COAL DEPOLYMERIZATION - LIQUEFACTION

Introduction

The Coal Depolymerization - Liquefaction (CDL) process is a variation of coal liquefaction which subjects the feed coal to depolymerization (separation of a single large molecule into two or more molecules of lower molecular weight) produced by certain low-temperature reactions, followed by hydroprocessing (improvement of hydrocarbon oil quality by removal of sulfur, nitrogen, oxygen, and metals) of the resulting coal-derived liquid [1]. This pretreatment of the coal by depolymerization allows milder reaction conditions and an improved product yield and distribution. The objective of the process is to produce primarily light liquid fuels (e.g., low molecular weight hydrocarbon oils), instead of the high boiling point liquids that are typically produced by conventional direct liquefaction and which must be extensively upgraded to form premium transportation fuels.

This concept is being proposed by the Department of Fuels Engineering, University of Utah, Salt Lake City, UT with the process currently supported (in part) by the Department of Energy. The process is being developed at the lab scale.

The coal refinery based on the CDL process is shown in Figure 1. The necessary inputs to this coal refinery would include run-of-mine coal, phentydrone, ferric chloride catalyst, 10 percent methanolic-potassium hydroxide (KOH) solution, mesitylene solvent, nitrogen, hydrogen, and electricity, while major products would include gasoline, kerosene and hydrocarbon oils (light gas oil, heavy gas oil and vacuum gas oil).

Detailed Process Description

In the Coal Preparation section, the run-of-mine coal is pulverized after which the the powdered coal is pre-extracted with THF (phentydrone, $C_{13}H_{12}O$) to yield small amounts of solubles (including resins). The pre-extracted coal is then intercalated (i.e., deep-seated impregenation of a compound within the coal macro structure) using catalytic amounts (3 to 20 percent) of ferric chloride ($FeCl_3$). The resulting coal-$FeCl_3$ intercalate is hydrotreated (using hydrogen) under mild conditions (below 550°F), to partially depolymerize the coal by bond cleavage caused by destructive hydrogenation (breakup of the coal molecule due to the addition of hydrogen). The ferric chloride is back extracted and recycled to the Intercalation step.

The partially depolymerized coal is further depolymerized by base-catalyzed hydrolysis (decomposition of the molecular structure by the action of water) under supercritical conditions. The properties of supercritical substances lie between those of a liquid and those of a gas. Supercritical methods require high-temperature and high-pressure conditions. The supercritical state is preferred because within it, the substance is most reactive, being more mobile than a liquid and denser than a vapor, making it an ideal agent for extraction. The depolymerizing agent is a 10 percent methanolic-potassium hydroxide solution. The resulting coal extract is charged to the Base-Catalyzed Depolymerization (BCD) reactor with the KOH solution and pressurized by N_2 to approximately 1,000 psig and heated to approximately 480 to 550°F.

The product stream from the BCD reactor is dissolved using mesitylene (C_9H_{12}) as the solvent and hydroprocessed using either a sulfided cobalt-molybenum or ruthenium-molybenum catalyst. The hydroprocessing step is performed at about 700°F and under a hydrogen partial pressure of about 2700 psia. This results in the removal of heteroatoms (in the form of H_2S, NH_3, COS, etc.) by exhaustive hydrodeoxygenation, hydrodesulfurization and hydrodenitrogenation, yielding a light (molecular weight in the range of 100 to 270) hydrocarbon oil. The low molecular weight oils are then fractionated (separated) into different boiling fractions in the Fractionation section.

It would be expected that the off-gas from the Hydroprocessing step would be processed to remove the sulfur compounds (H_2S, COS) and ammonia. This may entail absorption of the sulfur compounds within the off-gas by either a physical or chemical solvent followed by concentration in the gas stream. The concentrated sulfur gas stream may then converted into elemental sulfur. The ammonia could be separated from the gaseous stream containing the sulfur compounds, for potential sale as anhydrous ammonia.

Types of Feed Coal

The CDL process is a form of low-temperature coal solubilization based on the application of selective chemical-catalytic reactions. As such, it should theoretically be able to process most types of coals, with the processing conditions for the stepwise depolymerization-liquefaction process varying based on the type and rank of coal to be treated [2].

The types of coals for which the CDL process has been applied are shown in Table 1, with the product distribution given in Table 2. It can be seen that the coals range from low to high in sulfur content and in rank, with the overall conversion of the different coals into hydrocarbon oils on the order of 80 to 90 percent (by weight). This is in comparison with approximately 10 percent of the starting (non-depolymerized) coal which is converted into light hydrocarbons oils after direct hydroprocessing [3].

Products

The main products from this coal refinery range in boiling point (and molecular weight) from gasoline to vacuum gas oil. The majority of the liquid products are concentrated at the lower boiling range, with over 50 percent boiling below 620°F. This minimizes further processing of the coal-derived liquid required to form premium transportation fuels, such as by cracking of the heavy hydrocarbons to decrease the molecular weight and boiling point.

Integration of the required cleanup of the off-gas streams from the different process units may allow the production of (elemental) sulfur and anhydrous ammonia, which are marketable byproducts.

Likely Applications

This coal refinery involves many chemical processing steps which are fairly complex and technically advanced in nature. This process would therefore most likely be used in conjunction with a major chemical processing facility such as a petroleum-based refinery, to take advantage of similar operating experience and utilization of similar processing units (hydrotreating, hydroprocessing, fractionation) to decrease capital and operating costs.

Status of Development

Lab scale experiments (using less than a kilogram of coal) have been performed, under a series of different operating conditions (temperature, pressure) for the individual process units, to determine the specific conditions required for the different coal types. More severe operating conditions (higher temperature for base-catalyzed depolymerization, longer time for hydrotreatment, higher hydrotreatment temperature) increased the yield of light oils and improved the product distribution due to decreased percentage of "heavies" (asphalt compounds) [3, 4]. The effect of ferric chloride concentration on the effectiveness of coal solubilization appears to go through a maximum and is dependent on the type and rank of the coal studied [4].

Environmental Aspects

The available literature does not address this issue but it is to be expected that atmospheric emissions to be in accordance with the minimum level of control allowed by NSPS. The acid gases generated within this coal refinery would most likely be removed by the appropriate technology.

One possible concern is the use of ferric chloride as the catalyst in the intercalation stage. Ferric chloride has been designated a hazardous substance under 40 CFR 116 [5], and its discharge is thereby regulated.

Research Needs

A more reactive depolymerizing agent (other than the 10 percent methanolic KOH solution) or a more active catalyst for the hydroprocessing step would lead to milder operating conditions for the overall process.

Determination of a catalyst with a greater selectivity to form lighter hydrocarbon oils would result in increased production of the more valuable liquid products.

This process (apparently) operates in the batch mode. Development of a continuous flow process, in which the coal is continuously fed into the system and products continuously generated, would potentially allow sharing of similar process units in a petroleum-based refinery.

Partial depolymerization of the coal is by ferric chloride, which may attack stainless steel due to chloride stress corrosion. Appropriate metallurgical studies to determine the degree of corrosive activity during the ferric chloride intercalation process may be warranted.

Table 1: Analysis of Feed Coals Used in CDL Research

COAL	Illinois No. 6 Bituminous	Pittsburgh No. 8 Bituminous	Helper, Utah Bituminous	Fruitland, New Mex. Bituminous	Beulah Zap Lignite
ULTIMATE ANALYSIS (weight percentage; moisture, ash-free basis)					
Carbon	78.9	76.9	81.1	78.7	72.6
Hydrogen	5.4	5.5	6.0	6.0	5.2
Sulfur	4.4	6.7	0.5	1.0	1.2
Nitrogen	1.2	1.5	1.1	1.6	1.3
Oxygen	10.0	9.4	11.3	12.7	19.7
Chlorine	0.1	0.1	< 0.1	0.1	< 0.1
Heating Value, Btu/lb	14,200	15,200	14,500	14,300	12,200

Table 2: Product Distribution from the CDL Process [1, 3, 4]

COAL	Illinois No. 6	Pittsburgh No.8	Helper, UT	Beulah Zap
PRODUCT YIELD (weight percentage)				
Gasoline (< 400°F)	15.1	20.3	18.7	Not Given
Kerosene (400 to 525°F)	29.8	25.9	27.2	Not Given
Gas Oil (525 to 620°F)	12.2	24.6	9.4	Not Given
Heavy Gas Oil (620 to 750°F)	3.5	13.7	7.7	Not Given
Vacuum Gas Oil (750 to 1000°F)	13.3	8.4	13.9	Not Given
Total Distillables	73.9(a)	92.9	76.9(b)	86 to 93

(a) Later results state amount of distillables to be 86 to 91 percent [1]
(b) Later results for a similar bituminous coal (Blind Canyon, UT) indicates that a conversion to total distillables between 83 to 90 percent [1]

Table 3: Composition of Hydroprocessed Product for Different Feed Coals [3, 4]

ELEMENTAL COMPOSITION (weight percentage, dry basis)					
COAL	Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen
Illinois No. 6	87.09	11.55	0.07	< 0.01	1.29
Fruitland, NM	88.38	10.04	0.35	< 0.01	1.23
Helper, UT	88.97	9.83	0.21	< 0.01	0.99

References

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2. J. Shabtai *et al.*, "Low Temperature Depolymerization and Liquefaction of U.S. Coals," in *Proceedings of the Sixth Annual International Pittsburgh Coal Conference,* Pittsburgh, PA, p. 103 (September 1989)

3. J. Shabtai and T. Skulthai, "Low-Temperature Coal Depolymerization. 6 . Conversion of an Illinois No. 6 Coal to a Light Hydrocarbon Oil," paper presented at the 1987 International Conference on Coal Science, Amsterdam, The Netherlands, October 26 to 30, 1987

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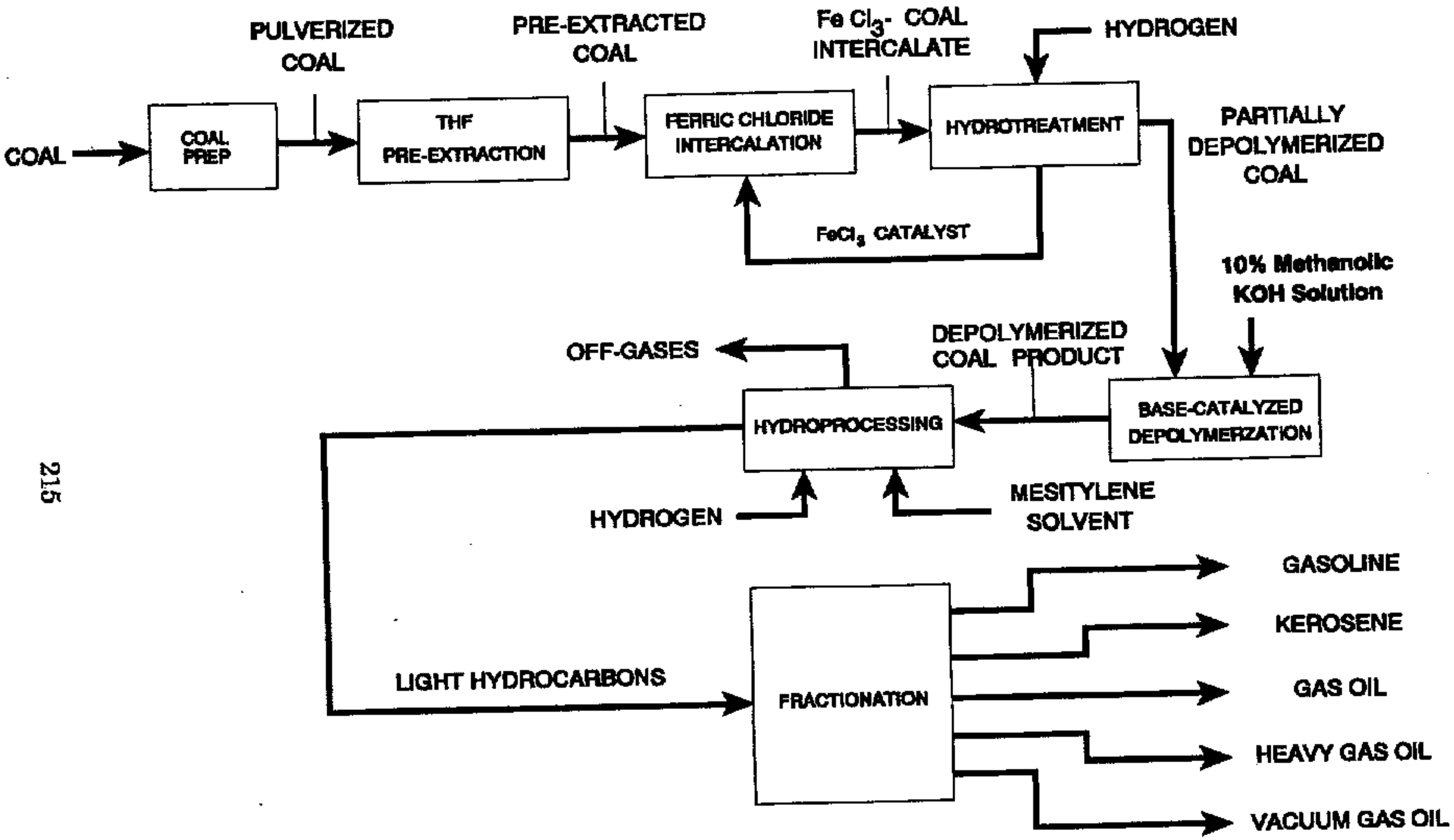

Figure 1: Overall Block Diagram of the Coal Depolymerization-Liquefaction Concept

COAL/OIL COPROCESSING

Introduction

Coal/oil coprocessing is the simultaneous reaction of coal (which can be possibly high in sulfur and nitrogen content) and poor-quality petroleum feedstocks, in the presence of hydrogen, to produce clean distillable liquids and fuel gases [1]. The process produces high-quality, environmentally acceptable fuel products from poor quality and low cost feedstocks. The liquids that are generated are low in sulfur, nitrogen, and trace metals and can be used directly as clean power plant fuels or upgraded (with conventional petroleum refinery technology) to produce transportation fuels. The petroleum feedstocks used are of such a low quality that it is too costly to process them further in a conventional petroleum refinery. Blending the poor quality petroleum feedstock with low cost coal appears to be a more economical route [2]. Due to this, it appears that coal/oil coprocessing has the potential to extend existing petroleum reserves in the U.S. and reduce dependence on imported oil. The process can generate five barrels of petroleum products from one ton of coal (for a 2:3 mixture of coal and petroleum residuum) which leads to a high-added value to the coal.

The coal liquids produced are more aromatic than petroleum products so that the octane number of gasoline produced using coprocessed liquids will be higher. An unique feature of coal/oil coprocessing is that the quality of the product slate is better than would be expected from either the coal or petroleum feeds [1,3].

Historically, coprocessing is not a new concept, as the approach was used by Germany during World War II to convert residues, coal, and coal tars to transportation fuels [1]. An earlier variation of coal/oil coprocessing was the H-Coal process [4], in which the recycled heavy distillate oil produced within the H-Coal process was used as the coal solvent, instead of the petroleum feedstock which is used in current applications of coal/oil coprocessing. A review of the coal/oil coprocessing concept up to 1986 can be found in reference [5].

The coal refinery based on coal/oil coprocessing is shown in Figure 1. The system would integrate the generation of the required process steam together with the production of chemical feedstocks. The necessary inputs to this coal refinery would be run-of-mine coal, petroleum feedstock, natural gas, electricity and water, while major products would include liquid hydrocarbons (naphtha, middle distillates, vacuum gas oil), and light fuel gases (propane, butane). Sulfur and ammonia would be byproducts of this coal refinery.

Detailed Process Description

The technology is a variation of direct liquefaction, in that coal is liquefied as the petroleum feedstock is hydrocracked (hydrocracking is a process by which high molecular weight compounds are cracked through addition of hydrogen to produce compounds of lower molecular weight and boiling point [3]. Coprocessing effectively upgrades and demetallizes the petroleum feed, allowing most metals to be removed with the unconverted coal and ash. A major difference between direct liquefaction and coprocessing is that the petroleum feed is still hydrogen-rich (H/C = 1.5) in comparison to coal (H/C = 0.8) so that the amount of hydrogen required for coal/oil coprocessing is less than that required for direct coal liquefaction (in which only coal is used). An advantage of coal/oil coprocessing is that its liquid products are more petroleum-like than coal liquids from direct coal liquefaction, which may tend to accelerate the introduction of these coal-derived liquids into existing markets. In addition, the similarity of coal/oil coprocessing technology with present petroleum-based technology would appear to be a major benefit.

The technical aspects of several coprocessing arrangements have been assessed by the Burns and Roe Services Corporation [6], and it was concluded that the one furthest developed is Hydrocarbon Research, Inc.'s (HRI) two-stage catalytic process. A detailed flow diagram based on this design is given in Figure 1, with all streams based on 100 pounds of run-of-mine coal. The inlet coal is taken to be eastern Applachian bituminous (Ohio Numbers 5 and 6), with a higher heating value of 13,476 Btu per pound (dry basis) and a sulfur content of 3.0 percent by weight (see Table 1 for more details), while the petroleum feedstock is Cold Lake atmospheric petroleum residuum (see Table 2). Coal/oil coprocessing is by two-stage catalytic ebullated-bed (in essence a bubbling three-phase reactor) processing of coal. It should be noted that this coal refinery design burns the vacuum bottoms to produce steam; other options exist in which the vacuum bottoms may be partially oxidized to partially satisfy the process hydrogen requirement, or to produce a petroleum coke [7].

In the Coal Preparation section, run-of-mine coal is prepared by crushing and grinding to reduce the coal to a particle size less than 30 mesh (0.0234 inch). If the moisture content of the feed coal is greater than 8 percent, drying will be required. No further coal preparation for impurity removal is expected as the ash will be captured by the vacuum bottoms, and coal-bound sulfur and nitrogen will be converted to H_2S (and ultimately to elemental sulfur) and ammonia respectively, in the ebullating bed reactors.

The prepared coal is then slurried with the petroleum feed and recycled heavy distillate oil. The petroleum feed could include atmospheric residuum, vacuum residuum, Fluid Catalytic Cracker (FCC) clarified slurry oil, heavy crudes or tar sands bitumen, and shale oil [1,3]. The slurry is compressed to reaction pressure (approximately 2,000 psig) and mixed with gaseous hydrogen. This mixture is then preheated and fed into the first of two ebullated-bed catalytic reactors along with fresh and recycle hydrogen. Typically, the temperature of the mixture entering the reactor is 750 to 825°F. In the reactor, the coal, oil, and hydrogen react in the presence of catalyst particles by way of hydrocracking, hydrodesulfurization (removal of sulfur and breakdown of oil), and hydrogenation (addition of hydrogen to form saturated hydrocarbons). The catalyst particles within the ebullating bed are suspended in a flowing mixture of gas, liquid and solid coal with the catalyst continuously removed and replaced by fresh catalyst without process shutdown. The effluent from the first-stage reactor flows into the second-stage reactor; two-stage operation is utilized as it allows milder conditions (lower temperatures and pressures) than a single-stage operation as well as increased product yield and reduced hydrogen consumption.

The gas and liquid effluents from the second-stage are separated by a series of pressure reductions and cooling steps. A portion of the gaseous effluent is treated to recover hydrogen which is recycled back to the slurry mixture. The gases are cleaned in the Acid Gas Removal section and separated into propane, butane, and process fuel gas (typically H_2, CH_4, C_2H_6, and inerts) by a series of compression and cooling steps in the Light Ends Recovery section. The process fuel gas is used to partially satisfy process energy requirements and as a supplement to the natural gas in the formation of hydrogen (in the Steam Reforming section).

The liquid product from the Coprocessing section is fractionated to recover distillate liquids and non-distillate product (vacuum bottoms stream). The non-distillate product stream contains all unconverted residual oil, unconverted coal, and ash. The distillate product is sent to the Atmospheric Fractionator, which separates the feed by distillation into a naphtha (C_4 to 350° F boiling point) stream and a middle distillate (350 to 650°F boiling point) stream.

The non-distillate product is sent to the Vacuum Fractionator, so as to allow boiling and separation of materials that would decompose if boiled at atmospheric pressure. The non-distillate product is separated into a vacuum gas oil (750 to 975°F boiling point) and a vacuum bottoms. The vacuum bottoms is a solid material at ambient temperature, and is solidified, flaked and combusted in the Bottom Processing section (typically, an atmospheric fluidized bed combustor, AFBC) to meet the steam requirement for this coal refinery. This accomplishes the disposal of the bottoms material in an environmentally secure manner while producing heat for the coal refinery. A solids waste stream containing the ash and spent limestone from the AFBC requires disposal in an environmentally responsible manner.

Hydrogen is consumed within the coal/oil coprocessing step. The hydrogen requirement is met in the Steam Reforming section using natural gas. Hydrogen is formed by the following reaction at high temperatures (1,400 to 1,500°F):

$$CH_4 + H_2O \rightarrow CO + 3\,H_2$$

with the CO converted to H_2 by the Shift Conversion reaction:

$$CO + H_2O \rightarrow CO_2 + H_2$$

The CO_2 in the product gas is removed.

The water-soluble waste streams are collected into a sour water stream, which is then treated by conventional means (i.e., passing steam through the heated sour water to remove dissolved gases such as ammonia and hydrogen sulfide). The ammonia is separated from the hydrogen sulfide and sold as an anhydrous ammonia byproduct. The H_2S-containing streams from throughout the coal refinery are combined and treated in the acid gas removal area to separate the light gases from the hydrogen sulfide. The H_2S stream from the acid gas removal area is mixed with the H_2S from the sour water stripping area and sent to the Claus® Plant. Here the H_2S stream is converted to elemental sulfur which will be sold as a byproduct, and a vent gas that will contain very low quantities of SO_2.

Types of Feed Coal

Coal/oil coprocessing is a variation of direct coal liquefaction, for which it has been established that bituminous, sub-bituminous, and lignitic coals may be converted to liquids [8]. The types of coal for which coal/oil coprocessing research has been pursued are given in Table 3. It can be seen that they range from lignite to bituminous [3], and from low to high in sulfur content. It would be expected that anthracitic coal would not be used for coprocessing, as anthracite tends to produce essentially all gas during direct liquefaction [8], and so would be more difficult to liquefy [5].

In general, high rank bituminous coals require more severe operating conditions than lower rank coals, with high volatile bituminous coals giving the highest liquid yields. Younger, lower rank subbituminous coals or lignites liquefy most readily but achieve lower yields and a higher proportion of gases [5].

Bituminous coals tend to produce the highest liquid yields during direct liquefaction, which is not seen for coal/oil coprocessing (see Table 4) [3]. (The negative values in Table 4 for the Heavy Distillate category denote that the heavy distillate was converted into the lighter components). The results in Table 4 indicate that more middle distillates are produced than naphtha, presumably due to the petroleum feedstock being of lower quality by containing a relatively high percentage of heavier (high molecular weight) components. Experimental results have shown that lignite to be more reactive than bituminous coal, with bituminous coal more reactive than sub-bituminous coal [3]. Coal conversions have been shown to be generally lower for lignites (90 percent versus 95 percent for Ohio No. 5/6) with the product quality more aliphatic (i.e., greater hydrogen content and lower density) [7].

Products

The main products from this coal refinery are naphtha, middle distillate, vacuum gas oil, and liquified petroleum gases (butane and propane). Marketable byproducts include sulfur and ammonia. The product output is shown in Table 5 in terms of pounds product per pound of feed coal. This analysis indicates that for the feed rates given in Figure 1 that 0.33 pounds of naphtha, 0.72 pounds of middle distillate, and 0.34 pounds of vacuum gas oil are produced from one pound of feed coal.

The overall yield and distribution of liquid products is dependent upon the operating conditions (reactor temperature and pressure), percent coal in fresh feed, recycle ratio, characteristics of the petroleum feedstock, catalyst effects, and coal type [3].

Likely Applications

Naphtha typically is a liquid intermediate in petroleum refining but is also used as a commercial solvent (for the paint and cleaners industries) and a turpentine substitute [9]. The naphtha can be further treated by catalytic reforming in a conventional petroleum refinery to produce high octane reformate, suitable for blending to motor gasoline or petrochemical feedstock. The middle distillate could be blended to an existing No. 2 diesel fuel supply, sold "as is" as turbine fuel for utility peaking applications, or after mild hydrotreating to lower the levels of sulfur and nitrogen, sold as a finished No. 2 diesel fuel. The vacuum gas oil could be used in utility turbines due to its low sulfur and nitrogen content (0.15 to 0.55 weight percent sulfur; 0.3 to 0.5 weight percent nitrogen), or as a premium quality low sulfur No. 6 fuel oil. When used with low-NO_x burner designs, these fuel products can be used as an alternative fuel while reducing the emissions of NO_x and SO_2 to extremely low levels.

The liquified petroleum gases (butane, propane) have many various uses, in the fields of domestic (as appliance fuel), industrial, commercial, agricultural (grain drying, orchard heating, etc.) and internal combustion engines (fork lift trucks, etc.) [10]. Butane is also used as a component in gasoline manufacture.

This coal refinery involves many chemical processing steps which are fairly complex and technically advanced in nature, with a product output similar to a conventional petroleum refinery. This process would therefore most likely be used in conjunction with a major chemical processing facility such as a petroleum-based refinery, to take advantage of similar operating experience and utilization of similar processing units (hydrotreating, hydroprocessing, fractionation) to decrease capital and operating costs.

Status of Development

Coal/oil coprocessing integrates the commercially available petroleum residuum hydroprocessing technology with coal liquefaction. Hydroprocessing improves the quality of various petroleum products or cracks heavy carbonaceous materials to lower boiling, more-valuable products. Resid hydroprocessing in the petroleum industry can be accomplished by way of a three-phase ebullating bed reactor, where catalyst particles are suspended in a flowing mixture of gas and liquid [2, 11]. This technology has been used commercially for more than 20 years to catalytically upgrade residual and heavy oils. Two of the successful, commercially proven resid hydrocracking technologies are the H-OIL [12] and LC-FINING [13] processes.

Ebullating-bed technology has also been used in direct liquefaction of coal. Lummus-Crest used LC-FINER reactors as the second stage together with a short-contact-time thermal first stage in their Integrated Two-Stage Liquefaction (ITSL) process. At the Wilsonville coal liquefaction test facility, H-OIL reactors are used for both first and second stages, while Hydrocarbon Research Incorporated (HRI) uses two close-coupled H-OIL reactors in their Catalytic Two-Stage Liquefaction process [14]. A similar process for coal liquefaction which utilizes ebullating-bed technology is the H-Coal Process [4]. The H-Coal Process converts coal into liquid fuels by slurrying the coal with a solvent distilled from the synthetic fuel oil produced from the coal. A 250 tons per day pilot plant was constructed and operated at Catlettsburg, KY, adjacent to the Ashland Oil, Inc. refinery, to determine the technical and economic feasibility of the process.

Ohio Clean Fuels, Inc. was awarded $45 million under the U.S. Department of Energy (DOE) Clean Coal Technology Program for demonstration of a Prototype Commercial Coal/Oil Co-Processing plant based on the HRI technology [15]. The project was one of the new technologies selected by DOE in the first Clean Coal Technology solicitation. The plant, proposed for a Warren, Ohio site, was designed to process 800 tons of Ohio No. 5/6 high-sulfur bituminous coal and 8,675 barrels of oil per day to produce 12,280 barrels of clean distillate and 57 tons of sulfur per day. As a part of this project, several bench-scale (50 lbs per day) tests and an extended run with a 3 ton per day fully-integrated pilot plant (PP) were performed. Steady-state reaction conditions, yields, and product quality were verified during a 45 day PP run [16]. The PP demonstration run appeared to confirm that the process could simultaneously upgrade high-sulfur coal and heavy oil to light products at a 90 percent conversion rate (see Table 6, from reference [17]). Process confirmation testing was felt to be successfully concluded in May 1989, but the balance of the project was suspended pending identification of a primary customer of the proposed plant's fuel product. The project was eventually terminated in June 1991, prior to the start of detailed design.

A proposal for a prototype (1,200 ton per day of Ohio No. 6 coal; 20,000 barrels per day of Alberta heavy oil) demonstration plant of this coal refinery concept has been submitted by Frontier Energy Corporation in the fourth Clean Coal Technology solicitation [18].

The process economics of coal/oil coprocessing have been studied extensively [3, 6, 7, 14, 18]. Because of the groundwork established in the past, if coprocessing could be shown to be technically and economically feasible on a plant scale operation, it would present an alternative to direct coal liquefaction that could deliver acceptable coal-derived liquids to the market place at an earlier date than could coal liquefaction technology.

Environmental Aspects

Coal/oil coprocessing theoretically can convert high-sulfur, high nitrogen coal into liquid fuels that are low in sulfur, nitrogen, and trace metals, and high in heating value. The majority of the sulfur (over 80 percent) and nitrogen (over 70 percent) in the feed coal and petroleum feedstock is recovered from the process in the form of elemental sulfur and ammonia, avoiding the generation of SO_2 and NO_x which would result if either the feed coal or petroleum feedstock was directly combusted without significant emission control.

The main environmental intrusions from this coal refinery include atmospheric emissions of SO_2 and NO_x, particulates (from the combustion of the vacuum bottoms), and solid wastes. The major atmospheric emissions result from the combustion of the vacuum bottoms to generate steam. The emission rate for SO_2 is approximately 0.0003 lb per lb of feed coal (assuming 90 percent sulfur capture within the AFBC), while for NO_x it is approximately 0.0001 lb per lb of feed coal. The SO_2 and NO_x emission rates would be in accordance with the minimum level of control allowed by NSPS.

The average solid waste production rate would be on the order of 0.19 lb per lb of feed coal. The solid waste would include the bottom and fly ash from the feed coal, spent sorbent (in the form of calcium sulfate, calcium carbonate, etc.), and the mineral content of the petroleum feedstock. It is expected that the toxicity of the solid waste would not differ greatly from that generated from a typical petroleum refinery or a coal-fired power plant.

Long-term buildup of carbon dioxide (CO_2) has been thought to lead to climatic change due to a warming of the earth's lower atmosphere. Carbon dioxide is generated by the calcination of the limestone used in the AFBC:

$$CaCO_3 \rightarrow CaO + CO_2$$

by the combustion process within the AFBC, and by the two reactions used in the Steam Reforming section to produce the hydrogen requirement for the process. The CO_2 which is produced within the AFBC (approximately 0.8 lb per lb of feed coal) would be released to the atmosphere. It may be possible to extract the CO_2 produced by steam reforming of natural gas (0.7 lb per lb of feed coal) for storage and possible compression to 1,500-2,000 psig for pipeline delivery.

Research Needs

Bench-scale and PDU data indicate the coal/oil coprocessing concept to be technically well grounded. A reliable, efficient and economical unit would be demonstrated by the successful integration of the many process steps of the different technologies at production rates approaching commercial scale.

Better understanding of the interactions between the two feedstocks (coal, oil) may lead to potential process improvements. One improvement is the possible large scale recovery of the spent catalyst particles from the ebullating beds through regeneration.

Table 1: Coal/Oil Coprocessing Plant Feed Coal Analysis

CONSTITUENTS IN COAL (weight percentage)						
Carbon	Hydrogen	Oxygen	Sulfur	Nitrogen	Chlorine	Ash
75.3	5.6	7.6	3.0	1.6	0.1	6.8
Free Moisture (weight percent)			3.1			
Higher Heating Value (dry basis)			13,476 Btu per lb			

Table 2: Coal/Oil Coprocessing Plant Feed Petroleum Feedstock Analysis

CONSTITUENTS IN OIL (weight percentage)				
Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen
83.7	10.2	0.5	5.2	0.4
Specific Gravity		1.025		
° API		6.6		

Table 3: Analysis of Feed Coals Used in Research

COAL	Ohio No. 5/6 Bituminous	Illinois No. 6 Bituminous	Alberta Sub-bituminous	Texas Lignite
Carbon, weight %	75.3	69.9	67.9	63.3
Hydrogen, weight %	5.6	4.9	4.7	5.3
Sulfur, weight %	3.0	3.7	0.5	1.2
Nitrogen, weight %	1.6	1.4	1.4	1.2
Ash, weight %	6.8	11.7	8.2	12.0
Oxygen, weight %	7.6	7.7	17.3	17.0
Moisture, weight %	3.1	4.9	9.9	30.0
Heating Value, Btu/lb (dry basis)	13,476	12,332	11,394	11,010

Table 4: Distillable Oil Composition as a Function of Coal Type and Percent Coal in Feed

COAL	Ohio No. 5/6 Bituminous	Alberta Sub-bituminous	Ohio No. 5/6 Bituminous	Alberta Sub-bituminous
Coal in Feed, weight %	33.0		50.0	
DISTILLABLE OIL COMPOSITION (lb per lb moisture, ash-free coal)				
Total Oil Yield	0.59	0.65	0.61	0.59
Naphtha	0.27	0.3	0.37	0.35
Middle Distillate	0.64	0.53	0.57	0.41
Heavy Distillate	-0.32	-0.18	-0.33	-0.17

Table 5: Product Output from Example Coal/Oil Coprocessing Coal Refinery

PRODUCT OUTPUT (lb product per lb feed coal)						
Naphtha	Middle Distillate	Vacuum Gas Oil	Butane, C_4H_{10}	Propane, C_3H_8	Sulfur	Ammonia, NH_3
0.33	0.72	0.35	0.02	0.04	0.06	0.02

Table 6: Performance of Process Demonstration Unit Run

PROPERTY	YIELD (weight percent on feed)
975° F plus conversion	88.1
Coal conversion	95.2
Hydrogen consumption	4.5
Desulfurization	82.4
Denitrogenation	78.3
Demetallization	99.0
PRODUCT YIELD (weight percent on feed)	
C_1 to C_3	6.9
C_4 to 350° F	16.5
350 to 650° F	44.1
650 to 975° F	16.5
975° F plus (includes solids)	12.7

Table 5: Product Output from Example Coal/Oil Coprocessing Coal Refinery

PRODUCT	OUTPUT (lb product per lb coal)
Naphtha	0.33
Middle Distillate	0.72
Vacuum Gas Oil	0.35
Butane, C4H10	0.02
Propane, C3H8	0.04
Sulfur, elemental	0.06
Ammonia, NH3	0.02

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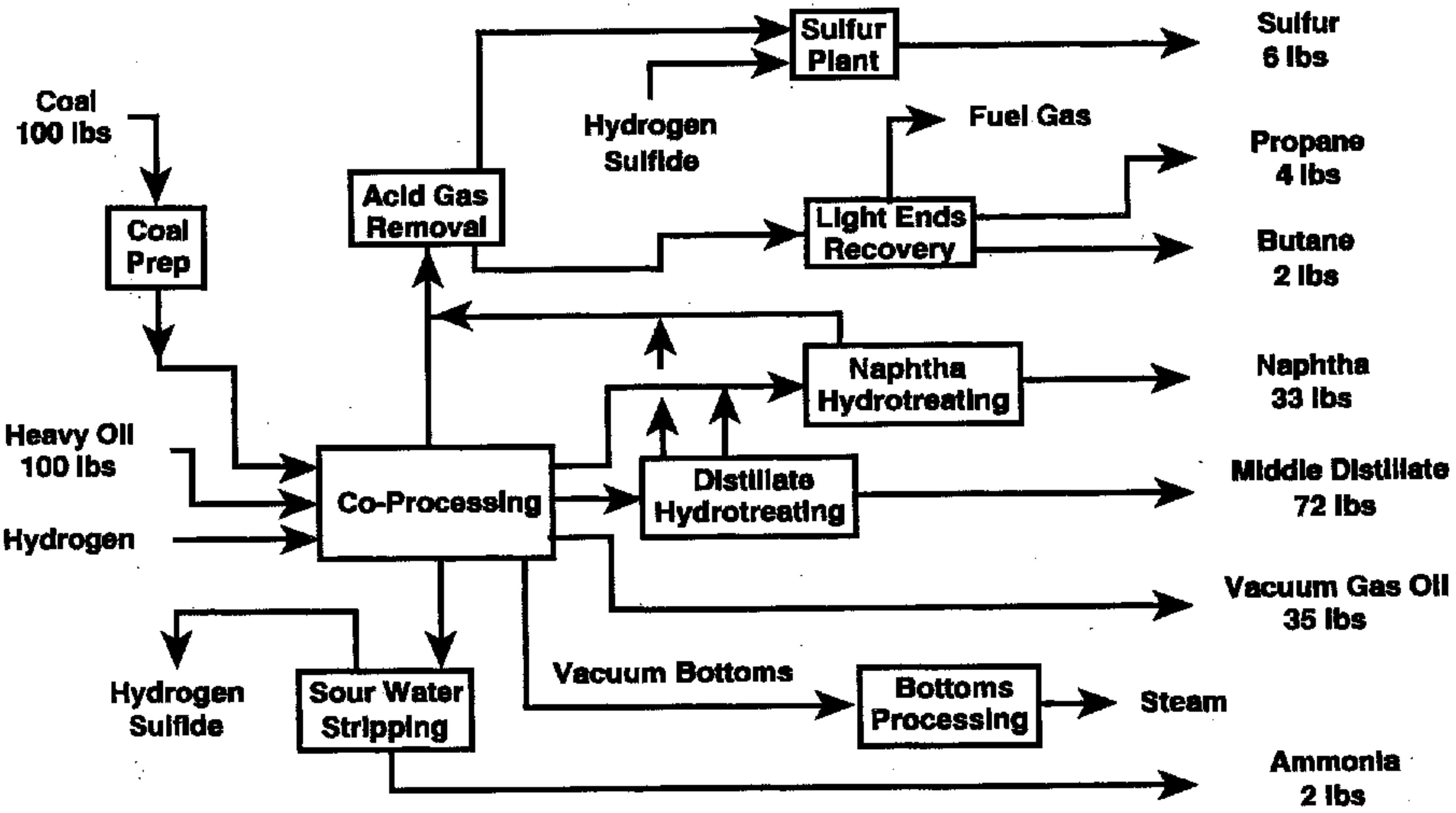

FIGURE 1: Overall Block Diagram of the Coal/Oil Coprocessing Concept