HIGH BTU GAS FROM COAL: STATUS AND PROSPECTS

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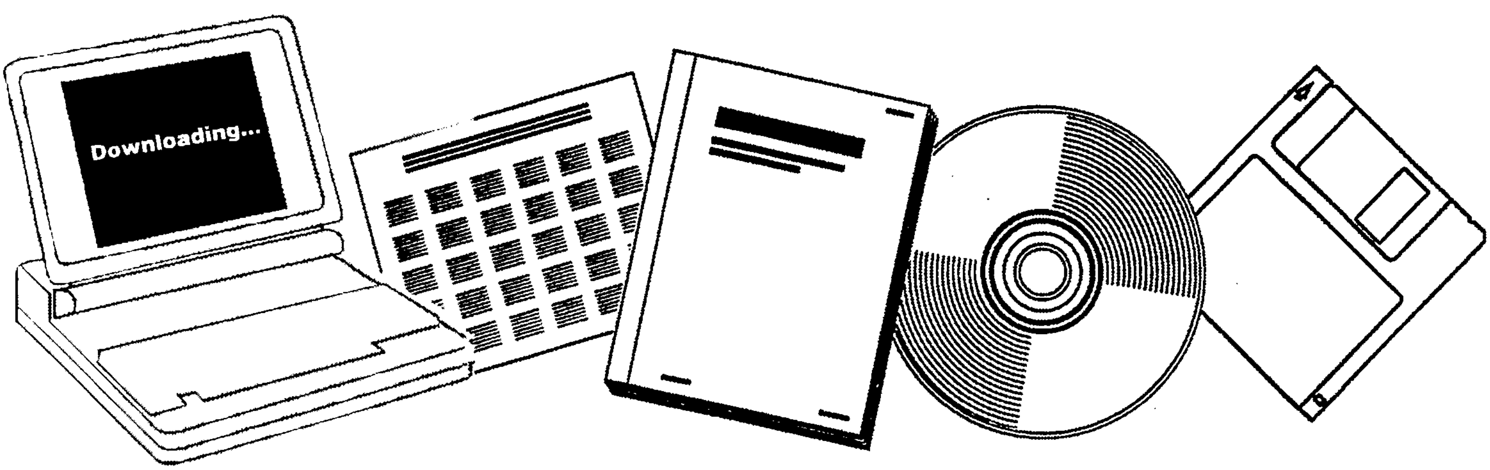

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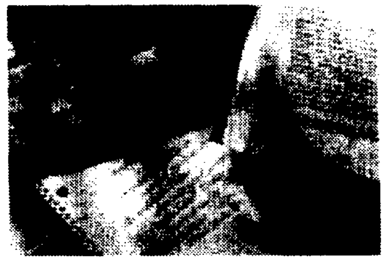

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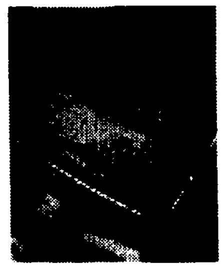

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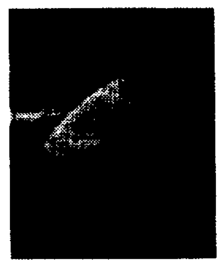

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By
W. P. Haynes
A. J. Forney

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Pittsburgh Energy Research Center
Energy Research and Development Administration
Pittsburgh, Pennsylvania

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HIGH BTU GAS FROM COAL: STATUS AND PROSPECTS

W. P. Haynes[1/] and A. J. Forney[2/]

The decreasing supply of natural gas in this country and the growing need for clean fuels has resulted in accelerating the development of numerous processes to make high-Btu gas from coal. Thus far, there are only two processes being offered commercially for making substitute natural gas (SNG) or high-Btu gas from coal--the Lurgi (1) (2)[3/] and the Koppers-Totzek processes (3) (4).

The Fossil Energy Division of the Energy Research and Development Administration (ERDA) is authorized to fund development of technology for converting coal to substitute natural gas (SNG) on a scale as large as demonstration plant size as well as pilot plant size. ERDA, in cooperation with the American Gas Association, is funding pilot plant development work on three processes for making pipeline gas from coal: the Hygas process, the CO_2-acceptor process, and the Bigas process. ERDA is also funding independently the development of the SYNTHANE process which was originated by the Bureau of Mines. The intent of the ERDA program is to support the development of a process to the point at which sufficient design and engineering data are available for industry to construct and operate a commercial sized facility with acceptable risk. The choice of such a process can be made only after data from competitive pilot plants are available.

The projected demonstration-sized plant will be versatile and is expected to incorporate the most promising parts of the various processes currently under study. If necessary, it will demonstrate more than one gasifier design. The high-Btu gasification program of ERDA includes support studies on individual sub systems. It includes studies on materials of construction, and it funds engineering evaluation studies. Total funding authorized for the program on development of high-Btu gas from coal processes is 59.8 million dollars for FY 1975, (5) and it is expected to increase significantly in FY 1976.

Private industry groups also have funded developmental work on gasifiers, methanation systems, and co-product processes that yield both liquid and high-Btu gases.

This report will attempt to give an overview of the status and prospects of the various endeavors to develop commercially viable processes for converting coal to high-Btu pipeline gas.

[1/] Supervisory chemical engineer
[2/] Research supervisor
[3/] Underlined numbers in parenthesis refer to items in the list of references at the end of this report.

General Process Scheme for Making SNG From Coal

Coal to SNG processes with variations follow a general process scheme such as that shown in figure 1. In the general scheme, after the coal is mined and stored at the plant site, there are nine basic process steps leading to the SNG product and seven auxilliary steps to furnish utilities and environmental protection.

In general, the major development effort is being expended upon advancing the coal pretreatment and gasification steps. Next in urgency is the development of a commercially viable catalytic methanation system. Considerable effort also is being expended on advancing those steps affording environmental protection. Although the remaining parts of the overall process are either commercially available or can be designed and constructed at an acceptable level of risk, efforts to improve these process steps are continuing.

Coal-To-SNG Processes, Their Status and Prospects

The processes considered to be the most advanced--i.e., they are either commercially available or they are at the pilot plant stage--are listed in figure 2 along the sponsoring firm. The status and prospects of these processes will be discussed. To provide a reference point, a description of the Lurgi process will be given first.

A. Lurgi Dry Ash Gasification Process

A simplified drawing of the Lurgi process is shown in figure 3.

Non-caking coal sized and dried is fed by means of lock hoppers to the gasifier where gasification occurs at 350 to 450 psi. Crude gas leaving the gasifier at temperatures between 700° F and 1100° F (depending on type of coal) contains tar, oil, naphtha, phenols, ammonia, plus coal and ash dust. Quenching with water removes tar and oil. Part of the gas passes through a shift converter. The catalyst used in the shift converter also promotes desulfurization of light oils in the gas. Gas coming out of the shift converter is washed for the removal of naptha and unsaturated hydrocarbons, then CO_2, H_2S, and COS are removed by absorption with cold methanol. Total sulfur content is reduced to less than 0.1 ppm. The gas is methanated in a series of two adiabatic reactors, and 970 Btu/scf pipeline gas is produced by final CO_2 removal and dehydration.

Condensate or gas liquor from the gasifier contains phenols, ammonia, fatty acids and other solubles. The Phenosolvan process step removes the phenols and ammonia, leaving less than 20 ppm phenols and less than 60 ppm ammonia. The water finally undergoes biological treatment to meet regulations for disposal of waste water. Regenerator off-gas from the methane scrubbing step is treated in a Stretford process step to recover free sulfur.

There are several fuel options for the high pressure steam generation step (not shown on the flowsheet)

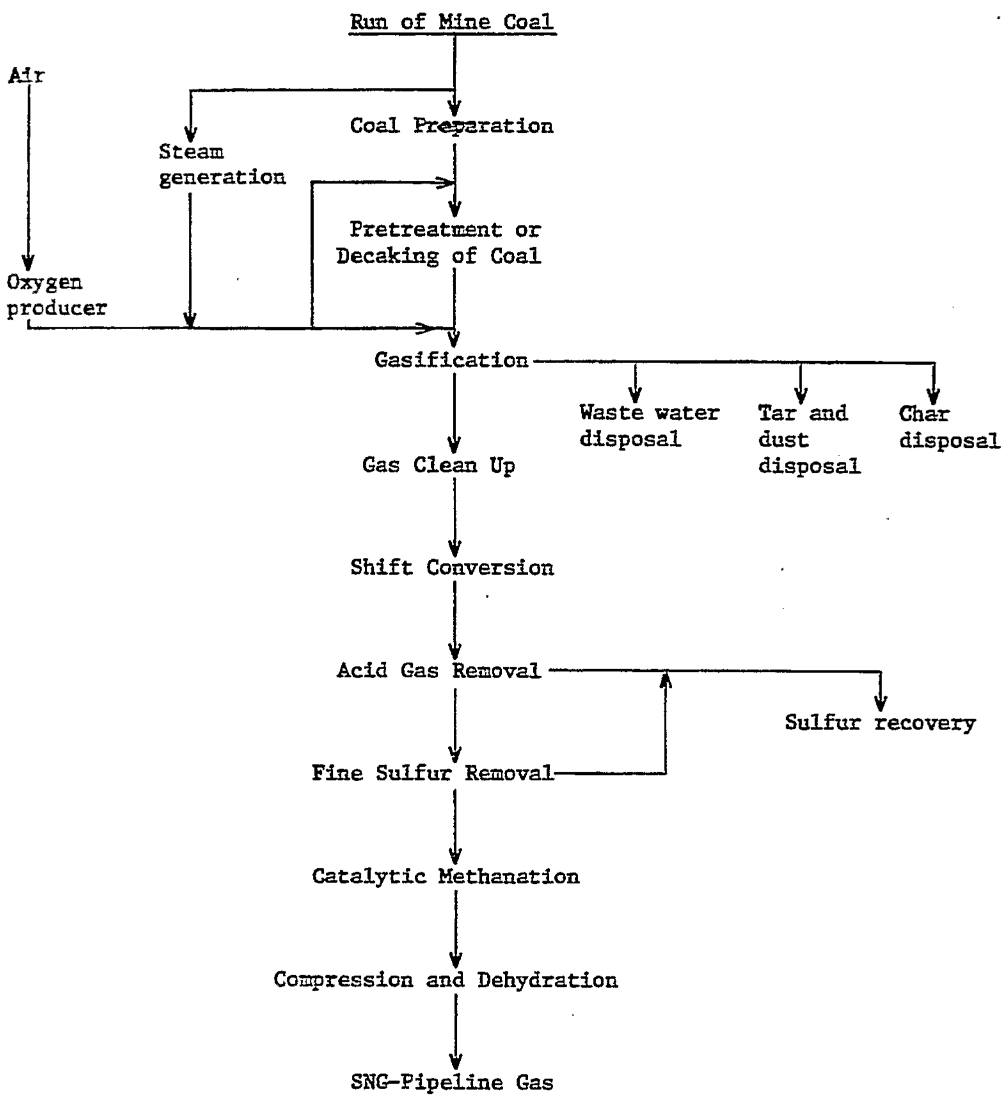

FIGURE 1. - A GENERALIZED PROCESS SCHEME FOR MAKING SNG FROM COAL.

FIGURE 2. - Gasifiers for Coal-To-SNG Processes

Commercial Scale

Process	Sponsoring Firm
Lurgi	American Lurgi Corporation
Koppers-Totzek	Koppers Company, Inc.
Winkler	Davy Power Gas

Pilot Plant Scale

Hygas	Institute of Gas Technology (IGT)
CO_2 Acceptor	Conoco Coal Development Company
SYNTHANE	Pittsburgh Energy Research Center
Bigas	Bituminous Coal Research, Inc. (BCR)
Agglomerated Ash	Union Carbide

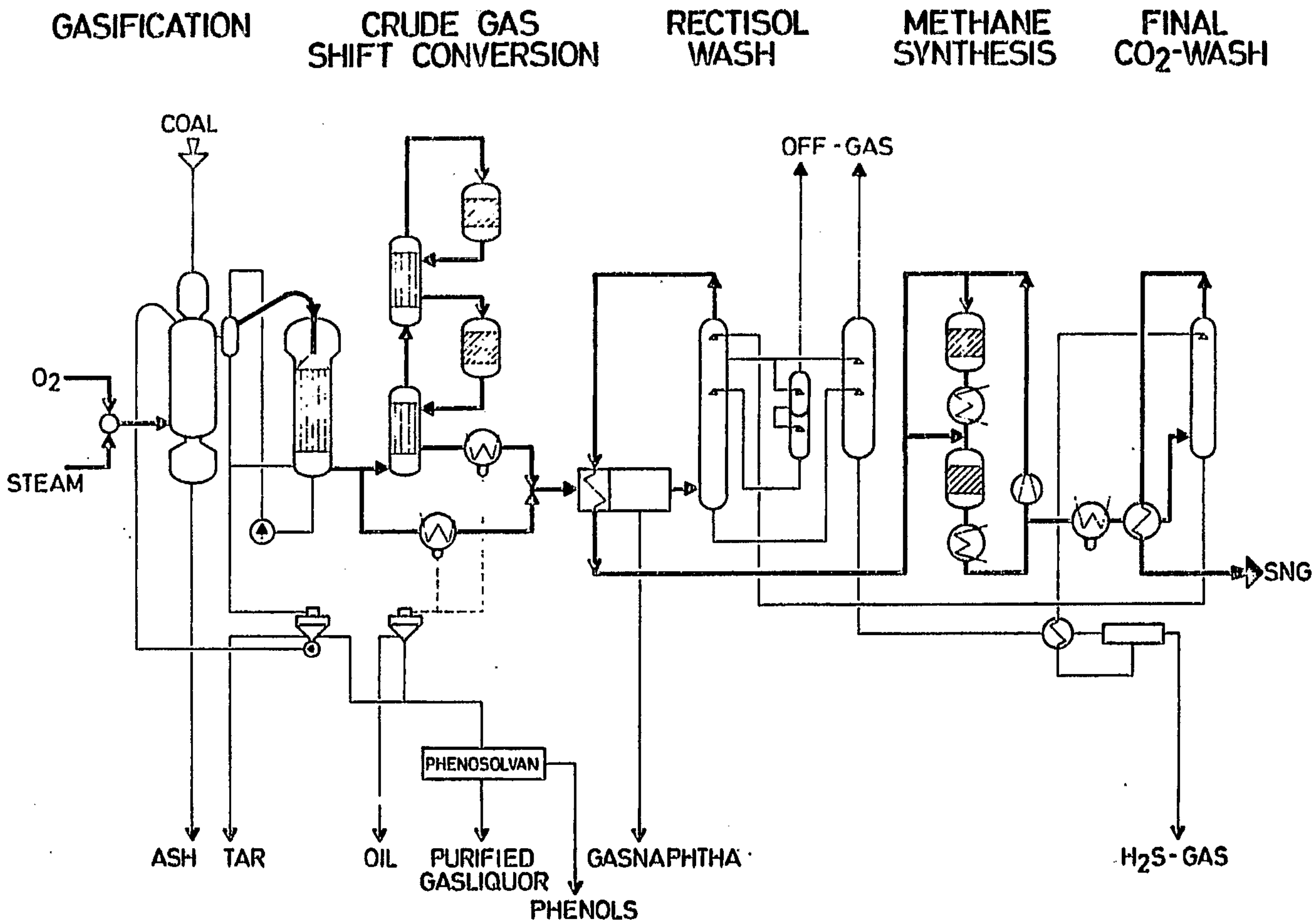

FIGURE 3. - LURGI PROCESS TO PRODUCE S.N.G. FROM COAL

(a) coal fines from coal preparation and tars from the tar separator;
(b) SNG product;
(c) partially purified synthesis gas; and
(d) air blown Lurgi.

Because of the inability of the Lurgi gasifier to operate on coal fines (<3/16 in.) the WESCO project (6) has selected option (a)-- the burning of coal fines along with the tars and removing the SO_2 from the stack gas. Agglomerating the fines to a suitable size briquette for a Lurgi feed is a promising concept but currently has not proved encouraging because of the characteristics of the coal and insufficient high quality binder from the tar fraction.

Major features of the Lurgi gasifier are shown in figure 4. A lock hopper is used to pressurize the coal fed to the gasifier bed. A water jacket surrounds the fuel bed and protects the outer pressure shell from overheating. Steam generated in the jacket is added to the gasification steam. A rotating distributor distributes the coal evenly across the bed and breaks up the coal as it carbonizes to prevent agglomeration. As the coal descends, devolatization occurs initially and is then accompanied by gasification in the temperature range of 1150° F to 1400° F. Residence time is about 1 hour. Steam is the source of hydrogen. About 84 percent of the coal is gasified; combustion of the rest of the char with oxygen at the bottom of the gasifier supplies the heat required. A revolving grate at the base of the reactor supports the fuel bed, removes the ash, and introduces the steam and oxygen mixture. Ash is depressurized and removed from the gasifier through the bottom lock hopper. The gas outlet has a mechanical scraper (not shown) to remove deposits which might accumulate and restrict the flow. The gas is quenched and scrubbed by a spray of recirculated gas liquor. Part of the gas liquor is sprayed over the top of the fuel bed to reduce dust carryover.

Overall thermal efficiency of the process going from coal to SNG is estimated to be 52.9 percent if gas generated from coal fines is used to raise steam and 55.1 percent if the fines are burned directly. These efficiencies would increase approximately 7 percent if the tar products, naptha, and phenols were used as fuel. For the WESCO project, 30 gasifiers (27 on-stream and 3 on stand-by) would be required to produce 250 million scf per day SNG from 25,100 tons per day of subbituminous 8,300 Btu/lb coal. Current maximum gasifier diameter is 12 ft (3.65 meters). To increase the unit capacity, designs are being completed for a new 16.4 ft (5 meter) diameter gasifier which is to be tested in Sasolburg, South Africa in 1976 and be ready for commercial application in 1978.

Lurgi gasifiers are currently limited to the use of 1-3/4 inch to 3/16 inch sized, non-caking coal. Recent tests conducted at Westfield, Scotland, however suggest that with a specially designed stirring arm and with proper modifications in operating conditions, it may be possible to operate with highly caking coals.

One of the main advantages of this gasifier is its countercurrent operation which favors a moderately high methane concentration (about 10 percent) in

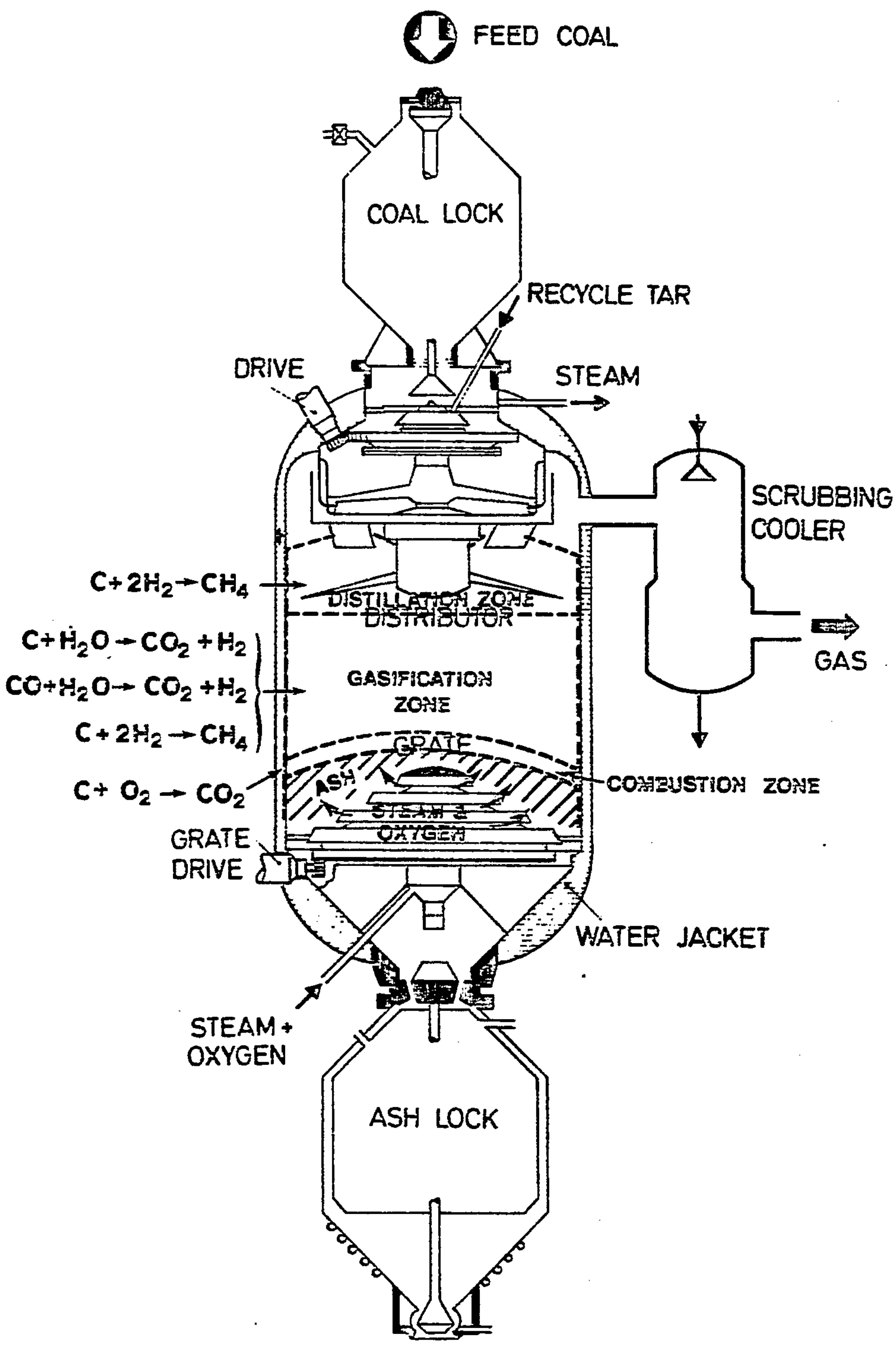

FIGURE 4. - LURGI GASIFIER

the dry raw product gas. This amounts to about 40 percent of the methane in the final SNG product. A relatively low oxygen rate (about 0.27 lb/lb coal) fed to the gasifier is required for gasifying the reactive western coals.

Lurgi has reported demonstration tests on a pelletted nickel methanation catalyst that has operated 4000 hours and has a projected life of over 16,000 hours (7). The catalyst has operated with inlet temperature of 200 to 300° C and outlet temperatures as high as 450° to 470° C. They found that water vapor tends to accelerate catalyst deactivation.

In view of all the proven steps of the Lurgi process, its prospects for a high on-stream time factor are very good. The gasification stream factor is expected to be well over 90 percent. Current commercial plans call for a total of about 10 Lurgi plants. Estimated capital costs (1973 basis) run at about .6 billion dollars increasing about .1 billion for mining. Estimated average unit cost of the gas over a 25-year plant life is $1.17 per mscf. (8)

B. Koppers-Totzek

The Koppers-Totzek (K-T) process offered commercially as a route to SNG, employs process steps similar to that of the Lurgi process. The main difference lies in its gasifier (see figure 5). The gasifier is refractory lined and operates under slagging conditions. Pulverized coal (70 percent through 200 mesh) is entrained in the oxygen-steam feed stream and is burned producing a flame zone temperature of about 3500° F. Endothermic reactions between the carbon and the steam reduce the flame temperature to about 2700° F, producing CO and H_2. Gasification occurs at atmospheric pressure with an oxygen feed of .58 to .8 lb/lb coal and a steam feed rate of 0.15 to .34 lb/lb coal to produce a 303 Btu/scf synthesis gas with a very low methane content (0.10 percent). About 50 to 70 percent of the slag drops into the water quench tank and is disposed of; the remainder is entrained in the gas stream. Steam fed to the gasifier comes from the water jacket. High gasification temperature, turbulence, and small coal size results in extremely rapid gasification rates and high coal throughputs as residence time is only about 1 second. A four-burner unit will gasify 850 tons of coal per day; 27, four-headed K-T gasifiers (includes three stand-by units) would be required for a 250 billion Btu per day SNG plant.

As shown in figure 6, gas leaving the gasifier is quenched with water to solidify entrained molten ash, and prevent it from solidifying on the walls of the waste heat boiler. After passing through a waste heat boiler the gas is scrubbed to reduce entrained solids to 0.002 grams per scf. Electrostatic precipitators may reduce particulates to 0.0001 grain per scf. The scrubbed gas is compressed to 450 psig and purified to remove hydrogen sulfide and a controlled quantity of carbon dioxide. The purified gas is shifted and then methanated in three stages, converting the waste heat to 1500 psi steam and using no gas recycle. The methanated gas is purified to remove the remaining CO_2 and dehydrated to produce pipeline gas. Overall thermal efficiency for this process is about 50 percent.

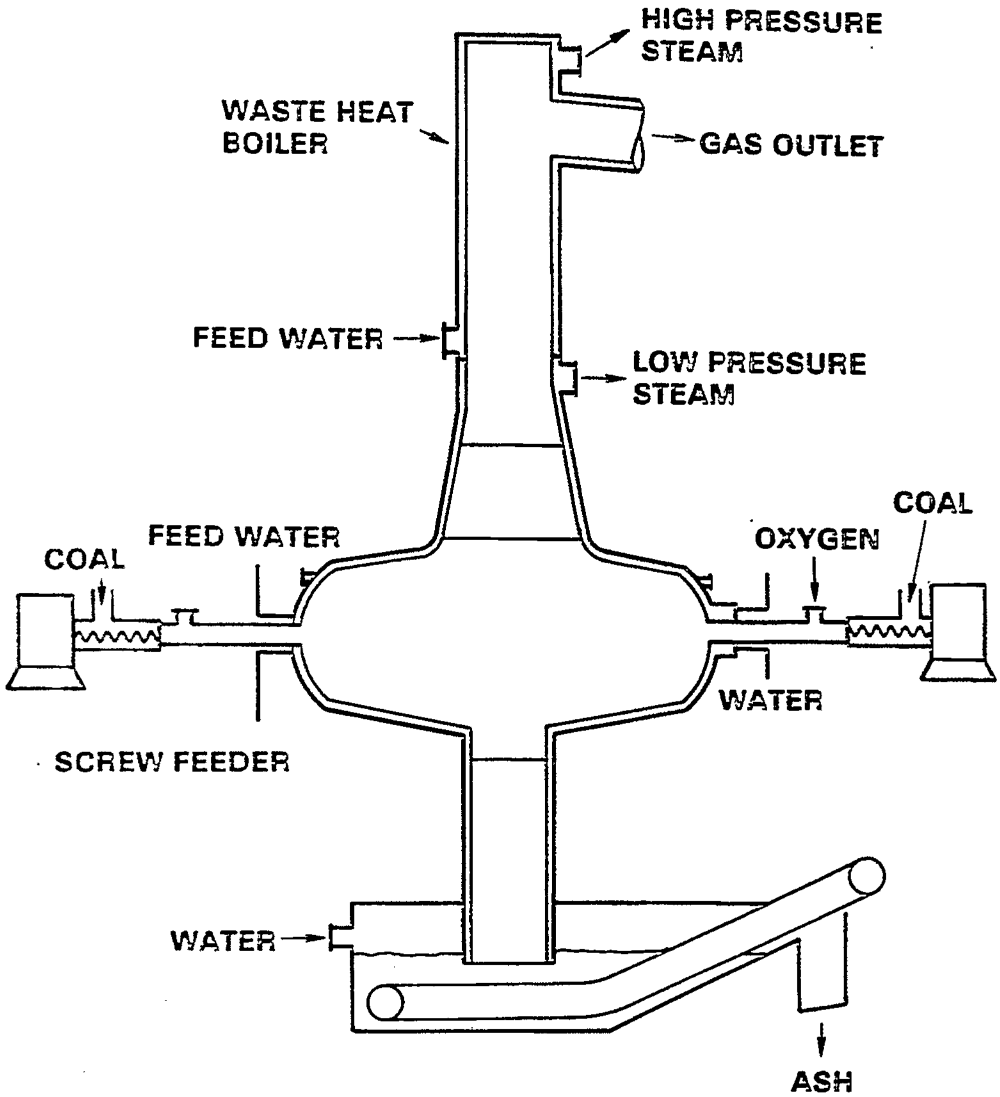

FIGURE 5. - KOPPERS-TOTZEK GASIFIER

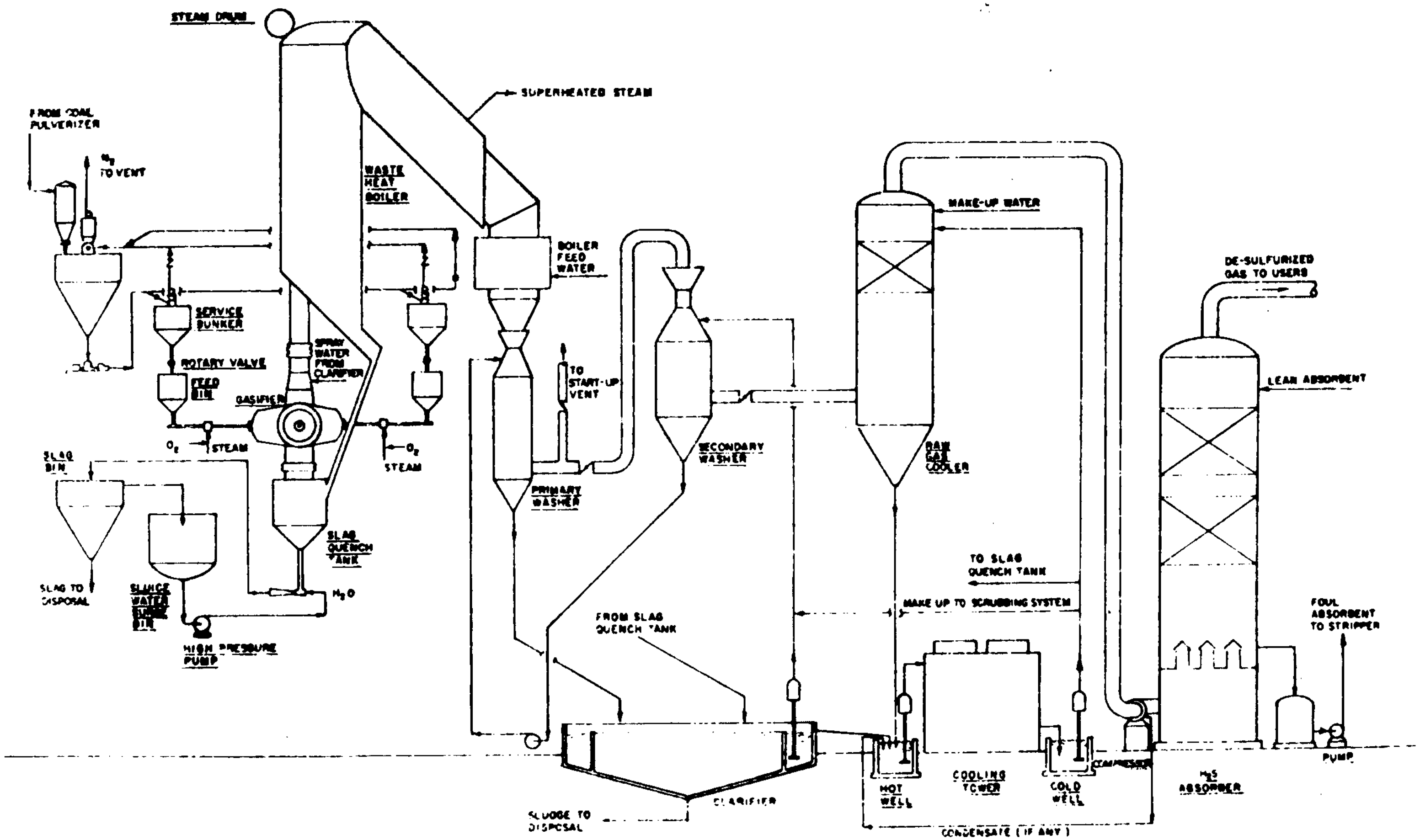

FIGURE 6. - KOPPERS-TOTZEK GASIFIER SYSTEM.

Particulate-laden water from the gas cleaning and cooling systems is treated in a clarifier for ash removal and then returned for further gas scrubbing duty. The K-T process offers the advantage of comparatively clean gas and water product streams. The sulfur in the gas is only H_2S and COS which are readily removed. No objectionable water contaminants such as phenols, tars, oils and pyridine are present in the circulating quench waters. This makes the water easily treated and reused.

Estimated average selling price of SNG over a 20-year life is \$1.86/mm Btu (3) of \$1.81 per mscf of 975 Btu/mscf gas. Price is based on October 1974 prices and wages, western coal at \$7/ton, and \$400,000,000 plant investment for \$250 billion Btu/day plant.

It is realized that gasification at higher pressures (15 to 30 atm) would bring an estimated 12 percent decrease in energy consumption and a 5 percent decrease in selling price. A pressurized version of the K-T process therefore is under development in a joint venture by Koppers and Shell International. To increase gasifier throughput, a 6-headed atmospheric pressure gasifier is in the final design stage and is expected to double the throughput of the present 4-head gasifier. A 6-ton per day SNG demonstration plant complete with the shift and methanation units is planned to be erected and operated in Germany.

Prospects for a commercialized K-T coal to SNG plant should improve greatly if a high on-stream factor of the gasifier is retained after incorporating the planned improvements.

C. Winkler Gasifier

To date there is no complete coal-to-SNG process offered commercially that employs a Winkler gasifier. The gasifier itself is offered commercially for operation at pressures up to 3 atmospheres (9). As shown in figure 7, the Winkler gasifier is a fluidized bed system; it is refractory lined and 13- to 15-feet in diameter. Dried, crushed coal (3/8" x 0) is fed to the unit. Steam and oxygen are fed in the bottom of the fluid bed. Gasification temperature is in the range of 1500° to 1850° F depending upon the type of coal. Because of the high temperatures, all tars and heavy hydrocarbons are reacted. About 70 percent of the ash is carried over by gas and 30 percent of it is removed from the bottom of the gasifier by the ash screw. Unreacted carbon carried over by gas is converted by secondary steam and oxygen in the space above the fluidized bed. As a result maximum temperature occurs above the fluidized bed. To prevent ash particles from melting and forming deposits in the exit duct the gas is cooled by a radiant boiler section (not shown) before it leaves the gasifier. Methane content of the raw gas is low, on the order of 2 percent; therefore considerable upgrading of the gas would be required to obtain SNG product.

The unit can process 1500 ton per day of coal and reports an average on-stream factor of over 91 percent. Thus, if 20,000 ton/day of coal were to be gasified for conversion to SNG, about 13 Winklers plus 2 spares would be required. Although coals containing up to 8 percent moisture can be handled without drying, the system cannot gasify the caking coals without prior treatment to destroy the caking characteristic.

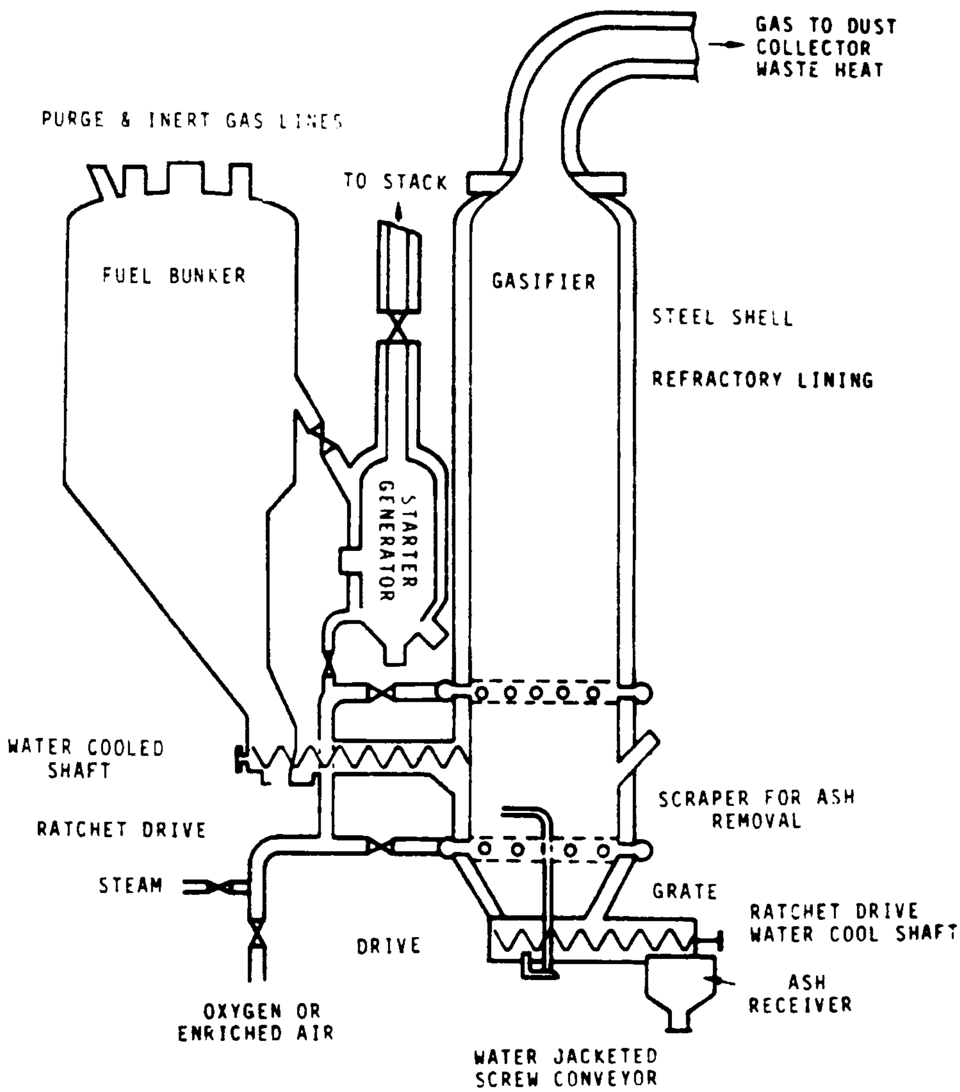

FIGURE 7. - WINKLER GASIFIER

Pilot Plant Studies

A. Hygas

The main feature of the Hygas process is the staged hydrogasification reactor where a large amount of the coal is converted directly to methane by two-stage counter current reaction with hydrogen at 1100 psi pressure. Major features of the hydrogasification system shown, in figure 8, are as follows: Lignite (12 x 0 mesh size) slurried with light oil is fed to the fluidized bed slurry drier where rising 1200° F product gas from the first stage hydrogasification section vaporizes the slurry at about 600° F. The dried coal descends to the bottom of the first stage hydrogasification section where it is entrained upward and reacted with hot (about 1750° F) rising gases from the second stage hydrogasification section. After disengagement, the partially gasified char flows down to the second stage hydrogasification section which is a fluidized bed reactor where the char then reacts further with hydrogen-rich gas fed at the bottom of the second stage.

The IGT pilot plant hydrogasifier has a design capacity of 3 ton per hour to produce 1.5 million scf per day of pipeline gas. The reactor is a 66-inch inside diameter by 134 ft long water jacketed vessel. Space between the pipes and the shell in the upper part of the reactor is pressurized with nitrogen.

There are three alternative methods of generating the required hydrogen as follows:

(1) Electrothermal gasification--char from the bottom stage of hydrogasifier is gasified with steam at 1900° F in a fluid-bed electrothermal gasifier. The required heat is supplied by passing electrical current through the fluid bed. Residual char from the electrothermal gasifier goes to the power plant for generation of electricity and hydrogen-rich gas is sent to the hydrogasifier. A 30-inch diameter electrothermal pilot plant reactor was erected at IGT. Batch tests demonstrated the feasibility of computer control of either current flow of power input to the varying resistive load of the fluid bed. The large power requirement (700 kw in these tests) adversely affects the economics of this scheme.

(2) Oxygen-steam gasification (illustrated in figure 8)--the char from hydrogasification is gasified in a fluid bed with oxygen and steam at 1900° F to form synthesis gas which mainly consists of hydrogen and carbon monoxide. Required heat is supplied by combustion of a portion of char with oxygen. Tests have demonstrated the feasibility of the integrated steam-oxygen gasifier. The raw dry product gas from the hydrogasifier should contain about 19 percent methane.

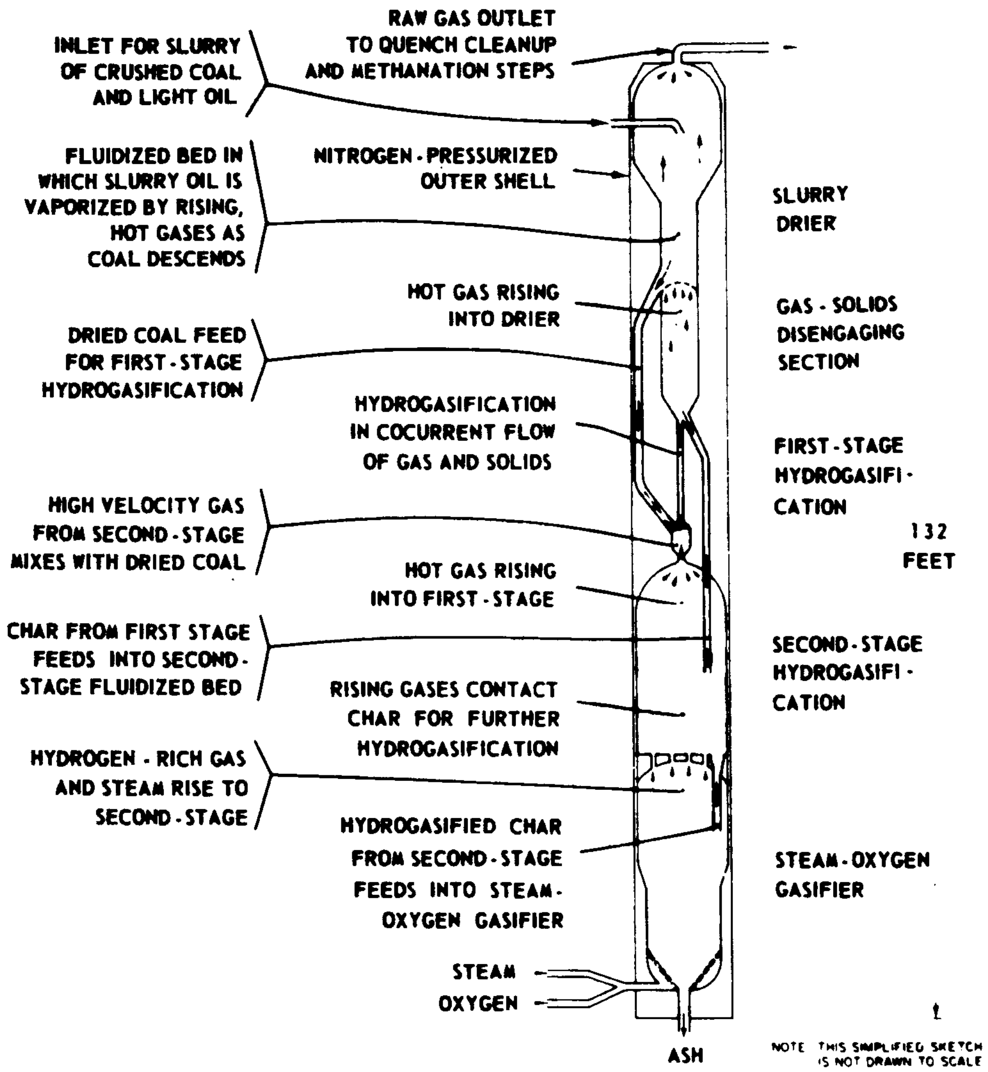

FIGURE 8. - IGT PILOT PLANT HYDROGASIFICATION REACTOR SECTION

(3) Steam-iron process--the hydrogen required for the hydrogasification is produced by the reaction of steam with FeO at 1500° F and 1500 psi in an oxidizer unit. The resulting iron oxide, Fe_3O_4, is returned to a reducer where it reacts with hot producer gas made from char in an airblown gasifier to make FeO for recycle. The spent product gas is cooled to 1200° F and expanded to recover energy for air inlet compression. Figure 9 shows the 1200 psi pilot plant system under construction at IGT. Both the product and steam-iron reactors are water jacketed. Producer temperatures will be as high as 2100° F. Although the slurry feed system will be used to feed the char to the producer during independent testing, hot dry char from the second stage hydrogasifier will feed into the producer by gravity flow when the steam-iron unit is integrated with the hydrogasifier as shown in figure 10. Separate studies on fluidization and transport of iron ore, and fluid-bed gasification of Husky lignite char and evaporization of char-water slurry in a tubing coil have established the needed plant design parameters (10).

Tests simulating a steam-hydrogen feed from a steam-iron plant have yielded a raw hydrogasification product gas containing up to about 39 percent methane dry basis and about 40 percent carbon gasification.

A serious difficulty encountered in maintaining good solids flow in the hydrogasifier system was corrected by preventing the accumulation and recirculation of coal fine between the slurry dryer and the first stage. This was accomplished by extending the 30 inch diameter section of the slurry dryer all the way to the top exit nozzle and eliminating the top expansion zone where settling out of the fines previously occurred. An external cyclone was installed to remove fines prior to quenching.

Mechanical problems of pumping up to 50 wt-percent coal slurries into the system have been solved (11). A linear velocity of over 5 ft/sec is sufficient to prevent settling of coal particles. Reciprocating mud pumps with Stellite coated valve seats deliver the slurry under pressure. Aromatic oil with a low heat of vaporization was the slurry medium used in initial tests when a hydrogen-steam mixture was the feed in the hydrogasifier. However, present operation with hydrogen fed directly via synthesis gas from a steam-oxygen gasification step should produce a larger amount of heat sufficient to use a water slurry instead of an oil slurry.

The char or ash is satisfactorily let down to atmospheric pressure in a 25 percent solids water slurry through a tungston carbide coated Willis choke valve. (11).

If a caking bituminous coal such as Illinois No. 6 is to be used, it is to be pretreated in air and at atmospheric pressure before it is fed into the hydrogasifier to prevent caking. The IGT pretreater was fitted with special heat exchanger coils to control the bed temperature adequately (about 800° F max). Tests on Illinois No. 6 coal have been successful in reducing the Free Swelling Index of the coal from 5 and 7 to 1 or less.

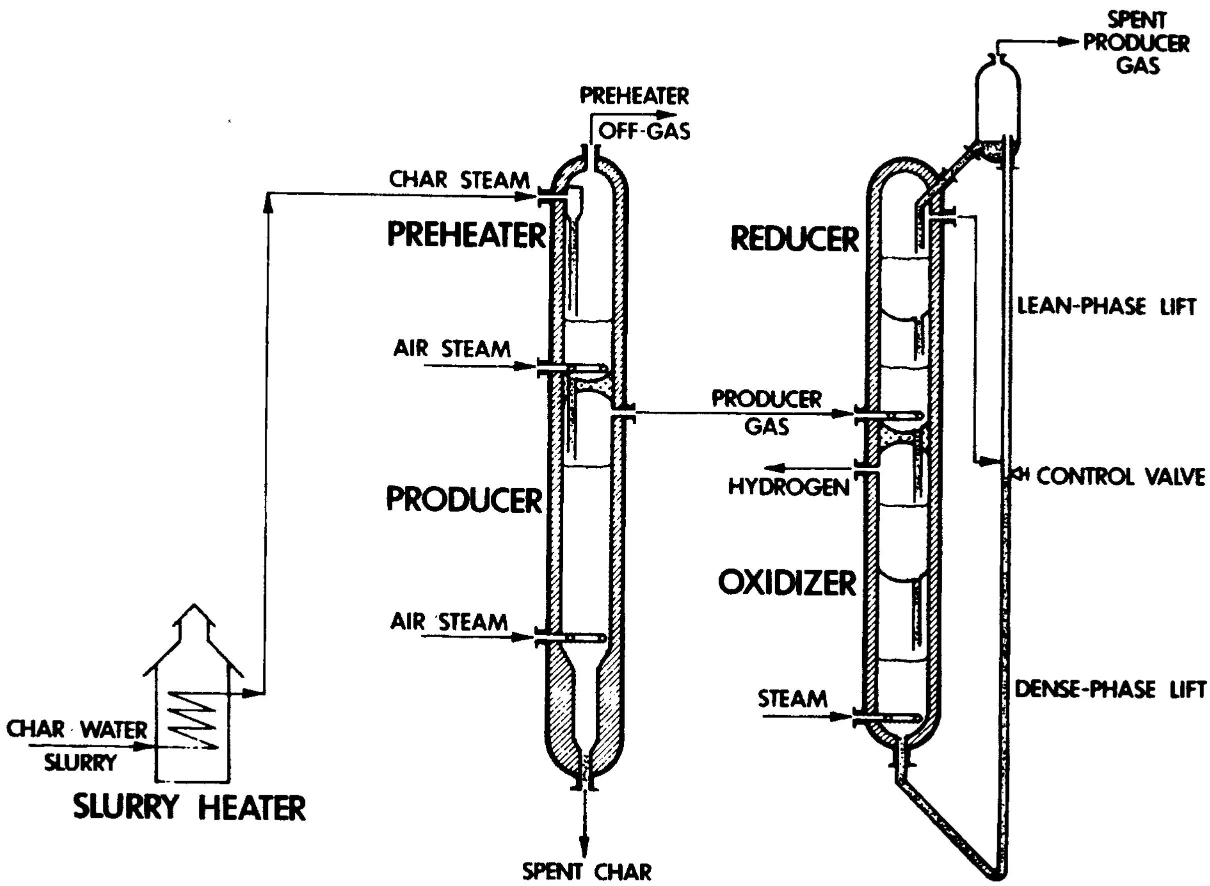

FIGURE 9. - STEAM-IRON PILOT PLANT REACTOR SYSTEM

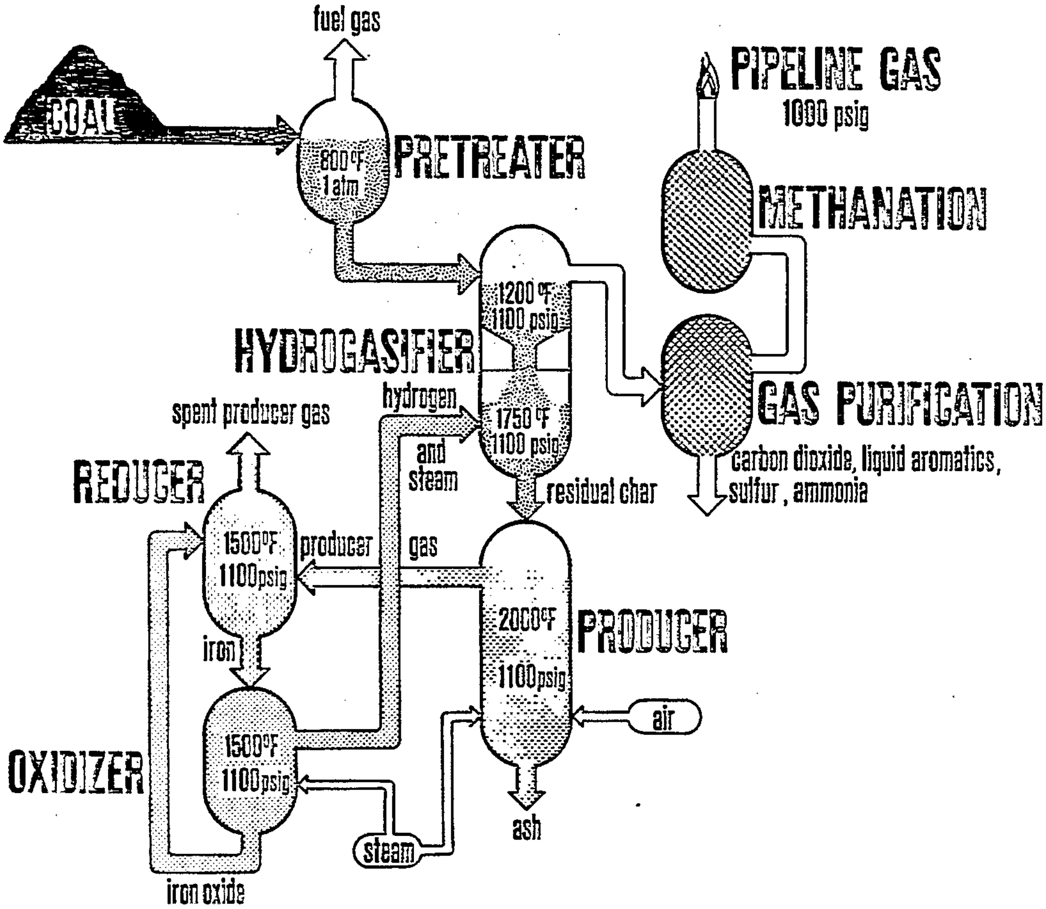

FIGURE 10. - IGT HYGAS PROCESS (HYDROGEN GENERATION BY THE STEAM-IRON PROCESS)

In the pilot plant a diglycolamine-water scrubber reduces CO_2 and sulfur compounds, respectively, to about 150 and .18 ppm. Further scrubbing with caustic solution reduces these values further to <50 pm CO_2 and 0.00 ppm sulfur compounds. Two stage methanation operating at 3600 and 2300 hourly space velocities in the first and second stages respectively, yield a product gas with < 0.1 percent CO. While no shift conversion unit is used in the pilot plant, the commercial plant will have a shift conversion step.

Design work has begun for an 80 million scf per day demonstration plant. It is projected that it can be built in 1978. For a 250 million scf per day commercial plant, coal requirements are expected to be low, about 15,000 ton per day. The projected number of 24-ft diameter hydrogasifiers required would be two with no spares. (12)

B. CO_2 Acceptor Process

The CO_2 Acceptor Process operates with a circulating lime-bearing material called the acceptor which supplies the heat for the gasification reaction at 1450° to 1570° F. As shown in the diagram in figure 11, the acceptor which can be either limestone or dolomite supplies the heat in the gasifier by the reaction:

$$CaO + CO_2 \rightarrow CaCO_3 \qquad \Delta H = -76,200 \text{ Btu/lb mole } (77° \text{ F})$$

The acceptor reaction in reversed at about 1850° F in the regenerator where heat is supplied by burning residual char from the gasifier with air. Ash containing about 5 percent sulfur is removed from the regenerator by elutriation, collected and discharged via an external cyclone and lockhopper system, and is to be treated for sulfur recovery. Seals between the gasifier and regenerator are maintained by purged standlegs of solids. Deactivated acceptor is withdrawn from the gasifier, and make-up acceptor is added in the return line to the regenerator. The product gas is cooled, washed and cleaned up for methanation, compression and drying. Flue gas from the regenerator is to be cleaned up and used to power a gas turbine.

Design throughput for the pilot plant gasifier is 40 ton per day dried lignite to produce 2 million scf per day of raw gas or a yield of 50 mscf/ton of coal fed. A 30 ton per day (dried lignite) test operation at 150 psi was reported to yield 45.6 mscf/ton (12). Fluidized velocities in the gasifier and regenerator were 1.12 and 2.87 ft/sec respectively.

The coal to SNG thermal efficiency is 77 percent. The dry raw product gas contains 14 percent methane. It also has a H_2/CO mole ratio of more than 3 which eliminates the need for a shift converter. This process, therefore, successfully produces a suitable synthesis gas for conversion to pipeline gas without the use of oxygen.

Other salient features of the process are: (1) carbon conversion is nearly complete with the ash containing less than 1 percent of the carbon in the feed; (2) concentrations of CO_2 and H_2S in the product gas are

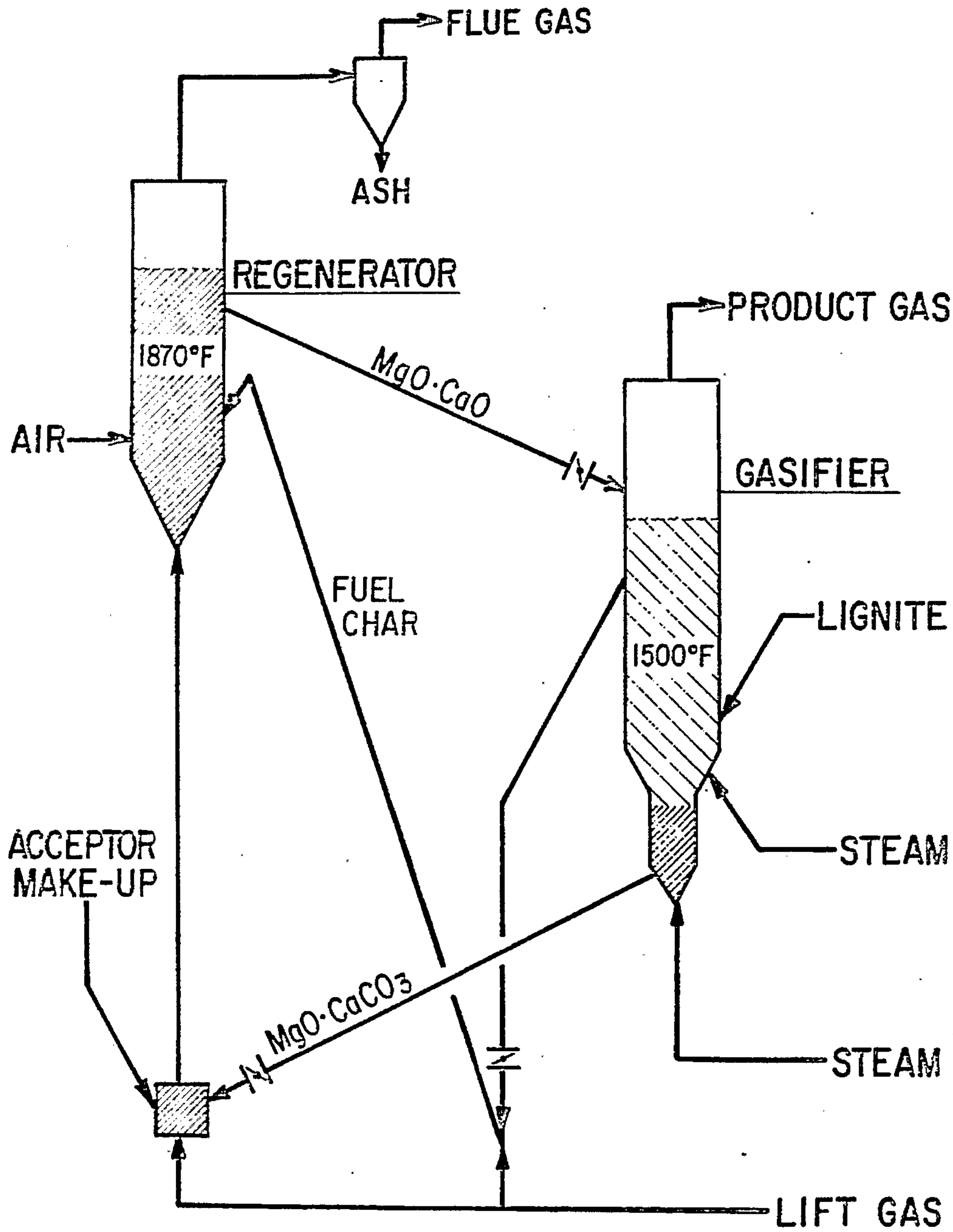

FIGURE 11. - CO_2 ACCEPTOR PROCESS DIAGRAM

comparatively low (10.9 and .13 percent respectively compared, for example, with 14.7 and 0.6 percent for a Lurgi gas) reducing gas clean-up requirements; (3) to avoid slagging of the fuel char and to prolong the activity of the acceptor, gasification and regeneration temperatures are limited. This in turn requires that the coal feed be limited to the reactive lignites and subbituminous coals.

Feasibility of this unit was demonstrated with 252 hours of integrated operation and 171 hours of operation in which the circulating acceptor supplied the entire gasification heat. Advancements reported in the development of the system include: (1) an improved start-up procedure in which attrition resistant dead-burned dolomite is used as the starting acceptor instead of the more active but more friable fresh calcined dolomite; (2) use of a Velva lignite feed was successfully demonstrated with no evidence of tar or pitch formation in the gasifier. Lignite char was used in initial tests; (3) erosion in the acceptor lift line was overcome through redesigning and coating the expansion joint with Stellite and limiting the carrier gas velocity; (4) corrosion of start-up heater pipes (Incoloy 800) by sulfur and by carburization-decarburization reactions has been arrested by addition of steam and by zinc oxide removal of sulfur compounds from the recycled preheat gas; (5) unstable operation due to accumulation of a narrow size fraction (28 x 65 mesh) of solids is expected to be overcome by installation of a char removal system which should control the accumulation of intermediate fines; and (6) refractory failures associated with porous block insulation in both gasifier and regenerator were overcome by using a hard face castable backed by an insulating castable.

Future goals of the CO_2 acceptor program include the following: (1) conduct an extended run of 30 days duration; (2) conduct tests on lignites with higher sodium content (5 to 14 percent); (3) operate with a Montana subbituminous coal; (4) test an alternate acceptor (Minnekahta limestone); and (5) install and operate a packed bed methanation reactor system utilizing a Dowtherm cooling system.

Based on a 59-foot diameter gasifier, only one unit will be required for a commercial plant operation (12).

C. SYNTHANE Process

A simplified flowsheet of the SYNTHANE process is shown in figure 12.

Coal crushed to 20 x 0 mesh size is pressurized and fed through lock-hoppers to the pretreater. About 12 percent of the total steam and oxygen required in the process is fed to the pretreater to destroy any caking characteristics of the coal. The operating temperature of this stage is about 800° F. Coal is partially devolatilized and its caking tendency is destroyed. Coal along with any separated volatile matter and excess steam is fed to the top of the fluid-bed gasifier and a mixture of steam and oxygen is introduced at the bottom. The

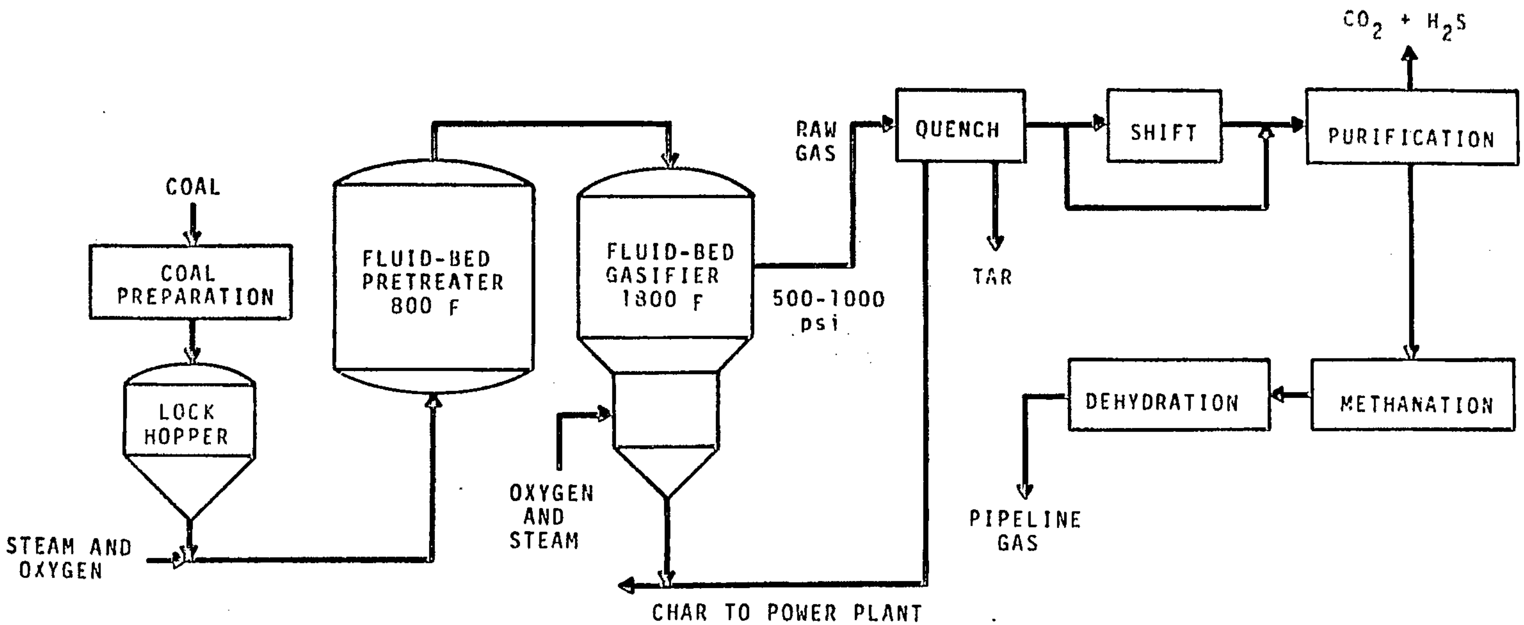

FIGURE 12. - SYNTHANE PROCESS

gasifier operates at about 1800° F and 500-1000 psi to produce a gas containing about 18 percent methane dry basis. Char, containing roughly 30 percent of the carbon from the original coal is removed combined with residual dust and tar collected from the raw product gas and sent to the power plant.

Dust particles are removed from the raw gas by internal cyclones and returned to the bed. Residual dust, tar, and excess water vapor are removed by water wash. The cleaned gas goes to the shift converter, where the overall H_2/CO ratio is raised from (1.7) to (3.1). Gas from the shift converter is treated by hot carbonate scrubbing to remove CO_2 and H_2S and is further purified with activated carbon to get sulfur below 0.1 ppm. The purified gas is methanated and dehydrated to produce pipeline gas. About 68 percent of the total methane produced is made in the gasifier; the rest is produced by methanation.

Based on a highly caking Pittsburgh seam coal, a 250 million scf per day plant would require about 14,250 ton per day coal feed. With 927 Btu/scf product gas the overall thermal efficiency is about 60 percent. (14) Oxygen and steam requirements per pound of coal feed are 0.26 lb and 0.9 lb, respectively.

Two methods are being developed to convert the bulk of the CO in the purified product synthesis gas to methane: the hot gas recycle (HGR) reactor (15), and the tube-wall reactor (TWR) (16). Both presently use Raney nickel catalyst flame-sprayed into tubes or onto plates and partially extracted. The HGR reactor utilizes the heat capacity of the recycle gas to remove the exothermic heat of reaction (65 Btu/cu ft of H_2 + CO) converted from the catalyst. In the TWR system, the heat of reaction is transferred through the sprayed tube wall to a boiling heat transfer liquid. Both operate at about 750° F and the highest pressure used has been 400 psig. Results so far indicate a catalyst life exceeding six months and a feed gas throughput of 2000 scf per cubic foot of reactor volume.

A second stage adiabatic methanator employing pelleted nickel catalyst will insure complete conversion of the CO to methane.

Efforts are being made to develop improved methods of spray coating of Raney nickel and to minimize deactivation of the catalyst.

A 3-ton-per-hour SYNTHANE pilot plant gasifier has a unique char cooling system that will be demonstrated. A diagram of the char cooler is shown in figure 13. Hot 1800° F char from the cone of the cone of the gasifier is sparged with steam while it flows by gravity into the fluidized bed of the char cooler. Char flow rate is controlled by varying the pressure difference between the char cooler and the gasifier bed. Cooling of the char to 600° F is achieved by vaporization of water spray in the fluid bed. The generated steam goes through filters to the shift reactor. The char overflows to the ash lock hopper for discharge into a water quench and slurry system for separation by filtration.

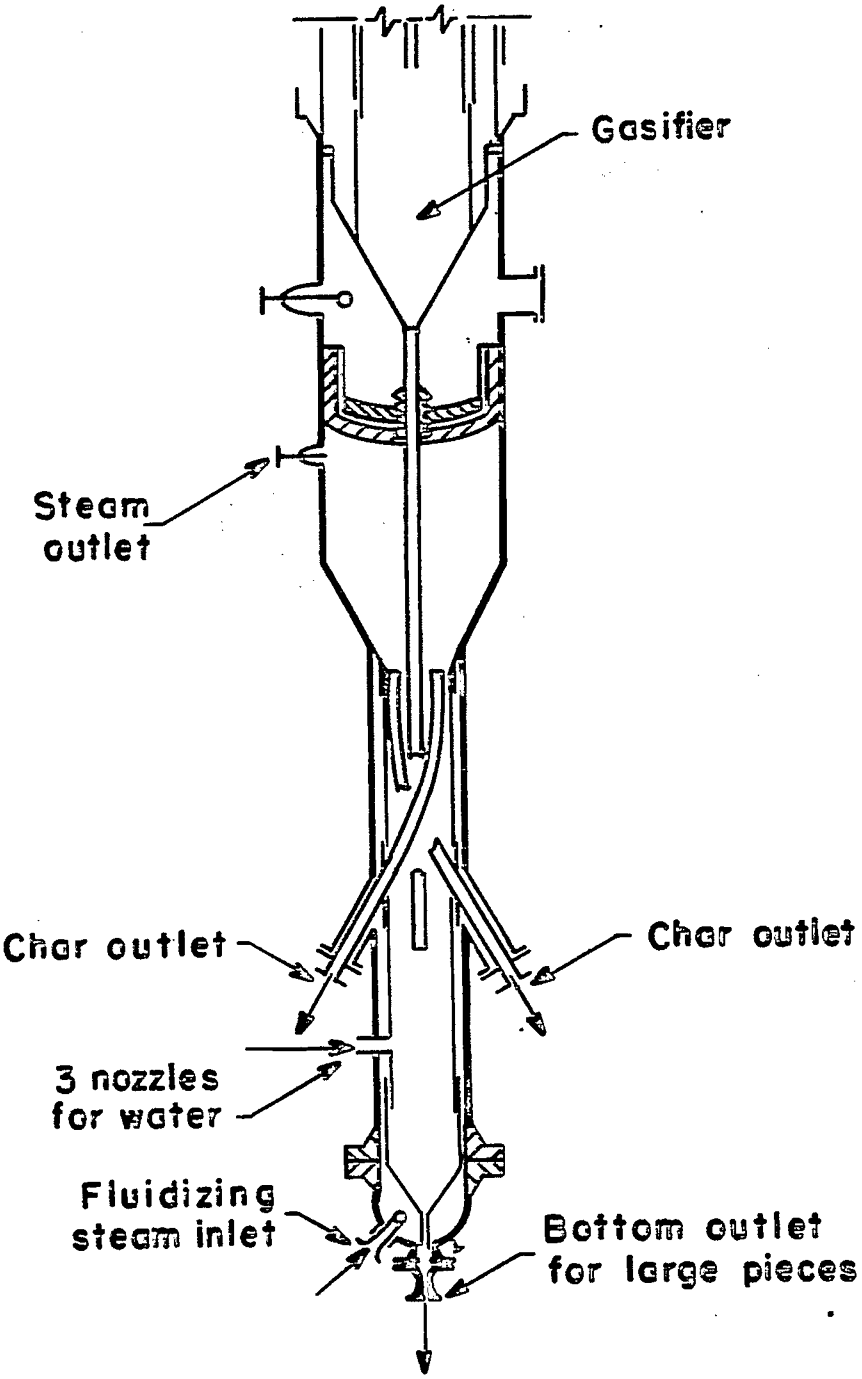

FIGURE 13. - CHAR COOLER

Condensate water from the gasifier is similar to weak ammonia liquor from a coke plant, as shown in table 1. One waste water treatment process based on commercial experience has been proposed (17). It involves the steps of ammonia, stripping and lime treatment to remove the ammonia, final tar separation, biological treatment to reduce phenols to about 0.1 mg/l and thiocyanates by 70 percent, and finally a polishing step using gasifier char to remove the remaining phenols.

TABLE 1. - Byproduct water analysis from SYNTHANE gasification of various coals, mg/l (except pH)

	Coke plant	Illinois No. 6 coal	Wyoming subbituminous coal	Illinois char	North Dakota lignite	Western Kentucky coal	Pittsburgh seam coal
pH..............	9	8.6	8.7	7.9	9.2	8.9	9.3
Suspended solids,	50	600	140	24	64	55	23
Phenol..........	2,000	2,000	6,000	200	6,600	3,700	1,700
COD.............	7,000	15,000	43,000	1,700	38,000	19,000	19,000
Thiocyanate.....	1,000	152	23	21	22	200	188
Cyanide.........	100	0.6	0.23	0.1	0.1	0.5	0.6
NH_3.............	5,000	1/ 8,000	9,520	2,500	7,200	10,000	11,000
Chloride........	-	500	-	31	-	-	-
Carbonate.......	-	2/ 6,000	-	-	-	-	-
Bicarbonate.....	-	2/ 11,000	-	-	-	-	-
Total sulfur....	-	3/ 1,400	-	-	-	-	-

[1] 85 percent free NH_3.
[2] Not from same analysis.
[3] S = 400
SO_3 = 300
SO_4 = 1,400
S_2O_3 = 1,000

The treated cooling water can then be used as cooling water make-up. An estimated 1974 operations cost of the water treatment step was $0.014 per mscf SNG produced. Research is being conducted to develop improved water treatment processes with the possibility of adding a step to recover the phenols.

A Stretford sulfur recovery system is used to convert the H_2S contained in the regenerator acid gases to free sulfur. Part of the sulfur-free CO_2 is recompressed to pressurize the lock hoppers. Char from high sulfur (4 percent sulfur) feed coals when burned to generate steam will require flue gas treatment for removal of SO_x.

The 72 ton-per-day pilot plant has been constructed. It should be ready to start a test campaign for demonstrating all its features by the spring of 1976.

A commercial size plant would require three 31-foot-diameter reactors with no spares (12).

D. Bi-Gas

In the Bi-gas process (18) a pulverized coal (70 percent through 200 mesh) is slurried in water, pressurized to over 1500 psi, spray dried, and separated in a cyclone separator. Figure 14 shows the dried pressurized coal being fed along with steam to the upper zone of the reactor and rapidly heated to 1700° F by 2700° F gas from the lower part of the reactor. Devolatilization of the coal occurs within 8 to 10 seconds residence time to produce methane, synthesis gas, and char. The char leaves the top of the gasifier with the raw product gas, is quenched to 800° F by atomized water, separated in an external cyclone, and returned to the lower section of the gasifier by a steam eductor. The char is gasified to H_2 and CO at 2700° F in the lower section with steam and 1200° F oxygen. Molten slag forms in the 2700° F zone, drains into quench water, and is depressurized and discharged via dual weigh hoppers. Burners assist in keeping the slag molten and free flowing. A slag breaker is provided to knock off any stalactites which may form.

Further gasifier details may be seen in the diagram of the pilot plant gasifier shown in figure 15 (19). In the upper gasifier zone (Zone II) concentric injector feed nozzles are spaced around the shell so that the feed streams do not strike the refractory but impinge in the gas space. The entering coal particle velocity is about 20 to 30 ft/sec; exiting char particle velocity is 1.5 to 2.5 ft/sec. In the slagging zone (Zone I), the three char burner nozzles are arranged to fire cyclonically in a 6-inch diameter circle. The temperature of the 5-foot diameter pressure shell is controlled to below the 650° F maximum by verticle close-spaced water-cooled tubes which are in turn covered with insulation and dense refractory. An inner 24-inch diameter wall of vertical water-cooled tubes covèred with dense refractory forms the Zone I furnace wall that is suitable for molten slag service commonly found in cyclone furnaces and other slag tap furnaces. The throat restriction between Zones I and II serves to prevent raw coal from dropping down into Zone I and to aid in separation of molten slag before the gas enters Zone II.

The raw gas from the gasifier, containing about 15 percent methane (dry basis) and 25 percent water vapor, in quenched by hot condensate to 660° F leaving sufficient steam for the shift reaction. After the shift reaction, a Selexol physical absorption process is used to remove H_2S and CO_2. A Claus process can recover sulfur from the H_2S. Fluid-bed methanation followed by additional CO_2 removal by Selexol yields the final SNG.

The Bi-gas gasifier can operate with caking coals as well as non-caking coals and requires no pretreatment step. Oxygen and steam feed requirements

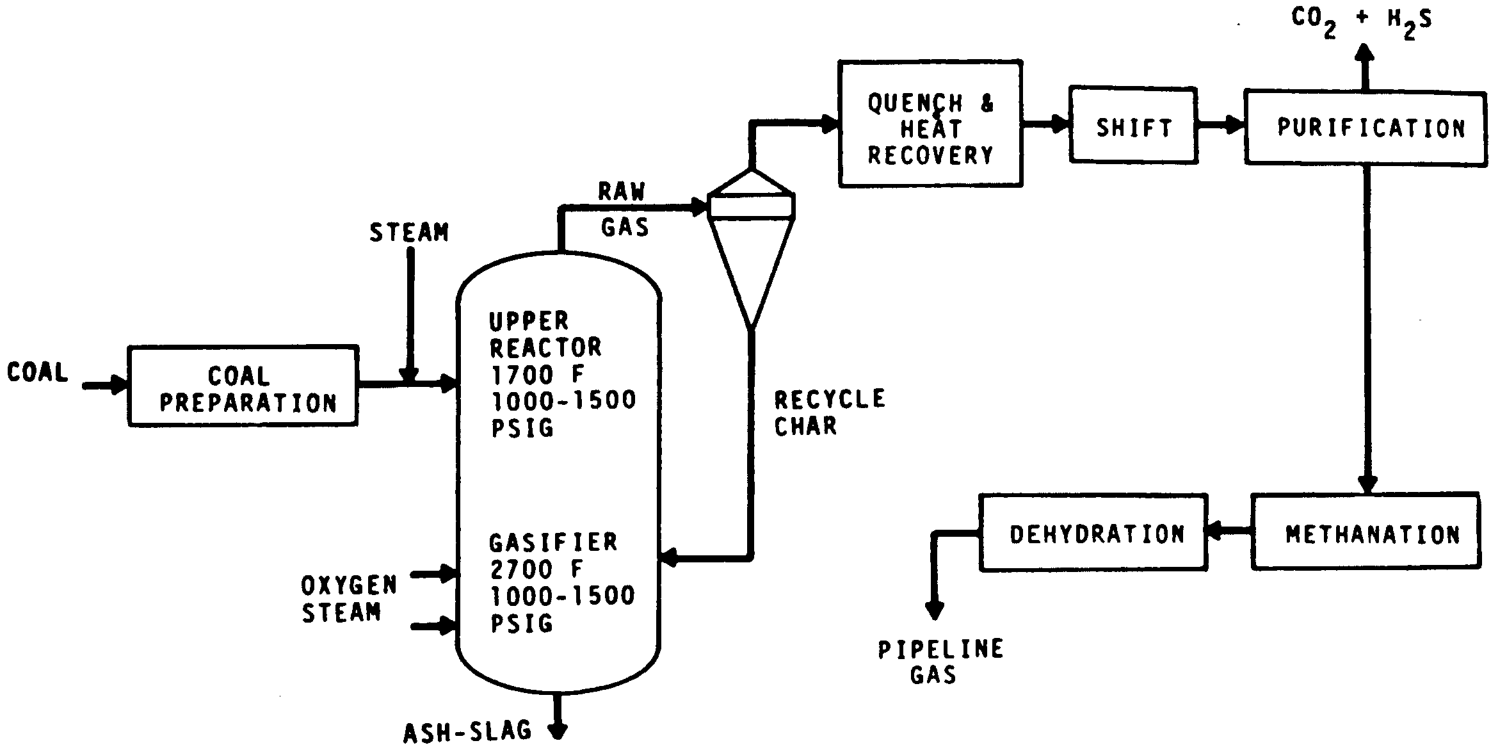

FIGURE 14. - BIGAS PROCESS

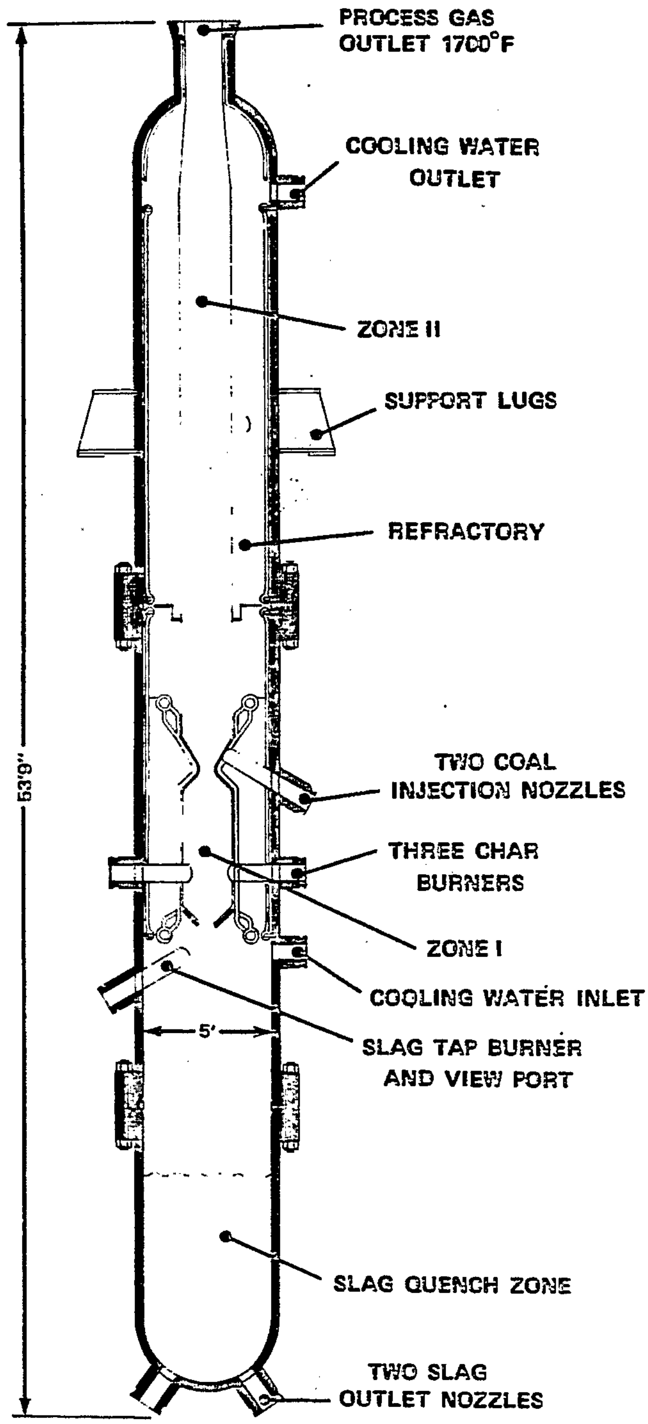

FIGURE 15. - BI-GAS REACTOR

per pound of coal feed are 165 pound and 1.2 pound respectively. No tars or oils are formed, and carbon gasification is essentially complete.

Tests conducted at 1000 psi and temperature ranges from 850° to 970° F in a fluid-bed methanator with a 1 cubic foot charge of catalyst typically yielded product gases containing 1.0 tp 2.3 percent CO at about 1750 hourly space velocity (20). Catalyst attrition loss was about 0.2 percent per day. The search continues for improved catalyst.

Construction of the 5-ton-per-day pilot plant is scheduled to be completed in 1975.

A commercial design of the Bi-gas gasifier requires only one reactor per plant. Inside diameters of the upper and lower stages are 9-feet and 12-feet respectively (12).

E. Agglomerating Burner--Gasification

Consideration is being given to using an agglomerating burner--gasifier (ABG) system for generating synthesis gas for the production of SNG, because it is a system that does not use oxygen. The ABG system operates on the principles of generating hot ash agglomerates in a fluidized bed coal or char burner and using the hot (about 2050° F) agglomerates to supply the heat to a fluidized bed steam coal gasifier at about 1800° F. At 100 psi operation, the raw synthesis gas is expected to contain about 2-1/2 percent methane (dry basis) and 18 percent water vapor. Flue gas from the agglomerating burner powers a gas turbine which in turn drives the compressor for the combustion air supply and the compressor for pressurizing purified synthesis gas prior to methanation.

A 25-ton-per-day pilot plant ABG system is under construction to operate at 100 psi (21). Figure 16 is a simplified diagram that shows some of the principle features of the pilot plant unit (compressors and gas turbines are not shown). Non-caking coal or pretreated coal sized to 8 x 100 mesh, is fed to the gasifier in the tapered intermediate zone of the gasifier at about the same level for entry of the hot agglomerates from the burner. The agglomerates are carried by steam into the intermediate zone where intimate mixing of the feed streams will occur. Net flow of the coal char is to the top of the gasifier bed where it is discharged. Net flow of agglomerates will be downward to the bottom of the gasifier where it will be discharged at about 1500° to 1600° F. In the lower zone of the gasifier an upward flow of superheated steam strips the carbon from the ash agglomerate. The ash agglomerates are continuously transferred to the bottom of the bottom of the burner with an air lift.

In concept, char from the gasifier is sent continuously to the burner for combustion; but the simplified pilot plant will initially use coal (less than 100 mesh) as the burner fuel, and the gasifier char will be discharged through a lock hopper system.

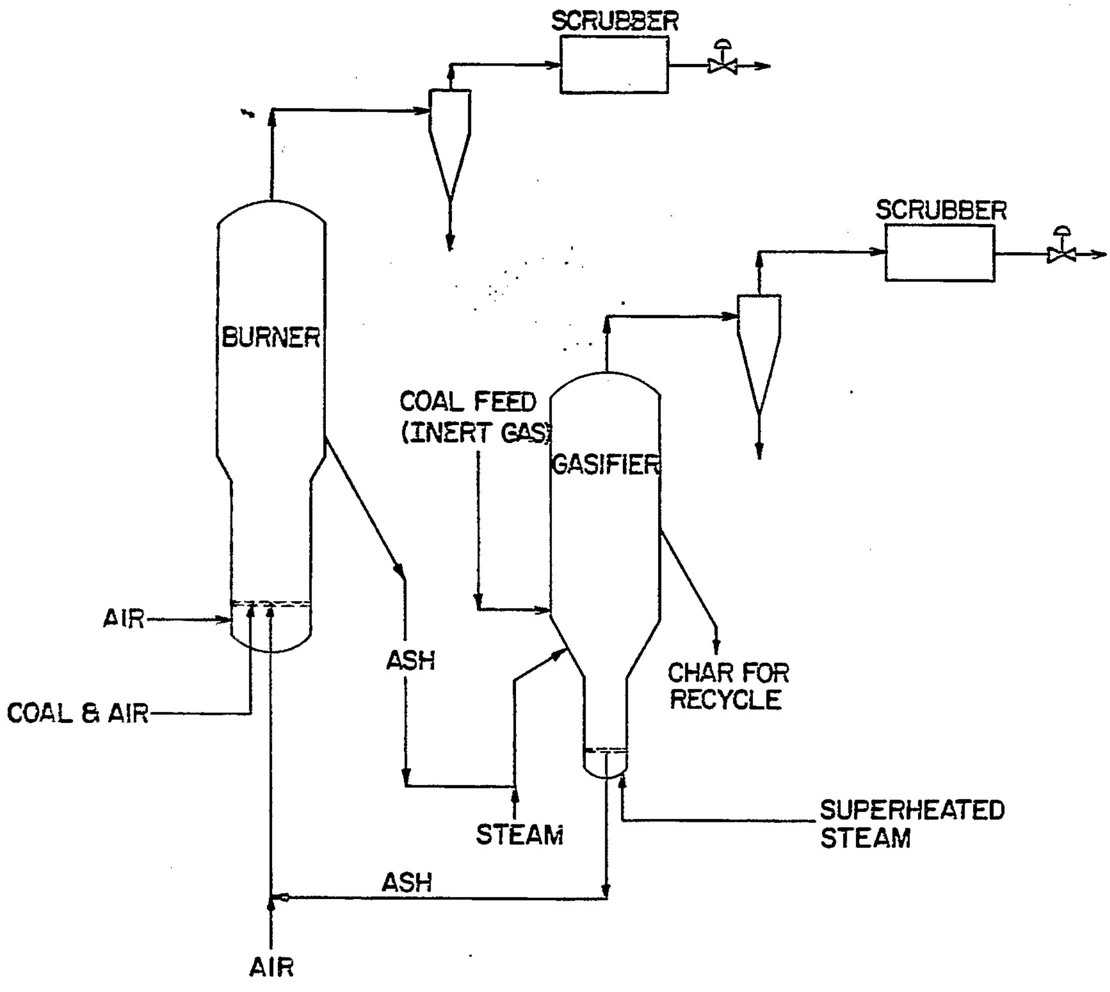

FIGURE 16. - BURNER-GASIFIER FEED AND CIRCULATION SYSTEM

Combustion of the coal reheats the upward flowing agglomerates until they overflow from the top of the fluidized bed to return to the gasifier. Agglomerate circulation will be about 20 tons per hour.

Completion of construction of the pilot plant, due early in 1975, is behind schedule. Initial operation will first establish whether sufficient solids circulation can be achieved. Next the gasifier and burner will be operated. Some of the system features to be evaluated include (1) effect of 100 psi pressure on general operation; (2) effectiveness of seal purging of the solids transfer line; (3) stability of control of solids flow; (4) erosion resistance of ceramic coated valves in the solids transfer lines; and (5) durability of a purged refractory slip joint located underneath a metallic expansion joint bellows and (6) suitability of the burners off-gas as a turbine motive fluid.

A projected commercial plant design specified two ABG units per plant. The diameters of the gasifier and burner are 62 feet and 30 feet respectively. (12)

Other Studies

Development of the Hydrane Hydrogasifier is continuing (22) in the laboratory scale at the Pittsburgh Energy Research Center of ERDA Fossil Energy. The Hydrane reactor is a two-stage system that is fed with unpretreated coal and hydrogen at 1000 psi and 1650° F. The hydrogen comes from steam-oxygen gasification of the residual char followed by shift conversion and the removal of acid gases. About 95 percent of the methane in the SNG product would be produced in the Hydrane unit. The Hydrane process is being evaluated for feasibility and for determination of the minimum scale-up size for a process development unit. Location selected for the process development unit is the Morgantown Energy Research Center of ERDA.

Rather extensive developments have been reported at a recent symposium (23) on the field of catalytic methanation of synthesis gas, as follows: (1) a 4-inch diameter liquid phase methanator operated at up to 1000 psi with particles of nickel catalyst submerged in circulating cooling liquid has achieved 95- to 98-percent conversion of a 20 percent CO feed gas. A pilot plant unit has been designed by Chem Systems and is under construction and (2) Ralph M. Parsons Co. reported 400 psi pilot plant data for their RMP process where the shift conversion step is combined with catalytic methanation in a series of six steps, each step followed by an intermediate heat removal step. The first four reaction temperatures are held at 1000° F and the final two at 600° F and 500° F. These steps are followed by CO_2 removal and a final (7th) methanation step to yield the SNG product. The feed gas may contain 50 percent CO and 50 percent H_2, but it must be desulfurized.

In addition to process development efforts other important areas of concern in developing a viable SNG for coal industry include development of improved auxilliary equipment such as a grinder, power recovery turbines, valves, filters, etc. Development of improved materials of construction is also being studied. Screening and evaluation tests are being conducted in the search for new ceramics and new alloys (24).

In summary, a multi-faceted research and development program to overcome the economic and technological programs involved in efficiently converting coal to SNG is being pursued vigorously by industry and the Federal government. Solutions to most of the major problems are expected within a few short years.

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