APPENDIX C

LIQUEFACTION COAL REFINERY CONCEPTS

AGFLOTHERM

Introduction

The Agflotherm process simultaneously beneficiates subbituminous coal and upgrades bitumen and/or heavy oil. The process is basically the agglomeration of low quality coal using a bridging liquid, typically bitumen (a semi-solid cementitious material derived from petroleum) or a low-quality heavy oil, followed by thermal treatment of the agglomerated coal to recover and convert part of the bridging liquid into a distillable oil. The result is a solid fuel with low ash and moisture content, high heating value, and good handling properties (i.e., uniform particle size and quality), and a processed oil with a low heteroatom content, reduced density and viscosity [1].

This concept has been advanced by the Alberta Research Council, of Alberta, Canada. Work is continuing under the sponsorship of a number of U.S. and Canadian organizations, including the Electric Power Research Institute (EPRI) [2].

The coal refinery based on the Agflotherm process is shown in Figure 1. The necessary inputs to this coal refinery include run-of-mine coal, bitumen/heavy oil, water, and steam (or nitrogen), while products would include a high calorific solid fuel, and a high quality oil that could be further processed by conventional petroleum refining technology.

Detailed Process Description

This variation of oil-agglomeration coal cleaning relies on the difference in surface properties of coal and its impurities. Agglomeration is a size enlargement process by which small particles gather into larger, permanent masses in which the original particles can still be identified [3]. The organic carbonaceous matter in coal is generally oleophilic (readily wetted by oil) while most of the inorganic coal constituents (excluding pyrite) are hydrophilic (readily wetted by water). When finely ground coal, water, and oil are mixed together, the organic coal particles become coated with a thin layer of oil (also known as the bridging liquid) and collect as clusters (i.e., agglomerate). The inorganic impurities remain suspended in the aqueous phase but rapidly settle out. The agglomerated coal can then be separated from the water and the unagglomerated particles.

In the Coal Preparation section, run-of-mine coal is pulverized to a nominal particle size of 100 percent minus 0.024 inch (average diameter approximately 0.008 inch) to liberate mineral matter and pyrite. The coal is then slurried with water in the Coal Slurry section to form a dilute suspension (approximately 25 to 35 weight percent coal). The coal water slurry then passes to the Mixing section where a second liquid (bitumen and/or heavy oil; approximately 0.5 to 5.0 percent of dry coal) is added to the coal water slurry. High shear mixing (by agitation, in-line high shear mixers, or other mechanical means) is performed to ensure preferential wetting of the coal by the oil. The finely divided organic coal particles in the water suspension agglomerate and separate from the suspending liquid (water) due to the addition of the small amount of the second liquid which preferentially wets the organic coal particles but is immiscible with water. After agitation, compact spherical clusters of the organic coal particles (called microagglomerates) held together by the oil are formed. The mineral matter and pyrite remain suspended in the water.

The entire mixture of microagglomerates and water with mineral matter and pyrite is passed to the Agglomerate Separation section, which could be either a flotation cell or a series of screens. The stream containing the mineral matter, pyrite and water (called tailings) separate from the microagglomerates. The mineral matter and pyrite are then separated from the water (one possible method is by a hydroclone, a liquid-solid cyclone), with the water recycled back to the Coal Slurry section and the coal impurities headed for disposal in an environmentally responsible manner.

The microagglomerates are then collected, washed, and dewatered using screens after which they are sent to the Size Enlargement area. Their size is increased, either by mechanical means such as pressure compaction or by extended agitation while adding additional bridging liquid (5 to 25 weight percent of dry coal). The amount of bridging liquid is always greater than that required in the production of the microagglomerates. Large-scale clusters, called macroagglomerates, are formed. The bridging liquid above that required for particle cohesion is to be recovered.

In the Oil Recovery section, the macroagglomerates are subjected to a short residence time (on the order of 10 minutes) thermal treatment at elevated temperature and under an atmosphere of inert gas. The optimum thermal treatment was found to occur at 660°F with steam more effective in oil recovery than a nitrogen purge [2,4]. The thermal treatment unit could either be a rotary kiln-type preheater (a type of solids-drying equipment) or a fluidized bed [4]. Thermal treatment was found to improve the water repellant properties and strength of the macroagglomerates while recovering a portion of the valuable bridging oil used in the agglomeration steps.

Types of Feed Coal

The process has been applied to lignitic, subbituminous, and bituminous coals. The types of coal for which the Agflotherm process has been pursued are given in Table 1 [2, 5]. In general, all the coals tested could be beneficiated by oil agglomeration to a certain degree [2]. Application of this process to subbituminous and high volatile bituminous coals resulted in a solid fuel with excellent properties. However, modification of the agglomeration technique was required for lignite, as initial experiments showed its agglomerating properties to be poor with respect to the other coals studied. This is due to the efficiency and selectivity of the oil agglomeration technique in general decreasing with decreasing coal rank.

For all coals tested, oil agglomeration resulted in a solid product fuel with low ash and moisture contents and higher heating value than the original coal. The amount of increase was found to depend on the degree of ash removal, amount of bridging liquid added, and moisture rejection [2].

During the agglomeration process, water is displaced as the bridging liquid becomes adsorbed on the surface of the organic coal particles. The chemical and physical properties of the bridging liquid have a greater affinity for reactive coal sites than does water. Subsequent thermal treatment of the macroagglomerates derived from subbituminous coals led to a solid fuel with a lowered moisture capacity (defined as the amount of moisture in equilibrium with air at 68°F and 95 percent humidity) than the original coal.

In general, a lower viscosity for the bridging liquid produced solids with lower final ash contents while a more viscous bridging liquid produced larger agglomerates. Highly viscous liquids form immobile liquid bridges where the binding strength between individual particles within the cluster is much larger, allowing larger agglomerates to form without subsequent breakage [6].

For bitumen and very heavy oils, the recovery of bridging liquid is approximately 50 percent, with significantly higher yields for heavy oils which are less dense and viscous [4]. The amount of bridging liquid required was found to depend on the rank of coal, its surface properties, particle size distribution of the original coal, its ash content, and the size of agglomerates to be produced [7].

Products

The main products from this coal refinery are a solid fuel and a processed oil (i.e., recovered bridging liquid). The properties of the agglomerated solid fuel and the recovered bridging liquid are given in Tables 1 and 2, respectively. It can be seen in Table 1 that the solid fuel has a higher calorific value and lower in mineral matter (ash) and moisture. The recovery of combustibles (e.g., energy recovery, measured in Btus) is very high. The product solid is stable, transportable, dust-free, and has a low moisture content.

The bridging liquid recovered by thermal treatment does not contain any of the higher-boiling fractions (asphaltenes) associated with heavy oils, as the heavier oil components remain with the solid fuel. In addition, the processed oil is lower in heteroatom (nitrogen, sulfur) content, density, and viscosity than the initial oil (see Table 2). Certain properties of the recovered oil (i.e., API gravity, viscosity) in Table 2 are comparable to a No. 2 fuel oil [2], but further upgrading of the processed oil is required for heteroatom removal.

Likely Applications

The Agflotherm process could potentially be useful in powerplant pre-combustion fuel preparation to meet environmental requirements for existing units. The solid fuel can be used in existing plants to reduce emissions when used with current post-combustion cleaning techniques which eliminates the necessity of extensive retrofits. The processed oil would be sold to help the process economics. It is to be expected that the commercial entities primarily interested in this coal refinery concept would be mine owners (of low-quality coal who wish to upgrade their raw coal into a more marketable product) and fuel vendors (to enter into new markets).

Status of Development

Research to study oil agglomeration for the beneficiation of coal and coal fines by the Alberta Research Council started in August 1984 [2]. Low rank subbituminous coals were shown in a 2 liter batch stirred tank system to be successfully agglomerated with liquids made up primarily of bitumen, heavy oil, and heavy refinery residue [2, 4]. The process was successfully scaled-up to a larger batch stirred tank (approximate volume of 160 liters) and a continuous closed-loop pipeline system (approximate total volume of 46 liters) [2].

The process was judged by EPRI to have a potential for commercialization. A joint U.S.-Canadian consortium was established in 1990 to fund the next stage of development which is a 6 ton per day continuous coal-oil agglomeration bench-scale unit [2].

Environmental Aspects

The Agflotherm process removes pyritic sulfur and mineral matter from the feed coal. The mineral matter content can be greatly reduced by this process, as shown in Table 1. A Pennsylvania coal with a particle size less than 100 mesh (0.0059 inch) having an initial sulfur content of 1.13 percent (0.57 percent pyritic, 0.56 percent organic sulfur) was beneficiated to a final sulfur content of 0.8 percent, with a combustible recovery of over 92 percent [7]. Precombustion recovery of sulfur decreases the amount of SO_2 emitted which would result if the original feed coal had been directly combusted without significant emission control.

One environmental intrusion from this coal refinery would be the oil content in the tailings stream. The oil content can approach 12 milligrams per liter of solution (1×10^{-5} lbs oil per lb solution) [2]. To avoid the continuous buildup of oil in the slurry water, the system would have to be periodically blowndown, with the release presumably sent to a wastewater treatment to remove immiscible and dissolved oils, and any dissolved sulfides.

Research Needs

The success of the oil agglomeration step is heavily dependent on proper selection of bridging liquid. Further study of coal/heavy oil surface chemistry, as it pertains to oil agglomeration of coal, may help to predict how a given coal and heavy oil will behave when processed using the technology of this coal refinery.

Further process development work at a level approaching commercial operation would establish the economic viability of this coal refinery concept sited within this country.

Table 1: Agglomeration Tests Using Different Coals

COALS TESTED	Wyodak subbituminous	Kemmerer subbituminous	Blind Canyon bituminous	Texas lignite
BRIDGING LIQUID (weight percentage)				
Constituents	Heavy oil + Diesel oil (1:1)	Bitumen + Diesel oil (1:1)	Maja oil + Diesel oil (1:1)	Coal-derived tar
Quantity (dry basis)	14.9	20.8	13.9	17.4
FEED COAL (weight percentage)				
Moisture content, (as received)	21.2	17.7	4.4	22.3
Moisture capacity	29.3	20.6	5.2	25.5
Ash content (dry basis)	7.2	5.4	15.9	26.9
Heating Value (Btu/lb, as received)	9,220	10,220	11,630	7,020
AGGLOMERATE PRODUCT (weight percentage)				
Moisture content, after air drying	3.9	4.5	Not Given	Not Given
Moisture capacity	21.6	14.0	Not Given	18.3
Ash content (dry basis)	5.4	3.6	6.5	18.8
Recovery of Combustibles	100.0	99.0	95.9	95.7
Heating Value (Btu/lb, air dry)	11,740	12,780	13,980	10,810

Table 2: Comparison of Oils Distilled from Original Bridging Liquid with Oils Recovered After Heat Treatment of Agglomerates at 660°F

BRIDGING OIL	Bitumen + Diesel (3:2)		Bitumen + Diesel (4:1)	
Treatment	Standard Distillation[(a)]	Oil from Agglomerates[(b)]	Standard Distillation[(a)]	Oil from Agglomerates[(b)]
Yield (weight percent)	74.1	64.6	62.0	53.6
Density (g/ml at 60°F)	0.8920	0.8973	0.9126	0.9156
API Gravity[(c)]	27.1	26.1	23.5	23.1
PROXIMATE ANALYSIS (weight percentage)				
Carbon	87.1	85.9	85.6	85.7
Hydrogen	12.2	11.8	11.9	11.7
Sulfur	1.2	1.0	1.9	1.6
Nitrogen	291 ppm	244 ppm	505 ppm	329 ppm

Standard Distillation[(a)]:	fraction of the original bridging oil mixture that boils below 977°F
Oil from Agglomerates[(b)]:	fraction of original oil used in agglomerates formation recovered during thermal treatment
API Gravity[(c)]:	term used in the petroleum industry to report the density of petroleum products

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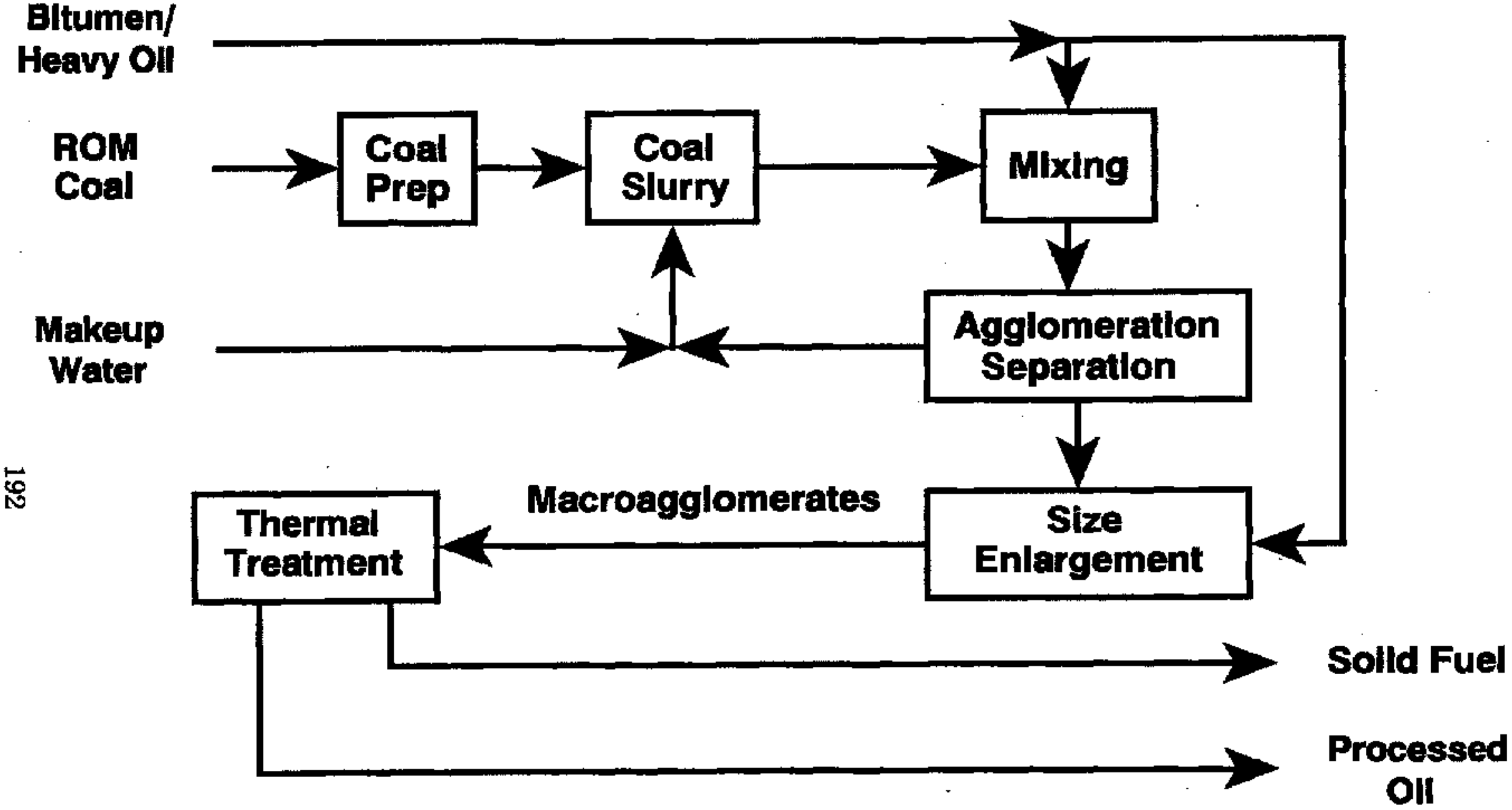

FIGURE 1: Overall Block Diagram of the Agflotherm Concept

CATALYTIC TWO-STAGE LIQUEFACTION

Introduction

The Catalytic Two-Stage Liquefaction (CTSL) process converts coal by direct liquefaction, with direct liquefaction defined to be the reaction of coal with hydrogen [1]. The coal is slurried with a process-derived solvent that donates hydrogen to the coal. The conversion step is typically carried out at less severe conditions (temperature, pressure) for a two-stage process than for a single-stage, resulting in greater liquid yields and more efficient hydrogen utilization. Higher temperatures lead to increased liquefaction and upgrading reaction rates, but the greater degree of thermal cracking (breaking larger molecules into two or more smaller molecules due to the application of heat) leads to excessive hydrocarbon gas formation which increases hydrogen consumption and is unfavorable for making a hydrogen-donor solvent. The two-stage process is designed to control the reaction rates of hydrogenation, cracking, and liquefaction and improve the quality of the hydrogen-donor solvent.

The CTSL process essentially liquefies coal by solubilizing and cracking hydrocarbons in the coal using a catalytic ebullated-bed (see discussion on Coal/Oil Coprocessing for more details) first-stage reactor with the coal-derived liquids that are produced subsequently hydrogenated (addition of hydrogen within the macrostructure) to form distillable liquids in the second-stage reactor. The coal-derived liquid product can then be upgraded to premium transportation fuels either by conventional petroleum refinery operations or by coupling in-line hydrotreating (improving oil quality through removal of sulfur, nitrogen, oxygen and metals, in addition to cracking heavy carbonaceous materials to lower boiling, more-valuable products) within the process. The goal of direct liquefaction is to convert coal from a heterogeneous, combustible solid to a clean, liquid fuel that would rival (petroleum-based) oil in utility and flexibility [2].

The coal liquids are typically more aromatic in nature than petroleum liquids and those which are in the naphtha boiling range (less than 350°F) possess excellent reformability (i.e., containing molecules in the gasoline range with high octane numbers such as methyl cyclohexane, etc.) characteristics so that a high octane unleaded gasoline can be produced [3].

Direct liquefaction has as its basis the discovery by Friedrich Bergius in 1913 that treating coal with hydrogen at high temperature and pressure in the presence of a catalyst produced a liquid similar to crude petroleum. Germany during World War II applied the Bergius process to produce up to 25.5 million barrels of synthetic petroleum per year from coal. A review of the direct liquefaction concept up to 1990 is given in reference [4].

The CTSL process is a variation on the Integrated Two-Stage Liquefaction (ITSL) technology [5], but with the deasher unit placed after the second reactor and both reactors operating at similar pressures so that closer-coupling of the system is achieved.

The coal refinery based on the CTSL process is shown in Figure 1 [3]. The system would integrate the generation of the hydrogen and steam requirements together with the production of chemical feedstocks. The necessary inputs to this coal refinery include run-of-mine coal, air, natural gas, electricity, and water, while major products would include liquid hydrocarbon fuels (unleaded gasoline, No. 2 diesel, utility turbine fuel) and potentially LPG (propane). Sulfur and ammonia would be byproducts of this coal refinery.

Detailed Process Description

The CTSL process is a modern version of direct liquefaction, which uses moderate temperatures and pressures to convert coal into coal-derived liquids. This process would also include upgrading of the whole liquid product from the Liquefaction section (typically in the naphtha and middle distillate boiling ranges) to produce end-use products such as gasoline and diesel fuel. A detailed flow diagram is given in Figure 1. The feed coal is taken to be a Wyoming subbituminous coal (Clovis Point Mine), with a higher heating value of 11,351 Btu per pound (dry basis) and an approximate sulfur content of 1.1 percent by weight (see Table 1 for more details). It should be noted that this concept utilizes the propane as a plant fuel gas for internal plant consumption; another option may be to separate the propane from the other light fuel gases (methane, ethane, etc.) for potential utilization as a marketable product. In addition, the fuel gas is utilized for steam production with coal potentially required for hydrogen production; another option is to combust the coal in an atmospheric fluidized bed combustor (AFBC) for steam generation, while using the clean (sulfur-free) fuel gas for the hydrogen requirement. The ultimate use of the propane and fuel gas streams would most likely depend on the overall efficiency and economics.

In the Coal Preparation section, run-of-mine coal is prepared by crushing to achieve a particle size in the range of 200 mesh (0.0029 inch) [6] to 30 mesh (0.0234 inch) [3]. The moisture content of the run-of-mine coal is greater than 5 weight percent so that drying is required, which will involve heating the coal using hot combustion gases. No further coal preparation for impurity removal is required, as the coal-bound nitrogen and sulfur are converted to ammonia (NH_3) and hydrogen sulfide (H_2S) in the Liquefaction section and later removed, and the ash will be retained in the ash concentrate stream from the Deashing unit.

The prepared coal is then fed into the Liquefaction section, for which the unit operations are shown in block diagram form in Figure 2 and the individual subprocesses in Figure 3 [7]. The crushed and dried coal is slurried with the recycle process derived solvent in the Slurry Mixer, as shown in Figure 3. The recycle process derived solvent includes the stream originating from the Flash Separation system, the hydrotreated resid (which contains some ash and unconverted coal), a portion of the hydrotreated product solvent, and the deashed hydrotreated resid (DHTR). This slurry is then mixed with hydrogen and preheated before entering the 1st-Stage (catalytic) Reactor.

As stated previously, the 1st-Stage reactor is an ebullated-bed, in which catalyst (typically alumina-supported transition metals [6]) with an average reactor temperature of 750 to 810° F and an inlet hydrogen partial pressure of 2,300 to 2,500 psig [6]. The 1st-Stage Reactor carries out the thermal dissolution of the coal, with a high degree of coal conversion (up to 90 percent) into liquid and gaseous products [6]. The products from the reactor which are gases, distillate, resid, unconverted coal, and ash pass directly to the second reactor.

The purpose of the 2nd-Stage Reactor is to further upgrade the coal-derived liquids through catalytic hydrogenation, and removal of the remaining heteroatoms. The first-stage product is mixed with a preheated hydrogen stream before entering the 2nd-Stage Reactor. The second-stage products are separated into gases, distillate, recycle solvent, and hydrotreated resid (containing ash and unconverted coal) feed for the Deashing unit. The distillate is to be fed into the Product Upgrading section for further treatment to generate the major products of this coal refinery concept.

The relationship of the temperature of the 2nd-Stage Reactor with respect to the first-stage will depend upon the design: the Hydrocarbon Research, Inc. design has the temperature of the second reactor higher than the first, to promote resid hydrocracking in the second reactor while generating a recycle solvent [8], while the design at the Wilsonville Advanced Coal Liquefaction Facility has the temperature of the first reactor greater than the second so that most of the thermal cracking occurs in the first reactor with solvent hydrogenation in the second reactor [1].

The Deashing unit separates the hydrotreated resid into an ash concentrate stream (containing unconverted coal, ash, and some residuum) and a solids-free residuum liquid stream which becomes part of the recycle process derived solvent used for slurrying the prepared coal.

The coal-derived liquid from the Liquefaction section is catalytically hydrotreated (breakdown of large molecules to lower boiling ones while increasing the hydrogen content) at high severity in the Product Upgrading area or by in-line hydrotreating (not used in this case). The hydrotreated product is then fractionated (separated) to recover light naphtha (C_5 to 180° F), heavy naphtha (boiling point of 180 to 350° F), middle distillate (350 to 650° F) and heavy gas oil (boiling point above 650° F). The light naphtha is suitable as a blending component of premium gasoline, while the middle distillate goes to diesel fuel, and heavy gas oil to turbine fuel. The heavy naphtha is catalytically reformed (conversion of molecules in the gasoline boiling range to others with high octane) with the high octane reformate blended to gasoline.

The raw gas streams from the Liquefaction and Product Upgrading sections are cleaned in the Acid Gas Removal section and separated into propane (C_3), butane (C_4), 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 steam requirements, while the butane is recycled to the Product Upgrading section for blending to gasoline. Natural gas is used to satisfy the overall coal refinery steam requirement.

The hydrogen that is unconverted in the Liquefaction section is recovered and recycled back into the process. The makeup hydrogen requirement for the CTSL process is met by utilization of the ash concentrate stream from the Deashing unit using noncatalytic partial oxidation and by steam reforming of natural gas, both of which are technologies that have been applied in conventional petroleum refining [9]. The ash concentrate is gasified at elevated temperatures and pressures in the presence of oxygen and steam. The primary products of gasification are H_2, CO, CO_2, and H_2O. The hydrogen content is catalytically increased at lower temperature by application of the shift conversion reaction to convert CO to H_2:

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

with the CO_2 removed from the product gas to give hydrogen of 95 to 98 percent purity. A solid waste containing the ash content of the coal results from the partial oxidation unit which will be handled in an environmentally responsible manner. The natural gas (mainly methane) and steam are converted to hydrogen by the reverse of the methanation reaction:

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

followed by shift conversion of CO to H_2 and removal of CO_2.

The oxygen required for the gasification of the ash concentrate is generated by the Air Separation plant, with excess nitrogen available for export as a marketable product, if warranted by the process economics. Otherwise, the excess nitrogen is vented to the atmosphere.

The water-soluble waste streams from all process units are collected into a sour water stream, which is then treated by conventional means (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 waste water is further treated (in a phenolsolvan unit) to remove dissolved phenols, which can then be sold as a byproduct or utilized for steam generation. Another option to decrease the amount of waste water treated is to use a portion of the waste water to slurry the ash concentrate stream.

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 Sulfur Removal area (typically a Claus® unit). Here the H_2S stream is converted to elemental sulfur which will be sold as a byproduct.

Types of Feed Coal

The CTSL process is a variation of direct coal liquefaction, for which it has been established that bituminous, subbituminous, and lignitic coals may be converted to liquids [10]. It would be expected that anthracitic coal would not be used as it tends to produce greater volumes of hydrocarbon gas during direct liquefaction, leading to greater consumption of expensive hydrogen. Some of the coal types for which CTSL process research has been pursued are given in Table 2 [1, 6, 7, 11], and range from low to high in ash and sulfur content. Research with lignitic coal has also been performed [6].

In general, high volatile bituminous coals give the highest liquid yields, with distillate yields of up to 78 percent, while yields with subbituminous coal are in the range of 62 percent (see Table 3, [1, 6]). Comparison of the product distribution for the Wyoming subbituminous with an Illinois No. 6 bituminous coal (given in Table 4, [1, 3]) indicates that a higher coal conversion and a greater percentage of lighter hydrocarbon oils is generated for the more volatile bituminous coal. The equivalent liquid yield for a moisture-ash free bituminous coal (such as Illinois No.6 and Ohio No. 6) is approximately 5 barrels per ton coal [1]. Research in deep-cleaned (mineral matter less than 10 percent) coals has been performed to improve distillate yield and reduce energy rejection, with results indicating that coal conversion decreases with increasing feed coal ash content [6].

The overall yield and distribution of liquid products will depend on the degree of hydrogenation within the thermal liquefaction reactor, which itself is a function of the severity of operating conditions such as temperature, pressure, catalyst type, catalyst age, catalyst charge and withdrawal, and coal feed rate [12].

Products

The main products from this coal refinery are premium unleaded gasoline, No. 2 diesel fuel, and utility turbine fuel. Marketable byproducts include elemental sulfur and anhydrous ammonia. The product output is shown in Table 5 in terms of pounds product per pound of feed coal (dry basis). This analysis indicates that for the feed rates given in Figure 1 for a Wyoming subbituminous coal that 1.3 barrels of premium gasoline, 2.4 barrels of diesel fuel, and 0.1 barrels of turbine fuel can be produced from one ton of feed coal (dry basis).

Likely Applications

The premium grade unleaded gasoline has a research octane number (RON) of 95, making it suitable for blending in existing gasoline stocks. The physical properties of the diesel product from this coal refinery compare favorably with the requirements for a commercial grade No. 2 diesel fuel oil (as shown in Table 6 [3, 11, 13]). The turbine fuel product (°API of 17; sulfur content of 0.1 weight percent; nitrogen content of 0.2 to 0.4 weight percent) from this coal refinery concept can potentially be used as a heavy gas turbine fuel oil.

Given that the product output (comprising mainly transportation fuels) and certain processes (i.e., the Product Upgrading area) of this coal refinery concept closely resembles that of a conventional petroleum refinery, it would be expected that the principal commercial entity interested in pursuing this concept would be the petroleum industry.

Status of Development

The development of the Catalytic Two-Stage Liquefaction (CTSL) process started in 1985 with all process development unit (PDU) programs using two catalyst stages.

The Wilsonville Advanced Coal Liquefaction Facility's 6 ton per day PDU was modified in 1982 to run in the ITSL (integrated two-stage liquefaction) mode, by using an H-Oil [14] ebullated bed hydrotreating unit for the second-stage reactor. The deasher was then placed after the second reactor (whereas before it was located between the first and second stages) and with both reactors operating at similar pressures, the Wilsonville arrangement became a close coupled system, called the Close-Coupled Integrated Two-Stage Liquefaction (CC-ITSL) process. Wilsonville prefers to have most of the thermal cracking occur in the first reactor with solvent hydrogenation in the second, so that the temperature of the first reactor (800 to 820°F) is greater than that of the second (760 to 795°F) [1, 4].

Hydrocarbon Research Inc., (HRI) in 1982 initiated the development of the CTSL process, by operating a catalytic system with lower conversions in the first ebullated bed (at 750°F) and the second reactor operating at about 800°F to hydrocrack the residuum stream to make an aromatic process solvent for recycle and coal slurrying. The hydrocarbon gas yield was lowered by 50 percent (liquid-to-gas ratio of 12 to 1, [15]), leading to lower hydrogen consumption. A different catalyst type (Ni-Mo) was also used for the hydrogenation of the residuum, to maximize conversion.

In 1985, Wilsonville was converted to a Close-Coupled Catalytic Two-Stage Liquefaction (CC-CTSL) mode, by installing a second ebullated bed reactor [16].

Recent research work has centered on the variation of the severity of operating conditions [11], evaluation of different catalysts [11], catalyst deactivation [7], and the use of cleaned coals [6].

Sensitivity analyses have been performed on the overall process economics as a function of coal type, production rate, and favourability of different processes (e.g., hydrogen production by steam reforming of natural gas versus partial oxidation using coal; extent of deashing) in references [1] and [3].

Environmental Aspects

The CTSL process can theoretically convert high-sulfur, high-nitrogen coal into a suite of liquid fuels that are low in sulfur and nitrogen. The majority of the sulfur in the coal is converted into H2S gas, which is subsequently reacted to form elemental sulfur, while the nitrogen in the coal is converted into ammonia, avoiding the large scale generation of SO2 and NOx if the feed coal had been directly combusted.

The main environmental intrusions from this coal refinery would include atmospheric emissions of SO2 and NOx, and solid wastes. The extent of the atmospheric emissions will depend in part on whether coal is used for steam and/or hydrogen generation, as opposed to use of cleaner, but more expensive, natural gas. It would be expected that the SO2 and NOx emission rates to be in compliance with the minimum level of control allowed by NSPS.

The majority of the solid waste would result from the gasification of the ash concentrate, with the slag (ash) from the coal as the residue (in this case, 0.09 lb per lb feed coal, dry basis). The slag has been judged nonhazardous [17]. Other solid wastes would be typical of a conventional petroleum refinery, such as sludges and solids recovered from waste treatment processes.

Carbon dioxide, CO2, is produced by the partial oxidation of the ash concentrate, steam reforming of natural gas to generate hydrogen, and from the Liquefaction and Product Upgrading processes. Approximately 0.7 pounds of CO2 are produced per pound of feed coal [3], with the majority (over 90 percent) due to hydrogen generation. The CO2 generated during hydrogen production (~ 0.65 lb per lb feed coal) theoretically can be captured during purification of the resulting hydrogen stream, thereby potentially becoming a marketable byproduct. The majority of the CO2 produced in the Liquefaction and Product Upgrading processes (~ 0.05 lb per lb feed coal) would collect in the Sour Water stream and therefore be vented to the atmosphere.

Research Needs

Direct coal liquefaction has the potential of converting coal to liquid fuels at a cost of $30 per barrel. If this conversion cost is achieved through pilot plant and engineering studies, then engineering and new concepts could be tested on a new 50 to 200 pound per day process development unit (PDU) on a continuous-flow integrated basis at a scale smaller than the Wilsonville 6 ton per day PDU.

This concept used pulverized run-of-mine coal as the feed to the liquefaction plant. Integration of advanced coal cleaning techniques to reduce the mineral matter content with the above processes has been assessed [18]. The potential economic benefits that may accrue from integration of advanced coal cleaning with the CTSL process may warrant further investigation.

A major proportion of the cost of a liquefaction plant is due to hydrogen production. The cost of coal liquids would be reduced if hydrogen could be generated at a lower cost or if the hydrogen consumption could be reduced by making less hydrocarbon gas.

The liquefaction of coal is directly related to the effectiveness of the catalysis process. Novel unconventional catalyst systems may increase the performance above that seen for conventional supported catalysts.

Table 1: CTSL Process Feed Coal Analysis

ULTIMATE ANALYSIS (weight percentage, dry basis)					
Carbon	Hydrogen	Oxygen	Sulfur	Nitrogen	Ash
65.36	4.81	19.0	1.13	0.92	8.78
Free Moisture (weight percent)			30.0		
Higher Heating Value (dry basis)			11,351 Btu per lb		

Table 2: Analysis of Feed Coals Used in Research

ULTIMATE ANALYSIS (weight percentage, dry basis)			
COAL	Illinois No. 6 Bituminous	Ohio No. 6 Bituminous	Wyodak Subbituminous
Carbon	69.9	71.4	63.1
Hydrogen	4.9	4.7	4.5
Oxygen	17.3	7.7	18.7
Sulfur	3.7	3.2	0.9
Nitrogen	1.4	1.4	1.1
Ash	11.7	11.6	11.8
PHYSICAL PROPERTIES			
Free Moisture, weight percent	4.9	9.9	29.8
Heating Value, Btu/lb dry	12,300	13,500	11,700

Table 3: Component Yield as a Function of Type of Coal

YIELD (weight percentage; moisture, ash-free basis)			
COAL	Illinois No. 6 Bituminous	Ohio No. 6 Bituminous(a)	Wyodak Subbituminous
C_1 to C_3	7.4	8.3	11
C_4 plus Distillate	66.5	68.7	60
Resid	1.4	7.3	2

Ohio No. 6 Bituminous(a): Coal cleaned to an ash content of 6.2 percent

Table 4: Product Distribution as a Function of Type of Coal

YIELD (weight percentage, dry basis)		
COAL	Wyoming Subbituminous	Illinois No. 6 Bituminous
Light Gases (C_1 to C_3)	7.3	6.6
C_4 to 390°F	17.4	18.2
390 to 650°F	27.8	32.6
650 to 975°F	10.9	16.4
975°F plus oil	4.2	12.6
PROCESS PERFORMANCE (weight percentage; moisture, ash-free basis)		
C_4 to 975°F	56.1	67.2
Coal Conversion	90.1	94.8

Table 5: Product Output from Example CTSL Coal Refinery

PRODUCT	OUTPUT (lb product per lb moisture-free coal)
Premium Gasoline	0.17
Diesel Fuel	0.37
Utility Turbine Fuel	0.02
Sulfur, elemental	0.01
Ammonia, NH_3	0.008
Nitrogen, N_2 (*)	0.42

(*) Nitrogen can be a potential byproduct, if warranted by the process economics

Table 6: Comparison of Properties of Coal-Derived Diesel Product with Commercial No. 2 Diesel Fuel Oil(a)

FUEL TYPE	Boiling Range (°F)	°API	Sulfur (weight %)	Cetane No.	Nitrogen (weight %)
Diesel Product	350 to 650	32	0.1	> 40	0.1 to 0.2
Commercial No. 2 Fuel Oil(a)	540 to 640	36	0.5 (maximum)	40 (minimum)	----

(a) Derived from petroleum

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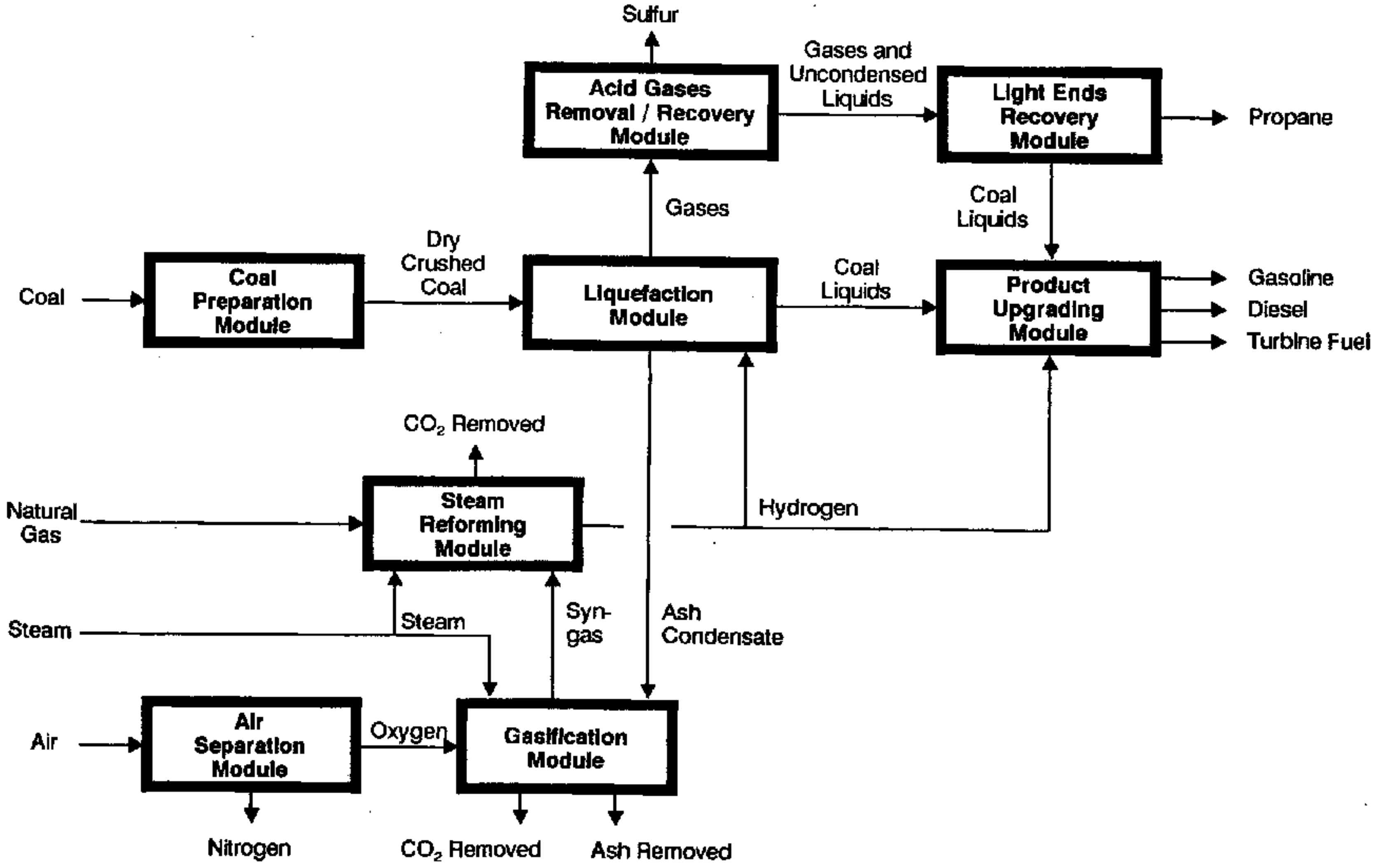

Figure 1: Overall Block Diagram of CTSL Concept

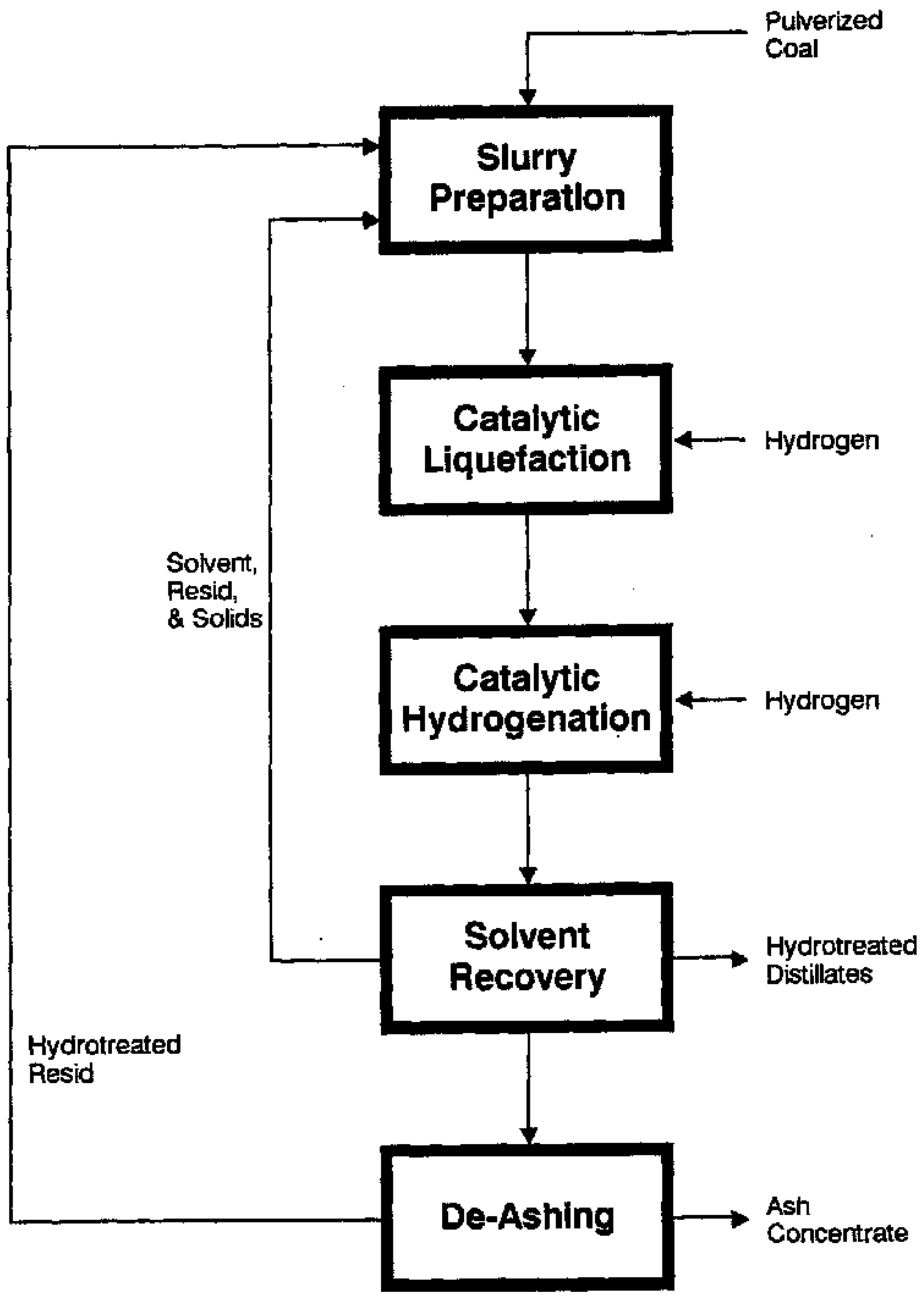

Figure 2: Unit Operations of the CSTL Concept

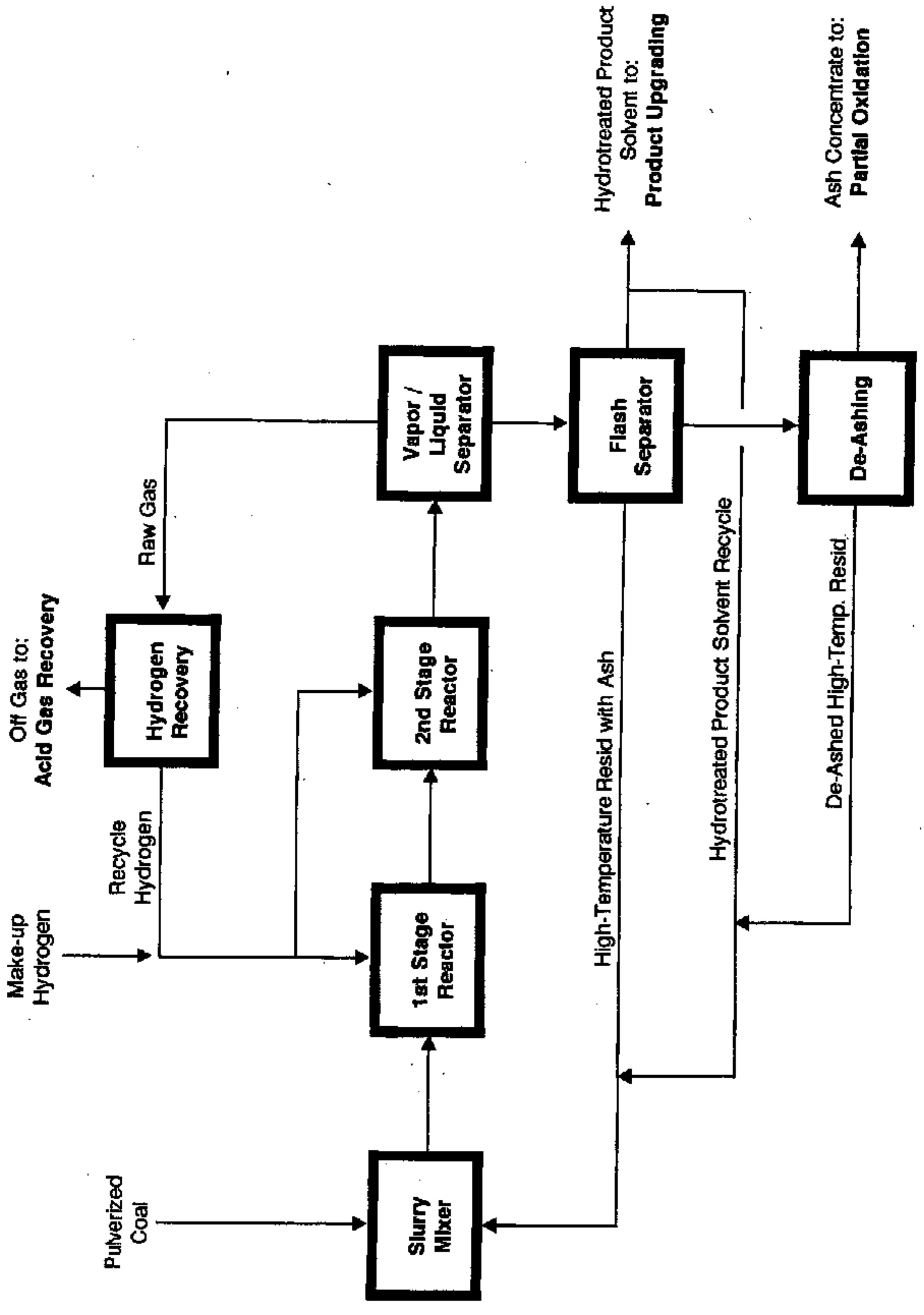

Figure 3: Schematic Flow Diagram of the CSTL Liquefaction Module