

## ENHANCED COAL LIQUEFACTION BY RAPID HEATING

### Introduction

The objective of the Enhanced Coal Liquefaction by Rapid Heating concept is to develop a technique by which coal can be liquefied to high conversions while reducing the amount of hydrogen required to achieve this.

This concept is proposed by the Department of Fuels Engineering of the University of Utah, at Salt Lake City, Utah with the process currently supported in part by the Department of Energy. The research program is currently in its third year of a four year funding scenario and is being developed at the bench scale level.

Pending patent applications limit the amount of information available about this process.

### Process Description

Coal is pulverized to a powder state and the coal particles heated with hot hydrogen gas in turbulent flow, thereby achieving coal particle heatup rates on the order of thousands of degrees Fahrenheit per second. A catalyst hydrogenates (addition of hydrogen) the coal to liquids in 5 to 10 seconds. More than 70 percent (by weight) of the initial coal is converted into liquids, with hydrocarbon gas formation restricted to less than 0.7 to 1 pound gas produced per 10 pounds of liquid. This is in comparison to a hydrocarbon gas evolution rate of 2 to 3 pounds per 10 pounds of liquids typically seen in other liquefaction processes. The low amount of product gas reduces consumption of hydrogen thereby improving the process economics, as equipment for hydrogen production can be 40 percent of the capital cost of a liquefaction plant.

### Research Needs

Further process development work would establish the technical and economical viability of this process. A market assessment of the products expected of the process would establish the merits of possible commercialization of this coal refinery concept.

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## LIQUID SOLVENT EXTRACTION

### Introduction

The Liquid Solvent Extraction (LSE) process utilizes noncatalytic direct hydrogenation coal liquefaction and involves contacting coal at temperatures up to 950 °F with a "donor" solvent; i.e., a solvent that is capable of transferring labile (readily removable) hydrogen atoms to the coal and thereby maximizing the fraction of coal that goes into solution [1]. The LSE process essentially involves two stages, with the first stage being low-pressure thermal liquefaction and high temperature filtration, with catalytic hydrocracking (heavy distillate hydrocarbons converted under hydrogen pressure into lower molecular weight products) of the coal extract as the second. A two-staged conversion system is used due to the recognition that coal liquefaction takes place in two stages, consisting of coal dissolution followed by upgrading of the solubilized products [2]. The goal of the process is to economically produce premium liquid transportation fuels such as gasoline and diesel from any type of coal [3].

This concept has been proposed by the British Coal Corporation, of Great Britain. The process is being demonstrated at the pilot plant level in a liquefaction plant at Point of Ayr, North Wales.

The coal refinery based on the LSE process 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, air and natural gas (to form hydrogen), while the major products would include liquid hydrocarbons (naphtha, distillate), LPG (propane, butane), and potentially SNG (synthetic natural gas) and coke. Sulfur and ammonia would be byproducts of this coal refinery.

### Detailed Process Description

The technology is a variation of earlier processes for direct liquefaction [4], in that coal is dissolved at low pressures that eliminate the need for hydrogen or a suitable catalyst. In addition, unlike most current U.S. processes, de-ashing of the dissolved coal extract is by filtration. A detailed process flow diagram outlining these points is given in Figure 1.

In the Coal Preparation section, the run-of-mine coal is pulverized and then dried. The pulverized and dried coal is then slurried with the process-derived hydrogen-donor solvent from the Distillation section. The slurry is pressurized to 290 psia (to limit undue evaporation of the process solvent) and heated, then fed into the Digester at 770°F. Coal is hydrogenated (addition of hydrogen) due to the presence of the donor solvent, dissolving up to 95 percent of the coal. The resulting digest is then cooled to about 570°F, depressurized, and filtered to remove the coal mineral matter,

undissolved coal, and metals in the Cooler and Filtration unit. The resulting liquid from the filtering step is called the "coal extract solution." A portion of the light solvent from the Flash Vessel is used as the wash oil for the filtration step, to displace the coal extract solution. The residual wash oil is recovered by vacuum drying of the filter cake. The washing and vacuum drying steps minimize the amount of coal extract solution and light solvent that remain within the filter cake.

The coal extract solution is then pressurized to approximately 3,045 psia, mixed with hydrogen gas, and preheated to 570° F before being fed into the catalytic Hydrocracking section operating at 750 to 850° F. Hydrocracking is by ebullated bed reactor (for more details, see discussion on Coal/Oil Coprocessing), yielding products of lower molecular weight and boiling point; hydrocracking decreases the molecular weight of aromatic compounds like those seen in coal-derived liquids resulting in lower-boiling products. The product stream from the Hydrocracker is fed into the Distillation section, to fractionate (separate) the product stream into its individual components. The three main products are LPG (propane, butane); naphtha (boiling point below 320° F); and middle distillate (boiling point between 320 and 570° F). The process-derived hydrogen-donor solvent (boiling point between 570 and 840° F) is recovered for recycling back to slurry the coal. In addition, some solid pitch (boiling point above 840° F) is produced as a byproduct. The remaining byproducts from the Distillation section consist of a light hydrocarbon fuel gas (i.e., CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, etc.) and acid gases (H<sub>2</sub>S, NH<sub>3</sub>). This sour gas is processed in the Gas Treatment section, to produce a fuel gas stream and elemental sulfur and ammonia as byproducts. The fuel gas stream may be gasified to produce hydrogen or processed to become SNG (synthetic natural gas; see discussion on Coproduction of SNG, Electricity, Methanol and Chemical Intermediates).

A major consideration in coal liquefaction is the relatively large amount of hydrogen that must be added to remove hereatoms (i.e., H<sub>2</sub>S, NH<sub>3</sub>) and to convert material containing about 5 percent hydrogen by weight to products with 12 to 14 percent hydrogen. The generation of hydrogen is accomplished by gasification of the intermediate byproducts of this coal refinery (fuel gas, steam, filter cake, and pitch) and supplemented by coal if required. The required oxygen for the Gasifier unit is produced from air in the Air Separation plant (not shown in Figure 1). A portion of the nitrogen generated in the Air Separation plant is used for process requirements, with the remainder vented to the atmosphere. The flue gases from the Gasifier (containing primarily CO, H<sub>2</sub> and CO<sub>2</sub>, with trace amounts of CH<sub>4</sub> and other light gases) would be converted into additional hydrogen (for more details, see discussion on Dual Production of Ammonia and Electricity) and the impurities contained in the sour gas stream removed in the Gas Treatment section.

The process steam requirement is supplied by combustion of the pitch, which also generates electricity for the plant requirement. The pitch however may be processed in the Delayed Coker section where the high-molecular weight pitch is thermally

cracked to a lighter distillate oil and a solid coke. The coke may be used to generate hydrogen in the Gasification section, or upgraded to a higher-value product.

Saturated hydrocarbons (which do not contain labile hydrogen) may build up in the process-derived hydrogen-donor solvent, resulting in lower rates of coal dissolution. The donor solvent would then be sent to the Sat Coker unit to adjust its saturates (paraffinic) content; the products from the Sat Coker unit are olefinic (unsaturated open-chain hydrocarbons containing at least one double bond) in nature. This step helps produce a process-derived solvent with labile hydrogen.

### **Types of Feed Coal**

Solvent extraction has been shown to have the ability to convert any grade of coal below anthracite (from lignite to low-volatile bituminous) to usable liquid products [5]. The LSE technology was developed for processing bituminous coal, but has been successfully applied to subbituminous coals and lignites [6, 7], although the overall efficiency of the process will decrease with these lower rank coals.

The LSE process is similar to the Solvent-Refined-Coal (SCR-II) process with a major difference being filters instead of the Kerr-McGee critical solvent de-ashing (CSD) process are used. The SCR-II process has liquefied both U.S. bituminous (including Illinois No. 6 and Kentucky No. 9) and subbituminous coals [5]. Therefore, it may be expected that this coal refinery concept based on the Liquid Solvent Extraction process can be successfully applied in this country.

### **Products**

The main products from this example coal refinery are LPG (propane, butane), naphtha, and middle distillate. The LSE process can generate a favourable liquid-to-gas ratio of about 4.8 (at 750 to 800°F, ~ 300 psia) [2]. The essentially nitrogen-free distillate can be made into a high-octane, lead-free petrol and a low-sulfur low-pour-point diesel. The sulfur content of the diesel fuel produced is approximately 0.05 percent, with a benzene content in the range of 1 to 3 percent, both of which are less than in typical diesel fuels produced from crude oil [8].

The process is reported to yield approximately 4 pounds of LPG, 17 pounds of gasoline, and 17 pounds of diesel from 100 pounds of coal (containing 10 percent moisture and 10 percent mineral matter) [7]. The yield of the individual products depend on process characteristics such as Digester temperature, residence time of the donor solvent in the Digester, and coal type.

## **Likely Applications**

Naphtha is an intermediate in petroleum refining and can be further treated to produce high octane reformat, for blending in motor gasoline or as a petrochemical feedstock. The octane number of the reformat is reported to be in excess of 100, making it suitable for blending to premium gasoline [7]. The middle distillate fuel oil can be used by electric utility and industrial markets that traditionally use petroleum fuels or can be upgraded for use as diesel fuel.

The current coal refinery concept gasifies the coke for hydrogen generation; other designs being investigated involve the creation of graphite and high-purity electrode cokes for the aluminum and steel industries [9].

Commercial transportation fuel suppliers may be interested in pursuing this coal refinery concept so as to assure an indigenous source of fuel from coal. Given that the product output and certain processes of this coal refinery concept closely resembles a conventional petroleum refinery, the primary entity that would be interested would be the petroleum refining industry.

## **Status of Development**

The Liquid Solvent Extraction process is currently being demonstrated at a 2.5 ton per day pilot liquefaction plant at Point of Ayr, North Wales [6]. After extensive development in a 20 lb per day bench unit, the LSE process was scaled up to 2.5 tons per day [9]. One difference in the process design between bench and pilot scale is that a fixed bed hydrocracker was employed during bench-scale, while an ebullated-bed reactor will be used in the pilot plant. The project is designed to validate the process's technical and commercial feasibility, evaluate the performance of catalysts during long operating periods, and provide samples for testing in automobiles engines and design data for commercial coal-derived liquefaction plants that could consume between 5 and 6 million tons per year of coal. The pilot plant will operate during a three-year period beginning in 1991.

Detailed mathematical models of laboratory, pilot and commercial scale (50,000 barrels per day) facilities have been developed to aid in process optimization and scaleup [7]. Estimates of the overall process economics are given in references [3] and [6].

## **Environmental Aspects**

The LSE process extracts the majority of the sulfur and nitrogen from the parent coal in the form of elemental sulfur and ammonia. Recovery is claimed to be greater than 96 percent [10]. In addition, other pollutants contained within the coal, including heavy metals, are also removed.

The main environmental intrusions from this coal refinery include atmospheric emissions of  $\text{SO}_2$  and  $\text{NO}_x$  from the combustion of the pitch and the gasification of the filter cake (and potentially coal), and solid wastes. The solid wastes would consist of ash and refuse from the parent coal, and sludges and solids recovered from waste treatment processes, with ash as the major component. Solid waste disposal has been stated to be an environmental concern for this coal refinery [8], as the leachate from the solid waste may require extensive controls.

The majority of the  $\text{CO}_2$  produced from this coal refinery would be due to the combustion of the pitch for steam and power generation, and from the gasification (of the coke, filter cake and potentially coal) and subsequent water-gas shift of the synthesis gas. It may be possible to capture a large percentage of this  $\text{CO}_2$  production within the Gas Treatment section, through the proper choice of the acid-gas treatment process (e.g., see Table 11.5 of reference [11]).

### **Research Needs**

The cost of producing hydrogen is an important component in the overall cost of coal-derived liquids by the direct liquefaction process, with hydrogen generation possibly contributing as much as 20 percent of the total capital cost of the coal liquefaction plant. Development of a method of low cost hydrogen production from coal would reduce the potential economic liabilities of this coal refinery concept.

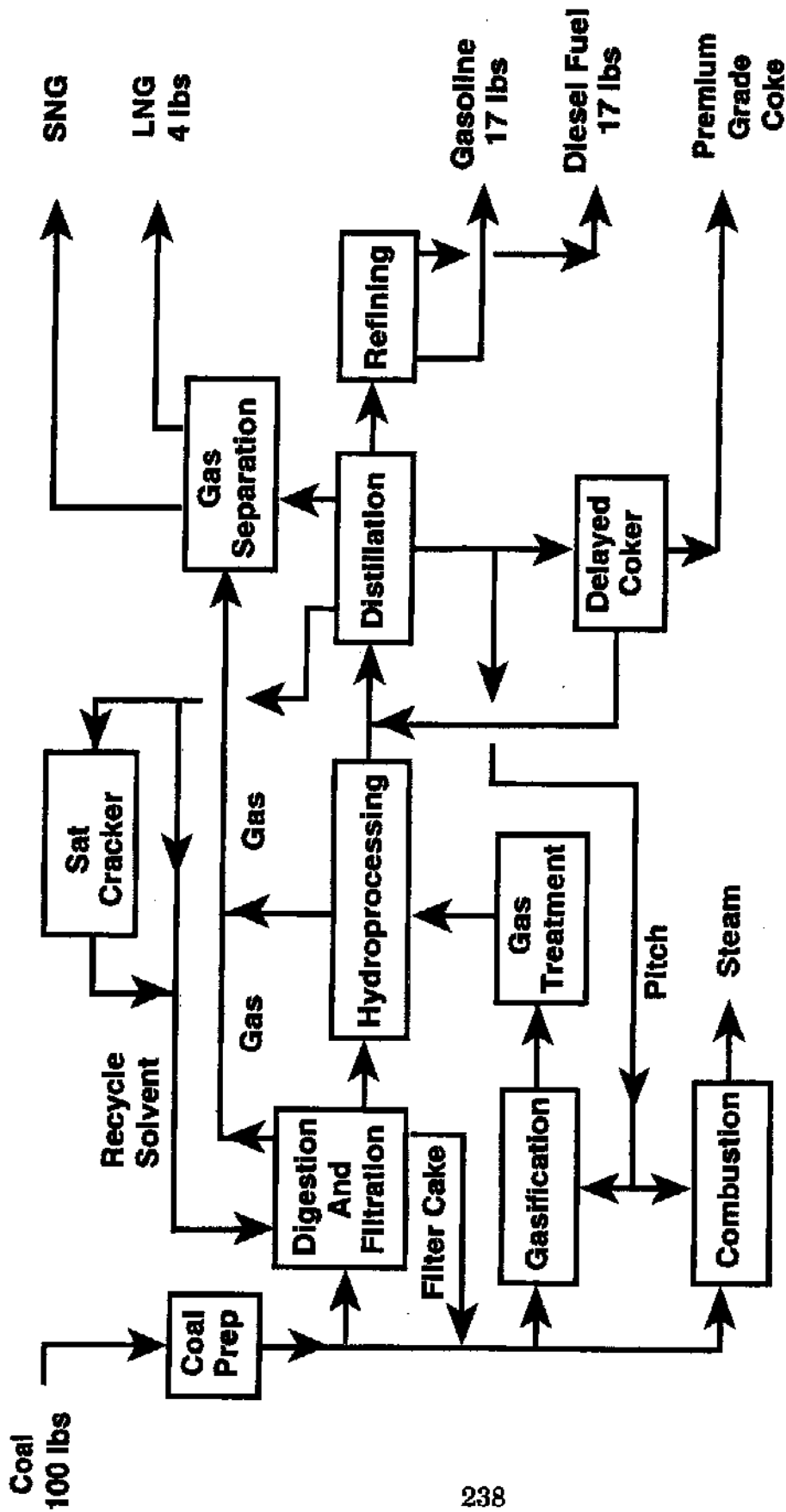
The process model for determination of the overall economics was based on predictions of product yields from fixed-bed hydrocracker experiments. Experimental results from the pilot scale ebullated-bed hydrocracker (when available) would allow recomputation of the overall economics to more accurately reflect the potential commercial-scale application of this coal refinery concept.

Revision of the economic model to include any possible revenue generated from byproduct sales, and better estimates of the possible construction period and plant reliability, after construction and operation of the pilot facility would help to establish the economic viability of this coal refinery concept.

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**FIGURE 1: Overall Block Diagram of the Liquid Solvent Extraction Concept**

## NEDOL

### Introduction

The NEDOL process involves the combination of three processes similar to U.S. processes: direct hydrogenation (H-Coal, [1]), solvolysis (ITSL, [2]), and solvent extraction (EDS, [3]) [4]. Integration of the different features of the three processes is felt to have many advantages such as greater flexibility in the mix of distillate products and less-severe reaction conditions than single stage liquefaction processes.

This concept is being developed by the New Energy Development Organization (NEDO) of Japan. The process is being demonstrated at the process development unit level.

The coal refinery based on the NEDOL (1 ton per day) process development unit is shown in Figure 1. The system essentially consists of a thermal dissolution stage as used in the ITSL process, and a solvent hydrogenation stage as used in the EDS process. The necessary inputs to this coal refinery would include coal, makeup hydrogen (or natural gas/some other fuel for hydrogen generation), synthetic pyrite catalyst, and electricity, while major products include naphtha, middle distillates and potentially LPG. Potential byproducts include (elemental) sulfur and ammonia.

### Detailed Process Description

The run-of-mine is pulverized to become a powder in the Coal Preparation section, after which the powdered coal is slurried (mixed) with the recycle solvent and a synthetic pyrite catalyst (at approximately 3 percent by weight) in the Slurry Mixer. The slurry is pressurized to approximately 2,400 psig after which hydrogen is added at an (approximate) ratio of 55 standard cubic feet (SCF) of hydrogen to one pound of slurry. The gas-liquid mixture is then heated to 840°F, and fed into the Thermal Dissolution (liquefaction) reactor. The mixture reacts for one hour at about 800 to 860°F, and 2,200 to 2,900 psia [5]. The coal liquefies and the heteroatoms in the coal are converted into hydrogen compounds such as H<sub>2</sub>S and NH<sub>3</sub>. The vapor (raw gas) and liquid are flash separated in the Vapor/Liquid Separator with the coal-derived liquid fed into the Fractionation (distillation) section.

The coal liquids are separated into a light oil (product naphtha), a middle distillate (which may be a product or recycled as part of the recycle solvent stream to maximize production of naphtha), a heavy oil stream which is to be catalytically hydrogenated (addition of hydrogen to the molecule of an unsaturated hydrocarbon), and a residue stream. The residue stream will typically contain ash and unconverted coal and could be a potential fuel source, such as for the production of process steam within an atmospheric fluidized bed combustor (AFBC). Use of the residue stream as a fuel would

increase the overall process efficiency and potentially decrease the amount of solid wastes from this coal refinery.

The heavy oil (and possibly the middle distillate) product from the Fractionation section is pressurized to approximately 1,400 psig and hydrogen added at an (approximate) ratio of 28 SCF per pound liquid. The gas-liquid mixture is heated to 625 to 700° F and fed into the Solvent Hydrogenation unit. The Solvent Hydrogenation unit is a fixed-bed reactor, containing Ni-Mo/Al<sub>2</sub>O<sub>3</sub> catalyst with which the heavy distillate is hydrogenated at about 600 to 750° F and 1,400 to 2,200 psia at a space time of 1 hour (i.e., 1 hour required to process one reactor volume). The product stream is fractionated (separated) to produce naphtha and the recycle solvent with which to slurry the coal.

The raw gas from the Vapor/Liquid Separator is treated to recover hydrogen which is recycled back to the slurry mixture and the heavy distillate. The gases would be cleaned in the Acid Gas Removal section to produce (elemental) sulfur and anhydrous ammonia, and possibly separated into propane, butane, and process fuel gas (typically CH<sub>4</sub>, H<sub>2</sub>, C<sub>2</sub>H<sub>6</sub>, and inerts) by a series of compression and cooling steps in the Light Ends Recovery section. The process fuel gas could be utilized to partially satisfy the process steam requirement and in the generation of hydrogen.

If the process hydrogen requirement could not be fulfilled due to insufficient fuel gas production, natural gas (or potentially coal) could be used to form hydrogen in the Steam Reforming section.

### **Types of Feed Coal**

The NEDOL process has been designed to liquefy bituminous coal. Extension to other types of coal has not been demonstrated.

### **Products**

The main products from this example refinery include naphtha and middle distillates, with a targeted yield of over 50 percent naphtha plus middle distillate having been established [5]. The naphtha and middle distillate may require further upgrading (by hydrotreatment) in order to produce a suitable petrochemical feedstock. LPG is a potential major product, unless the process economics indicate that it would be best utilized in hydrogen production. Elemental sulfur and anhydrous ammonia could be potential byproducts from this coal refinery concept.

### **Likely Applications**

Naphtha is an intermediate in petroleum refining and can be further treated to produce high octane reformat, for blending in motor gasoline or as a petrochemical

feedstock. The middle distillate could be sold as a turbine fuel for peaking applications or after mild hydrotreating to lower the levels of sulfur and nitrogen could be sold as a finished No. 2 diesel fuel.

The commercial entities that may be interested in this coal refinery concept may include the petrochemical industry, commercial transportation fuel suppliers, and primarily the petroleum refining industry (because many of the process steps and technologies are similar to petroleum refining).

### **Status of Development**

A 150 ton per day coal liquefaction plant is scheduled to begin construction in 1991 with operation to begin in 1994. Supporting research is being conducted to obtain process and engineering data for the larger scale plant, with research performed in 3 units: a 0.1 ton per day bench scale unit, a 1 ton per day process development unit and a 1 ton per day process supporting unit [4].

### **Environmental Aspects**

Environmental intrusions from this coal refinery can be expected to be comparatively small, with atmospheric emissions potentially consisting of  $\text{SO}_2$ ,  $\text{NO}_x$  and particulates from the combustion of the residue for steam generation. It is expected that the  $\text{SO}_2$  and  $\text{NO}_x$  emission rates would be in accordance with the minimum level of control allowed by NSPS.

The solid wastes would include the bottom and fly ash from the feed coal, spent sorbent from the AFBC, and possibly the synthetic pyrite catalyst, if it is shown that the economics for its recovery and regeneration are unfavourable.

### **Research Needs**

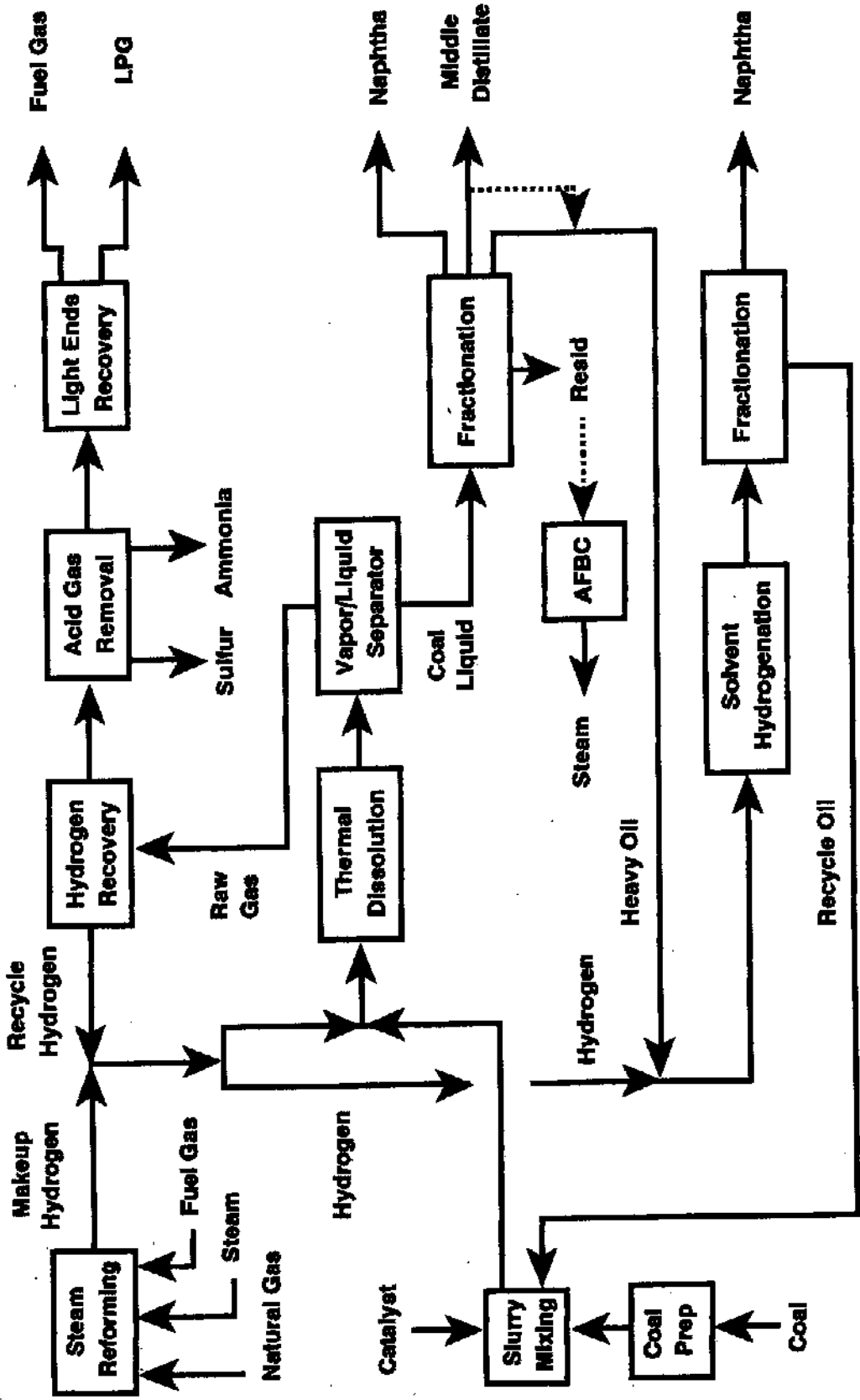
The process development units (apparently) do not integrate processes that would fully utilize the raw gas stream emitted from the Vapor/Liquid Separator. Integration of these processes with the remainder of the system may potentially lead to a more thermally efficient design.

The NEDOL process is currently limited to bituminous coals, and further research may be warranted to extend the technology to other coal types.

The process is still at the PDU level. Successful scaleup would allow commercial-scale demonstration of this process and aid in establishing the overall process economics

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**FIGURE 1: Overall Block Diagram for the NEDOL Concept**

## NIPPON BROWN COAL LIQUEFACTION

### Introduction

The Nippon Brown Coal Liquefaction (BCL) process involves the two-stage liquefaction of high-moisture Victorian (Australia) brown coal (lignite), and consists of four unit processes: dewatering, primary hydrogenation (PH), deashing and secondary hydrogenation [1]. The project is specifically designed to liquefy Victorian brown coal.

This concept is being developed by the Nippon Brown Coal Liquefaction Co, Ltd., which is a consortium of five Japanese companies - Kobe Steel, Mitsubishi Kasei, Nissho Iwai, Idemitsu Kosan and Asia Oil [2]. The process is being demonstrated at the pilot plant level in a liquefaction plant at Morwell, Victoria. The project has been financed by the Japanese government through the New Energy Development Organization (NEDO) in cooperation with the Victorian state government. The major thrust is to more fully utilize Victorian lignite as the brown coal reserves in Victoria are estimated at 202 billion tons, of which 43 billion is thought to be economically and readily recoverable. The brown coal has a high moisture content (on the order of 60 percent), making it uneconomical to transport and only less than 2 percent of the coal has been used since mining began in 1921 [3].

The coal refinery based on the BCL process is shown in Figure 1. The system would integrate the production of the required hydrogen together with the production of chemical feedstocks. The necessary inputs to this coal refinery would be run-of-mine brown coal, disposable catalyst, natural gas (for hydrogen generation), and electricity, while the major products would include liquid hydrocarbons (naphtha, middle distillate) which can be upgraded to produce premium transportation fuels such as gasoline and diesel.

### Detailed Process Description

The BCL process consists of two-stage hydrogenation with deashing between stages. The major differences with respect to current U.S. liquefaction technology are in the use of a cheap disposable iron-based catalyst which is not regenerated (unlike the higher-cost but regeneratable catalysts employed in this country) and the emphasis on fixed-bed instead of ebullated-bed reactors. These points are more fully outlined in the following text and in Figure 1.

In the Coal Preparation section, the run-of-mine brown coal is pulverized to below a particle size of about 0.003 inches, mixed with the disposable once-through iron catalyst and dried by a special dewatering system developed for the BCL process. It is necessary to remove the water from the coal due to the high moisture content (about 60 percent by weight) and because the water will vaporize in the Primary Hydrogenation section and raise the pressure. The dewatered coal/catalyst mixture (moisture

content of approximately 14 percent by weight) is then slurried with the process-derived hydrogen-donor solvent (i.e., solvent-refined coal bottoms and hydrogenated deashed oil) in the Slurry Mixer. The slurry is then pressurized (to about 2,150 to 3,000 psia), mixed with hydrogen and heated to about 800 to 850° F. The resulting mixture is thermally dissolved and hydrogenated in the Primary Hydrogenation (PH) section. The PH section can consist of a series of reactors designed to assure complete mixing, with the hydrogenation carried out in suspension using the high-performance disposable catalyst. The off-gases containing light hydrocarbons (i.e., CO, CH<sub>4</sub>, etc.) and water vapor are separated from the coal-derived liquid in the Gas-Liquid Separator, with the resulting coal-derived liquid fractionated (separated) in the Solvent Recovery Distillation section to produce a light distillate fraction (boiling point below 360° F), solvent (boiling point between 360 and 790° F), and a non-distillable fraction (coal liquid bottoms, boiling point above 790° F). The solvent is highly aromatic, with a ratio of approximately 0.7 to 0.8 aromatic carbon atoms to total number of carbon atoms [4]. The coal liquid bottoms is a solid at room temperature and only becomes liquid at high temperature. The light distillate fraction is removed as (product) naphtha, while a portion of the solvent and coal liquid bottoms is recycled to become part of the process-derived hydrogen-donor stream.

Two-stage liquefaction requires a separation of nondistillable liquid product from the ash and unconverted coal. The majority of the coal liquid bottoms are treated to separate the non-distillable liquid intermediate from the residue (coal ash, unconverted coal and disposable catalyst) in the Solvent Deashing unit. The deashed oil is then mixed with the rest of the solvent fraction from the PH section and fed into the Secondary Hydrogenation section. Fixed-bed hydrogenation is carried out in the secondary stage, using a high durability Ni-Mo/Al<sub>2</sub>O<sub>3</sub> catalyst. The resulting product liquid is fractionated in the Secondary Distillation section to produce naphtha and middle distillate (boiling point between 360 and 480° F) as products and high-boiling oils from the catalysis stage (hydrogenated deashed oil, boiling point above 480° F). The hydrogenated deashed oil is recycled together with the solvent and coal liquid bottoms to the Slurry Mixer.

The hydrogen requirement would be partially satisfied through steam reforming of the off-gases from the Gas-Liquid Separator (using steam and the light hydrocarbons).

Typical operating conditions for the Primary Hydrogenation stage based on the 0.1 ton per day (TPD) process development unit (PDU) are given in Table 1 [5].

### **Types of Feed Coal**

The BCL process has been designed to liquefy only one type of coal: Victorian (Australia) brown coal. Its properties are shown in Table 2 [6]. Extension of the process to other types of coal has not been demonstrated.



## **Products**

The main products from this example refinery are naphtha and middle distillates; typical properties of the two products are given in Table 3 [4]. Most of the product is obtained in the first stage and upgraded by the Ni-Mo/Al<sub>2</sub>O<sub>3</sub> catalyst in the second stage [7]. The targeted liquid yield is 50 percent naphtha and middle distillate [8]. Initial indications are that the process can produce one barrel of naphtha (light oil) or middle distillate from a ton of raw brown coal [2].

## **Likely Applications**

Naphtha is an intermediate in petroleum refining and can be further treated to produce high octane reformat, for blending in motor gasoline or as a petrochemical feedstock.

The commercial entities that may be interested in pursuing this coal refinery concept may include the petrochemical industry (to assure a continuous supply of industrial chemicals, based on coal) and commercial transportation fuel suppliers. The primary entity that would be interested would be the petroleum refining industry.

## **Status of Development**

The Brown Coal Liquefaction process is being demonstrated at a 50 ton per day pilot liquefaction plant at Morwell, Victoria. The BCL process has been successfully developed and scaled-up from research and development works in a 5 liter autoclave to 0.1 and 0.5 ton per day process development units and then to the 50 ton per day pilot plant [1]. Basic data was collected on the liquefaction stage from the smaller scale units to further develop the BCL process. The 50 ton per day plant started operation in Morwell in 1987, with the plant achieving continuous operation for up to 1,700 hours [3]. Operations were terminated in October 1990, and the unit is being disassembled.

The next stage in scaleup will be a demonstration plant that will process 10,000 tons of raw brown coal per day to produce 10,000 barrels per day of oil, followed by full production of 100,000 tons per day [9]. The full-scale conversion plant is not expected to be fully operational until at least the year 2007. It has been stated that due to prior experience and the relatively moderate reaction conditions that future scale-up to commercial operation will be relatively easy [3].

Estimates of the required selling price of the coal liquids generated by this process is given in reference [2].

## **Environmental Aspects**

The available literature does not address this issue but it is to be expected that the acid gases generated during hydroliquefaction ( $H_2S$ ,  $NH_3$ ) would be removed by the appropriate technology (absorption by a physical or chemical solvent followed by concentration into a gaseous stream for further processing and removal). One possible concern is the disposal of the catalyst for the Primary Hydrogenation stage but the catalyst would most likely be a "red mud" (an iron oxide byproduct of bauxite processing) for which disposal techniques are known and well-established. The solid wastes generated by this coal refinery concept would consist of ash and refuse removed from the parent coal and sludges and solids recovered from waste water treatment processes. It would be expected that the amount of solid wastes to be relatively small due to the low ash content (less than 3 percent on a moisture, ash-free basis) of Victorian brown coal.

## **Research Needs**

A throw-away catalyst of relatively low activity is used in the primary hydrogenation stage. Development of a catalyst with a higher activity and selectivity and lower rate of deactivation may be warranted.

The BCL process has been designed and applied only for lignitic coal from Victoria, Australia. Its applicability to reactive lignite coals from the U.S., such as Big Brown lignite (Texas), etc., has not been demonstrated.

Optimization of the liquefaction technology may warrant the development of new pressure vessels, pumps and valves required for scale-up to the commercial plant.

The lignite for which the BCL process was designed contains approximately 60 to 70 percent water by weight. This water is evaporated away but potentially is a source of hydrogen. The moisture within the coal may be utilized for hydrogen production by the development of a suitable catalyst.

Table 1: Typical Operating Conditions for the 0.1 TPD PDU Unit [5]

Pressure (psia)	Temperature (°F)	Solvent/maf Coal (weight/weight)	Catalyst (weight % on maf coal)
150 to 350	800 to 880	2.5 to 3	3 to 5

Table 2: Properties of Victorian Brown Coal [2]

PROXIMATE ANALYSIS (weight percentage)				
Moisture		Ash		Volatile Matter
62.2		1.1		20.5
ULTIMATE ANALYSIS (weight percent; moisture, ash-free basis)				
Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen
67.1	4.7	0.4	0.3	27.5

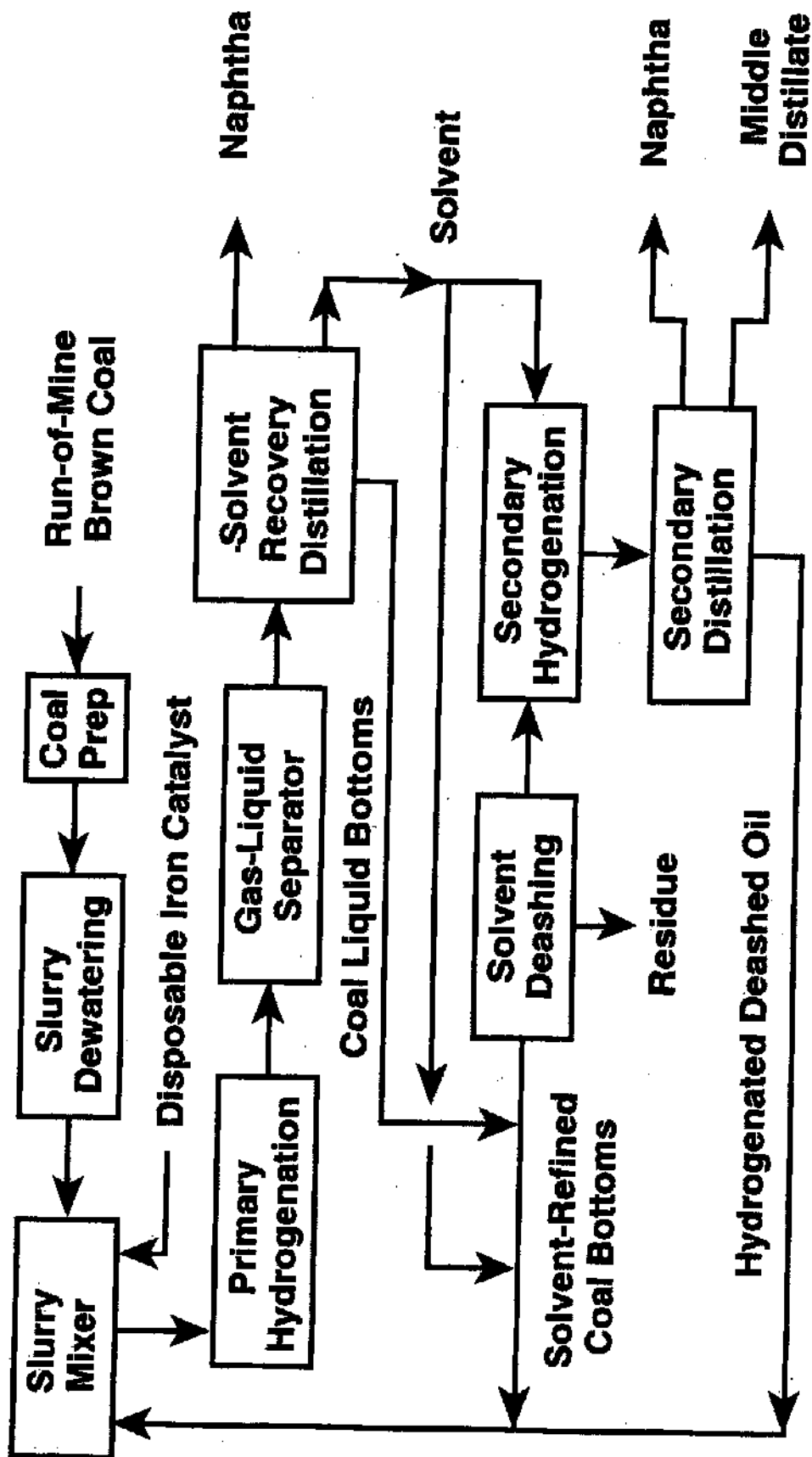
Table 3: Properties of the Naphtha and Middle Distillate Products [4]

ULTIMATE ANALYSIS OF PRODUCT NAPHTHA (weight percent)				
Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen
86	12	0.6	0.1	1.4

ULTIMATE ANALYSIS OF MIDDLE DISTILLATE PRODUCT (weight percent)				
Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen
88 to 89	10.5 to 11	0.05 to 0.06	0.01 to 0.02	0.2 to 0.6

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**FIGURE 1: Overall Block Diagram of the Nippon Brown Coal Liquefaction Concept**

**APPENDIX D**

**BIOPROCESSING COAL REFINERY CONCEPTS**

## BIOCONVERSION OF COAL TO ALCOHOL FUELS

### Introduction and Description

The bioprocessing of coal to produce gaseous and liquid fuels is an active research area. Various cultures and consortia of microbes are being examined for conversion efficiency and specificity of product [1, 2]. Microbial conversion for alcohol production has certain advantages over chemical conversion. Substantial energy and potential equipment savings are possible. The potential savings in equipment result from a decrease in pressure and temperature in contrast with conventional chemical processes for producing the same products from coal. A major disadvantage of bioprocessing is that the low reaction rate necessitates large and specially configured processing vessels. Nutrient and sterility requirements are also disadvantages. In general however, research progress has engendered enthusiasm among the investigators for a future role of bioprocessing in the production of alcohol fuels.

The possible process steps, microbial organisms, and process conditions for connecting coal to alcohols are many. Broadly speaking, they can be grouped into indirect or direct conversion.

The indirect method starts with coal derived, raw synthesis gas ( $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ ). Microorganisms are used to produce alcohols from this synthesis gas. With the proper strain of microorganisms and conditions, an alcohol consisting principally of a single type can be produced. The reactions of interest usually involve anaerobic organisms because the rate of reaction is advantageous as compared to aerobic organisms. Cultures capable of producing ethanol from raw, synthesis gas in a continuous reactor have been studied. One early culture isolated from animal waste produced significant ethanol but also a considerable fraction of acetate (in acetic acid) [3]. Other cultures have also been able to produce alcohols.

The direct method, where one set of microorganisms liquifies the coal and another set produces the alcohol fuels, also has potential. One of the difficulties in this approach is to find organisms that are compatible with liquified coal products. Because coal has such a large and varied chemical structure, many of the liquified products are toxic to the microorganisms producing the ethanol. Also, the rates of reaction appear to be very low.

Researchers at the University of Arkansas have suggested a coal refinery concept with two variations [3, 4]. One variation uses a three step approach. In the first step, coal is pulverized and then liquified through the use of microorganisms. As noted above, there are currently many difficulties with reaction rates, costs, etc., that must be overcome for this step to be feasible. However, if liquid yields from this first step were such that the liquids could be economically upgraded into higher-value products, this microorganism liquefaction step could be attractive in its own right. The next step involves

the oxidation of the liquid to provide gaseous products which are used as feed material for the third step where microorganisms are used to produce the alcohol fuel. The second variation on this concept is an indirect method where conventional coal gasification is used to produce the synthesis gas and then microorganisms are used to convert this gas into alcohol fuels. Detailed descriptions of these steps are lacking. In general, an inhibitor is used to suppress methane formation. Ethanol is the main alcohol formed, however, acetic acid is also a product.

### **Status and Research Needs**

Lignites and subbituminous coals were used in one study of a novel approach for coal bioconversion to alcohol fuels [5]. Soluble coal products were produced using hydrogen peroxide pretreatment plus biosolubilization using select microbial cultures. Low production of solubilized carbon indicated that, perhaps, research efforts should be oriented toward conversion of untreated coal directly to alcohols. Preliminary calculations indicate a coal carbon conversion to ethanol of 35 percent to 45 percent can be attained with untreated coal.

An obstacle to commercialization of bioprocessing to produce alcohol fuels is the unknown cost. Reaction rates between coal and the microbes must be improved. Currently, the estimated bioreactor size is very large for a commercial production facility and is a key factor in any economics. Cost estimates are highly uncertain because of the extrapolation of these data to designs that will result in conversion times of minutes rather than the hours currently experienced. The economics of alcohol production from coal depend on specific engineering definition and on associated production cost estimates.

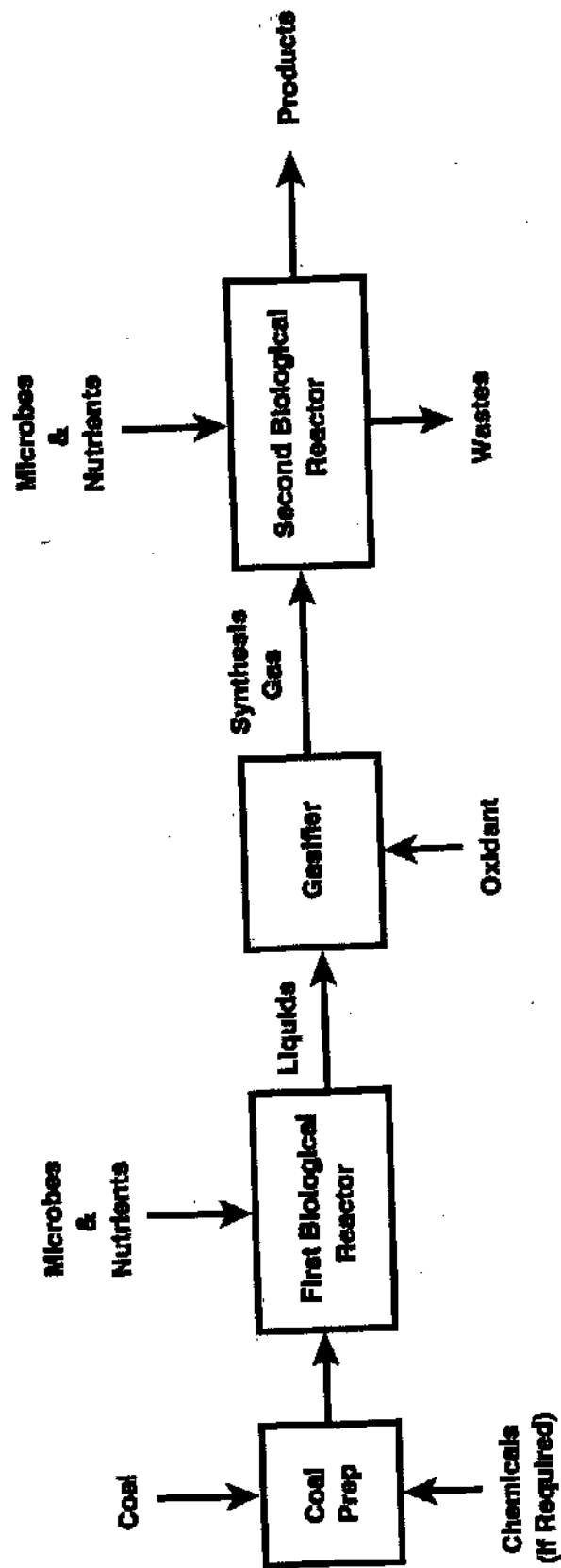
Further research on the selection of microorganisms, optimum configurations, and system requirements could help to establish conditions that might lead to an economical commercial process.

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**FIGURE 1: Overall Block Diagram of the Bioconversion of Coal to Alcohol Fuels Concept**

## LIGNITE REFINERY

### Introduction

In the last ten years, there has been a growing interest in the bioprocessing of coal [1, 2]. The U.S. Department of Energy (DOE) and the Electric Power Research Institute (EPRI) have committed significant funds to this growing field. The research efforts on the use of microorganisms has included work on (a) the reduction of sulfur in coal, (b) the conversion of syngas to liquid and gaseous fuels and chemicals, and (c) the solubilizing of coal and its conversion to liquid or gaseous fuels and chemicals. In the third category, a Texas utility, because of its interest in natural gas and the availability of a large amount of lignite, undertook a resource study linked with bioprocessing to produce gas and liquids.

This economic study of a "lignite refinery," which incorporates bioprocessing, was carried out by Houston Power and Light Company [3]. Heat and material balances were calculated, equipment sized, and capital and operating costs estimated.

Texas lignite, which is about 50 percent water and ash, was the input fuel. The main products from this concept were methane, benzene, toluene, xylene, and heavy oil. The concept did not meet their prestudy economic criterion of 15 percent return on investment, but one of the cases did produce a positive cash flow. Process efficiency improvement and product price increases are needed to meet the economic criterion.

### Process Description

Lignite and sodium carbonate are slurried with water and heated to about 480°F. This material, under about 600 psi pressure, is fed to a pretreatment vessel where the lignite breaks down and water soluble organic compounds are formed. Material from the vessel is reduced in temperature and centrifuged. A solids stream containing ash and unreacted lignite is removed from the circuit. The liquid, which contains organic acids, is sent to a Benzene, Toluene and Xylene (BTX) reactor. A simplified flow schematic of this process is shown in Figure 1.

In the BTX reactor, nominal BTX is formed from about 50 percent of the acids. This mixture is then cooled and sent to a "phase separator" where the acids and BTX are partitioned. BTX is sent to a distillation module where the nominal benzene, toluene, xylene, and heavy oils are separated. The acids are sent to a bioreactor.

Nutrients and a pH buffer are pumped, along with the acids and other dissolved species, to an underground cavern containing the microorganisms. Here, the organisms feed on acids to produce methane and CO<sub>2</sub> under approximately 650 psi pressure. The gas phase containing most of methane and some of the CO<sub>2</sub> is sent to a methane purification circuit. The liquid phase containing the waste and some of the CO<sub>2</sub> is sent to

a flash unit. The CO<sub>2</sub> is recovered and is assumed to be a marketable product. The liquid is filtered for solids removal and the filtrate is partially recycled and the remainder treated as plant effluent.

The concept described here has been simplified by a number of assumptions. Development work, of course, is required to demonstrate an overall working system versus analytical design using experimental data for some process steps. The study examined a biogasification alternative as well as a refinery.

### **Process Feed Materials**

The refinery design is based on a Texas lignite feed of 20,000 tons per day. This lignite consists of 32 percent moisture, 16 percent ash, 27 percent volatiles, 24 percent fixed carbon and 1 plus percent sulfur. About one tenth pound of sodium carbonate is used per pound of lignite. Phosphoric acid and urea are added nutrients to the bio-reactor to makeup the phosphorus and nitrogen deficiency in the feed material.

### **Products**

Pipeline quality CO<sub>2</sub> and methane are produced along with aromatic hydrocarbons. Approximately 700 tons of methane and 2,100 tons of CO<sub>2</sub> are produced per 20,000 tons of lignite. Nominal BTX production is 1,300 tons per 20,000 tons of lignite. Some heavier liquids are also produced. Although not used in the study, the residue from the centrifugal separation appears to have a significant energy content and could be used as a boiler fuel.

### **Likely Applications**

This concept was investigated by an electric utility having interests in natural gas supply and biotechnology. The utility would probably be interested in pursuing this concept if it appeared attractive. Energy resource firms and gas transmission firms, could also be interested in applying the developed technology. Chemical firms with biotechnical expertise could also be interested, especially if they have uses for BTX materials in chemical manufacturing.

The CO<sub>2</sub> product would have its principal market in enhanced oil recovery activities. Methane, of course, has an existing market with a vast distribution system of pipelines.

### **Status**

A main purpose of the study discussed in Reference [3] was to investigate the feasibility of a methane-producing, bioprocess concept, using current technical information. One of several cases examined was the lignite refinery concept described here.

The use of lignite as the energy bearing material was central to the study. A multiproduct refinery concept was secondary in the economic analysis. As noted in Reference [3], the economics of the process were not attractive. Thus, the refinery concept does not have a development status per se because the results were not encouraging. However, the study did show that a refinery with its multiproducts was more economically attractive than biogasification alone.

### **Environmental Aspects**

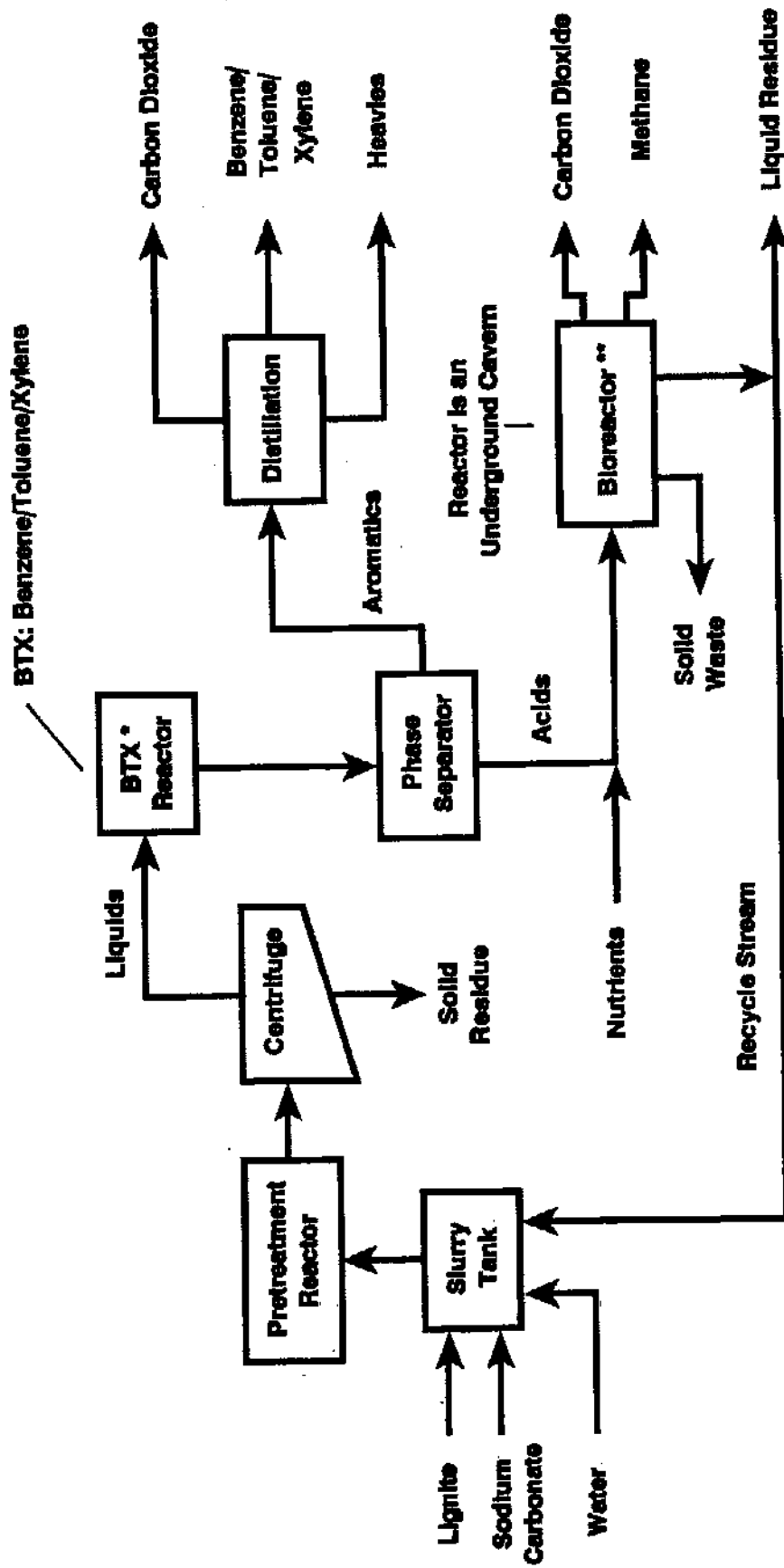
A discussion or examination of the environmental issues was not part of the lignite refinery study. However, issues about the disposal of the solid wastes, disposal of excess fluids (contaminated water) and potential subterranean water contamination from the bioreactor are certainly evident. The solid wastes could be reduced in volume by using the energy content for steam and electrical production. The economics of this use are uncertain and depend on actual energy content of the waste, as was noted in Reference [3]. A large amount (2,000 tons per day of sodium carbonate is fed to the plant) of sodium bearing water must be disposed of.

### **Research Needs**

This coal refinery concept is an early state of evolution and there is a need to focus efforts on process improvements. Although the analysis in the study was based on the most accurate information available, further research would be required to predict yields of products under various conditions.

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**FIGURE 1: Overall Block Diagram of the Lignite Refinery Concept**