flow from the bubbling bed into the fast fluidized bed and not vice-versa. Both air streams are metered with ASME nozzles and incorporate control valves for adjusting the flow velocities in the fast fluidizing and bubbling bed sections of the combustor.

Crushed and sieved coal is fed from a pressurized hopper via a screw feeder pneumatically into the dense portion of the fast fluidized bed, using metered high pressure air. Sized limestone, stored in a separate hopper, is fed simultaneously into the air stream, conveying the coal into the bed. Both coal and limestone feed systems have been calibrated individually.

Figure 1. Schematic of 4-Inch Internal Diameter Circulating Fluidized Bed Combustor

Two quartz, glass-lined observation ports, one located in the dense bed at the bottom, and the other located near the top in the dilute phase or transport section of the bed, serve for visual monitoring of the combustion process. The circulating fluidized bed combustor is instrumented with chromel-alumel thermocouples at various positions for measuring temperature. The thermocouples are connected to a selector switch and, thence, to a digital readout meter.

Solids too small to be captured by the hot cyclone are trapped in a multiclone, mounted at the hot cyclone exit. In the present system, these multiclone solids are not reinjected into the bed. The multiclone solids are later analyzed for heat content, using an adiabatic calorimeter. Combustion gases are drawn off from a point at the exit of the multiclone, filtered through 2-5 micron particulate filters, and conveyed via heated lines to an instrument panel for determining gas composition. Carbon monoxide and carbon dioxide are measured with Beckman NDIR analyzers, oxygen with a Beckman 755 paramagnetic analyzer, oxides of nitrogen, NO_x , with a Thermoelectron 10 AR chemiluminescent analyzer and sulfur dioxide with a Beckman IR analyzer. HCl is measured with a Thermoelectron gas filter correlation hydrogen chloride analyzer.

II. Test Procedures

CFBC Combustion and Emissions Tests

The combustion testing of the pellets involves the following steps:

- * The CO , CO_2 , O_2 , NO_x and SO_2 analyzers are calibrated at the beginning and at several times during a test burn.
- * The CFBC combustor is filled with the proper amount of bed material (sand or limestone).
- * The propane preheat system is fired the bed material and unit is brought up to about $1100\text{--}1200^\circ\text{F}$. This step takes several hours.
- * Coal and limestone hoppers are filled with prepared standard coal and limestone sorbent, respectively.
- * The coal feed is initiated and the CFBC unit is brought up to operating temperatures of around 1500°F on the standard coal. The operation of all sampling and control systems are checked.
- * For tests with the parent coals and the devolatilized chars, typical values of operating variables are as follows:

fluidization velocity 9 ft/sec
Ca/s ratio 1-4
Bed temperature $\approx 1450\text{--}1650^\circ\text{F}$

These parameters are kept constant with all the fuels, so that comparison of the combustion and emissions parameters can be made under identical conditions of operation.

- * No additional limestone sorbent will be injected during initial tests. If SO_2 emissions are higher than EPA limits, further tests will be conducted with limestone injection.
- * Six to ten test runs are planned to be made. Each test run is made after the combustor has reached steady state conditions. Combustor steady state conditions are usually achieved after 30-48 hours of operation. Where test fuel supplies are limited, the procedure adopted is to first bring the combustor to steady state operation on the standard coal or another Illinois coal, and then change the fuel feed to the test coal, only for the duration of the steady state data acquisition period.
- * The variables measured during a test include:
 - fuel and air mass flows
 - air superficial velocity
 - bed temperature
 - other temperatures at various combustor locations
 - combustion gas analysis comprised of CO , CO_2 , O_2 , NO_x , HCl and SO_2 emissions

- test duration time
- quantity of ash collected in cyclones during test period

Combustion generated ash and spent limestone from the experiments are analyzed. The heat content of the elutriated unburnt carbon is determined from calorimetry tests. Spent limestone and ash are prepared on metal stubs and subjected to energy dispersive x-ray (EDX) analysis to determine the elements present in the samples.

Sample Analysis

(a) Proximate and Ultimate Analyses

Proximate and ultimate analyses of the parent coals and chars are obtained using standard ASTM procedures at the Coal Technology Laboratory at Carterville, Illinois.

(b) Particle Size Analysis

Particle size analysis in the range below 125 microns is measured utilizing a Leeds and Northrop Microtrak Model 7995-10 particle size analyzer. A schematic of the instrument is shown in Figure 2. In this version of the instrument, a laser beam is projected through a transparent cell that contains a stream of moving particles suspended in a liquid. Light rays that strike particles are scattered through angles that are inversely proportional to their sizes. The rotating optical filter transmits light at a number of predetermined angles and directs it to a photodetector. Electrical signals proportional to the transmitted light flux values are processed by a microcomputer system to form a multichannel histogram of the particle size distribution.

Figure 2. Schematic of Microtrak Particle Size Measurement System

(c) Mineral Matter Analysis

The mineral matter analysis of the coal in the pellet fuels and the reference Illinois No. 6 coal is conducted with a Hitachi H-600 analytical electron microscope operating both in the transmission and the scanning-transmission electron microscopy (STEM) modes. With STEM, a Tracor-Northern energy dispersive x-ray (EDX) Model 5500 analysis system was employed. The specimen samples were mounted on adhesive copper grids and examined at 100 kV in the electron microscope. The samples were uncoated.

Data Analysis

From the measured data the following parameters will be computed:

- * excess-air ratios
- * Ca/s mole ratios
- * carbon conversion efficiency
- * sulfur capture efficiency %
- * SO₂ emissions levels in lb/10⁶ Btu
- * NO_x emissions levels in lbs/10⁶ Btu
- * HCl emissions levels in lbs/10⁶ Btu
- * carbon balances

RESULTS AND DISCUSSION

1. Fuels Procurement and Analysis

Two coals and chars produced from them by Foster Wheeler Development Corporation (FWDC) in their pyrolyzer have been initially selected for experimentation under this program. One of the coals is from an Illinois No. 6 seam, and the other is from a Pittsburgh No. 8 seam. The chars are produced by FWDC and have been collected from the primary cyclone drain location of the pyrolyzer. These fuels have already been supplied by FWDC under the previous year's work funded by ICCI.

2. Elemental Analysis of the Fuels

The elemental analysis of the coals and chars are given in Table 1.

Table 1

Elemental Analysis of Test Fuels

	Illinois No. 6 Coal	Pittsburgh No. 8 Coal	Illinois coal char	Pittsburgh coal char
Carbon	63.90	67.73	63.12	61.00
Hydrogen	4.41	4.31	0.56	0.49
Nitrogen	1.54	1.86	0.92	1.08
Sulfur	2.66	4.44	1.78	3.31
Oxygen	8.18	4.74	----	----
Ash	11.00	13.78	32.83	33.12
Moisture	8.31	3.14	0.94	1.00
HHV Btu/lb	11,550	12,200	10,035	9,420

3. Fuels Particle Size Analysis

The particle size analysis of the parent coals are shown in Figures 3 and 4. Figures 3 and 4 show that 30% of the Illinois and Pittsburgh parent coals are less than 100 microns in size and that their size distribution is essentially similar. This increases the elutriation rates of these coals and could result in lower conversion efficiencies.

The particle size distribution for the Illinois and Pittsburgh coal chars collected at the primary cyclone drain are shown in Figures 5 and 6, respectively. Fully 50% of the Illinois sample is less than 200 microns in size. The primary cyclone Pittsburgh char is even smaller, with 50% less than 100 microns.

4. Pellet Composition and Structure

During this quarter, pellets were made with the chars mixed in various proportions with the parent coals. Typical mixtures were

20% coal, 80% char
30% coal, 70% char
0% coal, 100% char

Fifteen percent cornstarch was used as the binder. These pellets were made with very low compaction pressures to determine the influence of pressure on mechanical strength. The size of the pellets was typically 0.25 inch diameter and 0.5 inches long. Figures 7-9 show pellets made in the above proportions with one of the chars.

In other tests, pellets are being made with higher compaction pressures. Also, gob waste coal is used instead of the parent coal as the blending fuel.

During the next quarter, the proper proportions for the pellet composition will be determined. Also, wood lignin as binder will be investigated. With data from these bench scale pellet experiments, larger quantities of pellets for combustion testing will be produced using commercial pellet forming mill services.

CONCLUSIONS

During this quarter, chars obtained from Foster Wheeler Development Corporation and produced in their pyrolyzer under DOE contract have been analyzed, and sample pellets have been made using cornstarch as binder. The fuel blended with the char was the original parent coal.

During the next quarter, wood lignin as a binder will be investigated. The use of gob coal as a blending fuel will be studied. The final specifications for the pellets will be determined, and sufficient quantities of pellets will be made using commercial pellet mill services in preparation for combustion testing.

Figure 3. Particle Size Analysis of Illinois No. 6 Parent Coal

Figure 4. Particle Size Analysis of Pittsburgh No. 8 Parent Coal

Figure 5. Particle Size Analysis of Pyrolyzer Char from Illinois Coal

Figure 6. Particle Size Analysis of Pyrolyzer Char from Pittsburgh Coal

Figure 7. Char Pellets Containing 20% Coal and 15% Cornstarch as Binder

Figure 8. Char Pellets Containing 30% Coal and 15% Cornstarch as Binder

Figure 9. Char Pellets with No Blended Coal; 15% Cornstarch Used as Binder

DISCLAIMER STATEMENT

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