

EPA-650/2-74-009-h

August 1975

Environmental Protection Technology Series

EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES

GASIFICATION: SECTION 6. HYGAS PROCESS



U.S. Environmental Protection Agency
Office of Research and Development
Washington, D. C. 20460

EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES

GASIFICATION: SECTION 6. HYGAS PROCESS

by

C. E. Jahnig

Exxon Research and Engineering Company
P. O. Box 8
Linden, New Jersey 07036

Contract No. 68-02-0629
ROAP No. 21ADD-023
Program Element No. 1AB013

EPA Project Officer: William J. Rhodes

Industrial Environmental Research Laboratory
Office of Energy, Minerals, and Industry
Research Triangle Park, North Carolina 27711

Prepared for

U.S. ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF RESEARCH AND DEVELOPMENT
WASHINGTON, D.C. 20460

August 1975

EPA REVIEW NOTICE

This report has been reviewed by the National Environmental Research Center - Research Triangle Park, Office of Research and Development, EPA, and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the Environmental Protection Agency, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

RESEARCH REPORTING SERIES

Research reports of the Office of Research and Development, U.S. Environmental Protection Agency, have been grouped into series. These broad categories were established to facilitate further development and application of environmental technology. Elimination of traditional grouping was consciously planned to foster technology transfer and maximum interface in related fields. These series are:

1. ENVIRONMENTAL HEALTH EFFECTS RESEARCH
2. ENVIRONMENTAL PROTECTION TECHNOLOGY
3. ECOLOGICAL RESEARCH
4. ENVIRONMENTAL MONITORING
5. SOCIOECONOMIC ENVIRONMENTAL STUDIES
6. SCIENTIFIC AND TECHNICAL ASSESSMENT REPORTS
9. MISCELLANEOUS

This report has been assigned to the ENVIRONMENTAL PROTECTION TECHNOLOGY series. This series describes research performed to develop and demonstrate instrumentation, equipment and methodology to repair or prevent environmental degradation from point and non-point sources of pollution. This work provides the new or improved technology required for the control and treatment of pollution sources to meet environmental quality standards.

This document is available to the public for sale through the National Technical Information Service, Springfield, Virginia 22161.

Publication No. EPA-650/2-74-009-h

TABLE OF CONTENTS

	<u>Page</u>
1. SUMMARY	1
2. INTRODUCTION	2
3. SELECTION OF BASIS	4
4. PROCESS DESCRIPTION.	6
4.1 Coal Preparation.	6
4.2 Gasification.	8
4.3 Quench and Dust Removal	8
4.4 Shift Conversion and Cooling.	10
4.5 Acid Gas Treatment.	10
4.6 Methanation and Drying.	11
4.7 Auxiliary Facilities.	11
5. EFFLUENTS TO ATMOSPHERE.	14
5.1 Coal Preparation.	14
5.2 Gasification.	22
5.3 Quench and Dust Removal	23
5.4 Shift Conversion and Cooling.	23
5.5 Acid Gas Treatment.	24
5.6 Methanation and Drying.	25
5.7 Auxiliary Facilities.	25
6. EFFLUENTS - LIQUIDS AND SOLIDS	28
6.1 Coal Preparation.	28
6.2 Gasification.	29
6.3 Quench and Dust Removal	29
6.4 Shift Conversion and Cooling.	30
6.5 Acid Gas Treatment.	30
6.6 Methanation and Drying.	31
6.7 Auxiliary Facilities.	31
7. SULFUR BALANCE	34

TABLE OF CONTENTS (Cont'd)

	<u>Page</u>
8. THERMAL EFFICIENCY.	36
9. TRACE ELEMENTS.	38
10. TECHNOLOGY NEEDS.	41
11. PROCESS DETAILS	45
12. QUALIFICATIONS.	50
13. BIBLIOGRAPHY.	51

LIST OF TABLES

<u>No.</u>		<u>Page</u>
1	EFFLUENTS AND STREAMS FOR HYGAS PROCESS.	16
2	THERMAL EFFICIENCY HYGAS PROCESS	37
3	TRACE ELEMENTS - ESTIMATED VOLATILITY.	39
4	TECHNOLOGY NEEDS	42
5	COAL FEED AND PRODUCTS - HYGAS PLANT	46
6	STEAM BALANCE HYGAS PROCESS.	47
7	ELECTRIC POWER CONSUMPTION HYGAS PROCESS	48
8	WATER BALANCE HYGAS PROCESS.	49

LIST OF FIGURES

<u>No.</u>		<u>Page</u>
1	FLOW PLAN FOR HYGAS PROCESS.	7
2	HYGAS REACTOR.	9
3	EFFLUENTS AND STREAMS FOR HYGAS PROCESS.	15
4	SULFUR BALANCE - HYGAS PROCESS	35

1. SUMMARY

The HYGAS process being developed by the Institute of Gas Technology has been reviewed from the standpoint of its potential for affecting the environment. The quantities of solid, liquid and gaseous effluents have been estimated where possible, as well as the thermal efficiency of the process. For the purpose of reduced environmental impact, a number of possible process modifications or alternatives which could facilitate pollution control or increase thermal efficiency have been proposed, and new technology needs have been pointed out.

2. INTRODUCTION

Along with improved control of air and water pollution, the country is faced with urgent needs for energy sources. To improve the energy situation, intensive efforts are under way to upgrade coal, the most plentiful domestic fuel, to liquid and gaseous fuels which give less pollution. Other processes are intended to convert liquid fuels to gas. A few of the coal gasification processes are already commercially proven, and several others are being developed in large pilot plants. These programs are extensive and will cost millions of dollars, but this is warranted by the projected high cost for commercial gasification plants and the wide application expected in order to meet national needs. Coal conversion is faced with potential pollution problems that are common to coal-burning electric utility power plants in addition to pollution problems peculiar to the conversion process. It is thus important to examine the various conversion processes from the standpoint of pollution and thermal efficiencies and these should be compared with direct coal utilization when applicable. This type of examination is needed well before plans are initiated for commercial applications. Therefore, the Environmental Protection Agency arranged for such a study to be made by Exxon Research & Engineering Company under Contract No. EPA-68-02-0629, using all available non-proprietary information.

The present study under the contract involves preliminary design work to assure that conversion processes are free from pollution where pollution abatement techniques are available, to determine the overall efficiency of the processes and to point out areas where present technology and information are not available to assure that the processes are non-polluting.

All significant input streams to the processes must be defined, as well as all effluents and their compositions. This requires complete mass and energy balances to define all gas, liquid, and solid streams. With this information, facilities for control of pollution can be examined and modified as required to meet environmental objectives. Thermal efficiency is also calculated, since it indicates the amount of waste heat that must be rejected to ambient air and water and is related to the total pollution caused by the production of a given quantity of clean fuel. Alternatively, it is a way of estimating the amount of raw fuel resources that are consumed in making the relatively pollution-free fuel. At this time of energy shortage this is an important consideration. Suggestions are included concerning technology gaps that exist for techniques to control pollution or conserve energy. Maximum use was made of the literature and information available from developers. Visits with some of the developers were made, when it appeared warranted, to develop and up-date published information. Not included in this study are such areas as cost, economics, operability, etc. Coal mining and general offsite facilities are not within the scope of this study.

Other previous studies in this program to examine environmental aspects of fossil-fuel conversion processes covered various methods for gasifying coal to make synthetic natural gas or low Btu gas. Reports have been issued on the Koppers, Synthane, Lurgi, CO₂ Acceptor and BIGAS processes (1,2,3,4,5).

In the area of coal liquefaction, reports have been issued on the COED process of FMC (6) to make gas, tar, and char, as well as on the SRC process of Pittsburg & Midway Coal Mining Company to make a heavy liquid clean boiler fuel (7).

The present report covers our environmental evaluation of the HYGAS process to gasify coal and make synthetic pipeline gas, based to a large extent on information in reference 8, as part of a study made for EPA. This reference gives flow rates and compositions for oxygen-steam gasification, as well as total utilities for a complete plant. A breakdown of utilities requirements was not given and had to be estimated, as was the case with certain flow rates and compositions.

Our calculations included weight balances on individual elements, heat balances, and thermodynamic considerations. Some additional information on the process is given in other publications (9-18) and an engineering analysis for a commercial HYGAS plant has been projected (18), although these do not use the present route of gasifying with oxygen and steam. Information on the U-Gas System is also given in references 8, 13, 16 and 17. The U-Gas process is incorporated into the overall plant design in the present HYGAS study, but a separate report will be issued to cover it in more detail.

This particular design omits pretreating of coal to destroy caking properties, and has been used for the present study as suggested by the Institute of Gas Technology. Although pretreating is not required if the process is used on a non-caking coal, it should be emphasized that with caking coals pretreating may be necessary, in which case extensive additional facilities would have to be added beyond those considered in our present study. Also, pretreating generates a very large volume of raw gas that must be cleaned up and used, produces by-product tar liquids, and releases a large amount of heat (14).

3. SELECTION OF BASIS

In the HYGAS development, various alternatives have been considered for generating hydrogen or synthesis gas to use in the gasifier (9,11,15). One method is electrothermal, in which spent char is supplied by electrodes whereby an electric current flows through the fluidized bed of char. An alternative source of hydrogen is based on the steam-iron process, in which steam reacts with iron to form hydrogen and iron oxide. The latter is circulated to a separate vessel where it is regenerated using low Btu gas formed by reacting air with char. These are not the routes selected for the latest pilot plant design; however, the steam-iron route may be of interest in the longer range picture.

Attention is now focused on steam-oxygen gasification for the HYGAS process, together with methanation. The gasifier operates at 1,200 psig and the coal feed is pressurized by pumping it as an oil slurry, rather than using lock hoppers.

Coal-oil slurry at 1,200 psig or higher is dried by evaporation in a fluidized bed at 600°F and the coal then flows to a 1250°F bed, followed by one at 1750°F, and is finally gasified at 1900°F with steam oxygen. The feed coal passes through coking zones of increasing severity, consequently appreciable by-product liquids are formed. Most of the tar present in the raw gas is condensed in the 600°F drying zone and returned along with the coal feed back to the 1250°F zone, where the tar can be cracked to lighter liquid.

Except when a non-caking coal feed was used, pretreating of the coal feed to eliminate caking tendency was included in previous publications on the HYGAS process (9,11). Since caking coal would agglomerate in the fluid beds and cause plugging, pretreatment was considered to be necessary. The method of pretreating is to blow air through a fluid bed of coal particles at about 750-800°F, whereby much of the volatile matter is removed and some by-product tar is recovered (14). Oxidation destroys caking properties. It also releases a very large amount of heat which is recovered and used to make steam.

Pretreated coal must then be cooled in order to form a slurry for pumping at 400°F or less. The cooling step might be avoided if pretreating were carried out at gasifier pressure, but then the air for pretreating would have to be compressed, and a very large volume of air is required. Pretreating uses about 375 MM SCFD of air for a plant making 250 MM SCFD of synthetic natural gas (SNG). Off-gas from pretreating has a heating value of only 39 Btu per cubic foot but can be used as fuel, after clean-up (12). Tar yield from pretreating, if it were used, is estimated to be 630 tpd.

Looking into the future, it is the hope of IGT that modifications to the process can be made so that pretreating will no longer be necessary. Therefore, they recommended that pretreating be omitted from our study case and on this basis we have used a HYGAS design without coal pretreatment. It should be pointed out that if pretreatment is required, then the plant will look considerably different and will include large complex pretreatment facilities generating a large amount of heat, as well as a large volume of low Btu gas (e.g., 39 Btu per cf) which would have to be processed to remove tar, sulfur, and dust and consumed within the process.

To make the plant complete and self-sufficient, the necessary auxiliaries have been included, such as a sulfur plant, an oxygen plant, and all utilities. Clean low Btu fuel gas for the boiler furnace and for coal drying is manufactured using the IGT U-Gas process, based on steam-air gasification of coal. Since information on this system was incomplete, some of the flow rates and balances were calculated or estimated, in order to allow defining environmental controls and effects for the U-Gas operation.

4. PROCESS DESCRIPTION

The process makes 260 MM SCFD of pipeline gas (SNG) from Illinois No. 6 coal by gasifying it with medium Btu gas (mainly CO plus H₂ and steam) in a series of countercurrent fluidized zones. Residual char is then gasified with oxygen and steam in a bottom zone to provide gas for gasification in the upper zones. Carbon content of the rejected char may be 10-30 wt. %.

Raw gas is cleaned-up, shifted, and methanated. Operating pressure is sufficiently high so that compression of the product gas is avoided. The method of pressurizing coal feed involves slurrying it with light oil by-product, pumping to high pressure, and evaporating the slurry to dryness by direct contact with hot raw gas in a fluidized bed.

A block flow diagram of the processing steps is shown in Figure 1, together with major flow rates and operating conditions. The process can conveniently be sub-divided into a sequence of operations, each of which will be described in the following sub-sections of the report: (1) Coal Preparation, (2) Gasification, (3) Quench and Dust Removal, (4) Shift Conversion and Cooling, (5) Acid Gas Removal, (6) Methanation and (7) Auxiliary Facilities.

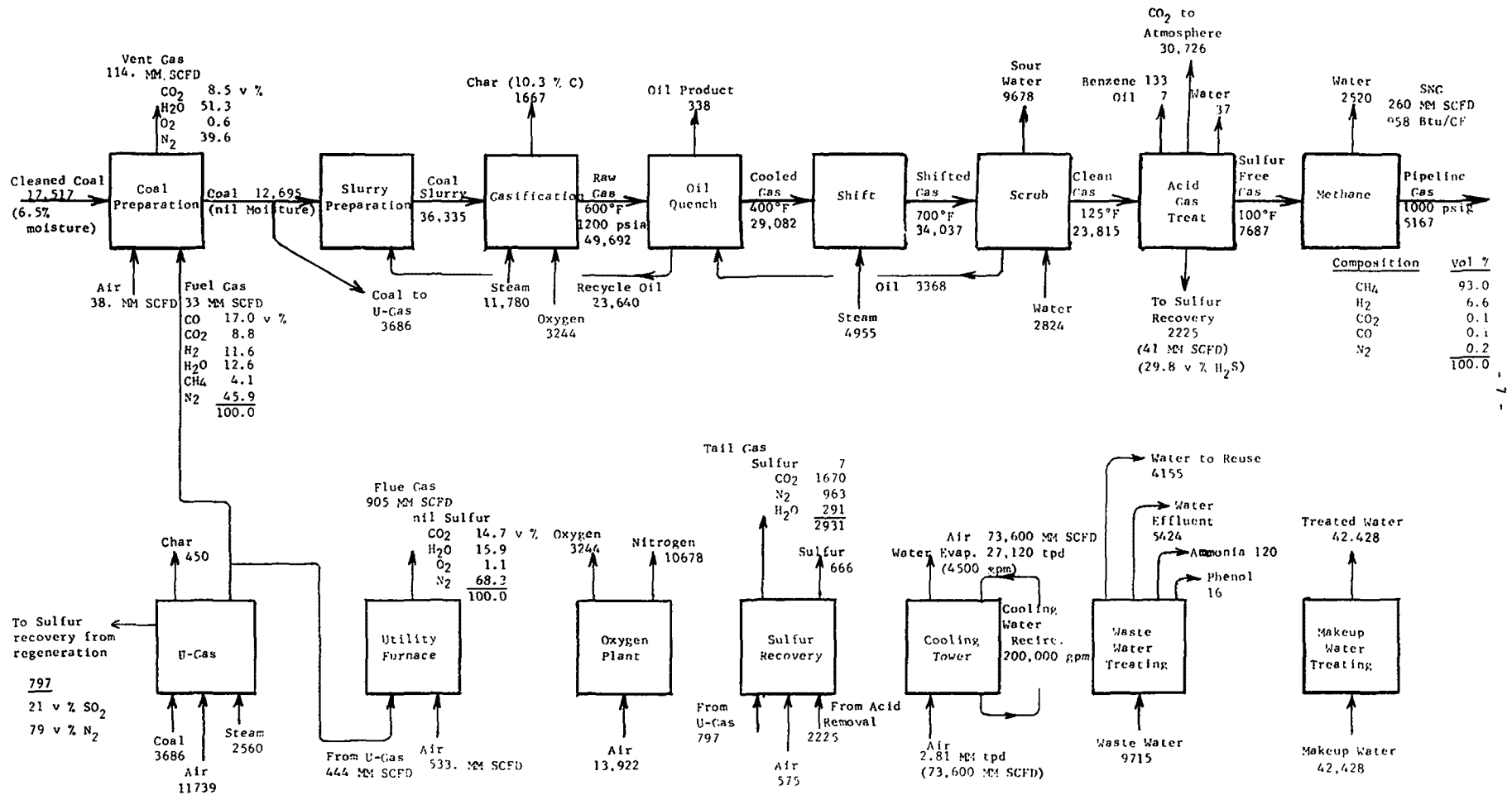
4.1 Coal Preparation

These facilities include storage and handling, crushing, and drying. It is assumed that cleaned coal is delivered, the separation of refuse and washing having been done at the mine or elsewhere with suitable disposal of waste and environmental controls. Coal feed, amounting to 17,517 tons/day (6.48% moisture), is received and 30 days storage is provided. Information on the coal feed is given in Table 1. Since the storage pile is very large, roughly 15 acres at 25 ft high, protection will be needed to control dust nuisance due to wind, while rain run off should be collected and cleaned up to supply makeup water for the plant.

Crushing is the next step in coal preparation, to reduce the coal feed to minus 8 mesh. Crushed coal is then dried to negligible moisture content in a fluid bed drier fired with part of the low Btu gas produced by the U-Gas system. The latter also supplies clean gas fuel for generating utilities, and consumes 22.5% of the total coal used by the plant.

Dried coal going to gasification is pressurized by mixing with oil to form a slurry which is pumped to about 1200 psia. Theoretical power for pumping is about 4500 horsepower. Oil is vaporized and recovered when the slurry is subsequently dried in an upper zone of the

FIGURE 1
FLOW PLAN FOR HYGAS PROCESS



Note: Numbers are tons/day except as noted

HYGAS reactor. Sufficient oil is thereby recycled to give a slurry containing 35% coal/65% oil, and cooling is provided so that temperature of the recycle oil is 400°F.

It should be emphasized again that this specific study case does not include pretreating to destroy caking properties of the coal feed.

4.2 Gasification

The HYGAS reactor has four zones, down through which the coal passes in series countercurrent to rising gas. These include an initial drying zone, followed by gasification zones at increasing temperature and severity. Figure 2 shows this arrangement (18). Slurry feed is dried in the upper bed at 600°F using heat in the raw gas. Vaporized oil is condensed and most of it is recycled to slurry preparation, but part of it is withdrawn as net product.

Dry coal then flows to the next bed at 1250°F where partial gasification occurs. Volatiles will be released from the coal at this temperature, including gas, oil, and tar. The oil can leave as vapor from the upper bed and be condensed for use in slurry preparation. However, heavy tar will condense in the upper bed and remain on the coal which is fed to the 1250°F bed. It will, therefore, tend to recycle between these two beds and build up until it is destroyed by cracking and coking in the 1250°F zone.

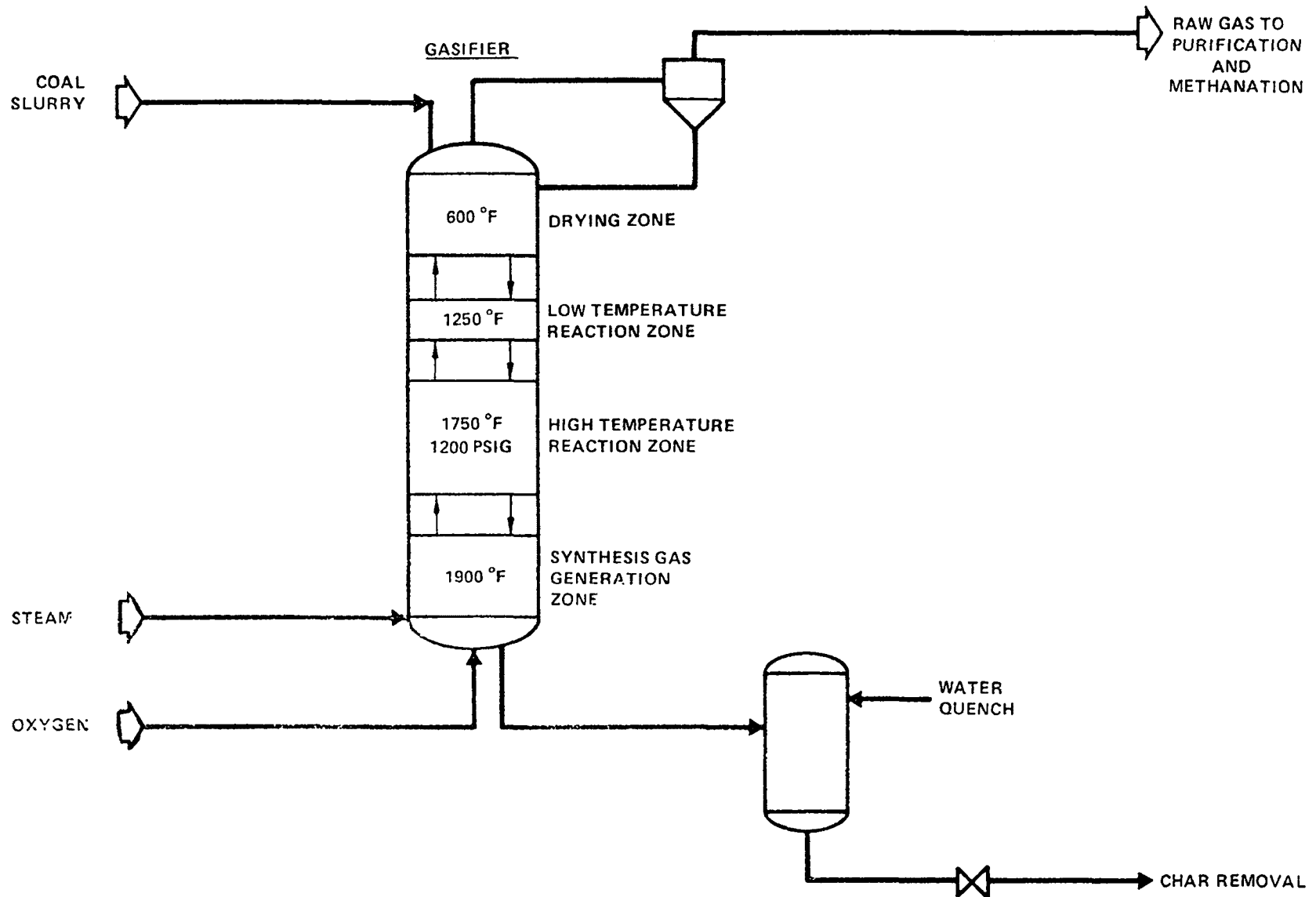
Char passes next to a bed at 1750°F, and then to the bottom zone where steam and oxygen are added for final gasification. Residual char rejected from this lower zone may contain 10-30% carbon, corresponding to 2-7% of the original carbon contained in the coal feed. The char is slurried in water, depressured, and discharged through lock hoppers.

The countercurrent contacting between gas and char provided by this multibed arrangement results in a considerable saving in oxygen. Of the total methane in the product, 58% is formed in the gasifier by the favorable effects of high pressure, temperature gradient, and the contribution from volatile matter in the coal feed.

4.3 Quench and Dust Removal

Raw gas leaving the upper drying bed of the gasifier at 600°F, is cooled to 400°F by contact with a recirculating oil stream, whereby most of the oil is condensed out and returned to slurry preparation. Temperature is maintained high enough to avoid condensing water which could cause emulsion problems; moreover, the steam is needed for the subsequent shift reaction. Heat removed in this cooling operation can be used to generate low pressure steam by recirculating the 400°F oil through waste heat boilers.

FIGURE 2
HYGAS REACTOR
(From Reference 18)



When the oil is condensed upon cooling, most of the dust in the raw gas leaving the drying bed will also be removed. Since the condensed oil is recycled and used for slurring coal feed, the fines will also be recycled and buildup in concentration, unless some provision is made to purge them from the system.

4.4 Shift Conversion and Cooling

The next step in gas handling is shift conversion, to react part of the CO with steam and thereby increase the H_2/CO ratio to 3/1 as needed for methanation. A sulfur resistant shift catalyst such as cobalt-molybdenum is used, and one-third of the raw gas bypasses the catalytic reactor. The catalyst is also exposed to oil vapors contained in the gas, and operates at about 700°F.

After shift conversion, the gas is cooled to condense most of the moisture, and at the same time remove ammonia, phenols, cyanides, and light oils, etc. This sour water is cleaned up for reuse by extraction and stripping, which operations will be described in Section 4.7 Auxiliary Facilities. The light oil condensed at this point is separated from the sour water and removed as a by-product, or it may be recycled to slurry preparation.

4.5 Acid Gas Treatment

At this point, the gas still contains various contaminants that must be removed, such as: H_2S , COS, CO_2 , and condensable hydrocarbons. The required cleanup is accomplished by scrubbing with refrigerated methanol, using the Rectisol process. Gases containing the sulfur compounds removed in the Rectisol unit are sent to a Claus plant for sulfur recovery. The Claus plant also provides incineration of COS and combustibles on this stream.

Most of the CO_2 is removed as a separate stream in the Rectisol regeneration, and indicated to be discharged to the atmosphere. However, this vent stream is shown as containing over 2.0 vol. % of combustibles, most of which is ethane; consequently, it will require further cleanup or incineration. While sulfur content is indicated to be low, nil H_2S and 300 ppm COS, other detailed evaluations of similar Rectisol applications show that additional controls will be needed--as a minimum, incineration, and possibly a modified processing scheme using a different type of sulfur plant (3,19,20,21).

It is not clear that any one simple process for acid gas treatment available today can simultaneously meet the targets of a highly concentrated stream to the sulfur plant, together with a CO_2 waste stream that is clean enough to discharge directly to the atmosphere, without further treatment such as sulfur cleanup or incineration. Therefore it appears that additional facilities will be needed, such as adsorption by molecular sieves or activated carbon to clean up the CO_2 vent stream.

A guard bed, for example of zinc oxide, is used to remove remaining traces of sulfur in the clean gas, so as to protect the methanation catalyst, which is extremely sensitive to sulfur poisoning. Reheating is needed since the guard bed operates at about 600°F, and can be provided by heat exchange with gas leaving the methanator. Such preheat is also needed to initiate the methanation reaction when this is carried out in a fixed bed of catalyst.

4.6 Methanation and Drying

Fixed bed catalytic reactors with conventional nickel base catalyst are used to react CO and H₂ to form methane and water. Operating temperature is 550-900°F. Outlet gas at 900°F is recycled to the inlet through waste heat boilers which generate steam, thereby recovering the large exothermic heat of reaction. Heat release amounts to 954 MM Btu/hr, which can generate about 1 million lb/hr of high pressure steam.

Water formed by the methanation reaction is condensed and recovered when the product gas is cooled, providing 200,000 lb/hr of clean condensate suitable for boiler feed water makeup. Final drying of the gas is effected by scrubbing with glycol, to meet pipeline specifications of 7 lb/MM SCF. The product specification of 0.10 vol. % CO maximum is met by providing effective control of methanation and excess hydrogen, leaving 6.5 vol. % hydrogen in the product gas. High heating value is then 960 Btu/CF.

4.7 Auxiliary Facilities

To make the plant complete and self-sufficient, various utilities and auxiliary facilities are needed in addition to the main gasification process. A Claus plant is used for sulfur recovery on a concentrated stream from acid gas removal, with tail gas cleanup by incineration followed by scrubbing with sulfite to remove SO₂, using the Wellman-Lord process (8). The Rectisol design basis provided shows 29.8 vol. % H₂S in the feed to the Claus plant, while at the same time the CO₂ vent gas contains no H₂S and 300 ppm of carbonyl sulfide. This would represent a very desirable high concentration of feed to the sulfur plant together with complete removal of H₂S from the CO₂ vent gas, although the latter contains an excessive amount of COS plus 2 vol. % combustibles, so it would require incineration.

Oxygen for gasification is supplied by a conventional air separation plant. While it does not generate contaminated waste streams, it is a large consumer of utilities, with a correspondingly large impact on thermal efficiency for the overall process.

Large amounts of steam and power are needed in the process. These are supplied by a utilities system fired with clean gas fuel manufactured by the U-Gas process being developed by The Institute of Gas Technology. This U-Gas process has been described in the literature (13,16,17).

In the U-Gas process, coal feed goes first to a pretreating reactor to destroy caking properties (14). Here it is contacted with air at 750-800°F in a fluid bed to give partial oxidation, accompanied by a decrease in volatiles. A very large amount of heat is released, which is used to generate steam. Hot char then goes to a second reactor where it is gasified with steam and air at 1800°F and 300 psia in a fluid bed. Off gas from pretreating, with a high heating value of only 39 Btu/CF, contains tar and sulfur, so it is mixed with hot gases from the gasifier in order to destroy the tar.

Sulfur removal is provided at high temperature by contacting the gas with a "molten metal", which is regenerated in a separate zone by reacting with air to form a concentrated SO₂ stream that is sent to the sulfur plant.

After further clean up by cooling to condense water and by scrubbing, the gas is used as clean fuel for coal drying, furnaces, and gas turbines.

A combined cycle system is used to maximize efficiency by first burning the high pressure fuel gas from the U-Gas unit for use in a gas turbine, and then discharging the hot exhaust to a boiler furnace which supplies process steam. Combined cycle systems may be a very effective way to supply by-product power for the oxygen plant compressors and for generating electricity.

Water treatment is an important part of the process. As in the Lurgi and Synthane gasification processes, considerable oil, phenols, etc., leave the HYGAS reactor and must be removed and disposed of in the gas cleanup section. A similar arrangement is used for this purpose, consisting primarily of a Rectisol unit for gas cleanup, and a Phenosolvan unit to remove phenols from the sour water. The latter effectively removes low molecular weight phenols as a by-product, but may be less satisfactory on higher molecular weight phenols. Treated water from Phenosolvan then passes to a sour water stripper which removes ammonia as a by-product, and H₂S which is sent to the sulfur plant. On the HYGAS process, details are not available for utilities used by the Rectisol and Phenosolvan units. Therefore, these were estimated by using information from the Lurgi plant design prepared for the El Paso project, which includes similar operations and processing units.

Other auxiliary facilities include treatment of makeup water, boiler feed water preparation, storage of by-product oil, phenol, ammonia, and sulfur, as well as ash disposal, and a cooling water circuit with cooling tower. While the original design showed no net water effluent

from the plant, this would lead to unacceptable buildup of dissolved solids, etc., in the cooling water circuit, since salts in the makeup water build up due to evaporation in the cooling tower and there is no way for them to leave the system. Therefore, we have added a nominal amount of discharge, and have increased the makeup water requirement accordingly.

Information is not available on sour water composition in the HYGAS process, but pilot plant data have been reported (22) for the Synthane process which may be comparable. Some results have also been reported for a commercial Koppers-Totzek plant (23). In the original HYGAS design, gas liquor from the Phenosolvan unit was processed for ammonia recovery and then sent directly to the cooling tower. However, it is estimated that this water may contain 100 ppm or more each of residual ammonia, phenols, and fatty acids, together with some H_2S left after sour water stripping. A similar situation in the design for the El Paso gasification project shows 100 ppm of free ammonia and 500 ppm of phenols in the treated water (3). These materials might be stripped out in the cooling tower, causing undesirable odors and contamination of the large volume of air flowing through.

Experience shows that part of the contaminants in the water can be removed by biological action in the cooling water circuit. This necessarily results in generation of cellular material, sludge, and algae, which foul the cooling tower and exchangers; consequently, additives are usually introduced to inhibit biological action. In order to avoid such complications, the design was modified to process the water in a biox unit before sending it to the cooling tower.

5. EFFLUENTS TO ATMOSPHERE

Environmental aspects of the process will now be discussed, together with possible control techniques in order to assure adequate pollution control. The various streams will be considered as shown in Figure 3, in the order of processing steps used in the section on Process Description. Table 1 shows the amounts and characteristics of all effluents from the process and auxiliary facilities.

5.1 Coal Preparation

A first consideration is the handling and storage of large amounts of coal feed. Delivered coal must be loaded on conveyors, with transfer to and from storage piles. Such operations necessarily tend to create problems due to noise, dust nuisance, and spills. These facilities should be enclosed as much as possible, with plans and equipment provided for cleanup. A dust collection system is desirable, operating at below atmospheric pressure to collect vent gas and pass it through bag filters. Storage piles are an additional concern since wind can disperse the fine particles. In some cases consideration has been given to covering the coal pile, or coating it with for example heavy tar. The pile is very large, over 500,000 tons for 30 days storage, requiring an area of about 10 acres. Coal piles are also liable to spontaneous combustion, calling for special attention and plans for control, together with provision for extinguishing fires if they occur (24). The obnoxious fumes, sulfur, and odor from this type of fire is well known. Previous reports in this series include further discussion of the general subject (e.g. 5) but for any specific project, a very careful and thorough evaluation and definition of facilities is needed.

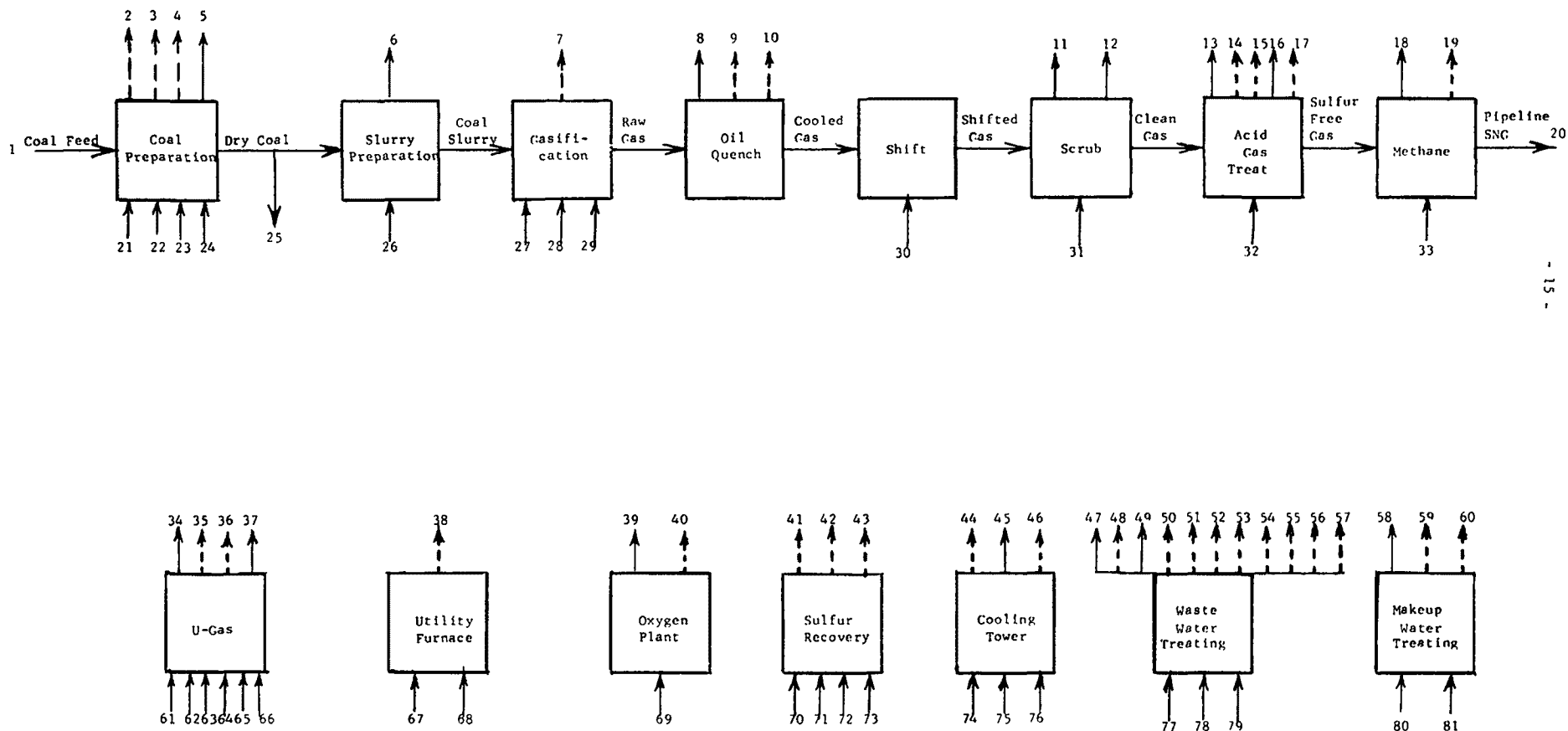
Noise control should be carefully considered since it is often a serious problem in solids handling and size reduction. If the grinding equipment is within a building, the process area may be shielded from undue noise but additional precautions are needed from the standpoint of personnel inside the building.

It should be noted that the present design is based on receiving cleaned coal, so that environmental considerations for the cleaning operation will be transferred to a different location. Coal cleaning and washing results in rejection of a large amount of refuse and fines, often 25% of the mined coal, with major environmental impacts as discussed in previous reports in this series.

Coal is crushed through 8 mesh and fed to a fluid bed dryer where essentially all moisture is removed. Since the fluid bed provides good contacting and temperature control, the heating gas can be introduced at a relatively high temperature without overheating coal particles and releasing volatiles. To maximize fuel efficiency, combustion should be with minimum excess air (e.g. 10%) and dryer offgas can be recycled to temper the hot gas to about 1000°F before it enters the fluid bed. Low excess air also decreases the volume of vent gas compared to some other drying systems that may use as much as 100% excess air in order to facilitate

FIGURE 3

EFFLUENTS AND STREAMS FOR HYGAS PROCESS



Note: Effluents released to the environment are shown by heavy dashed line; other streams are returned to the process. See Table 1 for details.

TABLE 1

EFFLUENTS AND STREAMS FOR HYGAS PROCESS

<u>Stream</u>	<u>Identification</u>	<u>Flow Rate Tons/Day</u>	<u>Comments</u>
1	Coal Feed	17,517	Total coal to plant - 6.5% moisture
*2	Wind	----	Wind can pick up and disperse fines from coal storage and handling.
*3	Rain	e.g. 6" in 24 hrs.	Rain will wash fines from coal preparation area and should be collected and sent to separate storm storage pond.
*4	Flue Gas	114 MM SCFD	Vent gas from coal dryer. Clean gas fuel is fired, but dust must be recovered, e.g., by bag filters, scrubbing, etc.
5	Coal Fines	e.g. 175	Fines recovered from dryer gas may be returned to U-Gas agglomerating reactor.
6	Flash Gas	----	Gas and vapors released when hot recycle oil is depressured and mixed with coal feed, must be recovered and returned to system.
*7	Char Slurry	1667 Char	Due to low density and open structure of char, as much as 16,000 tons/day of water may be needed to form a fluid slurry. Part of this water may be recovered by draining for reuse, but much of it may be retained in the char structure.
8	Quench Oil	23,640	Light oil recycled to slurry preparation.
*9	By-Product Oil	338	Net light oil by-product.
*10	Fine Solids	----	Purge of ash, coal fines, volatile trace elements, etc. that accumulate in oil quench recycle system and must be purged to prevent undue buildup.
11	Light Oil	3368	Oil vapors are condensed in scrubber, separated from water layer, and returned to quench system.
12	Sour Water	9678	Scrubber water containing compounds of sulfur, nitrogen, and oxygen. Processed in waste water cleanup section for reuse.
13	Sulfurous Gases	2225	From acid gas treatment - sent to sulfur plant. Based on 30% H ₂ S content.

TABLE 1 (Cont'd)

EFFLUENTS AND STREAMS FOR HYGAS PROCESS

<u>Stream</u>	<u>Identification</u>	<u>Flow Rate Tons/Day</u>	<u>Comments</u>
*14	CO ₂ Vent Gas	13,726	Must be discharged to the atmosphere but requires cleanup to remove combustibles by incineration or adsorption, etc.- could be treated by passing through utility furnace.
*15	By-Product Oil	140	Mainly benzene. Separated in Rectisol unit and removed as by-product.
16	Waste Water	37	Separated from gas in Rectisol operation and sent to waste water treating.
*17	Chemical Purge	----	Methanol consumption in Rectisol unit is estimated at 13 tons/day and definition is needed as to where it leaves.
18	Condensate	2520	Clean water produced by methanation reaction - used for boiler feed water.
*19	Water Purge	----	Removed in glycol dryer to meet SNG requirement.
20	Pipeline Gas	5167	Product SNG, 960 Btu/CF HHV. (260 MM SCFD).
21	Wind	----	Wind action on coal storage and handling area.
22	Rain	e.g. 6" in 24 hrs.	Rain onto storage pile can pick up acids, organics, fines, etc.
23	Gas Fuel	33 MM SCFD	Clean low Btu gas fired on coal dryer (from U-Gas).
24	Air	38 MM SCFD	Combustion air to coal dryer.
25	Coal	3686	Dry coal to U-Gas unit.
26	Oil	23,640	Quench oil recycled to slurry preparation.
27	Steam	11,780	High pressure steam to HYGAS reactor.
28	Oxygen	3244	Oxygen to HYGAS reactor.
29	Water	e.g. 16,000	Water used to quench and slurry spent char for depressuring and disposal. (See item 7).
30	Steam	4955	Added to shift reactor to convert CO to CO ₂ + H ₂ .

TABLE 1 (Cont'd)
EFFLUENTS AND STREAM FOR HYGAS PROCESS

<u>Stream</u>	<u>Identification</u>	<u>Flow Rate Tons/Day</u>	<u>Comments</u>
31	Water	2824	Recirculated water added to scrubber for dilution.
32	Chemicals	13	Methyl alcohol makeup added to Rectisol unit.
33	Glycol	----	Small amount of makeup to glycol dryer on product gas.
34	Fuel Gas	482 MM SCFD	Clean gas to supply plant fuel requirements. Made in U-Gas process by coal gasification with steam and air.
*35	Char Slurry	450 Char	Spent char from U-Gas unit is quenched and slurried in water for disposal. See item 7.
*36	Dust	----	Recovered from product gas on U-Gas unit, as required to meet gas turbine requirements and emission standards. May be returned to system. May contain some metal or chemical used to desulfurize raw gas.
37	SO ₂ Stream	797	From regeneration of sulfur acceptor on U-Gas unit. 21 vol. % SO ₂ and 79 vol. % N ₂ .
*38	Flue Gas	905 MM SCFD	Flue gas from utility furnace after combined cycle turbines. Should be low in sulfur and dust, but NO _x should be controlled.
39	Oxygen	3244	From oxygen plant - used in gasifier.
*40	Nitrogen	10678	Waste nitrogen to atmosphere. Should be clean.
*41	Tail Gas	2931	Waste gas from sulfur plant after tail gas cleanup by Wellman-Lord process.
*42	Sulfur	666	By-product sulfur from sulfur plant.
*43	Chemical Purge	----	Chemicals are used in sulfur plant. Sulfite scrubbing for tail gas cleanup requires purge containing sodium sulfate etc. May go to waste water treating.

TABLE 1 (Cont'd)

EFFLUENTS AND STREAMS FOR HYGAS PROCESS

<u>Stream</u>	<u>Identification</u>	<u>Flow Rate Tons/Day</u>	<u>Comments</u>
*44	Air	73,600 MM SCFD	Air from cooling tower containing evaporated water (27,120 tons/day). Will also carry mist of water drops which may amount to 1200 tons/day.
45	Cooling Water	200,000 gpm	Water from cooling tower recirculated to process heat exchangers.
*46	Chemical	----	Chemicals are used in cooling water circuit to control corrosion, algae, etc. and must appear in the effluent.
47	Treated Water	4155	Water for reuse after treatment.
*48	Net Water Discharge	5424	Water effluent from plant to reject soluble salts. Disposal of this stream may present problems and more definitive information on composition is needed.
49	H ₂ S Stream	----	Gases from sour water stripper sent to sulfur plant for incineration and recovery.
*50	By-Product Ammonia	120	Recovered from sour water using Phosam process.
*51	Phenol	16	By-product recovered from sour water using Phenosolvan process.
*52	Oil	----	Recovered in oil separator on waste water treating system.
*53	Sludge	20-60	Cellular material from biox reactions. Should be incinerated to avoid odor problems.
*54	Chemicals	----	Used in Phosam process. Purge streams must be defined so that disposal can be specified.
*55	Trace Elements	See Text Section 9	Volatile trace element will accumulate in cleanup system and must be deactivated and disposed of.
*56	Water Evaporation	See item 7	Evaporation from pond where char slurry is sent for draining and drying. May be odor problem.
*57	Spent Char	2117 dry	Combined drained char from HYGAS and U-Gas units. Will also contain moisture.

TABLE 1 (Cont'd)

EFFLUENTS AND STREAMS FOR HYGAS PROCESS

<u>Stream</u>	<u>Identification</u>	<u>Flow Rate Tons/Day</u>	<u>Comments</u>
58	Treated Water	42,428	Make up water to plant.
*59	Sludge	e.g. 10-20	From water treating using lime, alum, etc.
*60	Chemical Wastes	----	From water treating, including acid and caustic use in boiler feed water demineralization.
61	Coal	3286	Feed to U-Gas unit.
62	Air	11,739	Air for gasification in U-Gas unit.
63	Steam	2560	To U-Gas unit.
64	Air	651	Used to regenerate sulfur acceptor that removes sulfur from raw gas on U-Gas unit. (Gas is sent to Claus unit for sulfur recovery.)
65	Quench Water	ca. 4000	Used to quench and slurry spent char for transport to settling pond. See item 7.
66	Sulfur Acceptor	----	Makeup metal or chemical used to remove sulfur from raw gas.
67	Fuel Gas	444 MM SCFD	Clean fuel gas to utility boiler.
68	Air	533 MM SCFD	Combustion air to utility boiler.
69	Air	13,922	Air used to make oxygen.
70	H ₂ S Stream	2,225	From Rectisol unit for acid gas treatment of raw gas.
71	SO ₂ Stream	797	From air-regeneration of sulfur acceptor in U-Gas unit.
72	Air	575	Supplemental air needed to complete Claus reaction.
73	Chemicals	----	Sodium sulfite tec. used in Wellman-Lord unit for fuel gas cleanup.
74	Air	73,600 MM SCFD (2.8 MM Tons/Day)	Air to cooling tower.
75	Water	200,000 gpm	Cooling water circulated through cooling tower.
76	Chemicals	----	Treating agents used in cooling water circuit, e.g. chlorine to control algae and chromate to control corrosion.

TABLE 1 (Cont'd)

EFFLUENTS AND STREAMS FOR HYGAS PROCESS

<u>Stream</u>	<u>Identification</u>	<u>Flow Rate Tons/Day</u>	<u>Comments</u>
77	Waste Water	9715	Sour water from gas cleanup system to be processed for reuse.
78	Chemical	----	Makeup to phenol extraction system (Phenosolvan). May be isobutyl ether.
79	Chemical	----	Makeup to ammonia recovery unit (Phosam) e.g., phosphate.
80	Water	42,428	Makeup water to plant.
81	Chemicals	----	Used for treating makeup water. Includes lime, alum, ion exchange resin, sulfuric acid, and caustic.

* These streams are emitted to the environment, others are returned to the process.

drying. Low excess air results in higher moisture content of the gas, but this disadvantage is more than offset by decreases in fuel consumption and volume of vent gas to be cleaned up. Volume of dryer vent gas is 114. MM SCFD, or about half as much as the SNG product. Moisture content is 51 vol. %. Although the drying system is not described in the references, the fluid bed can be operated with a coal temperature of 250-300°F to give adequate drying, but maximum allowable coal preheat is limited by the need for forming a slurry at 400°F for pumping.

About 77% of the dried coal goes to gasification, while the remainder is used to supply plant fuel requirement after being processed in a U-Gas unit which converts the coal to clean low Btu gas fuel. Part of this gas is used as fuel for coal drying, consequently the dryer vent gas does not require sulfur removal, although dust cleanup is needed and can be provided by bag filters, scrubbing, or electrostatic precipitators. Odor of the dryer vent gas may be a problem, particularly if the operation is on lignite or a reactive coal. This question needs to be answered in the pilot plant program.

The next step in this section of the process is to mix recycle oil with the coal feed for pumping into the high pressure gasifier. Any flash gas released during slurry formation must be recovered and used, or incinerated. Slurry concentration is about 35 wt.% coal/65 wt.% oil, and reciprocating pumps are indicated to raise the pressure to 1200 psia.

5.2 Gasification

A series of complex operations are involved in gasification, including handling of slurry and hot fluidized solids at very high temperatures. Careful consideration should be given to potential problems due to leaks, spills, upsets, etc., as well as scheduled shutdown and maintenance. The major effluent from gasification during normal operation is the residual char, containing ash that comes in with the coal feed.

This char, containing 10-30% carbon, is quenched with water, depressured by lock hoppers, and sent to an ash settling pond. Steam formed by this quenching operation may contain particulates and other contaminants, consequently it should be returned to the process or collected for disposal. The amount of steam is approximately 50,000 lb/hr. In the operation as intended, there should be no serious emissions to the atmosphere; however, the system may be difficult to operate and maintain due to plugging or erosion of valves, and any failures or upsets could cause serious environmental emissions.

Ash in the water slurry is recovered in a settling pond, which is drained so that semi-dry ash can be removed for burial. Although wet ash is not dusty, parts of the settling basin or spills on the ground can dry out and become a dust nuisance, as has happened in the past.

The nature of this ash or char warrants further discussion. If the original coal particles maintain their size during gasification, then their density will decrease as the carbonaceous content falls to the indicated 10.3% carbon. On this basis, a char density of only 11.24 lb/cu. ft.

is calculated for a coal feed of 1.40 specific gravity and 11.54 wt.% ash. Obviously, the particles will not only be light, but friable as well. Therefore, additional information should be obtained on the depressuring, handling, and disposal operations in order to assure that problems are satisfactorily resolved.

If the char particles break up, then very fine dust may result, with complications in the ash handling and disposal. Alternative approaches to this problem have used sintering of fly ash or an agglomerating fluid bed system (25,26), or a slagging gasifier (1,5).

Attrition of particles in the HYGAS reactor will generate fines that are carried up with the gas stream. These fines will probably be removed rather completely by the oil quench system, thus tending to build up in concentration in the recycle oil unless they are removed by agglomeration, filtration, or other means. Similarly, volatile trace elements such as arsenic, lead, and cadmium will accumulate in the recycle oil, requiring separation as will be discussed in the section on trace elements.

5.3 Quench and Dust Removal

Gas from the gasifier, together with slurry oil evaporated in the drying zone is quenched to 400°F by direct contact with recirculated oil, the heat being used to generate steam in waste heat boilers. Most of the oil vapor in the entering gas is condensed and recycled to slurry preparation, while at the same time particulates and condensibles such as certain trace elements will be removed from the gas and accumulate in the oil. While most of the oil is recycled, part of it must be withdrawn as product and can be expected to contain toxic elements such as arsenic, lead, and cadmium, as well as particulates, phenols, sulfur and nitrogen compounds, etc. Therefore properties of this oil and its projected use need further evaluation to define what treatment may be required to make it suitable for use as fuel, or as a raw material for refining.

Due to safety considerations and the possibility of emulsions, quench temperature is maintained above the water dew point. Steam will not condense unless a high concentration of steam inadvertently occurs, as for example during startup or upsets.

5.4 Shift Conversion and Cooling

The next step in the gas processing sequence is to shift the CO by reaction with steam to make hydrogen. This catalytic operation may also give some hydrogenation to help remove olefins, cyanides, and oil vapor. It may also be possible to modify the shift reactor so as to hydrolyze carbonyl sulfide and other compounds to form H₂S which is removed more easily in acid gas treatment. Some deposition of trace elements and coke is expected on the shift catalyst, consequently it should be reprocessed periodically or properly disposed of.

In the scrubbing operation to remove dust, the gas is cooled to 125°F, condensing out most of the water vapor remaining in the shifted gas. Ammonia, cyanides, phenols, oil, etc., will be present in the sour water, similar to the cases of Lurgi (3) and Synthane (2), in fact the

processing sequence is quite similar to proposed plants using Lurgi technology (3,19,20). The major effluent from this area is the gas liquor, handling of which will be discussed in Section 6.4, but it should be noted that when this water is depressured, gases are released that must be recovered or incinerated. In addition, sour water stripping produces by-product ammonia, and H_2S which can be sent to sulfur recovery.

5.5 Acid Gas Treatment

In this section of the process, the bulk of contaminants remaining in the gas are removed. Major constituents are acid gases, H_2S (1.44 vol. %) and CO_2 (30.85 vol. %), while minor contaminants include HCN, ammonia, light hydrocarbons, naphtha, etc. Ideally, acid gas treatment should remove all contaminants to a low level, while giving both a concentrated sulfur containing stream to the sulfur plant together with a CO_2 waste stream that is sufficiently pure so that it can be vented directly to the atmosphere without further treatment.

The IGT design uses a Lurgi Rectisol system for acid gas treatment, based on scrubbing with refrigerated methanol. The design shows 30 vol. % concentration of sulfur compounds in the gas fed to the Claus Plant, which represents a desirably high concentration to allow efficient sulfur recovery. Other Rectisol designs for commercial projects (19) show much lower concentrations, less than 2 vol. %. These designs remove H_2S and CO_2 together, resulting in a dilute H_2S stream that is not suitable for a conventional Claus plant. It is understood that the Rectisol process can be designed to remove the H_2S as a separate stream at high concentration, although the utilities consumption may be increased. In any detailed specific evaluation, special attention must be given to design basis to assure that the process efficiency used is consistent with costs and economics, utilities consumption, and environmental effects.

The CO_2 rejected to the atmosphere is a very large stream, the tons/day exceeding total coal used by the plant. Therefore, it must be particularly free of undesirable contaminants. Unfortunately, methanol scrubbing is indicated to give about 2% combustibles in this CO_2 waste stream, including 1.46% ethane, and 300 ppm carbon monoxide, and 300 ppm COS. Other related information (3, 19, 20) confirms that the CO_2 waste will require further cleanup, possibly with incineration. Sulfur content at 300 ppm is moderate, amounting to less than 1% of sulfur in the coal to gasification, and would appear acceptable in some cases depending on standards that apply for a specific location, at least if it is in a less objectionable form such as SO_2 .

Combustible content is more of a problem. Heating value in the CO_2 vent gas is about 270 MM Btu/hr., or 2% of the heating value in the coal to gasification, so it should not be wasted. On the other hand, extraneous fuel, or effective preheat, would be needed to maintain a minimum incineration temperature of say 1500°F, which corresponds to a heat load of 440 MM Btu/hr.

Other methods of acid gas removal, such as scrubbing with amine or hot carbonate, also have difficulty in providing a CO_2 purge stream that is clean enough to vent directly to the atmosphere. Further work on this problem would be very desirable, for example to develop a simple inexpensive way to clean up the CO_2 vent stream. Contaminants to remove include combustibles, carbon monoxide, plus carbonyl sulfide and other sulfur compounds.

One possible approach is to pass the CO_2 stream through the combustion zone of a furnace, such as the utility boiler. This would provide incineration of combustibles with recovery of useful heat. If the boiler already had stack gas cleanup, sulfur emissions would also then be controlled, so that requirements in acid gas treatment would be less stringent. A further advantage is that combustion temperature would be decreased by the added CO_2 stream, thereby decreasing NO_x formation. A disadvantage however is that the CO_2 stream is large, increasing the total volume of flue gas by about one-third. Assurance is needed that the CO_2 vent stream will be clean, so further study and evaluation of alternatives is called for.

While the major gas emissions have been discussed, it should be recognized that there can be other effects associated with depressuring or handling other streams in acid gas treatment, such as separated water, oil, etc., or waste chemicals that may be discharged. Also, utilities consumption for acid gas treatment is large; it is often the largest single consumer in the plant.

5.6 Methanation and Drying

As covered by the process description in Section 4.6, methanation and drying is carried out in a closed system, with no streams normally emitted to the atmosphere. The large heat release is used to make steam, generally by recirculating reactor outlet gas through waste heat boilers. Careful attention is required in design and operation to control leaks from this system. In addition, gas released when depressuring water produced, or when depressuring equipment for maintenance should be collected and recovered or incinerated.

5.7 Auxiliary Facilities

The complete plant includes auxiliary facilities one of which is a sulfur plant to make by-product sulfur from sulfur compounds removed in various cleanup operations. A Claus plant is used for this purpose, with tail gas cleanup by sulfite scrubbing using the Wellman-Lord process.

Feed to the Claus plant is mainly an H_2S stream from acid gas treatment, plus an SO_2 stream from the U-Gas unit producing clean plant fuel. The former contains 30 vol. % sulfur compounds (nearly all H_2S), while the latter has 21% SO_2 plus 79% N_2 from regenerating the molten metal used to desulfurize the fuel gas. Some additional SO_2 comes from the tail gas cleanup system.

These feed streams are combined and reacted with additional air to form sulfur which is recovered. Tail gas cleanup is specified to give a total sulfur of 250 ppm as SO_2 in the gas released to the atmosphere, which should be satisfactory for most plant locations. There are no other primary emissions from the sulfur plant, but special precautions should be taken to control leaks, vents from sulfur storage, etc., and to avoid offensive odors. H_2S is appreciably soluble in molten sulfur, but there are well established techniques for control.

Oxygen production is not expected to cause undesirable emissions to the air, the main effluent being waste nitrogen. However, being a large energy consumer, it affects the size of the utilities system and the amount of waste heat to be dissipated to the environment.

A major concern on emissions to the air is from the system used to provide plant utilities. The utility boiler is an important factor, discharging a volume of flue gas 3.5 times the volume of SNG product. In this case it is expected to be free of particulates and very low in sulfur, since clean gas fuel is supplied from the U-Gas system. There is still the question of NO_x formation, although this will tend to be lowered by the fact that low Btu fuel is used (151 Btu/scf), thereby giving a relatively low flame temperature. The exact amount will depend on the furnace design, use of staged combustion, etc., and should be defined for specific process applications to be sure that applicable standards are met.

A large part of the fuel gas will be used first to drive gas turbines as part of the "combined cycle" operation. Since this improves efficiency, it should tend to reduce plant emissions.

Clean low Btu gas for plant fuel is provided by the U-Gas unit which gasifies high sulfur coal using air and steam. Required cleanup facilities are included in this section to remove sulfur, tar, dust, and other pollutants. The large size of this operation makes it especially important in evaluating environmental impacts. The primary gas product is contained and treated within the system, without specific emissions to the atmosphere. However, contaminants are withdrawn as a liquid effluent containing water, sulfur, nitrogen, and oxygen compounds, as well as oil, tar, and particulates. This stream must be contained and handled in a way to avoid undesirable emissions or odors. In addition, spent char which is withdrawn from the U-Gas unit could cause a dust nuisance unless proper precautions are taken in handling and disposal. It is presumably similar to char from the main gasifier, so a common disposal system might be used.

In addition, there is a drift loss due to mist carried out by the air. A typical estimate of this would be about 200,000 lb/hr, although it could be reduced considerably by using some of the new techniques that are being developed to control drift loss from cooling towers (27). Drift can cause deposits in the nearby area due to dissolved solids in the cooling water. Careful consideration should also be given to the potential fog problem or plume associated with cooling towers due to condensation under unfavorable atmospheric conditions. One way to avoid the plume is to provide reheat on the air leaving the cooling tower, but this will not normally be warranted. It may be that these problems can be taken care of by proper design and placement of the cooling tower.

Normally, there will not be contaminants introduced into the cooling water circuit that might be stripped out by the air flowing through the cooling tower. However, experience has shown that leaks can be expected

in exchangers used in cooling water service, especially at high pressures such as the 1000 psig in this process. Leaks, for example, in exchangers on sour water service could introduce sulfur, cyanide, and ammonia into the cooling water, which would then be stripped out into the air. Special precautions and possibly monitoring equipment may be needed from this standpoint.

In the areas related to use of water, by far the largest effluent to the atmosphere is from the cooling tower. Flow of air through the cooling tower is 74,000 MM SCFD, or nearly 300 times the volume of product SNG. The volume of air passing through the cooling tower is so large that every precaution should be taken to see that it does not inadvertently become contaminated.

6. EFFLUENTS - LIQUIDS AND SOLIDS

As in the preceeding section, effluents will be discussed in the order in which they appear on the Figure 1 flowplan. Individual streams are all identified on Figure 3 and described in Table 1.

6.1 Coal Preparation

This particular design assumes that the coal has been cleaned before delivery, consequently the rejection of rock, gangue, or tailings does not appear on the flowsheet. In applications where coal cleaning must be provided at the plant, a considerable amount of refuse will have to be disposed of. Similar designs for other processes (5) show for example 20% refuse on delivered run of mine coal, equivalent to over 800 acre-ft/yr to dispose of. Moreover, coal washing requires a large volume of water which must be sent to a tailing pond, cleaned up, and reused. Leaching from solids, for example, by rain and seepage from ponds are potential problem areas.

A further consideration on the coal preparation area is with regard to the coal storage pile. The design includes storage for 30 days minimum, or about 500,000 tons; so the coal storage pile will cover a very large area. Rain run off can lead to undesirable effluents. A large part of the rain can run off quickly and carry suspended particles, while the remainder will have a long contact time with the coal and can pick up metals, acids, and organics. Therefore, rain run off from the storage area should be collected in storm sewers and sent to a separate storm pond. With a certain amount of treatment, this water can then be used as make-up for the process. Control of seepage may be desirable on the pond, and particularly on the coal storage area, using for example a layer of concrete, plastic or clay.

Coal drying can also contribute effluents. The drying gas will pick up coal fines, which should be recovered by filtering, scrubbing, or electrostatic precipitation for reuse in the process. Fines are undesirable in the coal feed to HYGAS in that they readily blow out of the initial drying bed and accumulate in the oil recycled to slurry preparation. One possible use for the coal fines is as fuel in the coal dryer, to the extent permitted by sulfur emission. Using some fines as fuel in the utility furnace might be satisfactory, although fly ash recovery may have to be added. Perhaps a better use for the fines is to gasify them in the U-Gas system so that all fine ash is recovered. Adding the fines as a separate stream to the oxygen gasifier at the bottom of the HYGAS reactor would also consume them, while assuring that all ash is recovered.

In slurry preparation, coal from drying is mixed with hot recycle oil. The latter must be depressured from over 1000 psig, which will no doubt release vapors. These should be condensed and returned to the system. Similarly, any residual moisture in the coal that flashes during slurry preparation should be collected and returned to the process.

6.2 Gasification

The major effluent from this area of the plant is spent char, which serves to reject ash brought in with the coal feed. Some unreacted carbon is also rejected in the char. Hot char from the gasifier is handled by quenching in water, forming steam which is presumably returned to the gasifier, and a water slurry (25% solids) which is depressured across an oil field type choke (28). The slurry goes to a settling pond from which water is recycled to the quench system. At intervals, the pond is drained so that wet ash can be reclaimed for ultimate disposal offsite. There is no water effluent from the ash system other than that retained by the ash, but this may contain soluble salts or trace elements so further information should be obtained on leachables from the wet ash. Exposure to air may be a factor, and tests are needed to define to what extent leaching by rain or ground water may be a problem when the char is disposed of by burial or as fill. Potential leaching of calcium chloride, magnesium sulfate, compounds of iron, manganese, fluorine, etc., are of concern.

It has been indicated that the char resembles activated carbon in that it has adsorptive properties, and will remove phenol from waste water. If used in this manner, the char could be regenerated, for example, by returning it to the gasifier. In some cases it may be preferable to discard without regenerating spent char used in waste water treating, in which case additional assurance is needed that adsorbed materials will not find their way into natural waters and cause problems.

Char from the gasifier is expected to have a low density and to be friable. Severe turbulence associated with depressuring the slurry may create very fine particles. Therefore, careful consideration is needed of potential problems due to particulates in drainage water, in addition to the potential leaching problem mentioned earlier. When the wet ash dries out, dusting could also be a nuisance and requires evaluation, since this has sometimes been a problem.

6.3 Quench and Dust Removal

In this section of the process, raw gas is cooled by direct contact with product oil which is recirculated and cooled. At the same time, particulates in the raw gas will be removed. While most of the oil is used to slurry the coal feed, a stream of by-product oil is also withdrawn corresponding to the net yield of oil from the coal gasification reaction. This by-product oil, amounting to 338 tons/day is the only major effluent from this section of the process. One possibility is to use it for fuel, but considerably more information would be needed to determine whether it can be burned directly, or whether it will first need further treatments to remove contaminants.

As discussed in Section 4.3, the oil will pick up fines from the gas, and if ash in the oil is over 0.2%, then it would exceed the 0.1 lb. of particulates per MM Btu specified for large stationery boilers. Ash content may well be excessive since this is the only purge of fines that are carried out in the raw gas leaving the drying bed of the gasifier. An ash

content of 0.2% in the by-product oil would correspond to only .05 wt. % of the ash in the coal fed to gasification, so to the extent that net entrainment of coal fines exceeds this value, ash removal from the by-product oil would be needed.

A further concern is the amount of other contaminants picked up in the quench oil. It is known that many trace elements such as Hg, As, Cd, Sb, etc., are partially volatile at gasification conditions. When the gas is cooled, some of them may drop out, and accumulate in the gas handling system, for example, in the quench oil. Some plant data on coal gasification has shown as much as 30-50 ppm each of arsenic and lead in oil or tar by-products, raising major questions regarding subsequent use and disposal of such materials. The subject is discussed further in Section 9 dealing with trace elements.

6.4 Shift Conversion and Cooling

Shift conversion does not involve primary emissions or effluents, although some trace elements and tarry materials may accumulate on the fixed bed of catalyst used in this operation. However, subsequent cooling and scrubbing of the gas condenses a large amount of sour water which must be cleaned up and reused or disposed of. Some oil is also condensed and it is returned to the oil quench system, after separation from the water layer. If there is residual dust in the gas leaving the oil quench system, it will also be removed in the scrubber.

As in other gasification processes previously evaluated for environmental aspects, cleanup of the water layer, commonly called gas liquor, is a formidable challenge. At this point in time, not enough information is available on the HYGAS process to define the kinds or amounts of contaminants in the condensate from scrubbing. It is known that various sulfur compounds including H_2S will be present, as well as nitrogen compounds such as ammonia, cyanides, etc., oxygen compounds such as phenols and fatty acids, together with thiocyanates, chlorides, and other products of interaction. In addition some of the volatile trace elements will also appear in the sour water, particularly chlorine and fluorine, although their chemical form is uncertain.

It is apparent that a great deal of additional information needs to be obtained in pilot plant operations to define the problem adequately so that effective measures for environmental controls can be specified. Phenols can be largely separated by extraction (e.g. Phenosolvan process), while sour water stripping will remove NH_3 and H_2S for recovery. Biological oxidation (biox) may then be used for further cleanup of waste water, followed by filtration, activated carbon, etc., as required.

6.5 Acid Gas Treatment

The primary liquid effluent from acid gas treatment is naphtha and oil which is recovered from the gas by scrubbing with refrigerated methanol. This oil is returned to the oil quench system, and eventually withdrawn as a by-product. As a result of the high cracking severity that it has been exposed to in the gasification reactor, it will consist mainly of aromatics such as benzene and should be useful as a raw material for making chemicals or motor gasoline. Benzene is toxic, so proper precautions are needed in its handling and storage.

There is also a small amount of water rejected from acid gas treatment, which can be combined with sour water from scrubbing for treatment. Methanol makeup is needed on the Rectisol unit, a typical figure being .03 lb/MSCF feed gas, which corresponds to 13 tons/day of methanol. It is not specified as to where this leaves the system, but it probably is mostly in the rejected water, and can be consumed in the biox operation. There should be no significant solid effluents from acid gas treatment, since char and ash particles will be removed efficiently in the scrubber. The large effluents of gases were covered in Section 5.5.

6.6 Methanation and Drying

The methanation reaction also produces a large amount of water, which is condensed and used for makeup to steam boilers. The amount, 2520 tons/day, is large relative to the net waste water effluent of 5424 tons/day and thus makes an important contribution in the overall water balance. It is very clean condensate, free of sulfur and dissolved solids, so little or no treating is required on it. When it is condensed at high pressure, some methane will be dissolved in it and may have to be removed. If the water is depressured, most of the methane will be released and should be recovered and returned to the system, or incinerated.

6.7 Auxiliary Facilities

One of the auxiliary facilities is a Claus plant for sulfur recovery. The sulfur by-product may be handled as either a solid or a liquid, and methods are well established for handling it in a clean safe manner, for example with regard to odors, dust, H_2S release from molten sulfur, etc. No other solid or liquid effluents are expected from the Claus plant itself, except for catalyst which is replaced periodically. There is also a separate system for tail gas cleanup using the Wellman-Lord process based on scrubbing with a sodium sulfite solution. Since some of the sulfite is oxidized to sulfate, a purge stream of the solution is withdrawn and must be disposed of. If 10% of the sulfur in the Claus plant feed were to leave as sodium sulfate, the latter would amount to 300 tons/day of salt. In addition, there may be sodium sulfite present, particularly if purge represents part of the circulating solution. If only sodium sulfate is purged, it might be sold, otherwise there can be a sizeable disposal problem.

On the oxygen plant there are no major liquid or solid effluents, although a small amount of water may be condensed from the entering air and recovered for use as boiler feed water.

The utility furnace burns clean gas fuel and the major effluents are gases, with no ash or slag. There will be water blowdown from the boilers, but this can be added to the cooling water to provide makeup. It is assumed that char from the HYGAS and U-Gas reactors will be low enough in carbon content so that it can be discarded. If it were necessary to burn it to recover heating value, then recovery of particulates would be needed on that operation. The indicated carbon content of 10.3% on char corresponds to about 1.5% of the heating value in the coal feed, while if the carbon content were 30% it would then represent 5.7% of the heating value in the coal feed and recovery of this would be desirable.

Clean gas fuel for the utility boiler and the coal dryer is supplied by a U-Gas unit, which includes scrubbing to remove particulates and acid gas treatment to remove sulfur. A major liquid effluent from U-Gas is sour water from scrubbing to remove particulates. It will be similar to the sour water from HYGAS and contain a variety of sulfur, nitrogen and oxygen compounds. Particulates could be removed by filtration and settling, and the sour water might then be treated along with sour water from gasification. There may also be tar and oil to remove in the U-Gas cleanup system, since the operation uses pretreatment of the coal feed to destroy caking properties. Pretreating generates considerable tar, perhaps 4 wt. % on coal feed. Most of this will be destroyed in the U-Gas design which provides for passing the pretreater off-gas through the gasification reactor where it is held at about 1500°F for 10-15 seconds to destroy tar (16). Even so, some oil or benzene may appear in the gas cleanup system, especially during startup or upsets.

Another effluent from the U-Gas unit is spent char or ash. As discussed earlier in this subsection, it is important that the carbon content be low enough so that it does not result in a significant loss of the heating value in the coal feed. A carbon content of 10% or less would be desirable from this viewpoint, but may cause operability problems due to low particle density, or due to disintegration of the ash.

The cooling water circuit has a very important impact on the water effluent from the plant. The large amount of evaporation serves to concentrate dissolved solids in the cooling water circuit. Moreover, chemicals are added to the cooling water system, such as chromates to inhibit corrosion of exchangers and equipment, chlorine to suppress algae growth, or other additives. These chemicals appear in the water blow-down from the cooling tower, and when they go the biox system can interfere with its operation. Biological processes are often inhibited by less than 1 ppm of chromium or copper, for example. In general, water blowdown will not be a direct effluent to the environment, but rather will be processed first through waste water treatment.

The major stream to waste water treating is gas liquor from cooling and scrubbing the raw gas, after extraction of phenols and after sour water stripping. Residual amounts of phenols, H_2S , NH_3 etc. are then further decreased by biological oxidation.

Operability considerations make it necessary to have a net discharge of waste water from the process in order to purge dissolved solids. Some of these enter in the makeup water as sodium salts while others may be formed during water treating or softening operations, or by leaching from ash or refuse. Due to evaporation in the cooling tower, dissolved solids can buildup in concentration to a level approaching brackish water which would not be acceptable for discharge from the plant at inland locations. For the basis shown on Figure 1, the amount of water evaporated is 5 times the net effluent; consequently, dissolved solids will increase by a factor of 6 at least over that in the makeup water. One approach to this problem is to evaporate the water effluent to dryness (e.g., in a pond) and store the salts or dispose of them in the ocean.

Many trace elements are partially volatile in the gasifiers and must be removed in the gas cleanup systems, thereby showing up in the net effluents of liquids and solids from the plant. To a large extent, these show up in the waste water (e.g., fluorine, arsenic, and chlorine). At this time there is almost no information available to define the problem or to outline treating methods that would recover or deactivate harmful materials. It is unlikely that conventional waste water treating will be satisfactory for this purpose. The subject will be discussed further in Section 10 on trace elements.

A solid effluent from water treating is sludge from biox, which may be several hundred tons/day of material having a high water content, and difficult to filter. It might be disposed of as land fill if odor problems can be controlled, or it could be incinerated although extraneous fuel may be necessary.

The facilities for treating makeup water also generate sludge, as well as various liquid effluents. In this case the sludge is innocuous and can be buried or disposed of with the char. Demineralization of boiler feed water usually uses ion exchange resins which are back washed with acid or alkali. These effluents can be combined and neutralized, but still contribute to total dissolved solids in the waste water. In brief, all chemicals used by the plant must also leave in some stream, and in many cases they will leave in the waste water.

7. SULFUR BALANCE

Of the sulfur entering in the coal feed, nearly all of it appears in the raw gas leaving the gasifiers, from which it can be separated and sent to the Claus plant for sulfur recovery. The latter gives 99% sulfur recovery with tail gas cleanup. The HYGAS design is based on essentially complete recovery of all sulfur compounds by the Rectisol unit, so that sulfur emission in the CO₂ vent gas is very low—much less than 1% of the sulfur entering with the coal. In addition, the gas stream sent to the Claus plant is very high in sulfur, equivalent to 30 vol. % H₂S, giving a high sulfur recovery. There is some question as to whether this is completely consistent with the utilities consumption listed in Section 11. In the absence of detailed numbers, the latter were taken from the design for the El Paso Lurgi Plant, which gave only 1% H₂S in the gas to sulfur recovery and may therefore have relatively lower consumption of utilities.

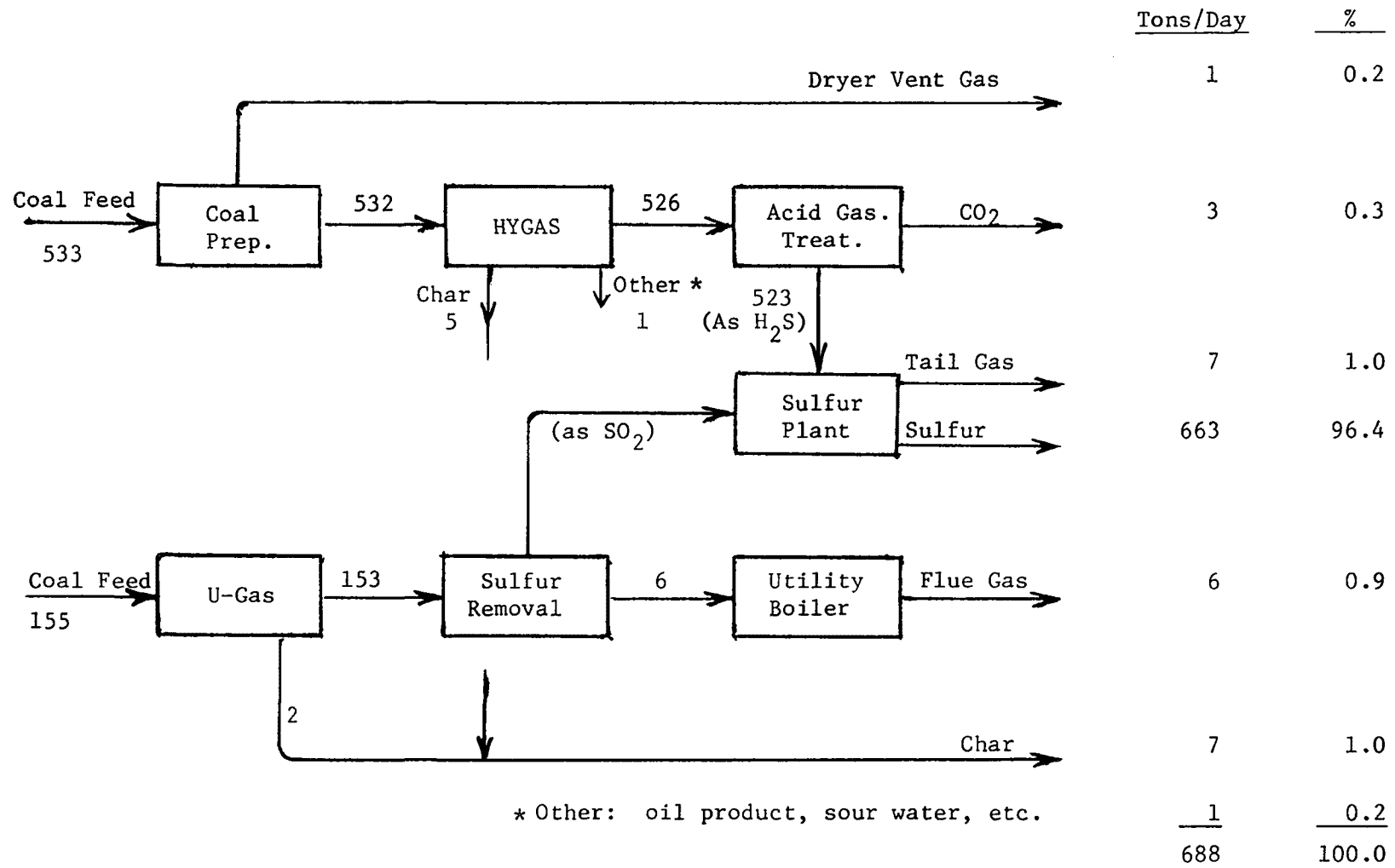
Overall sulfur balance for the plant is summarized in the diagram on Figure 4, including the U-Gas unit supplying clean fuel gas to coal drying and the utilities area. Sulfur is removed in the U-Gas system using a molten metal, which is regenerated by blowing with air to form SO₂ which goes to the sulfur plant. This system is presumed to give about 96% sulfur removal including compounds such as COS, etc.

While effective control of sulfur emissions and high sulfur recovery are indicated by the numbers on Figure 4, supporting confirmation of the design basis will be essential in any actual commercial application.

FIGURE 4

SULFUR BALANCE - HYGAS PROCESS

(Numbers are Tons/Day Sulfur)



8. THERMAL EFFICIENCY

It is of interest to compare the heating value of pipeline gas product versus that of total coal consumed by the plant including auxiliaries. This represents the thermal efficiency for the process and relates to raw materials consumed, as well as to the amount of waste heat that must be rejected to the environment, or that appears in by-products such as oil. In order to be meaningful the calculation must include all processing, auxiliaries, environmental controls, utilities, etc., needed to make the plant self-sufficient. In this HYGAS design thermal efficiency is 64.6% for the complete plant, as shown in Table 2.

Oil by-product is included, and adds 4.3% to thermal efficiency. It is also informative to look at where losses in efficiency occur, so that their relative importance can be examined. The lower part of Table 2 shows a breakdown of losses. Evaporation of water in the cooling tower is by far the largest, followed by sensible heat transferred to the air as it passes through the cooling tower.

The tabulated numbers assume that rejected char contains only 10.3% carbon. If this low level is difficult to achieve or results in excessive attrition within the gasifier, then thermal efficiency might decrease. Thus if the char contained 30% carbon, loss via this stream would increase to 5.7% of the high heating value of total coal used.

In the energy balances used for this study, all electric power needed for the plant is generated onsite, and no power is purchased.

TABLE 2
THERMAL EFFICIENCY
HYGAS PROCESS

	<u>Tons/Day</u>	<u>10⁹ Btu/Day</u>	<u>%</u>
<u>In:</u> Coal to Hygas	12,695	320	77.5
Coal to U-Gas	<u>3,686</u>	<u>93</u>	<u>22.5</u>
	16,381	413	100.0
<u>Out:</u> Pipeline Gas	5,167	249	60.3
Oil By-Products	<u>478</u>	<u>18</u>	<u>4.3</u>
	5,645	267	64.6
<u>Losses:</u>			
Char (10.3% Carbon)	2,117	6.3	1.5
CO ₂ Vent Gas	13,726	6.5	1.6
Dryer Vent Gas	3,647	1.3	0.3
Cooling Tower Evap.	27,120	57.0	13.8
Cooling Tower Sens. Heat	2,800,000	29.2	7.1
Furnace Flue Gas	34,280	10.0	2.4
By-Products:			
Sulfur	666	5.3	1.3
Ammonia	135	2.6	0.6
Phenol	16	0.5	0.1
Other Losses	--	<u>27.3</u>	<u>6.7</u>
		146	35.4

9. TRACE ELEMENTS

Coal contains many trace elements present in less than 1% concentration that need to be carefully considered from the standpoint of potential impact on the environment. Many of these may volatilize to a small or large extent during processing, and many of the volatile components can be highly toxic. This is especially true for mercury, selenium, arsenic, molybdenum, lead, cadmium, beryllium and fluorine. The fate of trace elements in coal conversion operations, such as gasification or liquefaction, can be very different than experienced in conventional coal fired furnaces. One reason is that the conversion operations take place in a reducing atmosphere, whereas in combustion the conditions are always oxidizing. This maintains the trace elements in an oxidized condition such that they may have more tendency to combine or dissolve in the major ash components such as silica and alumina. On the other hand, the reducing atmosphere present in coal conversion may form compounds such as hydrides, carbonyls or sulfides which may be more volatile. Studies on coal fired furnaces have indicated that smaller particles in fly ash contain a higher concentration of trace elements, presumably due to volatilization of these elements in the combustion zone and their subsequent condensation and collection on the fly ash particles (29). Other studies on coal fired furnaces are pertinent (30,31,32) and some of these report mass balances on trace elements around the furnaces (33).

Considerable information is available on the analyses of coal, including trace constituents, and these data have been assembled and evaluated (34,35). A few experimental studies have been made to determine what happens to various trace elements during gasification (2,37,38). As expected, these show a very appreciable amount of volatilization on certain elements. As an order of magnitude, in this specific HYGAS design each 10 ppm of element volatilized would amount to about 240 pounds per day.

Results on the fate of trace elements in commercial gasification plants is rather limited, but an effort was made to assemble and evaluate the available data. Some trace elements show up in unexpected places, thus analyses of tar from gasification show up to 50 parts per million of lead, while oil samples from gasification show 30 parts per million of arsenic. This raises environmental and safety questions on subsequent use of such materials. One further example is a report of about 200 parts per million of titanium in the heavy liquid or tar produced in a coal liquefaction pilot plant (7). While titanium is not now considered to be one of the more toxic elements, further consideration of the implications is called for.

In order to make the picture on trace metals more meaningful, the approximate degree of volatilization shown for various elements has been combined with their corresponding concentration in a hypothetical coal (as typical), giving an estimate of the pounds per day of each element that might be carried out with the hot gases leaving the gasifier. Results are shown in Table 3 in the order of decreasing volatility. Looking at the

TABLE 3
TRACE ELEMENTS - ESTIMATED VOLATILITY

	<u>Hypothetical Coal ppm</u>	<u>% Volatile*</u>	<u>lb/day**</u>
Cl	1500	90+	32,400
Hg	0.2	90+	4
Se	2.2	74	39
As	31	65	484
Pb	7.7	63	116
Cd	0.14	62	2
Sb	0.15	33	1
V	35	30	252
Ni	14	24	81
Be	2	18	9
Cr	22	nil	nil
Zn	44	e.g. 10	106
B	165	e.g. 10	396
F	85	e.g. 10	204
Ti	340	e.g. 10	816

* Volatility based mainly on gasification experiments (38) but chlorine is taken from **combustion** tests, while zinc, boron, and fluorine were taken at 10% for illustration in absence of data.

** Estimated volatility for 12,000 tons/day of coal to gasification.

estimated amounts that may be carried overhead, it becomes immediately apparent that there can be a very real problem. For each element the net amount carried out in the gas leaving the gasifier should be collected, removed from the system, and disposed of in an acceptable manner. Environmental controls will be needed if the effluent contains excessive amounts of undesirable components, particularly if these are toxic elements. In the case of zinc, boron and fluorine the degree of volatilization has not yet been determined, but they would be expected to be rather volatile. Even if only 10% of the total amount is volatile, there will be large quantities to remove in the gas cleaning operation and to dispose of.

A complication that has not generally been recognized, occurs in the gas cleanup section due to the volatility of trace elements. These may be carried out with the raw gas, and removed in the gas cleanup facilities when the gas is cooled and scrubbed. In any event, they do not remain in the pipeline gas, and it follows that they must leave the system at some point. Compounds such as cyanides might be destroyed by recycling to the process (e.g., the gasifier), but this can not be the case for elements such as arsenic, lead, chlorine, etc. Neither will they disappear in the biox unit. Therefore provision will be needed to separate and recover them, or to deactivate them for disposal in a satisfactory manner. As can be seen from Table 3, the combined amounts of all volatile portions of potentially undesirable trace elements can present a formidable disposal problem.

The preceding discussion has been directed primarily at trace elements that are partially volatilized during gasification and thereby carried into the gas cleaning section. Consideration must also be given to trace metals that are not volatilized and leave in the solid effluents from the plant, one of which is the char from gasification. Undesirable elements might be leached out of this char since it is handled as a water slurry, and the char will ultimately be exposed to leaching by ground water when it is disposed of as land fill or to the mine. Sufficient information is not now available to evaluate the potential problems associated with char disposal, and additional information and evaluation is needed. The situation may be quite different from the ash rejected from coal fired furnaces, since the char is produced in a reducing atmosphere rather than an oxidizing one. Background information on slag from blast furnaces used in the steel industry may be pertinent from this standpoint, since the blast furnace operates with a reducing atmosphere. However, a large amount of limestone is also added to the blast furnace, consequently the nature of the slag will be different.

10. TECHNOLOGY NEEDS

From this review and examination of environmental aspects of the HYGAS process, a number of areas have been defined where further information is needed in order to evaluate the situation or where additional studies or experimental work could lead to a significant improvement from the standpoint of environmental controls, energy consumption, or thermal efficiency of the process. Items of this nature will be discussed in this section of the report, and a summary is shown in Table 4.

Any coal conversion operation has solid refuse to be disposed of. Rejected char in the present design generates over 3,000 acre feet per year of refuse. More work is needed in order to define methods of disposal that do not create problems due to leaching of acids or sulfur which could contaminate natural water. In addition, adequate controls are needed with regard to the potential dust nuisance and washing away of particulates, since the char is expected to have a low density, and be quite friable. In many cases the material may be suitable for land fill with revegetation. Although there is already some general background on this subject, specific information is needed on each coal for each process, and each specific location in order to allow thorough planning to be sure that disposal will be environmentally sound.

Coal drying is used on most coal conversion processes; consequently, considerable effort is warranted to optimize the operation from the standpoints of fuel consumption, dust recovery, and volume of vent gas to be handled. It will often be attractive to burn high sulfur coal rather than clean gas fuel and to include facilities to remove sulfur from the vent gases, since this gas must be processed in any event for dust removal.

The need for a simple, efficient means of feeding coal to the high pressure gasifier has been apparent and has received considerable study. For pressure levels of 400-500 psig, lock hoppers have been used satisfactorily, although they are expensive. For systems at 1,000 psig, it may be attractive to pump an oil or water slurry of the coal in order to pressurize it. A water slurry could be particularly attractive if it is possible to then evaporate the water at high pressure and thereby supply steam to the gasifier (5).

In the area of acid gas removal, scrubbing with refrigerated methanol may give satisfactory cleanup of the gas but utilities consumption is high, and the CO₂ vent stream requires further treatment to remove combustibles, and possibly also COS. Systems based on amine or hot carbonate are not completely satisfactory and leave room for improvement. Amine scrubbing is not effective on carbonyl sulfide, and it is often difficult to provide a highly concentrated stream of H₂S to send to the sulfur plant. In addition the CO₂ stream vented to the atmosphere may contain too much sulfur. Adsorption/oxidation systems are often not effective on carbonyl sulfide and in any event do not remove CO₂ as required; and therefore, additional processing is needed. The available

TABLE 4
TECHNOLOGY NEEDS

- Environmentally sound disposal of large amounts of char from gasification, with regard to dust, leaching and sediment, trace elements, land use, etc.
- An optimized design for coal drying to use low excess air and give maximum allowable coal preheat, with good dust recovery.
- An improved system to feed coal into high pressure zones, for example using a piston feeder on oil or water slurry. If water is used, then it might be evaporated in a heated fluid bed to make steam for gasifier, and preheat the coal fed to gasification.
- A simpler and more efficient process for acid gas removal which would provide an H_2S stream of high concentration (e.g. 50 vol. %) to the sulfur plant, while giving a separate clean stream of CO_2 that can be vented directly to the air. Desirable features to include:
 - Good sulfur cleanup, to a few ppm
 - A clean CO_2 vent stream that does not require incineration or cleanup
 - Low utilities consumption
 - Little or no chemical purges to dispose of
- An effective process to remove sulfur at high temperature could lead to improvements such as reacting CO directly with steam to form methane, and thereby avoid cooling and reheating the gas as in present designs.
- Ways to treat COS, CS_2 , thiophene, etc., that are usually present and may not be handled effectively by many acid gas removal processes. Hydrolysis to H_2S is one approach, and would assure that COS, for example, does not escape with the CO_2 vent stream.
- Sour water cleanup techniques need evaluation and demonstration. There is a great need for a practical way to evaporate sour water to make steam for use in the gasifier, and a fluid bed system appears promising.
- Information on trace elements and techniques for their disposal.
 - Extent of volatility for specific process and coal.
 - Where they appear in gas cleanup system, and in what form. They may collect on the char or shift catalyst, in sour water or acid gas removal, or in the by-product oil.
 - Many trace elements may be toxic and require separation and decontamination treatment before disposal.
 - Since trace elements contained in the coal feed must leave the system at some point, specific means must be defined either to recover them, or dispose of them in an environmentally acceptable manner.
 - Leaching may occur on the rejected char. Information is needed to define the potential problems and to devise environmentally sound disposal techniques.

systems for acid gas removal have very high utility requirements, causing a significant loss in thermal efficiency for conversion of coal to clean fuel products. In addition there is often a waste stream of chemical scrubbing medium which may be difficult and expensive to dispose of.

Desirable objectives for an acid gas removal process can be summarized as follows: (a) good cleanup of all forms of sulfur to give a stream high in sulfur concentration for processing in a Claus type sulfur plant, (b) effective CO₂ removal while producing a vent stream satisfactorily low in sulfur and pollutants, (c) low utility and energy consumption, and (d) no waste streams that present a disposal problem.

The need for a simple, effective method to clean up sour water for reuse is another item that is common to most fossil fuel conversion operations. Sour water generally contains sulfur compounds, ammonia, H₂S, phenol, thiocyanates, cyanides, traces of oil, etc. These are generally present in too high a concentration to allow going directly to biological oxidation, but their concentration is often too low to make recovery attractive. Particulates, if present, further complicate the processing of sour water. Usual techniques for clean up include sour water stripping to remove H₂S and ammonia, and in addition, extraction may be required to remove phenols and similar compounds. Such operations are large consumers of utilities and have a large effect on overall thermal efficiency.

As in most gasification processes, the amount of sour water produced is less than the amount of steam fed to gasification plus shift conversion, which suggests a way to dispose of sour water. One approach is to vaporize the sour water to make steam which can be used in the gasifier. In this case, compounds such as phenol should be destroyed and reach equilibrium concentration in the circulating sour water. It may not be practical to vaporize sour water in conventional equipment such as exchangers, due to severe fouling and corrosion problems. Therefore, new techniques may be required, and one possibility would be to vaporize the sour water by injecting it into a hot bed of fluidized solids (5).

On trace elements, information is needed on the amount vaporized in the gasifier and what happens to them, where they separate out, and in what form, so that techniques can be worked out for recovering or disposing of the materials. Again specific information is needed for each coal and for each coal conversion process since operating conditions differ. In many cases, the trace elements may tend to recycle within the system and build up in concentration. This offers an interesting opportunity to perhaps recover some of them as useful by-products. The toxic nature of many of the volatile elements should be given careful consideration from the standpoint of emissions to the environment, as well as protection of personnel during operation and maintenance of the plant. Carcinogenicity of coal tar and other compounds present in trace amounts or formed during start up or upsets should also be evaluated.

Protection of personnel, especially during maintenance operations should be given careful attention, which will require that additional information be obtained. Thus, toxic elements that vaporize in the gasifier may condense in equipment such as piping and exchangers where they could create hazards during cleaning operations.

In an actual application, the net water effluent from the plant will have to be cleaned up. Water make-up that is brought to the plant will contain dissolved solids including sodium and calcium salts. Calcium salts may be precipitated during the water treating operation to form a sludge which can be disposed of with the other waste solids, but the fate of the sodium salts in the make-up water calls for further study. These will leave with the blowdown from the cooling tower. If the concentration of dissolved solids is too high in this blowdown water to allow discharging it to the river, then some suitable method of disposal will have to be worked out. On one proposed commercial plant, this has been handled by using an evaporation pond where the water is evaporated to dryness. The salts accumulate and will ultimately have to be disposed of. If they cannot be used or sold then it would seem logical to store them, or dispose of them in the ocean. It would be desirable to define alternative technology for disposing of or using more effectively the final waste water discharge from a plant.

11. PROCESS DETAILS

Further information on the basis used in this evaluation is given in Tables 5-8, which show coal feed and products for the plant, as well as utilities consumption. The latter were not given for the basic design, which was not specified in sufficient detail to allow calculating them; however, the overall process is very similar to the plant design of the El Paso Natural Gas project for New Mexico based on Lurgi gasifiers (3), which therefore provides a sound basis for utilities required on the HYGAS process. In fact, both designs are sized to make the same amount of SNG. The gas cleanup systems are also quite similar using the Rectisol process for acid gas treatment, and the Phenosolvan process to extract phenols. Since the El Paso design gives only about 1% H₂S in the stream to sulfur recovery compared to the 30% assumed for HYGAS, the utilities consumption in acid gas treatment may be higher than we have used.

Operating pressure is higher for HYGAS, 1200 psia versus about 500 psia for Lurgi, which increases the power required for oxygen compression and coal feeding. Also, the steam fed to the gasifier cannot be used first in bleeder turbines to generate power as is done in the El Paso design. Offsetting these factors is the saving gained by eliminating the product SNG compressor, made possible by operating the HYGAS process at higher than pipeline pressure.

In both cases, clean gas for plant fuel is made by air gasification of coal, and this gas is used in a combined cycle to generate useful power from gas turbines before it is used in furnaces. All electrical power used by the plant is generated onsite and allowed for in the energy balances. No power is purchased.

Rough estimates of differences between the two processes show that utilities requirements will be about the same although some modifications to the El Paso utility consumptions were made where practical. There will be some debit for the HYGAS process in that high level heat in the raw gas from gasification is degraded in temperature level by the drying bed and oil quench system, so that it cannot be used to make high pressure steam.

TABLE 5

COAL FEED AND PRODUCTS - HYGAS PLANT

	<u>tons/day</u>
<u>Coal Feed</u> - Dry Basis to HYGAS	12,695
(Illinois No. 6) to U-Gas	<u>3,686</u>
	16,381

<u>Coal Analysis</u> - Dry Basis	<u>Wt. %</u>
Carbon	69.40
Hydrogen	4.80
Oxygen	8.71
Nitrogen	1.35
Sulfur	4.20
Ash	<u>11.54</u>
	100.00

High Heating Value 12,600 Btu/lb

Pipeline Gas 250 MM SCFD @ 1000 psig
(H.H.V. 960 Btu/SCF)

<u>Char</u>	<u>Tons/Day</u>
From HYGAS	1,667
From U-Gas	<u>450</u>
	2,117

(10.3% Carbon, 0.3% Sulfur, 89.4% Ash, H.H.V. 1500 Btu/lb)

Product Oil 478 Tons/Day (H.H.V. 18,800 Btu/lb)

By-Products

Sulfur	663 tons/day
Ammonia	120 tons/day
Phenol	16 tons/day

TABLE 6
ESTIMATED STEAM BALANCE HYGAS PROCESS*

<u>Source</u>		<u>Use</u>	
	<u>1500 Psia (955°F)</u>		
Power Boiler	1489 M Lb/Hr	Electrical Generator	1105 M Lb/Hr
		Pipeline Compressor	384
	<u>1100 Psia (930°F)</u>		
Methanation and Superheater	1354 M Lb/Hr	Pipeline Compressor	571 M Lb/Hr
		Methanation Recycle Compressor	784
	<u>500 Psia (752°F)</u>		
Gasifier Jacket (O ₂ Blown)	171 M Lb/Hr	Gasifier (O ₂ Blown)	1762 M Lb/Hr
Pipeline and Methanation Compressors	1738	Gasifier (Air Blown)	312
Gasifier Jacket (Air Blown)	54	O ₂ Plant Turbine	132
Power Generator	842	Lock Gas Compressor (O ₂ Blown)	118
		Lock Gas Compressor (Air Blown)	35
		Oxygen Compressor	314
		Air Compressor (Air Blown)	132
	<u>112 Psia (336°F)</u>		
Waste Heat Boiler (O ₂ Blown)	741 M Lb/Hr	Phenosolvan	32 M Lb/Hr
		Rectisol	20
		Stretford Plant	21
		Refrigeration Compressor	435
		Condenser	234

* Note: This steam balance is from the Lurgi study using Lurgi gasifiers (3), in order to provide a complete picture. Overall steam balance would be about the same for the HYGAS process, although there would be some increases and decreases as discussed in the text. Steam production from gasifier jacket assumed to be similar for HYGAS plus U-Gas combination.

TABLE 7
ELECTRIC POWER CONSUMPTION
HYGAS PROCESS*

	<u>KW</u>
Coal Preparation	6,000
Gas Purification	13,200
Sulfur Plant	4,200
Gas Liquor Treating	3,600
Cooling Water System	7,000
Power Plant	8,000
U-Gas e.g.	3,000
Other e.g.	<u>12,000</u>
	57,000*

* All power consumed is supplied by onsite generation using appropriate facilities, and necessary fuel requirements, etc., are included in overall balances for plant.

TABLE 8
WATER BALANCE
HYGAS PROCESS

<u>Consumed</u>	<u>Tons/Day</u>	<u>gpm</u>
Steam to Gasifier	11,780	1,963
Steam to Shift	4,955	826
Water to Scrubber	2,824	470
Steam to U-Gas	2,560	427
Evaporated in Cooling Tower	27,120	4,520
Net Discharge from Plant	<u>5,424</u>	<u>904</u>
	54,663	9,110
<u>Recovered</u>		
From Scrubber	9,678	1,613
From Methanation	2,520	420
From Acid Gas Treatment	<u>37</u>	<u>6</u>
	12,235	2,039
<u>Net Makeup Water Required</u>	42,428	7,071

Note: Cooling water circulation rate will be
roughly 200,000 gpm.

12. QUALIFICATIONS

A major qualification of this study is that it assumes that no pretreat of the coal will be needed. The results therefore apply to those applications which use a non-caking coal. It is the hope of the Institute of Gas Technology that the process can eventually be used on caking coal without pretreatment, but additional development and demonstration will be needed to confirm this.

The basis used does not include coal cleaning, which is a very important factor in environmental impact of coal conversion technology, and has been included in some other studies in this series (7). Refuse from coal cleaning may be 20-25% of the coal as mined, presenting a sizeable disposal problem. Although it has not been included, suitable provision will have to be defined and evaluated in order to have a viable and complete gasification project.

As pointed out in connection with acid gas treatment, the very desirable high concentration of 30% H_2S shown as feed to the Claus plant is not confirmed by results from some operating plants and projected plant designs; consequently, further consideration and evaluation of this feature is needed.

An important basis item that can have a large effect on thermal efficiency is the carbon content assumed for the char withdrawn from the HYGAS and U-Gas reactors. Unused carbon in the rejected char is a direct loss of heating value in the coal feed. A higher than minimum amount of residual carbon may be set to control attrition and dusting in the fluidized or suspended solids reaction system, as well as by the reaction kinetics and extent of staging in gasification. A carbon content of 10.3% has been assumed, but other publications show higher values, and at 30% carbon content in char the number for thermal efficiency would be 4.2% lower.

Clean gas for plant fuel is provided by the U-Gas process, which is also in the development stage. Full use is made of the combined cycle operation using gas turbines to supply power, and thereby gain a significant increase in efficiency for generating utilities. Combined cycles are being explored intensively, although they are not used conventionally yet by public utilities or industry.

In view of these and other factors used in setting a study basis, great caution should be exercised in attempting to make comparisons between processes, since they must be on a strictly comparable basis if the results are to be meaningful. Variations that must be taken into account include coal type and sulfur content, plant location, amount of emissions relative to permissible, production and disposition of tar and other by-products, utilities systems, use of air cooling, and scope of project.

13. BIBLIOGRAPHY

1. Magee, E. M., et al., "Evaluation of Pollution Control in Fossil Fuel Conversion Process, Gasification: Section 1: Koppers-Totzek Process," EPA-650/2-74-009a, January 1974.
2. Kalfadelis, C. D., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes, Gasification: Section 2: Synthane Process," EPA-650/2-74-009b, June 1974.
3. Shaw, H., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes, Gasification: Section 3: Lurgi Process," EPA-650/2-74-009c, July 1974.
4. Jahnig, C. E., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes, Gasification Section 4: CO₂ Acceptor Process," EPA-650/2-74-009d, December 1974.
5. Jahnig, C. E., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes," Gasification: Section 5: BI-GAS Process," EPA-650/2-74-009g, April 1975.
6. Kalfadelis, C. D., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes," Liquefaction: Section 1: COED Process," EPA-650/2-74-009e, January 1975.
7. Jahnig, C. E., "Evaluation of Pollution Control in Fossil Fuel Conversion Processes, Liquefaction: Section 2: Solvent Refined Coal Process," EPA-650/2-74-009f
8. Study Made for EPA by Booz Allen and Hamilton Co. on "Pollution Control in Fossil Fuel Conversion Processes," Report EPA 9075-015, March 1974 Sections VII and XIV.
9. Schora, F. C., et al., "The HYGAS Process," IGT Clean Fuels Symposium, Chicago, Ill., Sept. 1973.
10. Schora, F. C., "Clean Energy from Coal," Intersociety Energy Conversion Conference, San Francisco, Calif., August 26-30, 1974.
11. Lee, B. S., "Status of HYGAS Process," Fifth Synthetic Pipeline Gas Symposium, Chicago, Ill., October 29-31, 1973.
12. Schora, F. C., et al., "Analysis of a HYGAS Coal Gasification Plant Design," AIChE 65th Annual Meeting, New York City, Nov. 27-30, 1972.
13. Bodle, W. W., et al., "Clean Fuels from Coal," The Oil Gas Journal August 26, 1974, pg. 73-88.
14. Kavlic, V. J., et al., "Coal Pretreatment in Fluidized Bed," American Chemical Society, Division of Fuel Chemistry, September 1966.

15. Lee, B. S., "Status of HYGAS, Electrothermal Gasification, and Steam Oxygen Gasification Programs," Fourth Synthetic Pipeline Gas Symposium, Chicago, Ill., Oct. 30-31, 1972.
16. Loeding, J. W., et al., "The U-Gas Process," Chemical Engineering Progress 71, (4) pg. 85, (April 1975).
17. Loeding, J. W., et al., "Clean Fuels from Coal Symposium," Institute of Gas Technology, Chicago, Sept. 10-14, 1973.
18. Bolln, J. J., "Commercial Concept Designs," Fifth Synthetic Pipeline Gas Symposium, Chicago, Ill., Oct. 29-31, 1973.
19. Gibson, C. R., et al., "Environmental Aspects of El Paso's Burnham/Coal Gasification Complex," EPA Symposium: Environmental Aspects of Fuel Conversion Technology, St. Louis, Mo., May 1974, EPA-650/2-74-118.
20. Berty, T. E., et al., "Environmental Aspects of the WESCO Coal Gasification Plant," EPA Symposium: Environmental Aspects of Fuel Conversion Technology, St. Louis, Mo., May 1974, EPA-650/2-74-118.
21. El Paso New Mexico Coal Gasification Project Environmental Statement 1974.
22. Forney, A. J., et al., "Analyses of Tars, Chars, Gases, and Water Found in Effluents from the Synthane Process," EPA-650/2-74-118.
23. Farnsworth, J. F., et al., "Clean Environment with Koppers-Totzek Process," EPA-650/2-74-118.
24. Colgate, J. L., et al., "Gob Pile Stabilization, Reclamation, and Utilization," Office of Coal Research R&D Report No. 75, 1973.
25. Goldberger, W. M., "The Union Carbide Coal Gasification Process," Fourth Synthetic Pipeline Gas Symposium, Chicago, Ill., Oct. 30-31, 1972, pg. 257.
26. Corder, W. C., "The Union Carbide/Battelle Coal Gasification Process," Fifth Synthetic Pipeline Gas Symposium, Chicago, Ill. Oct. 29-31, 1973, pg. 107.
27. Furlong, E., "Cooling Tower Operations," Environmental Science and Technology 8, (8) pg. 712 (Aug. 1974).
28. Lee, B. S., "Slurry Feeding of Coal Gasifiers," Chemical Engineering Progress 71, (4), pg. 81, (April 1975).

29. Lee, R. E., et al., "Trace Metal Pollution in the Environment," Journal of Air Pollution Control, 23, (10), October 1973.
30. Schultz, H., Hattman, E. A., and Booker, W. B., ACS Div. of Fuel Chem., Vol. 8, No. 4, p. 108, August 1973.
31. Billings, C. E., Sacco, A. M., Matson, W. R., Griffin, R. M., Coniglio, W. R., and Harley, R. A., "Mercury Balance on a Large Pulverized Coal-Fired Furnace," J. Air Poll. Control Association, Vol. 23, No. 9, September 1973, p. 773.
32. Schultz, Hyman et al., "The Fate of Some Trace Elements During Coal Pretreatment and Combustion," ACS Div. Fuel Chem. 8, (4), p. 108, August 1973.
33. Bolton, N. E., et al., "Trace Element Mass Balance Around a Coal-Fired Stream Plant," NCS Div. Fuel Chem., 18, (4), p. 114, August 1973.
34. Magee, E. M., Hall, H. J., and Varga, G. M., Jr., "Potential Pollutants in Fossil Fuels," EPA-R2-73-249, June 1973.
35. Trace Elements and Potential Toxic Effects in Fossil Fuels H. J. Hall, EPA Symposium "Environmental Aspects of Fuel Conversion Technology," St. Louis, Mo., May 1974, EPA-650/2-74-118.
36. Ruch, R. R., et. al., "Occurrence and Distribution of Potentially Volatile Trace Elements in Coal," EPA-650/2-74-054, July 1974.
37. Attari, A., "The Fate of Trace Constituents of Coal During Gasification," EPA Report 650/2-73-004, August 1973.
38. Attari, A., et al., "Fate of Trace Constituents of Coal During Gasification," (Part II), Presented at Amercian Chemical Society Meeting, Div. of Fuel Chem., Phil., PA., April 6-11, 1975.

TECHNICAL REPORT DATA (Please read Instructions on the reverse before completing)		
1. REPORT NO. EPA-650/2-74-009-h	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE Evaluation of Pollution Control in Fossil Fuel Conversion Processes; Gasification: Section 6. HYGAS Process		5. REPORT DATE August 1975
		6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) C. E. Jahnig		8. PERFORMING ORGANIZATION REPORT NO. Exxon/GRU.11DJ.75
9. PERFORMING ORGANIZATION NAME AND ADDRESS Exxon Research and Engineering Company P.O. Box 8 Linden, NJ 07036		10. PROGRAM ELEMENT NO. 1AB013: ROAP 21ADD-023
		11. CONTRACT/GRANT NO. 68-02-0629
12. SPONSORING AGENCY NAME AND ADDRESS EPA, Office of Research and Development Industrial Environmental Research Laboratory Research Triangle Park, NC 27711		13. TYPE OF REPORT AND PERIOD COVERED Final
		14. SPONSORING AGENCY CODE
15. SUPPLEMENTARY NOTES		
16. ABSTRACT <p>The report gives results of a review of the HYGAS process being developed by the Institute of Gas Technology, from the standpoint of its potential for affecting the environment. The quantities of solid, liquid, and gaseous effluents have been estimated where possible, as well as the thermal efficiency of the process. For the purpose of reduced environmental impact, a number of possible process modifications or alternatives which could facilitate pollution control or increase thermal efficiency have been proposed, and new technology needs have been pointed out.</p>		
17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Air Pollution Coal Gasification Fossil Fuels Thermal Efficiency Trace Elements	Air Pollution Control Stationary Sources Clean Fuels HYGAS Process Fuel Gas Research Needs	13B 13H 21D 20M
18. DISTRIBUTION STATEMENT Unlimited	19. SECURITY CLASS (This Report) Unclassified	21. NO. OF PAGES 60
	20. SECURITY CLASS (This page) Unclassified	22. PRICE

ENVIRONMENTAL PROTECTION AGENCY
Technical Publications Branch
Office of Administration
Research Triangle Park, North Carolina 27711

OFFICIAL BUSINESS

AN EQUAL OPPORTUNITY EMPLOYER

POSTAGE AND FEES PAID
ENVIRONMENTAL PROTECTION AGENCY
EPA - 335



Return this sheet if you do NOT wish to receive this material ☐
or if change of address is needed ☐. (Indicate change, including
ZIP code.)

PUBLICATION NO. EPA-650/2-74-009-h