COAL LIQUID AND COKE MILD GASIFICATION

Introduction

The concept based on the Coal Liquid and Coke Mild Gasification process (CLC) is an example of a coal refinery in the Devolatilization category. Significant liquid yields can in general be achieved using high-volatile bituminous coals under mild gasification conditions. However, high-volatile bituminous coals are also caking coals, which when rapidly heated, first soften, become sticky, swell and then agglomerate or "cake" into a coherent porous, carbon-rich solid mass called 'clinkers'. Typically, special precautions have to be taken when feeding caking coals into a hot mild gasification reactor to prevent plugging. The CLC process is a potential method by which to convert caking coals into coal liquids and coke by continuous mild gasification.

This coal refinery concept is being advanced by CLC Associates, Inc. of Bristol, Virginia. Essentially, coal is processed under mild conditions using an externally-heated screw pyrolyzer to extract liquids from coal and create char. The externally-heated screw pyrolyzer is a fairly simple system in that recirculation of solids (e.g., for heating purposes) is not required. The liquids are further processed into transportation fuels (gasoline, diesel). The char formed from caking coal is upgraded into form coke usable for the reduction of iron ore in a blast furnace.

The coal refinery concept based on the CLC process is shown in Figure 1. The system would integrate the generation of char, which is then processed into form coke, with the evolution of coal-derived liquids. The necessary inputs to this coal refinery would be cleaned process coal, cleaned binder coal (a coking coal), a source of process heat (generated from the non-condensible gases), water, air, and nitrogen (purge gas), while major products could include transportation fuel-blend stock derived from the coal liquid, and form coke.

Detailed Process Description

The process feed coal is an Eastern high-volatile highly caking bituminous coal; a representative coal is East Kentucky H&K Williamson #2 bituminous (sulfur 1.0 weight percent, ash 6.9 weight percent, on a dry basis). The feed coal is ground to minus ¼-inch size, preheated to drive off moisture and then fed under a nitrogen blanket into the Pyrolyzer.

The Pyrolyzer consists of a pair of interfolded screw conveyors enclosed in an insulated chamber. The outside of the Pyrolyzer unit as well as the hollow screw shaft is heated by radiant gas heaters [1]. Coal is mechanically moved through the Pyrolyzer and heated to a temperature between 800 and 1,200°F (typically 1,000°F) in the absence of air. Coal devolatilizes into char and a gaseous phase containing water vapor, vaporized hydrocarbon liquids, and non-condensible gases (primarily H_2, CO, CH_4, etc.).

The screw design for the Pyrolyzer provides a approximate residence time of 20 minutes and a slow heating rate which is believed to optimize liquid product quality [2].

Hot char is discharged to a char cooler, while the vapors from the coal are condensed in the Gas/Liquid Separator to recover the liquid hydrocarbons and water. A part of the non-condensible gases are recycled to satisfy the process heat requirement, with the remainder initially burned in an incinerator. Eventually, the excess gas may be used as a feedstock for production of coal-derived liquids (e.g., methanol, diesel fuel oil), or used to generate steam and power in a waste heat boiler with associated cogeneration equipment.

The coal-derived liquids are separated from the water in the Oil/Water Separator. The waste water from the Oil/Water Separator will be burned with the excess non-condensible gases in the Incinerator, to mitigate any environmental problems. As an alternative, the waste water may be treated using conventional technology to separate out useful chemicals, such as phenolic compounds, etc. The crude coal-derived liquids may be upgraded into transportation-fuel blend stock in the Fuel Refining section [3].

The char exiting the Pyrolyzer enters a water-cooled screw char cooler where it is cooled below its ignition temperature (in the range of 500°F). The char is then ground (delumped) and mixed with binder compounds (such as pitch, coal tar, "binder" coking coal, etc.) in order to impart higher strength. The mixture is formed into moist, raw (green) briquettes in the Briquetter section; the briquetting process, in essence, is one in which the char particles, covered with adhesive, are pressed into molds. The briquettes are fed into the Coker unit, which is a specially designed rotary hearth oven, and are calcined at about 2,000°F over a 100 minute duration. Volatiles and water are driven off, to produce high-quality blast furnace formed coke of the desired density. The hot coke is then quenched before storage. The evolved volatile material is burned in the Coker, providing the fuel input to the Coker unit, with the excess going to the Pyrolyzer furnace.

Air pollution control equipment (Scrubber unit) remove SO_2 and particulates from the many flue gas streams from the process before the flue gas exhausts through the stack.

Types of Feed Coal

This process has been designed to primarily process Eastern high-volatile caking-type bituminous coals. In principle, any high-volatile coal (such as from the Midwest) may be successfully processed as long as the volatility of the coal is such that reasonable liquid yields can be achieved at moderate temperatures. The operating conditions and design details of the process will depend on the process coal feedstock.

Products

Char represents approximately about 60 to 70 weight percent of coal feedstock in yield, and is the major product of this coal refinery concept. Therefore, 1000 pounds of process coal is converted to approximately 600 pounds of char; addition of approximately 400 pounds of binder (coking) coal, pitch and coal tar results in 1,000 pounds of the "green" briquettes. The final form coke yield is approximately 800 pounds. Based on a commercial plant processing about 2,000 tons per day of process coal, approximately 84,000 gallons of crude coal liquids would be produced (1 barrel of liquid per ton of coal) [4]. This translates into the coproduction of 0.8 pound of form coke and 0.04 gallons of crude coal liquids from 1 pound of process caking coal (and 0.4 pounds of binder coal).

Likely Applications

The commercial organizations that may be interested in pursuing this coal refinery concept include mine owners (to produce a higher value product distribution from the parent coal), and the steel industry (to assure a dependable, indigenous supply of form coke).

The coal-derived liquids can be blended with petroleum stocks in special formulations developed at Coal Technology Corporation (CTC) to produce gasoline and diesel fuels [1, 5]. Commercial fuel suppliers may be interested in this concept, to displace petroleum in transportation and heating fuels.

Status of Development

During the period of 1983 to 1990, many mild gasification tests have been performed on the pilot scale [6]. The batch-operated 100 pound per hour pilot unit consists of vertical retort tubes contained within a furnace box. The liquid yield was on the order of 8 to 10 percent of the dry feed coal. Currently, CTC is operating a pilot plant unit with a 1,000 pound per hour coal feed rate. This unit has been in operation since March 1991 and is designed to supply scale-up data.

Coking experiments have been performed in a briquetting coking oven (inner volume of 90 cubic feet) designed and constructed by CTC [1]. High-quality coke could not be made from char alone, requiring binder materials to attain the desired coke strength and reactivity. It was established that the binder materials had to be mixed with the char and formed into briquettes before coking was performed. The effect of amount of char, binder coal, and other binders was studied as well as the effect of final coke oven temperature and heatup rate.

A proposal for a prototype 1,500 ton per day demonstration plant of this coal refinery concept has been submitted for the fourth solicitation of the Clean Coal Technology program [3].

Economic viability of the process has been preliminarily demonstrated based on pilot scale operating data [1].

Research Needs

Production of form coke from other coal types may warrant investigation into the suitable operating conditions (for parameters such as briquetting temperature, binder selection, particle size, binder compositions).

The coal-derived liquids from this coal refinery concept are currently destined for the transportation market. Investigation of non-fuel applications (one option may be to recover the high value liquids such as the unsaturated hydrocarbon and/or naphthenic components) may ultimately result in better overall process economics.

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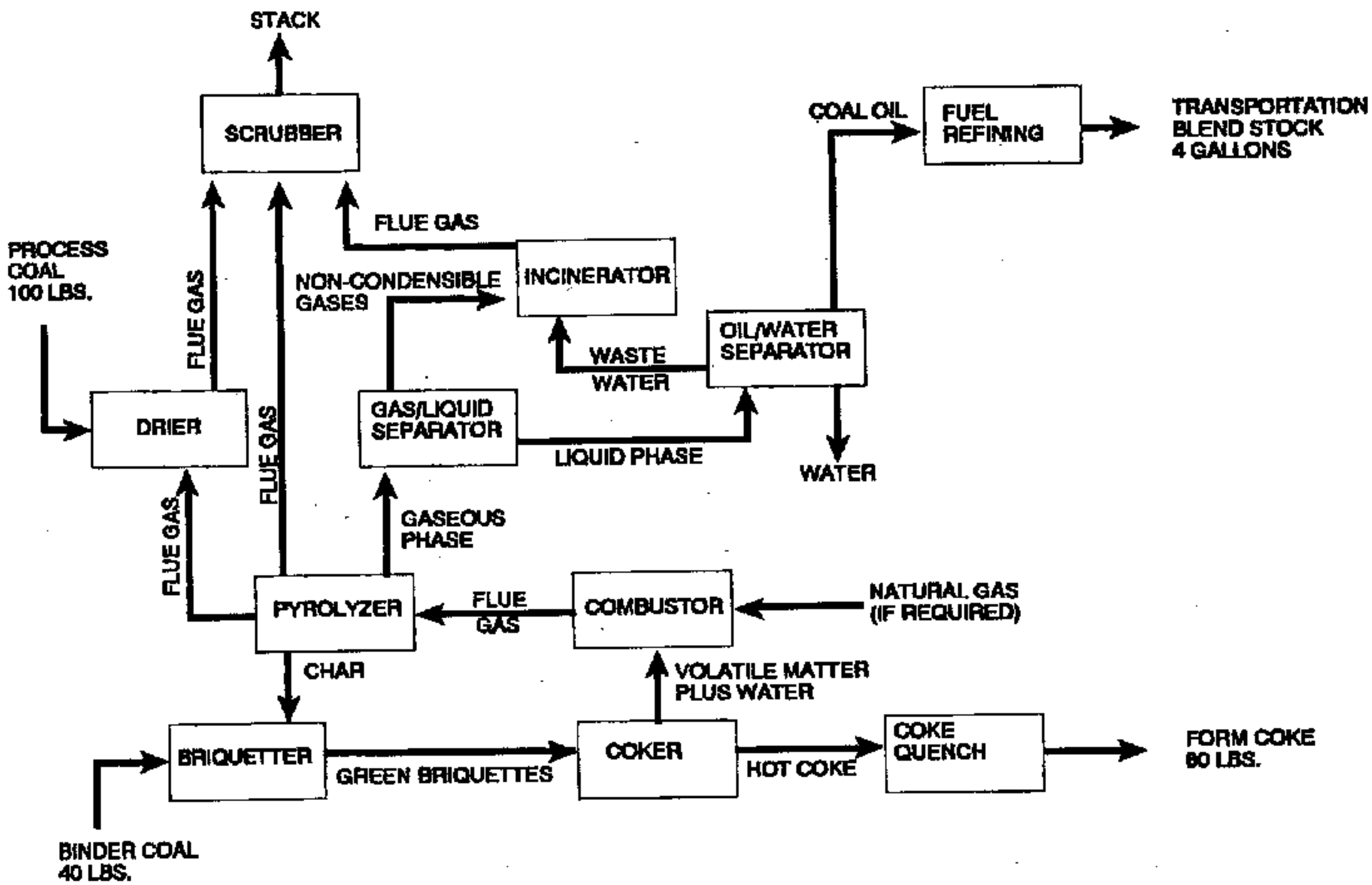

FIGURE 1: Overall Block Diagram of the Coal Liquid and Coke Mild Gasification Concept

ENCOAL MILD GASIFICATION

Introduction

In the ENCOAL process, coal is treated by exposing it to moderate temperatures and near atmospheric pressure. A low-sulfur solid with a high heating value (called Process Derived Fuel or PDF) and a low-sulfur, heavy-hydrocarbon liquid (a Coal Derived Liquid or CDL) are produced. Both of these products are marketable.

This concept was proposed (and subsequently selected by DOE) in the third Clean Coal Technology solicitation by the ENCOAL Corporation, a wholly-owned subsidiary of the Shell Mining Company of Houston, Texas. An integral part of the ENCOAL concept is the Liquids From Coal (LFC) technology developed by Shell Mining Company and SGI International of La Jolla, California [1, 2].

Process Description

The ENCOAL process uses moderate temperatures (about 1,000°F) and low pressures (about atmospheric) to devolatilize coal into solid and liquid products. A simplified flow diagram for this process is given in Figure 1. Run-of-mine coal is screened to yield a 2-inch by ⅛-inch feed which is sent to a rotary grate dryer where it is heated and dried by a hot gas stream. The temperature of the inlet gas and its residence time in the dryer were selected so as to dry the coal without initiating significant chemical changes in doing so. The temperature of the coal is controlled so that the coal does not produce significant quantities of methane, carbon monoxide, or carbon dioxide during the drying process. Solids from the dryer are then sent to the pyrolyzer where they are heated by a hot gas recycle stream to about 1,000°F. In the pyrolyzer, the feed coal is devolatilized and partially decomposed to create the two fuel forms. The remaining free water is removed from the coal along with some of the sulfur, depending on the form of the sulfur in the coal feed stock. A control system has been developed to optimize the product specifications based on market needs and the composition of the feed stock. The gas from the pyrolyzer goes to a cyclone for removal of particulates and is then cooled to stop further pyrolysis reactions and to condense the desired coal liquids. Conditions in the condenser are controlled so as to condense only the coal liquids and not the water contained in the pyrolysis gas.

Most of the uncondensed gases exiting the condenser are recycled directly to the pyrolyzer. A portion of these gases are burned in a pyrolyzer combustor prior to being blended with the recycle gases. In this way, the temperature of the recycle gases entering the pyrolyzer is maintained at the desired 1,000°F. The remaining uncondensed gas is sent to the dryer combustor where it is burned and blended with the dryer recycle gas to control the dryer temperature at the desired level.

Off-gases from the dryer are treated in a wet scrubber for removal of particulates and in a horizontal scrubber for removal of sulfur oxides. The cleaned gas is discharged through a stack while the spent solution from the horizontal scrubber is sent to an evaporation pond.

Solids from the pyrolyzer are quenched to stop the pyrolysis reactions. They are cooled further and sent to temporary storage. A dust suppressant is then added to these solids prior to transport to the end-user.

Coal Feeds

To date, most of the work on the ENCOAL process has been with a Powder River Basin sub-bituminous coals. Process design details, product yields, and economics will be dependent on the coal feedstock but these factors have not yet been evaluated in detail. Table 1 shows analyses of Buckskin Mine coal feed and resulting PDF.

Products

The marketable products from the ENCOAL process includes both a solid and a liquid. Some low-Btu gases are used internal to the process to provide heat at various points. The ENCOAL solids account for approximately 50 percent of the original mass feed and are purported to be of lower sulfur content and higher heating value than the original feedstock due to the chemical changes that occur during the pyrolysis process. The solid fuel is a stable fuel similar in composition and handling properties to eastern bituminous coals. As shown in Table 1, the PDF has an energy content of about 12,000 Btu/lb and has retained sufficient volatiles to allow its use as a boiler fuel. The liquid product is similar to a low-sulfur No. 6 fuel oil. The liquid yield is approximately one-half barrel per ton of coal feed depending on the properties of the coal feed. Current plans call for selling the liquid product for burning rather than upgrading it into higher valued liquid products. Table 2 provides a comparison of CDL to No. 6 low- and high-sulfur fuel oil. Overall yields of approximately 60 percent by weight and 90 percent by heating value have been claimed for this concept [3, 4].

Likely Applications

The solid and liquid fuel produced in the ENCOAL concept could be used in both commercial and electric utility applications. The liquids could be used to displace petroleum-derived No. 6 fuel oil, while the solid fuel could be used to aid in complying with emission control requirements and to provide a more uniform fuel for use in boilers or furnaces.

Several commercial entities could potentially be interested in owning and operating a coal refinery using the ENCOAL process. Mine owners could use it to add value to their fuel and to expand their markets. Electric utilities could use both the liquid and solid fuels for economic operation and environmental compliance reasons. A commercial fuel supplier could potentially be interested for the purpose of providing fuels to its customers. As with any process, the economics of the ENCOAL process and those of its alternatives will dictate the ultimate applications of this refinery concept.

Due to the low liquid yield, commercial operations will focus on simply marketing the CDL product as a premium industrial fuel and directly displacing currently used petroleum-derived No. 6 fuel oils. It is expected that no further upgrading is required to compete in this market.

Status of Development

The mild gasification process used in the ENCOAL Project was originally developed by SGI International of La Jolla, California and the Shell Mining Company in the mid-1980s. Development included establishment of process conditions and product yield/quality profiles through step-wise scale-up of equipment to an eventual 2.5 tons per day PDU scale. This development identified the acceptability of many commercially available equipment items which have been used extensively in allied industries like coking and kilning, where the equipment is currently operational at scales much larger than the 1,000 tons per day size of the Clean Coal Technology demonstration plant. Data acquired from the PDU established the critical process conditions from which control and operating parameters were obtained, and thus permitted design of the much larger demonstration plant. Since the temperature and pressure conditions are significantly milder and less severe to equipment than in similar coking operations, the scale-up design factor of approximately 350 is not considered unreasonable from economic and technical points of view. Operation of a fully integrated plant during 1992-94 is the primary technical objective of the Clean Coal project, which will provide sufficient data for the parallel design of a still larger facility. Construction of this much larger plant (which would process approximately 20,000 tons of coal per day) is planned for the mid-1990s by ENCOAL, and this commercial-scale plant will more fully establish the life of all components and economics of the overall process.

Environmental Aspects

Sulfur in the coal will be released during the devolatilization process and will become part of the process gas stream sent to the condenser. While some of this sulfur may be contained with the CDL, most of it will remain in gaseous form. It will be converted to sulfur dioxide in the pyrolyzer and dryer combustors to be ultimately removed in the horizontal scrubber using a water-based sodium carbonate solution. The spent solution from this scrubber is discharged to a pond where it is evaporated.

Nitrogen oxides are created during the burning of the gases in the pyrolyzer and dryer combustors. Controlled temperatures and special burner designs will be used to control the formation of nitrogen oxides in these units.

There are no reported waste water or toxic solid wastes generated in this process. An environmental assessment was conducted for the Clean Coal demonstration project, and a finding of 'no significant impact' was reached.

Research Needs

Commerciallization of the ENCOAL process will be aided by continued operation of the Clean Coal demonstration plant as an entry-level commercial facility. Parallel and ongoing research by the industrial participants on the impacts of coal type on the design, operation, and yields of the ENCOAL process will establish the full potential of the concept.

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Table 1: PDF Quality Profile

	Buckskin Mine Coal Feed	
	Coal	PDF
Heating Value, Btu/lb	8,370	11,900
SO_2 potential, lb/MM Btu	1.2	1.1
Moisture, weight %	30	4
Ash, weight %	5	10
Volatiles, weight %	29	19 *
Fixed Carbon, weight %	36	67
Sulfur, weight %	0.50	0.49
Ash Fusion Temp., °F	2,220	2,220

* Variable, Controllable by Process

Table 2: CDL Quality Profile

	Petroleum-Derived Fuel Oil		
	No. 6 Low Sulfur	No. 6 High Sulfur	CDL
API Gravity	4-6°	4-6°	6-8°
Sulfur, weight %	0.8	4.0	0.3
Nitrogen, weight %	0.3	0.2	0.6
Oxygen, weight %	0.6	0.4	6.2
Pour Point, °F	50	50	96
Viscosity @ 122°F, cs	200-635	200-635	88.1

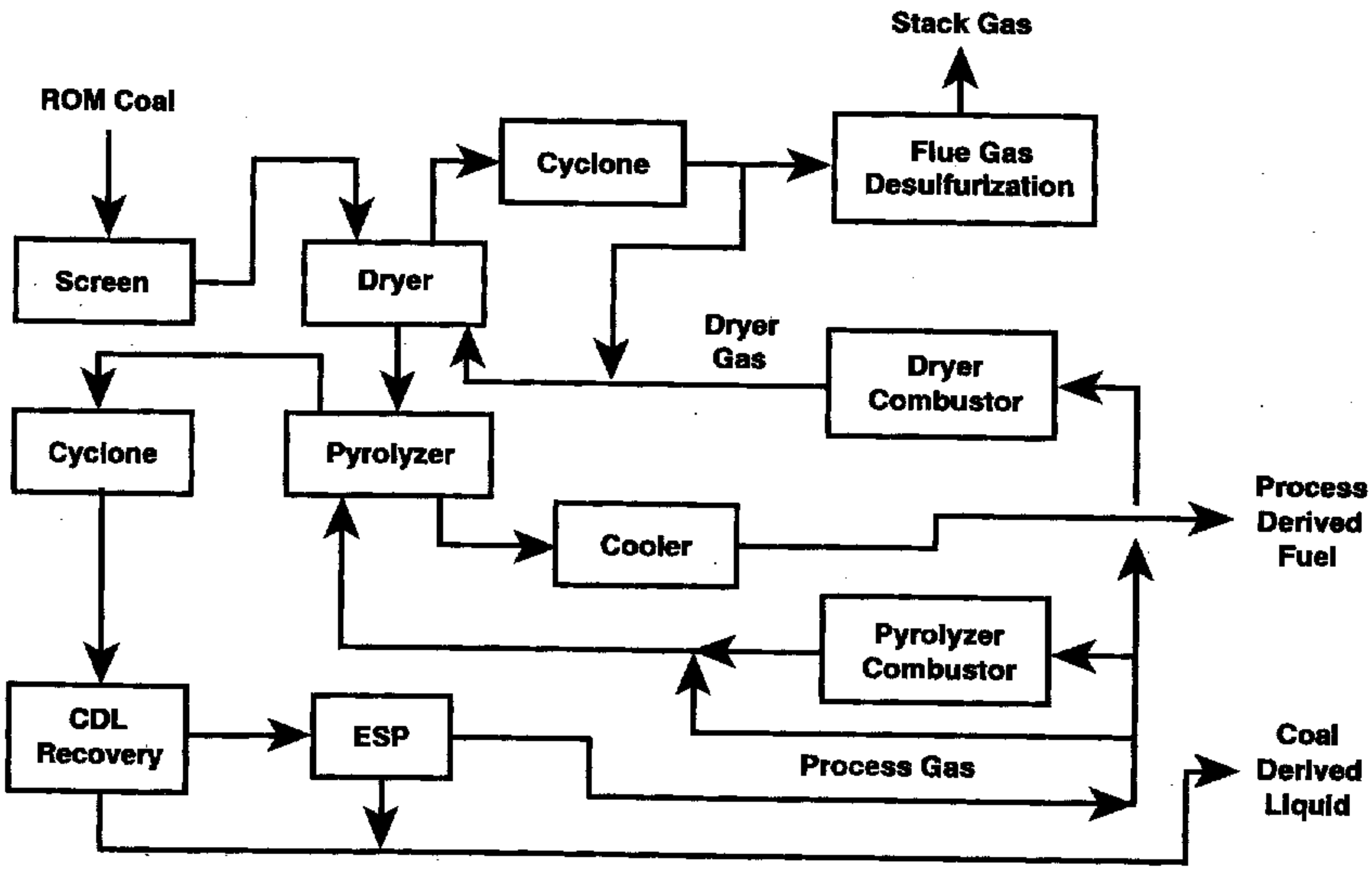

FIGURE 1: Overall Block Diagram of the ENCOAL Concept

HYDROCARB

Introduction

The Hydrocarb concept has two principal steps; the first is the hydropyrolysis of coal to produce methane and the second step involves the decomposition of the methane to carbon and hydrogen. Some of the hydrogen is recycled to the hydropyrolysis reactor. A potential third step converts the normally small amount of carbon monoxide in the outlet pyrolysis stream and hydrogen to methanol. The main products are carbon, hydrogen and some methane. The carbon is the main energy bearing material and is in a particulate form similar to a pure carbon black. All coals and carbonaceous materials including biomass, MSW, sewage sludge, tar and char can be used as the input feedstock material [1, 2, 3, 4]. The process involves high temperatures (800°C to 1,150°C) and moderate pressure (about 50 atmospheres). The process described herein omits the third methanol producing step.

The process has undergone analysis and economic evaluation. Carbon black can be slurried with fluids such as water, alcohols, or hydrocarbons to produce a fuel material for power production, for heat engines (turbines and diesels) [1], for transportation, and for other commercial commodity uses as carbon black [3]. Brookhaven National Laboratory has been the site of initial developmental efforts for this coal refinery concept.

Process Description

The goal for this coal refinery concept is to produce carbon and hydrogen from coal in an energy efficient process. A simplified process flow schematic is given in Fig. 1. The analysis of this process is based on a large facility using 25,000 tons per day of coal, which on an energy basis, is approximately equivalent to a 100,000 barrel day petroleum refinery [3, 4].

As shown in the schematic, the coal is prepared for the hydropyrolysis reactor by heating to partially dry it. The heated coal is then fed to the hydropyrolysis reactor. In this vessel, the coal is hyropyrolyzed in the presence of hydrogen to produce methane. Limestone is also fed into the vessel to react with the hydrogen sulfide and fix the sulfur as calcium sulfide. Circulating aluminum oxide (Al_2O_3) balls at 800°C, are used to establish a temperature level and to transport the major portion of the heat of reaction by their 300°C temperature rise in a moving bed reactor. The mass flow of the $A1_2O_3$ is about twenty times that of the coal. The product, or "make," gas is approximately 50 percent by volume methane, a large amount of hydrogen, and up to equilibrium amounts of H_2O, CO, CO_2, N_2, and NH_3. The make gas, after being reduced in temperature in a regenerative cooler, is sent to a partial condenser. The purpose of this condenser is to further cool the gas so that some of the water vapor will condense. The dried gas is pumped through a heater to the second major processing vessel, the methane pyrolysis reactor.

The pyrolysis reactor thermally breaks down a large fraction of the methane into carbon and hydrogen. The gas temperature at the inlet is 750°C and is 1,100°C at the outlet.

This pyrolysis reaction is endothermic with the heat being supplied by Al_2O_3 balls that exit the hydropyrolysis reactor at 1,100°C and are heated to 1,150°C after being separated from the ash. These balls leave the reactor with the carbon at 800°C. The carbon (0.1 to 3 micron in size) is separated from the balls and fed to the carboline-fuel-making operations. This carbon-black-like material is then mixed with a carrier fluid (e.g. water, methanol, or a combination of these) to produce the energy fluid desired.

The carbon, when mixed with 40 percent water, has a heat value of about 96,000 Btu per gallon [1]. Higher volumetric heating values can be obtained by mixing the carbon with methanol. A value of 140,000 Btu per gallon can be formulated, which is about the same as diesel oil. A stabilizing material is used in these mixtures [3].

The ash, CaS, and CaO are removed from the system when the accompanying Al_2O_3 balls are cleaned after leaving the hydropyrolysis reactor. This separation and cleaning is carried out at 1,100°C and about 50 atmospheres pressure.

Coal Feed or Other Feedstocks

It is reported that the starting carbonaceous material can include all grades of coal (lignite, bituminous, anthracite) as well as other energy source materials such as tars, heavy residual oils, oil shale, biomass (wood), MSW etc. [1, 2, 3, 4]. Adjustments in process flows and equipment are needed depending on the composition of various feedstocks.

Products

Pure carbon in the micron size range is the main product. This product can be used in a manner similar to pulverized coal or in briquetted form for use in stationary power furnaces or in slurry form mixed with water or methanol. When the carbon is combined with methanol, the mixture has a volumetric heat content similar to gasoline or diesel oil and might be useable as a fuel in heat engines, turbines, and diesels [3].

The high-Btu purge gas, which is mainly hydrogen and methane could be used for heating or production of electric power. With the impurities separated from this purge gas, a quality hydrogen gas could be provided to industry.

Likely Applications

The siting for a conceptual coal refinery based on the Hydrocarb process is envisioned to be located at a coal mine-mouth with the product clean carbon fuel and slurry coproducts distributed to the utility and transportation sectors. The ash, sulfur and other impurities remain at the mine site. The fuel users can burn the clean fuel products in a manner similar to oil and gas without the need for ash disposal or stack gas cleaning. This process would most likely be commercially used by large energy concerns such as coal or chemical companies.

Status of Development

The conceptual process has been designed and analyzed. More information is needed on the supporting systems. Laboratory process data are available [2]. Preliminary combustion tests of carbon and slurry fuel have been carried out [2, 4].

Environmental Aspects

The process produces clean fuels. During process operation, ash and other wastes are produced and must be disposed of. Care in the disposal of the CaS must be used and the CaS will have to be oxidized to produce a stable sulfate. In general, environmental aspects are not expected to cause a concern.

Research Needs

Economic assessment of the potential for this concept is required. A PDU could help to determine the operating and design parameters that are optimum for this process. Combustion and engine tests with the Hydrocarb fuels are required to define whether any changes are needed to use this fuel in conventional combustion and engine equipment, and to estimate the value of these fuels in such equipment. Such efforts are not expected to be large in comparison with the development of the coal refinery concept itself.

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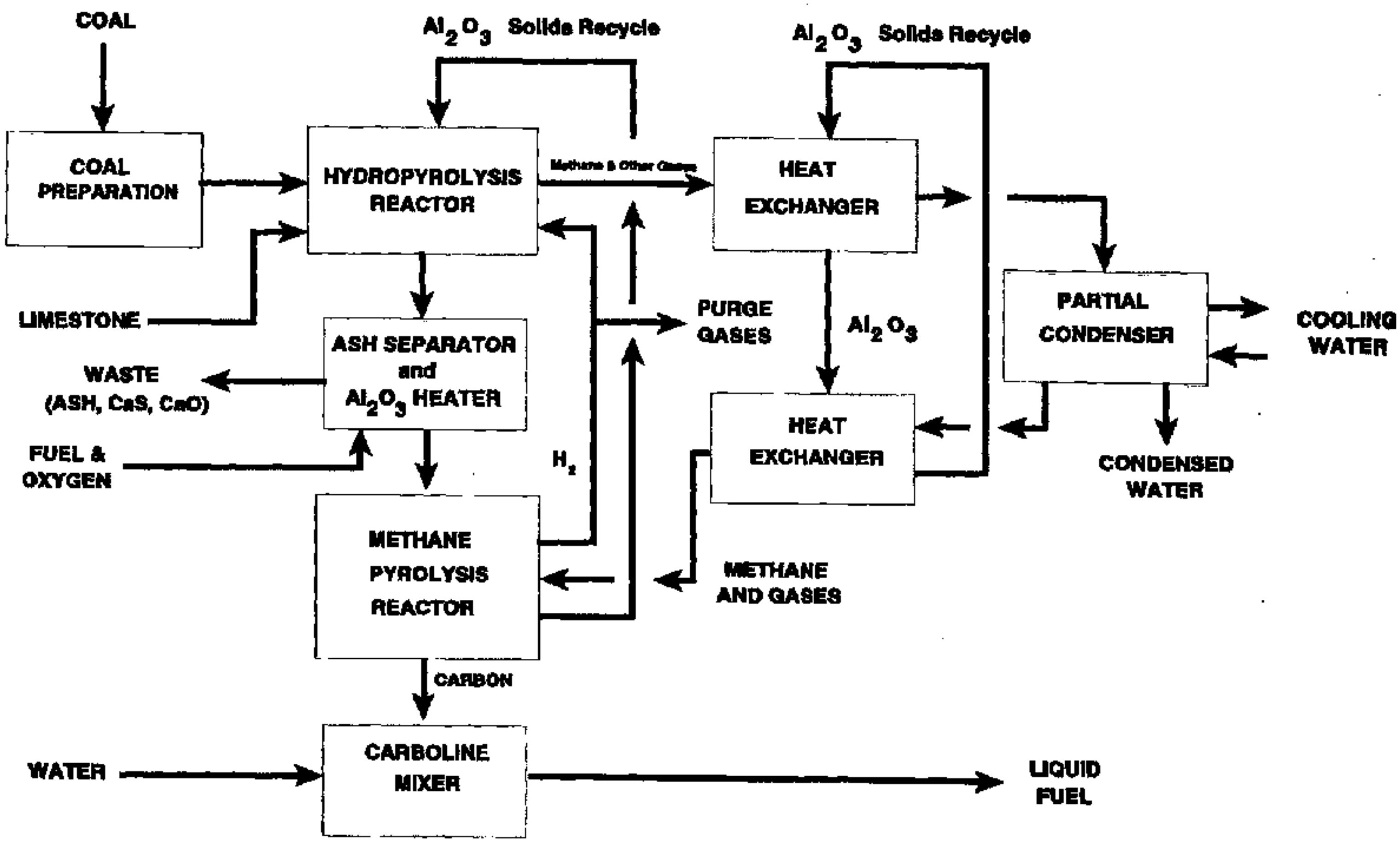

FIGURE 1: Overall Block Diagram of the Hydrocarb Concept

IGT MILD GASIFICATION

Introduction

The Institute of Gas Technology (IGT) Mild Gasification (IMG) concept for production of form coke and coal liquids is an example of a coal refinery in the Devolatilization category. The concept is capable of processing both eastern caking and western non-caking coals, and is designed to offer options in the product slate by varying the process conditions and by blending different feed coals. Depending on the feed coal characteristics and the operating conditions, the char can be used as an improved fuel for power generation or hot-briquetted to make form coke for steel-making blast furnaces or for foundry cupola operations. The hot briquetting process offers options for blending various chars, coals and other additives (like alloying agents) to tailor the properties of the form coke. The liquids, which can be processed as feedstocks for chemicals (e.g., BTX, phenol, cresols, xylenols, naphthalene, indene), pitch for use as an electrode binder in the aluminum industry, and fuels. The mild gasification and briquetting processes are done entirely within closed vessels which offer significant advantages over conventional coking practices for control of fugitive emissions. The IMG project team, consisting of Kerr-McGee Coal Corporation (KMCC), IGT, Southern Illinois University at Carbondale (SIUC), Bechtel Group, Inc., and the Illinois Department of Energy and Natural Resources, is negotiating award of a cost-shared DOE contract to construct and operate a 24 tons per day mild gasification PDU.

Under an earlier DOE contract, a project team consisting of IGT, Peabody Holding Company, Inc., and Bechtel Group, Inc. completed a literature and market survey [1] which formed the basis for the selection of the IGT mild gasification reactor configuration and projected co-product slate; designed, constructed, and operated a 100 pounds per hour Process Research Unit (PRU) to validate the design and obtain a data base with bituminous coals [2]; completed a char upgrading study [3]; conducted system integration studies that included PRU operation with full-stream condensate recovery; and completed the conceptual design of a 24 tons per day PDU [4]. Studies also included PRU tests with a subbituminous coal.

Process Description

The IMG reactor consists of coaxial fluidized- and entrained-bed reaction stages. The coarse coal fraction is fed into the fluidized bed with a gas distributor designed to promote rapid mixing and dispersal of the incoming coal throughout the bed of char. The fines fraction of the coal (-60 mesh) is fed to the entrained section, with lower residence time sufficient for conversion of the smaller particles. Figure 1 shows a block flow diagram of the process as configured for the 24 tons per day PDU to be constructed and operated at SIUC. In a commercial process, the incineration/scrubbing unit would be replaced by an optimized gas cleanup system, and product gases in excess of that required for heating fluidization gases would be utilized as an energy source for other plant requirements.

The feed coal is dried and preheated with hot flue gases, then conveyed by entrainment to a classifier, where the coarse and fine fractions are separated and fed separately to the two sections of the mild gasifier. The coal is dried to about 8 percent to 12 percent moisture and preheated to 500° F, just below the point where caking coals begin to exhibit fluidity and stickiness. The fluidized bed consists of devolatilized char, including partially combusted char which is recycled into the reactor for heat input. Fluidization is provided by partially combusted product gases, which provide a neutral or slightly reducing atmosphere in the reactor. Spent char is discharged from a bed overflow, and char fines are removed from the overhead product gas by sequential cyclones and, optionally, a hot gas filter. The solids-free hot product gas stream passes through a gas-fired thermocracker in order to upgrade the properties of the oils/tars in the vapor phase. Oils/tars are then condensed by sequential cooling in a series of spray towers to recover heavy oil/pitch, middle oil, and light oil. Water, primarily from coal devolatilization, is separated and recycled to the gasifier for incineration of contaminants.

In the preferred process scheme which upgrades the mild gasification char to metallurgical form coke, the hot (1,000°F) char is blended with raw caking coal in a specially designed heater/mixer. An *in-situ* binder is produced by the softening of the raw coal, and volatiles from this step are removed prior to hot briquetting of the fluid mixture in a roll press. The "green" briquettes are then fed to a vertical shaft furnace where they are calcined at about 1,800°F to develop the proper strength and reactivity properties. The volatiles from the heating, mixing, and calcination steps are processed along with the volatiles from mild gasification to recover oils/tars and fuel gases.

Coal Feeds

Four coals were tested in the mild gasification PRU. The proximate and ultimate analyses of samples of the four coals are shown in Table 1.

The Illinois No. 6 coal was from the high-moisture fines (less than ⅛-inch) stream of the Peabody Coal Company Randolph Preparation Plant, and is a typical high-sulfur, high-ash Illinois coal used primarily as utility fuel. The West Virginia coal is also from the high-moisture fines stream of a Peabody Preparation plant at the Wells Complex in Boone County near Wharton, West Virginia. The West Virginia coal is a more highly caking coal (free swelling index 5 to 8) than the Illinois No. 6, and is typical of Eastern medium-sulfur (about 1 percent) low-ash (about 6 percent) coals. The Rochelle subbituminous coal is a low-sulfur utility coal from the Powder River Basin, and is fairly typical of Western coals, having very low sulfur (0.14 percent), low ash (5.5 percent), and a high oxygen content (about 20 percent). The moisture content of this coal is also typically high (about 16 percent). The second Illinois No. 6 coal is an atypical low-sulfur, low-ash variety from a mine in Sesser, Illinois, which is used for coke blending. The moderate caking properties of the coal are typical of Illinois No. 6 bituminous coals.

The results of tests with the Randolph Plant Illinois No. 6 coal were used as a basis for the design of the 24 tons per day PDU, which will primarily test Illinois and West Virginia bituminous coals from KMCC mines.

Table 1. ANALYSES OF COALS TESTED IN IGT MILD GASIFICATION PRU

	Ill No. 6 (Randolph)	West Va (Wells)	Wyoming (Rochelle)	Ill No. 6 (Sesser)
Proximate	----- Weight percent as fed to reactor -----			
Moisture	5.3	1.4	16.2	6.6
Volatile Matter	34.7	30.6	35.9	31.6
Ash	15.0	6.4	4.6	4.2
Fixed Carbon	45.0	61.6	43.3	57.5
Total	100.0	100.0	100.0	100.0
Ultimate	--------- Weight percent dry basis ---------			
Ash	15.8	6.4	5.5	4.5
Carbon	64.3	80.2	68.3	78.2
Hydrogen	4.2	5.0	4.2	5.0
Nitrogen	1.4	1.7	1.1	2.0
Sulfur	3.9	1.3	0.14	1.1
Oxygen (by difference)	10.4	5.4	20.8	9.3
Total	100.0	100.0	100.0	100.0

Products

The criteria for co-product selection were identified in the market survey [1]. Table 2 shows the yield distribution of the raw product streams for three of the four coals tested in the PRU [2]. The PRU tests also showed the temperature dependency of the yield structure with the bituminous coals, with char yield decreasing monotonically, oils/tars yields reaching a maximum at about 1,100°F, and gas yield increasing monotonically with increasing temperature over the ranges studied. Table 3 shows typical compositions of feed coal, char, and oils/tars from Illinois No. 6 coal subjected to mild gasification at 1,183°F. The data show the degree of desulfurization of the products which typically takes place in mild gasification.

Because char is the major co-product, comprising 54 percent to 76 percent of the total co-product stream, its beneficial utilization is critical to commercialization of IMG technology. Char upgrading studies were completed under the PRU project, and the results were reported in the *Char Upgrading Studies* Topical Report [3].

Table 2. CO-PRODUCT YIELDS FROM IGT MILD GASIFICATION PRU

	Illinois No. 6 Bituminous	West Virginia Bituminous	Wyoming Subbituminous
Temperature, °F	1,035 - 1,390	1,049 - 1,149	1,062
Co-Product Yields, weight percent dry coal			
Char	54.3 - 66.7	65.7 - 76.0	66.2
Oils/Tars	13.5 - 28.7	18.0 - 29.4	13.2
Gas	6.7 - 19.4	5.2 - 7.8	12.9

Table 3. TYPICAL COAL, CHAR, AND OILS/TARS ANALYSES (Test MG-16, 1,183°F)

Elemental Analysis	Feed Coal	Char	Oils/Tars
Carbon	63.86	68.73	74.24
Hydrogen	3.99	0.58	6.74
Nitrogen	1.33	1.15	0.78
Sulfur	3.60	2.11	1.60
Oxygen (by difference)	11.07	2.42	16.23
Ash	16.15	25.01	0.42
	100.00	100.00	100.00

Consultations with an Industrial Project Advisory Group (IPAG), consisting of representatives of steel companies, coke suppliers, tar processors, foundries, and other potential consumers of co-products, led to the conclusion that the use of mild gasification char to make form coke for blast furnaces and foundries should be given the highest priority. The total potential market for this end use is about 29 million tons per year. In order of decreasing priority, the other potential char markets are: (1) production of smokeless fuel as an alternative to firewood for domestic heating, and (2) steam activation of char for industrial water treatment applications (1 to 2 million tons per year).

A form coke co-product from the IMG char has two sub-markets. The larger of the two markets for form coke is in blast furnace production of iron, with a current annual consumption of about 27 million tons of coke per year. A smaller market of about 1.8 million tons per year of foundry coke is used in cupolas for re-melting and alloying iron for castings. The cost of coke from various suppliers is $90 to $150 per ton today, so a suitable form coke from IMG would present an excellent value-added product. In addition, the IMG process offers continuous form coke production with superior environmental control that is difficult and costly to achieve in present coke oven batteries. The existing coking plants in the United States are reaching the limits of useful age, and environmentally acceptable methods to produce supplementary supplies of coke rapidly are urgently needed. An assured domestic supply of form coke from a continuous, environmentally safe process would have significant benefits for the steel and coal companies and the nation.

Form coke made from IMG char can meet the requirements for coke properties. In general, coke needs a strength sufficient to support the burden in the blast furnace, and also has to provide a certain bulk porosity for gas, liquid metal, and flux flows. In addition to these properties, the coke must also meet a reactivity criterion based on the reaction of carbon with carbon dioxide, and its sulfur and ash contents should be low. Blast furnace coke is usually produced in the coke oven by selectively blending several coking coals, usually a high-volatile and a low-volatile coal, to make a strong structure with a desired reactivity. IMG chars can be produced and blended in a similar manner with better control of the process conditions and emissions than attainable in coke ovens.

Mild gasification oils/tars, which comprise 13 percent to 29 percent of the co-products, can also be upgraded into value-added products. Different fractions of the condensable hydrocarbons can be used as binders for electrodes in the steel and aluminum industries (market size 435,000 tons per year), in roofing and road paving industries (330,000 tons per year), as pitch coke for electrodes (155,000 tons per year), as chemical feedstocks used for production of plastics, paints, adhesives, dye intermediates, insecticides, surfactants, etc. (at least 15 million tons per year), and as fuel for peaking turbines (0.9 million tons per year). The consensus of IPAG members is that pitch-based products -- binders and pitch coke -- should be the highest priority targeted products from the oils/tars, followed by chemical feedstocks and fuel uses. It is apparent from the PRU data that the IMG pitch requires post-treatment to increase aromaticity and remove heteroatoms in order to be acceptable for electrode binders. On the basis of PRU test results and consultation with IPAG members, the IMG system design now includes a on-stream precondensation thermal upgrading step, and the liquids recovery system was designed to sequentially condense oils/tars in decreasing boiling-range fractions to maximize the QI content of the pitch, thus reducing the off-stream upgrading requirements for meeting electrode binder pitch specifications. These steps have been integrated into the mild gasification PDU to be constructed at SIUC. Post-condensation heat-treating and catalytic dehydrogenation methods are also applicable to upgrade a high-temperature pitch from mild gasification, if necessary.

Until the advent of abundant cheap petroleum, coal was the primary source of many chemicals for industry. Yields of BTX and phenols are higher from IMG than from coke ovens. These chemical feedstocks are widely used as starting materials to ultimately make plastics, synthetic fibers, and building materials. Some of the major products include phenolic resins, nylons, polycarbonates, polyesters, and plasticizers for PVC. The markets for these chemicals are almost entirely dominated by petrochemicals, however. Some chemicals, like naphthalene, are still produced in significant amounts from coal tar. Naphthalene is an alternative for *o*-xylene in the manufacture of phthalic anhydride, which is a feedstock for polyester production. Other coal-based chemicals of interest are indane and indene, which are valuable feedstocks for manufacture of specialty polymers. The economics of separation technology and current market conditions will be the dominant factor in determining the desirability of recovering these chemicals for revenue rather than selling mixed fractions to tar processors at lower prices.

IMG liquids, or some fraction thereof, also have potential for being converted into transportation fuels. The middle to heavy distillates, covering the boiling range of 390 to 650°F, can be used as a diesel fuel blending stock, although upgrading is necessary to remove sulfur, nitrogen, and oxygen, both for reducing emissions and for stabilizing the fuel against undesirable degradation reactions during storage. For example, the diesel-oil fractions from the Coalite process which have an acceptable cetane number have been used to fuel city buses in Bolsover, England [1]. Another fuel application for IMG liquids with minimal upgrading would be in gas turbines, which have less stringent operating requirements than internal combustion engines. Turbines may use many types of fuel. The primary limitation on the use of any fuel in a gas turbine is based on emissions, which can be predicted in a straightforward way from the fuel properties.

Based on these factors, a potential market for middle- to heavy-oil fractions could be power-generation peaking turbines. Those installations which can tolerate a low-grade fuel such as the IMG liquids, however, would not command a premium price, and would at best supply an outlet for liquid fractions that are not used for producing value-added products.

The 5 to 19 percent yield of fuel gas co-product, after the addition of combustion gases for part of the reactor heat and fluidization gas supply, is a low- to medium-Btu gas. The current projected mass and energy balances suggest that in-plant utilization of the entire fuel gases for energy needs will be the primary end use. Depending on plant siting, it is possible that some surplus fuel gas may be used for power generation or steam production for sale.

Likely Applications

The most likely IMG applications are led by the char products which, according to the market assessment, are:

- Iron-making blast furnace form coke,
- Iron-melting/casting foundry form coke,
- Smokeless fuel for industrial use or for export, and
- Char for fuel or gasification.

The most probable IMG location will be at a coal preparation plant to take advantage of excess energy from IMG for additional coal drying at the prep plant. Other advantages of siting an IMG installation at a coal prep plant are the in-place transportation infrastructure and nearby locations of coal tar processors.

Status of Development

IGT has built and operated a coaxial 8-inch/4-inch-ID isothermal PRU at its Energy Development Center in Chicago, Illinois. The PRU was operated successfully for 26 steady-state tests, including closed material balances, with three bituminous coals and one subbituminous coal [2, 4].

In the course of PRU testing and data analysis, two key items were discovered. First, it was found that the relationship of incoming coal feed rate to the mass inventory of char in the fluidized bed was critical in controlling agglomeration with caking coals using a single fluidized bed. Secondly, it was found that a minor portion of the char product, reheated by partial combustion, could supply adequate process heat for the fluidized bed. The continuous processing of caking coals in a single reactor has been the major achievement of the PRU test program.

The PRU has produced char and liquids for bench-scale evaluation. The char has been processed into form coke briquettes on a small scale, and these were tested for strength, density, and reactivity. The liquids were evaluated for fractionation into fuels and chemical feedstocks. The results of these product evaluations have been used to update the design of a 24 tons per day PDU to be constructed at SIUC under a cost-sharing agreement with DOE.

The overall strategy for commercialization of the proposed IMG concept consists of stepwise scale-up to a demonstration plant. The IMG PDU scheduled for construction at SIUC is an adiabatic system which will provide data necessary for process scale-up that is not currently available from the existing isothermal PRU. A block flow diagram of the PDU system was shown in Figure 1. The PDU can also supply larger batches of co-products, approximately 15 tons per day of char and 22 barrels per day of condensable hydrocarbons, to permit the extensive larger scale testing of these co-products for value-added uses. The 24 tons per day unit will feature the flexibility to handle all types of coals and evaluate many possible end uses for the char and condensables. Char from the PDU will be evaluated for its performance in manufacture of form coke and as a utility fuel. Based on the process and co-products test data and evaluations thus obtained, a revised market survey will be done, which should further improve the reliability of projections for introducing the co-products into the marketplace.

With the successful completion of the PDU, it is the project team's intention to seek support within the Clean Coal Technology program to utilize the experience from the PDU to install a "first-of-a-kind" commercial demonstration plant of 500 tons per day coal capacity. This plant size represents half the capacity of a single-train commercial scale gasifier envisioned in a three-train, one million tons per year plant. Successful operation of the 500 tons per day plant will present the developer an acceptable level of risk for building the first large commercial plant.

One candidate location for the demonstration plant is the Southern Illinois University at Carbondale (SIUC) campus. This site is about 50 miles from KMCC's Galatia mine, which would be the source of the test coals. Coal from the preparation plant could be transported by trucks to the SIUC facility. During the initial operation of the demonstration plant, the product char would be burned in a closely coupled circulating fluidized-bed (CFB) boiler to generate steam and electricity for the campus. The condensable hydrocarbons would be transported to the St. Louis metropolitan area where markets for the co-products already exist. The coal tar processing facilities of Reilly Industries located at Granite City, Illinois, is a prospective candidate to receive the oils and tars and test their processing into marketable products. Fine coal from West Virginia mines would also be tested in the 500 tons per day plant to evaluate the form coke option. The CFB boiler would also be designed to burn coal, so that the char from the mild gasifier could be utilized to produce sufficient quantities of form-coke for testing by the steel industry. Initially, the CFB boiler would be fueled by IMG char. During the latter stages of the test, the boiler would be fueled by coal and the gasifier char trucked to an off site location for briquetting and form coke preparation. Quantities of form coke would be accumulated for a full-scale blast furnace test.

Environmental Aspects

One of the major advantages of mild gasification for the production of form coke in this country is the environmental advantages compared to conventional slot-oven coking operations. Mild gasification takes place in a continuous closed system, thus minimizing fugitive emissions associated with batch operations. It is also compatible with state-of-the-art gas cleaning technology to maintain SO_2 and NO_x emissions well below allowable limits. Wastewater emissions are expected to be very small because the process uses no steam and the only water generated is that from coal drying and devolatilization. The small amount of wastewater recovered from condensable streams will be recycled to the gasifier to further reduce contaminants, and the net wastewater can be treated by conventional means prior to discharge. There are no other waste streams expected from the process.

Research Needs

The primary thrust of future research should be directed towards co-product upgrading and industrial testing. The 24 tons per day PDU will produce large quantities of char, form coke, and oils/tars for this purpose. Form coke will be evaluated by conventional testing of physicochemical properties. For more direct evaluation, form coke will be tested by General Motors in a 60-inch Pellet Technology Corporation cupola. Oils/tars from the PDU, produced with and without operating the on-stream thermo-cracker, will be evaluated and processed by Reilly Industries.

In addition to these planned activities in the PDU program, the IMG process could further benefit from research in the following specific areas:

- Desulfurization of char and/or briquettes made therefrom, in order to extend the resource base for form coke to include higher-sulfur coals; funding has been solicited by IGT from the State of Illinois for one such study using process-derived gases.

- Formulation of foundry coke briquettes containing alloying additives, concentrating on the effects of such additives on the strength and reactivity properties.

- Alternative methods of upgrading oils/tars, either on- or off-stream, to produce higher-value products; the emphasis should be on producing high-quality binder pitch with high aromaticity, low sulfur, and favorable physical properties.

- Separation technologies to recover high-value chemicals from mild gasification oils/tars, including advanced processing schemes combining fractional distillation with other technologies such as solvent extraction or solid adsorption.

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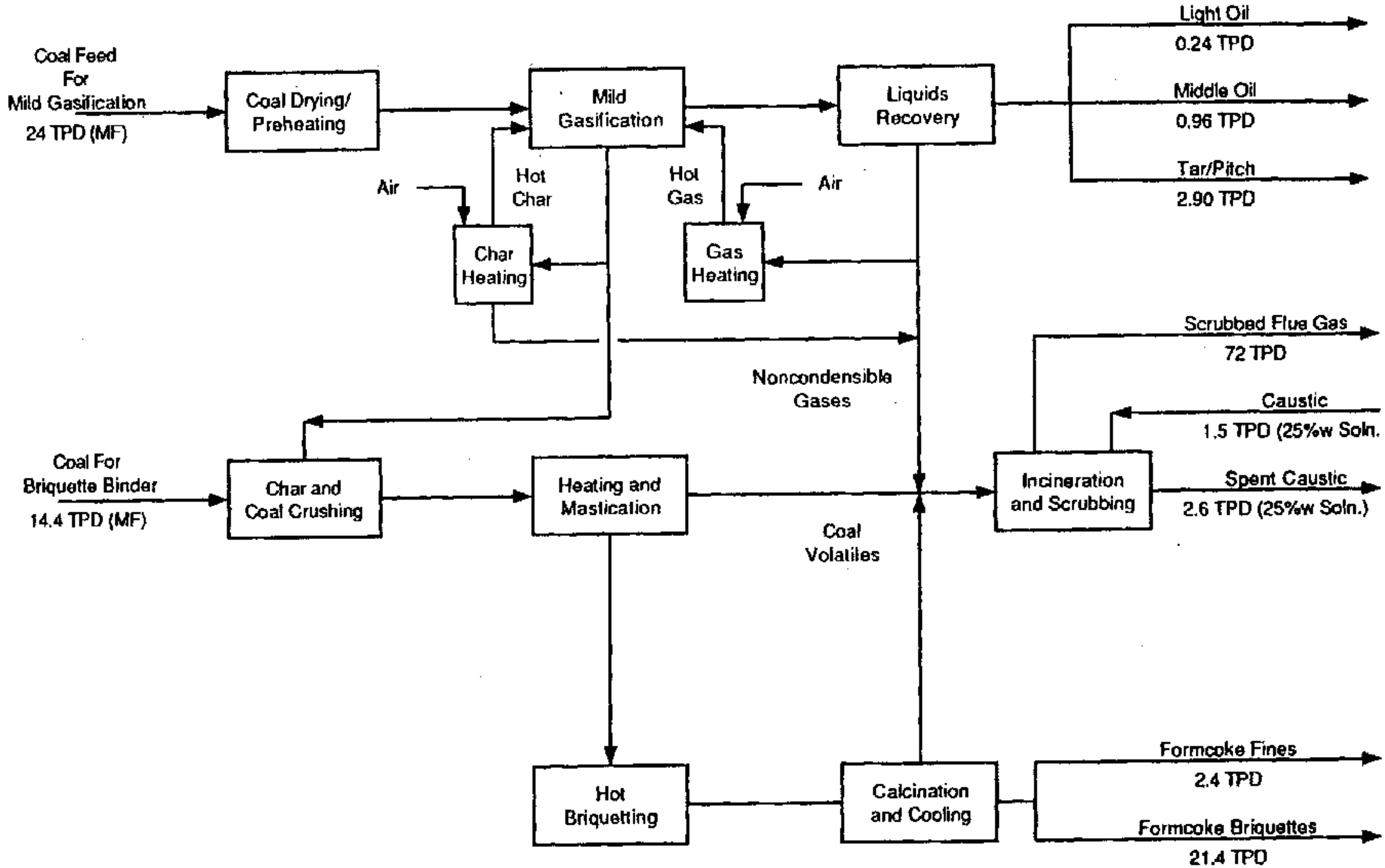

Figure 1. BLOCK FLOW DIAGRAM OF MILD GASIFICATION PDU