

## METHANOL FROM COAL

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## METHANOL FROM COAL

## INTRODUCTION

Destructive distillation of wood originally provided the source of methanol alcohol. Other developments in the wood industry, namely plywood, for which nearly 40% of the production of methanol finds its way with forms of formaldehyde and associated resins have promoted its use and applications.

U.S. annual production in 1973 was close to one billion gallons or the equivalent of a 10,000 tons per day plant. The corresponding world production was 25,000 tons per day. By reason of its price and availability in recent years, natural gas was the preferred feedstock.

Some of the properties of methanol are given in Table 1.

Table 1      Properties and Other Data on Methanol

Composition	CH <sub>3</sub> OH		
Appearance	Clear, colorless		
Density	.792		
Vapor Density	1.11		
Boiling Point	148 <sup>o</sup> F		
Flash Point	52 <sup>o</sup> F		
Ignition Temperature	800 <sup>o</sup> F		
Explosive Limits (air)	Lower	6.0%	
	Upper	36.5%	
Calorific Values HHV	BTU	BTU	BTU
	per lb.	per gal.	per bbl.
Fuel Grade Methanol	9,760	64,771	2.72 million
No. 2 Fuel Oil	19,000	135,000	5.77 million
Western Coal-dry basis	10,345		

## Conversion Factors:

$$\begin{aligned}
 1 \text{ Ton} &= 301.7 \text{ gal.} = 19.5\text{MM BTU} \\
 \$30.28 \text{ per ton} &- 10 \text{ c/gal.} = 1.54 \text{ \$/MM BTU}
 \end{aligned}$$

Recent developments have brought about a situation that gaseous feedstocks no longer prevail as a readily available raw material and alternative sources must be considered for the production of synthesis gas, coal being a strong contender.

The extent of the coal reserves has been compared by Linden<sup>1</sup> with other sources of energy and represents a substantial percentage of the recoverable fossil fuel reserves in the U.S.

Table 11                      U.S. Fossil Fuel Reserves

	<u>Proven &amp; Currently Recoverable</u>	<u>Estimated Total Remaining Recoverable</u>
Natural Gas	0.3 Q	1.5 Q
Natural Gas Liquids		0.1 Q
Crude Oil	0.2 Q	2.2 Q
Shale	0.4 Q	6.0 Q
Coal	4.3 Q	28.9 Q
Total	5.2	38.7

$$Q = 10^{18} \text{ BTU}$$

In items of heating value, coal represents 75 to 80 percent of these resources. At the current rate of consumption, 650 million tons per annum, these coal reserves can last well into the next century.

In comparison, a 5000 ton per day fuel grade methanol plant would consume about 4 million tons per annum of coal and give a daily output of about the equivalent of 90-95 MMSCFD of natural gas.

The applications of fuel grade methanol as alternate fuels have been reported extensively in the literature. Recent interest as a feed for gas turbines has been reported by Power & Marine Systems a subsidiary of United Technologies Corporation with tests at the St. Petersburg installation of Florida Power Corporation.

#### METHANOL FROM COAL

##### Historical Background

The price and availability of natural gas led to the early introduction in the U.S. of this material as a feedstock for the production of synthetic methanol. The conversion of the 30 ton per day plant in Peoria in 1932 to a methanol-from-natural gas unit was the forerunner of the industries transformation leading to plants up to the 2,000 tons per day range which have been in successful operation over the last few years.

Prior to the advent of natural gas, solid fuels had been the main sources of the raw material for the production of synthesis gas. In areas such as Europe, Asia and S. Africa where natural gas was not available, coal became established as the backbone of the ammonia and methanol industries and where suitable economics prevail these plants continue to operate. Recently a plant was commissioned in Modderfontein which manufactured both ammonia and methanol from coal. Hence, the technology is still available and can be readily updated to

suit U.S. conditions. Present designs incorporate the improvements which have evolved since the early 1920's when the plants first came into operations with high pressure synthesis processes.

The last unit to employ coal as a raw material for methanol synthesis in the U.S. was started up in Belle, W. Virginia using a B & W/DuPont oxygen blown gasifier and operated for about 15 months, gasifying approximately 400 T/D of coal to produce 24 MMSCFD of synthesis gas; about 1/3 of the output required for the 285 ton/day high pressure methanol facility that was in operation on that site.

Other installations that manufactured methanol from coal were located in Billingham Heysham and Dowlais in the United Kingdom (cyclic water-gas gassification with H.P. methanol). Leuna in E. Germany (Winkler gassification with H.P. methanol and Mayengibe Paris, France (Koppers-Totzek).

#### Methanol Synthesis

The growth of the synthetic methanol business from one million gallons per annum in 1927 to 80 million gallons per annum in 1947 has been described by Kastens, Dudley and Troeltzsch.

Recent interest in methanol as an alternate fuel has resulted in a number of conferences and papers. A critical analysis of these latest developments was given by McGhee<sup>4</sup> at the Engineering Foundation Conference in New Hampshire in July 1974. Whilst emphasizing the reduction in energy requirements from 4 to 2 BTU per BTU of methanol, his paper contains a useful bibliography on the new low pressure 50/100 atm process, as compared to the high pressure up to 1,000 atm processes.

The amount of natural gas being flared was mentioned by Harrison<sup>5</sup> in a recent symposium on synthetic fuels. This gas could be utilized as part of a concept of transporting energy in the form of fuel grade methanol as opposed to LNG and it has been demonstrated that designs of units of 5,000 tons per day capacity are perfectly feasible and incorporate features of plants already in existence. In addition, checks with equipment suppliers indicate that such items as the methanol converter can be manufactured in the sizes required for a jumbo methanol unit.

Schemes have been proposed for up to 25,000 tons per day of methanol (corresponding to 480 billion BTU/day of product). These should be compared with the coal to SNG projects based on western coals which have normal capacities of 250 billion BTU/day of pipeline quality gas at 1,000 psi.

Later in this paper we will touch briefly on the economics of these large size units and the change in emphasis in capital investment which is about 20% for methanol synthesis in a coal based unit compared with about 40% in a gas based unit.

### Coal Gasification

Early gasification processes date back to the last century; for example in 1883 Ludwig Mond designed a producer gas unit employing air as the gasifying medium for 200 tons/day of coal. This type of unit became the forerunner for the semi-water gas plant designed to produce synthesis gas at low pressure to feed the Haber process for ammonia which went into operation in Germany at Oppau in 1913. Synthetic methanol was first produced on the industrial scale by BASF in Germany in 1923. Coke from gas ovens often was the most popular feedstock for these cyclic plants of which well over 1,000 units were put in operation.

Gasification of inferior fuels such as brown coals and lignite developed with the invention of the fluidized bed Winkler process in 1926 which also had the advantage of being a continuous process. To date, there have been built 36 units in 16 plants around the world.

Some of the disadvantages of gas compression from atmospheric pressure was overcome by the development between 1933/36 of the Lurgi process. This is somewhat offset by the high methane content of the gas produced as well as the need for extra equipment to deal with naphthas, tars and phenols which appear as by-products. To date, about 63 units in 13 plants have been put into operation and there has been considerable interest in the pilot plant work carried out on the Lurgi plant in Westfield Scotland to methanate the gas to produce SNG.

Where oxygen and steam are used as the gasification medium, the Koppers-Totzek process can handle most types of coal in the entrained fuel gasifier which requires pulverized fuel. The higher temperature of gasification, 2200°F, results in no heavy hydrocarbon being carried forward, and the gas, after purification, can be utilized for production of methanol or ammonia. Fifty-two units of this type have been reported in 20 plants worldwide.

Although there are many other processes in different stages of development, it can be seen that coal gasification is not a new and untried field of operation and indeed as recently as 1955 has been used for the production of synthesis gas in the U.S.

To prepare the raw gas from the gasification section for methanol synthesis requires several additional processing steps, namely:

1. compression
2. shift conversion
3. acid gas removal

all of which have been in commercial operation for many years. Each of these processing steps can be found in any modern day ammonia or methanol plant, regardless of the feedstock being processed.

The exact combination of process steps to convert coal to methanol will vary depending upon the gasification scheme selected and the economics of by-product production. Figures 1, 2, 3 and 4 present four possible alternatives.

As a typical example, the following (see Figure 5) is a more detailed description of a methanol from coal facility based on the Winkler gasification technology available from Davy Powergas:

#### Winkler Coal Gasification

**Coal Preparation.** The run-of-mine coal from storage pile is conveyed to the crushers where the coal is crushed to a particle size of  $\frac{3}{8}$ " x 0. If predrying of the coal is required, fluid bed dryers may be utilized to reduce the moisture level to that required. In the dryer, hot air, heated by the combustion of coal, is used to fluidize the coal and supply the heat necessary for drying. Most of the dried coal is removed directly from the fluid bed. However, a portion is entrained in the hot gases leaving the dryer. A cyclone recovers most of the entrained coal, and it is returned to the dryer product coal and conveyed to the gasification section. The hot gases from cyclone are scrubbed with water for particulate matter removal before venting to atmosphere.

**Coal Gasification.** The coal feed is conveyed to the gasifier through lock hoppers and screw conveyors. The gasifier is maintained as a fluidized bed operation under moderate pressure. A mixture of steam and oxygen is injected at several points within the bed to gasify the coal while steam alone is injected into the bottom most level to fluidize the coal and to cool the larger ash particles discharging from the gasifier bottom. The high bed temperatures, typically 1700-2200°F, are obtained by the partial combustion of the coal's carbon and contained hydrocarbons. Due to the relatively high temperature of gasification, the tars, gaseous hydrocarbons and carbon present in the coal are converted to carbon monoxide, hydrogen, and carbon dioxide. Only a small percentage of methane remains in the raw product.

The primary coal gasification reactions are:



At a constant coal feed rate to the Winkler generator, the ratio of oxygen and steam to coal is controlled to maintain the desired bed temperature. Optimum bed temperature is a compromise between product gas calorific value, carbon efficiency and overall thermal efficiency, but is limited by the ash softening temperature. If the ash softening temperature is exceeded, the ash may fuse and agglomerate, thus upsetting the fluidization characteristics of the bed and possibly plugging the reactor.

As a result of the fluidication, the particles of ash and their contained carbon are segregated according to size and specific gravity, i.e., the heavier particles fall down through the fluidized bed and pass into the ash discharge unit at the bottom of the generator while the lighter particles are carried up out of the bed by the product gas. Approximately fifty to seventy-five percent of the incoming ash will be entrained in the hot product gases leaving the top of the Winkler generator. The exact quantity entrained with a given gas velocity, is primarily dependent upon the particle size distribution of the feed coal. Since the height of the fluidized bed is relatively small compared to the total height of the generator, the upper or major portion of the generator is available to perform two other functions; firstly, to further gasify any entrained carbon particles, and secondly, to effect a separation of any heavier solid material. To aid this further gasification, a portion of the steam and oxygen is added to the generator near the upper limit of the fluid bed.

The unreacted carbon in the discharged ash is a function of gasifier temperature and coal reactivity. Generally, reactivity varies inversely with geological age, lignites being the most reactive.

#### Gas Cooling and Particulate Removal

The hot gases leaving the generator pass through the heat recovery train where heat is removed from the gas by generating and super-heating high pressure steam and preheating boiler feedwater. The hot Winkler product gas is cooled to approximately 300°F in the heat recovery train. High pressure steam in excess of that required by the process is generated and therefore available to drive the product gas compressors and/or the compressors in the air separation plant.

The heavier char particles leaving the bottom of the gasifier pass out of the system and the balance of the char is carried out of the gasifier in the overhead product gas. The bulk of this char is removed in the heat recovery train and in cyclones. In combination, the heat recovery train and cyclones are designed to remove approximately 85% of the entrained solids. The char thus removed will be utilized as supplementary fuel in the boiler house. The gas from the cyclone flows through the venturi scrubber where the remaining char is removed to a level of 1 grain/1,000 SCF.

#### Production of Methanol

The methanol plant contains the following sections to process the raw gas from the coal gasification plant:

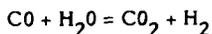
1. Raw Gas Compression
2. Gas Shift Conversion
3. Acid Gas Removal

4. Methanol Synthesis and Purification
5. Sulfur Recovery
6. Oxygen Production for Coal Gasification
7. Offsites

A description of each section is as follows:

Raw Gas Compression. The methanol synthesis is conducted at approximately 100 Ats. and it has been found that it is more economical to compress the gas exiting the gasification unit prior to further processing. The cooled and the dust free gas from the coal gasification section is compressed to 1480 psig in steam turbine driven compressors.

Gas Shift Conversion. The purpose of the shift conversion step is to adjust the ratio of carbon monoxide to hydrogen to that required for the methanol synthesis according to the exothermic shift reaction:



An exit CO content of 6.4 volume percent in the shifted gas is achieved in a one stage shift conversion reactor and a part of the compressed gas is bypassed around the shift reactor, cooled and mixed with the cooled shift effluent to yield an average CO content of approximately 20%. The compressed gas to the shift reactor is mixed with the shift reaction steam to give a 1.0 ratio of steam to the dry gas and preheated by heat exchange with the hot effluent gases from the shift reactor prior to its introduction to the reactor which contains a sulfur resistant catalyst. From the interchanger, the shift effluent is cooled in a series of waste heat recovery exchangers.

Acid Gas Removal. The Rectisol process has been selected to remove the acid gases as it has the advantage of using the plant product as the scrubbing medium. This process was developed by Linde and Lurgi in Germany and plants presently in operation have been designed by both companies.

The Rectisol process absorbs  $\text{CO}_2$  and  $\text{H}_2\text{S}$  from the shifted synthesis gas stream using methanol.  $\text{CO}_2$  is rejected to the atmosphere and a hydrogen sulfide rich gas is available for the sulfur recovery in a Claus Plant. These streams are obtained by selectively regenerating the methanol from the absorber in a two stage regenerator system. Low pressure nitrogen from the air separation section is used to strip carbon dioxide from the rich methanol solution in the first stage regenerator. Stripped solution from the first stage regenerator is stripped of its hydrogen sulfide by a steam heated reboiler in the second stage regenerator. The  $\text{H}_2\text{S}$  stream flows to the Claus unit for sulfur recovery and the purified synthesis gas is now ready for methanol synthesis. The carbon dioxide content of the synthesis gas is controlled by mixing a desulfurized side stream of high carbon dioxide content from the absorber with the absorber overhead.

**Methanol Synthesis and Purification.** The unique feature of this process is the synthesis step, utilizing a copper based catalyst specially developed by ICI, which gives good yields of methanol at low temperatures (410°F to 520°F). The high activity of the catalyst at low temperature permits the reaction to be carried out at pressures as low as 750 psig and is the key to the economy of the process. By-product formation is minimized as a result of the low operating temperature, thus leading to high process material efficiencies.

Final traces of sulfur are removed from the synthesis gas by a bed of zinc oxide after preheating to desulfurization temperature. A bed of chloride catch is also provided to prevent chloride poisoning of the synthesis catalyst. After cooling, the make-up synthesis gas enters the synthesis loop at the inlet of the circulator compressor. The mixture of unconverted gas and fresh make-up gas is preheated to reaction temperature in the converter interchanger by the hot gases leaving the converter. The methanol synthesis converter is a pressure vessel containing a single bed of catalyst. Temperature control is effected by injecting cold gas at appropriate levels into the catalyst bed.

The feed gas and the cold shot gas combine and react to form methanol as they pass downwards over the catalyst. The converter exit gas is first cooled in the converter interchanger and subsequently in the crude methanol condenser where the crude methanol product is condensed. Crude product is separated in the high pressure separator.

The non-reactive components of the make-up gas, methane and nitrogen are purged from the synthesis loop between the separator and the point of make-up gas addition and are subsequently used as boiler fuel.

The crude methanol collected in the separator is let down in a single stage to the let down vessel and the resultant product passes to the distillation plant. To provide some independent operation of the synthesis and distillation units, the crude can alternatively be pumped to crude methanol storage.

Flash gas from the letdown vessel (mostly dissolved gases) is mixed with the synthesis loop purge stream and used as fuel.

The crude methanol is processed in a single column system to fuel grade methanol, and the overall efficiency of the distillation system is expected to be 99% at the end of life conditions.

The upper section of the column removes the light ends, principally dimethyl ether, methyl formate, aldehydes, ketones, and lower paraffin hydrocarbons while the sections of the column below the feed tray is designed to remove water. Fusel oil (predominately alcohols such as isobutanol) is purged from a tray near the base of the column. In order to reduce organic and thermal losses in the effluent water stream, the fusel oil is subsequently blended back into the product fuel grade methanol. The fusel oil may be utilized as fuel if chemical grade methanol is the desired product.

Methanol product is removed from the top section of the column, cooled, and pumped to storage.

Sulfur Recovery. Sulfur recovery will be accomplished in a standard Claus unit equipped with a Wellman-Lord  $\text{SO}_2$  recovery unit. The W-L system has been included so that the tail gas leaving the plant will be in compliance with environmental regulations.

Oxygen Production. The methanol plant of 5000 TPD capacity from coal requires large tonnage oxygen for coal gasification and, hence, it is economical to include an oxygen production unit in the facilities. Standard cryogenic air separation units producing 1600 TPD of oxygen can be used. The by-product nitrogen from this unit will be utilized in the plant for purging, char conveying and rectisol unit methanol stripping.

Offsites. The offsites section consists of those facilities to provide all services to the other sections. The major systems are water treatment, cooling tower, boiler feedwater, and the coal fired high pressure boilers. The coal fired boiler is a conventional pulverized coal boiler, but will also burn the purge gases from the methanol loop and all of the dry char from the Winkler gasifiers. The boiler package includes an electrostatic precipitator for particulate removal, but does not include facilities to remove sulfur dioxide from the boiler flue gases.

#### Design Basis

##### Product Specifications

###### Fuel Grade Methanol

###### Analysis

Methanol plus organics, Wt. %	99.5 minimum
Water, Wt. %	0.5 minimum
Higher heating value, BTU/lb.	9,725

###### Conditions

State	Liquid
Pressure, psig	100
Temperature, °F	110
Delivered to Battery Limits for storage	

##### Raw Material Specifications

###### Coal

## Ultimate analysis, Wt. %

Water	16.50
Ash	18.87
Carbon	48.36
Hydrogen	3.61
Nitrogen	0.86
Sulfur	0.57
Oxygen	<u>11.23</u>
Total	100.00

Higher heating value, Dry, BTU/lb.	10,345
Ash softening point, °F	2,282
Ash melting point, °F	2,597
Ash flow point, °F	2,723

Raw Material, Utility and Product Rates

When producing the design 5,000 TPD of methanol (99.5% purity), the battery limit flows for the unit herein described are as follows:

Imports

## Coal

Process Coal	M lb/hr	896
Boiler Coal	M lb/hr	<u>129</u>
	Total	1,025
Raw Water	M lb/hr	3,047
Electric Power	MKWH	57
NaOH	lb/hr	45

Exports

Methanol	ton/hr	208
Sulfur	ton/hr	2
Wet char	M lb/hr	27
Boiler Ash	M lb/hr	52
Water Effluent	M lb/hr	1,300

DAILY OPERATING COST DATA

Methanol Production, 5 Tons/Day 5,000

	<u>Unit Cost</u>	<u>\$/Day</u>
Raw Water	1.5¢/1,000 lb	1,097
Electric Power	1¢/KWH	13,637
Caustic Soda	6¢/lb	65
Total, \$/Day		14,799
Coal	\$3/S Ton	36,913
	\$6/S Ton	73,825
	\$9/S Ton	110,738
	\$12/S Ton	147,650
	\$15/S Ton	184,563

No credit has been taken for sulfur production.

#### Economics

Using the design just described, a capital estimate of the facility was made based on January 1975 prices and the following overall economics were calculated:

Capital Cost, MM\$	270	
No. Gasifier Trains	8	
No. Operators/Day (4 shifts)	80	
		<u>\$/Day</u>
Maintenance Materials & Labor @ 4%		32,700
Taxes & Insurance @ 2.5%		20,500
Administrative Overhead @ 2.5%		20,500
Oper. Labor & \$10,000/Manyear		2,400
Capitalization		
Utility		
- 65% Debt @ 10%	62,500	
- 35% Equity @ 12% DCF	62,400	
- Incl. 20 yr Straight Line Dept.		
Total Fixed Charges, \$/Day		201,000
Total Fixed Charges, \$/ST MeOH		40.20
Total Direct Charges, \$/ST MeOH		2.96
Coal	\$3/ST, \$/ST MeOH	7.38
	\$6/ST, \$/ST MeOH	14.77
	\$9/ST, \$/ST MeOH	22.15
	\$12/ST, \$/ST MeOH	29.53
	\$15/ST, \$/ST MeOH	36.91

### PRESSURIZATION OF THE WINKLER PROCESS

The Winkler process for the gasification of coal has provided gas for fuel or power, for synthesis of methanol and ammonia, for Bergius-Hydrogenation, and for the production of hydrogen in Europe and Asia when coal was the only raw material available. All of those commercial installations were designed and operated so that, after cooling and particulate removal, the product gas would be delivered at nominally atmospheric pressure. The reason for the low pressure operation was that, at that time, German regulations favored atmospheric pressure plants, i.e., operating pressures above 1.5 ata (7 psig) created problems in the areas of materials and government supervision.

In mid 1972, Davy Powergas undertook a study to determine whether or not a Winkler gasifier operating under these proven conditions would be competitive with other available technology. Within the accuracy of the estimates made, none of the commercial processes appeared decisively better than any of the others and hence, review was begun of the Winkler process to determine what constituted its limitations for current U.S. conditions. It became rather obvious that the biggest deficit was the low pressure of operation since the detrimental effects of low pressure operation are threefold:

1. Large size equipment to handle the large volume of gas.
2. High capital cost attributable to the product compression station.
3. High daily operating cost of power for product compression

Process economic studies were conducted to find the optimum pressure ranges and to assess the magnitude of savings attributable to pressure operations. Three separate studies were completed as defined below:

- Case I - Effect of gasification pressure on the production cost of low/medium BTU fuel gas using air/oxygen as the gasifying medium. The final fuel gas is delivered at 210 psig, with a maximum of 100 ppm of sulfur.
- Case II - Effect of gasification pressure on the production cost of 5,000 STPD of methanol.
- Case III - Effect of gasification pressure on the production cost of 1200 STPD of ammonia.

The general conclusions were encouraging as outlined below.

1. General Conclusions

It may be concluded that an increase in gasifier operating pressure results in a decrease in the overall cost of product. It becomes apparent that the greatest savings may be attributed to a decrease in the number of gasification trains required for a given output. Although there are significant savings in the compression requirements for the higher pressure case, the number of gasifiers remains constant. Despite the fact that the size of each gasifier has decreased, the increase in cost of the higher pressure design outweighs the net savings in compression, resulting in a slight increase in overall production cost.

The greatest decrease in the number of gasifiers occurs between 20 psia and 43 psia, for example, the plant for the production of 5,000 ST/D of methanol would require 12 gasifiers at 20 psia as compared with 8 at 43 psia.

From examination of graphs, it was deduced that the optimum operating pressure for the gasifier, depending upon the final product, lies between 120 and 180 psia. The one case which does not necessarily follow these conclusions is the integration of a Winkler system with a combined cycle power plant. In this case the air compression is provided by the gas turbine--compressor system, and as a result, the optimum gasification pressure is the highest permitted by the air delivery pressure.

## 2. Methanol Systems

Figures 6 and 7 present the cost of producing 5,000 ST/D of methanol from western sub-bituminous coal. Since methanol may be considered either a bulk chemical, or a liquid chemical fuel, the production costs reflect both typical utility financing (Figure 6) and typical industrial financing (Figure 7). Although there are but small savings in production cost over the range of gasifier pressures of 43-213 psia, the minimum production cost occurs at approximately 150 psia generator pressure.

The improvement in capital and operating costs resulting from pressurization is shown clearly in Figure 8 which also compares alternative processes such as steam reforming and partial oxidation for the production of synthesis gas.

## ACKNOWLEDGEMENTS

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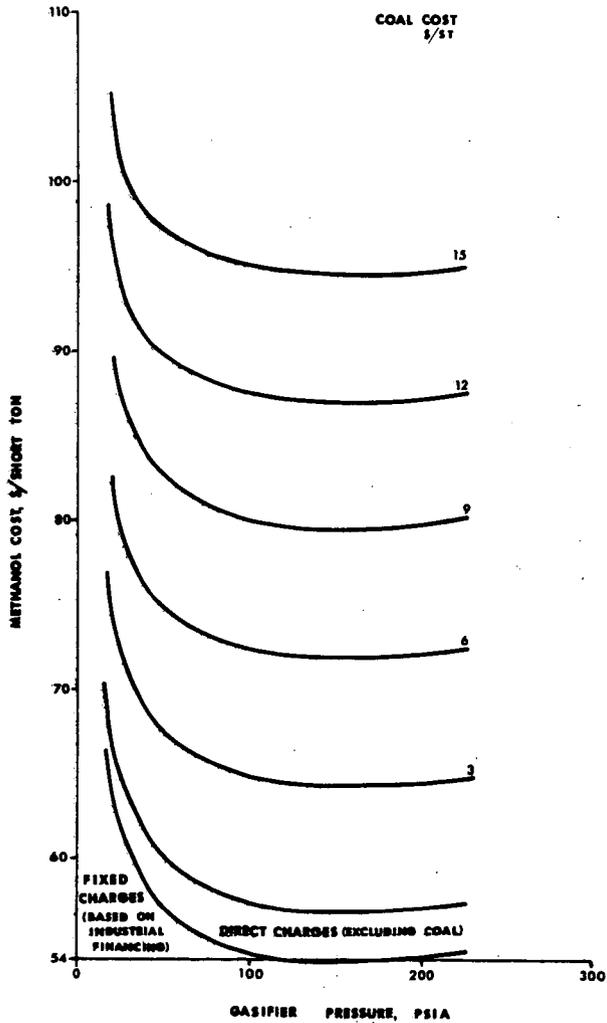
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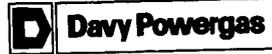
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GRAPH 4

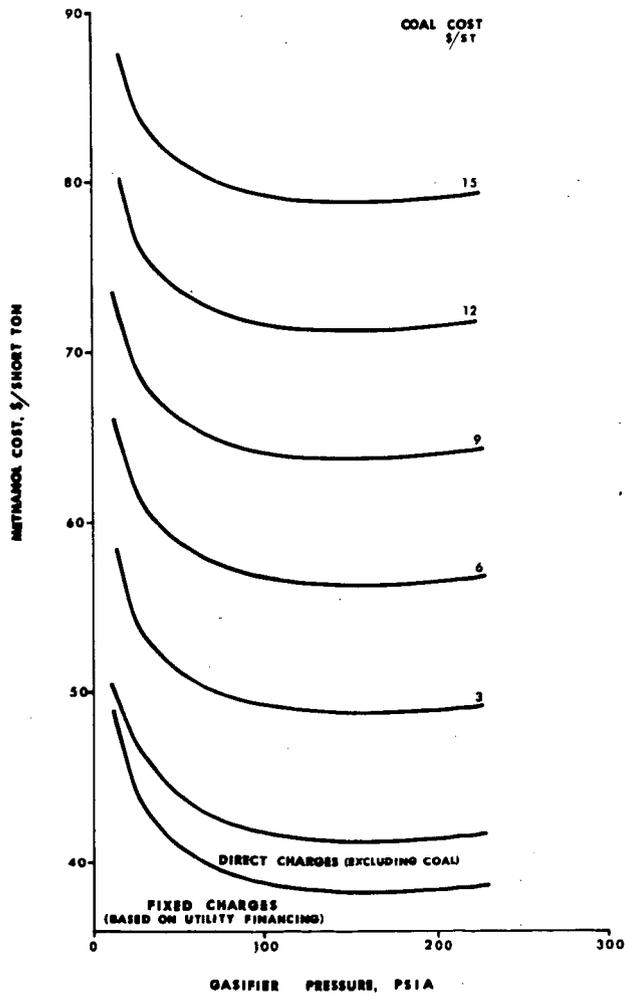
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BY THE WINKLER PROCESS

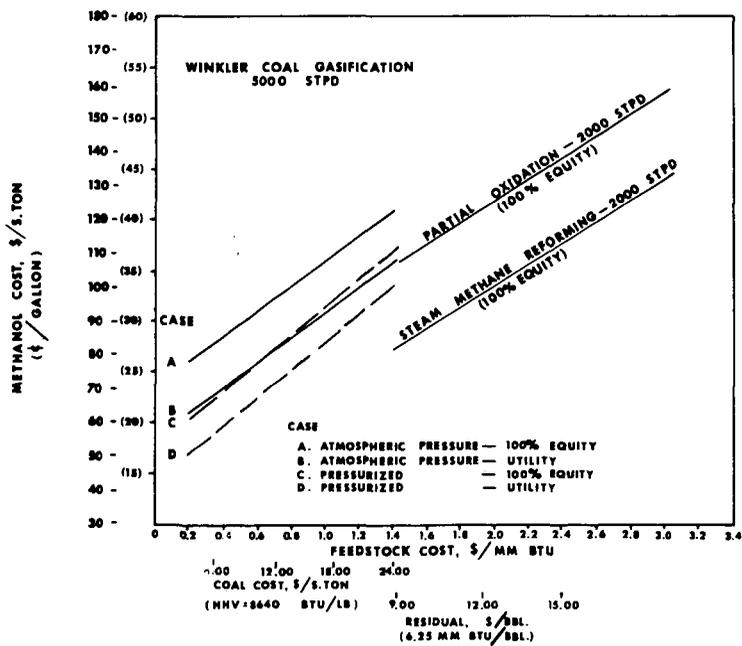




GRAPH 3

METHANOL FROM COAL  
BY THE WINKLER PROCESS




**Davy Powergas**
**COST TO PRODUCE METHANOL  
BY ALTERNATIVE PROCESSES**


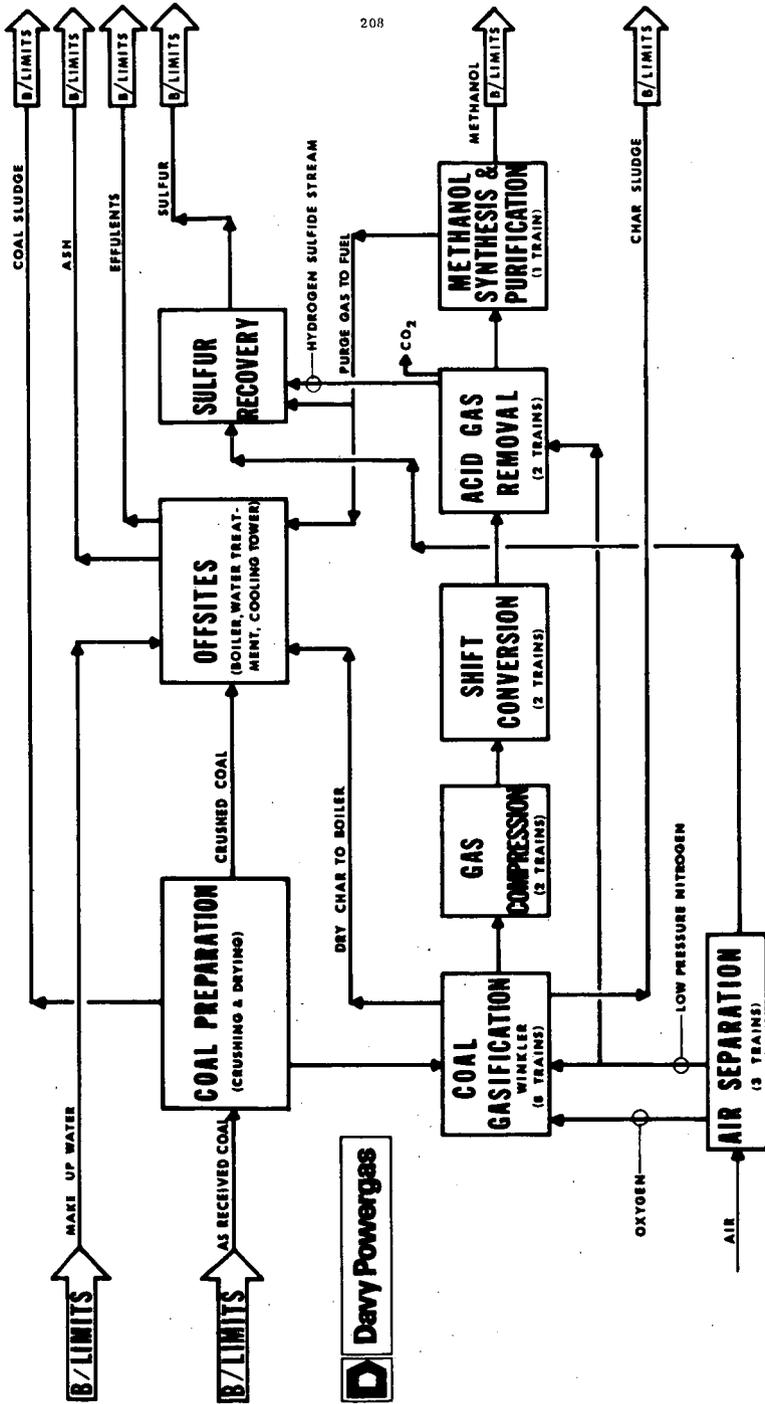


Fig. 5 Typical Methanol From Coal Facility Based On Winkler Gasification.

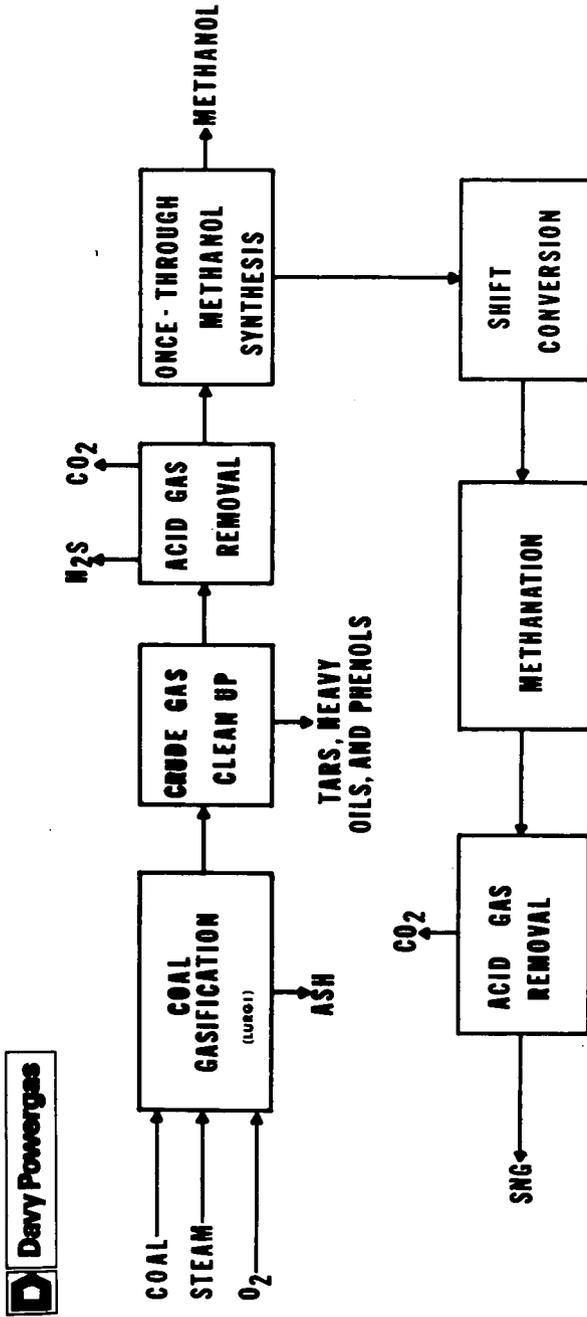


FIGURE 4



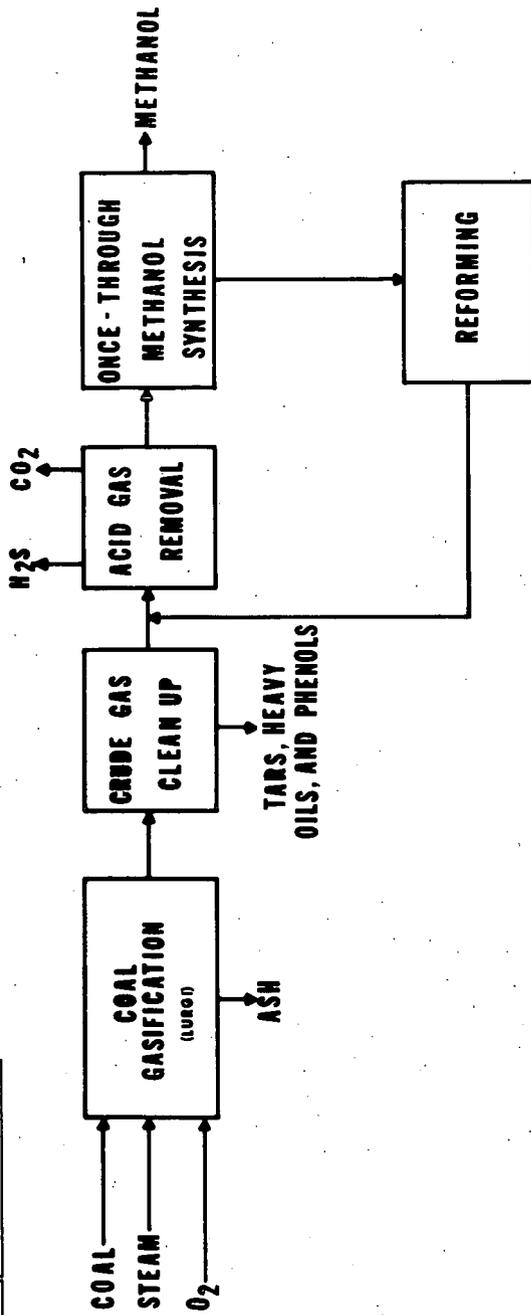


FIGURE 3

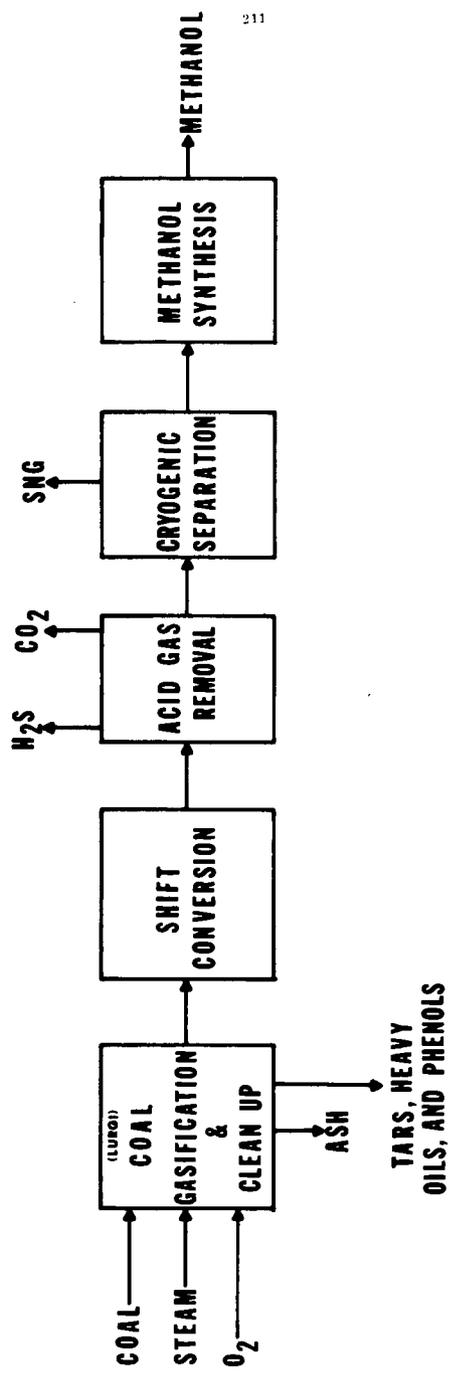


FIGURE 2

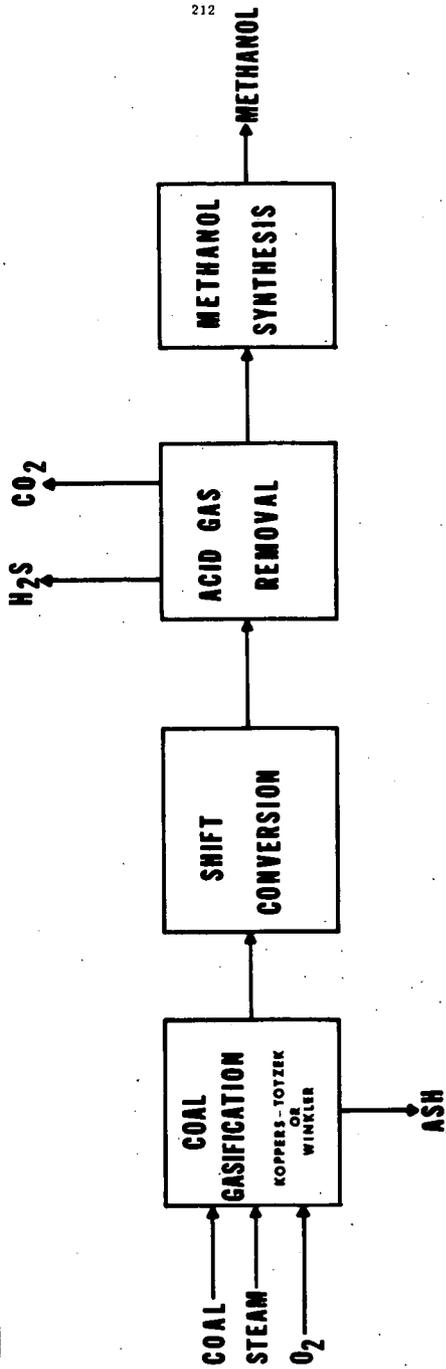


FIGURE 1