MARSHALL OWEN

Introduction

The Marshall Owen concept is an example of a coal refinery in the Devolatilization category. The Marshall Owen carbonization (the heating of bituminous coal or mixtures of different ranks of bituminous coals in the absence of air to form char) process is licensed from the Coalite Group PLC (Derbyshire, U.K.) through the Rexco company [1]. The Coalite process is a mild gasification process that has been in commercial operation in the U.K. for over 50 years [2]. The products of the Coalite process are a smokeless solid fuel for domestic heating, and coal-derived liquids for chemical feedstock, and industrial and transportation fuels. Char is the main product, although tar, light oil, ammonia liquor, and a high-Btu fuel gas are produced. The coal-derived liquids from the Coalite process have been further refined into commodity chemicals such as ammonium sulfate, benzene, toluene, xylene, naphthalene, pyridine, phenanthrene, anthracene, creosote, road tars, roofing pitches, and pipeline enamels [3].

This coal refinery concept is being pursued by the Marshall Owen Enterprises of Reno, Nevada and is shown in Figure 1. The necessary inputs to this coal refinery would include run-of-mine coal, air, and water, while major products include a smokeless solid fuel (i.e., Coalite) and coal-derived liquids which can be further upgraded to value-added products.

Detailed Process Description

This process is based on an updated version of the original Coalite process [3], which employs low temperature carbonization of coal in a vertical retort. The original Coalite process operates in a batch mode, unlike the Marshall Owen process which would operate on a continuous basis. An advantage of continuous operation is that product streams of uniform quality can be generated, unlike batch operation.

This mild gasification process employs continuous retorts which essentially are brick-lined cylinders, approximately 40 feet in height, with a proprietary configuration for heating the coal and extracting the volatiles that are generated. Coal is fed into the continuous retorts through a slide valve system. The coal passes through the retort and is converted into char in the carbonizing zone. The hot product gas (1,200 to 1,300° F) generated during carbonization contains tar oil products and light hydrocarbons, and exits the retort. The retort is heated by radiant energy, supplied by hot gas circulating around the outside of the vertical tubes. This hot gas is generated by the combustion of the process fuel gas with the flue gas used to preheat the incoming combustion air stream.

The hot product gas is sent to the Vapor/Liquid Separation section, where the product gas is cooled to remove tars and liquors. The remaining gas contains light hydrocarbons and a relatively high heat content (approximately 700 Btu per cubic foot [4]). A portion of this fuel gas stream is sent to the vertical retort section to supply the indirect heating for the coal carbonization reaction. The remainder can be utilized for on-site heating or for steam production leading to electric generation.

The hot char is removed from the retort at a rate controlled by an adjustable automatic extraction system. The cold fuel gas stream is sent to the retort to cool the char, after which the char is quenched.

Types of Feed Coal

Carbonization using vertical retorts requires careful selection of the coal. A coal that is too highly swelling could cause difficulty during discharge of the resulting char, while a coal that is too weakly caking would produce a smokeless fuel which is too friable and soft to withstand breakage during handling [3]. The Coalite process uses medium coking British coal whose properties are similar to those of U.S. Eastern bituminous coals [5].

Marshall Owen Enterprises has proposed a demonstration of the process which would use a high-volatile low-sulfur (0.6 to 0.7 percent) bituminous non-caking coal supplied by Bear Coal Co. of Somerset, CO [1].

Products

The main products from this coal refinery are smokeless solid fuel, crude tar, crude light oil, and high-Btu fuel gas [5]. An approximate product distribution is given in Table 1 [1, 2, 4].

The smokeless fuel produced by this process would have an open porous structure and a volatile matter content in the range of 9 to 12 weight percent (typical physical properties of the Coalite smokeless fuel are given in Table 2 [6]). The volatile matter in the smokeless fuel would be rich in hydrogen, which produces an almost non-luminous flame when burned.

The characteristics of the whole Coalite liquid and the middle distillate (boiling point between 350 to 650°F) fraction separated from it are given in Table 3 [5]. The distribution of chemical feedstocks that can be produced from the Coalite liquid is shown in Table 4 [7]. The amount of condensed carbon (coal fines) in the Coalite liquid is quite high (about 3.5 percent) requiring distillation to separate out the unconverted coal and to give the proper boiling range. The sulfur content is quite high (about 1 percent) with the majority of the Coalite liquid relatively high-boiling so that cracking to decrease the molecular weight followed by upgrading (e.g. hydrotreating for sulfur removal) to meet product quality specifications will be required.

It has been demonstrated that the the Coalite liquid can be processed into high energy density fuels [5].

Likely Applications

The economics of the carbonizing process is such that the plant should be located in the immediate vicinity of the coalfield which can provide suitable coal for the process, to minimize coal transportation costs [3]. The demonstration plant proposed by Marshall Owen Enterprises is to be located within 1 mile of the mine supplying the coal [1]. Since the commercial plant is to be located as close to the mine pithead, mine owners and operators would be one commercial entity interested in this coal refinery concept. Mine owners could use it to add value to their coal and to expand their markets.

The chemical and petroleum industries would have the capability for upgrading the raw coal-derived liquid into value-added products. In addition, the electric utility industry may be interested in this concept to assure a solid fuel with uniform properties.

Status of Development

The Coalite process has been in commercial operation for over 50 years. Application of this technology to U.S. coals may only require pilot tests to establish the operating conditions to assure sufficient reliability and product quality.

The process economics, including the required capital investment and the projected selling price of the smokeless fuel, are given in reference [1].

Environmental Aspects

The fuel gas produced during carbonization is composed of H_2, CO, CH_4, and other hydrocarbons, tars, and oils, with some H_2S, ammonia, and hydrogen cyanide. The potential environmental intrusions from this coal refinery include emissions of volatile organic compounds into the atmosphere, SO_2, and ammonia. Typically, the hydrogen sulfide (H_2S), ammonia, and hydrogen cyanide are removed by absorption with ammonia and water. The absorbent is then steam stripped to produce a concentrated stream of hydrogen sulfide, ammonia, and hydrogen cyanide which is passed over a catalyst to convert the ammonia and hydrogen cyanide to nitrogen and hydrogen. A commercial system (such as the Claus® process) then destroys the hydrogen sulfide and produces elemental sulfur. It would be expected that greater than 80 percent of the sulfur from the process will be removed, leading to an expected SO_2 emission rate of 0.4 percent [1].

Research Needs

The Marshall Owen concept is fairly wasteful of energy in that the significant energy that exists in the flue gas streams is not taken advantage of (e.g., for steam production, etc.). Modifications to the process design given in Figure 1 may potentially result in a higher thermal efficiency for the process.

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Table 1: Product Output for Marshall Owen Concept Coal Refinery

PRODUCT	OUTPUT (lb per lb coal)
Char (Coalite fuel)	0.68 to 0.75
Liquid Hydrocarbons	0.16 to 0.2
Product Fuel Gas (700 Btu per scf)	0.1 to 0.12

Table 2: Characteristics of Typical Coalite Smokeless Fuel

PROXIMATE ANALYSIS (weight percentage)			
Ash	Volatile Matter	Fixed Carbon	Total Moisture
12	9 to 12	61 to 64	15

Table 3: Characteristics of the Whole (Dewatered) Coalite Liquid and its Middle Distillate

LIQUID:	Whole Dewatered Coalite	Middle Distillate
ULTIMATE ANALYSIS (weight percentage; dry basis)		
Carbon	83.56	84.00
Hydrogen	8.55	9.26
Nitrogen	1.10	0.81
Sulfur	0.85	0.73
Oxygen	5.94	5.20
PHYSICAL PROPERTIES		
H/C Atomic Ratio	1.23	1.32
Con Carbon Residue, %	3.52	< 0.1

Table 4: Distillable Oil Composition of Whole Coalite Liquid

PRODUCT OUTPUT (weight percentage)				
Naphtha (< 350°F)	Mid Distillate (350 - 650°F)	Heavy Ends (> 650°F)	Coal Fines	Water
5	45	43	2	5

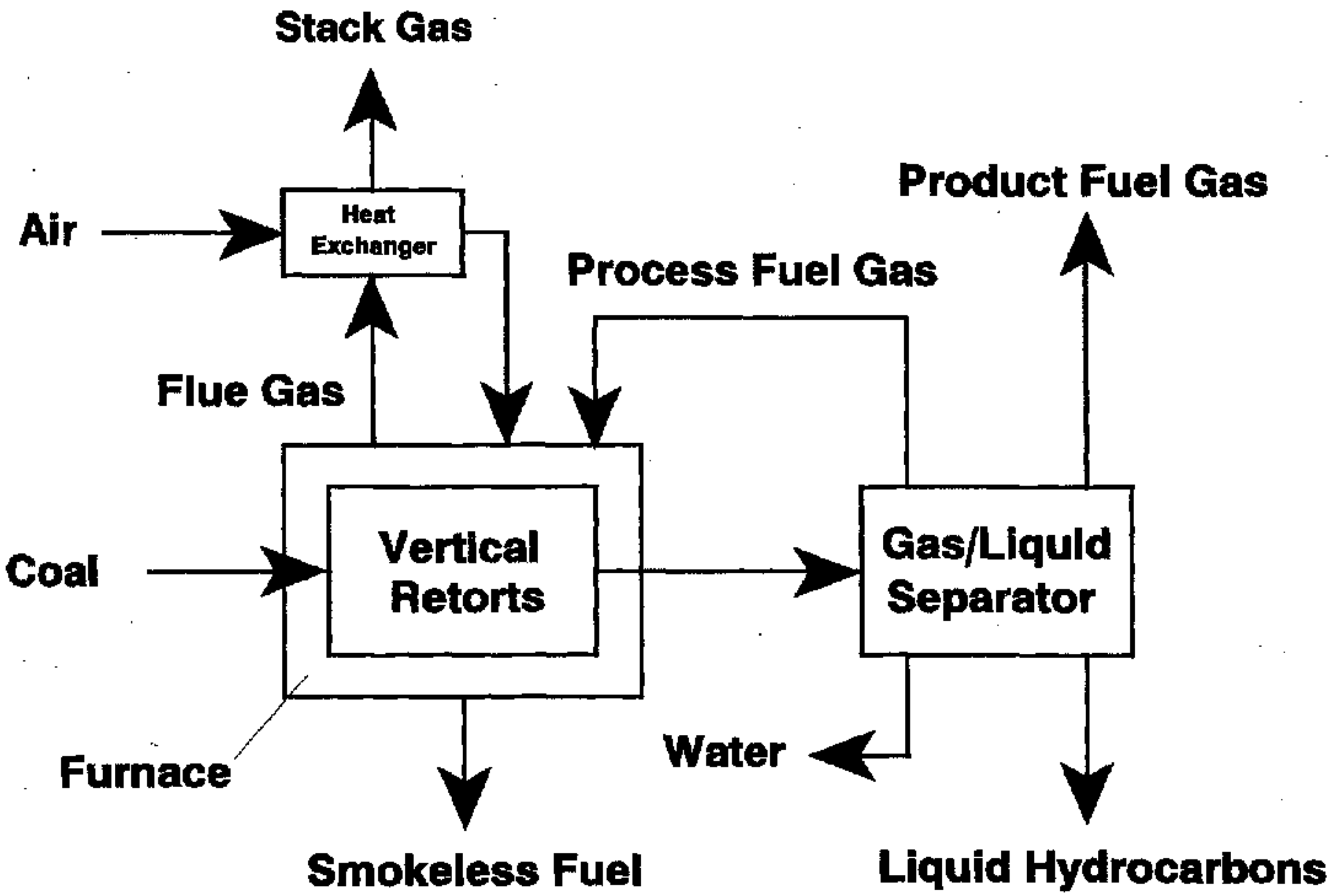

FIGURE 1: Overall Block Diagram of the Marshall Owen Concept

SFUEL

Introduction

The SFuel concept would integrate the pyrolysis of coal with production of a smokeless solid fuel and advanced electric power generation. The smokeless fuel (called SFuel) is intended for domestic and electric utility consumption, being more thermally efficient than direct combustion of coal while also minimizing environmental intrusions.

This concept has been advanced by the Institute of Chemical Processing of Coal (Poland) [1]. Key differences between this coal refinery concept and others in the Devolatilization category are its implementation of circulating fluidized bed (CFB) technology for coal pyrolysis and the integration of the pyrolyzer and a CFB combustor in an advanced design for electric generation [2]. This design overcomes an inherent limitation of using CFB's for power production: the turbine inlet temperature is typically limited by the operating temperature of the CFB, which is in the neighborhood of 1,850°F while the inlet temperature for gas turbines can be as high as 2,300°F. The SFuel concept partial gasifies the coal to form char and fuel gas, with subsequent firing of the fuel gas in a gas turbine at temperatures above the operating temperature of the CFB.

The coal refinery based on the SFuel Process is shown in Figure 1. The necessary inputs to this coal refinery would include run-of-mine coal, air, water, and dolomite (for sorbent purposes), while major products include a smokeless solid fuel and excess electricity.

Detailed Process Description

Run-of-mine coal is ground to a particle size of about 0.2-inch and dried in the Coal Preparation section. The prepared coal is then fed into the air-fed Circulating Fluidized Bed Reactor (CFBR) operating at temperatures of about 1,750°F. The coal thermally decomposes (partially gasifies) to char and a fuel gas which is burned in the Gas Turbine section. The oxidant for the CFBR is air, resulting in a lower-heating value fuel gas but avoiding the expense of an air-separation plant required for an oxygen feed. Sulfur is removed within the CFBR by the addition of dolomite, avoiding more extensive downstream processing.

In the Briquetting section, the hot (1,530°F) solid char is mixed with preheated (300°F) caking coal by use of an externally heated screw mixer. The temperature of the approximate 50/50 percent mix reaches the range of 790 to 850°F, at which point the caking coal softens and becomes plastic, sticking to the char. The hot mixture is then roll-pressed at high pressure (approximately 4,400 psia) into briquettes of smokeless fuel (SFuel).

The temperature of the raw gas stream exiting the CFBR must be lowered before it can be cleaned of particulates, alkali and other contaminants (tar) in the Gas Cleanup section. A Waste-Heat Boiler cools the raw gas to a temperature of about 660°F while generating steam. The steam could be used for additional electric generation, process heat, or for industrial heating purposes. A portion of the cooled raw gas stream is sent to heat the externally heated screw feeder in the Briquetting section while the majority is sent to the Gas Cleanup section. The heat evolved in the Gas Cleanup section preheats the dolomite going to the CFBR, increasing the thermal efficiency of the overall process. The pressure of the fuel gas is too great for introduction into the gas turbines to burn, so it must be decreased by expansion; this reduction in pressure can be converted into mechanical energy which is further converted into electrical energy.

Combustion air for the pressurized combustor is provided by a gas-turbine Compressor. The coal-derived fuel gas is a mixture of the fuel components H_2, CO, CH_4, higher hydrocarbons, and diluents CO_2, N_2, and H_2O with a heating value around 140 Btu per standard cubic foot (a low-Btu gas). The compressed air is mixed and burned with the fuel gas in a topping Combustor (afterburner) and the resulting hot gases expand through the Gas Turbine Generator at a temperature of 2,100°F to produce approximately a third (0.34 kWh per lb processed coal) of the electric power produced in the overall concept.

The flue gases from the Gas Turbine Generator contain significant energy that can be utilized. These exhaust gases contain oxygen in the range of 14 to 15 percent and are fed into the second CFB with processed coal and dolomite. Steam is produced from heat exchanger tubes located within the CFB and sent to the Steam Turbine. The majority of the electric power (0.68 kWh per lb processed coal) for this coal refinery is generated in the Steam Turbine. The solid waste stream exiting the CFB contains ash and spent sorbent.

This advanced, highly integrated design is expected to achieve a heat rate of around 8,100 to 8,500 Btu per kWh (thermal efficiency of 40 to 42 percent) assuming a gas-turbine inlet temperature of 2,100°F. This is to be compared with a pulverized-coal plant having current pollution control technology which may have a thermal efficiency as low as 37 percent [3]. Increasing the allowable inlet temperature to 2,300°F, which is exhibited by the new generation gas turbines [2], will increase the thermal efficiency, potentially achieving heat rates below 8,000 Btu per kWh.

Types of Feed Coal

The characteristics of the pyrolysis reaction to form char and fuel gas have been tested in a pilot plant CFBR [1]. The results are given in Table 1, together with the characteristics of the Polish feed coals used. These Polish bituminous hard coals are relatively low in sulfur and ash content, similar in characteristics with dewatered low-sulfur Western (i.e., Powder River Basin, Helper Utah high-volatile bituminous) and high-quality Eastern Applachian (i.e., Upper Kittanning West Virginia low-volatile bituminous, Ohio No. 5 & 6 bituminous) coals.

It would therefore be reasonable to expect that this coal refinery concept can be successfully applied in this country.

Products

The main products of this coal refinery are a smokeless fuel and electricity. This analysis indicates that 0.55 pounds of smokeless fuel and 1.02 kWh of electricity are produced from one pound of processed coal (shown in Table 2).

The parameters upon which the ratio of the amount of smokeless fuel to electricity will depend include the air-to-processed coal ratio, coal type, initial coal particle size, ratio of coal fed to the CFBR versus the CFB, and operating parameters such as the feed rate of processed coal to the CFBR and the CFBR operating temperature [4].

Likely Applications

The SFuel concept is envisioned by the Institute of Chemical Processing of Coal to replace coal used in domestic consumption and power production with a smokeless solid fuel in Poland. Poland is somewhat unique with respect to the U.S. in that about 90 percent of the energy market in Poland is satisfied by utilization of coal. The availability of high-quality hard coal is expected to decrease in the near future in Poland. This concept would substitute hard coal with the smokeless fuel.

In the U.S., the use of a smokeless char fuel as a replacement for wood has been advocated to decrease carbon monoxide and particulate emissions from wood-burning stoves and fireplaces [5]. The smokeless fuel would look like a coal that has been melted, and is lighter than the parent coal and dust-free in handling. In addition, the smokeless fuel would provide a more uniform fuel than wood.

The commercial entities that potentially could be interested in this coal refinery concept would include coal mine owners (to add value to the coal and to expand their markets), electric utilities (by using the char as a compliance fuel), and independent power producers.

Status of Development

A 7.2 ton per day CFB test reactor has been constructed and a series of experiments performed to determine the effect of basic parameters on the individual yields of char and gas, temperature distribution within the CFB, and distribution of the light gases that are evolved (see Table 1). The parameter which had the greatest effect on the overall performance is the air-to-coal ratio fed into the CFB, as it determines the circulation rate of the solids around the CFB (and therefore their residence time) and the degree by which secondary reactions (i.e., char gasification to form light gases; oxidation of CO and H_2 to CO_2 and H_2O) can proceed.

A 200 ton per day pilot plant is being commissioned, with startup expected in 1993.

Environmental Aspects

The SFuel concept can theoretically convert high-sulfur coal into a char that is low in sulfur. Typically, up to 90 percent of the sulfur in the coal is captured by the dolomite in the form of calcium sulfate ($CaSO_4$) while the sulfur compounds in the gas phase can be converted into elemental sulfur so that the emission rate of SO_2 can be expected to be quite low.

The main environmental intrusions from this coal refinery include the nitrous oxides produced in the combustor and the solid wastes from the second CFB. Steam or water can be employed to suppress NO_x formation by up to 70 percent in the fuel gas combustor; NO_x emissions levels of between 30 to 70 ppm at 15 percent excess O_2 can be achieved using current combustor technology [2].

The solid waste would include the bottom and fly ash from the feed coal, and spent sorbent along with some unreacted lime. It is expected that the toxicity of the solid waste would not differ greatly from that generated from a coal-fired power plant using fluidized bed technology [6].

Research Needs

The operating characteristics of the CFBR have been tested at the pilot scale. It however remains to demonstrate that coal can be efficiently and reliably pyrolyzed using circulating fluidized bed technology on a commercial scale. Also, demonstration of the successful integration of the individual process components may be warranted.

The use of air in the CFBR results in a fuel gas with a low Btu value. Potential application of hot gas cleanup technology in this coal refinery concept would avoid major sensible heat loss in the fuel gas (which constitutes a high proportion of the fuel's energy content).

Development of process models that simulate the effect of process variables may aid in determining the most efficient and cost effective design for this coal refinery concept.

The percentage of higher hydrocarbons in the fuel gas ranges between 4 and 8 volume percent. Identification of the chemical compounds present in the fuel gas stream may potentially allow their conversion into value-added products (such as methanol and oxygenates) or sale as LPG.

Table 1: Results of Pyrolysis in Pilot Plant CFBR

COAL:	Wieczorek	Siemianow	Moszczen	Myslowice	Staszic
ULTIMATE ANALYSIS (weight percent, dry basis)					
Carbon	73.18	71.4	82.26	68.09	74.1
Hydrogen	4.26	4.6	3.83	4.17	4.61
Nitrogen	1.59	1.33	1.38	1.43	1.13
Oxygen	10.5	12.26	4.62	10.56	10.21
Sulfur	0.87	1.11	0.36	0.51	0.65
Ash	9.6	9.3	7.55	15.24	9.3
OPERATING CONDITIONS					
Air/Coal Ratio (lb per lb coal)	1.04	0.98	0.85	1.18	1.33
Pyrolysis Temp (°F)	1,690	1,690	1,725	1,725	1,780
YIELD (lb per lb dry coal)					
Char	0.566	0.465	0.675	0.605	0.46
Gas	1.442	1.49	1.156	1.542	1.838
GAS COMPOSITION (volume percent, dry basis)					
H_2	7.2	6.5	7.7	5.2	5.0
CO	9.1	8.1	5.2	7.2	9.3
CH_4	4.9	4.0	4.6	3.9	3.8
C_nH_m	5.2	8.4	6.6	5.9	4.6
CO_2	14.1	15.5	13.8	13.0	12.5
N_2	59.5	57.5	62.1	64.8	64.8
Heating Value (Btu per scf)	180	155	120	120	130

Table 2: Product Output from Example SFuel Concept Coal Refinery

PRODUCT	OUTPUT (lb per lb of processed coal)
Smokeless Solid Fuel (SFuel)	0.55
Electricity	1.02 kWh per lb processed coal

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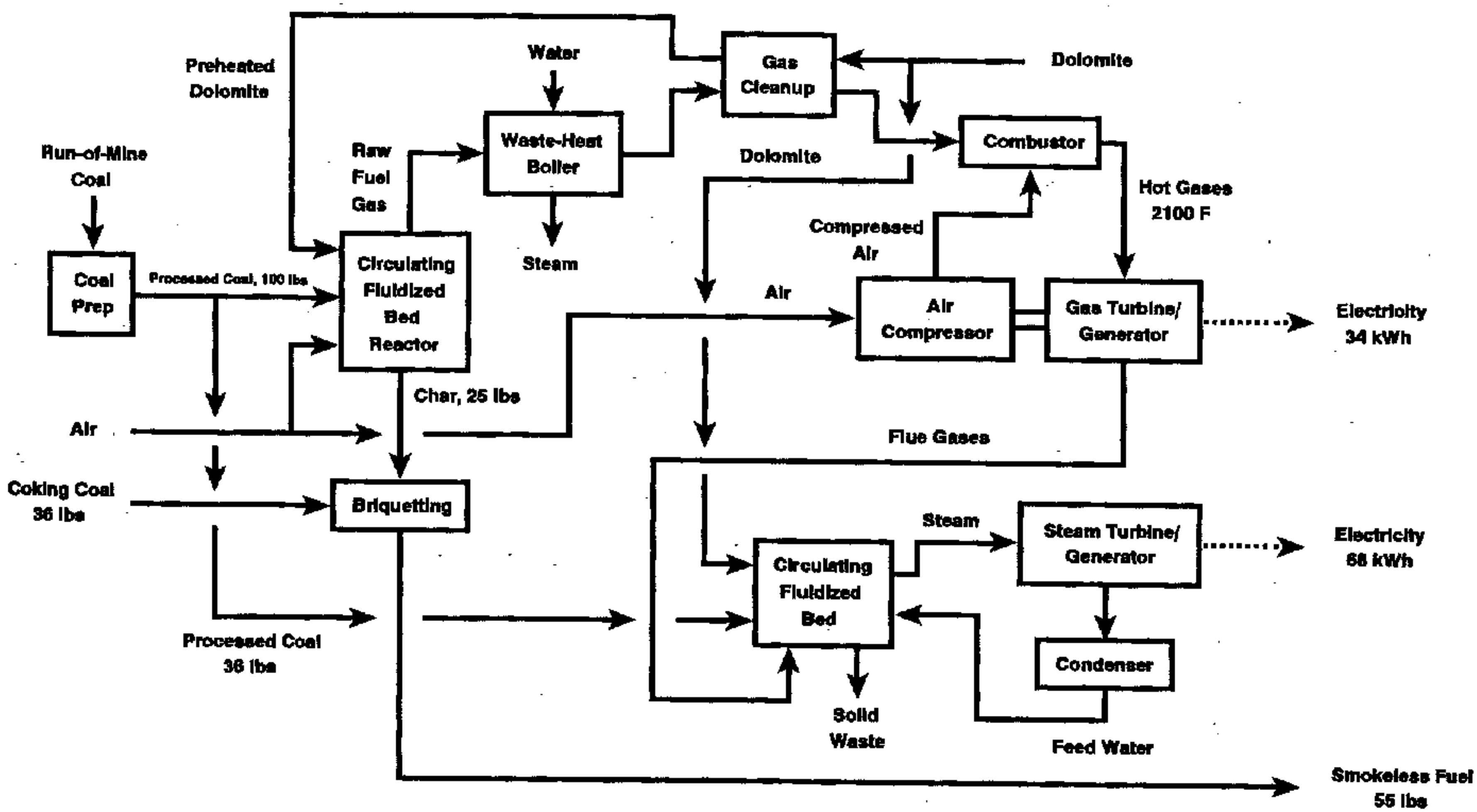

FIGURE 1: Overall Block Diagram of the SFuel Concept

UNDEERC-AMAX MILD GASIFICATION

Introduction

This concept is under development by the Energy and Environmental Research Center (EERC) of the University of North Dakota and AMAX R&D Center of Golden, Colorado (AMAX). This process development effort is targeted to produce metallurgical coke substitute, carbon black, activated carbon, diesel fuel additives, chemical feedstock materials, and pure chemicals [1].

This concept is being developed using a 100 pounds per hour process research unit (PRU). A high-sulfur midwestern bituminous coal and a low-rank, low-sulfur western coal are being evaluated in the PRU. The PRU consists of a fast fluidized-bed carbonizer designed to optimize coal liquids production and a bubbling fluidized-bed calciner to reduce volatile matter and sulfur in the char to acceptable levels for production of metallurgical coke. Condensables are collected in three unit operations: a high-temperature tar venturi scrubber, a sieve tower, and a water venturi scrubber. This process concept is similar to the petroleum-refining process in that several coal products are produced that meet the needs of different end users. The capability to alter product distributions, either by changing feedstocks or processing conditions, permits timely response to the ever-changing market [2].

Detailed Process Description

The PRU was constructed to provide a database for an integrated mild gasification system operating on specific design coals and to produce char and liquid products for upgrade testing and market evaluation. The unit was designed to process 100 pounds per hour (dry basis) of feed coal and to incorporate capabilities for drying, carbonizing, and calcining caking and noncaking coals in fluid-bed reactors and for separating char, liquid, and gaseous products. The unit was designed to enable both integrated and independent operation of the carbonizer and calciner process sections. Design of a conceptual commercial plant, which will process 1,000 tons per day of coal, is presently underway.

Heat for carbonization is principally supplied by hot flue gas from combustion of natural gas. Figure 1 is a diagram of the PRU carbonization area and shows the major components of the system. For operation with Wyodak coal, the coal is screened to a size of ¼-inch by 0. The carbonizer was designed as a spouted-bed gasifier. The operative principle allowing use of caking coal in this design is the dilution of the entering coal by internal recycle of char to the bottom of the tapered bed, where high velocity and low-bed density reduce tendency for agglomeration. Char residence time can be varied, however, the design residence time is 30 minutes. The carbonizer operates at temperatures of 900 to 1,500°F. Feed coal is entrained and fed into the bottom of the reactor using nitrogen preheated to 650°F. Char can be drawn from a variety of locations and injected into a nitrogen-purged tote bin [1].

The condensables (liquids) quench and separation system was designed to produce tar and oil fractions that meet primary product requirements. Ideally, the quench system should produce no waste-water condensables. Tar and oil recovery from the gas stream is based on direct contact tar venturi scrubber and sieve tower, respectively. Water from the gas stream is condensed in the direct contact venturi scrubber.

Types of Feed Coal

Bench-scale tests were conducted at the EERC on three different coals: Indiana No. 3 bituminous, Wyodak subbituminous, and Texas lignite. These tests were carried out at various temperatures and with different fractions of steam included in the nitrogen gas used for heating the coal. Design feed coals for the PRU testing are Wyodak subbituminous, Indiana No. 3 bituminous, and Illinois No. 6 bituminous.

Test results have confirmed that liquids and char yields are dependent on the coal feedstock used and the gasification conditions.

Products

Char yield represents about 59 to 70 weight percent of the moisture and ash- free (MAF) coal fed to the reactor for all the three aforementioned coals. Liquid yields were found to be 14 to 18 percent MAF for coal Indiana No. 3 bituminous, 13 to 14 percent MAF for the Texas lignite, and 8 to 10 percent MAF for the Wyodak subbituminous coal. These yields are based on the data obtained in bench-scale and PRU tests.

The ash and sulfur contents of the char are important characteristics in determining the feasibility of using the char in metallurgical applications. Typical specifications for metallurgical coke are a maximum of 10 percent ash and 1 percent sulfur. Based on the PRU tests, the volatile content of char produced at 1,100°F ranged in from 15 percent for the subbituminous coal to 13 percent for the bituminous coal. Ash contents were 10 percent and 19 percent, respectively, for the subbituminous and bituminous coal chars, while sulfur contents were 0.4 percent and 3 percent, respectively.

The Pellet Technology Corporation (PTC) utilized Wyodak char from an EERC test performed under conditions similar to those employed for the PRU test as a metallurgical coke substitute in iron ore-reducing tests. The PTC tests were performed using pellets made from Wyodak char, iron ore, lime, and silica. Pellets made with a 10 weight percent binder comprised of calcium oxide and silica exhibited satisfactory strength, density, and abrasion resistance. The results of the ore reduction tests, in which the pellets were subjected to temperatures of 3,500°F and 2,700°F, showed that iron ore reduction times for char-iron pellets could be reduced by as much as 80 percent compared to reduction times required for coke-iron ore pellets.

Three distinct boiling point fractions of liquids were collected from the PRU condensation train. With subbituminous coal, the tar scrubber yielded liquids ranging in boiling point from 715 to 930°F, the sieve liquids ranged from 390 to 880°F, and water scrubber liquids ranged from 340 to 825°F. With the bituminous coal, the tar scrubber yielded liquids ranging in boiling point from 750 to 950°F, and the combined sieve tower/water scrubber liquids ranged from 340 to 860°F.

Likely Applications

The economic feasibility of mild gasification is contingent upon obtaining a premium market for the char, probably as a metallurgical coke substitute in the U.S. steel production industry which is showing signs of economic recovery and is, therefore, in a position to take advantage of new technology. A new iron-making process developed by PTC can use mild gasification char in highly reactive iron ore/char pellets that greatly increases throughput in a conventional blast furnace. The pellets can also be used in smaller, more-efficient ore reduction equipment such as hot blast cupolas and direct reduction systems with a market potential of up to 20 million tons per year [3]. The mild gasification concept can be economically integrated with the existing power generating stations by gasifying their coal-fines waste to produce value-added products. Liquids and gas produced can also be used for additional electric power production.

The best opportunity for liquids in the transportation fuels market may be as a fuel additive for medium-speed railroad diesel engines. Other possibilities for condensibles include their use as feedstocks for production of carbon black (used in rubber goods, pigments, printer's ink, and in the production of carbon electrodes for aluminum ore reduction), creosote, cresylic acid, pitch, rubber-processing oil, and as a briquetting binder and a coal dust suppressant. Perhaps the best use of mild gasification process gas (from a 1,000 ton per day facility) will be as on-site plant fuel, with excess used for cogeneration of electricity.

Status of Development

During a period of 1987 to 1991, development activities included a market assessment, a process literature survey, 1 pound per hour bench-scale tests, and 100 pound per hour PRU tests. Nine shakedown tests were performed on the PRU during 1990. The last two PRU tests were performed on Wyodak coal and Indiana No. 3 coal, respectively, to achieve process and operational data and material balance data.

Additional tests are planned for in-situ sulfur capture and integrated operation of the PRU for char upgrading and acquisition of more-comprehensive technical data.

Concurrently, technical and economic evaluation of the EERC-AMAX mild gasification coal refinery concept is underway based on a 1,000 ton per day conceptual plant design.

Research Needs

Additional investigations in the following related areas could be beneficial to this mild gasification concept: (a) evaluation of mild gasification char in iron ore reduction processes; (b) further studies of methods to upgrade coal-derived liquids, and evaluations of these liquids as commercial products; (c) further development of methods for identification and quantification of sulfur species in coal-derived liquids, and subsequent development of better ways to remove sulfur-containing heteroatomic species; (d) based on the available data base, determination of the technical and economic feasibility of a commercial-scale mild gasification plant using this process.

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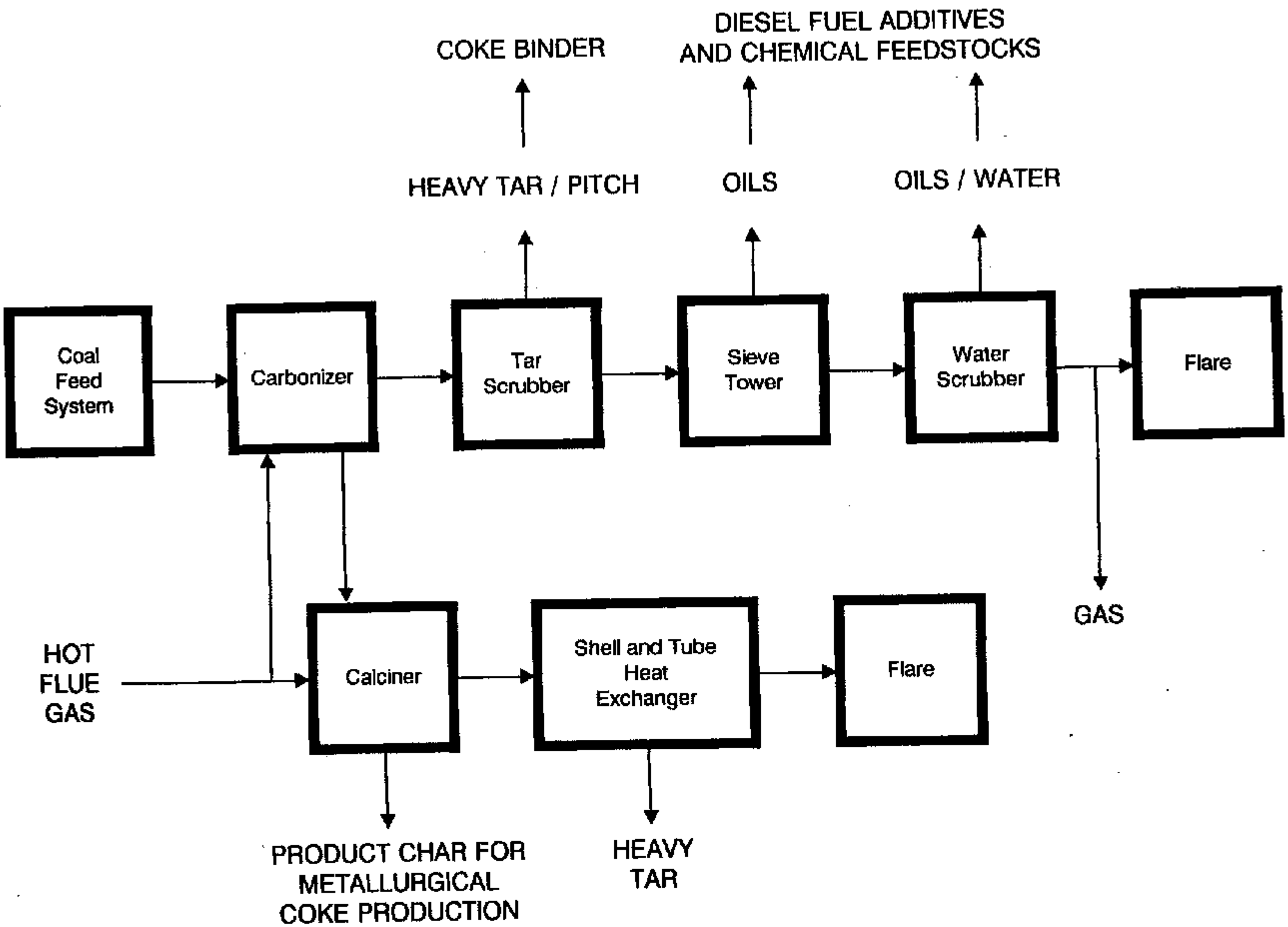
Figure 1: UNDEERC-AMAX MILD GASIFICATION PRU



WRI-AMAX MILD GASIFICATION

Introduction

As with all processes in the Devolatilization category, the WRI-AMAX concept uses moderate temperature and pressure conditions to devolatilize the coal thereby producing some liquids and a solid char. Unlike many other concepts in this category however, the WRI-AMAX concept upgrades the char into higher value products such as carbon powder for use in rubber tires or as anode carbon for use in aluminum production. Each of these products has a value considerably greater than the basic char which is typically used as a fuel in electric utility or industrial boilers.

This concept is under development through a collaborative effort among the Western Research Institute (WRI) of Laramie, Wyoming, AMAX R&D Center of Golden, Colorado, and the Riley Stoker Corporation (RSC) of Worcester, Massachusetts. It has been supported by the DOE Office of Fossil Energy.

Process Description

The WRI-AMAX process is illustrated in Fig. 1 where a conceptual flowsheet for a 1000 ton-per-day commercial plant is shown. This conceptual design is based on using Powder River Basin subbituminous coal as the feedstock. Use of other coals would necessitate some differences in the facility design although the basic overall process would be essentially the same.

As seen in this figure (taken from Ref. 1), the coal is crushed and sized to less than 0.15 inch and then sent to an inclined fluid-bed reactor for drying. Here, the coal is exposed to a flowing stream of CO_2 and is heated to about 500° F thereby decreasing the moisture content of the coal from about 30 percent (typical of Western subbituminous coal) to about 2 percent. This drying process helps to increase the liquid yield from the mild gasification process and also reduces difficulties resulting from agglomeration and swelling of caking coals. Gas exiting the dryer is cleaned and cooled to condense the water. The CO_2 is reheated and recycled.

After leaving the dryer, the coal goes to another inclined fluid-bed mild gasification reactor where it is heated in the absence of air and at ambient pressure to a temperature in the range of 1,000 to 1,200°F. Heating is provided by gases from an atmospheric fluidized-bed combustor (AFBC) and, perhaps, steam if it is found that steam could enhance the liquid yield and reduce the sulfur content of the char. Economic considerations (e.g., the value of the liquid products) would also play a role in deciding on the use of steam in this manner. At the temperature in the mild-gasification reactor, most of the volatiles are driven off leaving a solid char. These released volatiles are condensed in stages to yield three liquid products: dirty pitch, clean pitch, and oil. The volatiles remaining in gaseous form can be used for several purposes.

Part of this gas is preheated and recycled to the mild-gasification reactor where it heats the coal while providing the necessary reducing atmosphere in the reactor. Because this gas stream contains a mixture of CO, H_2, CH_4, and CO_2, it can be burned to provide heat for the char-to-carbon process used to upgrade the char. As shown in the figure, part of this stream is bled off for this purpose.

A distinguishing feature of the WRI-AMAX concept is that the char is upgraded to produce a product having a value greater than that of the basic char. This upgrading is accomplished through a char-to-carbon (CTC) process under development at AMAX. In this process, the char from the mild-gasification reactor is sent to tandem reactor systems. In the first reactor (a methanation or hydrogenation reactor), the char is heated to a temperature of 1,200 to 1,400°F by using a recirculating gas rich in hydrogen. Steam may also be used to heat the char to the necessary temperature. At this temperature and in the presence of hydrogen, the carbon in the char is converted to methane (CH_4) while the spent char retains most of the ash that has been carried over from the mild-gasification reactor with the char. The methane-rich gas exiting the first reactor is cleaned of any entrained ash and sent to the second reactor (a methane cracking reactor) where it is heated to a temperature 2,400°F to 2,600°F by direct firing of some gas from the mild-gasification reactor and a bleed stream from the CTC loop. At this temperature, the methane will decompose to yield pure carbon and hydrogen. This hydrogen is recycled back to the first CTC reactor for use in converting char to methane. It may be noted that no hydrogen is consumed in this two-stage CTC process. In fact, due to the hydrogen released from the char, the CTC process is a net hydrogen producer. The two refractory lined reactors used in the methanation process and the methane cracking process will alternate duty in a cyclic manner.

Ash from the char-to-methane reactor is collected along with unconverted carbon and sent to the AFBC unit where it is burned to produce high-pressure steam used in the production of electricity. This AFBC also burns fines from the various particulate collection devices throughout the plant and also serves to process any organic contaminated water or other wastes from the plant. In addition to electricity, the AFBC also provides heat for the drying and mild gasification steps in the WRI-AMAX process.

Coal Feeds

Tests have been done at WRI with the subbituminous coal form AMAX's Eagle Butte Mine and are reported in Refs. 1-3. The results of these tests were used in preparing the conceptual design for the 1000 ton/day facility that is the basis for much of the information presented herein.

As a consequence of the test results conducted at WRI, conceptual design work, process yield and economic analyses have concentrated on the use of subbituminous coal from the Eagle Butte Mine in the Powder River Basin near Gillette, Wyoming. In theory, any coal could be used in the WRI-AMAX concept although the yields and char properties would vary and these would impact the design and operation of certain components in the plant. For example, the use of high-sulfur bituminous coals from the Illinois Basin may require that precleaning of the coal be done before it is sent to the mild gasification reactor. This step, which could be done with heavy-media cyclones, could reduce sulfur and ash contents of the coal to levels that can be handled within the basic WRI-AMAX concept.

Products

A principal feature of the WRI-AMAX concept as compared to other mild gasification concepts is the upgrading of the char to a more valuable product. The product slate can be somewhat altered depending on the coal feedstock and the value of different possible products. Reference 1 presents a product slate for the Eagle Butte subbituminous coal. A plant capacity of 1,000 ton per day and an annual capacity factor of 90 percent were used in this analysis.

One of the liquid products from this concept is referred to as "dirty pitch." Approximately 15,000 tons of this product (which may contain some particulates) would be produced per year in the reference plant. This liquid can be sprayed on dried coal to minimize moisture reabsorption and dust formation that tend to occur during shipping. It would also help to suppress the self-ignition tendencies of sub-bituminous coals. Reference 1 notes that a petroleum-based product costing \$100-\$150 per ton (\$0.40 - \$0.60 per gallon) is currently being used at the 1 million ton/year AMAX drying plant at the Belle Ayr Mine in the Powder River Basin for this purpose.

Approximately 20,000 tons per year of "clean pitch" would be produced. This liquid would be upgraded to meet the specifications of coke-oven pitch used by aluminum companies in the manufacture of carbon anodes. Coke-oven pitch is presently imported at a price of \$150-\$250/ton.

The third liquid is an oil product that can be upgraded and blended with diesel fuel for use in operating the heavy equipment at the plant and/or the mine. If excess quantities of this product are produced, it could be exported. Approximately 25,000 tons of this product would be produced annually. In theory, these oil products could be upgraded to highly refined products such as gasoline. In general however, the quantities produced from mild-gasification processes are too small to justify the expensive upgrading and refining that would be needed. A possible market for these oil products with minimal upgrading is railroad diesel fuel although additional characterization of the performance of diesels with such a fuel must be completed.

The solid product from the WRI-AMAX coal refinery is a high-purity carbon powder. Approximately 95,000 tons of this powder would be produced annually from the reference plant. This product could be used as carbon black in the rubber-tire industry (where the current price is in the range of $400 to $600/ton) or as an anode carbon in the aluminum industry where the market value is typically $115 to $175/ton although some site-specific costs are as high as $200/ton.

An alternative use for the carbon produced in this process is as a premium fuel that is ash, sulfur, and moisture free. A price of approximately $100/ton (about $3/million Btu) has been projected for this type of fuel. This market can be expected to grow as environmental regulations for electric power plants and industrial facilities become more stringent.

Gaseous products will be used internally to provide heat for drying and in the carbon reactors. Excess heat from the AFBC and the methanation reactor is used to raise steam to be used in electric power production. Approximately 10 MW of electric power (beyond that used in the plant itself) would be produced in the reference plant.

Likely Applications

As presently envisioned, the WRI-AMAX concept is most likely to be used in conjunction with a mining operation. The reference case commercial plant discussed herein is largely based on AMAX needs in their coal drying and metals production industries. Other possible owners or operators of a coal refinery based on this concept include mine owners and fuel vendors who wish to upgrade the raw coal into a more marketable product.

Status of Development

Several coal drying tests and mild gasification tests have been conducted with some of these being summarized in Refs. 1-4. These tests were at bench-scale and helped to determine the design and operating parameters such as bed temperature, coal particle size, and moisture content. It was determined that a gas temperature of 575°F and a 2-5 minute residence time in the dryer would produce a coal of less than one percent moisture.

Bench-scale tests were also done the evaluate the effect of design and operating conditions on the liquid yields and on the composition of the char and gas produced. These tests led to the development of the inclined fluid-bed mild-gasification reactor with the coal and gases configured in a cross-flow mode. This arrangement provides high rates of heat and mass transfer, a short gas residence time, and a long solid residence time. These design parameters lead to more rapid removal of the devolatilization products thereby minimizing secondary reactions and increasing liquid yield, particularly that of the clean pitch.

Char upgrading tests were also done at the bench scale (50-100 grams of char) and are summarized in Refs. 1-3. The AMAX process used for upgrading the char is proprietary with patents pending so that few details are publicly available. Results of tests done on subbituminous coal however showed that high-purity carbon (in excess of 99 percent pure) could be attained at temperatures in the range of 2,400-2,600°F. It was also found that by increasing the temperature and decreasing the residence time, the carbon produced in the CTC process would have particle sizes typical of the carbon black used in the rubber tire industry.

Impacts of selected coal feedstocks on the product yields and on design features have also been investigated in these bench-scale tests.

The bench-scale tests noted above provided the basis for Process Demonstration Units (PDUs) equivalent to about 100 pounds of coal per hour. A mild gasification PDU at WRI began operation in the summer of 1989. A 50 pound-per-hour CTC Process Research Unit (PRU) is at AMAX R&D.

AMAX has done additional process development work along with product evaluations, market analyses, and economic feasibility studies for a 1,000 ton/day commercial plant. These analyses serve as the basis for the "Products" and "Economics" discussions contained herein.

Economic analyses have been done for the WRI/AMAX concept and are discussed in Refs. 1 and 4.

Environmental Aspects

Environmental releases from this plant are anticipated to be comparatively small. Atmospheric emissions will consist principally of nitrogen and carbon dioxide with some sulfur dioxide. Experimental results reported on Ref. 5 show that during the CTC process, most of the sulfur remains in the char rather than being converted to hydrogen sulfide. The SO_2 emissions will be controlled by feeding limestone into the AFBC unit. As noted earlier, the AFBC will also be used in the treatment of organically contaminated water and other waste products. The main solid waste will be the ash and sulfated lime (along with some unreacted lime) from the AFBC. This solid waste could be disposed of at the mine site.

Research Needs

Further operation of the PDUs for the mild gasification and the CTC units will help to reduce the uncertainties in the performance of these units. Integration of the major components into a single system rather than operating as individual units as is currently the case could also help to advance this concept. Some of these integration effects could be examined in a facility constructed at the pilot scale of 10-20 tons/day.

Optimum design and operating conditions can be determined for each coal type after the influence of different coal types has been more thoroughly investigated. Economic analyses could then be done to evaluate the feasibility of using different coal types and/or the economic conditions that would be necessary for the processes to become economically viable. Such analyses could indicate, for example, how the inclusion of a precleaning step impacts the overall process economics for the Indiana No. 3 coal used in some of the early testing of this process.

Additional economic studies could help in the evaluation of the impacts of changing design and operating parameters and on the value of the products. For example, it may be advantageous to use more heat recuperators or steam generators at various points in the facility than are used in the current conceptual design with Wyodak subbituminous coal. Some environmental systems such as waste water treatment and atmospheric emission control systems must be more thoroughly examined and incorporated into the facility design, operations, and economic considerations.

References

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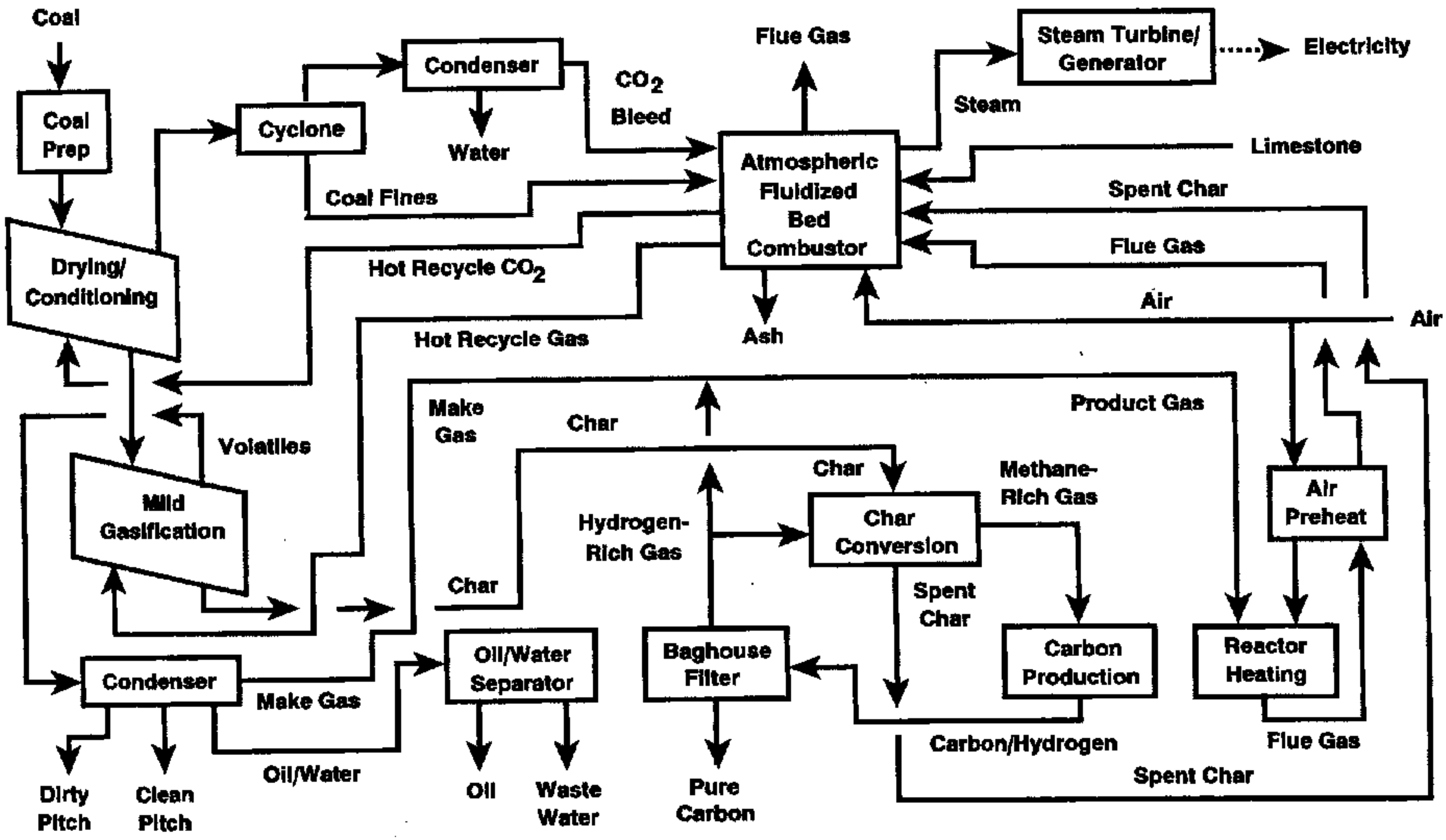

FIGURE 1: Overall Block Diagram of the WRI-AMAX Mild Gasification Concept