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## COAL CONVERSION TECHNOLOGY

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### Abstract

Energy consumption is rapidly increasing throughout the world and the United States is no exception. Efforts to reduce the dependence on oil imports have focused on utilization of our coal resources. This paper examines various coal conversion processes and presents a method for evaluating their contribution to energy production.

### 1. INTRODUCTION

Demand for energy in all forms is rapidly increasing. The United States gas and oil consumption over the past 20 years is representative of this increase and is shown in Figure 1.

This increased demand in the face of a limited supply has been one of the factors driving up the cost of both gas and oil. Despite the great increase in both the amount of gas and oil used and its high cost, there are large reserves of coal in this country that remain relatively untapped as energy sources.

One of the major reasons for this lack of interest in coal is the Clean Air Amendment Act of 1970. The Environmental Protection Agency suggested ambient standards for sulfuroxide content in the air, and these were incorporated into the Clean Air Act in 1970. The primary standards (i.e. health related standards) set by the 1970

Amendment specified an annual mean output of 80 milligrams of  $\text{SO}_x$  per cubic meter and a maximum output of 365 milligrams per cubic meter for twenty-four hours. These standards have often posed problems for facilities with conventional coal-fired boilers.

One way of meeting these standards is by burning low sulfur coal. However, domestic reserves of low sulfur coal are extremely limited. Another alternative is stack scrubbing. Much advancement has been made in this technology, to the point where the Environmental Protection Agency is recommending the use of scrubbers as a solution to the pollution problems of utility companies. However, in the opinion of Donald Cook, Chairman of American Electric Power Company, the work has not yet reached optimum costs, reliability, and feasibility.

A third alternate is to convert existing high sulfur coal into a Synthetic Natural Gas. This alternate provides clean fuel which is acceptable within air quality regulations and standards. There is an additional benefit from such a conversion in that it may help close the gap in natural gas supply and demand. Figure 2 represents the sources of U.S. natural gas supply and illustrates the need for a fuel source to replace shrinking domestic production.

Coal gasification is being studied worldwide and the importance and urgency of the research is becoming increasingly evident. As stated by Dr. Abbas Fallah (Iran), Hormoz Petroleum Co.:

"...petroleum is a raw material too valuable to be burnt for its destined use as feedstock for chemical and petrochemical industries... We should immediately devote full attention to development of new technologies for coal gasification/liquifaction.."

From the study of the gasification and desulfurization of coal, many different processes have emerged. Each of these processes has singular characteristics, advantages, and disadvantages. There are four classifications of coal conversion processes: pyrolysis, solvation, hydrogenation, and production of synthesis gas. Nearly thirty processes have been developed within these categories.

The purpose of this paper is to present a structured comparison of coal conversion processes. This comparison will take into account not only the quantitative characteristics of the process, but also the qualitative factors that could affect the success of the conversion of coal to a clean, convenient fuel or to a synthetic feedstock.

There are three distinct types of process characteristics which are involved in the

comparison. These are as follows:

- (1) Operating costs and revenues
- (2) Process efficiencies
- (3) Qualitative desirability factors

The first two of the above items are self-explanatory. The third item consists of investigating and comparing such things as:

- (1) Sensitivity to product prices
- (2) Public acceptance
- (3) Labor requirements
- (4) Etc.

This paper uses a structured evaluation model to compare existing coal conversion processes and illustrates how a more in-depth process comparison can be made.

## 2. DISCUSSION

### 2.1 PREVIOUS DEVELOPMENTS IN COAL PROCESSING

The first gas was made commercially from coal in the nineteenth century. This gas was produced by heating coal in the absence of air. Before being replaced by electricity late in the century, this gas was used for lighting cities, homes, and buildings. Afterwards, it was primarily used for cooking.(1) One by one, markets for both coal and gas made from coal disappeared as natural gas became more available.

Many companies worked on coal gasification during the mid-twentieth century, but most of these companies ran into problems which proved either insurmountable or uneconomical to solve. As natural gas shortages evolved, interest in coal conversion has awakened with renewed vigor as scientists strive to discover relief from the energy shortage.

## 2.2 DESCRIPTION OF COAL CONVERSION PROCESSES

Many types of coal conversion processes have been studied. Some of the processes have many variations, such as the number of stages, the temperatures, and the pressures of operation.

Scientists have succeeded in producing a synthetic natural gas from coal and have developed methods of refining the gases produced in certain of the conversion processes into methane. Figure 3 represents steps involved in gasification. Other conversion processes using coal as a raw material, result in the formation of synthetic crude oils which are suitable refinery feedstocks.

### 2.2.1 Processes Used in the Production of Synthetic Natural Gas

The following processes include all of the basic technology; however, some variations which have been made on certain processes were eliminated to prevent repetition.

Pyrolysis reactor of Garrett Research and Development Co. This experimental system included a one-inch diameter by eleven-foot reactor, coal feeder, product collection equipment, and gas sampling apparatus. The reaction temperature ranged upward from 1,500°F (below the ash-softening temperature of the char), and heat was supplied by electricity. Sub-bituminous coal (<200 mesh) was fed horizontally to the reactor then transported upward in dilute phase with nitrogen. Each run took about four hours. A filter bag and water cooled condensers were used to remove the tar. Product char was removed by cyclones.(2)

At 1,700°F, and after recycling the tar to the reactor for extensive cracking, the total equivalent yield was approximately

8,500 standard cubic feet of pipeline gas per ton of coal.

"From a commercial standpoint, sulfur reduction is most meaningfully expressed on an equal BTU basis, defined as: Sulfur reduction (equal BTU basis) =  $100 \times (\text{lb. S/BTU of coal} - \text{lb. S/BTU of char}) / (\text{lb. S/BTU of coal})$ . On this basis, sulfur reduction of 30% to 45% were obtained."(3)

Both the pipeline gas and the char have high heating values compared to those of other processes.

Clean Coke process-carbonizing and hydrogenating of U.S. Steel Corp. This process may be used to produce clean coke, low-sulfur liquids, and gaseous fuel byproducts from high-sulfur, high-ash coals.

This process combines carbonization and hydrogenation of coal. After sizing in a coal-preparation plant, part of the coal is processed through a carbonization unit. Here, the coal is devolatilized and partially desulfurized. The product is used to provide the base material for coke production. The rest of the coal is slurried with a carrier oil and hydrogenated to convert most of the coal to liquids. These liquids are processed into low-sulfur liquid fuels, chemical feedstocks, and three oil fractions that are recycled to other process areas. The char and pitch coke is slurried with one of these oils, formed into pellets and baked to produce metallurgical coke with a low-sulfur content. The vapors of the coke-preparation are collected and returned to the process.

Preliminary evaluations show that a plant constructed to process 6.5 million tons/yr of as-mined coal would produce 2.2 million tons/yr of coke pellets, 2.3 billion lb/yr of chemicals, 8 million gallons/yr of liquid fuels and approximately 6 trillion BTU/yr of fuel gas.

Lurgi process. Among the few processes currently in commercial operation is the Lurgi process. This is a fixed-bed process in which a sized, non-coking coal is fed into a pressure gasifier of up to twelve feet in diameter. The gasifier uses a rotating grate underneath the coal bed for feeding steam and oxygen which cool the grate and prevent clinkering of the ash. Coal is spread evenly over the bed by a distributor at the top of the gasifier where temperatures range from 500°F to 800°F. The rotating grate at the bottom allows ash to be collected in a hopper. The temperatures at the bottom of the gasifier are less than 2000°F. Raw gases leave at the top at 850°F and are scrubbed and cooled.(1)

The counter-current flow of reactants in the fixed-bed reactor allows the efficient use of the heat that is released during the oxidation of the coal near the base of the gasifier. Since this method also operates under pressure, the reported thermal efficiencies are on the order of about 70%.

Koppers-Totzek Process. This process contains an entrained bed of reactants: coal, steam, and oxygen. Two or four opposing burners may be used for commercial gasifiers. Four burners can handle up to 850 tons/day of coal. The raw gas leaves the gasifier at temperatures up to 3,300°F. Therefore, the consumption of oxygen per unit of gas is significantly higher than for fixed-bed reactors. There is a slag collected at the bottom of the gasifier.

Any rank or type of coal may be gasified by the Koppers-Totzek process. All of the coal, even the fines, may be used. Since there are no phenols, tars, or light oils produced during the operation, there are fewer environmental problems than with other processes. The thermal efficiencies

of the process are reported to be about 77%.

Winkler process. This atmospheric, fluid-bed gasifier uses oxygen and steam as media. Temperatures for the operation range from 1,500°F to 1,850°F. Unreacted carbon, ash, and product gas are carried out of the bed. The unreacted carbon is reacted with more steam and oxygen in the disengaging space above the fluid bed. The gases are cooled by a radiant boiler in the upper portion of the gasifier.

Sixteen plants, in a number of countries, use this process. The largest plant has a capacity of 1.1 million standard cubic feet per hour.

This process can handle all sizes of coal, but it cannot handle a strongly coking coal which is not pre-treated. The process boasts very few environmental problems and has an average oxygen consumption compared to other processes. The overall thermal efficiencies are reported to be about 75%.(1)

Hygas process. The Office of Coal Research has sponsored the development of the Hygas process by the Institute of Gas Technology. At the present time, a large pilot plant is being tested.(4) A high-BTU gas is produced by the process by reacting hydrogen (supplied by steam-carbon and water-gas shift reactions and by reacting steam with char at 1,900°F) with coal at 1,000 to 1,500 pounds per square inch. Coal is fed into the hydrogasifier at the top, and hydrogen is fed in at the top, and steam is fed in at the bottom. The hydrogasifier is made up of two fluid beds. The upper bed operates at 1,200°F and the lower bed at 1,700°F. The reaction rate of the process and the amount of methane at equilibrium in the product gas is optimized by this method.(1)

There are three methods which may be used to produce the hydrogen needed for the Hygas process. These are electrothermal, steam-oxygen, and steam-iron. The three methods were proposed by the Institute of Gas Technology.

CO<sub>2</sub>-Acceptor process. The CO<sub>2</sub>-Acceptor process was also introduced by the Office of Coal Research and is being tested in a pilot plant. In this process, coal is fed into the gasifier and, after being devolatilized, is reacted with steam in a fluid-bed gasifier. Operating pressure ranges from 150-300 pounds per square inch. Hot dolomite is introduced into this reaction. The dolomite provides heat to the steam-carbon reaction by absorbing carbon dioxide formed by the decarboxylation of the lignite feed.(4) The product gas leaves at the top of the gasifier. The spent dolomite and unreacted char are removed at the bottom. Then the unreacted carbon is burned with air and the heat produced carbonates, and the dolomite is regenerated.

Synthane process. This two-stage process has been tested only in small pilot plants; however, there is a 70 ton per day plant scheduled for completion by late 1974. This method used a pressurized gasifier developed by the Bureau of Mines, in which the coking properties of the coal are destroyed with oxygen and steam. This may be accomplished by either a free-fall stage or in a fluidized bed. The coal is carbonized and gasified with steam and oxygen in the lower section. Product gas leaves at the top, and char and ash are removed at the bottom. The operating temperature at the top of the bed is about 1,100°F and ranges from 1,750°F - 1,850°F at the bottom.

Bigas process. The Bigas process involves a super-pressure method developed by Bituminous Coal Research, Inc. The process uses a two-stage, entrained bed. In the upper section, coal is heated by hot gases produced in the lower section. The distillation gases from coal carbonization leave the gasifier along with the gases produced in the lower section of the vessel. The raw gas and part of the char is recycled to the lower section where it reacts in an entrained state with steam and oxygen to produce synthesis gas(1) The lower section operates at 2800°F and the upper vessel operates at temperatures ranging from 1,400°F to 1,700°F.

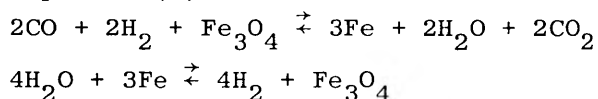
Union Carbide-Battelle process. This process is operated at 100 pounds per square inch pressure and requires no oxygen. In one stage (of the two-stage process), a fluid-bed combustor is used to burn part of the carbon with air to form coal ash agglomerates. A gasification reaction between steam and carbon takes place in a second vessel where the hot pellets are circulated to provide the heat for the reaction. Some of the pellets are recirculated and reheated in the first vessel.

Hydrane process. The U.S. Bureau of Mines developed this hydrogasification step, which is very similar to the Synthane process. A pressurized gasifier, using steam and oxygen as the reactants, is used as the second stage of a counter-current process to react devolatilized char from the first stage with hydrogen at 1,650°F.

Atgas process. This process was developed by the Applied Technology Corporation. It is a low-pressure process which uses a molten iron bath and limestone with either air or oxygen for the reaction. Using air, the process produces a sulfur-free, low-BTU gas. When oxygen is used in the reaction, a medium or high-BTU gas is produced.

Molten Salt process. This process, developed by the M. W. Kellogg Co., employs a molten-carbonate gasification system. Gasification is accomplished by steam-carbon and carbon-oxygen reactions, which may occur simultaneously or in a divided vessel.

Steam-Iron process. This process is a hydrogasification process which produces carbon oxides, hydrogen, and nitrogen by passing steam and air over a bed of hot carbon. When these gases come into contact with iron oxide, iron is formed. Then steam is passed over the iron, resulting in the production of hydrogen. This process lends itself to a continuous process and studies are underway in this area. The following equations represent the process(5):



Work on this process was sponsored by the Fuel Gas Associates, an organization that represents several energy companies.(4)

Bituminous Coal Research process. In this process, ground, dried coal is gasified at 1,100 pounds per square inch (gauge) with oxygen to produce methane. Heat from this stage is used to supply the steam carbon reaction heat in the gasifier. The process involves a shift converter, gas purification, and methanation. The process requires oxygen for the gasification and heat for the slag of coal ash at the bottom of the gasifier.(4)

Methanation process. After coal has been gasified, it is possible to produce a largely increased amount of methane from the products of this gasification. The process involves the production of methane from hydrogen and carbon dioxide by use of a catalyst. Figure 4 illustrates the methanation process.(4)

Toscoal process. This process is an adaptation of technology utilized in the production of oil from oil shale. Partially heated feed is heated to carbonization temperature by contact with heated ceramic balls. A trommel screen is used to separate the solid carbonization residue from the ceramic balls, which are recycled for reheating. The hot residue is cooled and this heat is used to produce steam needed to remove tar and water from gaseous components of product stream from pyrolysis drum.(6)

## 2.2.2 Processes Used in the Production of Synthetic Crude Oil

Much current research is concerned with coal liquefaction due to the need for substitute refinery feedstocks and clean fuel for direct use in boilers.(1) Some of the processes discussed are in operation; some are in development.

Bergius process. This is a German process which boasts 55% overall efficiency.(1) The hydrogasification technique reacts a mixture of finely ground coal and a hydrocarbon liquid with the hydrogen at 850°F and 10,000 pounds per square inch. The product is separated into light, middle, and bottom portions. The middle portion requires further refinement using a catalyst. The bottom portion is strained and the liquid being used in the feed mixture. This process is very expensive.

COED process. The Char Oil Energy Development Process(7) employs a multi-stage fluidized-bed pyrolysis to yield a synthetic crude oil, a char product and a gas stream which can be processed to produce hydrogen, fuel gas, and liquid hydrocarbons. The process uses various numbers of states depending upon the type of coals. The product oil is hydrotreated to produce a synthetic crude. This process will

easily lend itself to commercial use if the economics permit.

Fischer-Tropsch process. This process is a synthesis gas process in which coal is gasified completely to a product of a state which depends on the variable ratio of hydrogen to carbon monoxide. Different type products may be purified and passed over different catalysts in a temperature range of 570°F to 640°F at a pressure of about 450 pounds per square inch. The result is a mixture of paraffinic and olefinic products. The process has an overall conversion efficiency of about 38%. One existing process employs fixed-bed Lurgi gasifiers. "The major engineering problem is the removal of the large volumes of heat that are released when the gas is converted to a liquid by the catalyst."(1)

Project gasoline process. This process produces a refinery feedstock by first converting coal to liquid by hydrogen-transfer recycle solvent. Then a fluid-bed catalyst reactor is used to react the liquid product with hydrogen. The solvent is separated from this final product and recycled.

H-Coal process. This process is a Hydrocarbon Research, Inc. variation of the H-Oil process. A hydrogenator using an expensive cobalt molybdate catalyst produces a liquid-solid mixture and hydrogen. A flashdrum is used to treat the product. The ultimate product is a low-sulfur refinery feedstock.

Synthoil process. Like the H-Coal process, the Synthoil process is a low-ash, low-sulfur process. The hydrodesulfurization uses the turbulent flow of hydrogen (an excess amount of recycled hydrogen) to move a slurry of coal in a recycled portion of the product oil through a bed of cobalt-molybdate pellets at 2,000 to 4,000

pounds per square inch and 800°F. The sulfur is removed in the form of hydrogen sulfide which is converted to elemental sulfur for storage.

As the process may be operated at a much less extreme pressure than the Bergius process, the resultant produce should be much less costly.(1)

Pamco process. Another low-sulfur, low-ash fuel process, the Pamco process involves the hydrogenation of finely ground coal dissolved in a recycle stream at 1000 pounds per square inch. The product is a solid at room temperature and a liquid at 350°F; therefore, it may be burned as a boiler fuel if preheated. Two pilot plants which use this process are now being tested.

### 2.2.3 Overview of Process Capabilities

Even with this vast amount of research being carried on in the field of coal conversion, it is difficult to secure information on the processes, their efficiencies, and their economics. Many processes, such as Cogas, are in the development phase in which the security of proprietary information is of the utmost importance. For various types of coal, frequently reported efficiencies for coal gasification processes range from 56% to 77% thermal efficiency, with capital investment ranging from \$275 million to \$490 million in 1975 dollars. For coal liquifaction, reported efficiencies range from 60% to 75% and investment ranges from \$265 million to \$570 million. The processes for the production of methanol from coal have reported thermal efficiencies of 60% to 67% and capital investments of \$318 million to \$470 million.

### 2.3 DEVELOPMENT OF A SELECTION MODEL

Economic analysis is a wide-spread policy for decision-making. However, when making a management or engineering decision, one



should realize that, if there are many factors which are pertinent to the decision, it is frequently very difficult to quantify all of these factors in dollars or any other common denominator.

Energy independence is very important to this country's future. Coal conversion is an alternative route to this goal. However, when considering the increasing awareness of industries' social responsibilities, it becomes obvious that economics is only one basis for evaluating coal conversion alternatives. In addition to an economic analysis, one must study the efficiency of the processes and all intangible factors. Utility theory can be used to analyze these factors.

The first step in this analysis is a determination of the intangible factors which could possibly affect the decision-making situation. See example below.

TABLE I  
DESIRABILITY FACTORS

- (1) Reliability (maintainability)
- (2) Reserve Situation
- (3) How Well Process Meets Pollution Requirements (Environmental)
- (4) Do Products Meet Market Requirements
- (5) Are There Markets For By-Products
- (6) How Easily is Product Transported
- (7) Input Requirement
- (8) Adaptability
- (9) Health and Safety
- (10) Residue Disposal
- (11) Management
- (12) Public Acceptance
- (13) Capacity Expansion
- (14) Labor Requirement
- (15) Plant Siting
- (16) Conversion Technology
- (17) Ecological Efficiency
- (18) Back-up and Storage
- (19) Sensitivity to Product Prices

A set of criteria for each factor should be established. For example, operating costs should be minimized. The criteria should be established according to the situation specifications. These criteria will be used in evaluating each process with respect to the given factor.

In any given situation, some factors will be more important to the decision-maker than other factors. Since all factors are not of equal importance, it is necessary to assign importance ratings(8) which will be used to rank the factors according to their relative importance. A scale of 0 to 1 or 0 to 100 may be used.

These ratings may be adjusted in order to improve the consistency of the importance ratings. Since there are 19 factors to be considered in the utility function to be used in evaluating coal conversion process, the following adjustment procedure is recommended:(8)

- (1) List the factors in order of descending importance.
- (2) Select one factor at random and assign an importance rating of 100 to it.
- (3) Assign each remaining factor to one of several groups of about equal size. Add the selected (Step 2) objective to each group.
- (4) Assign importance ratings to each factor by groups, keeping the rating of the selected objective at 100, and arrange in descending order.
- (5) Compare the importance of the first factor in each group to the importance of all the rest together and follow the procedure outlined here. (Do not change the rating of 100 assigned to the selected factor.)

- (a) If the first factor is more important than all the rest of the factors together, adjust its rating so that it is greater than the sum of the ratings of all of the other factors.
- (b) If the first factor is of equal importance to all of the other factors put together, adjust its rating so that it is equal to the sum of the ratings of all of the other factors in the group.
- (c) If the first factor is of less importance than all of the other factors put together, adjust its rating so that it is less than the sum of the ratings of all the other factors in the group.
- (6) If 5a or 5b is the case, omit the factor in question and apply the fifth step to the next lower factor in the list.
- (7) If 5c is the case, compare the importance of the factor in question to the sum of the importance ratings of all but the lowest factor in the list and proceed as in the fifth step. If 5c is still applicable, compare the factor in question to take the sum of all but the lowest two ratings, and so on, until the factor in question is being compared to the sum of the two ratings closest to the sum of the factor in question. At this time, proceed to Step 6.
- (8) Continue the procedure until the rating of the third from the lowest factor has been compared with the sum of the two lowest ratings in the group.
- (9) Make a combined list of the factors in order of descending importance. Adjust any difference in ranking from the initial list if that list is thought to be correct.
- (10) Find the sum of all of the ratings and divide this into each rating and multiply by 100 in order to rank the importance of each factor on a scale of 0 to 100.
- After the desirability factors have been properly weighted according to their relative importance, a measure of the desirability of each process may be given for each factor. An overall weighted utility score may be calculated to indicate the desirability of each process using the formula:(9)
- $$U = \sum_{i=1}^N \left( W_i \sum_{j=1}^M D_{j,i} \right)$$
- where  $U$  = utility  
 $i = 1, 2, 3, \dots, N$  desirability factors  
 $W_i$  = the weighted importance rating for  $i^{\text{th}}$  factor  
 $j = 1, 2, 3, \dots, M$  alternative processes  
 $D_j$  = the desirability score for the  $j^{\text{th}}$  alternative process
- This model is a simple linear non-interacting mathematical model.
- The last step in the development of the utility function is to list the processes in descending order according to their overall utility scores.
- ### 3. CONCLUSION
- Coal conversion will play a major role in our future. This paper presents a thorough model for examining alternative routes to synthetic gas and oil from coal.

Efficient, practical coal conversion is one major step toward self-reliance.

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#### 5. BIOGRAPHIES

Karen A. Dollar attended the University of Missouri-Rolla where she received her Bachelor of Science Degree in Engineering Management in May 1974 and her Master of Science in Engineering Management in May 1975. She currently resides in Wilmington, Delaware where she is employed as a chemical engineer in the Field Engineering Section of the Engineering Service Department of the E. I. du Pont de Nemours & Co., Inc.

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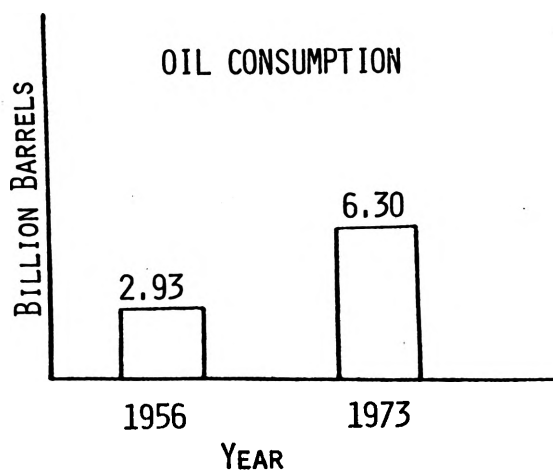
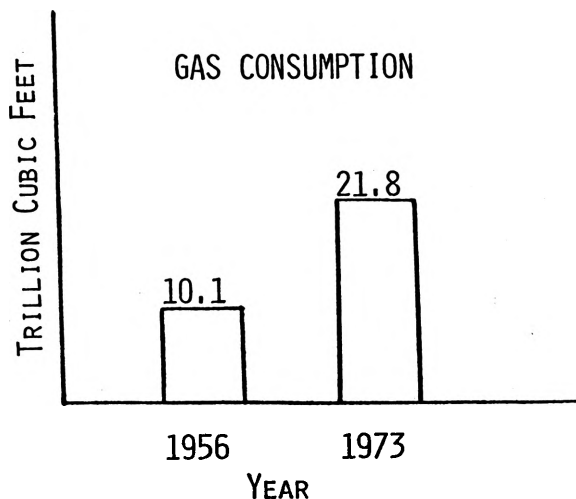


FIGURE 1.

## U.S. GAS SUPPLY

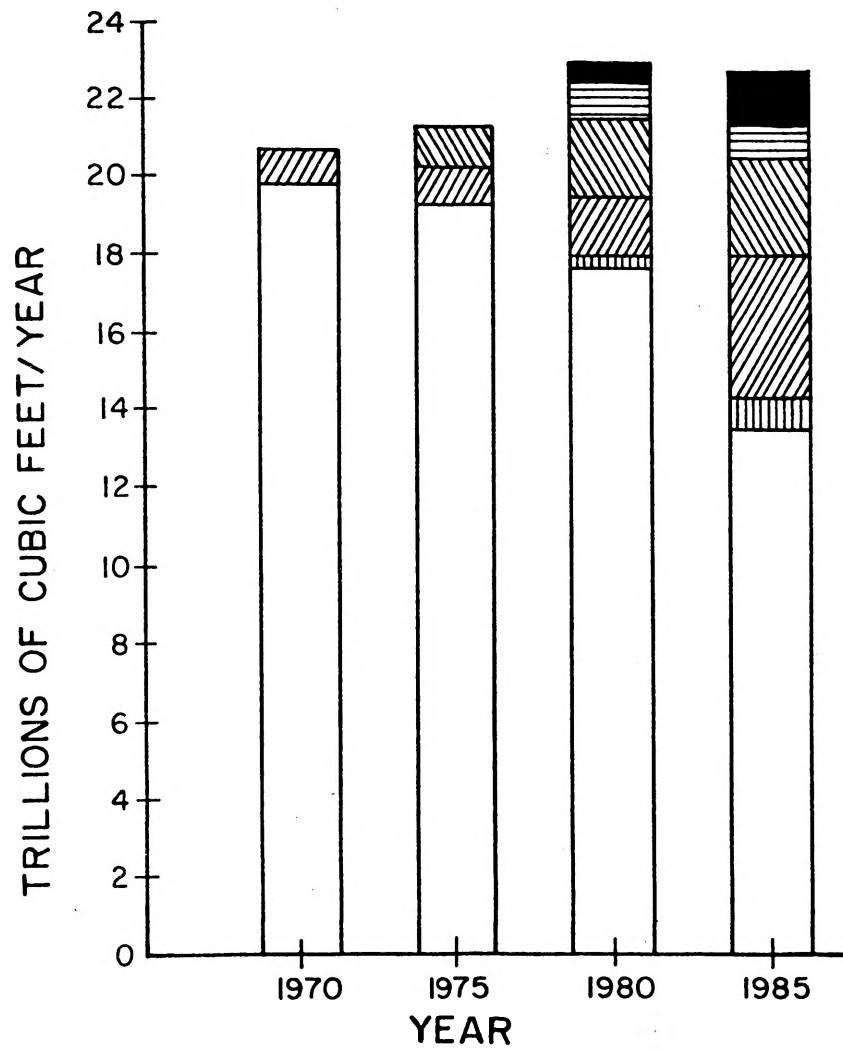
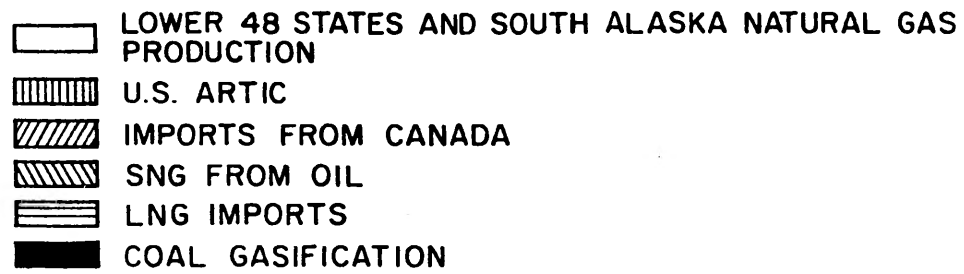


FIGURE 2.

# GENERAL GASIFICATION PROCESS STEPS

1.  $\text{COAL} \rightarrow \text{C} + \text{CH}_4 + \text{a mixture of liquids and gases}$
2.  $\text{C} + 2\text{H}_2 \rightleftharpoons \text{CH}_4$
3.  $\text{C} + 2\text{H}_2\text{O} \rightleftharpoons \text{CO} + \text{H}_2$
4.  $\text{C} + \text{O}_2 \rightleftharpoons \text{CO}_2$
5.  $\text{CO}_2 + \text{H}_2 \rightleftharpoons \text{CO} + \text{H}_2\text{O}$

Figure 3.

# GENERAL METHANATION PROCESS STEPS

1.  $\text{COAL} \xrightleftharpoons{1000-1500^\circ\text{F}} \text{CH}_4 + \text{C} + \Delta\text{H}$
2.  $\Delta\text{H} + \text{C} + \text{H}_2\text{O} \xrightarrow{1700^\circ\text{F}} \text{CO} + \text{H}_2$
3.  $\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{H}_2 + \text{CO}_2 + \Delta\text{H}$
4.  $\text{C} + 2\text{H}_2 \xrightleftharpoons{1700^\circ\text{F}} \text{CH}_4 + \Delta\text{H}$
5.  $3\text{H}_2 + \text{CO} \xrightleftharpoons{\text{Ni CAT. } 700^\circ\text{F}} \text{CH}_4 + \text{H}_2\text{O} + \Delta\text{H}$

Figure 4.