APPENDIX B

GASIFICATION COAL REFINERY CONCEPTS

COPRODUCTION OF ACETIC ANHYDRIDE, ACETIC ACID AND METHANOL

Introduction

Coproduction of acetic anhydride, $(CH_3CO)_2O$, acetic acid, CH_3COOH, and methanol, CH_3OH, is an example of a coal refinery in the Gasification category. In 1983, The Tennessee Eastman Company (TEC), a subsidiary of the Eastman Kodak Company, became the first manufacturer to develop and commercialize chemical technology using synthesis gas from coal for the production of acetic anhydride. Acetic anhydride is used by Eastman in the manufacture of photographic film base, cellulose plastics, acetate for cigarette filter tow and textile yarns, as well as coating chemicals [1].

This process is in commercial operation by the Tennessee Eastman Company of Kingsport, TN and was built solely with Kodak funds. The process substitutes coal-derived synthesis gas for ethylene or other hydrocarbon feedstocks as the raw material. The TEC plant converts 900 tons per day of coal into an annual production rate of 182,500 tons of methanol, 75,000 tons of acetic acid, and 250,000 tons of acetic anhydride. The chemicals produced would require the equivalent of one million barrels of oil per year using conventional technology [2], and so this process can be considered to be a step towards reducing our national dependence on foreign oil.

The coal refinery concept based on this process is shown in Figure 1. The system integrates the generation of synthesis gas by Texaco gasification technology with conversion of the resulting synthesis gas to industrial chemicals. The necessary inputs to this coal refinery include run-of-mine coal, air, and water, while major products include acetic anhydride, acetic acid and methanol. Byproducts of this coal refinery would include elemental sulfur (and potentially liquid nitrogen).

Detailed Process Description

This process uses several licensed units to obtain methanol and carbon monoxide as feedstock for the production of methyl acetate, CH_3COOCH_3, and acetic anhydride. The two-step process uses methyl acetate as an intermediate. Very high yields under moderate operating conditions (340 to 410° F, 600 to 1,200 psia) using rhodium catalysts have been obtained [3].

In the Coal Preparation section, run-of-mine coal is first crushed and then enters a wet grinding circuit after being mixed with the condensate from the ammonia stripper. The result is a coal slurry of 55 percent (or higher) by weight, which is ground to a fine consistency (typically in the range of 100 percent less than 0.055 inches). The slurried coal is pumped from the Coal Slurry section into the Gasifier section where it reacts with oxygen in an entrained-flow gasifier.

The entrained-flow gasifier was selected based on its high pressure, high temperature operation [2] with typical operating conditions at 600 to 950 psia and 2,400 to 2,600°F. High-pressure operation eliminates recompression of the syngas before methanol synthesis, while the higher temperatures minimizes production of unwanted methane and byproduct liquid tars.

Unlike conventional coal combustion processes which completely combust the coal with stoichiometric or greater amounts of air, the mixture of coal and oxygen within the gasifier is sub-stoichiometric in oxygen and reacts to form a fuel or product gas. This product gas is a mixture of hydrogen (H_2) and carbon monoxide (CO) instead of the carbon dioxide and water vapor that is formed by complete combustion. This mixture of H_2 and CO is called 'synthesis gas' (or 'syngas'). The mineral matter (ash) originally in the coal is removed as a molten slag. Most of the slag is quenched in a water pool at the bottom of the gasifier and removed from the gasifier as a water slurry. The slag-water slurry is then dewatered and the water clarified (i.e., solids removed) before being fed to the gasifier.

The required oxygen stream for the gasifier is produced from air in the Air Separation Plant. Nitrogen (containing less than 5 ppm oxygen) in excess of process requirements can be stored on-site as a liquid for sale as a marketable product or vented to the atmosphere, as warranted by the process economics.

The raw product gas is scrubbed with water to remove any carbon soot or slag particles and to cool the gas. A portion of the raw gas stream is sent to a water-gas shift reactor where the following reaction takes place:

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

to increase the hydrogen concentration at the expense of carbon monoxide. Low-pressure steam is generated as the raw syngas is cooled before removal of acid gas impurities by the Rectisol® process.

Gasification converts the sulfur in the coal to reduced sulfur compounds (H_2S, COS) instead of SO_2 as for combustion processes. The syngas stream is treated to remove the sulfur compounds and carbon dioxide by the Rectisol® process. The Rectisol® process uses cold methanol (at about -45°F) as the solvent to physically absorb hydrogen sulfide and carbon dioxide from the raw syngas. The solubilities of the impurities (H_2S, COS, and CO_2) increase with decreasing temperature while the solubilities of the desired product gases (CO and H_2) in the methanol solvent are not affected by low temperatures. It is for this reason that the raw syngas feed for the Rectisol® process is precooled and refrigerated methanol used as the solvent. Releasing the absorbed gases for treatment in the Sulfur Recovery unit involves increasing the methanol temperature to release the acid gases. Sulfur levels of less than 0.1 ppmv can be obtained in the treated syngas, with complete removal of organic sulfur compounds such as mercaptans, thiophene (C_4H_4S), carbonyl sulfide (COS), and carbon disulfide (CS_2) [4].

The extracted acid gases from the Rectisol® process 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. Approximately 99.7 percent of the sulfur contained in the coal is recovered as elemental sulfur. The elemental sulfur byproduct can be sold in the commercial marketplace.

Acetic anhydride synthesis requires a feed stream of pure carbon monoxide, while stoichiometry for methanol synthesis requires a hydrogen-to-carbon monoxide ratio of two:

$$CO + 2\,H_2 \rightarrow CH_3OH$$

The H_2-to-CO ratio of the clean unshifted syngas is in the range of 0.7 to 0.9 [5, 6], so that to produce a hydrogen-rich stream for methanol production, a portion of the clean syngas is cryogenically separated into a carbon monoxide feed stream for the Acetic Anhydride section and a hydrogen stream for the Methanol Synthesis area. The pure hydrogen stream from the CO/H_2 Separator is mixed with the remaining syngas to achieve the desired H_2-to-CO ratio of two.

Methanol is produced from carbon monoxide, carbon dioxide, and hydrogen using a low-pressure process licensed from Lurgi. The Lurgi process operates at about 735 to 1,470 psia and 450 to 510°F. The synthesis gas flows through proprietary catalyst-filled tubes contained in a fixed-bed arrangement within a tubular reactor [7]. The reaction mixture leaving the reactor is cooled to separate the crude methanol from any unreacted synthesis gas, with the syngas recycled back to the methanol synthesis reactor for further conversion. The crude methanol contains small percentages of water, alcohols, and low-boiling impurities and is subsequently distilled to produce pure methanol.

A portion of the product methanol stream is diverted towards the Methyl Acetate area. An Eastman-patented process esterifies the methanol with recovered acetic acid (a byproduct from cellulose esters manufacturing) to produce methyl acetate [8]:

$$CH_3COOH + CH_3OH \rightarrow CH_3COOCH_3 + H_2O$$

The above reaction takes place in a single 210 foot reactor-distillation tower in which acetic acid and methanol react in a series of countercurrent flashing stages. Purified methyl acetate is the overhead product of the reactor-distillation tower, with water as the bottom product. Any side products that are generated are removed through a side-draw system from the tower.

In the Acetic Anhydride Reactor system, methyl acetate is carbonylated (addition of a carbonyl group, a carbon-oxygen double bond, to acetic anhydride) with carbon monoxide to yield acetic anhydride:

$$CH_3COOCH_3 + CO \rightarrow (CH_3CO)_2O$$

The proprietary catalytic system and process for the conversion of methyl acetate to acetic anhydride was developed by TEC and Halcon International [2].

Part of the product acetic anhydride stream is reacted with methanol to coproduce acetic acid and methyl acetate:

$$(CH_3CO)_2O + CH_3OH \rightarrow CH_3COOH + CH_3COOCH_3$$

The methyl acetate so formed is recarbonylated in the Acetic Anhydride Reactor system to form acetic anhydride. The crude acetic acid and acetic anhydride streams are purified by distillation.

Types of Feed Coal

The process currently uses as its feedstock Eastern high-sulfur bituminous coal containing 10 to 15 percent ash.

A major component of the TEC process is the coal gasification plant employing a 900 ton per day 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 [5, 6, 9].

Products

The main products from this example coal refinery are acetic anhydride, acetic acid, and methanol. Marketable byproducts include (elemental) sulfur, dry ash, and slag. The TEC plant gasifies approximately 900 tons of coal per day to produce 500 million pounds of acetic anhydride, 365 million pounds of methanol, and 150 million pounds of acetic acid per year [1]. Assuming the reported 95.5 percent on-stream factor for the process [10], approximately 0.8 pounds of acetic anhydride, 0.58 pounds of methanol, and 0.24 pounds of acetic acid are produced from one pound of feed coal.

Likely Applications

The major product of this coal refinery is acetic anhydride, which is an important industrial reagent, chemical feedstock (e.g., for the preparation of acylbenzenes by Friedel-Crafts acylation [11]) and used by TEC primarily for the production of film and other cellulose acetate products. Methanol can be used as a transportation fuel, turbine (utility) fuel, or as a chemical feedstock (e.g., production of acetic acid by carbonylation of carbon monoxide; conversion into gasoline). Acetic acid is a high-volume industrial chemical intermediate with many applications.

Given that the products from this coal refinery can be considered to be industrial intermediate chemicals, it would be expected that the principal commercial entity interested in pursuing this coal refinery concept would be the chemicals industry.

Status of Development

This coal refinery concept has been in commercial operation at Kingsport, Tennessee since June, 1983. A review of the recent operating history of the plant can be found in reference [12]. The facility has exceeded the design rate for acetic anhydride and acetic acid for the period of 1986 to 1989, with the gasifier typically operating between 100 to 105 percent of its design rate. After a shakedown period to identify major problems after startup, the onstream time (a measure of the reliability of process operation) has been above 95 percent with a typical value of 97 percent. In May 1989, TEC announced plans to double the capacity of the plant [12].

Comparison of the economics of anhydride production for the TEC process versus the traditional process (which reacts ketene, $CH_2{=}C{=}O$, with acetic acid to form acetic anhydride) is given in reference [13].

Environmental Aspects

Minimal environmental impact is claimed for this process [1, 10]. The high temperature operation of the gasifier avoids the coproduction of environmentally undesirable byproducts such as tars or other hydrocarbons. The majority (approximately 99.7 percent) of the sulfur originally contained in the coal is recovered as elemental sulfur [1]. The offgases from the individual subprocesses are consumed for heat recovery and power generation. The flue gases emitted to the atmosphere are reported to be within the limits of NSPS, with Lowest Achievable Emission Rates (LAER) having been met [1, 10].

The aqueous waste streams containing hydrocarbons from the individual subprocesses are recycled back for reuse in coal slurry and scrubbing water makeup [14], so that the majority of the process water requirement is satisfied with only loss makeup necessary [14].

The solids (slag, flyash) removed from this process are reported to be suitable for disposal in nonhazardous landfills [10].

Research Needs

The feed coal for this coal refinery is an Eastern high-sulfur bituminous. Locating a commercial plant based on this process away from the Applachian coal fields would involve determination of the effect of different coal types on the process design and operation.

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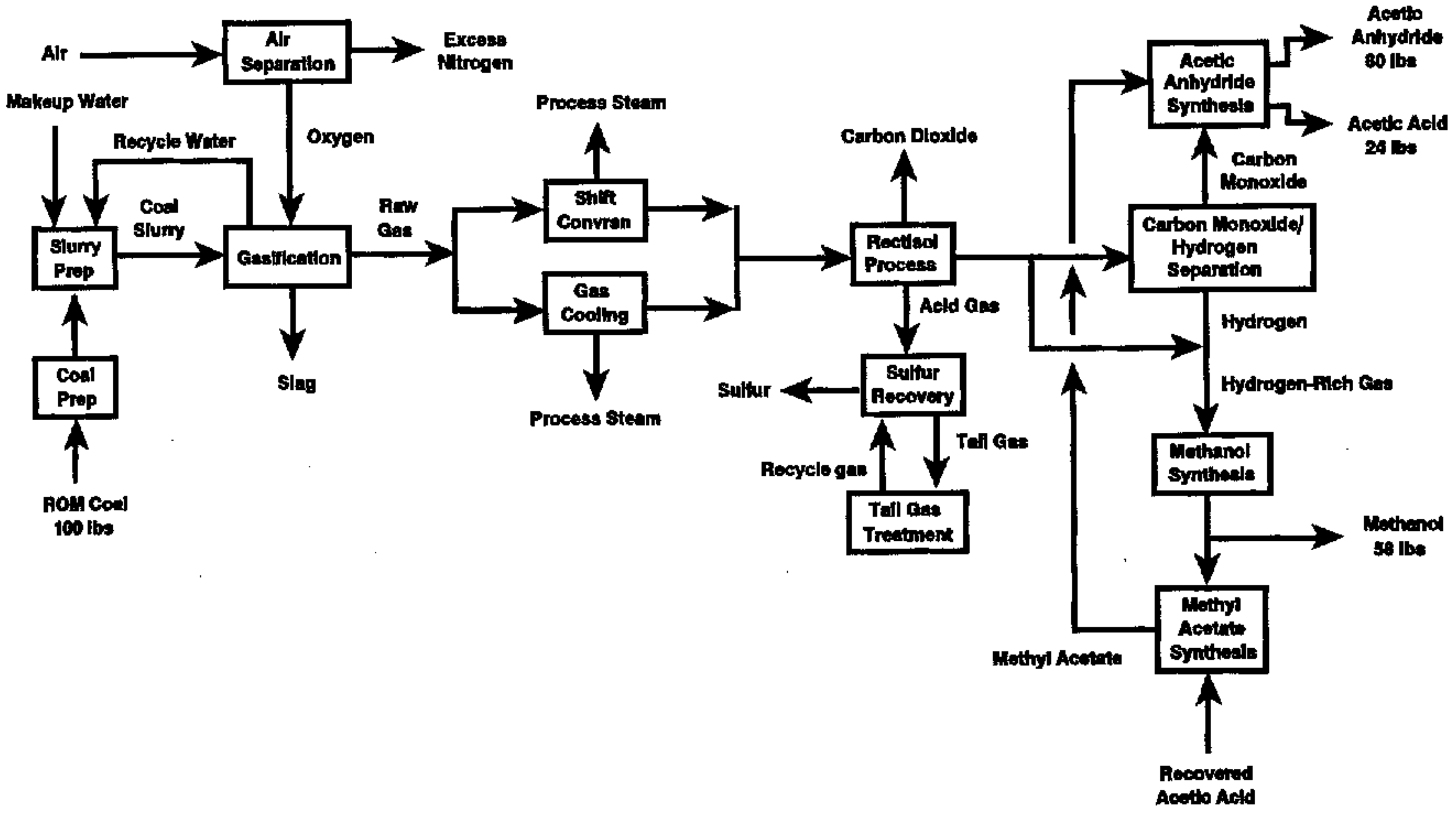

FIGURE 1: Overall Block Diagram of the Coproduction of Acetic Anhydride, Acetic Acid and Methanol Concept

COPRODUCTION OF METHANOL AND ELECTRICITY

Introduction

This coal refinery concept was developed by Air Products and Chemicals, Inc., and is an example of the Electric Power Research Institute's (EPRI's) definition of a coal refinery, in which electrical power generation becomes one element of a broader, integrated resource-processing capability due to the coproduction of commercial chemicals [1, 2]. It is based on the coproduction of electricity and methanol, with the conversion of the synthesis gas produced in an integrated coal gasification combined-cycle (IGCC) power plant to methanol. Gasification combined-cycle power plants have been stated to be the most likely candidate for new baseload electric generation in the near future [3]. The methanol synthesis step is integrated into an IGCC power plant in a single-pass (once-through) arrangement, so that a fraction of the coal-derived synthesis gas is converted to methanol. Under one scenario, methanol is produced during low demand periods and stored; when electric demand peaks, the methanol may be withdrawn and burned as a peaking fuel. This should lead to improved utilization of the power plant and provide methanol for use as a chemical feedstock or as a clean fuel to use in other power plants [4].

Methanol is one of the top ten organic chemicals in current production with an annual worldwide production rate of over 3 million tons [5]. Methanol production is one of the top three consumers of synthesis gas, with ammonia and Fischer-Tropsch liquids. Methanol has a high octane number and can be used as a raw transportation fuel. It is used directly to enhance the octane number of gasoline and in the production of methyl tertiary-butyl ether (MTBE), which is itself an excellent octane enhancer in gasoline. Methanol can itself be converted into gasoline; approximately 14,000 barrels per day are now being produced in New Zealand by the Mobil Methanol-to-Gasoline (MTG) process [6]. In addition, new chemical syntheses have been and are currently being developed to convert methanol into different chemical feedstocks, potentially displacing petroleum products as a feedstock (see Figure 1 [7]).

In the past, methanol was produced industrially from coal-derived synthesis gas for many years, and the technology was well established, with a 1940 U.S. production of about 130,000 gallons per day. Methanol production by coal was however displaced by low-cost natural gas and light petroleum distillates (naphtha, gas oils) after World War II [8]. Today, only one methanol production plant uses coal as a feedstock in the U.S. (Tennessee Eastman Company plant in Kingsport, TN).

The necessary inputs to this coal refinery include run-of-mine coal, air and water, while major products would include methanol and electricity. Sulfur and nitrogen (N_2) would be byproducts of this coal refinery.

Detailed Process Description

A detailed flow diagram is given in Figure 2, with all streams based on 100 pounds of run-of-mine coal fed. 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 3.9 percent by weight, dry basis (see Table 1 for more details). An entrained-flow gasifier is used to produce synthesis gas from coal.

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 600 to 950 psia and 2,400 to 2,600°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.

The gaseous fuel (energy content of about 250 Btu per standard cubic foot; referred to as medium-Btu or synthesis gas) must be cleaned of solids, sulfur compounds (H_2S, COS), and ammonia, prior to catalytic conversion to form methanol. The temperature of the synthesis gas is reduced by passing the hot synthesis gas through the Waste Heat Recovery & Gas Cooling section, which lowers the temperature of the syngas to about 70°F. The sensible heat is converted to high pressure saturated steam which is sent to the Heat Recovery Steam Generator. There the saturated steam is superheated and sent to the Steam Turbine Generator to produce electricity (0.54 kWh per lb of run-of-mine coal feed). The raw gas is freed of carbon soot and fly ash in a quenching zone. The soot is recycled to the Coal Preparation Section, so that the only major solids discharge from this coal refinery would be an easily handled slag. Over 99 percent sulfur removal is required to protect the methanol catalyst from possible poisoning.

The cooled synthesis gas contains sulfur compounds (H_2S, COS) that are removed in the sulfur removal section. Typically, the sulfur compounds within the synthesis gas are absorbed by a physical or chemical solvent and concentrated in a gaseous stream. The concentrated sulfur gas stream is then converted into elemental sulfur. The byproduct off-gases from the elemental sulfur recovery section are then processed. For this example, the physical absorption of sulfur is accomplished by a Selexol® acid gas removal unit, with a Claus® sulfur recovery unit to convert the gas stream into elemental sulfur, and a SCOT process for off-gas treatment [9]. The elemental sulfur byproduct can be sold in the commercial market.

The cleaned gas then passes through the once-through liquid phase methanol synthesis process, where approximately 9.6 percent of the coal-derived gas is converted to methanol at a reactor pressure of 750 psig [8].

The catalytic conversion of CO, CO_2 and H_2 to methanol (CH_3OH) is a very exothermic reaction, and involves the following reaction [10]:

$$CO_2 + 3\,H_2 \rightarrow CH_3OH + H_2O$$

The heat generated during methanol formation is considerable, with higher temperatures increasing the rate of methanol formation but also leading to side reactions to form methane, dimethyl ether, methyl formate, higher alcohols, and acetone. In addition, higher temperatures detrimentally affect catalyst life. The controlling mechanism for optimum methanol production is to maintain a high reaction temperature to achieve greater yields, but the reaction heat that is generated must be withdrawn quickly so as not to allow an undesirable rise in the reaction temperature.

Historically, high-pressure (3,000 to 6,000 psia) synthesis of methanol had been employed in the past [11]. The newer low-pressure and medium-pressure (450 to 1,500 psia) methanol synthesis processes require very high purity synthesis gas (so as to avoid poisoning the catalyst). There exists two distinctly different types of technologies that can be utilized to convert the synthesis gas into methanol. For this coal refinery concept, methanol synthesis is assumed to occur by the newer liquid-phase process due to its ability to directly process the CO-rich gas produced by advanced coal gasifiers [12] and potential lower costs than the traditional vapor-phase processes [13]. An analysis of combined-cycle generation with methanol production using conventional gas phase technology is given elsewhere [14]. Typically, the liquid-phase methanol process operates at a reactor pressure of 750 psig and temperature of 480° F.

An important advantage of liquid phase methanol formation is that the liquid medium in which the catalyst is suspended acts as a heat sink, so that the heat of reaction can be withdrawn quickly away from the catalyst surface. The temperature rise in the liquid phase is controlled by an internal heat exchanger, where steam is produced. Before entering the reactor, the synthesis gases pass through a final catalytic absorption stage (a carbonyl/sulfur guard bed) to ensure a high-quality synthesis gas as well as to prevent the breakthrough of sulfur. In the reactor, the gases bubble through a liquid phase containing the catalyst particles that convert the CO, CO_2 and H_2 into methanol and water. The resulting methanol exits the reactor in gaseous form, and is condensed and removed by the Vapor-Liquid Separator. Any slurry droplets entrained by the product gas flow are returned to the reactor by the cyclone separator, with the catalyst continuously removed and replaced by fresh catalyst without process shutdown.

The liquid-phase methanol process produces crude methanol of approximately 97.5 percent purity [15]. Higher alcohols (~ 1 percent) and water (1 to 3 percent) are retained in the fuel-grade product; the purity of the crude methanol is such that it can be used directly as a transportation fuel. If a higher purity level is desired (so as to produce a commercial grade methanol product for use as a chemical feedstock), the raw methanol product would be separated by a distillation plant.

The required oxygen stream for the gasifier is produced from air in the Air Separation Plant. Nitrogen (containing less than 5 ppm O_2) in excess of process requirements can be stored on-site as a liquid for sale as a marketable product. Another option (which is not applied here) is to return 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 unit thereby increasing electrical production [16].

The fuel for the Advanced Power Generation system is the portion of the synthesis gas that was unconverted after passage through the methanol synthesis reactor. The overall concept of the Advanced Power Generation system is similar to an Integrated Gasification Combined-Cycle (IGCC) plant [17]. The pressure of the unconverted synthesis gas is too great for introduction into the gas turbines to burn, and so it must be 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 (0.01 kWh per lb of run-of-mine coal fed). Compressed air is then mixed and burned with the synthesis gas and the resulting hot gases (at about 2,200° F) are expanded through the Gas Turbine Generator to produce the majority of the electric power (0.82 kWh per lb of run-of-mine coal fed).

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. This steam is sent to the Steam Turbine/Generator to produce electricity (0.53 kWh per lb of run-of-mine coal fed).

Based on this Texaco gasification design, the overall energy requirement for the methanol synthesis section is approximately 28 million Btu per ton of methanol produced, based on Illinois No. 6 Bituminous as the feed coal.

The electrical requirement for the overall process is approximately 0.21 kWh per lb of run-of-mine coal. The overall integration of this coal-based methanol plant allows utilization of the waste heat, with the majority of the power required for the individual compressors within the methanol synthesis plant supplied by high-pressure steam. The remainder is partially supplied by the 100 psig saturated steam. For this coal refinery design, an excess of 1.15 kWh per lb of run-of-mine coal is available for export as a saleable product.

Types of Feed Coal

This coal refinery concept has the ability to handle most coals, ranging from lignite to high-rank bituminous. Anthracite is typically not gasified due to its lower reactivity during gasification (requiring higher levels of steam and and oxygen consumption).

Coal characteristics such as reactivity, size consistency, ash-fusion temperature, grindability, and degree of coal caking can affect the choice and performance of a given gasifier.

Products

The main products from this example coal refinery are methanol (CH_3OH) and electricity. Marketable byproducts include sulfur (elemental) and liquid nitrogen (N_2). The product output is given in Table 3 in terms of pound product per pound of feed coal. This analysis shows that approximately 0.17 pounds of methanol and 1.15 kWh of electricity are produced from one pound of dry feed coal.

The product output depends on the type of feed coal (in that the coal affects the relative distribution of the light gases upon gasification) and the methanol synthesis technology.

Likely Applications

Methanol is one of the three primary basic chemicals (after ammonia and ethylene) and is a valuable chemical used in the chemical industry as a raw material. Traditional uses of methanol are in the synthesis of formaldehyde, methyl halides, methylamines, methyl methacrylate, dimethyl terephthalate, and as a solvent. Newer uses of methanol are in the synthesis of oxygenated chemicals: MTBE (methyl tertiary-butyl ether), acetic acid, acetic anhydride, ethanol, vinyl acetate and ethylene glycol. Potential uses of methanol are in the synthesis of petrochemical feedstocks: ethylene, propylene, butenes, benzene, toluene and xylene [7]. Approximately 50 percent of all methanol produced is processed to become formaldehyde [11]. In 1987, imports accounted for approximately 37 percent of this country's annual methanol requirement (approximately 1.8 billion gallons) [18]. Given that methanol is an industrial intermediate chemical, it would be expected that the chemicals industry may be interested in pursuing this coal refinery concept.

In the energy sector, methanol can be used as a transportation fuel, as a turbine fuel (e.g., for peak-shaving purposes in power generation in highly populated areas) or can be converted into gasoline with technology developed by Mobil Corporation [6]. In 1988, approximately 390 million gallons of methanol were converted into methyl t-butyl ether and blended into 112 billion gallons of gasoline used into this country [19]. A recent report by the Department of Energy (DOE) stated that methanol is an alternative fuel (to gasoline) that can see widespread use as a transportation fuel in the U.S. before the turn of the century [20]. The commercial transportation fuel suppliers industry may be interested in pursuing this coal refinery concept.

It has been stated that in the long run, electric utilities will replace natural gas with coal-derived gas produced in gasification combined cycle power plants. Coproducing 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 [4]. It is therefore to be expected that the most likely commercial entity interested in owning and operating a coal refinery employing once-through methanol production with electric generation would be within the electric utility market. Coproduced methanol could also serve as the fuel for multiple fuel cells in urban locations, thereby achieving the many advantages of dispersed generation.

Elemental sulfur has many uses, such as production of sulfuric acid and other inorganic chemicals. Liquid nitrogen (which contains argon and other noble gases) can be used in cryogenic applications, as an industrial gas, and in enhanced oil recovery.

Status of Development

A variety of gasification processes can be used for combined-cycle power generation, with the overall layout and operating conditions dependent on the gasifier type. EPRI has identified the Texaco, DOW, British Gas/Lurgi, and Shell combined-cycle gasifiers as the most likely commercial processes for electric utility application in the U.S.[21]. The Texaco gasifier used here for this example requires cooling of the syngas before acid gas cleanup. Other gasification technologies (e.g., the British Gas/Lurgi slagging gasifier) produce a lower-temperature syngas (800°F outlet temperature from the gasifier). However, the Texaco gasifier does not produce heavy tars and oils (due to the high gasifier temperature) typical of the British Gas/Lurgi technology which must be removed by cleaning and returned to the gasifier for further conversion.

There currently exists many commercial processes for methanol synthesis based on coal gasification; they include the Imperial Chemical Industries (ICI) Low-Pressure Process and the Lurgi Low-Pressure Process [8, 22]. This coal refinery concept uses the liquid phase process due to its flexibility in the syngas composition which does not limit the type of gasification technology or the coal type. Earlier demonstration runs by Air Products and Chemicals, Inc., at the Department of Energy's test facility in LaPorte, Texas showed that during a 124 day operation, the experimental methanol production facility produced more than 8,000 barrels of fuel-grade methanol. The methanol production rates averaged 60 to 70 barrels per day, about twice the facility's original 35 barrel per day design rate [23], with successful operation at high slurry concentration (50 percent by weight). Successful agreement of scaleup from bench scale to PDU operation has apparently been achieved.

The Liquid Phase Methanol Process had been proposed as an addition to the Great Plains Gasification Plant in Round 3 of the Clean Coal Technology program [24]. The plant was designed to produce 500 tons of methanol per day, using about 10 percent of the synthesis gas produced at Great Plains. An alternate site is however under consideration [25].

An economic analysis has been performed comparing the capital and levelized costs for an IGCC-only and an IGCC with once-through methanol production (IGCC/OTM) production plants in reference [13]. The analysis was based on the liquid phase methanol synthesis (LPMEOH) process for a 650-MW Texaco-based IGCC facility designed by Fluor [26].

Environmental Aspects

The main environmental intrusions from this coal refinery include atmospheric emissions of SO_2 and NO_x, and solid wastes. Integration of the IGCC portion within this coal refinery results in very low emissions of SO_2, NO_x and particulates. For this coal refinery, over 99 percent of the sulfur within the coal is captured, to eliminate potential poisoning of the methanol catalyst. The emission rate for SO_2 is approximately 0.0001 lb per lb of feed coal. The emission rate for NO_x is approximately 0.003 lb per lb of feed coal [17]. The SO_2 and NO_x emission rates would meet the limits allowed by NSPS. The release of particulates is negligible (0.0003 lb per lb feed coal, [27]) as a low level of particulate emissions is inherent in the Texaco gasification technology [16]. 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 [28].

The process water requirements are approximately one-third less than the requirements of conventional power plants (on the order of 0.7 gallons per lb dry feed coal, specific water consumption [17]) with no harmful pollutants detected [17, 27]. This value is dependent upon the type of gasifier and other design details.

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 [1]. 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 0.1 lb per lb feed coal.

Utilization of IGCC technology for electric power production can result in significant reductions in CO_2 emissions; an achieved thermal efficiency of 46 percent for an IGCC plant means a 15 percent reduction in the CO_2 emissions per kWh produced when compared with a conventional coal-fired power plant operating at 40 percent efficiency [28].

Research Needs

Bench-scale and PDU data indicate that the liquid phase methanol process to be technically well grounded. IGCC technology is near or at the commercial application stage, with several large IGCC projects having been launched during 1990 [29]. A reliable, efficient and economical unit would however be demonstrated by the successful integration of the many process steps of the different technologies.

The LPMEOH process has been proven technically using syngas blended from pure components. Demonstration of the process using actual synthesis gas from a coal gasifier and under variable load conditions would establish the overall technical viability of the LPMEOH Process for use in this coal refinery concept.

The overall economics will depend on the proportion of methanol to power production. Studies on the effect of coal type on methanol yield may lead to reduced levelized costs.

Catalyst deactivation rates of less than 0.2 percent per day have been achieved [30]. Process development work to lower the deactivation rate would improve the reliability and economics of the methanol production process.

Table 1: Feed Coal Analysis

ULTIMATE ANALYSIS (weight percentage; dry basis)						
Carbon	Hydrogen	Oxygen	Sulfur	Nitrogen	Chlorine	Ash
69.5	5.3	10.0	3.9	1.3	0.01	10.0
Free Moisture (weight %)			6.0			
Higher Heating Value (dry basis)			11,342 Btu per lb			

Table 2: Typical Yield of Various Compounds from Texaco Gasifier

COMPOUND	YIELD (mole percentage)
Carbon Monoxide, CO	41.0
Hydrogen, H_2	29.8
Carbon Dioxide, CO_2	10.2
Methane, CH_4	0.3
Nitrogen, N_2	0.7
Argon, Ar	0.1
Hydrogen Sulfide, H_2S	1.0
Carbon Oxysulfide, COS	0.1
Ammonia, NH_3	0.2
Water, H_2O	17.1

Table 3: Product Output for Coproduction of Methanol and Electricity

PRODUCT	OUTPUT (lb product per lb dry feed coal)
Methanol, CH_3OH	0.17
Electricity	1.15 kWh per lb dry feed coal
Elemental Sulfur	0.04
Liquid Nitrogen, N_2	2.99

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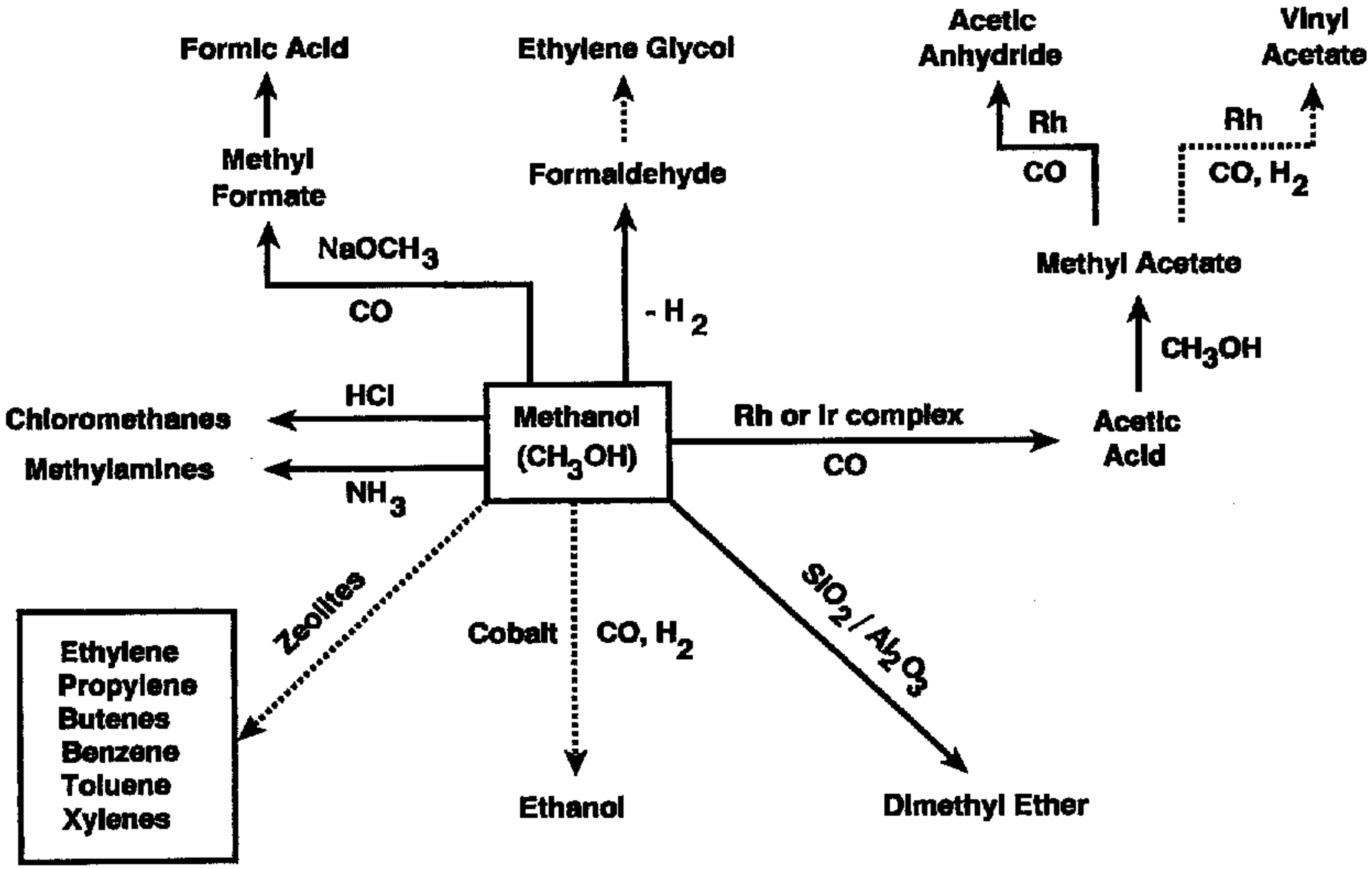

FIGURE 1: Some Chemicals from Methanol (- - - ➤ not yet commercial) [3]

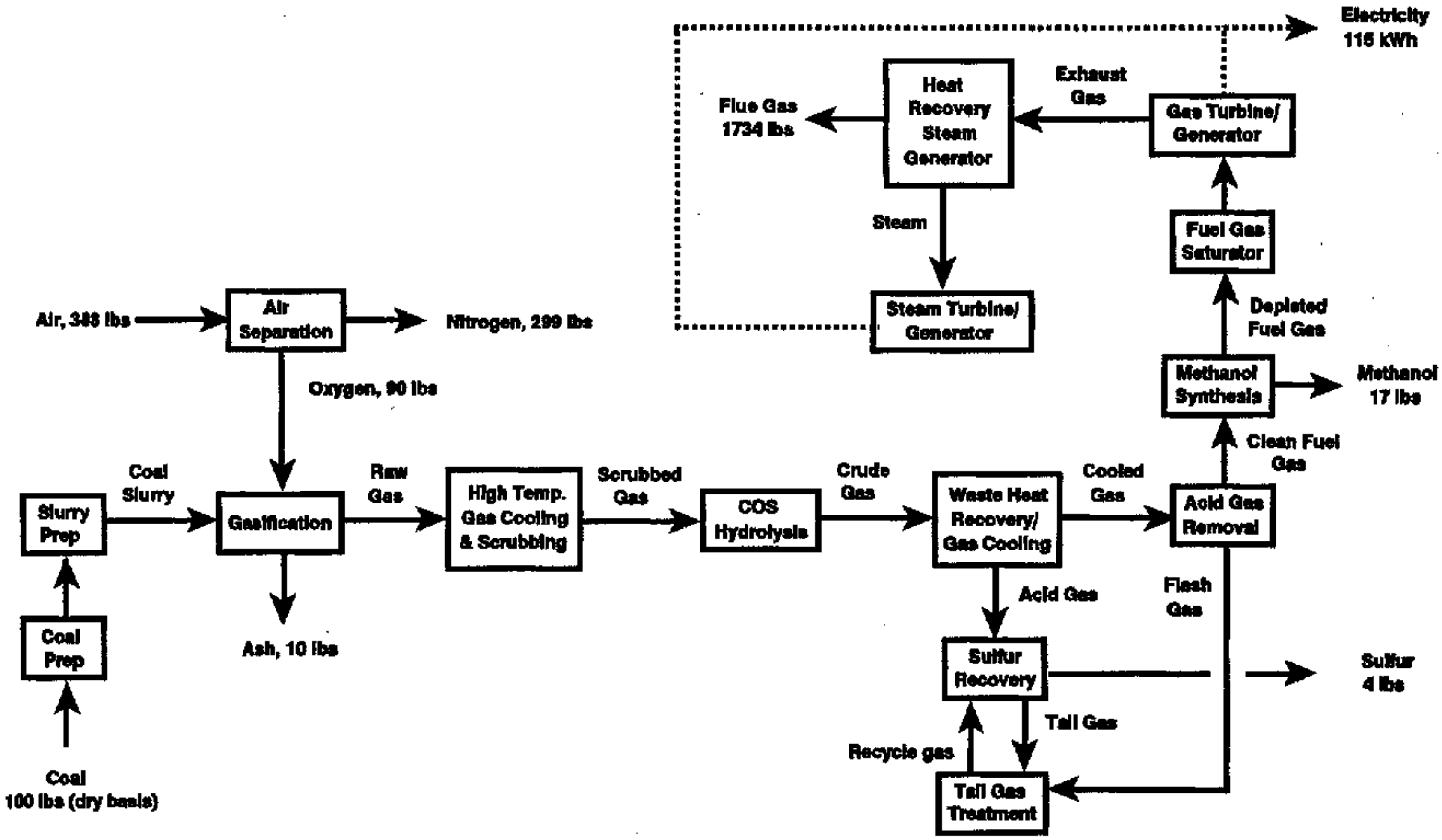

FIGURE 2: Overall Block Diagram of the Coproduction of Methanol and Electricity Concept