I. Introduction/Summary

The purpose of this paper is to estimate the thermal efficiency, capital investment and total cost of indirect coal liquefaction processes. Over the past five years many study designs have been performed on the production of methanol and other indirect liquids from coal. Some of these are original designs, while others are secondary studies, taking one or more original designs and adjusting economic parameters, etc. Figure 1 shows the available indirect coal liquefaction studies and their chronology and interrelationship. Since the secondary studies only modified the economic basis of the original studies referenced and not the basic design and each secondary study used a different basis, preventing intercomparison, this study will restrict itself to the original studies and attempt to place them all on one single comparable basis. The following is a list of these original studies:

"Screening Evaluations: Synthetic Liquid Fuels Manufacture," Ralph M. Parsons Company for EPRI, August, 1977, EPRI AF-523.[1] (This report estimates the cost of methanol from four different gasification technologies, Foster-Wheeler, BGC-Lurgi, Koppers-Totzek, and Texaco, with Chem Systems methanol synthesis. The study also looks at the Fischer-Tropsch process following BGC-Lurgi gasification.)

"Coal to Methanol Via New Processes Under Development: An Engineering and Economic Evaluation," C.F. Braun and Company for EPRI, October, 1979, EPRI AF-1227.[2] (This report covers two coal to methanol processes: Illinois No. 6 coal to methanol via Texaco gasification and Chem Systems methanol synthesis, and Wyodak coal to distillate fuel and vacuum residual oil via a non-catalytic hydroliquefaction process in which the residual oil is processed into methanol by the same process as the coal.)

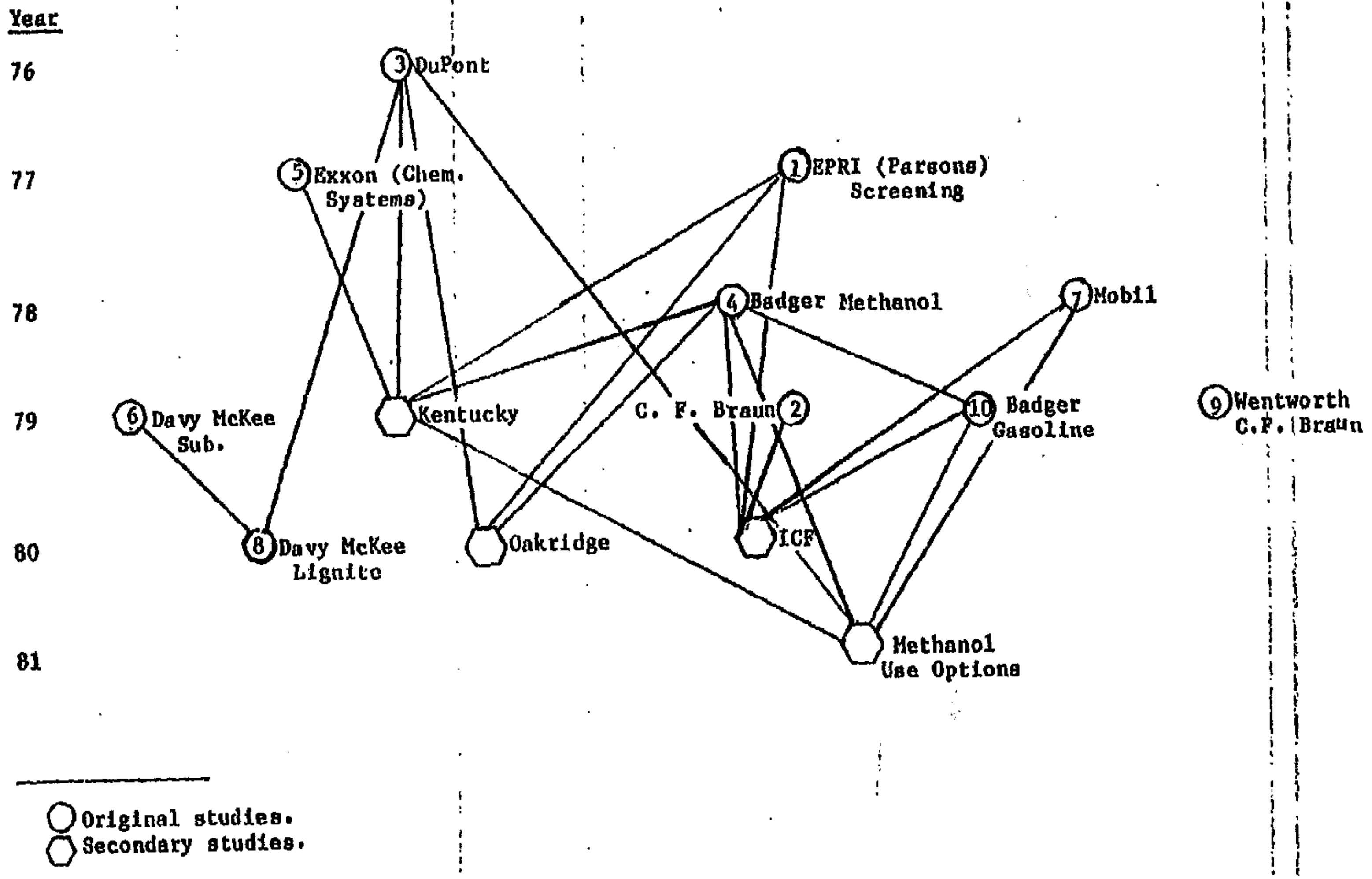
"Economic Feasibility Study, Fuel Grade Methanol From Coal For Office of Commercialization of the Energy Research and Development Administration," McGeorge, Arthur, DuPont Company, for ERDA, 1976, TID-27606.[3] (Eastern coal to methanol via Texaco gasification with ICI synthesis.)

"Conceptual Design of a Coal-To-Methanol Commercial Plant" (Vols. I-IV), Badger Plants, Inc., for DOE, February, 1978, FE-2416-24.[4] (Eastern coal-to-methanol via Lurgi "slag-bath" gasification and Lurgi low pressure methanol synthesis technology.)

"Production Economics for Hydrogen, Ammonia, and Methanol During the 1980-2000 Period," Cornell, H.G., Heinzelmann, F.J., and Nicholson, E.W.S., Exxon Research and Engineering Co., April, 1977.[5] (Eastern coal to methanol via Koppers-Totzek and Shell-Koppers gasification with ICI synthesis.)

Figure 1

Methanol Report "Tree"



"Methanol From Coal, An Adaptation from the Past," E.E. Bailey, (Davy McKee), presented at The Sixth Annual International Conference, Coal Gasification, Liquefaction and Conversion to Electricity, University of Pittsburgh, 1979.[6] (Subbituminous coal to methanol via Winkler gasification and ICI synthesis.)

"Research Guidance Studies to Assess Gasoline From Coal By Methanol-To-Gasoline and Sasol-Type Fischer-Tropsch Technologies - Final Report," Schreiner, Max, Mobile R&D Company, for DOE, August, 1978, FE-2447-13.[7] (Comparison of eastern coal to methanol and SNG, and gasoline and SNG by Lurgi gasification/Lurgi synthesis/Mobil MTG with gasoline from Lurgi gasification and Fischer-Tropsch synthesis.)

"Lignite-to-Methanol: An Engineering Evaluation of Winkler Gasification and ICI Methanol Synthesis Route," DM International, Inc. for EPRI, October 1980, EPRI AP-1592, Project 832-3.[8] (Lignite to methanol via modified Winkler and ICI synthesis.)

"Production of Methanol from Lignite," Wentworth Bros., Inc., and C.F. Braun and Co., for EPRI, September 1979, EPRI AF-1161, TPS-77-729.[9] (Lignite to methanol via Texaco gasification and WBI synthesis.)

"Conceptual Design of a Coal-to-Methanol-to-Gasoline Commercial Plant," for DOE, March 1979, FE-2416-43.[10] (Adds Mobil process to methanol design of study no. 4 above.)

Methanol: To estimate the cost of producing methanol, all of the design studies were: 1) normalized to a production yield of 50,000 fuel oil equivalent barrels per calendar day (FOEB/CD) and a common financial basis and 2) inflated to $1981, as discussed in a previous report.[10a] Of the thirteen designs contained in the above ten studies, nine used bituminous coal, two used subbituminous coal and two used lignite as a feedstock. The studies included eight different coal gasification technologies (Foster-Wheeler (1), BGC/Lurgi (1), Koppers-Totzek (2), Texaco (4), Lurgi (1), "Slag-Bath" (1), modified Winkler (2) and Koppers-Shell(1)) and four different types of methanol synthesis processes (Lurgi (2), ICI (5), Chem Systems (5), and Wentworth Bros. (1)). The Winkler, Lurgi and Koppers-Totzek gasifiers have been proven on a commercial scale and the Texaco process is very close to commercialization. Of the synthesis units, ICI and Lurgi are used extensively today. Wentworth Bros. claim that their process is commercial and Chem Systems is a new process which is still being tested.[21] Lurgi and ICI have been competing for the last ten years and both have highly developed processes, good efficiencies and, according to Parsons,[1] room for further improvement is small. In addition, Parsons states that the Chem Systems process only shows a slightly higher thermal efficiency and lower capital cost than the ICI system. Since the costs of the proven ICI and Lurgi synthesis processes are indistinguishable and it appears that the cost for the Chem Systems process is only slightly lower, it has been decided to place most of the emphasis here on the effect of the various gasification technologies costs which appear to be more significant.

The original ranges of product costs and capital costs reported by the thirteen studies are very large due at least in part to the large range in plant size ($3.74-12.55 per mBtu for product cost and $0.401-$5.05 billion for capital, $1981, for plants ranging from 2,000-58,000 ton per day of methanol). With this type of data it is very difficult to estimate the actual cost of methanol, let alone compare it with any other coal technologies. After normalizing the costs for the thirteen studies the ranges of costs are much smaller.

For bituminous coals the product cost ranged from $4.65-9.05 per mBtu for the low capital charge rate (CCR) and $8.14-12.54 per mBtu for the high CCR. The gasifiers used in these studies are Foster-Wheeler, BGC-Lurgi, Koppers-Totzek, Lurgi Slag Bath, and Texaco(2).

Of these gasifiers the Koppers-Totzek is proven, and the remainder represent advanced technology. The cost of methanol from these gasifiers are presented in Table 1. When using the Koppers-Totzek gasifier the cost ranges from $7.23-12.42/mBtu depending on the capital charge rate; for the Texaco gasifier the cost ranges from $5.90-6.48 and $9.44-10.41/mBtu; for the other advanced technology the cost ranges from $5.30-6.08 to $8.74-9.78/mBtu.

Both the Texaco and Koppers-Totzek gasifiers are entrained bed units which seems to emphasize the statement that entrained bed gasifiers are the only commercially-available (or near-commercial) reactors today which can economically gasify eastern bituminous coals (eight of the nine cases investigated used entrained bed gasifiers).

The range of instantaneous plant investment for the nine cases was $1.93-$2.92 billion (50,000 FOEB/CD plant). As shown in Table 1, the instantaneous plant investment for the methanol plant using bituminous coal ranged from $1.99 to $2.21 billion when the Texaco gasifier was used, $2.92 when the Koppers-Totzek gasifier was used, and ranged from $1.93-2.22 billion when the other advanced technology gasifiers were used.

The range of product and capital costs for methanol from subbituminous coals and lignite are smaller than that of bituminous. Of the two studies using subbituminous coals, one uses proven gasification and synthesis technology, Lurgi/Lurgi [7], while the other uses a gasification technology which the manufacturer claims is "here now," and a proven synthesis process, modified Winkler/ICI.[6] The average product cost range is fairly

Table 1

Product and Capital Costs of Selected Coal Liquefaction Processes(1981 Dollars)

		Product Cost ($/mBtu)		Capital Cost** (Billions of Dollars)
Indirect Liquefaction	Product Mix	11.5% CCR	30% CCR	
Process				
Texaco (Bituminous)[2,3]	100% MeOH*	5.90-6.48	9.44-10.41	1.99-2.21
Koppers (Bitum.)[1]	100% MeOH*	7.23	12.42	2.92
Advanced Technology (Bituminous)[1,4]	100% MeOH*	5.30-6.08	8.74-9.78	1.93-2.22
Lurgi (Subbit.)[7]	47.9% MeOH*	7.04	12.48	2.59
	49.7% SNG	5.63	9.98	
	2.4% Gasoline	7.04	12.48	
Modified Winkler (Lignite)[8]	100% MeOH*	5.70	9.56	2.17
Texaco (Lignite)	100% MeOH*	6.92	12.24	3.00
Lurgi-Mobil MTG (Subbit.)[7]	41.2% Reg. Gasoline	8.01	14.35	2.95
	53.3% SNG	6.41	11.48	
	5.5% LPG	6.25	11.20	
Mobil MTG Incremental Cost	85-90% Reg. Gasoline 10-15% LPG	1.45	2.87	0.68
Fischer Tropsch[7]	1.8% LPG	6.56	11.36	3.00
	64.5% SNG	6.82	11.80	
	2.6% Alcohols	8.52	14.75	
	25.3% Gasoline	8.52	14.75	
	4.6% Diesel Fuel	7.67	13.28	
	1.3% Heavy Fuel Oil	6.56	11.36	

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* MeOH = 95-98% methanol, 1-3% water, and the remainder higher alcohols.

** Instantaneous capital costs.

small, \$6.16-\$6.34 per mBtu for the low CCR and \$10.26-\$11.24 per mBtu for the high CCR. The instantaneous plant investment range is \$2.10-\$2.59 billion. Although the costs seem to compare favorably, only the Lurgi/Mobil prices are shown in Table 1. This is because the modified Winkler/ICI plant size had to be scaled up significantly where as the Lurgi/Mobil plant size was much closer to the selected 50,000 FOEB/CD and was therefore considered more accurate.

Four lignite cases were studied. However, two of these cases used Texaco gasifiers with coal slurry concentrations which are still in a developmental stage. The other cases involved the Texaco gasifier (at an appropriate coal slurry concentration) and the Winkler gasifier. At this slurry concentration the Texaco gasifier appeared to have a large economic disadvantage relative to the Winkler gasifier, so the Winkler was chosen as the best design. The resulting product costs for the low and high CCRs are \$5.70 and \$9.56/mBtu, respectively. The instantaneous investment plant cost is \$2.17 billion. These costs are shown in Table 1.

In summary, the prices which have been chosen for this study represent two commercially proven gasification technologies: Koppers-Totzek and Lurgi, a modified Winkler, for which its manufacturer will back financially, and the near commercial Texaco gasifier. For bituminous coals, the Koppers-Totzek prices are higher than Texaco's because the former operates at atmospheric pressure.

MTG: To evaluate the cost of producing gasoline from coal utilizing Mobil's methanol-to-gasoline (MTG) process, two different studies by Mobil and Badger were analyzed in the same manner as the methanol studies.[7,10] Gasoline costs from these two studies varied widely. Therefore, it was preassumed that incremental product and capital costs for Mobil's MTG gasoline relative to methanol could be determined from both studies and be more accurate since methanol costs (capital and product) were available for the same technology by the same designers.[7,4] When the cost of gasoline was compared to that of methanol, the incremental cost of gasoline for both studies was very close. Since the MTG process is a patent of Mobil's, it is believed that their study is more reliable; therefore their costs were used in preference to Badger's, which were slightly higher.

The Mobil study analyzed a few different cases with respect to the Mobil MTG process. The most economical was the case which produced gasoline and SNG as the major products. For this case, the average product cost ranged from \$7.06-12.65 depending on the CCR. The total instantaneous plant investment was \$2.95 billion. These costs are shown in Table 1. By comparing this case with Mobil's other case (methanol from Lurgi gasification of subbituminous coal) an incremental cost of gasoline relative to methanol was determined. Based on a 50,000 FOEB/CD MTG unit, the incremental cost of gasoline over methanol was found to range from $1.45-2.87 per mBtu depending on the CCR. The incremental instantaneous investment was found to be approximately $680 million for a plant producing all methanol and then gasoline. These costs are also shown in Table 1.

Fischer-Tropsch: There were two studies which investigated Fischer-Tropsch synthesis technology, Parsons and Mobil.[1,7] Since the Mobil study was based on a more thorough design than the Parsons study, its costs were used in preference. The instantaneous plant investment cost for the Mobil case was $3.00 billion. Its average product cost ranged from $7.60-13.38 per mBtu depending on the CCR. The costs of the products from this case are presented in Table 1. The Mobil study was also used to determine the average product cost difference between Fischer-Tropsch synthesis and methanol synthesis plants. The instantaneous plant investment difference is $355 million and the operating cost difference is $67 million with the Fischer-Tropsch case costing more. The figures translate into an average product cost difference of $1.00/mBtu.

II. History of Methanol Production

The primary source of all methanol prior to the 1920's was the destructive distillation of wood. In this pyrolysis process air was excluded while the wood was heated to a temperature of 160-400 degrees Centigrade. As the components of the wood heated they volatized and thermally decomposed. The products were separated into gases and a condensed liquid called pyroligneous acid. Upon further distillation this liquid could be separated into acetic acid, acetone and rather impure methanol. Since the yield was three to six gallons per ton of wood, the product was very expensive.[12]

During the pre-World War I period, the development of a synthetic methanol process began in Germany and France. Between 1910 and 1916 there were several patents issued in Europe describing the chemical reaction of carbon monoxide (CO) and hydrogen (H_2) to form alcohols, ketones, aldehydes, etc. The reaction was carried out at temperatures of 300 to 400 degress Centigrade and at pressures at or above 1500 psi. Catalysts containing chromium, zinc, maganese and cobalt, or their oxides were used to help the conversion of the carbon monoxide and hydrogen to methanol.[12]

In 1923, BASF in Germany became the first company to produce commercial-scale synthetic methanol. The U.S. started importing synthetic methanol produced from coal or coke in 1924. Soon Commercial Solvents Corporation and DuPont became interested and by 1928 each had a commercial plant producing methanol in the U.S.[12]

When coal and coke derived synthetic methanol hit the U.S. market there was an enormous difference between the price of natural and synthetic methanol. Natural methanol cost 68 cents per gallon while synthetic methanol could be made for 36 cents per gallon. The price competition was so great that the natural wood distillers united and managed to persuade the tariff commission to increase the import tariff to 18 cents per gallon. They also were able to get legislation passed which mandated the use of natural methanol to denature ethanol, thus securing a third of the total methanol market.[12]

The wood distillers managed to keep the price of natural methanol competitive for a number of years through consolidation and larger more efficient plants. However with the large discoveries of petroleum and natural gas and the mass production of high-purity methanol, the synthetic manufacturers were soon able to lower the price of methanol beyond reach, leaving natural methanol producers to their captive denaturant market.[23]

The first plants were built in conjunction with other plants to make use of carbon dioxide or hydrogen by-products. However as the demand for methanol grew, plants were built specifically for methanol production. The first feedstock to be gasified to carbon monoxide and hydrogen was coal. Later the feedstock was shifted to oil and then natural gas as large discoveries of these sources were made and their prices dropped. Natural gas was an ideal feedstock because it contained very little, if any, sulfur and its price was very low. Thus by the 1960's, synthetic methanol in the U.S. was almost entirely produced from natural gas utilizing the high-pressure methanol synthesis process.[23]

By 1967 the combination of a common feedstock, comparative technology and a competitive market had stabilized the price of methanol at 27 cents per gallon. However, in 1967, Imperial Chemical Industries (ICI) introduced a newly developed low-pressure synthesis process based on a copper-zinc-chromium catalyst in place of the zinc-chromium catalysts previously used. Since these copper catalysts were more reactive than the others, lower operating pressures and temperatures could be used. In fact, by the latter part of 1971, the selling price of methanol had dropped to 11 cents per gallon.[23]

In the 70's the increasing cost of production, the demand for low-sulfur natural gas and the OPEC oil embargo of 1973 brought into focus the energy crisis and the finite supply of fossil fuels. The tripling of oil prices and doubling of the cost of natural gas caused the price of methanol to triple between 1973 and 1975 (14 cents/gal to 42 cent/gal). Since 1975 the price of methanol has continued to increase with that of natural gas. The current price for methanol is between 72 and 80 cents per gallon.[23]

Between the time it was first introduced into the U.S. and today, methanol has exhibited a dramatic growth. For the first 45 years there was 13.7 percent average annual growth rate.[12] In the 1930's plant sizes ranged from 20 to 40 tons of methanol per day. By the early 50's the size has risen to 150-200 tons/day. In the 70's the capacity has gone from 1,500 to 2,000 tons/day to single trains of 5,000 tons/day.[23]

Methanol production in the United States is now near 4 million tons per year or about 70,000 barrels per day (BPD). Virtually all of this is produced from natural gas.[12] The natural gas (essentially pure methane) is reformed with steam to produce a synthesis gas consisting mainly of carbon monoxide and hydrogen. After purification, the synthesis gas is compressed and combined in a catalytic converter to produce methanol. The reaction is highly exothermic while the conversion per pass is relatively small (2-10 percent). Large volumes of unconverted gas are recycled through the converter in order to achieve high overall conversion and to assist in removing the exothermic heat of reaction. Overall CO conversions of 96 to 99 percent can be obtained.[13]

III. The Methanol Production Process

The basis of all processes for manufacturing synthetic methanol is the catalytic reaction of carbon monoxide and carbon dioxide with hydrogen to produce methanol.[12] These reactions are shown below.

Carbon Monoxide + Hydrogen = Methanol

$$CO + 2H_2 = CH_3OH \tag{1}$$

Carbon Dioxide + Hydrogen = Methanol + Water

$$CO_2 + 3H_2 = CH_3OH + H_2O \tag{2}$$

The source of carbon monoxide or carbon dioxide is usually derived from the partial combustion of a hydrocarbon fuel such as coal, coke, natural gas, naptha, or a heavy petroleum fraction.

The primary source of hydrogen is water and the hydrogen contained in the feedstock, which is the case of coal is very low (3-6 percent). The reactions shown in Equations (1) and (2) are carried out at pressures between 750 and 4500 psi at a temperature of 250 to 350 degrees Centigrade in the presence of a metal or metal oxide catalyst. The metals used depend upon the process, and are usually proprietary. Catalysts may contain zinc, chromium, or copper-based compounds or oxides.

A description of a typical coal to methanol process follows:[11]

Coal Receiving and Preparation: Vibrating feeders transport the coal to the sizing equipment, ring mill crushers and rod melts where the coal is sized for the specific gasifier in which it will be processed.

Coal Gasification: The coal is heated to very high temperatures and partially-oxidized to carbon monoxide and hydrogen in the presence of oxygen (or air) and steam. The majority of the sulfur is converted to hydrogen sulfide with some production of carbonyl sulfide. The nitrogen in the coal is converted to free nitrogen combined with some traces of ammonia and hydrogen cyanide. The ash is removed from the bottom in a dry or molten slag depending on the temperature and gasification technique used.

Gas Cooling: The hot raw gas is cooled and scrubbed with recycle gas liquor or sour water from the shift converter. Then the gas is cooled further in a heat exchanger where steam is produced by the waste heat.

Gas Shift: Here the ratio of hydrogen to carbon monoxide is increased by adding steam and pushing the following water-gas shift reaction to the right: $CO + H_2O = CO_2 + H_2$.

Acid Gas Removal: In this process the sulfur is removed from the synthesis gas to prevent poisoning of the methanol synthesis catalyst. In the Selexol process hydrogen sulfide is removed first, and then carbon dioxide and carbonyl sulfide are removed. In the following Rectisol process, naptha, HCN and water are removed by washing the gas with a small quantity of methanol.

Methanol Synthesis: In this stage clean shifted gas is catalytically converted into crude methanol by the following two reactions:

$$CO + 2H_2 = CH_3OH \text{ and } CO_2 + 3H_2 = CH_3OH + H_2O.$$

Auxiliary Facilities: The functional relationships of the auxiliary facilities to the major process areas are as follows:

Water Supply - provides for treatment, storage and distribution of process water requirements, including makeup cooling water.

Water Cooling - provides for treatment, storage and distribution of process cooling water.

Oxygen Production - cryogenically separates air into oxygen and nitrogen. Oxygen is used in coal gasification. Some of the nitrogen is used in carbon dioxide removal, the remainder being vented to the atmosphere.