

COPRODUCTION OF SNG, ELECTRICITY, METHANOL, AND CHEMICAL INTERMEDIATES

Introduction

The coproduction of SNG (substitute natural gas), electricity, methanol (CH_3OH), and chemical intermediates is an example of a coal refinery within the Gasification category. Natural gas provides nearly one-fourth of this Nation's energy and supplies nearly half of all end-use energy consumed in U.S. homes [1]. In the past, a form of fuel gas (called "manufactured" or "town" gas) was produced from coal. At its peak in the 1920 to 1940 period, coal carbonization was employed to produce "manufactured" gas in thousands of plants, throughout the world. The availability of petroleum oil products and natural gas at low prices after World War II led to the decline of this technology. However, serious shortages in natural gas in the early 1970's resulted in a revival of interest in alternative methods of natural gas production from coal.

This coal refinery concept is an alternative that integrates the generation of synthesis gas using Lurgi gasification technology, with on-site power generation utilizing the coal fines. The synthesis gas stream is subsequently converted to SNG and methanol. The necessary inputs to this coal refinery include run-of-mine coal, air, and water, while major products would include SNG, methanol, and electricity. Sulfur, anhydrous ammonia and liquid nitrogen would be marketable byproducts of this coal refinery. This concept is very close to the process at the Great Plains Coal Gasification Plant at Beulah, North Dakota. However, the power plant is not on the SNG site, nor is it dependent on the SNG process. Also, only limited methanol make-up is produced at Great Plains at the present time, for the Rectisol[®] acid gas removal system in use there.

Process Description

The flow diagram for this concept is given in Figure 1, with all streams based on 100 pounds of run-of-mine coal fed. The inlet coal is taken to be North Dakota lignite, with a higher heating value of approximately 7,000 Btu per lb (as received) and an as-received sulfur content of 0.68 percent by weight (see Table 1 for more details). Production of synthesis gas is by high-pressure Lurgi moving-bed gasification process because it is said to generate the highest percentage of methane in the product synthesis gas of all commercially available gasification technologies [2]. Pressurized coal gasification is preferred for methanol production [3] to minimize energy consumption per unit mass of methanol produced due to compression costs (the methanol synthesis step is typically carried out at pressures of 500 to 900 psig).

In the Coal Preparation section, the run-of-the-mine coal is crushed to produce the desired particle size (typically less than 2-inches). Advanced coal preparation methods to decrease the ash or moisture content are not required. The fines (particle

size less than ¼-inch) generated during the grinding process are separated from the larger coal particles and are to be combusted in the Advanced Power Generation section; the Lurgi gasifier has a limited tolerance for coal fines, as they tend to be entrained out of the reactor before reacting. The remaining sized coal (approximately 64 weight percent of the total coal feed) is sent to the Gasification section.

Sized coal is fed into the Gasification section where it exothermically (gives off heat) reacts with oxygen and steam. Steam is also required to moderate the temperature and avoid ash sintering. The reactive gases pass through the bed in a countercurrent fashion to the slowly descending coal, both devolatilizing and drying the incoming coal. Coal is heated very rapidly and converted to a combustible gas (composition shown in Table 2) and a residual ash by partial oxidation with steam and oxygen. The high steam-to-coal ratio required to prevent ash sintering and the relatively low gasifier temperatures produces a gas with high concentrations of CO₂, H₂, and CH₄ in comparison with entrained-bed gasification which produces a CO-rich gas. Appreciable amounts of condensable tars and tar oils byproducts are also formed. The raw synthesis gas exits the top of the Lurgi gasifier at approximately 850°F and 425 psig [4]. The ash is removed at the bottom of the gasifier through a lock hopper arrangement and quenched with water.

The required oxygen stream for the gasifier is produced from air in the Air Separation plant. Nitrogen in excess of process requirements is stored on-site as a liquid for sale as a marketable byproduct.

Downstream from the gasifier, the raw synthesis gas stream is split into two streams, with 33 percent sent for shift conversion. In the Shift Conversion area, carbon monoxide reacts with steam in the presence of a catalyst to form hydrogen and carbon dioxide:



to increase the ratio of hydrogen-to-carbon monoxide. The cobalt molybdenum-based shift conversion catalyst can operate at high temperatures without degradation and is not poisoned by the low levels of sulfur compounds in the synthesis gas. This leads to increased thermal efficiency of the process as acid gas cleanup (which requires cooling of the gas stream) can proceed after shift conversion.

During gasification, the inorganic constituents (sulfur, nitrogen) react to form a series of reduced sulfur compounds [primarily hydrogen sulfide (H₂S) and to a lesser amount, carbonyl sulfide (COS) and carbon disulfide (CS₂)] and ammonia (NH₃). These impurities as well as CO₂ have to be removed so as to comply with environmental legislation requirements and also to avoid poisoning of the catalysts used in methanol and methane synthesis. Technology for high-temperature, high-pressure synthesis gas cleanup is not commercially available so that the high-temperature synthesis gas must

be cooled before cleanup. Both synthesis gas streams pass through the Gas Cooling area, where the tars, tar oils and water condense and separate from the synthesis gas. The Acid Gas Removal section consists of a Rectisol® wash unit.

The Rectisol® process employs cold methanol (at about -45° F) to physically absorb the acid gases (H₂S, COS, CO₂) and so reduce the content of H₂S and COS to less than 1 ppm and CO₂ to less than 30 ppm. Also removed are liquid hydrocarbons such as naphtha. The acid gases which are extracted by the Rectisol® process from the raw synthesis gas are sent to the Sulfur Recovery section, namely a Stretford unit for conversion of the hydrogen sulfide to elemental sulfur. The Stretford process is used in the Great Plains gasification plant. In the Stretford process, hydrogen sulfide is absorbed in a liquid solution, oxidized to elemental sulfur and removed from the solution. Approximately 99 percent of the hydrogen sulfide can potentially be recovered by this process. The exit gas from the Stretford unit contains a small percentage of hydrocarbons (about 4 to 5 percent) and CO₂, and is combusted to generate steam.

A portion of the purified synthesis gas from the Rectisol® unit is diverted to generate methanol for make-up of losses in the Rectisol® unit, with the remainder sent for methanation. The hydrogen-to-carbon monoxide mole ratio of the purified synthesis gas stream from the Rectisol® unit is approximately 3:1, the minimum needed for methanation.

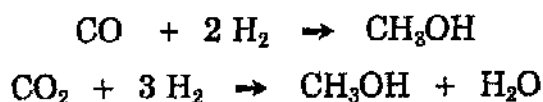
In the Methanation section, the H₂ and CO in the purified synthesis gas are combined catalytically in a series of fixed bed reactors, using a pelletized nickel catalyst, to produce methane, CH₄, and water:



The above reaction is quite exothermic and the heat generated by methanation is used to produce a large proportion of the steam requirement for the gasification reaction.

The majority of the water in the wet methanated gas is removed by cooling and glycol dehydration. The resulting product gas is sent to the Compression unit where it is compressed into pipeline quality SNG with a heating value of 970 Btu per standard cubic feet. Typical constituents of the SNG product stream are shown in Table 3.

The purified synthesis gas stream sent for methanol production first passes through a final catalytic absorption stage (a carbonyl/sulfur guard bed) in order to ensure a high-quality synthesis gas as well as to prevent the breakthrough of sulfur. The catalytic conversion of CO and H₂ to methanol is a very exothermic reaction, and involves the following reactions [5]:



The aqueous stream from the Gas Cooling area contains tars and tar oils, which are removed from the water in the Gas Liquor Separation area. The process water from the Gas Liquor Separation area is filtered and treated in the Phenosolvan unit to remove any dissolved organics such as phenolic compounds. The Phenosolvan process adds solvents such as isopropyl ether or butyl acetate to the process water, to attract phenols or other organics. The solvent and organics are then separated by distillation. The tar, tar oils, naphtha, and phenolic compounds could be refined into marketable chemicals but for this coal refinery are instead used to supply the steam requirement for this coal refinery. The ammonia within the process water from the Phenosolvan unit is removed for sale as a marketable byproduct.

The fines from the Coal Preparation section are combusted in the Pressurized Fluidized-Bed Combustor (PFBC) to generate electricity. An advantage of a pressurized fluidized-bed combustor is that it allows combined-cycle operation, generating power from both steam and the combustion gases. Also, additional pollution control equipment for SO_2 and NO_x is not required, due to the relatively low operating temperature (around $1,600^\circ\text{F}$) which limits formation of thermal NO_x . In fluidized-bed combustion, coals burn in a bed of rapidly moving solid particles suspended (i.e., fluidized) by an upward flow of air and combustion gases [7]. The sulfur in the coal is converted into SO_2 and reacts with the sorbent (limestone, CaCO_3 , or dolomite, $\text{MgCO}_3 \cdot \text{CaCO}_3$) injected into the PFBC to form inert calcium sulfate, CaSO_4 , a dry granular material. Reductions in the range of 90 to 95 percent of the SO_2 generated can be achieved. The combustion gases are cleaned of particulates, alkali, and other contaminants, and then expands through the Gas Turbine section. Approximately 23 percent (0.07 kWh per pound of feed coal) of the total electricity is produced in the Gas Turbine section.

The combustion gases exiting the Gas Turbine section contain significant energy that is used to generate steam in the Heat-Recovery Steam Generator (HRSG). Additional steam is also produced from heat exchangers located within the PFBC. This steam is used to generate electricity (0.23 kWh per pound of feed coal) in the (conventional) Steam-Turbine cycle.

Types of Feed Coal

Lurgi gasifiers have the ability to handle most coals, ranging from lignite to high-volatile bituminous. U.S. coals which have been successfully tested in large-scale Lurgi gasifiers include bituminous coals such as high-ash high-sulfur Pittsburgh No. 8, high-sulfur Illinois No. 6 and high-sulfur Kentucky No. 9, high-moisture low-sulfur subbituminous coals such as Powder River Basin, and high-moisture low-sulfur lignitic coals such as Texas and North Dakota [2].

Desirable coal characteristics include low free-swelling index, high ash softening temperature, and high coal reactivity. Low-rank coals from North Dakota, New Mexico, Texas, Montana, and Wyoming are reported to be well-suited for gasification by the Lurgi process [2].

The moving-bed technology used in Lurgi gasifiers typically have inherent problems handling coal fines [2]. Excessive fines in the feed coal leads to entrainment of the fines in the exiting synthesis gas stream and ultimately combining with the tars and tar oils. Generally, Lurgi gasifiers are operated with coal feeds containing less than 10 percent by weight of fines smaller than 1/4-inch.

Products

The main products from this example coal refinery are substitute natural gas (SNG), electricity, methanol (CH_3OH), and chemical byproducts. Marketable byproducts include (elemental) sulfur, liquid nitrogen, and anhydrous ammonia (NH_3). The product output is given in Table 4 in terms of pound of product per pound of feed coal. This analysis indicates that 2.5 cubic feet of SNG, 0.01 pounds of methanol, and 0.3 kWh of electricity are produced from one pound of feed coal.

The product output is dependent on the type of feed coal (as the amounts of different constituents in the feed coal determines the distribution of the synthesis gas, in addition to the characteristics given in the above section), the type of gasifier (if a Lurgi gasifier is not used), and the degree of integration of the individual process steps.

Likely Applications

The main product of this coal refinery is SNG which can be used as supplemental pipeline gas supplies. The industrial fuel gas market sector can benefit from the introduction of SNG, as the potential market in this sector is large due to the widespread existing use of natural gas by the manufacturing industries. The SNG can also be supplied to the residential and commercial sectors. Thus, one commercial sector that may be interested in this coal refinery concept is the (natural gas) fuel supplier industry.

This coal refinery also offers the possibility of recovering the following organic compounds that are generated during Lurgi gasification: carbon dioxide, phenol, BTX (benzene, toluene, xylene), cresols, and creosote. The potential sale of these compounds will depend on the amount of associated capital investment and the degree of success of the required recovery techniques. This coal refinery can therefore be considered not only as a synfuels complex but also a petrochemical complex. The petrochemical industry may be interested in pursuing this coal refinery concept given the projected decreasing availability of petroleum crude oil.

Status of Development

The first commercial plant based on Lurgi gasification technology was built in 1936 [2]. As of 1983, 164 commercial gasifiers based on Lurgi technology were built in 15 plants for 12 different customers [2]. The largest application of Lurgi gasification technology is in the South African Coal, Oil, and Gas Corp., Ltd. (SASOL) facilities, which uses 90 Lurgi gasifiers to convert 66,600 tons of coal per day to 86,000 barrels per day of liquids.

The Great Plains Synfuels Plant, built in the early 1980's at Beulah, ND, uses 14 Lurgi gasifiers to produce approximately 160 million cubic feet per day of SNG from about 17,000 tons per day (TPD) of North Dakota lignite [8]. It is the only large-scale coal gasification plant in the U.S. that produces a high-Btu SNG from coal. The synfuels plant began operation in the summer of 1984, and currently coproduces approximately 70 TPD of ammonia and 50 TPD of sulfur. Research is being focused on the economical recovery of cresols, xylenols, creosote and argon, with a \$20 million plant planned to recover 22,000 ton per year of phenol [9].

Environmental Aspects

The main environmental intrusions from this coal refinery include atmospheric emissions of SO_2 and NO_x , and solid wastes. The majority of the sulfur in the coal is removed, leading to an emission rate for SO_2 in the range of 0.001 pound per pound of feed coal [4]. Nitrous oxides (NO_x) are not formed in any appreciable amount during gasification, since most of the nitrogen in the coal is converted into ammonia. Also, the relatively low temperature within the PFBC limits formation of thermal NO_x . The NO_x emission rate is less than 0.001 pound per pound feed coal [4]. The SO_2 and NO_x emission rates would be in accordance with the New Source Performance Standards (NSPS).

Coal gasification plants utilize water as an indirect cooling agent as a means to quench the ash and raw synthesis gas. Approximately 0.3 gallons of makeup water to the cooling system are required per pound of feed coal [4].

This coal refinery generates a variety of solid wastes including coal ash (comparatively inert), spent sorbent from the PFBC, tar and oil sludge, and spent sludge from wastewater treatment. The amount of ash generated from this coal refinery is about 0.06 pounds per pound of feed coal.

Research Needs

This coal refinery integrates mostly commercially available technologies. Detailed economic studies have been reported for individual portions of this coal refinery concept but not for the total coal refinery. Determination of the overall economics

would establish the set of conditions that could merit full scale production of SNG, methanol and electricity by this coal refinery concept.

Table 1: Feed Coal Analysis

CONSTITUENTS IN COAL (weight percentage, dry basis)						
Carbon	Hydrogen	Oxygen	Sulfur	Nitrogen	Chlorine	Ash
65.54	4.65	17.75	1.07	1.18	0.09	9.72
Free Moisture (weight %)			36.21			
Higher Heating Value (dry basis)			11,000 Btu per lb			

Table 2: Typical Lurgi Gasifier Product Gas Composition

COMPOUND	YIELD, mole percent, ash-free basis
Carbon Monoxide, CO	8.09
Hydrogen, H ₂	20.12
Carbon Dioxide, CO ₂	16.85
Methane, CH ₄	5.6
Higher Hydrocarbons, C ₂ to C ₄	0.43
Nitrogen, N ₂	0.04
Argon, Ar	0.03
Hydrogen Sulfide, H ₂ S	0.18
Carbon Oxysulfide, COS	0.005
Ammonia, NH ₃	0.52
Water, H ₂ O	45.54
Tar and Tar Oils	2.56

Table 3: Typical Product SNG Composition

CONSTITUENTS IN SNG (volume percentage, dry basis)				
Methane, CH ₄	Hydrogen, H ₂	Carbon Monoxide, CO	Carbon Dioxide, CO ₂	Nitrogen, N ₂
95.9	3.0	0.05	0.4	0.6
Heating Value		970 Btu per standard cubic foot		

Table 4: Product Output from Example Coproduction of SNG, Methanol and Electricity

PRODUCT	OUTPUT (per lb feed coal)
Substitute Natural Gas, SNG	2.5 scf
Methanol, CH ₃ OH	0.01 lb
Electricity	0.3 kWh
Sulfur, elemental	0.004 lb
Ammonia, NH ₃	0.005 lb
Liquid Nitrogen, N ₂	0.35 lb

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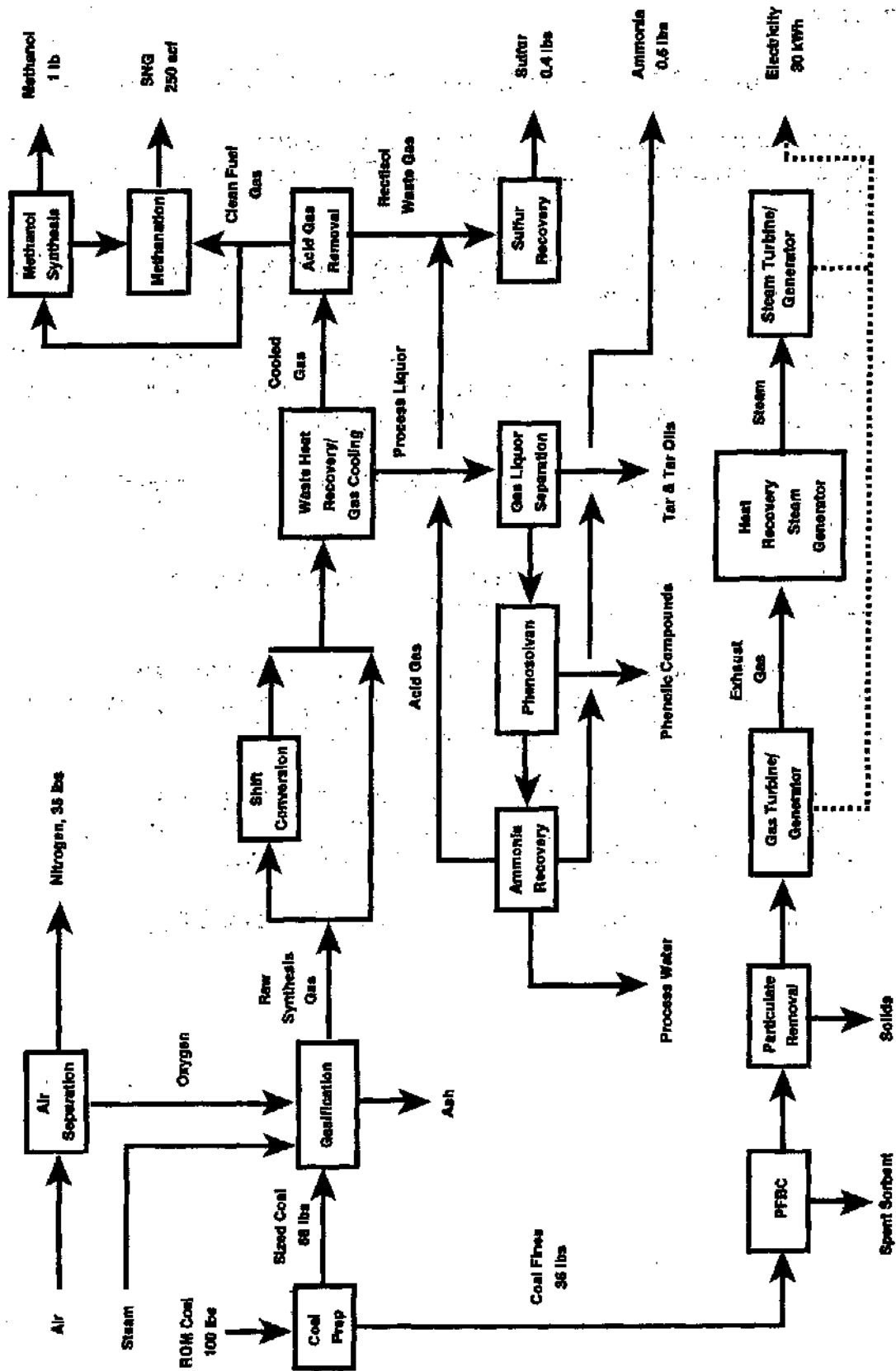


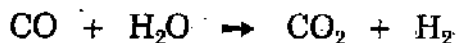
Figure 1: Overall Block Diagram for Coproduction of SNG, Methanol, Electricity, and Chemical Intermediates

COPRODUCTION OF UREA AND ELECTRICITY

Introduction

Coproduction of urea (NH_2CONH_2) and electricity is an example of a coal refinery in the Gasification category. At room temperature, urea is a colorless, odorless, and tasteless solid. During the period 1976 to 1980, urea became this nation's most important solid nitrogen fertilizer, with U.S. urea production more than doubling in one decade (1970 to 1980) [1].

This concept has been advanced by the Tennessee Valley Authority (TVA). A unique feature of this concept is the use of membrane technology to separate the synthesis gas into two streams, one hydrogen-rich and the other hydrogen-deficient. The hydrogen-rich stream is ultimately converted into urea. Another approach for urea production would be to convert the carbon monoxide in the synthesis gas to hydrogen by the water-gas shift reaction (see discussion on Dual Production of Ammonia and Electricity):



For every mole of hydrogen produced by water-gas shift, one mole of CO_2 is also generated. Process requirements for ammonia synthesis dictate that the carbon dioxide so produced should be scrubbed from the resulting gas stream. For power generation however, it is not necessary to remove any CO_2 , and so the overall efficiency of power production may potentially be greater for this coal refinery concept.

The coal refinery envisaged here is shown in Figure 1. This coal refinery is based on the coproduction of urea and electricity, with electrical generation by integrated coal gasification combined-cycle (IGCC). The necessary inputs to this coal refinery include run-of-mine coal, air (or oxygen), and water, while major products would include urea and electricity. Sulfur would be a byproduct of this coal refinery.

Detailed Process Description

The major process components of this coal refinery concept include a coal preparation step which results in a finely ground and slurried coal that is gasified to produce synthesis gas. Hydrogen is separated from the synthesis gas stream using membrane technology. The hydrogen first reacts with nitrogen to form ammonia, with the ammonia subsequently combined with CO_2 and converted to urea. The remaining portion of the synthesis gas stream is diverted to an integrated gasification combined-cycle power plant where a combustion turbine burns the synthesis gas to produce electricity and a steam turbine is powered by waste heat from the combustion turbine and the gasifier. Greater detail of the individual process components is given below.

In the Coal Preparation section, run-of-mine coal is first crushed and then sent to a wet grinding circuit where it is mixed with process water to form a coal slurry with a fine consistency. The slurried coal is pumped into the Gasification section.

In general, any second generation gasifier (e.g., Texaco, Shell, Dow) can be used for the coproduction of urea and electricity. However, the overall thermal and process efficiencies and the degree of synergism between the different process components will differ depending on the gasification technology. This coal refinery example is similar to TVA's experimental ammonia-from-coal facility [2, 3] and the commercial Ube Ammonia plant in Japan [14], both of which utilized a Texaco gasifier for the production of synthesis gas from coal. Typical operating conditions within the Texaco gasifier are 500 psig and 2,500°F. The chemical constituents within the coal react with oxygen to form light gases (primarily water vapor, carbon monoxide, hydrogen, and carbon dioxide), with the mineral matter (ash) removed as a molten slag. Most of the slag is quenched in a water pool in the bottom of the gasifier and removed from the gasifier via ash locks as a water slurry. The slag-water slurry is then dewatered and the water clarified (i.e., solids removed) before being recycled to the gasifier.

The raw gas stream has an energy content of about 250 Btu per standard cubic foot and is referred to as "medium-Btu" or synthesis gas. It must be cleaned of all solids, sulfur compounds, and ammonia that have been produced in the gasifier. The gas is freed of carbon soot and fly ash in a quenching zone. The soot is recycled to Coal Preparation section, so that the only major solids discharge from this coal refinery would be an easily handled slag. In addition, the temperature of the synthesis gas must be lowered so as to allow the synthesis gas to be cleaned. This is accomplished by passing the hot synthesis gas through the Waste Heat Recovery & Gas Cooling section, generating steam which may be sent to the Heat Recovery Steam Generator or directly to the Steam Turbine Generator, depending on the quality of the steam so produced.

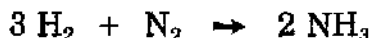
During coal gasification, essentially all of the sulfur in the coal volatilizes, with the predominant sulfur compound being hydrogen sulfide (H_2S) with smaller amounts of carbonyl sulfide (COS) and carbon disulfide (CS_2). These sulfur compounds are removed in the Acid Gas Removal section, so as to comply with environmental legislation and also to avoid poisoning of the catalyst used in ammonia synthesis. Typically, the sulfur compounds within the synthesis gas are absorbed by a physical or chemical solvent and concentrated in a gaseous stream [4]. In the Sulfur Recovery unit, the concentrated sulfur gas stream is converted into elemental sulfur, which can be sold as a byproduct. The gases from the Sulfur Recovery unit are processed to remove any traces of sulfur compounds before being vented to the atmosphere.

Carbon dioxide is a feedstock in urea synthesis. Its removal from the synthesis gas can be by any one of a number of licensed processes, e.g., hot potassium carbonate, MEA (monoethanolamine), Selexol®, etc. [4]. Impurities in the raw carbon dioxide stream are removed to meet urea feedstock specifications.

The synthesis gas stream from the Acid Gas Removal area is separated by membrane technology into two streams, a hydrogen-rich stream that is sent for ammonia synthesis and a hydrogen-deficient (i.e., CO-rich) stream that is sent to a combustion turbine for power generation.

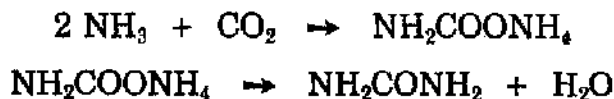
Membranes separate gases by their molecular size and gas solubility and are typically tailored to permit the passage of a particular gas only. Advantages of membrane technology are that it is potentially energy-efficient and that the required process equipment is simple and compact [5]. Hydrogen has an extremely small molecular size in comparison with the other synthesis gas constituents (CO, CH₄, CO₂) and so is more easily separated [6]. The hydrogen-rich gas stream passes through a zinc oxide bed to reduce the total sulfur content to approximately 0.1 ppm.

Ammonia is formed from hydrogen and nitrogen by the following reaction:



Nitrogen from the Air Separation section is added to the H₂-rich stream from the Membrane Separation section in the ratio of three volumes of H₂ to one volume of N₂. This gas mixture is compressed to the pressure required for ammonia synthesis, approximately 3,150 psia. The compressed gas mixture is then catalytically converted to ammonia in the Ammonia Synthesis section. The effluent vapors from the Ammonia Synthesis section are cooled to condense and separate the anhydrous liquid ammonia from the effluent. Conventional ammonia synthesis technology recycles the unreacted syngas back for further conversion, but instead this coal refinery concept sends the reactants not transformed into ammonia to power production.

Urea is produced from liquid ammonia and gaseous CO₂ at high pressure (2,350 to 3,675 psia) and temperature (340 to 400°F) [1]. The two feed components are delivered to the high pressure Urea Reactor, typically at a molar ratio greater than 2.5 NH₃:1 CO₂. Ammonia and CO₂ (from the Acid Gas Cleanup system) are catalytically combined in an exothermic reaction to form ammonium carbamate (NH₂COONH₄), which is kept at elevated temperature until water splits out, forming urea [7]:



Any excess CO₂ would be vented to the atmosphere.

The fuel for the power generation system is the hydrogen-deficient portion of the synthesis gas together with the unreacted gas stream from Urea Synthesis. The overall concept of power generation is similar to an Integrated Gasification Combined-Cycle (IGCC) plant. Essentially, electricity is produced both by burning the synthesis gas in

a combustion turbine and by using waste heat to boil water to drive a steam turbine. Compressed air is mixed and burned with the medium-Btu gas and the resulting hot gases are expanded through the Gas Turbine Generator to produce the majority of the electric power (typically two-thirds of the total power output).

The required oxygen and nitrogen streams are produced from air in the Air Separation Plant. Nitrogen in excess of process requirements is introduced into the Gas Turbine to act as a diluent (in place of steam) in the combustor to limit NO_x production and to increase the mass flow through the turbine thereby increasing electrical production [8].

The exhaust gases from the Gas Turbine Generator contain significant energy that can be utilized. These exhaust gases pass through the Heat Recovery Steam Generator, generating steam and depending on the gasification technology, superheating the steam from the Waste Heat Recovery & Gas Cooling system. The superheated steam is sent to the Steam Turbine Generator to generate additional electricity.

Types of Feed Coal

Theoretically, any type of coal can be gasified to produce synthesis gas which may be converted to urea. Coal characteristics such as reactivity, size consistency, ash-fusion temperature, grindability, and degree of coal caking can all affect the choice and performance of a given gasifier.

Gasification allows use of high-sulfur (low-cost) coals in an environmentally responsible manner without resort to extensive precombustion coal cleaning or post-combustion flue gas scrubbing.

Products

The main products from this coal refinery are urea and electricity. The factors determining the relative amount of electricity-to-urea include coal characteristics (such as the initial amounts of carbon and hydrogen in the coal), gasifier design (through the resulting synthesis gas composition and the overall thermal process efficiency), and the operating temperature of the Gas Turbine (higher temperatures allow greater conversion of the energy of the synthesis gas stream into electricity).

Marketable byproducts from the process can include sulfur and potentially CO_2 . The elemental sulfur can be combined with phosphate rock into a granular super phosphate fertilizer, thereby maximizing fertilizer production from this coal refinery concept. The current design is to vent the portion of the CO_2 stream not required for urea synthesis. One option, dependent on process economics, is to purify and compress this CO_2 stream for sale as a specialty gas or for utilization in enhanced oil recovery (EOR).

Likely Applications

Urea has been used commercially as a solid nitrogen fertilizer, cattle-feed supplement, in the production of various barbiturates (through reaction with malonic acid) and in the production of urethanes (through reaction with various alcohols). Elemental sulfur has many uses, such as production of sulfuric acid and other inorganic chemicals such as agricultural fertilizer.

Coproduction of electricity and chemical products in a fully integrated energy facility can potentially reduce the cost of generating electricity. Similar coal refinery concepts proposed for electric utilities are based on coproduction of electricity and a commercial chemical (typically methanol) which could be used as a turbine fuel for peak-shaving purposes in power generation. Even though this coal refinery concept produces urea which is not a turbine fuel, the electric utility industry may be interested in owning and operating this coal refinery concept.

Coal gasification to produce synthesis gas is important to the urea, and consequently to the nitrogen fertilizer, industry in many foreign countries. Given that expansion of foodstuff production is based on the availability of fertilizer for agricultural purposes, it would be expected that the commercial entity most interested in this coal refinery concept would be the industrial chemicals industry (more specifically, the fertilizer industry).

Status of Development

A 150 ton of coal per day Texaco gasifier was operated at the Muscle Shoals facility of TVA to produce synthesis gas. This gas was converted to hydrogen by downstream processing units. The hydrogen was then used to produce ammonia in an existing ammonia plant. A portion of the CO₂ stream from acid gas removal was cleaned to urea feedstock specifications for demonstration purposes. This demonstration facility was shut down in 1985 at the project conclusion. Subsequently, a commercial unit comprised of four 500 ton of coal per day Texaco gasifiers has been operated by Ube in Japan to produce ammonia [14].

A proposal for a prototype 2,300 ton of coal per day demonstration plant of this coal refinery concept has been submitted by TVA in the fourth solicitation of the Clean Coal Technology (CCT) program [9].

Environmental Aspects

The main environmental intrusions from this coal refinery include atmospheric emissions of SO₂ and NO_x, liquids, and solid wastes. Integration of the IGCC portion within this coal refinery results in very low emissions of SO₂, NO_x and particulates. For this coal refinery, over 99 percent of the coal-derived sulfur is captured to eliminate potential poisoning of the catalysts. It has been reported that the SO₂ and NO_x emission rates would meet Maximum Achievable Control Technology (MACT) requirements of the New Source Performance Standards (NSPS) [10]. Emissions of trace toxics, such as halogens, heavy metals, ammonia and organic compounds are believed to be extremely low as the metals are fixed in the slag, which is non-leachable [11].

The process water requirements for this coal refinery are reported to be approximately 60 percent less than for a conventional pulverized coal-fired power plant [10]. The urea synthesis process would generate water, as one mole of water is formed for each mole of urea produced. The water formed during urea synthesis would contain residual levels of ammonia and urea, which would be removed during wastewater treatment.

The average solid waste production rate would be approximately equal to the quantity of ash resulting from coal combustion. The slag that the coal refinery produces has been judged nonhazardous [10, 12]. The sulfur within the coal is recovered and sold; it therefore generates a revenue stream instead of a disposal concern.

Utilization of IGCC technology for electric power production can result in significant reductions in CO₂ emissions; a 10 to 15 percent reduction in CO₂ emission per kWh produced when compared with a conventional coal-fired power plant is reported for this coal refinery [10]. Further reductions in CO₂ emissions can be realized if the CO₂ is recovered from the effluent stream for urea synthesis.

Research Needs

Commercial processes exist for ammonia and urea synthesis from synthesis gas. IGCC technology is near or at the commercial application stage, with several large IGCC projects having been launched during 1990 [13]. A reliable, efficient and economical unit would be demonstrated by the successful integration of the many process steps of the different technologies.

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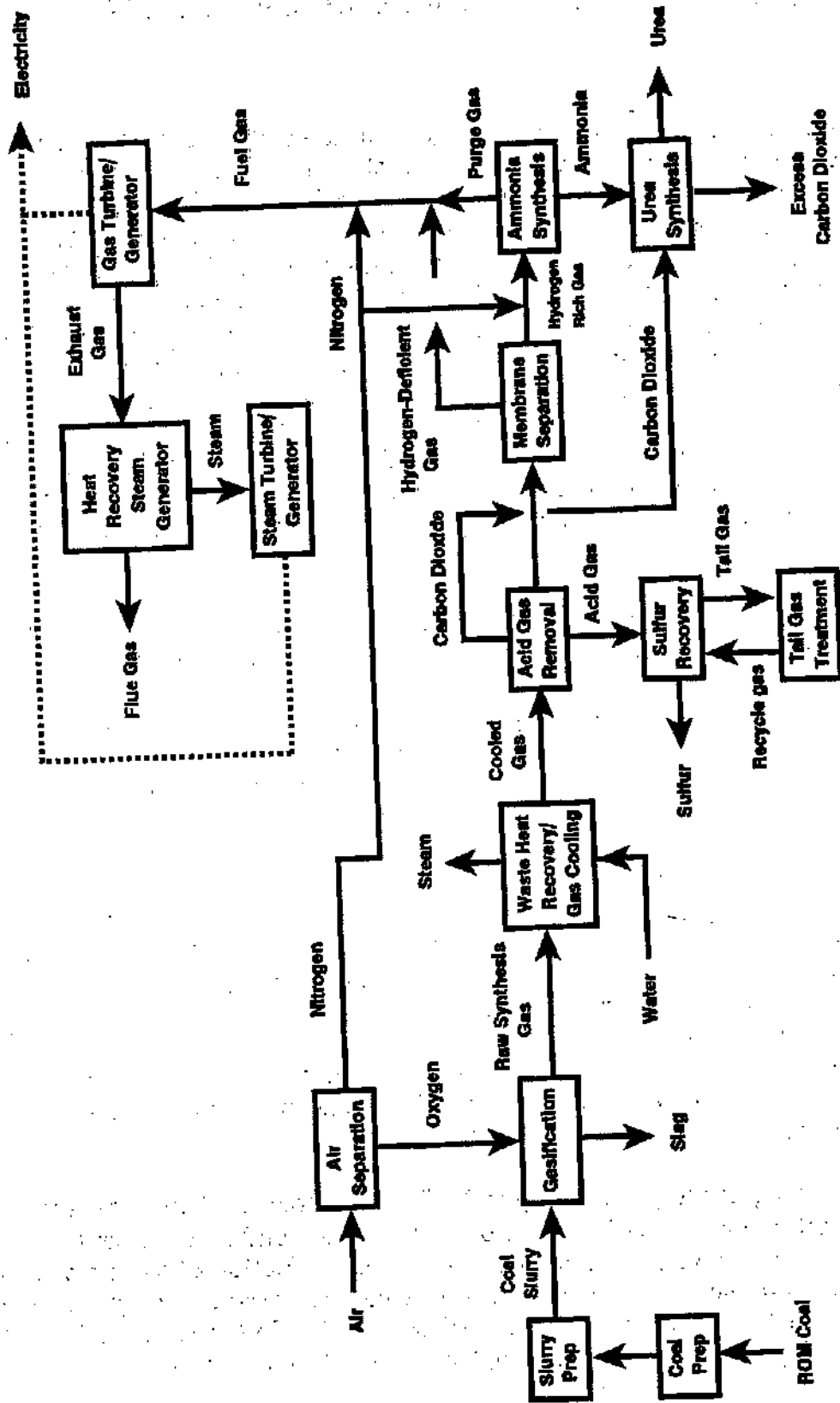


FIGURE 1: Overall Block Diagram of the Coproduction of Urea and Electricity Concept

DUAL PRODUCTION OF AMMONIA AND ELECTRICITY

Introduction

Dual production of ammonia and electricity is an example of a coal refinery in the Gasification category. Ammonia production is the second largest consumer of synthesis gas production for formation of chemicals by coal gasification, with Fischer-Tropsch products being the greatest [1]. A large proportion of the food and fiber produced in the U.S. is dependent on the use of fertilizers [2], with nitrogen as the nutrient of major importance. Practically all nitrogen fertilizer is made from ammonia, which is currently produced in the U.S. using natural gas. Historically, making ammonia from coal is an old concept but the process has not been economical in the U.S. due to the low price of natural gas. However, modern coal-based ammonia plants have been built in the Eastern Hemisphere and have successfully demonstrated the feasibility of large coal-to-ammonia/fertilizer plants [3, 4]. In this country, a 150 ton per day synthesis gas facility had been constructed and evaluated by TVA to determine the economic and technical aspects of converting coal to ammonia via coal gasification [5, 6].

The coal refinery envisaged here is shown in Figure 1. With its highly integrated energy recovery systems, the ammonia-from-coal plant would be a combination chemical plant and power generation system. It would be a "grass roots" facility, requiring only coal and water for operation and generating its own electric power. It would contain all the necessary facilities to gasify the coal, treat the resulting syngas, and convert the treated syngas to ammonia and electricity. The main products are ammonia and electricity, with sulfur, N_2 and CO_2 as marketable byproducts.

Detailed Process Description

The major process components of this coal refinery concept include a coal preparation step which results in a finely ground and slurried coal that is gasified to produce synthesis gas. The impurities of the synthesis gas are removed before the ammonia synthesis process. A portion of the synthesis gas produced in the gasifier is diverted to an advanced combined-cycle power plant to produce the required steam and electricity, with electricity available for export. Greater detail of the individual process components is given below.

A detailed flow diagram is given in Figure 1. The inlet coal is taken to be Illinois No. 6 Bituminous, with a higher heating value of 11,342 Btu/lb and a sulfur content of 4.2 percent by weight (see Table 1 for more details). The production of the synthesis gas from coal for this example is by a Texaco gasifier, which is currently being used for commercial-scale production of ammonia from coal by the Ube Industries, Limited of Japan (1,650 ton per day of coal) and in Lunan, China (400 ton per day of coal) [4, 7].

In the Coal Preparation section, run-of-mine coal is first crushed to less than ½-inch. It then enters a wet grinding circuit where it is mixed with the condensate from the ammonia stripper to form a coal slurry of 55 percent (or higher) by weight, and ground to a fine consistency (100 percent less than 0.055-inch). The slurried coal is pumped into the Gasification section where it reacts with the oxygen gas. Typical operating conditions within the Texaco gasifier are 850 psig and 2,400°F. The chemical constituents within the coal react to form light gases (shown in Table 2), with the mineral matter (ash) removed as a molten slag. Most of the slag is quenched in a water pool in the bottom of the gasifier and removed from the gasifier via ash locks as a water slurry. The slag-water slurry is then dewatered and the water clarified (i.e., solids removed) before being recycled to the gasifier.

In an alternate configuration, the raw gas stream passes from the gasifier to a radiant heat exchanger where the raw gas is cooled to about 1,400°F and then flows through a convection waste-heat boiler, generating saturated steam at 950 psig. The gas is freed of carbon soot and fly ash in a quenching zone. The soot is recycled to Coal Preparation section, so that the only major solids discharge from this coal refinery would be an easily handled slag.

The synthesis gas stream from the gasifier is then split into two, with 70 percent (by weight) sent to a combined-cycle power generation system and the rest as feedstock for ammonia synthesis. The following paragraphs deal with ammonia synthesis.

Ammonia is formed from hydrogen and nitrogen by the following reaction:



so that the desired product gas from the gasifier must be primarily hydrogen. The carbon monoxide contained in the raw gas stream is catalytically converted into additional hydrogen by the following water-gas shift reaction:



in the Shift Conversion section. The above reaction is quite exothermic, and the heat evolved during shift conversion together with the waste heat generated during gas cooling is used in the generation of low, medium, and high pressure steam.

The impurities (sulfur-containing compounds, CO₂) have to be removed so as to comply with environmental legislation requirements and also to avoid poisoning of the catalyst used in ammonia synthesis. The Enhanced Gas Cleanup and Purification section is composed of a Rectisol® wash unit, a molecular sieve unit, and a nitrogen wash unit.

The Rectisol® process employs cold methanol (at about -45° F) to physically absorb the acid gases (H₂S, COS, CO₂) and so reduce the content of H₂S and COS to less than 1 ppm and CO₂ to less than 30 ppm. The acid gases which are extracted by the Rectisol® process from the raw synthesis gas are sent to the Sulfur Recovery section, namely a Claus® unit for conversion of the hydrogen sulfide to elemental sulfur. A Shell Claus Off-Gas Treating (SCOT) unit removes the last traces of sulfur from the Claus® unit vent gas. Over 99 percent of the sulfur contained in the coal is recovered as elemental sulfur.

The treated gas from the Rectisol® wash unit contains low levels of CO₂ and methanol vapor which are partially removed by a molecular sieve unit. Molecular sieves are synthetic aluminosilicate adsorbents which possess high porosity and only adsorb small molecules. They are used primarily in gas treatment, as they can be made to be selective to the shape of molecules to be removed. A liquid nitrogen wash is then used to remove all gases other than H₂ that remain in the syngas after the molecular sieve unit. The remaining CO, CH₄, argon and a portion of the hydrogen are removed by passing the synthesis gas through a series of liquid nitrogen baths. The result is an essentially pure H₂ stream. The liquid nitrogen stream which contains CO, CH₄, argon and H₂ is not vented to the atmosphere but instead is considered a low-pressure fuel gas (higher heating value of about 160 Btu per standard cubic foot) and combusted to provide energy for the Claus® unit.

After passage through the Enhanced Gas Cleanup and Purification section, the synthesis gas contains very low concentrations of gases other than H₂ (less than 50 ppm CH₄ and argon, 5 ppm CO and O₂). Nitrogen is added to the H₂ stream in the ratio of three volumes of H₂ to one volume of N₂. This gas is then compressed to a pressure of about 3,150 psia. This high pressure is required for the ammonia synthesis reaction. The compressed gas mixture is then catalytically converted to ammonia in the Ammonia Synthesis section.

The required oxygen and nitrogen streams are produced from air in the Air Separation Plant. Nitrogen (containing less than 5 ppm O₂) in excess of process requirements can be stored on-site for sale as a marketable product. Another option (which is not applied here) is to introduce the nitrogen to the gas turbine to act as a diluent (in place of steam) in the combustor to limit NO_x production and to increase the mass flow through the turbine thereby increasing electrical production [8].

The integration of the coal-based ammonia plant allows utilization of the waste heat for the overall energy requirement of the ammonia synthesis process, so that only a small additional amount of power (approximately 0.02 kWh per lb of run-of-mine coal) need be produced in the Advanced Power Generation system. Most of the power required for the individual compressors within the ammonia synthesis plant is supplied by the 850 psig, 900° F steam. The majority of the rest is supplied by the 100 psig saturated steam.

The fuel for the Advanced Power Generation system is a portion of the synthesis gas produced in the gasifier. The overall concept of power generation is similar to an Integrated Gasification Combined-Cycle (IGCC) plant [9]. The gaseous fuel (energy content of about 250 Btu per standard cubic foot; referred to as "medium-Btu gas") must be cleaned of all solids, sulfur compounds, and ammonia that have been produced in the gasifier. In addition, the temperature of the synthesis gas must be lowered so as to allow the synthesis gas to be cleaned. This is accomplished by passing the hot synthesis gas through the Waste Heat Recovery & Gas Cooling section, lowering the temperature of the syngas to about 700°F. The sensible heat is recovered to generate saturated steam which is sent to the Heat Recovery Steam Generator.

The degree to which this portion of the synthesis gas needs to be cleaned is not as extensive as with the syngas stream for ammonia synthesis. The cooled synthesis gas is scrubbed of hydrogen sulfide (H_2S) and carbonyl sulfide (COS) by the Selexol® acid gas removal unit. In the Selexol® process, the sour gas enters an absorber tower after having been cooled. Almost all of the H_2S is removed by physical absorption as the sour gas flows up through the absorber tower. A syngas stream with low levels of contaminants is produced. About 15 percent of the CO_2 is removed, along with minor amounts of other gases. The absorbed gases are recovered by a series of pressure let-downs and stripping. A concentrated acid gas stream is produced that contains between 24 and 39 percent H_2S , which is routed to the Claus® unit. The sulfur-containing gases from the Selexol® unit together with the stream from the Enhanced Gas Cleanup and Purification section are converted to elemental sulfur in the molten state in the Claus® Sulfur unit. The solid sulfur can be sold in the commercial market.

The pressure of the cleaned synthesis gas is too great for introduction into the gas turbines to burn and is decreased to about 250 psia by expansion. The Fuel Gas Expansion Turbine converts this reduction in pressure into mechanical energy which is further converted into electrical energy (0.014 kWh per lb of run-of-mine coal feed). Compressed air is then mixed and burned with the synthesis gas and the resulting hot gases (at about 2,000°F) are expanded through the Gas Turbine Generator to produce the majority of the electric power (0.5 kWh per lb of run-of-mine coal feed).

The exhaust gases from the Gas Turbine Generator leave at around 1,000°F and therefore contain significant energy that can be utilized. These exhaust gases pass through the Heat Recovery Steam Generator, superheating the saturated steam from the Waste Heat Recovery & Gas Cooling system. The superheated steam is sent to the Steam Turbine Generator to produce electricity (0.37 kWh per lb of run-of-mine coal feed).

Based on this Texaco gasification design, the overall energy requirement for the ammonia synthesis section is approximately 39.3 million Btu for Illinois No. 6 Bituminous and 42 million Btu for high-sulfur, high-ash Australian coal per ton of ammonia

produced [4]. For a lignite-based plant, it would be about 4 to 6 percent higher than for Illinois No. 6 Bituminous.

The electrical requirement for the overall process is approximately 0.13 kWh per lb of run-of-mine coal. For this coal refinery design, an excess of 0.75 kWh per lb of run-of-mine coal is available for export as a saleable product.

Types of Feed Coal

Historically, the types of coals that have been used for ammonia synthesis are lignites (27 percent) and bituminous (73 percent) [1]. Theoretically, any type of coal, other than anthracite, can be gasified to produce synthesis gas which may be converted to ammonia. Anthracite is typically not gasified due to its lower reactivity during gasification (requiring higher levels of steam and oxygen consumption). Coal characteristics such as reactivity, size consistency, ash-fusion temperature, grindability, and degree of coal caking can all affect the choice and performance of a given gasifier.

A major component of this coal refinery is the coal gasification plant employing a Texaco gasifier. In theory, the entrained-flow configuration of the Texaco gasifier has the ability to gasify all ranks of coals. The feed coals that have been successfully gasified at the Cool Water demonstration plant (nominal capacity of approximately 1,000 tons of coal per day) of Texaco gasification technology include high-sulfur coals such as Illinois No. 6 bituminous (3.1 weight percent sulfur, dry basis) and Powhatan (5.1 weight percent sulfur, dry basis), low-sulfur western coals (Sufco; 0.35 weight percent sulfur, dry basis), and foreign coals such as high-ash, low-sulfur Lemington (Australian) coal [9, 10, 11].

The Ube Ammonia plant (production of one thousand tons of ammonia per day) has successfully gasified a variety of coals including low-sulfur (0.3 weight percent) high-ash (11.2 weight percent) Canadian coal, high-sulfur (4.2 weight percent) high-ash (15.0 weight percent) Australian coal, medium-sulfur (1.3 weight percent) high-ash (13.5 weight percent) South African coal, and low-sulfur (0.8 weight percent) low-ash (7.7 weight percent) Chinese coal [4].

Products

The main products from this example coal refinery are ammonia (NH_3) and electricity. Marketable byproducts from the process can include sulfur, N_2 and CO_2 . The product output is shown in Table 3 in terms of pounds product per pound of feed coal. This analysis indicates that approximately 0.17 pounds of ammonia and 0.75 kWh of electricity are produced from 1 pound of feed coal.

The values in Table 3 are in part dependent upon the type of feed coal (Illinois No. 6 Bituminous coal for this example) and the ratio of the amount of synthesis gas for ammonia production to that for power generation. The difference in ammonia output for a grass-roots ammonia synthesis plant between different gasification technologies is not significant (based on computations using reference [1]).

Likely Applications

Ammonia can be used directly as a fertilizer, catalytically oxidized to NO_2 for the production of nitric acid (HNO_3), or combined with nitric acid to form ammonium nitrate (NH_4NO_3) which in turn is used as a fertilizer or in explosives. Elemental sulfur has many uses, such as production of sulfuric acid and other inorganic chemicals. The high purity nitrogen and carbon dioxide streams have many uses, one of which is injection into oil wells for enhanced oil recovery [12] and as specialty gases for the industrial sector.

Coproduction of electricity and chemical products in a fully integrated energy facility has been projected to potentially reduce the cost of generating electricity by approximately 40 percent [13]. The EPRI concept is based on coproduction of electricity and a commercial chemical (typically methanol) which could be used as a turbine fuel for peak-shaving purposes in power generation [14]. Even though this coal refinery example does not fit the EPRI concept as ammonia is not a turbine fuel, the electric utility may be interested in owning and operating this coal refinery concept.

Coal gasification to produce synthesis gas is important to the ammonia and consequently to the fertilizer industry in many foreign countries. Given that expansion of foodstuff production is based on the availability of fertilizer for agricultural purposes, it would be expected that the commercial entity most interested in this coal refinery concept would be the industrial chemicals industry (more specifically, the fertilizer industry).

Status of Development

The two gasification technologies that were judged to be most suitable for ammonia synthesis are the Texaco gasifier and the slagging gasifier developed by British Gas/Lurgi [11]. The reasons are that operation of the gasifier at medium-to-high pressures requires lower coal consumption, reducing the number of gasifiers and plant size, and also that the ammonia synthesis process operates at high pressure.

A comparison of the relative capital costs between the Texaco and British Gas/Lurgi gasifiers indicated that the British Gas/Lurgi gasifier is more expensive than the Texaco gasifier, primarily due to the large amounts of methane formed in the British Gas/Lurgi system which must be converted into CO and H_2 for ammonia synthesis [15].

A 150 ton of coal per day Texaco gasifier was operated at the Muscle Shoals facility of the Tennessee Valley Authority (TVA) to produce synthesis gas, which was converted to hydrogen by downstream processing units. The hydrogen was then used to produce ammonia in an existing ammonia plant. This demonstration facility was shut down in 1985 at the project conclusion.

The first large-scale ammonia syngas plant based on Texaco gasification technology started operation in 1984 [4, 23]. The plant, owned by Ube Industries, Limited, of Japan, has successfully produced the rated capacity of 1,000 tons of ammonia from 1,500 tons of coal per day. The Texaco coal gasification unit replaced an existing steam reforming unit (e.g., catalytic generation of hydrogen from a hydrocarbon fuel using steam). The plant had been using a variety of petroleum-based feedstocks including petroleum coke, naphtha and LPG (liquified petroleum gas; propane, butane) for ammonia production before switching to coal.

Commercial scale demonstration of the Texaco technology for coal gasification is targeted to start in 1995, with construction at Freeport, MA of two Texaco oxygen-blown gasifiers, each with a 2,000 ton per day capacity [16]. The 440 MW IGCC power plant based on Texaco gasification technology is expected to have an initial plant heat rate of 9,400 Btu per kWh (thermal efficiency of about 36.3 percent) and will eventually be expanded from an initial 440 MW to 1,200 MW [17].

Environmental Aspects

The main environmental intrusions from this coal refinery include atmospheric emissions of SO_2 and NO_x , and solid wastes. The emission rate for SO_2 is approximately 0.0008 lb per lb of feed coal, while for NO_x it is approximately 0.0003 lb per lb of feed coal [18]. The SO_2 and NO_x emission rates would meet the minimum level of control allowed by NSPS. The release of particulates is negligible (0.0003 lb per lb feed coal, [19]) as a low level of particulate emissions is inherent in the Texaco gasification technology [8]. Emissions of trace toxics, such as halogens, heavy metals, ammonia and organic compounds are assumed to be extremely low as the metals are fixed in the slag, which is non-leachable [20].

The amount of slag produced from coal gasification is equal to the quantity of ash that results from burning coal. The slag that the coal refinery produces has been judged nonhazardous [14]. The sulfur within the coal is recovered and sold; it therefore generates a revenue stream instead of a disposal concern. The average solid waste production rate would be on the order of the slag production rate, which for this example is 0.23 lb per lb feed coal.

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. This coal refinery concept would extract the CO_2 produced by shift conversion and ammonia synthesis for storage

and possible compression to 2,000 psig for pipeline delivery (approximately 0.43 lb per lb of feed coal). For combined-cycle power generation, it is usually beneficial to leave carbon dioxide in the syngas, since it can release power in the gas turbine due to expansion, and reduces the requirement for NO_x control steam injection [21]. The CO₂ which is produced in the power generation section (approximately 1.5 lb per lb of feed coal) [18] is released to the atmosphere. Approximately 22 percent of the CO₂ generated by this coal refinery is captured, without release to the atmosphere.

Research Needs

Ammonia synthesis from coal is technically well grounded. IGCC technology is near or at the commercial application stage, with several large IGCC projects having been launched during 1990 [22]. A reliable, efficient and economical unit would be demonstrated by the successful integration of the many process steps of the different technologies.

The optimum ratio of the amount of synthesis gas for ammonia synthesis versus power generation would be determined by a detailed economic analysis which includes the results of recent on-going development leading to higher process efficiency.

Table 1: Feed Coal Analysis

ULTIMATE ANALYSIS (weight percentage, dry basis)						
Carbon	Hydrogen	Oxygen	Sulfur	Nitrogen	Chlorine	Ash
67.0	4.5	8.4	4.2	1.3	0.01	14.6
Free Moisture (weight percent)			6.0			
Higher Heating Value (dry basis)			11,342 Btu per lb			

Table 2: Typical Texaco Gasifier Product Gas Composition

COMPOUND	YIELD, mole percent ash-free basis
Carbon Monoxide, CO	30.60
Hydrogen, H ₂	23.80
Carbon Dioxide, CO ₂	12.90
Methane, CH ₄	0.26
Nitrogen, N ₂	0.36
Argon, Ar	0.11
Hydrogen Sulfide, H ₂ S	1.01
Carbon Oxysulfide, COS	0.067
Ammonia, NH ₃	0.09
Water, H ₂ O	30.80
TOTAL	100.00

Table 3: Product Output for Coproduction of Ammonia and Electricity

PRODUCT	OUTPUT (per lb coal)
Ammonia, NH ₃	0.17 lb
Electricity	0.75 kWh
Sulfur, elementary	0.05lb
Carbon Dioxide, CO ₂	2.16 lb
Nitrogen, N ₂	3.2 lb

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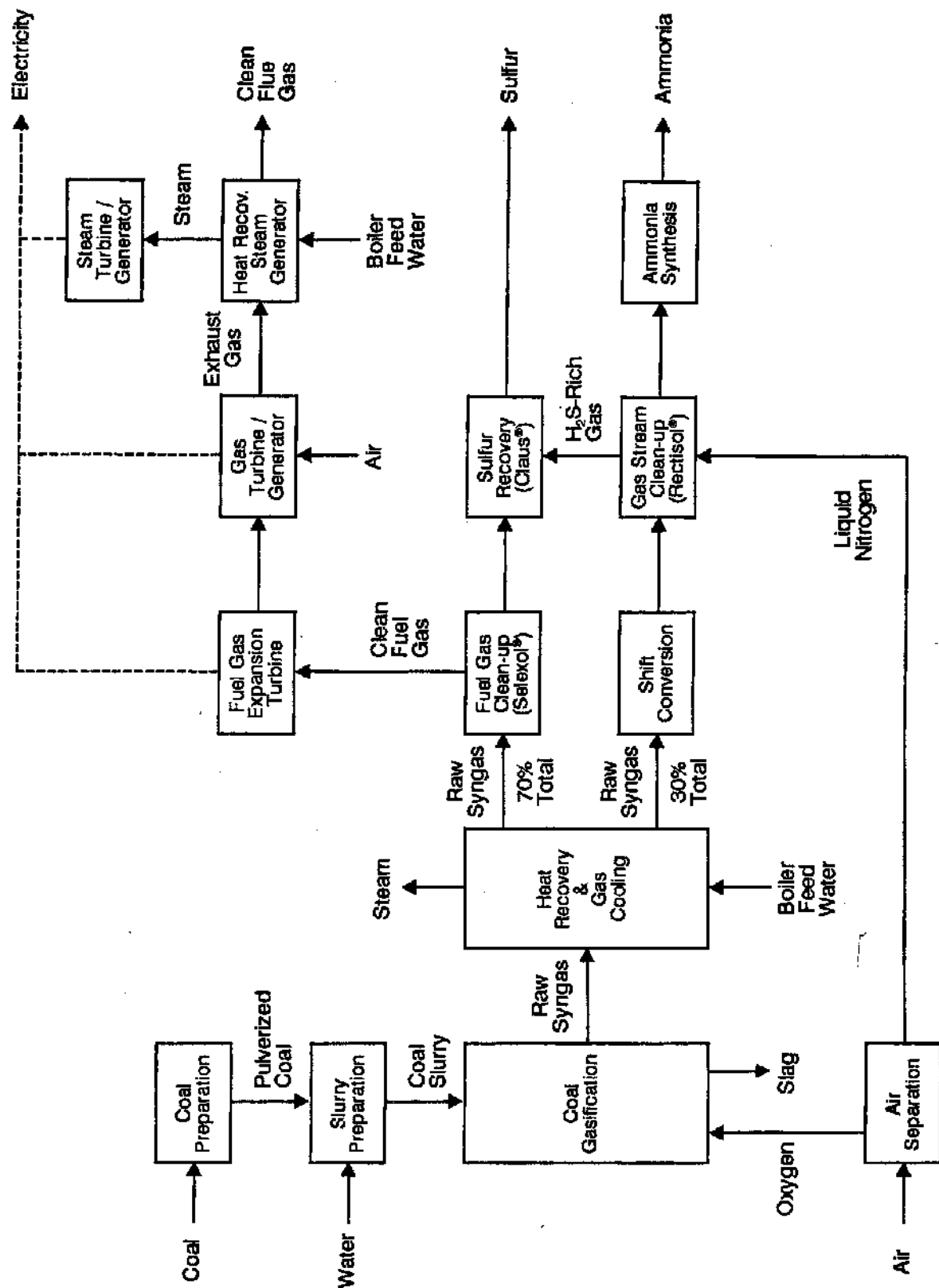


Figure 1: Dual Production of Ammonia and Electricity Concept