ABSTRACT

To gain market share, new uses must be found for Illinois coals in the largest end use area, namely power generation. One promising avenue in this direction is to ensure that these high sulfur coals can be utilized in the next generation of high efficiency power plants, which are based on combined cycles. This approach involves partial gasification of the coal to produce fuel gas for firing the topping cycle gas turbines, while the residual char is burnt in the gas turbine exhaust to produce steam for the Rankine bottoming cycle.

Combustion of the residual char from the pyrolyzer in a fluidized bed unit is an attractive way of utilizing this char. Areas of concern in the fluidized bed combustion of the high ash low volatility char are

1. Attainment of high carbon conversion efficiencies, because the char is of low density; and residence times for combustion need to be longer;

2. Reduction of nitrogen oxides; and

3. Efficient usage of the calcium present in the char to reduce sulfur compounds.

The overall objective of the project, therefore, was to attempt to solve some of these problems. To increase residence times, the char is pelletized with a gob waste coal, which also increases the overall volatiles content. Because of the char reduction properties, the oxides of nitrogen are also expected to be reduced.

In this project, the above advantages of increased carbon conversion efficiency and low oxides of sulfur and nitrogen emissions have been demonstrated in a bench scale circulating fluidized bed combustor. Char-gob waste pellet formation and compressive strength characteristics have been investigated, and the results show that about 10-15 weight percent of cornstarch binder produces the right compressive strength for pellet handling and feeding. Fluidized bed combustion tests have been performed with three pellet compositions and the gob coal. The results are summarized below.

1. Utilizing just the calcium present as calcium sulfide in the char to reduce sulfur dioxide emissions from the pellet requires careful selection of the char to gob waste coal mass ratio. In this case, a 70% char-40% gob coal gave the lowest sulfur dioxide emissions.

2. By incorporating additional limestone in the pellets, sulfur dioxide emissions are markedly reduced, and there is no difficulty in meeting environmental regulations.

3. Combustion efficiency of the pellets is extremely good being on the order of 98% or higher.

4. The char does indeed have a beneficial effect in reducing the overall oxides of nitrogen emissions.
EXECUTIVE SUMMARY

Advanced power generation schemes using a carbonizer in conjunction with pressurized fluidized bed combustors offer definite advantages in terms of efficiency gains compared to single cycle units. The fuel gas produced in the carbonizer can be utilized as the heat input to the Brayton topping cycle, while the residual char from the carbonizer can be burnt in the Brayton cycle exhaust gas to raise steam for the Rankine bottoming cycle. To investigate the suitability of Illinois and other coals for these advanced power cycles, an investigation was conducted recently under ICCI sponsorship in collaboration with Foster Wheeler Development Corporation, wherein chars taken from different locations in the carbonizer were combusted in an atmospheric fluidized bed combustor. The combustion efficiency and the sulfur dioxide and NOx emissions from the chars were measured. It was found that the combustion efficiency of the chars could be improved by increasing the combustor residence time of these fuels. The present work, which is an extension of the one mentioned above, investigates methods of improving the char burning characteristics while reducing sulfur dioxide and oxides of nitrogen emissions.

Thus, the research objectives 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 SO2, NOx, N2O, HCl, and other emissions.
3. To conduct similar experiments with the char alone.
4. To demonstrate the increased carbon conversion efficiency and lower SO2, NOx and other emissions obtainable from the char-waste coal pellets.

To achieve these objectives, the use of waste gob coal has been investigated as a blending component with the char. This will utilize a waste fuel while improving the combustion properties of the char.

Pellet Composition and Compressive Strength

Experiments were conducted to determine the various proportions of char, gob waste and binder needed to produce pellets suitable for combustion. In addition, both wood lignin and cornstarch were tested as binders. It was found that cornstarch is better suited for holding together the char-gob waste pellets than wood lignin. Different amounts of both distilled and tap water was used in the experiments to study if distilled water offered any advantages.

Two sizes of pellets were made. Single pellets 0.5 inches in diameter and 0.5 inches long were made in a die press to study the influence of

(a) die force
(b) water type (distilled vs. tap water)
(c) amount of binder

on the compression strength of the pellets. These pellets did not have any other blended fuel. This was done to obtain baseline data with the char alone before blending.

The results from these tests show that ordinary tap water is just as suitable for use in the pellet-forming process than distilled water. The data also indicates that for the low density, powdery pyrolyzer char used in the experiments, no gain in compressive strength is realized by increasing the binder concentration over 10% when the char is pelletized without any other blended fuel.
To investigate the influence of gob coal as a blending agent, a second type of pellet 0.125 inches in diameter, 0.3-0.5 inches long was made. Experiments were conducted with the char-gob coal pellets to investigate the influence of

(a) char/coal weight ratio

(b) binder concentration

on the pellet compression strength.

The results of these tests show that increasing the char content of the pellets in proportion to the gob coal reduces the compression strength. Increasing the percentage of binder, however, increases the pellet compressive strength. No limestone was used in these pellets as the char itself contains calcium sulfide resulting from the use of dolomite or limestone during the carbonizing process.

Having established the char-gob coal blending proportions and the binder concentration necessary for making pellets suitable for combustion, pellets were then made in larger quantities needed for the combustion tests. These pellets were 0.125 inches in diameter and 0.375 inches long, and were produced at the California Pellet Mill Company in Crawfordsville, Indiana.

Combustion and Emissions Characteristics of Pellets

The combustion and emissions properties of the pellets were studied in a bench scale circulating fluidized bed combustor. The CO, CO₂, SO₂, NOₓ and O₂ emissions were measured. The combustion efficiency was evaluated. The results of the combustion tests show that

1. The combustion tests show a synergistic effect on the release and subsequent capture of sulfur species. In the present tests, the 70% char-30% gob coal pellets yielded lower sulfur dioxide values than the 60-40 and the 80-20 char to coal ratio pellets.

2. Incorporation of additional limestone into the pellet matrix while forming the charcoal pellets has a much stronger effect on sulfur dioxide reduction than feeding the limestone separately. In the present tests, it was possible to meet regulations by adding limestone into the 80-20 char to coal ratio pellets at a Ca/S ratio of 2:1.

3. Oxides of nitrogen emissions are, in general, reduced by the incorporation of char in the pellet matrix.

4. Compared to the combustion efficiency of the char alone, the char-gob coal pellets yielded remarkably high values of combustion efficiency, on the order of 99%.

In summary, pelletizing carbonizer char with another fuel like coal, even if it is a waste gob coal, overcomes the difficulties present in burning the char alone, such as low residence time and high sulfur dioxide emissions.

Pelletizing increases the residence time and significantly increases combustion efficiency. To control sulfur dioxide emissions to acceptable levels, the present tests show that incorporation of additional limestone into the pellet matrix is an effective way of achieving this result.
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$_2$O, 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 (HIPPs) 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 that 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 in these high efficiency power 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$_2$O from char combustion

EXPERIMENTAL PROCEDURES

I. Equipment and Instrumentation

The experiments are being conducted in the 4-inch 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 multclone, mounted at the hot cyclone exit. In the present system, these multclone solids are not reinjected into the bed. The multclone solids are later analyzed for heat content, using an adiabatic calorimeter. Combustion gases are drawn off from a point at the exit of the multclone, filtered through 2-5 micron particulate filters, and conveyed via heated lines to an instru-
ment 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 Thermo Electron 10 AR chemiluminescent analyzer and sulfur dioxide with a Beckman IR analyzer. HCl is measured with a Thermo Electron 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, and the bed material and unit is brought up to about 1100-1200°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 standard coals and the char-coal waste pellets, typical values of operating variables are as follows:
  
  fluidization velocity 9 ft/sec  
  Ca/S ratio 1-4  
  bed temperature≈1450-1650°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.
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 Tracer-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 %
* \( \text{SO}_2 \) emissions levels in lbs/10^6 Btu
* \( \text{NO}_x \) emissions levels in lbs/10^6 Btu
* \( \text{HCl} \) emissions levels in lbs/10^6 Btu
* Carbon balances

RESULTS AND DISCUSSION

1. Fuels Used

The experiments were conducted with a char produced from an Illinois No. 6 seam as this was the largest quantity of char available. The char was supplied by Foster Wheeler Development Corporation (FWDC). This char was blended with a gob coal also obtained from an Illinois source. The gob coal is used by the Southern Illinois Power Cooperative power plant at the Lake of Egypt in Southern Illinois. The limestone blended with the char and waste coal in certain pellet formulations was obtained locally.

The char and gob coal were pelletized using cornstarch as binder. This is a fine ground, pregelatinized corn meal with a water solubility of 15-30%. It was procured from the Lanhoff Grain Company in Danville, Illinois, and goes by the trade name of PCF 1000.

2. Elemental Analysis of Fuels

The elemental analysis of the parent Illinois coal and the pyrolyzer char as supplied by FWDC are given in Table 1, which also gives the analysis of the gob coal blended in the pellets. The limestone used in certain of the pellet formulations contained 96.3% calcium carbonate, 1.2% magnesium carbonate and the rest impurities.

<table>
<thead>
<tr>
<th></th>
<th>Illinois No. 6 Coal</th>
<th>Illinois Coal Char</th>
<th>Gob Coal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon</td>
<td>63.90</td>
<td>63.12</td>
<td>49.13</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>4.41</td>
<td>0.56</td>
<td>4.60</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>1.54</td>
<td>0.92</td>
<td>0.93</td>
</tr>
<tr>
<td>Sulfur</td>
<td>2.66</td>
<td>1.78</td>
<td>2.50</td>
</tr>
<tr>
<td>Oxygen</td>
<td>8.18</td>
<td>-----</td>
<td>2.38</td>
</tr>
<tr>
<td>Ash</td>
<td>11.00</td>
<td>32.83</td>
<td>21.04</td>
</tr>
<tr>
<td>Moisture</td>
<td>8.31</td>
<td>0.94</td>
<td>19.42</td>
</tr>
<tr>
<td>HHV Btu/lb</td>
<td>11,550</td>
<td>10,035</td>
<td>8161.5</td>
</tr>
</tbody>
</table>
3. Pelletization of Carbonizer Char

(a) Pellets with No Binder

Tests were conducted to obtain baseline data on char pelletization. Since the char is of low density with high porosity, the strength of pellets formed from char will be different from that formed with coal. Hence, these tests were conducted with the char without adding any other fuel component to it to examine the compaction pressures necessary for forming the pellets.

The pellets were made using a mold and die press and were 0.5 inch in diameter and 0.3 to 0.5 inches long. The char was produced from an Illinois No. 6 coal in the Foster Wheeler Development Corporation pyrolyzer.

The char was mixed with water and pellets were made from it. Both tap water and distilled water were used to see if the water type had any influence on the pellets. The pellets were then dried and their compression strength determined. The binder used was cornstarch. The compaction force was varied during the tests.

Figures 3 and 4 show the influence of die force used while making the pellet on the compression pressure that the pellet was able to withstand before fracture. The compression pressure measured in lbs/in² (psi) has been divided by the ratio of the diameter to the height of the pellet (d/h) as shown in the figures. This is labeled as “Comp. Load/(d/h) ratio” in the graphs. The figures show that as the compaction force is increased from 1000 lbs to 3000 lbs, the maximum compression pressure increases. However, beyond a compaction pressure of 3000 lbs, the compression pressure actually decreases. As seen from the figures, the use of distilled water does not offer any significant advantages over ordinary tap water.

(b) Influence of Binder Concentration on Pellet Strength

To investigate the influence of binder concentration on pellet strength, char pellets were made with cornstarch binder. Binder concentrations by weight of 5, 10 and 15 percent were investigated. Again, the pellet size is a nominal 0.5 inch diameter and 0.25 to 0.5 inches in height. The compressive strength (psi) of the dried pellets (which initially had 23% water) is plotted divided by the diameter/height (d/h) ratio for these pellets in Figures 5-7.

Figure 5 shows that the compressive pressure/(d/h) ratio for the pellets with 5% binder reaches its maximum value of about 710 psi at a die pressure of 3000 lbs. Further increase in compaction pressure does not produce any appreciable change in the compression strength. When the binder concentration is increased to 10% by weight, Figure 6, the compression strength is about 1400 psi, even at the low compaction force of 1000 lbs, which is much higher than that of the 5% binder pellets. With further increase in compaction force to 5000 lbs, the maximum compression load in psi increased to 2550 psi. Increasing the compaction die force further, however, only brought about a reduction in the compression strength to about 1050 psi.

These results indicate that 10% by weight binder in the char pellet gives the pellet good abrasion and attrition resistance with the particles of the char bound together at their surfaces of contact with adequate amounts of binder. With only 5% binder, not enough binder is present to give this bonding strength to the particles of char. With compaction pressures of over 5000 lbs, the compression strength reduces, possibly because the char particles are crushed in the pellet forming process and thereby lose their strength. With increase in binder concentration to 15%, Figure 7, there is a competing effect seen at die forces of over 5000 lbs. The char particles are being crushed on the one hand by the high compaction pressure, while on the other hand binder is being forced into the pores of the
Figure 3. Effect of Die Force on Compression Strength of Char Pellets Formulated with Distilled Water
Figure 4. Effect of Compaction Force on Compression Strength of Char Pellets Formulated with Tap Water
Figure 5. Effect of Compaction Force on Compression Strength of Char Pellets with 5% Cornstarch Binder
67 wt% Char, 23 wt% Water, 10 wt% PCF1000

Max. Comp. Load/ (do/h0 Retio), (psi)

Die Force, (lbf) (Thousands)

Figure 6. Effect of Compaction Force on Compression Strength of Char Pellets Containing 10% Cornstarch Binder
Figure 7. Effect of Compaction Force on Compression Strength of Char Pellets Containing 15% Cornstarch Binder
Figure 8. Effect of Binder Concentration on Compression Strength of Char-Gob Waste Pellets
char as the compaction force is increased beyond 5000 lbs. Hence, we see an increase in compression strength beyond 5000 lbs compaction pressure, Figure 7.

3. Pelletization of Carbonizer Char-Gob Coal Waste Mixtures

Having established the characteristics of the pellets made with char alone, experiments were conducted with the formulation of the char-gob coal waste pellets. The gob coal used was that being burnt at the Southern Illinois Power Cooperative Plant, at the Lake of Egypt in Marion, Illinois. The percentages of char and gob coal used were as follows:

<table>
<thead>
<tr>
<th>Char %</th>
<th>Gob Coal %</th>
</tr>
</thead>
<tbody>
<tr>
<td>30%</td>
<td>70%</td>
</tr>
<tr>
<td>60%</td>
<td>40%</td>
</tr>
<tr>
<td>90%</td>
<td>10%</td>
</tr>
</tbody>
</table>

Binder concentrations of 5, 10 and 15% were used with each char/coal combination.

These pellets were 0.125 inches in diameter, 0.4 to 0.5 inches long. They were made by mixing the char, gob coal and binder together with adequate amount of water to form a putty-like mixture. This was then formed into pellets by an extrusion process. The pellets were then let to dry. The dried pellets were tested in a compression tester. This data is shown in Figure 8. Again, the data presented is the same as that of the previous figures. The compression pressure has been divided by the (d/h) ratio of the pellets.

The results of Figure 8 show that the strength of the char-gob waste pellets increases as the binder concentration increases. Also, pellets containing more char than coal have lower compression strength.

4. Combustion and Emissions Evaluation

For the combustion testing of the pellets, sufficient quantities were made at the California Pellet Mill Company in Crawfordsville, Indiana. Utilizing the data from the laboratory tests described in the last section, pellets were made in the following formulations for combustion testing.

<table>
<thead>
<tr>
<th>Char %</th>
<th>Gob Coal %</th>
<th>Additional Limestone</th>
</tr>
</thead>
<tbody>
<tr>
<td>A 60</td>
<td>40</td>
<td>No</td>
</tr>
<tr>
<td>B 70</td>
<td>30</td>
<td>No</td>
</tr>
<tr>
<td>C 80</td>
<td>20</td>
<td>No</td>
</tr>
<tr>
<td>D 80</td>
<td>20</td>
<td>Yes (Ca/S=2:1)</td>
</tr>
</tbody>
</table>

These pellets along with the gob coal were burnt in the CFBC unit.

(a) Sulfur Dioxide Emissions

Figures 9-12 show the sulfur dioxide emissions measured from the combustion of the pellets, while Figure 13 shows the SO₂ emissions for the gob coal alone. These results were obtained when the pellets and gob coal were burnt such that the excess air was 30% for each test. It is to be mentioned that the results presented are for comparison purposes only and that actual emissions from industrial size units would be different. It was found that, with no additional limestone added the sulfur dioxide emissions were quite high for the 60% char 40% coal pellets. The char does contain 1.78% sulfur in it as shown in Table 1. The ash in the char contains 5.04% sulfur in the form of calcium sulfide and almost 33% of the char is ash. The gob coal contains 21.04% ash and 2.53% sulfur and 24.52% volatiles. The kinetics of sulfur compounds released from the calcium sulfide of the char is much different from the kinetics of sulfur compounds released from the gob coal. These release rates depend on the temperatures, also. In addition, the sulfur compounds have to react with the calcium of the calcium sulfide in order to be converted to calcium sulfate. While the proximity of the calcium to the sulfur compounds in the
Figure 10. Effect of Bed Temperature on Sulfur Dioxide Emissions from 70-30 Char-to-Coal Ratio Pellets
Figure 11. Effect of Bed Temperature on Sulfur Dioxide Emissions from 80-20 Char-to-Coal Ratio Pellets (No Limestone in Pellets)
Figure 12. Effect of Bed Temperature on Sulfur Dioxide Emissions from 80-20 Char-to-Coal Ratio Pellets (Additional Limestone in Pellet; Ca/S=2:1)
Figure 13. Effect of Bed Temperature on Sulfur Dioxide Emissions from Gob Coal
pellets is good, it is known that high temperatures in the pellet are not conducive to sulfur oxides capture. These factors are illustrated in the results shown.

For the 60-40 pellets, the SO$_2$ emissions are quite high at normal fluidized bed operational temperatures of 1550°F. The addition of limestone fed separately (as shown by the triangle in Figure 9) reduces the SO$_2$ emissions to 3 lbs/10$^6$ Btu from 4.1 lbs/10$^6$ Btu at 1535°F. It is to be observed that these high SO$_2$ emissions result, because the fuels used are not high quality fuels. As seen from Figure 14, the char when burnt by itself yields about 2.5 lbs/10$^6$ Btu of sulfur dioxide. Gob combustion alone also yields high SO$_2$ emissions, Figure 13. Pelletized with the low quality gob coal, the SO$_2$ emissions are higher for the 60-40 pellet formulation, Figure 8, and the 80-20 pellet formulation, Figure 10, than for the 70-30 pellets. Since the sulfur release kinetics and capture mechanisms are quite complex, as mentioned above, the lower SO$_2$ emissions for the 70-30 pellet formulation perhaps indicates a synergistic effect, which tends to reduce SO$_2$ emissions.

In Figure 9-11, a data point is indicated, which illustrates the effect of feeding extra limestone at a Ca/S ratio of 2:1. The effect of adding this extra limestone is more pronounced when the SO$_2$ emissions are higher as in the case of the 60-40 and the 80-20 pellets.

However, if this extra limestone is mixed into the pellet matrix, there is a much stronger effect in reducing the SO$_2$ emissions. This is seen in Figure 12. This is possibly due to the lower temperatures in the pellet resulting from the endothermic calcining of the limestone. This effect promotes higher sulfur capture rates especially from the sulfur oxides resulting from the calcium sulfide decomposition. This is a very beneficial effect in utilizing the pyrolyzer char in pellet form. Figure 15 summarizes this data.

(b) Oxides of Nitrogen Emissions

The oxides of nitrogen (NO$_x$) emissions for the pelletized char are shown in Figures 16-18. Figure 19 shows the NO$_x$ emissions for the char alone, while Figure 20 shows that of the gob coal. Generally, the NO$_x$ emissions for the pellets are of the same order of magnitude. At fluidized bed temperatures, the NO$_x$ from the char alone, Figure 19, is lower than that from the pellets. This is to be expected from the strong reducing effect of the char. Hence, pelletizing the char with the gob coal should yield lower NO$_x$ values than from the gob alone. This was one of the objectives of this work, and Figures 16-18 do show this effect. The NO$_x$ emissions from the pellets are indeed lower than from the gob coal. However, it was found that in all the pellet combustion cases, the NO$_x$ levels reduced as the temperature increased, indicating that the char reduction mechanism is more effective at higher bed temperatures.

In the present tests, no hydrogen chloride emissions were detected.

(c) Combustion Efficiency

The results of the tests indicate that the carbon conversion efficiency of the char-gob coal pellets is extremely good. Previous tests showed that the carbon conversion efficiency of the char alone was on the order of 80-88%. This was because the char was of low density and low in volatiles, making it difficult to burn in the short residence times available to it in the combustor. These drawbacks are much ameliorated by pelletizing with the gob coal. Combustion efficiency values for the test fuels are as follows at 1550°F.

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Combustion Efficiency %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Char alone</td>
<td>86</td>
</tr>
<tr>
<td>60-40 pellets</td>
<td>98.5</td>
</tr>
<tr>
<td>70-30 pellets</td>
<td>99.3</td>
</tr>
<tr>
<td>80-20 pellets</td>
<td>98</td>
</tr>
<tr>
<td>Gob coal alone</td>
<td>98.6</td>
</tr>
</tbody>
</table>
Figure 14. Effect of Bed Temperature on Sulfur Dioxide Emissions from Char Alone
Figure 15. Comparison of Sulfur Dioxide Emission Data from the Pellets and the Gob Coal
Figure 16. Effect of Bed Temperature on Oxides of Nitrogen Emissions from 60-40 Char-to-Coal Ratio Pellets
Figure 17: Effect of Bed Temperature on Oxides of Nitrogen Emissions from 76-30 Char-to-Coal Ratio Pellets
Figure 18. Effect of Bed Temperature on Oxides of Nitrogen Emissions from 80-20 Char-to-Coal Ratio Pellets
Figure 20: Effect of Bed Temperature on Oxides of Nitrogen Emissions from Gob Coal
As seen from the above data, the objective of the research to demonstrate the higher carbon conversion efficiency of the char-gob coal pellets has been realized.

5. Combustion Residues Analysis

Figure 21 shows a typical histogram obtained from proton induced x-ray emission (PIXE) analysis of the combustion generated ash from the char combustion. The analysis was conducted by Elemental Analysis Corporation, Lexington, Kentucky. The results of comparing these histograms for the various fuels are discussed in this section. Samples of ash obtained at test temperatures of 1550-1600°F are studied by this method, to evaluate the transformations of sodium, potassium, iron, sulfur calcium, silicon, etc. As seen from the histogram, aluminum and silicon resulting from the aluminosilicate clays in the ash are the most abundant components of the ash. The next most abundant component is calcium. It was found that the concentrations of these components increase during combustion of the pellets as a result of the increased percentage of ash as present in the char-gob waste coal pellets compared to the char itself. When additional limestone is added to control sulfur dioxide emissions, the increase in calcium content in the ash increases proportionately. Also in these cases, the sulfur content of the ash increases. For both the char and the pellets, the combustion residues show an increased chlorine content, indicating that the chlorine released in gaseous form is negligible. Thus, no detectable levels of hydrogen chloride gas were observed. The potassium levels in the ash also increased from the pellet combustion. This indicates that potassium compounds react with other minerals and form salts retained in the ash. On the other hand, sodium levels decreased on combustion indicating that sodium compounds were released. In addition to these elements, the ash also contained appreciable amounts of iron, some titanium and small quantities of chromium, manganese, nickel, copper, zinc, bromine, strontium and lead. No arsenic was detected. Additional studies need to be performed to determine whether the levels of elements such as lead are harmful to the environment.

CONCLUSIONS

Work carried out in this project shows that

1. Carbonizer char obtained from the partial gasification of coal can be easily pelletized with waste gob coal with or without limestone addition.

2. Cornstarch is a better binder for these char-coal pellets than wood lignin.

3. Ten to fifteen percent of cornstarch by weight is adequate to give the char-coal pellets sufficient strength for handling and feeding.

4. The combustion tests show a synergistic effect on the release and subsequent capture of sulfur species. In the present tests, the 70% char-30% gob coal pellets yielded lower sulfur dioxide values than the 60-40 and the 80-20 char to coal ratio pellets.

5. Incorporation of additional limestone into the pellet matrix while forming the char-coal pellets has a much stronger effect on sulfur dioxide reduction than feeding the limestone separately. In the present tests, it was possible to meet regulations by adding limestone into the 80-20 char to coal ratio pellets at a Ca/S ratio of 2:1.

6. Oxides of nitrogen emissions are, in general, reduced by the incorporation of char in the pellet matrix.

7. Compared to the combustion efficiency of the char alone, the char-gob coal pellets yielded remarkably high values of combustion efficiency, on the order of 99%.

In summary, pelletizing carbonizer char with another fuel like coal, even if it is a waste gob coal, overcomes the difficulties present in burning the char alone, such as low residence time and high sulfur dioxide emissions.
Figure 21. Histogram of PIXE Data for Combustion Ash of the Illinois Coal Char
Pelletizing increases the residence time and significantly increases combustion efficiency. To control sulfur dioxide emissions to acceptable levels, the present tests show that incorporation of additional limestone into the pellet matrix is an effective way of achieving this result.

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Notice to Journalists and Publishers: If you borrow information from any part of this report, you must include a statement about the DOE and Illinois cost-sharing support of the project.
PROJECT MANAGEMENT REPORT
June 1 through August 31, 1995

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

DOE Cooperative Agreement Number: DE-FC22-92PC92521 (Year 3)
ICCI Project Number: 94-1/5.2A-1M
Principal Investigator: S. Rajan, Southern Illinois University at Carbondale
Project Manager: Frank Honea, Illinois Clean Coal Institute

Comments:
This project was completed on schedule and within cost.
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*Cumulative by Quarter
CUMULATIVE COSTS BY QUARTER - EXHIBIT C

Combustion of Char-Coal Waste Pellets for High Efficiency and Low NOx

Cumulative $ (thousands)

Months and Quarters

O = Projected Expenditures

Δ = Actual Expenditures

Total ICCI Award $66,112
The schedule for this one year project is shown below.

**PROJECT SCHEDULE**

A. Fuels Procurement
B. Fuels Analysis
C. Char-Coal Pellets Manufacture
D. CFBC Combustion Tests
E. Combustion Residues Analysis
F. Data Analysis
G. Final Report

Begin
Sept. 1,
1994