Experimental Development of a Multi-Solid Fluidized Bed Reactor Concept

Topical Report

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ABSTRACT

Battelle's Columbus Division is developing a coal mild gasification process based upon the Multi-Solid Fluidized bed reactor system to produce high quality liquid and gaseous products. This process uses 2-stages to gasify coal at high throughputs to produce a range of products in compact reactors without requiring an oxygen plant.

Preliminary bench scale experiments have shown that the relative amounts of liquid and gaseous products can be produced by adjustment of reactor temperature and conveying medium. Data generated on the process has shown that approximately 20 percent by weight of the incoming coal can be converted to liquid products and an additional 20 percent to a fuel gas with a heating value in the range of 600 Btu/SCF. The remainder of the coal is converted to a char coproduct that, if desired, can be used within the process to generate steam or be removed from the system as a product. The relative amounts of these coproducts can be adjusted by changing process operating conditions.

Sulfur in the coal exits the process primarily in the fuel gas. The fuel gas accounts for about 85 percent of the coal sulfur with another 10 percent contained in the liquid products. Typically, less than 5 percent of the coal sulfur exits from the combustion stage of the process.

The circulating medium in the MSFB process also provides unique potential for the process. By using catalytically active materials, the chemical composition of the liquids or gases produced can be potentially altered to minimize the amount of upgrading necessary. Chemically active circulating materials such as a sulfur scavenger to reduce or eliminate the requirement for sulfur removal from the products produced can also be used.

The relatively high gas velocities in the gasification reactor provide turbulent mixing in the base of the reactor thus reducing ash agglomeration and coal swelling problems. As a result the system can accept a range of coal types. This report describes initial tests in an integrated gasification unit as well as examines a preliminary process conceptual design.

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EXECUTIVE SUMMARY

The Multi-Solid Fluidized Bed (MSFB) gasifier provides a flexible advanced technology that produces high quality gaseous and liquid products at mild gasification conditions. The process utilizes a dual-circulating bed concept that can process coal at high throughputs, thus producing a range of products in compact reactors. The dual beds provide a means to produce high value liquid products while integrating the efficient utilization of char coproducts.

Battelle has undertaken a program to develop this technology for producing a range of products from high-quality liquids through gaseous fuels-to char and high temperature heat sources suitable for steam generation or process heating.

Objectives of Current Program Phase

The initial phase of the development program had as its objectives: (1) to obtain preliminary experimental data so a process conceptual design could be developed, (2) to generate comparative rate data on candidate coal feedstocks through thermogravimetric (TGA) analyses, and (3) to perform a preliminary process technical and economic analysis of an integrated system. These activities were planned to guide later phases of the program on a path toward commercialization of the technology. The continuing program objectives are to further develop the MSFB gasification process and to generate additional liquid production rate data. The other potential benefits of the process such as in-situ sulfur capture and product modification through catalytic operations will be investigated in later program phases. These objectives were met through continued operation in Battelle's integrated gasifier.

Status of Development Effort

A set of progressive bench-scale tests have generated initial performance data over a range of operating conditions. Initially, three raw

coals and two treated coals were evaluated in a TGA to determine relative reaction rates. Due to the inherently short residence times in the MSFB system, higher reactivity coals were expected to exhibit higher potential conversion to prime products. These tests provided the basis to determine which coal should initially be used for testing in the integrated gasification facility. Tests in a small, integrated gasification facility have demonstrated stable and continuous operation at high throughput (~ 1000 lb/hr ft²) with controlled solids circulation between the gasification and combustion reactors. Coal conversion to liquids was constant during most of the gasification tests at about 20 percent.

Increased liquid-product yield makes mild gasification an important step in producing transportation fuels from coal. More than 70 percent of the liquid products collected during the tests have a boiling point below 392 F (200 C) which suggests excellent potential for upgrading. In addition, the chemical makeup of the liquid product is low in oxygen thus further indicating straight forward upgrading.

Gaseous products are suitable for use as medium-Btu quality fuel (~ 600 Btu/SCF) or as feedstock to a chemical/petrochemical process. Carbon conversion to gas ranges from 5 to 25 percent as a function of gasifier temperature. At projected operating conditions of 1400 to 1600 F conversion to gas is about 20 percent.

An important process variable is the fate of sulfur species in the coal. Experimental results have shown that, unlike other mild gasification processes, nearly all of the coal sulfur is present in the fuel gas and the liquids with very little remaining in the char coproduct. Such a split of sulfur species greatly simplifies gas cleanup from the process. With other mild gasification processes, up to 50 percent of the coal sulfur would appear in the char coproduct.

A preliminary process design and economic evaluation was prepared for a 1000 tons of coal per day (TPD) mild coal gasification facility. The plant has the flexibility to produce a slate of products including 12,500 lb/hr of liquids at median 15 percent conversion to liquids and 179,500 SCFH of medium-Btu (650 Btu/SCF) gas. Char could be recovered or converted into steam or electricity depending upon the application. The projected capital

cost for the 1000 TPD plant is \$9 million without the cost of gas or steam turbines that might be desired.

Process Applications

The MSFB gasifier operates at 10 to 30 ft/sec superficial velocity to achieve turbulent mixing of coal and circulating solids. The prime products, liquid and fuel gas, are separated from the circulating solids in a cyclone and then go to product recovery and cleanup. The circulating solids are returned to the MSFB combustor which burns the unreacted char to provide heat for the gasifier. Integrated utilization of the char directly recovers energy from this gasification coproduct without requiring char cooling and handling equipment. Circulating solids are returned to the gasifier to control temperature and heat balance between the two vessels. The combustor flue gas is sent to a heat recovery system without diluting the prime products. Therefore, air can be used in the combustor with significant savings over systems that require oxygen and/or dilute the product gas system. The circulating solids can also serve the function of chemical reactants such as sulfur sorbent or catalytic agents to modify the product slate.

Fuel flexibility is provided by the turbulent mixing in the MSFB dense bed. Coal particle sizes up to 2 inches, including fines, can be fed into the MSFB gasifier reducing rejected material and fuel preparation costs. The integrated unit, for instance, has operated successfully with fine Illinois No. 6 coal without particle agglomeration that would normally be expected when using caking coals.

MSFB gasification has flexibility to satisfy several applications within the coal, chemical, petroleum, and utility industries. Each specific application has unique product requirements and operating conditions. An integrated coal production facility producing chemical feedstocks, transportation fuels, and electric power is a potential major change from present U.S. practice where the major coal users are utilities generating only power. As major coal companies increase their affiliation with petroleum and/or chemical companies, coal processing plants that produce high-value liquid products and fuel or synthesis gas while burning lower grade residues

to generate steam and/or electricity can become commonplace. As is the practice in the oil industry, total profit from the integrated operation would be maximized rather than trying to maximize yield from a single process step. The feasibility of using coal as a chemical and gasoline feedstock is realistic based on historical use of coal tar based chemicals. By use of systems such as the MSFB process, production of liquids suitable for upgrading to transportation fuels with valuable coproducts can be achieved economically and with environmental acceptability.

Future Activities

Continued testing and technical development of the MSFB gasification system is recommended to realize these benefits. The continued testing of additional coals as well as circulating agents for in-situ sulfur capture are recommended as steps toward the successful application of the process. Pilot plant testing is the next logical step in the development to provide detailed engineering data for scale-up and commercial application.

INTRODUCTION

Coal is a complex raw material, the potential of which has never been fully realized. In the early 20th century, coal was used as a fuel gas source and as a chemical feedstock. Current practice, however, due to the availability of natural gas and petroleum, has relegated coal to the role of a lower quality fuel in use primarily for power generation. As gas and oil become more scarce and expensive, coal must supply a wider slate of products. An integrated coal facility producing chemical feedstocks, transportation fuels, and electric power is a major change from present U.S. practice where the primary coal users are utilities generating a single product. The feasibility of using coal as a chemical and gasoline feedstock is realistic based on historical use of coal tar-based chemicals. New developments in technology are required to realize these goals. Battelle is developing such a system that can produce liquid fuels along with a slate of coproducts such as gas and high-value process heat. This development effort is part of the overall DOE mild gasification program. Mild gasification, in the MSFB process, includes rapid devolatilization in a continuous reactor to generate clean liquid and gaseous products. The Multi-Solid Fluidized Bed (MSFB) Coal Gasification process is a 2-stage circulating-bed coal system operating at high coal throughputs without an oxygen plant while producing a high liquid yield and a medium Btu (600 Btu/SCF) product gas. The basic concept is similar to previously developed MSFB combustion and biomass gasifier systems, and is conceptually illustrated in Figure 1.

The MSFB reactor, which operates at gas throughput velocities of 10 to 30 ft/sec utilizes a circulating burden of fine particles to transport heat between an exothermic zone and an endothermic zone. The MSFB has been commercially demonstrated as a steam generation process. When operating in this mode, the endothermic zone is an external heat exchanger in which the circulating burden is cooled by contact with boiler tubes. A similar circulating concept was subsequently applied to biomass gasification. In the biomass gasifier, the fuel is fed to the endothermic zone where it is partially gasified with steam and the residual char passes to a second reactor where it is combusted with air to provide heat for the gasifier. The present

FIGURE 1. MULTI-SOLID FLUIDIZED BED MILD GASIFICATION PROCESS

project is aimed at applying the same solids circulation principle to mild gasification of coal. This solids circulation can be used to modify liquid products produced catalytically as well as to capture sulfur within the gasification system itself. Since air is not used as a gasifying agent, condensible products are not degraded by oxidation. The gas leaving the gasifier is not diluted by nitrogen and combustion products, and therefore, is of much better quality than that produced by conventional air blown gasifiers. The elimination of an oxygen plant is a major process simplification and cost reduction. Compared with conventional fluid-bed air-blown gasifiers, reactor size is reduced due to high coal throughput/velocity resulting in additional capital cost savings. The high gas velocity produces turbulent mixing in the gasifier so that coal swelling and ash agglomeration problems are reduced. The system thus has fuel flexibility. The overall benefits of the MSFB process then are:

- Production of liquid products with valuable coproduct streams.
- Production of an undiluted fuel gas without the need for an associated oxygen plant.
- High coal throughputs resulting in compact reactors.
- Ability to circulate catalytically active or chemically active materials.
- Low capital investment.
- Ability to process a wide range of coal ranks without regard to agglomeration tendency.
- Efficient use of char coproduct within the process, if desired.

Background

Battelle initially developed the MSFB technology as a circulating fluidized bed coal combustor for industrial and utility boilers. MSFB combustors are commercially available from Battelle's licensees throughout the world. Operating experience from several large industrial boilers confirms the predicted performance and rapid acceptance by the users/owners. Many of

the technical benefits (throughput, mixing, fuel flexibility, reliability, etc.) that are being demonstrated in the combustion systems would directly apply to the coal mild gasification application once it has been developed.

The 2-stage Forest Residue Gasification system was subsequently developed under DOE funding for biomass feedstocks. This technology is being scaled-up for industrial demonstration and has significant advantages in cost, throughput, flexibility and product gas quality over alternative systems. The experience in integrating a circulating fluidized bed gasifier and combustor directly applies to the MSFB coal gasification system. The existing Process Research Unit used in the development of the Forest Residue Gasification system is planned as the experimental facility for a later phase of process development for the MSFB coal gasification process.

In addition to these applications, a 2-stage MSFB Oil Shale Retorting system was experimentally tested for DOE, again using the Forest Residue Gasification test facility. This work confirmed significant yield and cost advantages compared to current retorting techniques. A significant advantage in both the biomass gasification and shale oil processes was the rapid heating and short contact time of the product, which minimize thermal degradation.

These circulating fluidized bed developments provide an extensive background for investigating the MSFB coal gasification system.

System Concepts

The MSFB gasification system is generically similar to a fluidized bed cat-cracker. In the petroleum industry such reactor systems have been demonstrated to be an effective means for coupling endothermic and exothermic reaction zones. This process produces a variety of products depending on the conditions chosen for the two zones and the circulating catalyst.

The MSFB coal gasification process, when developed, can potentially serve as the same type of flexible reactor system. The potential then, would be a system that could produce a liquid product stream suitable as a refinery feedstock with various coproducts such as char, fuel gas, and process heat adjusted for particular processing requirements.

In the two reactor system illustrated in Figure 1, when operated with wood fuel, the amount of heat evolved by char combustion matches closely the heat needed for wood devolatilization and any imbalance can be corrected by minor changes in operating conditions. The concept of applying such a mild gasification process to coal is more complex because of the relatively large fixed carbon content of coal.

The simplest way of applying mild gasification would, on the surface appear to be to pyrolyze the coal and to burn the char in a steam generator. This method, however does not take advantage of the potential range of products possible from mild gasification. Figures 2 and 3 illustrate how, by using a 2-zone system such as the MSFB gasifier the potential exists to produce a variety of chemical and energy products.

Figure 2 is an outline flow sheet of a system in which the char is used for steam generation, and the fuel gas is used in a combined cycle power generating unit, which also utilizes heat recovered from the raw gas stream. In this case the liquid coproduct can be used as a peaking fuel to supplement the turbine fuel in times of heavy electric demand. Battelle's development partner during this program, Centerior Energy, intends to use the technology in this mode after pilot demonstration of the process.

Figure 3 shows another alternative use of the system with a partial oxidizer in place of the combustor producing additional fuel gas. This system then uses the additional energy available as char for production of additional fuel gas, thus, increasing the combined cycle capacity.

Figure 3 does not indicate the peripheral processes that would be involved in gas cleanup, ash disposal, liquids recovery, gas compression, etc., but in general these do not introduce critical complications. A separate catalytic liquefaction step may also be added to the complete gasification scheme, shown in Figure 3, which would permit recovery as liquid fuel of a higher fraction of the heating value of the coal. In this case the residual char would be gasified in air/steam or oxygen/steam, as shown in Figure 3. Gas from the partial oxidizer would then pass into a catalyst chamber for conversion to liquids. This would maximize the liquid yield at the expense of heat consumption in the system.

FIGURE 2. OUTLINE FLOW SHEET. MILD GASIFICATION FOR LIQUID YIELD AND STEAM POWER UNIT

FIGURE 3. OUTLINE FLOWSHEET. MILD GASIFICATION FOR LIQUID YIELD WITH GASIFICATION/COMBINED CYCLE POWER UNIT

By increasing the temperature in the first gasifier, it is possible to achieve higher conversion of fixed carbon at the expense of liquid production. Ultimately, if ash behavior were not prohibitive, the system could operate like the biomass gasifier already developed (in a thermally balanced mode), but at these high temperatures, the liquid yield would be reduced.

These schemes are relatively direct adaptations to conventional plants. In addition to the environmental controls afforded by fluidized bed combustion, an added benefit that may be expected is that nitrogen compounds would be expected to come off with the coal volatiles, permitting their removal before fuel gas combustion.

The practical value of the schemes outlined above depends critically on the quantity and quality of the liquids that can be produced. Devolatilization must be carried out under conditions giving a high yield of liquid products that are directly usable, or that can be modified by refinery treatment to produce high value end products. This yield may be increased by rapid processing to reduce product degradation, by use of catalytic agents, or by selection of the environment during devolatilization.

Objective

The overall development program has as its overall objective the determination of the preliminary technical and economic feasibility of the MSFB gasification system. The most recent program phase was to continue to meet this objective through activities in the following areas:

Subtask 4.2.1. Verification of Initial Test Data

Subtask 4.2.2. Testing of Additional Coal Types

Subtask 4.2.3. Evaluate In-Situ Sulfur Capture Methods

Subtask 4.2.4. Product Modification Through Catalytic Operation

Subtask 4.2.5. Product Modification Through Use of Recycled Product Gas

Previous program efforts consisted of three distinct tasks. These were: (1) to obtain comparative rate data on candidate coals through TGA testing; (2) to generate preliminary experimental data in a bench scale integrated facility so a preliminary conceptual design could be developed; and (3) to perform a preliminary process and economic analysis of the MSFB gasification process.

All of these objectives were planned to guide the later phase of the program toward commercialization of the technology.

Approach

This project phase included preparing a Test Plan, conducting integrated gasification tests in the small gasification unit, and revising the preliminary process design and economic analysis. The test plan proposed a progressive approach to screen selected coals and to examine key process variables in a limited test matrix.

Tests in the small integrated gasification facility allow a realistic and comprehensive screening of the operating conditions, and yield more complete data on the expected performance of the MSFB gasification system. The recent tests have confirmed earlier data that demonstrated the technical feasibility of the system.

A preliminary process design based upon MSFB gasification and the available data has been performed was reviewed by Centerior Energy Corporation and Thermo Electron Corporation. These teaming arrangements provide an "interested user" perspective to the power generation aspects of the evaluation based on the Integrated Gasification Combined Cycle concept. The evaluation considers a slate of coproducts that can be produced from the system.

The next phase of development of the project will continue testing in a larger Process Research Unit and investigate in-situ sulfur capture, catalytic modification of the coproducts, and further application of the technology to a variety of coals. Coal feed rate to this pilot plant will be approximately 100 to 500 lbs/hour. Operation in this unit will address the questions of coal particle size and residence time. The larger facility has relatively less heat loss and less dilution of the product stream from

nitrogen purges so that product recovery and yield should be determined more accurately than from the smaller unit. The tests run in the small integrated facility provide a confident basis for planning the tests in the PRU.

PROCESS DESCRIPTION

Battelle's 2-stage Multi-Solid Fluidized Bed (MSFB) Gasification process is based on advanced circulating fluidized bed technology (see Figure 1). The MSFB is a high-throughput device with potential to produce high-quality liquids and a medium-Btu gas by rapid devolatilization. The product gas is of sufficient quality that it can replace natural gas without extensive modifications to existing combustion equipment. Sulfur compounds present in the fuel gas must be removed from the fuel gas or from the products of combustion. These sulfur compounds present the only modifications necessary prior to use in natural gas-designed burners.

Coal is fed into the MSFB gasifier which operates at 10-30 ft/sec superficial gas velocity. Rapid heat-up of the charge is assured by the turbulent mixing and particle residence time in the gasifier zone which can be controlled by the gas velocity and circulation rates. The product gas is separated from the circulating solids in a cyclone and goes to product gas cleanup. Liquid products are condensed and separated for upgrading. The circulating solids are returned to a second MSFB operating as a combustor, which burns the unreacted char to provide heat for the gasifier. The reheated circulating solids are again recycled to the gasifier to transport thermal energy. A continuous heat balance between the two vessels is maintained by adjustment of the solids circulation rates which, in turn, provides control of the exothermic and endothermic processes. The combustor flue gas goes to a heat recovery system and is then exhausted from the process as a separate stream, thus not diluting the product gas. Therefore, air can be used in the combustor with significant savings over systems which require oxygen to avoid diluting the product gas stream.

The intensely turbulent mixing in the MSFB dense bed makes the system very tolerant to feed size. In combustion applications coal particle sizes up to 2-inch including fines can be fed into the MSFB, thus reducing

fuel preparation costs. Ash agglomeration problems are not experienced because of the turbulent mixing in the dense bed. Coal impregnated with lime has been found to provide enhanced reactivity, higher ash fusion temperature and in-situ sulfur capture.

Operating the MSFB Gasifier in the mild gasification mode, at temperatures ranging from 1200 F to 1500 F, is expected to provide rapid coal devolatilization with high liquid yield in a turbulent well mixed reactor. The reactor can use catalysts or in-situ sulfur sorbents as a circulating phase to produce high quality liquid and gaseous products which would require minimal upgrading. Flexibility exists to alter the product distribution to the products best suited for a specific application by variation of temperature, environment, throughput, and dense bed material. Char produced in the gasifier is used in the integrated combustor to produce heat for gasification and excess heat can be used to generate steam, electricity, or for other heating applications. Char handling and upgrading for subsequent use is not necessary as the circulating fluidized bed combustor is an integral part of the system.

The system is compact and the individual components can be shop fabricated for quality control and economic benefits. Modular unit design is consistent with phased construction or retrofit applications. The system can be fully integrated with automatic control systems for simplified operation.

EXPERIMENTAL FACILITIES

The initial experiments made in this Phase 1 program were conducted using a TGA (identical to the unit designed and built for METC in 1976) to obtain preliminary coal reactivity data followed by testing in an integrated coal gasification unit. The experimental equipment and general operating procedures for each are described below.

Thermogravimetric Analyzer (TGA)

The TGA, illustrated in Figure 4, consists of 2 zones; the upper zone houses the thermobalance assembly and the lower zone is used for conducting the reactions. The upper zone is constantly purged with helium during a gasification experiment to protect the thermobalance assembly from high temperatures. During preheating of the furnace the upper zone can be isolated from the lower zone by the use of a ball valve.

The reactor tube which is 3/4 inch I.D., 2-1/2 inch O.D., and 48 inches long is constructed of Haynes 188 superalloy. The sample basket is 5-3/4 inches long and has an inner diameter of 3/8 inch. It is made of 100 mesh stainless steel or rickel screen and is suspended by a 34-mil nichrome wire.

The sample basket can be raised and lowered at the rate of about 3/4 inch/second using a motor-operated windlass; less than a minute is required to lower the sample into the desired position. A dip tube that is constantly purged with helium extends from the upper zone to about 10 inches from the basket. The sample is not exposed to reaction conditions during most of the time it is being lowered into position, but is subjected to some heating during the last 10 to 15 seconds. Once the sample basket is in location, the sample mass is indicated by a transducer and the weight loss versus time is recorded.

Other parts of the system include a reactant gas preheater, a trap that can be operated at high temperature for capturing tar and particles, a back-pressure controller that can be operated up to 500 C, a condenser, and trap for condensate.

A proportional-integral-digital (PID) controller is used for controlling the furnace temperature at the desired value. The sample temperature is measured by a thermocouple placed 1/4 inch below the sample basket. An infrared detector is used to continuously monitor the methane concentration of the product gas. A gas chromatograph (GC) from Carle Instrument Company is used to determine the overall composition of product gases. Other analytical devices include gas flowmeters, a wet test meter, and a gas collection bag.

FIGURE 4. SCHEMATIC DIAGRAM OF THERMOGRAVIMETRIC ANALYZER (TGA)

The TGA was operated with a static gas environment within the reaction zone. This approach permits a more rapid screening of reactivity, as measured by sample weight loss versus time. The TGA data were used to provide an approximate indication of relative kinetic behavior. In this context, the use of more complicated, and therefore more expensive, test procedures was not warranted.

The general procedure for conducting a TGA test was to:

1. Preheat the furnace and reaction zone to the desired temperature while filling the reaction zone with the desired gaseous environment,
2. Place the prepared sample (approximately 2 grams) of coal in the sample basket and connect the basket to the suspension wire in the upper zone of the TGA,
3. Lower the sample into the lower reaction zone by operating the windlass,
4. Record the sample weight as a function of time; stop the test when the weight loss rate has stabilized,
5. Remove the sample and collect a bomb sample of the reaction zone gas for analyses,
6. Adjust conditions and inspect the suspension wire, replacing as required or following three tests, and prepare to repeat the sequence.

General experience in operating the TGA for this type of test indicate that mass balances typically do not provide good closure; sample quantities are very small, leading to significant imbalance due to minor analytical error. Because of this, the primary data obtained during TGA tests is the weight loss versus time to indicate coal reactivity.

Modular Integrated Gasification Unit

The small integrated gasification unit, illustrated in Figure 5, consists of a 2-inch diameter circulating bed gasifier, coupled to a 12-inch diameter bubbling bed combustor. The facility is constructed in such a way as

A

FIGURE 5. SMALL, INTEGRATED GASIFICATION UNIT

to allow rapid changes in configuration so that a number of process arrangements can be investigated within a short period of time. The 12-inch reactor is equipped with a natural gas fired start-up burner that supplies heated air to the reactor for initial heating of the bed. The air passes through a perforated plate distributor and then through the fluidized bed. For coal gasification experiments, the fluid bed was silica sand, a material that has been used successfully in the past as a heat transport medium.

The heated sand from the 12-inch reactor passes under gravity through a 2-inch diameter L-valve for solids flow control and then into the 2-inch circulating burden reactor. This reactor serves as the main gasification reactor for the program. The heated sand and the coal feed enter the base of the 2-inch reactor and are carried through the 10 foot length by a mixture of feed gas (steam or nitrogen) and product gas. The sand and unreacted char are separated from the gas phase by a cyclone and pass into the 12-inch reactor bed where the char is combusted with air. The product gas outlet passes on through a water quench, a xylene scrubbing system and then to a flare.

A 2-stage collection system was used to collect the liquid products from the integrated gasification unit. The water quench cooled the product stream to approximately 150 F and collected most of the heavy liquid fraction. The xylene scrubber absorbed the remaining hydrocarbons from the gas stream. This fraction of the condensed liquids requires additional processing to separate the xylene. During the most recent testing the xylene scrubber was replaced with a secondary water scrubber thus simplifying the recovery of the liquid products as shown in Figure 5A.

Both the 2- and 12-inch diameter reactors are constructed of type 330 stainless steel, a high temperature alloy, to allow rapid achievement of process conditions. To balance the inherently high heat losses in small-scale reactors, both reactors are surrounded by electric heaters and then a 3-inch layer of insulating wool blanket. This method has proven successful in producing a reactor that can accurately predict performance of large-scale gasification facilities.

Temperatures are measured at 20 locations in the facility. The 2-inch reactor contains 9 temperature points throughout its height and the 12-inch reactor contains 11 temperature points. The unit also measures system

pressure at a total of 16 different points thus providing an accurate temperature and pressure profile of the system.

A gas chromatograph and infrared analyzers for methane, CO, and O₂ provide on-line analysis of the two gas streams exiting the unit. A complete GC analysis is obtained at the rate of one sample every 20 minutes during testing to supplement the on-line analysis.

The integrated gasification facility was operated in the following manner:

1. Ignite the start-up burner and heat the reactor to a temperature of 1300 F.
2. Load a bed of sand into the 12-inch reactor and allow temperatures to recover.
3. Turn on the main gas to the 12-inch reactor and heat to the desired reaction temperature (usually 1600-1800 F).
4. Start steam or nitrogen flow to the 2-inch reactor, then start sand circulation to the 2-inch reactor.
5. Start the coal feed and adjust L valve flow to provide the required test conditions.

EXPERIMENTAL RESULTS

Tests were conducted in a TGA apparatus in order to determine the relative differences in the reactivities of the coals used and of the influence of calcium incorporation into the coal on reactivity. Direct utilization of TGA results to predict performance in reactors where the heat up rates are much faster than in the TGA and, where devolatilization is the main mode of conversion, is difficult. Nevertheless, TGA tests provide a rapid means of establishing relative reactivity differences in various coals as well as providing a continuous measure of the rate of conversion. Since the reaction rates in a TGA are continuously monitored, they provide insight into specific reaction mechanisms occurring during conversion not available from tests in a continuous gasification facility.

The results of TGA tests are by themselves unsuitable for use as scale-up data, and are not suited for collecting and measuring liquid

products. Therefore, continuous tests in an integrated gasification facility that is capable of providing steady-state data as well as the ability to collect enough liquid products for characterization are also necessary.

TGA Results

Illinois No. 6 coal was selected because of its frequent use for R&D activities. It represents the largest single coal deposit in the U.S. and is similar to many high-sulfur bituminous coals. The Wellmore No. 8 coal and the Mississippi lignite were chosen to compare test results with other DOE supported research studies. These specific coals have been tested recently by others and provide a broader reference for the present work.

Proximate and ultimate analyses of the three coals tested are included in Appendix A.

The objective of the TGA tests is to provide a relative screening of the feedstocks in terms of their reactivity and to examine the influence of temperature on conversion. During mild gasification these conditions are more moderate than in conventional gasification systems. In order to separate conversion due to devolatilization from conversion due to steam gasification, tests were conducted with both nitrogen and steam.

The TGA tests established the following:

- The relative conversions due to gasification and devolatilization.
- The relative reactivity of the different coals used and the effect of temperature on reactivity.
- Total conversion as a function of time.

In addition, previous TGA tests made with a simulated synthesis gas are compared with the present results.

In general, the TGA tests indicate that at the lower temperatures envisioned for mild gasification and with the high-throughput, low particle residence time reactor system employed, either reactive or catalyzed coals are necessary to achieve reasonable conversion.

Discussion of TGA Test Results

The primary parameters found, in the TGA experiments, to influence conversion were:

- Coal Type
- Temperature
- Calcium Addition
- Gasifying Medium (Steam or Nitrogen)

Figure 6 shows a typical TGA test trace result showing total weight losses as a function of time. This data is typical in that it suggests that, at constant steam partial pressures, the conversion rate is of the form

$$r = k (X - X_f)$$

where X_f represents the maximum fraction of carbon that can be converted and, as will be shown, is a function of coal type, temperature, calcium addition, and the method of incorporating the calcium into the coal.

Values of X_f were estimated from the plotted data by selecting the maximum conversion level achieved at any particular combination of TGA parameters. Table 1 summarizes the values of X_f .

The data indicate that conversion occurs both via devolatilization and gasification with steam. Between 1200 and 1500 F, the steam gasification rate is so slow that virtually all conversion is due to devolatilization. At 1500 F, gasification becomes significant and steam gasification becomes dominant at 1800 F. Comparison of these results with previous TGA tests with a simulated synthesis gas⁽¹⁾ show good agreement at the lower temperatures and also indicate that at the higher temperature, gasification reaction production (CO and H_2) can have a very significant retarding effect on the overall conversion rate.

FIGURE 6. TYPICAL TGA TRACE WEIGHT LOSS CURVE

TABLE 1. TGA TEST RESULTS

Run No.	Coal	Temp. (°F)	Reactant Gas	Weight Loss (%)	Weight Loss (%), MAF Basis
1-A	Wellmore	1800	Steam	43.7	46.6
2	Wellmore	1200	Steam	31.8	33.5
3	Wellmore	1200	Steam	32.6	34.4
4	Wellmore	1500	Steam	36.9	38.8
5	Wellmore	1800	N ₂	34.4	36.5
6	Wellmore	1200	N ₂	29.9	31.4
7-B	Miss Lignite	1200	Steam	63.2	49.6
8	Miss Lignite	1500	N ₂	67.1	56.6
9	Miss Lignite	1800	N ₂	69.4	60.9
10-A	Miss Lignite	1800	Steam	93.1	>100
11	Miss Lignite	1200	Steam	65.1	52.9
12	Miss Lignite	1500	Steam	86.2	91.6
13	Ill. #6	1200	N ₂	44.2	40.2
14	Ill. #6	1800	N ₂	49.9	47.4
15	Ill. #6	1200	Steam	45.1	41.4
16	Ill. #6	1500	Steam	49.5	46.9
17	Ill. #6	1800	Steam	89.8	98.2
18	CA Imp. I Ill.	1200	N ₂	39.0	44.1
19	CA Imp. I Ill.	1200	Steam	39.7	45.2
20	CA Imp. I Ill.	1800	Steam	87.3	>100
21	CA Imp. 3 Well.	1200	N ₂	30.5	31.7
22	CA Imp. 3 Well.	1200	Steam	26.3	25.7
23	CA Imp. 3 Well.	1800	Steam	86.1	>100
24	CA Mix	1800	Steam	80.7	92.9
25	CA Mix	1200	Steam	36.9	37.0
26	CA Mix	1200	N ₂	37.8	38.1

In the TGA tests, calcium also plays an important role through its influence on the gasification rate between 1500 and 1800 F.

Effect of Coal Type, Temperature, and Gasifying Medium

The coals selected for this study showed the expected variations in devolatilization and gasification reactivity. Figures 7 and 8 show how the ultimate MAF conversion level (X_f) with pure steam and nitrogen depend on temperature for each of the coals tested. These data allow the following conclusion to be drawn.

- The reactivity order for both gasification and devolatilization is Mississippi lignite > Illinois No. 6 > Wellmore.
- For the Mississippi lignite, steam gasification becomes extremely important above temperatures of 1200 F.
- For the Illinois No. 6 coal, steam gasification becomes the predominant means of conversion between 1500 and 1800 F.
- For the Wellmore coal, steam gasification becomes important in the neighborhood of 1800 F.
- Conversions due to devolatilization (N_2 atmosphere) were not greatly affected by temperatures above 1200 F.

Effect of Calcium Additives

One possible way to increase yield and control product distribution that may be especially important in mild gasification, is to utilize additives or catalysts. Therefore, tests were conducted using calcium incorporated into the coal either by physical mixing or by treatment of the coal with a calcium containing slurry. The tests with calcium were preliminary in the sense that only for the Wellmore coal was the physical mixture of coal and calcium compared with coal containing calcium incorporated by slurry treatment.

Only the Illinois No. 6 and the Wellmore coal were treated with calcium. In the case of the Illinois coal, the effect of calcium impregnation on the ultimate conversion was somewhat inconclusive in the sense that at

FIGURE 7. EFFECT OF GASIFYING MEDIUM ON ULTIMATE CARBON CONVERSION - ILLINOIS NO. 6 COAL

FIGURE 8. THE DEPENDENCE OF ULTIMATE CONVERSION ON COAL TYPE AND TEMPERATURE IN PURE STEAM AND NITROGEN ENVIRONMENT

1200 F calcium had no significant effect on either gasification or devolatilization conversion levels. On the other hand, at 1800 F complete conversion was achievable even without additives. Therefore, the strongest influence of calcium would exist over the temperature range between 1500 and 1800 F. The gasification rate at 1200 F however was much higher with the calcium impregnated coal roughly approximating the rate at 1800 F. With the least reactive Wellmore coal, the effect of calcium was pronounced. For example, with physically mixed calcium over 90 percent of the coal was converted compared to 47 percent without calcium. With the slurry incorporated calcium, complete conversion of Wellmore coal was achievable at 1800 F.

Significance of TGA Results

The TGA results can be simplistically interpreted in terms of a model that assumes that the ultimate conversion of coal [as defined by X_f in the devolatilization rate expression $r = k (X - X_f)$] is proportional to the number of bonds in the coal structure that are thermally ruptured which in turn is proportional to the gasifier temperature. A fraction of the broken bonds result in the evolution of volatile matter in the coal. However, reactive fragments formed by the breaking of bonds in the coal structure also recombine to form char. It is therefore speculated that the role of calcium in enhancing reactivity is that it poisons the recombination reactions that result in char formation.

From a design point of view, the TGA tests indicate that significant conversions can be achieved at milder conditions than employed in conventional gasification systems. The 2-zone process utilizes the char as fuel in high-temperature combustion whereas single stage gasification processes must be conducted at conditions where all of the char is consumed by gasification reactions. In addition, though the conversion rates in the TGA are rapid, the residence time required to reach the maximum conversion level is still on the order of minutes. Therefore a second stage following the initial dilute phase reaction zone can provide the residence time needed to achieve maximum total coal conversion levels.

Small Integrated Gasifier Results

A total of 21 tests have been run in the small integrated gasification facility. Three tests were conducted with lignite coal and the remainder with Illinois No. 6 seam coal. Of these Illinois coal tests, 11 were conducted during the first testing program. Table 2 summarizes the test data from these initial tests and Table 3 summarizes the results of the more recent verification experiments. Liquid product data are presented in Table 4 for the tests in which liquid samples could be quantified. During Tests 1 to 6, various methods of liquid collection were utilized to improve the liquid-collection efficiency. Tests 1, 2, and 3 were made using a simple water quench drum for collections, but during these tests significant amounts of condensed organics remained in the product gas as evidenced by blockage of the gas sampling lines.

A shell and tube product-gas cooler/condenser, followed by a xylene contacting tower, were installed prior to Test 4. This system, while demonstrating the ability to remove most of the liquids from the gas stream, still did not provide a means for collecting representative, analyzable samples of liquid materials. The liquid products were deposited as a film on the condenser walls.

Beginning with Test 7, a hybrid system was installed that incorporated the water drum quench, followed by the xylene scrubber as shown in Figure 5. Most of the liquid products, to date, have been collected in the water drum with a small fraction of the liquids collected from the xylene scrubber. In the most recent series of verification tests (beginning with test 22), the xylene scrubber was replaced with a simple water scrubber at the same location. This change simplified the analysis of the collected liquids, and proved to provide as complete a recovery of liquids as had been possible with xylene scrubbing.

TABLE 2. MSFB COAL GASIFICATION INTEGRATED UNIT TEST RESULTS

Test Number Test Date		1	2	3	4	5	6	7	8	9	10	11
		9/29/87	10/6/87	10/9/87	10/20/87	10/22/87	10/26/87	12/8/87	12/15/87	12/18/87	12/22/87	1/8
<u>Coal</u>												
- Type		III. #6	III. #6	III. #6	III. #6	III. #6	III. #6	III. #6	III. #6	Calcium Impregnated	III. #6	III. #6
- Feed Rate, lb/hr		12	20	20	20	13.4	15.2	15	12	22	12	17
<u>Gasifier</u>												
Temperature, °F		1320	1244	1360	1433	1480	1485	1460	1210	1220	1525	1641
- Fluidizing Gas		N ₂	N ₂	N ₂	N ₂	N ₂	Steam	Steam	Steam	Steam	Steam	Steam
- Superficial Velocity, FPS		7.4	13.0	23.4	25.4	19.5	-20	-20	-20	-20	-20	-20
<u>Product Gas Composition (Nitrogen Free, Mole Fraction)</u>												
- Hydrogen		0.059				0.298	0.305	0.348	0.138	0.280	0.380	0.4211
- Carbon Monoxide		0.223				0.228	0.228	0.167	0.170	0.138	0.195	0.203
- Carbon Dioxide		0.344				0.053	0.08	0.111	0.188	0.152	0.093	0.110
- Methane		0.345				0.289	0.29	0.300	0.409	0.352	0.262	0.217
- Ethane		0.012				0.025	0.027	0.018	0.047	0.039	0.012	0
- Ethylene		0.016				0.074	0.066	0.058	0.047	0.039	0.058	0.049
- Acetylene		0				0.023	0.05	0	0	0	0	0
- Heating Value Btu/SCF		487				659	665	599	672	623	565	500
- Carbon Conversion to Gas (%)		14.3				14.3	12.0	14.61	5.07	4.59	13.51	8.05
<u>Combustor</u>												
- Temperature, °F		1300	1365	1425	1500	1600	1520	1510	1390	1420	1630	1803
- Superficial Velocity, FPS		0.77	1.00	0.95	1.18	1.18	1.10	0.95	0.85	0.90	1.00	1.00

TABLE 3. MSFB COAL GASIFICATION INTEGRATED UNIT TEST RESULTS

Test No. Test Date	22 6/21/88	23 7/7/88	24 7/13/88	27 7/27/88	28 8/2/88	29 8/4/88	32A 8/31/88	32B 8/31/88	33 9/8/88	34 9/14/88	37 10/16/89	38 10/23/89
Coal Type	III. #6	III. #6	III. #6	III. #6	III. #6	III. #6	Lignite	Lignite	Lignite	Lignite	III. #6	III. #6
Feed Rate, lb/hr	11.94	12.24	10.08	24.6	14.3	20.27	15	15	12.47	13.13	12.4	16
Gasifier Temperature, F	1464	1282	1385	1851	1248	1842	1578	1581	1262	1289	1641	1698
Fluidizing Gas Velocity, fps	9.49	7.59	16.31	17.52	12.94	15.54	14.99	15.01	13.15	13.15	10.37	10.65
Product Gas Composition (Nitrogen Free, Mole Fraction)												
H ₂	34.84	30.78	39.52	41.39	17.39	44.42	42.19	43.28	25.83	33.87	47.29	48.61
C ₀	17.11	17.84	18.83	19.64	15.82	19.54	24.01	23.82	19.81	18.02	19.34	18.57
C ₀₂	10.09	8.78	9.04	8.57	18.34	10.26	22.13	21.58	35.32	32.51	8.85	15.01
H ₂ S	3.73	N/A	3.06	5.15	8.43	1	0	0	0	0	4.08	2.04
CH ₄	25.71	30.88	20.76	19.67	28.94	18.22	9.12	8.69	15.8	12.11	15.55	12.24
C ₂ H ₆	1.89	4.35	0.72	0.64	4.73	0.3	0.19	0.22	0.86	0.82	0.4	0.29
C ₂ H ₄	7.18	7.58	7.21	6.95	6.35	5.76	2.35	2.38	2.37	2.86	3.28	2.44
C ₂ H ₂	0.76	N/A	0.86	0	0	0.51	0.11	0.11	0	0	1.24	1.19
C ₃	N/A	3.26	N/A	0.1	3.27	N/A	N/A	N/A	N/A	0	N/A	N/A
C ₄	N/A	0.98	N/A	0	0.6	N/A	N/A	N/A	N/A	0	N/A	N/A
C ₅	N/A	0.16	N/A	0	0.07	N/A	N/A	N/A	N/A	0	N/A	N/A
Heating Value Value Incl. C ₃ to C ₅	577.31	688	540	519	586	497	349	349	361	348	451	484
Carbon Conv. to Gas (%)	639	789			729				29.6	35.7	23.88	31.04
Combustor Temperature, F	1580	1536	1680	1800	1390	var.	1736	1748	1386	1413	1751	1875

TABLE 4. YIELD OF PRIME PRODUCTS

Test No.	Temperature °F	% Total Carbon	Liquid Yield	Gas Yield	
			% Carbon Conversion	% Carbon Conversion	HHV Btu/lb*
1	1320		N/A	14.3	487
5	1480		N/A	14.3	659
6	1405		N/A	12.0	665
7	1460	24.80	9.87	14.61	599
8	1210	23.20	18.13	5.07	672
10	1525	22.76	9.25	13.51	565
11	1641	37.85	29.80	8.05	500
22	1463	39.31	20.43	18.88	577
23	1282		N/A	14.89	669
24	1585	46.28	20.09	26.19	553
27	1647	39.16	19.70	19.46	532
28	1236	26.21	16.6	9.61	586
37	1641	38.23	14.35	23.88	451
38	1698		N/A	31.04	404

* Add approximately 120 Btu/SCF to each entry for C₃ to C₅ components.

The test results are shown graphically in Figure 9. As can be seen, the liquid production rates with Illinois No. 6 seam coal are approximately constant regardless of gasifier temperature at about 18 percent of coal carbon. A slight increase is seen in the amount of liquid product generated as a result of the heat up rate of the coal particles as measured by the combustor temperature. The coal heat up rate also has an effect on the distribution of coal sulfur among the various coproducts as will be discussed below. These high production rates of liquids are significantly higher than those expected in a mild gasification process. Typical production rates from other mild gasifiers are on the order of 5 percent of the coal weight. Liquid production rates of 6.3 percent, for instance, were reported by United Coal Company⁽²⁾ in their mild gasification process.

These higher production rates in the MSFB process are caused by the rapid coal heat up rates achieved by direct mixing with incandescent sand particles rather than conventional coal heating methods. It has long been recognized that increasing the heat-up rate can significantly increase the amount of measured volatile content in a coal sample. This same type of phenomenon would likewise produce higher liquid yields from the integrated gasification system.

Carbon conversion to gas has been low for the tests run to date. These results are consistent with expectations for the temperatures at which the facility was operated. The heating value of the gas product has been consistent with expectations for a nitrogen free basis. The gas produced is very similar in composition to a coke oven gas-generated at these temperatures.⁽³⁾ Nitrogen is used for fluidizing gas, instrument purges and coal feed purging to simplify test operation and would not be used in a commercial facility. Tests using steam as the fluidizing gas showed gas yield and composition that was very similar to the N₂ tests.

The present facility and operating procedures limit the gasifier temperature to less than 1650 F. Attempts to increase the temperature are limited by system heat losses. The higher temperatures were achieved by oxygen enrichment of the combustion air to allow more fuel to be burned in the combustor at a maximum fluidizing velocity without elutriating bed material.

Operating experience in the small integrated gasification facility has been beneficial in establishing coal feeding and solids circulation techniques for the system. As expected, the small size of this facility limits the coal feed particle size and the mixing and residence time in the gasifier, but does provide a valuable tool in examining a range of process variables.

Gas Product

The relationship of carbon conversion to gas product yield as a function of gasifier temperature is shown in Figure 9. As expected, gas yields increase linearly with the gasifier temperature. As shown in Table 4, the heating value of the gas is consistently in the range of 550 to 650 Btu/SCF. In addition, some higher hydrocarbons (C_3 to C_5 alkanes and alkenes) are typically found in the gas raising the total effective heating value to about 700 Btu/SCF.

Liquid Product

Coal conversion to liquid products as a function of gasifier temperature is also found in Figure 9. The general expectation is that liquid yield decreases as the gasifier temperature increases. As discussed above, in the MSFB system, liquid yields remain relatively constant over the range of temperatures tested. This data is also shown as gallons of liquid/ton of coal fed in Figure 10.

Table 5 presents liquid product analyses for specific gravity, heating value and ultimate analysis. Figures 11 and 12 present boiling point ranges for some representative liquid samples. Complete analysis of every sample was not possible because the quantities produced were insufficient for all analyses.

The liquid fraction boiling below 492 F (200 C) is essentially constant at 70 to 80 percent of the material regardless of the gasifier temperature. As a result, it should be possible to upgrade these materials easily. The similarity between the sample collected from the water quench

CARBON CONVERSION TO LIQUIDS & GASES ILLINOIS #6 COAL

FIGURE 9. CARBON CONVERSION TO GAS VERSUS GASIFIER TEMPERATURE

Gallons Liquid per Ton

Versus Gasifier Temperature - Illinois #6

FIGURE 10. LIQUID PRODUCT YIELD - ILLINOIS NO. 6 COAL

TABLE 5. LIQUID PRODUCT ANALYSIS

Sample	Test 7		Test 8	
	12/9/87 Water Condenser Fraction	12/9/87 Xylene Scrubber Fraction	12/15/87 Water Condenser Fraction	12/15/87 Xylene Scrubber Fraction
Specific Gravity, @60 F	1.110	0.996	1.212	1.102
HHV, Btu/lb	16,180	16,989	11,679	15,961
Ultimate Analysis, Wt. %				
H	5.43	6.58	5.12	6.07
C	87.55	84.61	62.43	84.87
N	1.23	0.90	1.04	1.12
Cl	0.03	0.04	0.06	0.14
S	1.96	1.32	1.42	2.05
O (by difference)	3.81	6.53	15.56	5.68
Ash	0.02	0.06	14.43	0.21

CONDENSED PRODUCT BOILING POINTS

ILLINOIS # 6 COAL

TEST 8

TEST 7

FIGURE 11. COAL CONVERSION TO LIQUIDS VERSUS GASIFIER TEMPERATURE

CONDENSED PRODUCT BOILING POINTS

ILLINOIS #6 COAL

FIGURE 12. CONDENSED PRODUCT BOILING POINTS - ILLINOIS NO. 6 COAL

scrubber (W) and the xylene scrubber (X) is of interest. This may indicate that a rapid-quench water scrubber could be used commercially rather than a more complex scrubbing system.

Samples were also sent to Pacific Northwest Laboratory (PNL) for analysis by GC/MS. These samples identified, as expected, a wide variety of compounds in the materials. They also showed relatively low levels of oxygen and nitrogen which should simplify later upgrading of the materials. A copy of the Chromatographic data generated by PNL is found in Table 6.

Selected samples of the liquid products were analyzed by adsorption gas chromatography flame-ionization-detector and compared to a reference South Louisiana Crude Oil. This analysis determined the aliphatic and aromatic hydrocarbon content of the liquid products. Unfortunately, this technique could not analyze the heteroatoms so a qualitative analysis results. By comparison with the South Louisiana Crude Oil, the liquid products contained less aliphatic (straight chain hydrocarbon) materials and more aromatic material.

Fate of Sulfur Species

Evaluation of the data generated in the early tests on this program, showed that well over 90 percent of the coal sulfur (typically 95 to 96 percent) was present in the products from the gasifier (gas and liquid coproducts). By a detailed examination of the test data we found that the sulfur split (gasifier products vs combustor products) correlated with the liquid yield from the system. Liquid yield had previously been shown to correspond to heat up rate in the gasifier as discussed above. Higher heat up rates are achieved by combustor temperatures typical of projected full scale operating conditions (1900 F). We projected that the high heatup rates more effectively break apart the coal particles. Such breakage of the coal bonds makes both the organic and inorganic sulfur available to reaction, thus effectively removing sulfur from the char coproduct, and increases liquid product yields.

The final two tests during this phase of the program were designed to test this hypothesis. Combustor temperatures of over 1800 F were used and

TABLE 6. CHROMATOGRAPHIC DATA FOR MILD COAL GASIFICATION TARS

	A	B	D	E	F	G	I	J	K	L
1				MCG Test #37				MCG Test #22		
2	compound	mole	GC-MS	GC-FID	FID, Int	quantity	GC-MS	GC-FID	FID, Int	quantity
3		wi								
4	benzene ?	78		2.35	156	0.0006		2.29	48	0.0002
5	toluene ?	92		3.75	265	0.0010		3.71	227	0.0011
6	xylene	106	3.89	5.5	157	0.0006	3.94	5.47	355	0.0017
7	styrene	104	4.19	5.87	254	0.0009	4.22	5.85	279	0.0013
8	ethylbenzene	106		5.93	79	0.0003	4.24	5.9	178	0.0008
9	methylstyrene	118	5.78	7.75	151	0.0005	5.78	7.74	460	0.0022
10				7.8	198	0.0007		7.79	309	0.0015
11	indan		6.61	8.67	1536	0.0056	6.6	8.66	1760	0.0083
12	t-decalin	138	6.78	8.95	8112	0.0294	6.77	8.95	9806	0.0463
13	???	122				0		9.06	274	0.0013
14	paracresol	108				0	7.11	9.28	66	0.0003
15	methyl benzofuran		7.5	9.69	159	0.0006	7.52	9.67	449	0.0021
16	methylindene			10.47	65	0.0002	8.25	10.46	481	0.0023
17	methylindene			10.57	54	0.0002	8.36	10.56	509	0.0024
18	naphthalene	128	8.86	11.11	9221	0.0334	8.84	11.08	3261	0.0154
19	benzothiophene	134	8.96	11.21	147	0.0005	8.95	11.2	533	0.0025
20	dimethylbenzofuran		9.24	11.56	147	0.0005	9.31	11.52	253	0.0012
21	???	148		12.71	221	0.0008		12.7	209	0.0010
22	methyl naphthalene	142	10.48	12.82	2631	0.0095	10.58	12.82	2526	0.0119
23	methyl benzothiophene		10.58	12.94	247	0.0009	10.68	12.93	212	0.0010
24	methyl naphthalene	142	10.71	13.08	1666	0.0060	10.84	13.08	1973	0.0093
25	biphenyl	154	11.67	14.03	914	0.0033	11.89	14.02	476	0.0022
26	dimethyl naphthalene	156	12.02	14.42	546	0.0020	12.29	14.43	769	0.0036
27	cyclopentadien-1-ylbenzene	154				0		14.53	213	0.0010
28	dimethylnaphthalene	156	12.23	14.64	399	0.0014	in below	14.64	710	0.0034
29	" "	156	12.28	14.69	161	0.0006	12.54	14.69	397	0.0019
30	acenaphthene	154	12.37	14.77	516	0.0019	12.69	14.76	551	0.0026
31	dimethylnaphthalene	156		14.93	231	0.0008	12.84	14.92	587	0.0028
32	acenaphthylene	152	12.68	15.12	5247	0.0190	13.05	15.1	2499	0.0118
33	methyl biphenyl	166	13.11	15.49	245	0.0009	13.62	15.48	241	0.0011
34	cyanonaphthalene(?)		13.31	15.67	403	0.0015		15.61	130	0.0006
35	dibenzofuran	168	13.53	16	1329	0.0048	14.1	15.99	652	0.0031
36	naphthofuran(?)					0		16.29	281	0.0013
37	"					0		16.38	366	0.0017
38	phenalene		14.16	16.64	366	0.0013	15	16.64	685	0.0032
39	fluorene	166	14.42	16.89	2828	0.0102	15.22	16.88	1522	0.0072
40	???	166	14.66	17.15	421	0.0015	15.43	17.15	620	0.0029
41	dimethyl biphenyl	182	14.86	17.35	487	0.0018		17.35	624	0.0029
42	methylacenaphthylene	166	14.94	17.43	438	0.0016	16.16	17.43	658	0.0031
43	methyldibenzofuran	182	15.04	17.53	632	0.0023	16.38	17.52	637	0.0030
44	methyl fluorene	180	15.84	18.32	291	0.0011		18.32	479	0.0023
45	"	180				0		18.41	216	0.0010
46	"	180				0		18.55	471	0.0022

a temperature of 1600 to 1650 F was maintained in the gasifier. Coal feed rates were held between 15 and 20 lbs/hr in the 2 inch gasifier corresponding to a throughput of about 700 to 1000 lb/hr-ft².

During both of the tests we found a high percentage of the coal sulfur in the product gas (as H₂S) and only a small fraction of coal sulfur in the combustor flue gas (typically 1.5 to 5 percent of the coal sulfur). The precise split of sulfur species for the tests is shown below in Table 7.

TABLE 7. SULFUR SPECIES MEASURED IN MSFB GASIFICATION TESTS WITH ILLINOIS NO. 6 COAL

Test No.	Percentage of Coal Sulfur in Stream		
	Product Gas	Liquids*	Flue Gas
37	80.55	10.95	~5
38	82.49	10.95	1.5 to 6

* Based on typical liquid analysis

These results clearly verify both the hypothesis as to the mechanism of reactions as well as the previous data relative to the split of sulfur species possible in the MSFB Gasification process.

Testing of Additional Coal Types

A limited number of tests were made with a North Dakota lignite to demonstrate the ability of the MSFB process to handle a variety of coal feedstocks. This coal had an analysis as found in Table 8. The higher reactivity as compared to the Illinois coal led to interesting results from the gasification. The gaseous yield was found to be much higher than the Illinois coal and the liquid yield was found to be much lower. A gas yield of 69 percent was measured at the relatively modest temperature of 1580 F. Even at temperatures of 1260 F 30 percent of the carbon was converted to gas. This indicates that a high gas yield can be achieved as modest temperatures from

TABLE 8. INDIAN HEAD LIGNITE ANALYSIS

	As Received	Dry Basis
Proximate Analysis		
% Moisture	29.80	
Ash	6.41	9.13
Volatile	31.27	44.54
Fixed Carbon	32.52	46.33
Ultimate Analysis		
Carbon	45.90	65.39
Hydrogen	3.01	4.29
Nitrogen	0.63	0.90
Chlorine	--	--
Sulfur	0.51	0.72
Oxygen (Difference)	13.74	19.57
Btu/lb	7475	10,648

this type of coal thus broadening the range of applicability for the MSFB system. These results are shown in Figure 13.

Liquid production from the three lignite tests was below detection limits in the integrated gasifier as currently configured. As a result, to produce a liquid product lower temperatures (below 1200 F) will probably be required. However, the use of lignite in the MSFB system at the high gas yields measured provides the potential for an efficient IGCC process using lignitic coals as the fuel.

PRELIMINARY DESIGN AND ECONOMICS

A preliminary process conceptual design was generated using data from the integrated gasification tests. The design basis assumed a plant processing 1000 dry tons of Illinois No. 6 coal/day and a gasifier operating at 1200 F. Actual gas and liquid production levels measured in the integrated test unit were used to predict vessel sizes and product rates. A very compact gasifier is possible with the high specific coal throughputs possible in the MSFB system (1000 lb/hr-ft²). For the 1000 TPD plant, a gasifier diameter of only 7.3 feet is predicted. Major vessel sizes for the plant are shown in Table 9.

The plant will produce a slate of products as shown in Figure 14. In addition to the 12,500 lb/hr of liquids (15 percent of the coal by weight) produced at 1200 F in the gasifier, and the 179,500 SCFH (12.73 percent by weight) of scrubbed product gas, recoverable sensible energy in the form of hot flue gas and product gas are also available. In addition, if desired, on a site-specific basis about 26,000 lb/hr (260 MMBtu/hr) of

CARBON CONVERSION TO GAS VERSUS GASIFIER TEMPERATURE

LIGNITE GASIFICATION

FIGURE 13. CARBON CONVERSION TO GAS - LIGNITE COAL

TABLE 9. VESSEL DESIGN SIZES

Description	Size
Coal Feed System - Screw Feeder - Oversize Crusher	50 TPH
Inert Gas Supply	70 CFM
Gasifier - 1200 F - 2000 lb/hr-ft ² Tramp Material Removal at Base	7.3 ft diam x 100 ft
Gasifier Cyclone 57,500 ACFM Gas (Includes Condensibles) 1200 F	8.3 ft diam x 35.0 ft H Ref. Lined
Combustor 1950 F MSFB Type Water Walls	19.2 ft diam x 100 ft Ref. Lined Water Walls
Combustor Cyclone(s) 739,900 ACFM Gas	2 @20 ft diam x 80 ft L
Ash Cyclone(s)	2 @20 ft diam x 80 ft L
Gasifier Surge/Seal 5 min. Sand (63,630)	8.5 ft diam 15 ft high Ref. Lined
Prod. Gas Heat Recovery 9.42 MMBtu/hr to 750 F	Steam Gen. (W.H.B)
Combustor Surge/Seal 5 min. Sand	8.5 ft diam, 15 ft high Ref. Lined
Ash Collection/Cooling	22,000 PPH
Air Blower 5 psig Supply to Combustor	138,354 PPH
L-valve(s)	12 in. I.D. Refractory Lined

TABLE 9. (Continued)

Description	Size
Flue Gas Heat Recovery 370.8 MMBtu/hr to 250 F	Steam Gen. (W.H.B.)
Liquid Condense/Collection 12,336 SCFM Incl. - Water Pump - HX - Tank and Sprays	
Gas Cleanup 12,336 SCFM Incl. - Water Pump - HX - Tank and Sprays	
Waste Water Treatment	60 GPM
Start-up Burner	25 MMB/hr
Sand Makeup Hopper	80 ft ³
Dense Bed Makeup Hopper	50 ft ³
Conveying Blower	800 CFM
Steam Supply - L-Valve	1000 PPH
Refractory-Lined Pipe	300 ft
Gas Turbine/Compressor Generator	87.7 MMB/H
Steam Turbine/Generator	620,141 PPH

Stream numbers, , refer to preliminary heat and material balance.

FIGURE 14. PROCESS CONCEPTUAL DESIGN

char could be recovered from the process. For the purpose of the analysis, this char was used to generate steam in the combustor vessel.

A complete material balance is found in Table 10. The stream numbers correspond to those in Figure 14.

Installed equipment costs were estimated for the plant assuming the gaseous product as well as the available sensible energy would be used for power production. These figures are found in Table 11. The total projected capital cost for the 1000 TPD plant is \$25.4 MM. This includes turbine costs of \$16.4 MM. If strictly liquids, char and steam are the desired products, the costs are greatly reduced to about \$9 MM for the 1000 TPD plant.

If calculated on a dollars per installed kilowatt basis, the MSFB costs correspond to \$457.2/kW including the cost of turbines. This compares favorably with a Texaco plant costing \$957.1/kW.⁽⁸⁾ Since the Texaco process is a high pressure-oxygen blown process, the costs would be expected to be much higher. If an entrained bed, low pressure, air blown process were available, the costs would be projected to be \$573.8/kW -- still somewhat higher than the MSFB system.

A projected annual operating budget for the plant includes credits for the various products produced, and is found in Table 12. The overall process simplicity of the MSFB system, as has been demonstrated with commercial combustion systems, makes possible a small operating staff, reducing significantly a major cost element. Another significant reduction is possible since oxygen is not required in the MSFB system.

As a result, total operating costs are very low compared with conventional gasification plants using oxygen. Other savings can be realized by the elimination of a sulfur recovery plant. The circulating solids in the MSFB system can be a sulfur sorbent such as limestone which eliminates both the capital and operating costs of this complex gas scrubber equipment.

As is shown in Table 12, if a value of \$3.50/MM Btu is assigned to the gas and liquid coproduct streams for the case presented, and a value of \$4.50 for the steam product, overall annual operating credits of about \$27 MM is calculated. This magnitude of credits provides a significant \$10MM annual revenue to users of the technology. This analysis assumes gas, liquid and steam coproducts would be sold separately. If power were the main product, credits would amount to about \$48 MM annually, or an annual revenue of \$31MM.

[illegible]

TABLE 10. (Continued)

Stream No. Description Component	9 Cleaned Flue Gas		10 Ash		11 Solids to Gasifier		12 Air Purge	
	moles/hr	lb/hr	moles/hr	lb/hr	moles/hr	lb/hr	moles/hr	lb/hr
H ₂		0.00						0.00
CO		0.00						0.00
CO ₂		162355.60						0.00
CH ₄	3689.90							0.00
C ₂ H ₄		0.00						0.00
C ₂ H ₆		0.00						0.00
C ₂ H ₂		0.00						0.00
O ₂	457.19						0.37	11.92
N ₂	18000.44						1.40	39.22
H ₂ S		14630.08						0.00
H ₂ O	1763.97							0.00
Total	23911.50	31751.48	0.00	21250.00	0.00	0.00	1.77	51.14
SCF/hr	9082456.98	712749.35					672.00	
Temperature, F	1930.00						70.00	
Pressure, PSIG	2.88						5.00	
Coal								
Char								
Sand								
H ₂ O								
Ash								
Liquid Product				13416.67				
							763581.00	

TABLE 10. (Continued)

Stream No. Description Component	13 L Valve Control		14 Inert Gas Purge		15 Scrubber Outlet Gas		16 Dry Product Gas	
	moles/hr	lb/hr	moles/hr	lb/hr	moles/hr	lb/hr	moles/hr	lb/hr
H ₂	0.00	0.00	0.00	0.00	47.58	95.16	47.58	95.16
C ₀	0.00	0.00	0.00	0.00	58.60	1640.80	58.60	1640.80
C ₀₂	0.00	0.00	0.00	0.00	64.80	2851.20	64.80	2851.20
CH ₄	0.00	0.00	0.00	0.00	140.98	2255.68	140.98	2255.68
C ₂ H ₄	0.00	0.00	0.00	0.00	16.20	453.60	16.20	453.60
C ₂ H ₆	0.00	0.00	0.00	0.00	16.20	486.00	16.20	486.00
C ₂ H ₂	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
O ₂	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
N ₂	0.00	0.00	11.06	309.68	49.80	1394.40	49.80	1394.40
H ₂ S	0.00	0.00	0.00	0.00	115.22	3917.60	115.22	3917.60
H ₂ O	0.00	0.00	0.00	0.00	79.41	1429.38	0.00	0.00
Total	20.00	366.00	11.06	309.68	588.79	14523.82	588.79	13094.44
SCF/hr	20.00	366.00	4191.74	223152.70	223152.70	193056.31	193056.31	193056.31
Temperature, F	7500.00	7500.00	70.00	140.00	140.00	140.00	140.00	140.00
Pressure, PSIG	400.00	400.00	5.00	3.19	3.19	3.19	3.19	3.19
Coal	150.00	150.00						
Char								
Sand								
H ₂ O								
Ash								
Liquid Product								

TABLE 11. CAPITAL DESIGN COSTS

Description	Installed Cost \$ x 1000 4th Quarter 1987
Coal Feed System - Screw Feeder - Oversize Crusher	857
Inert Gas Supply	60
Gasifier - 1200 F 200 lb/hr-ft ² Tramp Material Removal at Base	288
Gasifier Cyclone 57,500 ACFM Gas (Includes Condensibles 1200 F)	50
Combustor 1950 F MSFB Type Water Walls	350
Combustor Cyclone(s) 739,900 ACFM Gas	50
Ash Cyclone(s)	87 each
Gasifier Surge/Seal 5 min. Sand (63,630 lb)	87 each
Product Gas Heat Recovery 9.42 MMBtu/hr to 750 F	38
Combustor Surge/Seal 5 min. Sand	240
Ash Collection/Cooling	38
Air Blower 5 psig Supply to Combustor	358
L-Valve(s)	121.5
Flue Gas Heat Recovery 370.8 MMBtu/hr to 250 F	71.8
	1,500

TABLE 11. (Continued)

Description	Installed Cost \$ x 1000
Liquid Condense/Collection 12,336 SCFM	293
Gas Cleanup	293
Waste Water Treatment	90
Start-up Burner	100
Sand Makeup Hopper	20
Dense Bed Makeup Hopper	15
Conveying Blower	10
Steam Supply - L-Valve	10
Refractory-Lined Pipe	102
Subtotal	5,303
Controls and Instrument	530
Electrical	530
Process Piping	530
Structure and Foundations	1,061
Total	7,954
Gas Turbine/Compressor Generator \$400/kW	Turb. 27,952
	Comp. 1,588
Steam Turbine/Generator \$290/kW	11,399
Total Gasif.	7,954
Design	1,034
Total	8,988
Including Turbines	49,927

TABLE 12. ANNUAL OPERATING COSTS - MSFB GASIFICATION

	\$K 100% On Stream
Coal - \$10/ton	14,600
Boiler Feed Water - \$1.50/1000 gal - 378,720 PPM	597
Power - 500 kW - 5¢/kW	219
Op. Labor - 2 men/shift - \$20/hr	467
M. Labor - 1 man/shift - \$20/hr	234
Op. Supplies (0.3 * Op. Labor)	140
M. Supplies (2½% Capital)	225
Taxes and Insurance (2½% Capital)	<u>225</u>
Total	16,707
Credits	
Gas and Liquids - \$3.50/MM Btu	10,756
Steam - \$4.50/MM Btu	<u>16,270</u>
Power Production - 109.2 MW	47,830
Power Revenue @5¢/kWh	

This preliminary evaluation shows that the MSFB gasification process should compete economically with current energy sources. This analysis will, of course, have to be updated as further data are generated in experimental operations.

CONCLUSIONS AND RECOMMENDATIONS

Accomplishments

Bench scale tests have demonstrated the technical feasibility of the MSFB system for coal mild gasification. Progressive tests in a TGA and a small integrated gasification facility provide initial performance data at selected test conditions. Continuous system operation is stable at high throughput (~ 1000 lb/hr-ft²) with controlled solids circulation. High quality gaseous and liquid products were produced simultaneously. Integrated utilization of the char directly recovers energy from this gasification coproduct without requiring char cooling and handling equipment. By handling ash alone, much smaller amounts of hot material will be discharged from the system.

Twenty-six TGA tests screened system operating conditions and performance for 3 coals, three gasifier temperatures, and steam and nitrogen fluidizing gas. TGA results are consistent and show good agreement with other published results. The TGA data provided the basis for the small integrated-gasifier test plan.

Twenty one tests were completed in the small integrated gasification facility which has a 2-inch circulating bed gasifier and a 12-inch fluidized bed combustor. Coal feeding, solids circulation and liquid product recovery were successfully demonstrated in this unit for coal feed rates between 10 to 20 lb/hr.

Liquid production rates were higher than expected. Liquid product yields of about 20 percent of the coal fed were achieved throughout the test program.

A preliminary process design and economic evaluation was generated to provide an initial assessment of the benefits of developing the MSFB

Gasification process. Estimated capital cost for a 1000 TPD facility is \$9 million. This reflects the high throughput, small reactor size and absence of an oxygen plant required for many conventional gasification systems.

Flexibility is a key feature in specifying the desired product distribution. Yield of the various products can be controlled by process operation to fit a specific application. Liquid products have excellent potential as chemical feedstocks and for upgrading to transportation fuels. Gaseous products are suitable for use as medium-Btu quality fuel (~600 Btu/SCF) or as feedstock to a chemical/petrochemical process. The circulating solids in the system can be a sulfur sorbent and/or a catalyst to increase or alter the yield and quality of prime products.

Conclusions

- The project data clearly establish the technical feasibility of the MSFB system for coal gasification and mild gasification. These data provide an initial indication of the system performance over a limited range of operating conditions. Further testing and optimization are required to fully quantify operating conditions and performance.
- Incomplete coal devolatilization in the integrated gasifier, compared to the TGA data, indicates potential for higher yields of liquid and gaseous products. Improved performance is expected from full scale gasifiers which can use larger coal particle sizes and dense bed material to increase the solids residence time in the MSFB gasifier. This expectation means that the reported results should be conservative.
- Coal feed rates of 12 to 22 lb/hr to the 2-inch gasifier are equivalent to throughputs over 1000 lb/hr-ft².
- Coal conversion to liquids was essentially constant at 20 percent for gasifier temperatures ranging from 1200 to 1650 F, respectively.
- Gas composition changes, primarily hydrogen and methane, are a function of gasifier temperature. Methane concentration is approximately 35 percent (volume) at 1200 F gasifier temperature and this is consistent with Babcock and Wilcox's data⁽³⁾ for typical coke oven gases. The gas yield generally increases with gasifier temperature except as noted during Test No. 11. This curve must be verified through generation of a broader range of data.

- MSFB Gasification has flexibility to fit several applications within the coal, chemical, petroleum, and utility industries. Each specific application has unique product requirements and operating conditions. The MSFB gasification system can meet these needs through the flexibility of producing a range of products along with the ability to internally modify these products as required to fit user requirements. These advantages are realized in compact equipment without an oxygen plant that will therefore be more cost effective than conventional processing schemes. Thus, this system can:
 - Produce liquid products with valuable coproduct streams
 - Produce an undiluted fuel gas without the need for oxygen
 - Achieve high coal throughputs resulting in compact reactors
 - Circulate catalytically active or chemically active materials to modify product stream properties
 - Require low capital investment
 - Process a wide range of coal ranks without regard to agglomeration tendency
 - Use char coproduct efficiently within the process, if desired.

Recommendation

The high liquid-product yields in the initial test series are significant findings for mild gasification tests. A better understanding of the operating conditions and mechanisms leading to these yields is a key to successfully capitalizing on the initial findings. Continued development of the MSFB Gasification system is recommended to establish a broader data base and realize the potential benefits from the system. In the initial program plan developed with DOE, the experimental testing would advance to the existing Process Research Unit (~100 lb/hr coal) at the conclusion of this program phase. Pilot plant data are necessary to provide a sound engineering basis for scale-up and technology demonstration.

Program Continuation Activities

Pilot plant testing, at approximately 100 lb/hr coal feed rate, to generate engineering data is necessary for scale-up and technology demonstration. Comprehensive pilot plant data increase the confidence in

preliminary design and economic evaluations to approach technology commercialization. Battelle's approach to commercial development includes an Architect/Engineer (A&E), equipment manufacturer(s) and a user. An integrated coal and chemical/petrochemical company represents an ideal user for this technology as they have the greatest incentive to generate feedstocks from coal as an alternative to crude oil or natural gas. User participation, during the PRU tests, greatly facilitates their technical background and transition to scale-up for a commercial demonstration. DOE's Clean Coal Technology Program provides a viable technique for technology transfer of an advanced system such as MSFB Gasification to industry.

An alternative approach to scale-up and commercial demonstration exists through repowering an electric utility unit. This application is of interest to both Centerior Energy Corporation and Thermo Electron Corporation and is being evaluated as part of their long-term planning.

Therefore, the PRU testing is necessary to implement MSFB gasification technology on a commercial scale. The results of the small integrated-gasifier tests will establish the specific operating conditions for PRU testing. The PRU will operate with larger coal particle size and increased residence time which is expected to improve prime product yield compared to the small integrated gasifier. We recommend that this PRU study be conducted after additional smaller scale tests have identified optimum processing conditions.

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APPENDIX A
COAL ANALYSIS

COMMERCIAL TESTING & ENGINEERING CO.

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OFFICE TEL. (312) 264-1173
TELEX: 283527

BATTELLE MEMORIAL INSTITUTE
Columbus Laboratories
505 King Avenue
Columbus, OH 43201

July 28, 1988

Sample Identification
by Battelle Memorial Inst.

ATTN: Ted L. Tewksbury, Rm. 13-3-022

Kind of sample
reported to us Coal

Illinois #6
7/15/88

Sample taken at Columbus Laboratories

Sample taken by Battelle Memorial Inst.

Date sampled 7/15/88

Shipper No. Z 4712
P.O. No. R 4475

Date received 7/20/88

Analysis report no. 71-59836

PROXIMATE ANALYSIS

	<u>As Received</u>	<u>Dry Basis</u>
% Moisture	11.50	xxxxx
% Ash	9.58	10.82
% Volatile	35.71	40.35
% Fixed Carbon	43.21	48.83
	<u>100.00</u>	<u>100.00</u>
Btu/lb.	10939	12361
% Sulfur	3.40	3.84

ULTIMATE ANALYSIS

	<u>As Received</u>	<u>Dry Basis</u>
% Moisture	11.50	xxxxx
% Carbon	60.44	68.29
% Hydrogen	4.43	5.01
% Nitrogen	1.19	1.35
% Chlorine	0.21	0.24
% Sulfur	3.40	3.84
% Ash	9.58	10.82
% Oxygen (diff)	9.25	10.45
	<u>100.00</u>	<u>100.00</u>

SULFUR FORMS

	<u>As Received</u>	<u>Dry Basis</u>
% Pyritic Sulfur	----	----
% Sulfate Sulfur	----	----
% Organic Sulfur	----	----
(Diff)	----	----
% Total Sulfur	----	----

FUSION TEMPERATURE OF ASH

	<u>Reducing</u>	<u>Oxidizing</u>
Initial Deformation	---- °F	---- °F
Softening (H = W)	---- °F	---- °F
Softening (H = ½ W)	---- °F	---- °F
Fluid	---- °F	---- °F

HARDGROVE GRINDABILITY INDEX = ---- at ---- % Moisture

% EQUILIBRIUM MOISTURE = ----

FREE SWELLING INDEX = ----

Respectfully submitted,
COMMERCIAL TESTING & ENGINEERING CO.

David W. Cox, Manager, South Holland Laboratory

Charter Member

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For Your Protection

DWC/ds

OVER 40 BRANCH LABORATORIES STRATEGICALLY LOCATED IN PRINCIPAL COAL MINING AREAS,
TIDEWATER AND GREAT LAKES PORTS, AND RIVER LOADING FACILITIES

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MANAGER
LOWEST DIVISION

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TELEX 283527

BATTELLE MEMORIAL INSTITUTE
Columbus Laboratories
505 King Avenue
Columbus, OH 43201

October 30, 1987

Sample identification
by Battelle Memorial Inst.

ATTN: Ted L. Tewksbury

Kind of sample
reported to us Coal

Ill #6
Rec'd 9/23/87
20 x 100

Sample taken at Columbus Laboratories

Sample taken by Battelle Memorial Inst.

Date sampled -----

Shipper No. Y-4777
P.O. No. P-5644

Date received 10/22/87

Analysis report no. 71-46207

PROXIMATE ANALYSIS

	As Received	Dry Basis
% Moisture	12.57	xxxxx
% Ash	8.77	10.03
% Volatile	36.18	41.38
% Fixed Carbon	42.48	48.59
	100.00	100.00
Btu. lb.	11173	12779
% Sulfur	3.51	4.01

ULTIMATE ANALYSIS

	As Received	Dry Basis
% Moisture	12.57	xxxxx
% Carbon	61.57	70.42
% Hydrogen	4.43	5.07
% Nitrogen	1.16	1.33
% Chlorine	-----	-----
% Sulfur	3.51	4.01
% Ash	8.77	10.03
% Oxygen (diff)	7.99	9.14
	100.00	100.00

SULFUR FORMS

	As Received	Dry Basis
% Pyritic Sulfur	1.32	1.51
% Sulfate Sulfur	0.01	0.01
% Organic Sulfur (Diff)	2.18	2.49
% Total Sulfur	-----	-----

FUSION TEMPERATURE OF ASH

	Reducing	Oxidizing
Initial Deformation	-----	-----
Softening (H=W)	-----	-----
Softening (H=1/2W)	-----	-----
Fluid	-----	-----

HARDGROVE GRINDABILITY INDEX = ----- at -----

% EQUILIBRIUM MOISTURE = -----

FREE SWELLING INDEX = -----

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COMMERCIAL TESTING & ENGINEERING CO.

David W. Cox
David W. Cox, Manager, South Holland Laboratory

Member

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BATTELLE MEMORIAL INSTITUTE
Columbus Laboratories
505 King Avenue
Columbus, OH 43201
ATTN: Frank W. Shirley

September 8, 1987

Sample identification
by Battelle Memorial Inst.

Kind of sample
reported to us Coal

Wellmore Coal
Head Sample

Sample taken at Columbus Laboratories

Sample taken by Battelle Memorial Inst.

Date sampled -----

Shipper No. Y-2926
P.O. No. P-0698

Date received 8/31/87

Analysis report no. 71-43422

PROXIMATE ANALYSIS

	As Received	Dry Basis
% Moisture	1.46	xxxxx
% Ash	7.90	8.02
% Volatile	31.12	31.58
% Fixed Carbon	59.52	60.40
	100.00	100.00
Btu/lb.	13920	14126
% Sulfur	1.25	1.27

ULTIMATE ANALYSIS

	As Received	Dry Basis
% Moisture	1.46	xxxxx
% Carbon	77.94	79.09
% Hydrogen	4.81	4.88
% Nitrogen	1.47	1.49
% Chlorine	-----	-----
% Sulfur	1.25	1.27
% Ash	7.90	8.02
% Oxygen (diff)	5.17	5.25
	100.00	100.00

SULFUR FORMS

	As Received	Dry Basis
% Pyritic Sulfur	-----	-----
% Sulfate Sulfur	-----	-----
% Organic Sulfur (Diff)	-----	-----
% Total Sulfur	-----	-----

FUSION TEMPERATURE OF ASH

	Reducing	Oxidizing
Initial Deformation	----- °F	----- °F
Softening (H = W)	----- °F	----- °F
Softening (H = 1/2 W)	----- °F	----- °F
Fluid	----- °F	----- °F

Ignited Basis

ROBROVE GRINDABILITY INDEX = ----- at ----- % Moisture %Sodium Oxide, Na₂O 0.90
%Potassium Oxide, K₂O 2.23

EQUILIBRIUM MOISTURE = -----

SWELLING INDEX = -----

Respectfully submitted,
COMMERCIAL TESTING & ENGINEERING CO.

David W. Cox

David W. Cox, Manager, South Holland Laboratory

Charter Member

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For Your Protection

OVER 40 BRANCH LABORATORIES STRATEGICALLY LOCATED IN PRINCIPAL COAL MINING AREAS,
TIDEWATER AND GREAT LAKES PORTS, AND RIVER LOADING FACILITIES

JWC/ds

COMMERCIAL TESTING & ENGINEERING CO.

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LLOYD W. TAYLOR III
MANAGER
MIDWEST DIVISION PLEASE ADDRESS ALL CORRESPONDENCE TO:
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SOUTH HOLLAND, IL 60473
OFFICE TEL. (312) 264-1173
TELEX: 283527

BATTELLE MEMORIAL INSTITUTE
Columbus Laboratories
505 King Avenue
Columbus, OH 43201

September 20, 1988

Sample Identification
by Battelle Memorial Inst.

ATTN: Ted L. Tewksbury, Rm., 13-3-022

Sample No. 2-6532
G-1159
Indian Head
8 x 20

Shipper No. Z-6532
P.O. No. R 8627

Kind of sample reported to us Coal
Sample taken at Columbus Laboratories
Sample taken by Battelle Memorial Inst.
Date sampled -----
Date received 9/8/88

Analysis report no. 71-62344

PROXIMATE ANALYSIS			ULTIMATE ANALYSIS		
	As Received	Dry Basis		As Received	Dry Basis
% Moisture	29.80	xxxxx	% Moisture	29.80	xxxxx
% Ash	6.41	9.13	% Carbon	45.90	65.39
% Volatile	31.27	44.54	% Hydrogen	3.01	4.29
% Fixed Carbon	32.52	46.33	% Nitrogen	0.63	0.90
	100.00	100.00	% Chlorine	-----	-----
			% Sulfur	0.51	0.72
Btu/lb.	7475	10648	% Ash	6.41	9.13
% Sulfur	0.51	0.72	% Oxygen (diff)	13.74	19.57
				100.00	100.00
SULFUR FORMS			FUSION TEMPERATURE OF ASH		
	As Received	Dry Basis		Reducing	Oxidizing
% Pyritic Sulfur	-----	-----	Initial Deformation	----- °F	----- °F
% Sulfate Sulfur	-----	-----	Softening (H=W)	----- °F	----- °F
% Organic Sulfur	-----	-----	Softening (H=½W)	----- °F	----- °F
(Diff)	-----	-----	Fluid	----- °F	----- °F
% Total Sulfur	-----	-----			

HARDGROVE GRINDABILITY INDEX = ----- at ----- % Moisture

% EQUILIBRIUM MOISTURE = -----

FREE SWELLING INDEX = -----

Respectfully submitted,
COMMERCIAL TESTING & ENGINEERING CO.

David W. Cox, Manager, South Holland Laboratory

Charter Member

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DWC/ds

OVER 40 BRANCH LABORATORIES STRATEGICALLY LOCATED IN PRINCIPAL COAL MINING AREAS,
TIDEWATER AND GREAT LAKES PORTS, AND RIVER LOADING FACILITIES

END

DATE FILMED

02 / 20 / 91