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EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES

GASIFICATION; SECTION I. LURGI PROCESS



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EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES

GASIFICATION; SECTION 1. LURGI PROCESS

by

H. Shaw and E. M. Magee

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

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EPA Project Officer: William J. Rhodes

Control Systems Laboratory
National Environmental Research Center
Research Triangle Park, North Carolina 27711

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SUMMARY

A process analysis of the Lurgi Dry Ash Gasification Process for high Btu gas was carried out. The process has been reviewed from the standpoint of its potential for affecting the environment. The waste stream compositions were calculated for a 250 MM scfd synthetic natural gas plant using a subbituminous coal. Thus, the quantities of solid, liquid, and gaseous pollutants were estimated, where possible. The thermal efficiency for various process alternatives was calculated. A number of process modifications which would reduce pollution and/or increase thermal efficiency were suggested. The technology needs to control pollution were assessed.

TABLE OF CONVERSION UNITS

<u>To Convert From</u>	<u>To</u>	<u>Multiply By</u>
Btu	Calories, kg	0.25198
Btu/pound	Calories, kg/kilogram	0.55552
Cubic feet/day	Cubic meters/day	0.028317
Feet	Meters	0.30480
Gallons/minute	Cubic meters/minute	0.0037854
Inches	Centimeters	2.5400
Pounds	Kilograms	0.45359
Pounds/Btu	Kilograms/calorie, .kg	1.8001
Pounds/hour	Kilograms/hour	0.45359
Pounds/square inch	Kilograms/square centimeter	0.070307
Tons	Metric tons	0.90719
Tons/day	Metric tons/day	0.90719

INTRODUCTION

A serious shortage of the more convenient and less costly fossil fuels is projected (1). Substantial fuel reserves which can be used directly in a way that does not harm the environment are similarly not available (2). A large effort is underway to develop technology to convert some of the large sources of domestic fuels to convenient and clean fuels (3). One of the most advanced technical efforts is in the area of converting high sulfur bearing coals to synthetic natural gas (4). There are other programs which are designed to convert coal to low sulfur fuel oil (5).

The Environmental Protection Agency has anticipated the need to produce convenient and environmentally acceptable fuels from fossil fuels which could be environmentally harmful. The contemplated processing plants for converting the less clean fuels would have the burden of removing the sulfur and other potential pollutants. Thus, the fuel conversion plant itself could become a source of pollution to the environment. Therefore, the time is ripe to assess the potential pollution problems that might be associated with such plants. If problems are anticipated at this time, then potential solutions can be developed prior to the construction of a commercial plant. An awareness of potential pollution problems will allow the developer to obviate most of the problems through proper design and construction.

The Environmental Protection Agency has awarded Contract No. EPA-68-02-0629 to evaluate the current status of fossil fuel conversion and/or treatment processes with respect to pollution control and thermal efficiency. Specifically, Exxon Research and Engineering Company is performing a detailed pollution control assessment of representative processes using non-proprietary information. As a result of this study the "technology needs" to minimize pollution will be delineated in order to allow sufficient time for research, development and design of adequate pollution control equipment in coal gasification processes.

All significant input streams to the processes must be defined, as well as all effluents and their compositions. Complete mass and energy balances are required to determine all gas, liquid, and solid streams. With this information, facilities for control of pollution can be examined and modified as required to meet Environmental Protection Agency 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 necessary to produce a given quantity of clean fuel. It is also a way of estimating the amount of raw fuel resources that are consumed in making the relatively pollution-free fuel. In view of the projected energy shortage this is an important consideration. Suggestions are included concerning technology gaps that exist for techniques to control pollution or conserve energy. Not included in this study are such areas as cost, economics, operability, etc. Coal mining and general offsite facilities are also not within the scope of this study.

Considerable assistance was received in making this study, and we wish to acknowledge the help and information furnished by EPA as well as that obtained from many specialists in Exxon Research and Engineering Company. Comments furnished by El Paso Natural Gas Company and by American Lurgi Corporation are also appreciated.

1. PROCESS DESCRIPTION

The present analysis of the Lurgi Dry Ash Gasification process draws heavily on the Stearns-Roger design for the El Paso Natural Gas Company (6). The location factors have been generalized in order to be consistent with the other coal gasification analyses that are being made. It should be emphasized that this work is not an attempt to analyze the plant of the El Paso Natural Gas Company since the design of that plant has been modified from that of the original FPC filing. This section is divided into two parts. One describes the equipment and processes associated with the SNG manufacture, and the other describes the auxiliary facilities that are required to make this plant self-supporting in utilities. A simplified process flow diagram is given in Figure 1 to help explain the interrelationships of the various flow streams and how they impact on potential pollutants. A detailed material balance is given in Appendix 1. It should be noted that this plant contains two gasification sections. SNG is produced in oxygen blown gasifiers (Section 1.1.3) and the power requirements for the plant are met with a low Btu gas that is produced in air blown gasifiers (Section 1.2.4).

1.1 Process Facilities

The Lurgi process has operations similar to other types of coal gasification processes, except for the gasification step itself. The gasification step in each case is peculiar to the process. In general, coal gasification involves getting coal from the mine, storing it, reducing its size to that necessary for gasification, and, possibly, pretreating the coal. The gasifier raw gas is generally processed through a shift reactor which converts carbon monoxide and steam to carbon dioxide and hydrogen. The hydrogen is necessary for a later step in methanation. This shift reaction is only applied to the raw gas if one desires to up-grade it to a synthetic natural gas (SNG) stream. For a low heating value gas, a water gas shift section is not required. In this Lurgi study, the assumption is that the gas will be up-graded to SNG. Following the shift there is a clean-up step to remove from the effluent gas all the H_2S and most of the CO_2 . The acid gases are then taken for sulphur production through a Claus plant or other sulfur recovery process. The last traces of sulfur are then removed from the gas purification product stream in order not to poison the methanation catalyst.

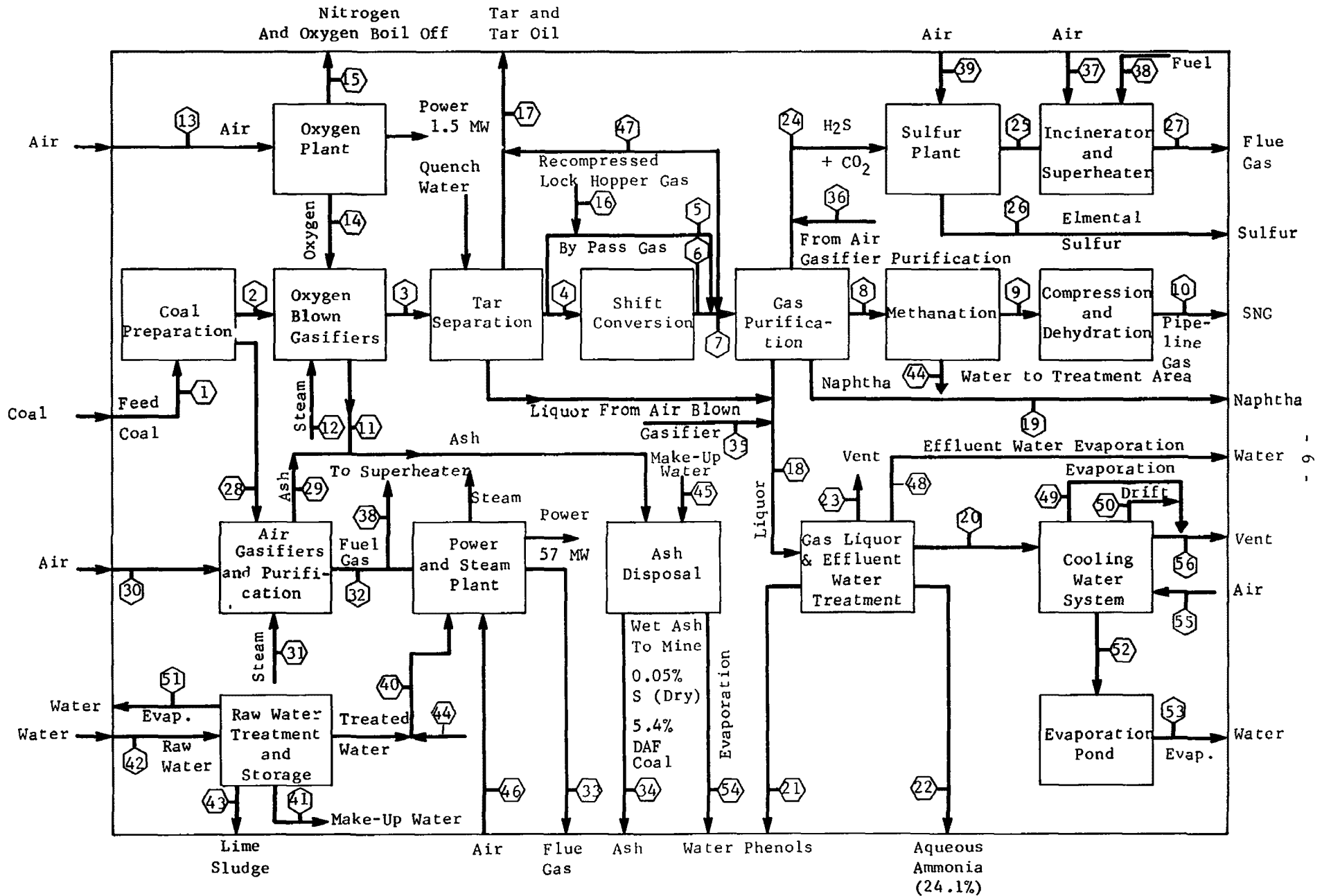
The next step is methanation, where three moles of hydrogen react with each mole of carbon monoxide to produce a mole of methane and a mole of steam. Considerable quantities of CO_2 also react to produce methane. These are highly exothermic reactions which produce a fair amount of the steam required in the plant. Following methanation there is a drying step and the gas is compressed to pipeline pressure.

1.1.1 Plant Site

The plant site for a 250 MM scfd SNG plant should be about 1000 acres and should be close to both a coal mine and a source of water. In general, the ash produced from the coal is returned to the mine for disposal. The coal requirement for the plant in the present study is 26,000 tons/day of Navajo sub-bituminous coal. The coal analysis is given in Tables 1 and 2.

Figure 1

PROCESS FLOW DIAGRAM FOR LURGI DRY ASH GASIFICATION PROCESS



Water make-up requirements are on the order of 59 MM lb/d or 4908 gpm. The plant is designed not to discharge any aqueous effluents.

1.1.2 Coal Storage and Pretreatment

The coal storage part of the plant does not involve coal cleaning, gangue removal or primary screening. All of these operations are assumed to have taken place at the mine. The coal from the mine is transported to the gasification plant by a continuous belt conveyor.

The typical properties of the Navajo Sub-bituminous Coal used in the design are given in Tables 1 and 2. (Also included in Table 2 are analyses by the Illinois Geological Survey furnished by EPA, of Navajo County Red Seam Coal.) The higher heating value (HHV) used in the design is 8872 Btu/lb of coal.

The sub-bituminous coal delivered to the gasification plant is crushed to 1-3/4" x 0. Six storage areas are used for stock piling. Each area is 1,750 ft. long x 124 ft. wide and contains roughly 120,000 tons of coal. Coal from the various storage piles is blended prior to feeding it to the gasifier in order to achieve proper heating value control (Btu control). An emergency stock pile and re-claiming facility are available to provide an additional 650,000 tons of coal. This will provide a 25 day supply of coal in cases of emergency.

A secondary screening facility is present at the gasification plant. The 1-3/4" x 0 coal is screened to produce two gasifier feed sizes (1-3/4" x 5/8" and 3/8" x 3/16"). Two sizes of coal are used as an economic measure to minimize size reduction and screening operations. All undersized material is conveyed at a rate of about 260 tons per hour to a briquetting plant. Briquettes are fabricated and sized to 1-3/4" x 5/8". The briquettes are mixed with the feed going to the gasifier. The briquetting plant contains mixers, coaters and compactors in order to mix the coal fines with a tar binder. (Revised designs (6) (October 1973) have eliminated the need for a briquetting plant.)

The coal preparation operations which are carried out at the gasification plant should be designed with proper dust control measures (7). Wet scrubber dust collectors should be installed in the screening and briquetting plant to eliminate dust and fuel emissions. Sprays should be used at transfer points for dust suppression. The disposal of the aqueous effluent from these scrubbers is analyzed in Section 2.2 (Water Pollution). The coal piles themselves should be designed and located in such a way as to minimize the dangers of spontaneous combustion (8,9). Other factors associated with rainfall on the coal pile should also be considered in order to avoid acid water drainage (10).

Table 1

NAVAJO SUB-BITUMINOUS COAL (6)

<u>Proximate Analysis</u>	<u>Weight %</u>
DAF Coal	66.2
Ash	17.3
Moisture	16.5
<u>Component Analysis (DAF Coal)</u>	
C	76.72
H	5.71
N	1.37
S	0.95
O	15.21
Trace Compounds	0.04
HHV Range 7500 To 10,250 Btu/lb	

Table 2

TYPICAL TRACE ELEMENT ANALYSIS
NAVAJO SUB-BITUMINOUS COAL (6)

	Trace Elements ppm by weight		IGS Data ⁽¹⁾
	Minimum	Maximum	
Sb	0.30	1.20	0.3
As	0.10	3.00	1.3
Bi	0.00	0.20	---
B	60.00	150.00	17.
Br	0.40	18.00	0.4
Cd	0.20	0.40	<0.2
F	200.00	780.00	39.
Ga	0.50	8.00	1.6
Ge	0.06	0.50	2.
Pb	1.40	4.00	4.
Hg	0.20	0.35	0.06
Ni	3.00	30.00	5.
Se	0.08	0.21	1.2
Zn	1.10	27.00	15.
Be	---	---	0.2
Co	---	---	7.
Cr	---	---	5.
Cu	---	---	22.
Mn	---	---	6.
Mo	---	---	2.
P	---	---	125.
Sn	---	---	<2.
V	---	---	17.
TOTAL	267.3	1023	
Fluorine + Boron	97.3%	90.7%	

(1) Data furnished by EPA from IGS Analyses of Navajo County Red Seam Coal.

Spontaneous combustion of coal is probably caused by the oxidation of the coal substance itself. The oxidation is influenced by such factors as moisture and pyrites. Other factors such as coal size, and in particular, the segregation of fines in the coal pile have a strong influence on the spontaneous combustion of coal. The rate of oxidation of most coals increases very slowly with temperature to about 160°F (11). If conditions of heat dissipation are particularly poor, temperature rises above this point and more rapid oxidation can occur, thus causing further increases in temperature until the coal ignition point is achieved.

In order to avoid spontaneous combustion of coal, certain rules should be followed. The coal should be stored in a pile in such a way as to avoid the segregation of lumps and fine coal. It is not advisable to pile the coal too high since this prevents the escape of heat from the region located in the center of the pile. Storage piles should be kept away from other external sources of heat. For long term storage it is advisable to compact the coal pile so as to avoid local regions where air and coal can interact, and to reduce dusting and wind losses. The temperature of the coal pile should be taken regularly, and if the temperature reaches about 160°F some preventive measures should be taken.

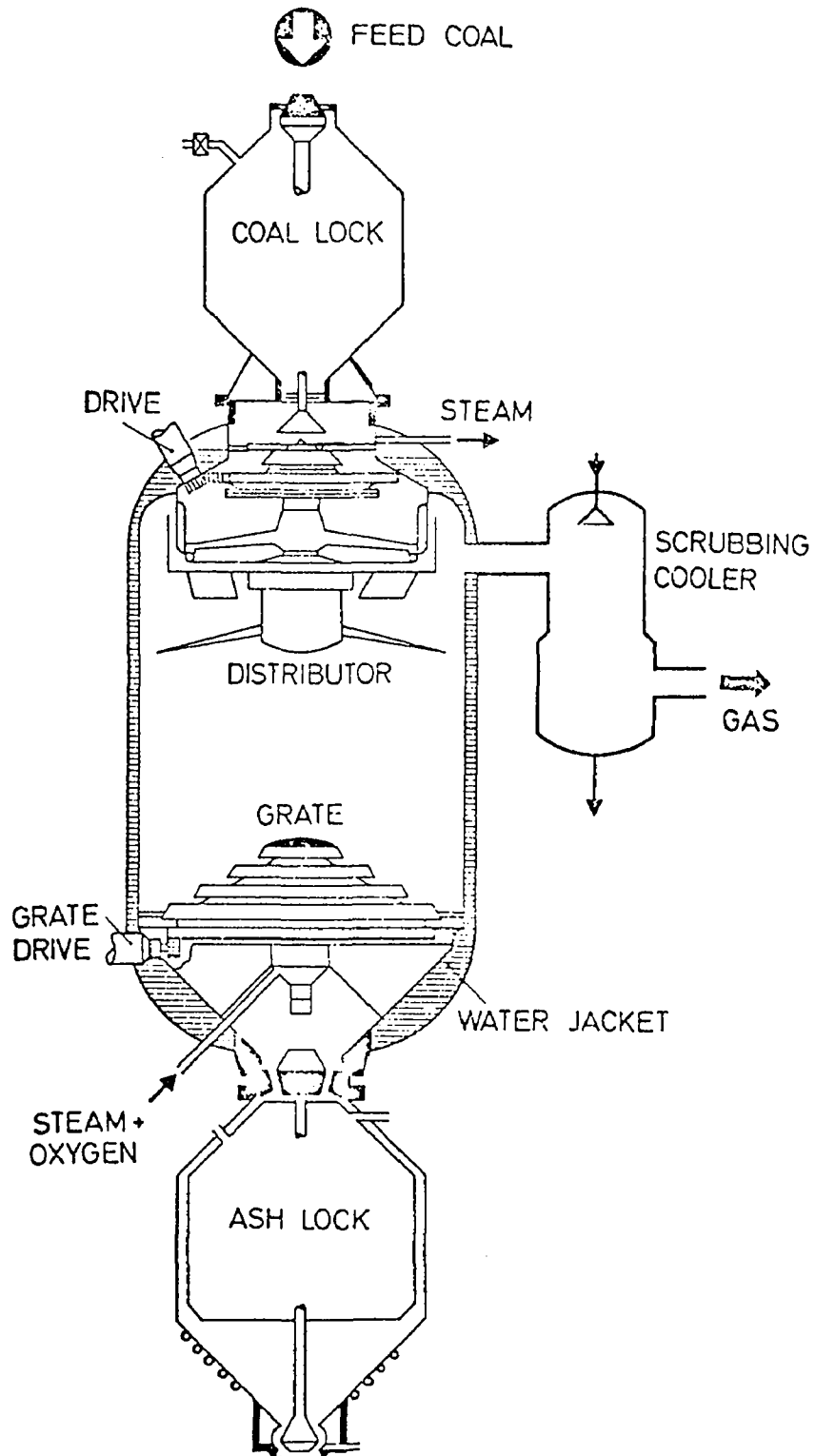
In all solids handling and processing, good housekeeping is essential. It should be a matter of policy in the plant to quickly contain and clean-up spills and leaks. This is generally required by proper safety procedures as well. In the outdoor coal storage and process areas any dust that is not contained can be picked up by the wind and spread promptly over the site. Specific clean-up equipment such as trucks, vacuum pick-ups, and hoses should be provided. Spraying water on the roads and hoses to flush dust to the storm sewer system should be done routinely.

Noise control is another environmental consideration which should be considered in the coal process area. Screening and briquetting are expected to be noisy operations. Most of the noise will be shielded from the public because these operations will be contained in a building. Special precautions will have to be taken to protect the personnel operating in that building.

1.1.3 Gasification

In the Lurgi Process, gasification takes place in a counter-current moving bed of coal at 420 psig. A cyclic mode of operation using a pressurized hopper is used to feed coal (12). The pressurizing medium is a slip stream of raw gas which is later recompressed and put back into the raw gas stream going to purification. The gasifier has a water jacket to protect the vessel and provide steam for gasification. Approximately 10% of the gasification steam requirement is provided in this manner. The internals of the gasifier are illustrated in Figure 2. They include blades to mechanically overcome caking, a moving grate on the bottom to remove the dry ash, and a mechanism to introduce steam and oxygen uniformly over the cross section of the gasifier.

Figure 2
LURGI GASIFIER (12)



In general there are three process zones in the gasifier. The first zone devolatilizes the coal. As the coal drops down it is met with hot synthesis gas coming up from the bottom causing devolatilization, thus removing hydrocarbons and methane from the coal. As the coal drops lower to the second zone, gasification occurs by the reaction of carbon with steam. Finally as the coal approaches the grate, carbon is burned to produce the heat required for the gasification process. The chemical reactions associated with these zones are listed in Table 3.

The top and middle zone temperatures are generally between 1100 and 1400°F, where the devolatilization and gasification take place. The gas leaves the bed between 700 and 1100°F depending on the rank of the coal. The effluent stream for the Navajo sub-bituminous coal will be approximately 850°F (13). The temperature of the ash is kept below the ash fusion temperature by introducing sufficient steam to avoid ash fusion. This is one of the advantages of this type of gasification. It is estimated that 1.41% of the DAF coal is not consumed and leaves with the ash. Thus 5.41% of the dry ash is coal, resulting in an ash sulfur content of 0.05%.

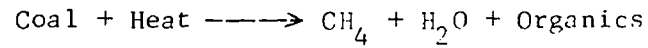
The gas stream leaving the Lurgi gasifier contains coal dust, oil, naphtha, phenol, ammonia, tar oil, ash, char and other constituents. This mixture goes through a scrubbing and cooling tower to remove the tar. The raw gas stream then goes through a waste heat boiler where the raw gas temperature is cooled to about 370°F. The boiler produces 112 psia steam for the Rectisol, Phenosolvan, and Stretford plants. The raw gas composition on a dry basis is as follows: 28.9 % CO₂, 0.32% H₂S, 0.40 % C₂H₄, 19.55 % CO, 38.81 % H₂, 11.09 CH₄, 0.31 % C₂H₆ and 0.32 % nitrogen plus argon. The raw gas stream after cooling is split into roughly two equal parts. Half of it goes through shift conversion to produce additional hydrogen which will be needed for methanation. The other half goes directly to the gas purification system. Any liquid that is condensed in the waste heat boiler and gas cooling section is sent to the gas liquor separation unit.

The coal lock hopper gas is compressed and mixed with the stream that goes directly to purification. This lock hopper gas stream is mixed with other vent streams which contain sufficient quantities of carbon monoxide and methane to warrant its re-introduction into the raw gas stream.

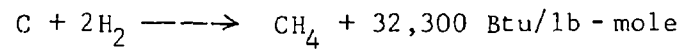
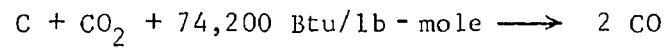
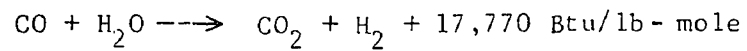
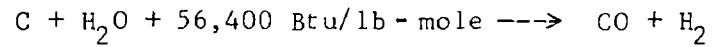
Table 3

CHEMICAL REACTIONS IN LURGI GASIFIER

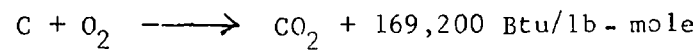
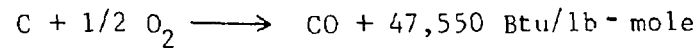
Devolatilization and Drying



Gasification



Partial Combustion

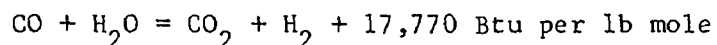


1.1.4 Tar Separation

The water that was used to initially quench the gas as it comes out of the gasifier becomes a gas liquor. The gas liquor cools the crude gas mixture to a temperature at which it is saturated with water. This gas liquor is then flashed, and the tar is removed out of the bottom. The top phase is then sent to water purification. The gas liquor flash tanks will also receive the aqueous effluent from the cooling area prior to the shift reactor. In the gas liquor purification system, dissolved phenol and ammonia are removed for subsequent by-product recovery value.

1.1.5 Shift Conversion

Slightly less than half of the total crude gas is sent to the shift conversion section. The crude gas will be cooled in a waste heat boiler generating steam at about 76 psia. This is the gas that goes to the shift reactor section. The shift reactors are designed to produce hydrogen by the "water-gas shift" reaction. This exothermic reaction has the following stoichiometry:



The shift gas feed is quenched and washed in a countercurrent water tower. The washed gas is heated and passed through a pre-reactor to remove carbon containing residues. The heated gas will be shifted in a series of reactors resulting in 77.2% conversion of carbon monoxide. The equilibrium temperature at which the 77.2% of the CO would be converted in this system is 800°F. Shift reactors generally operate between 700 and 1000°F. The shift section is designed to produce a ratio of over three moles of hydrogen to each mole of carbon monoxide in the total gas stream for methanation. In this design the ratio of H_2 :CO going to methanation is 3.7.

The hot gas liquor and tar which are condensed during cooling in the waste heat boiler are sent to the tar separation units. The product stream from shift conversion is then mixed with the by-pass gas stream from the gasification unit and is cooled and sent to gas purification. Since the shift reaction is fairly exothermic, a fair quantity of heat is recovered prior to the low temperature gas purification step. Heat is also recovered from the crude gas stream that does not go through the shift reactors.

1.1.6 Gas Purification

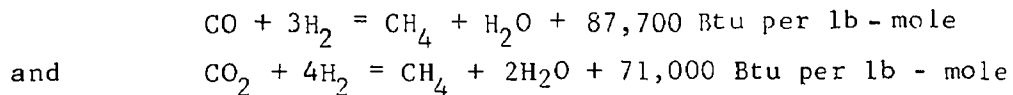
The effluent stream from the shift reactor section is combined with the other half of the raw gas and the recompressed lock hopper gas, and is then sent to the purification system. The mixed gas stream is cooled to low temperature in order to go into the Rectisol system (15).

The Rectisol process is a low temperature methanol wash process which removes acid gases such as H_2S , COS and CO_2 down to a level of about 0.1 vppm. (The process guarantee for Rectisol is 0.2 vppm.) The gas purification system is also used for drying and reducing the CO_2 level prior to final pipeline compression. The efficiency of methanol absorption increases considerably with decreasing temperature. The lowest temperature used in the process is on the order of $-75^{\circ}F$. The first vessel in the Rectisol unit is a prewash tower which strips out naphtha and cools the raw gas. The absorber then removes H_2S and COS down to about 0.1 vppm. Roughly 88% of the CO_2 is also absorbed at this time. The effluent raw gas from the methanol refrigerated absorption column is used to cool the incoming acid gas stream. This sulfur free gas stream is then sent to the methanation area.

All the acid gas streams are combined into a single stream and delivered to the sulfur recovery plant. The sulfur plant stream also includes the carbon dioxide that is removed after methanation. The acid gases from the cold methanol are recovered in a multi-stage operation. The acid gas containing stream is regenerated by step-wise expansion. The last step is a vacuum distillation. The stream to the sulfur plant contains, in addition to the acid gases, a fair amount of product hydrocarbons and carbon monoxide which will ultimately be burned in the incinerator. The Rectisol process is one of the major power consumers in this gasification scheme. About 23% of the power output is used in the refrigeration and compression stages of the process. A mechanical compression refrigeration cycle is used which provides refrigeration at two temperatures: high level refrigeration at $32^{\circ}F$ and $-50^{\circ}F$ which is used for the acid gas treatment. The $32^{\circ}F$ methanol stream is used mostly for removing water vapor.

1.1.7 Methanation

The feed gas leaving the acid gas purification system is preheated with product gas leaving the methanation reaction section. The chemical reactions involved in methanation are



Methanation catalysts are known to be extremely sensitive to poisoning by sulfur (16). The fresh feed is therefore treated with zinc oxide beds prior to exposure to the catalyst. Zinc oxide is known to be an effective remover of trace quantities of sulfur. A fraction of the methanated product is recycled and mixed with the feed to dilute the concentration of reactants in the feed. This type of operation helps maintain the methanation reactors close to equilibrium. The heat of reaction that is generated by the synthesis of methane is removed by converting boiler feed water to process steam. This steam is used for gasification and in other parts of the plant.

The Westfield Lurgi Plant (14) found excessive quantities of nickel carbonyl in its product gas. An active-carbon bed was installed to remove this material. The origin of the nickel carbonyl has not been established. Due to process and environmental considerations, this should be checked.

1.1.8 Compression and Dehydration

The product gas from the methanation reaction section leaves at approximately 225 psia and 800°F. The stream is cooled and is sent to a final product condensate separator. The water is recovered and is sent to the raw water treatment plant. The gas is cooled to 90°F and is then recompressed from 225 to 500 psia. This stream is then sent back to acid gas removal systems for CO₂ and water removal. The effluent from the gas purification system is then sent to the second stage of the compressor where the pressure is boosted to 915 psia to meet pipeline requirements. Air cooling is used to cool the compressor effluent gas prior to delivery to the pipeline. The pipeline gas stream contains 2.01% CO₂, 0.75% H₂, 95.96% CH₄, 0.12% CO, and 1.16% N₂ and Ar. The net flow of gas is 250.7 million scfd. The SNG has a higher heating value of 972 Btu/scf.

1.2 Auxiliary Facilities

In addition to the basic process facilities described above a number of auxiliary facilities are required to make the plant run efficiently and to remove pollutants. These will be described in this section.

1.2.1 Oxygen Plant

Three oxygen plants are required in this process to produce 6,000 tons per day of 98% pure oxygen. Approximately 444,000 scfm of air are compressed to 90 psia with three parallel centrifugal compressors (17). In so doing, the moisture content of the air is condensed and is available for process use.

Assuming an average gas temperature of 60°F and 50% relative humidity, the amount of water removed from the air is 11,190 lb/hr. Of this amount of water, approximately 9,600 lb/hr is available for use in the plant. Although the oxygen plant consumes 2.94 megawatts of electricity, it generates 1.5 megawatts by expansion of the cold nitrogen waste product. The oxygen plant is, of course, a net energy consumer. It uses 25% of the fuel gas produced in the air gasifier to operate the air compressors. (See section 1.2.4).

The oxygen plant effluent stream contains 429 ppm CO₂, 0.2% H₂O, 0.9% O₂, and 98.9% N₂. As this stream evaporates from the cold box it is taken through a gas turbine expander and generates 1.5 megawatts of electrical power. The oxygen stream is compressed to 500 psia and sent to the gasification unit.

1.2.2 Sulfur Plant

The H₂S effluent stream from the acid gas purification system (Rectisol) described in Section 1.1.6, and the H₂S from the acid gas treatment plant (hot potassium carbonate) from fuel gas production described in Section 1.2.4, are sent to a Stretford sulfur recovery plant. The Stretford process (18) was chosen by Stearns-Roger (6) for sulfur recovery in this plant because the total percentage of sulfur in the input stream is only 1%. It is not practical to use a Claus Plant for less than 10% H₂S; capital and operating costs increase drastically as throughput volume increases (19). Roughly, 94% of the sulfur that comes into this unit is removed and high quality elemental sulfur is produced. The effluent stream contains 741 ppm of sulfur as H₂S and COS. (In a later design of the plant (6) the Stretford unit is shown removing 99% of the sulfur.) This stream is combined with fuel gas and is incinerated in the superheater fire box. The overall sulfur balance for the gasification complex is given in Table 4. This sulfur balance does not include very minor streams, such as those that reacted with ZnO in the methanation guard chamber. These are insignificant from the viewpoint of sulfur recovery but are important from a pollution viewpoint.

The acid gas entering the Stretford unit is treated with a water solution containing sodium carbonate, sodium vanadate, anthraquinone disulfonic acid (ADA), citric acid, and traces of chelated iron at 80°F and a pH of 8.5. The H₂S is oxidized by the vanadate to form elemental sulfur. The vanadium, which is reduced by the sulfur reaction, is then reoxidized by the ADA to the pentavalent state. This reaction occurs in the absorber using air as the oxidizing medium. The liquid containing elemental sulfur passes to an oxidizer where ADA is reoxidized by air. The elemental sulfur/air froth overflows to a holding tank. The reoxidized solution is recycled back to the absorber. The sulfur is recovered from the sulfur froth by filtration, centrifugation or floatation. A typical Stretford solution purge contains sodium salts of anthraquinone disulfonate, metavanadate, citrate, thiosulfate and thiocyanate for which acceptable disposal must be arranged.

Table 4
SULFUR BALANCE (1)

<u>Source</u>		
Coal	13601 Lb/Hr	100%
<u>Distribution</u>		
Sulfur Product	13161 Lb/Hr	89.4%
Tar and Tar Oil Naphtha	232	1.7
Naphtha	9	0.1
Ash	192	1.4
Incineration	791	5.8
Power Plant	216	1.6
TOTAL	13601	100

(1) Numbers are rounded off and do not include lesser quantities of sulfur in minor streams.

Overall chemical equations can be written as $2 \text{H}_2\text{S} + \text{O}_2 = 2 \text{H}_2\text{O} + 2\text{S}$. COS goes through the Stretford sulfur production plant essentially unaltered and comes out in the gas effluent. The product sulfur solidifies at ambient temperature and is stored in a curbed storage area. A fair fraction of the Stretford solution must be disposed of daily. This is due to the formation of the dissolved solids that finally build up to such a level that they interfere with the reaction. These solids are primarily sodium thiocyanate and sodium thiosulfate. The thiocyanate is produced from any hydrogen cyanide left in the gas after Rectisol purification. The sodium thiosulfate results from the reaction $2 \text{NaHS} + \text{SO}_2 = \text{Na}_2\text{S}_2\text{O}_3 + \text{H}_2\text{O}$ in the oxidizer.

The properly designed Stretford unit should have provisions for removing the hydrogen cyanide in the gas prior to treating the sulfur gas stream with the absorbing columns (20). If HCN is not removed, then there are two methods of operation that have proven successful (21). One is to keep on making up Stretford feed in order to maintain the concentration of solids at 25%. The other is to allow the concentration of solids to build up to 40% and then dump the complete charge. The disposal of this effluent is a problem. It contains a fair amount of thiocyanate and thiosulfate ions. In view of the large amount of sulfur that leaves in the Stretford gaseous effluent as designed, it might be advisable to add a second stage to reduce the sulfur even further. Stack gas scrubbing may be necessary on the incinerator/superheater. This is discussed in the following section.

1.2.3 Incineration

The effluent stream from the Stretford sulfur plant is sent to incineration. The incinerator superheater fire box consumes about 13.7% of the product gas from the air gasification section. This corresponds to 44.9 MM scfd. This stream which consists essentially of 96% carbon dioxide will have a total flow of 367 MM scfd on a dry basis, and a higher heating value of 29 Btu/scf. Approximately 321 M lb/hr of air will be required to completely burn the Stretford effluent stream. The combined effluents from incineration and superheating come out of a common stack. The flue gas composition will be 62.5% CO_2 , 7.4% H_2O , 295 ppm SO_2 , 76.5 ppm COS, 57.5 ppm NO_x , 0.3% O_2 , and 29.8% N_2 . The total amount of heat input into the incinerator/superheater is approximately 872 million Btu/hr. Thus, the equivalent pounds of SO_2 per million Btu emitted are 1.82. Some flue gas desulfurization method may have to be applied to this gas stream to reduce the level of SO_x to one that is more environmentally acceptable. The NO_x level, on the other hand, would meet the standard of 0.2 lb of NO_2 per MM Btu set for boilers of greater than 250 MM Btu/hr heat input. The superheater is used to make 1100 psia steam to operate the pipeline SNG compressor and the methanation recycle compressor.

1.2.4 Power and Steam Production

The power requirements for the gasification complex are met with a boiler-gas turbine combined cycle fired with a low Btu gas produced in a Lurgi gasifier using air (22). The Navajo coal is gasified at about 285 psig. The method of operating the ten gasifiers (9 on stream and one on stand-by) is similar to that previously described in Section 1.1.3. The raw gas produced goes through a tar separation unit and then through an acid gas treatment section. The raw gas is desulfurized using a hot potassium carbonate system. The H_2S and CO_2 from the hot potassium carbonate system is sent to the Stretford unit and combined with the Rectisol effluent in order to produce elemental sulfur.

The same type of coal preparation mentioned previously is used for this gasification. The lock hopper vent gas is compressed and combined with the raw gas prior to acid gas treatment. In this system, hot compressed air and steam are mixed and introduced through the bottom grate. The ash is removed and combined with the ash from the oxygen gasifier in the ash quench pond. The ash slurry is transported back to the mine for ultimate disposal. Approximately 327 MM scfd of dry fuel gas is thus produced. The fuel gas composition is 5% CO_2 , 220 ppm H_2S , .28% C_2H_4 , 18.8% CO , 24.7% H_2 , 6.4% CH_4 , 0.4% C_2H_6 and 44.4% N_2 . The gas has a higher heating value (HHV) of 230 Btu/scf.

The flue gas is used in a combined cycle operation. Approximately 1/4 of the total gas is sent to gas turbines to operate the oxygen plant compressors. The rest of the fuel gas stream is heated in a fuel gas fired heater prior to going through a fuel gas expander. The effluent stream from the expander is used to fire the fuel gas heater, steam superheater, incinerator, and the power boiler. The fuel gas distribution is given in Table 5.

The flue gas composition coming from the power plant stack (which accounts for roughly 86% of the total fuel gas consumed in the plant) consists of 11.5% CO_2 , 16.6% H_2O , 74 ppm SO_2 , 128 ppm NO_x , 1.3% O_2 , and 70.6% N_2 . The boiler accounts for 2,700 MM Btu/hr, thus producing emission levels of 0.16 lb of SO_2 per MM Btu, and 0.2 lb of NO_x per MM Btu. The flue gas should be kept warm to avoid condensation in the stack or in the immediate vicinity of the effluent.

The overall electrical power balance is given in Table 6 and the plant steam balance is given in Table 7.

Table 5

FUEL GAS DISTRIBUTION

<u>Source</u>	Flow Rate <u>MM SCFD (Dry)</u>	Heat Rate <u>MM BTU/Hr</u>	Distribution <u>%</u>
Clean Fuel Gas (contains 1.4 wt % Tar Oil Naphtha)	326.8	3129	100
<u>Use</u>			
Gas Turbines	82.1	786.1	25.1
Fuel Gas Heater	19.5	186.7	6.0
Steam Superheater	44.9	430.0	13.7
Power Boiler	180.2	1725.4	55.2

Table 6

ELECTRICAL BALANCE

	<u>Consumed</u>	
	<u>MW</u>	<u>%</u>
Coal Preparation	6.65	11.4
Gas Purification	13.20	22.6
Sulfur Recovery	4.10	7.0
Cooling Tower	5.00	8.6
Power Plant	8.07	13.8
Fuel Gas Production	4.23	7.2
Other	17.25	29.4
TOTAL	58.5	100

	<u>Generated</u>	
Oxygen Plant	1.5	2.6
Power Plant	57.0	97.4
TOTAL	58.5	100

Table 7

STEAM BALANCE

<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
	<u>17.5 Psia (221°F)</u>		
Water Gas Shift Deaerator	2908 M LB/Hr	Methanation Waste Heat Boiler	1368 M Lb/Hr
		Shift Waste Heat Boiler	527
		Waste Heat Boiler (O ₂ Blown)	748
		Rectisol	92
		Gasifier Jacket (O ₂ Blown)	173

1.2.5 Raw Water Treatment

Raw water is supplied to a 21 day hold up storage reservoir from a major source such as a lake or river. The capacity of the reservoir is 185 million gallons, and it occupies a site of 28 acres by 30 feet deep. The reservoir serves various functions which include a place to settle silt and provide water for fire control. The reservoir should be lined to avoid seepage (23). The rate of evaporation from the reservoir is 145 gpm. Raw water strainers are placed on the inlet to the pumps going to the raw water treatment section.

Approximately 4900 gpm of raw water are pumped out of the reservoir to the treatment section. An additional 600 gpm are recycled from the methanation reaction and condensate from the oxygen plant. After the water is strained to remove silt, it is pumped to a lime treater where it is treated and clarified. The water in the clarifier is treated with alum and polymers. The effluents from the clarifier are drained to a clear-well where they are temporarily stored. The water from the clear-well is pumped through anthracite pressure filters. Approximately 4500 gpm are sent to demineralization. Of this amount 3900 gpm go in to become feed water for steam production. The demineralization section blowdown consisting of 551 gpm is sent to the ash quench area. Roughly 1/3 of the latter amount of water is taken back to the mine as part of the ash slurry for ultimate disposal. The process condensate aerator is used to remove hydrocarbons as well as carbon dioxide which might be dissolved in the water. The effluent from the condensate aerating vessel is mixed with the demineralizer effluent. The total demineralizer effluent flow rate is therefore approximately 4500 gpm. The pressure filter requires roughly 300 gpm of back wash which is sent back into the reservoir. The reservoir capacity is sized so that all the silt can be collected over the life of the project which is roughly 25 years. A good description of the raw water treatment steps is given in the Betz Handbook (24).

Approximately 2 tons per hour of water treating chemicals will have to be disposed of from the raw water treatment section. Most of these chemicals are sent to the evaporation pond and stored there for the life of the project. Roughly 1000 lb per hour of water treating chemical wastes are chemicals associated with the demineralization section. The demineralization waste stream contains caustic, sulfuric acid and resins. The internal water cooling system also requires chemical treatment.

The plant is designed to use 130,000 gpm of cooling water. This system removes 1170 MM Btu/hr. Water is designed to leave the cooling water system at 75°F and is returned at 93°F. The cooling water make-up requirement is approximately 2.2% of the circulation or 2810 gpm. Most of this make-up is supplied from the effluent water treatment area. The cooling water is supplied by three 5-cell cross-flow cooling towers. The cooling water is treated with chemicals in order to control corrosion, scale formation, plant growth and pH (25). The cooling towers are designed for a wet bulb temperature of 67°F,

Table 8

WATER BALANCE

Consumed

Reaction	1971 GPM	31.7%
Evaporation	3543	57.0
Vent	79	1.3
Drift	260	4.2
Ammonia By-Product	106	1.7
Wet Ash	145	2.3
Fuel and Incineration	108	1.7
TOTAL	6212	100

Supplied

Raw River Water Reservoir	4908 GPM	79.0%
Coal	713	11.5
Produced in Methanation	591	9.5
Oxygen Plant Condensate	0 to 19	--
TOTAL	6212 to 6231	100

allowing an 8°F approach to the designed condition. The cooling tower blowdown, consisting of only 210 gpm, is sent to the evaporation pond. Drift loss from the cooling towers is 260 gpm. The chemicals that are added to the cooling tower include an antifoam package, a biological control package, a scale and corrosion control package, and sulfuric acid for pH control. The overall plant water balance is given in Table 8.

1.2.6 Gas Liquor Treatment and Effluent Water Treatment

The aqueous streams condensed from the coal gasification and gas processing areas by scrubbing and cooling the crude gas stream are called the gas liquor. Gas liquor is collected in one central area coming from gasification, shift, gas purification, and fuel gas synthesis. Before all of these aqueous streams are collected, all of the tar, the tar oil naphtha, and naphtha will have been collected and stored for by-product value. Gas liquor streams will contain all of the ammonia and phenols that are produced in gasification. In addition to these by-products, the gas liquor will also contain carbon dioxide, hydrogen sulfide, trace quantities of hydrogen cyanide, and other trace components.

The incoming gas liquor stream is filtered to remove suspended matter such as coal dust and ash. Disposition of the filtered solid material may be a problem as it will be contaminated with traces of materials from the gas liquor. The liquid is then mixed with an organic solvent (isopropyl ether) in an extractor in order to dissolve the phenol. The Phenosolvan process (26) (Lurgi proprietary process) is an integral part of the gas liquor treatment section. The phenol solvent mixture is collected and fed to solvent distillation columns where crude phenol is recovered as the bottom product, and the solvent as the overhead product. The solvent is then recycled to extractors after removing some of the contained water. The raffinate is stripped with fuel gas to remove traces of solvent which are picked up in the extraction step. The fuel gas is scrubbed with crude phenol product to recover the solvent. Finally, the phenol solvent mixture is distilled in the solvent recovery stripper to produce the crude phenol product, and the solvent is recycled to the extraction step. The solvent free raffinate is heated and stream stripped to remove carbon dioxide, hydrogen sulfide, and ammonia.

The effluent stream from the steam stripper is air cooled and sent to the deacidifier reboiler. The carbon dioxide and hydrogen sulfide coming off the reboiler are recompressed and treated in the Rectisol process. The ammonia is collected as a 24.1 wt % aqueous solution. Some of the vent gas associated with collecting the ammonia in solution is sent to incineration. The bottoms from the steam heated ammonia stripper go to the effluent water treatment section after air cooling.

The effluent water treatment system, biological treatment (biox), (27) is used to reduce the phenol and ammonia concentrations in the effluent from the gas liquor so that the water can be reused as cooling tower make-up. The biox system is also used to treat sanitary sewage discharge and discharge from the API separator. Approximately 2900 gpm of effluent come from the gas liquor treatment area, and 110 gpm come from all the other feed streams. These two streams are treated in series. The first section treats the gas liquor effluent in an aeration basin followed by a settling basin. The second section treats the effluent from the first section, as well as the 110 gpm from all other streams in the same way. Thus, the second treatment area acts as a polishing section for the effluent water treatment plant.

In the aeration basin, air is introduced near the bottom of the tank in order to mix the contents of the tank and maintain a surplus of reserved oxygen. Also, micro-organisms as well as nutrients are introduced to digest the organic material. The mixed liquor from the aeration basin overflows into the second basin. The activated sludge settles to the bottom of the basin and the supernatant liquid is then sent to the polishing aeration basin. The polishing aeration and settling basins operate in the same manner. The sludge on the bottom of the settling basin is collected from both areas. Part of the sludge is returned to the aeration basin as required to maintain biological activity. The rest is sent to the ash disposal area for ultimate disposal to the mine. The purified liquid from the polishing settling basin is filtered and sent to the cooling tower sump.

The relatively low flow rate stream, 110 gpm, that is estimated for all streams other than gas liquor effluent includes water from the API separator, the sanitary sewer system and the storm drain system. Good design practice would dictate that this stream be fed to the biox units from a holding pond in order to provide a fairly uniform quality of water and thus not disturb biological activity. Similarly, in cases of flow disruptions or upsets in the Phenosolvan process and/or the sour water stripper for ammonia, the effluent should not be sent to the biox units in order not to disrupt their biological activity. A stand-by carbon or charcoal bed might be used to reduce the concentration of phenol and ammonia to levels that can be tolerated by biox.

1.2.7 Ash Disposal

Dry ash produced from both the oxygen blown gasifier and the air blown gasifier is quenched with demineralizer blowdown water. The water is used to reduce the ash temperature and to avoid dust problems in transporting the ash. Quenched wet ash is sent from the ash hopper through a drag conveyor to the belt conveyor for ultimate disposal to the mine. Additional ash slurry that is carried with the steam produced in the quench goes to a bin lock condensor as well as to a cyclone separator, followed by a droplet separator, and finally through an ash slurry thickener. The

de-watered ash is then conveyed back to the mine on the belt conveyor together with the ash from the ash hopper. A total of 466,700 lb/hr of wet ash is transferred. Of that amount roughly 73,000 lb/hr is water, 20,000 lb/hr is the equivalent of dry ash free coal, and 374,000 lb/hr is ash. The sulfur content of this material is approximately 0.05%. In addition to the ash, some spent chemicals and sludge from the water effluent treatment plant are also sent to the mine for burial. The total quantity of additional material will not add more than 0.5 wt % to the mass going back to the mine.

2. ENVIRONMENTAL CONSIDERATIONS

The potential pollution problems associated with the Lurgi dry ash gasification process are analyzed in this section. Where applicable, technically feasible alternatives are suggested. This part of the report is subdivided into three sections. These sections include a detailed analysis of air, liquid, and solid effluents for the plant as a whole. They also illustrate that most of the known potential pollution problems can be handled. Due to lack of data on the potentially harmful effects of trace elements, only a general discussion is presented. The areas where additional technical information is needed to assess the pollution potential will be discussed in a later part of this report.

2.1 Air Emissions

This section deals with the environmental aspects of process and utility effluents that end up in the air. The sources of NO_x and SO_x emissions are described, and quantitative estimates of their levels are made. Emissions of particulates and trace elements are also estimated but no quantitative estimates can be deduced with the presently available information. Other air effluents such as carbon monoxide and hydrocarbons will be discussed briefly. Figure 3 summarizes the gaseous effluents from the process.

2.1.1 Oxides of Nitrogen

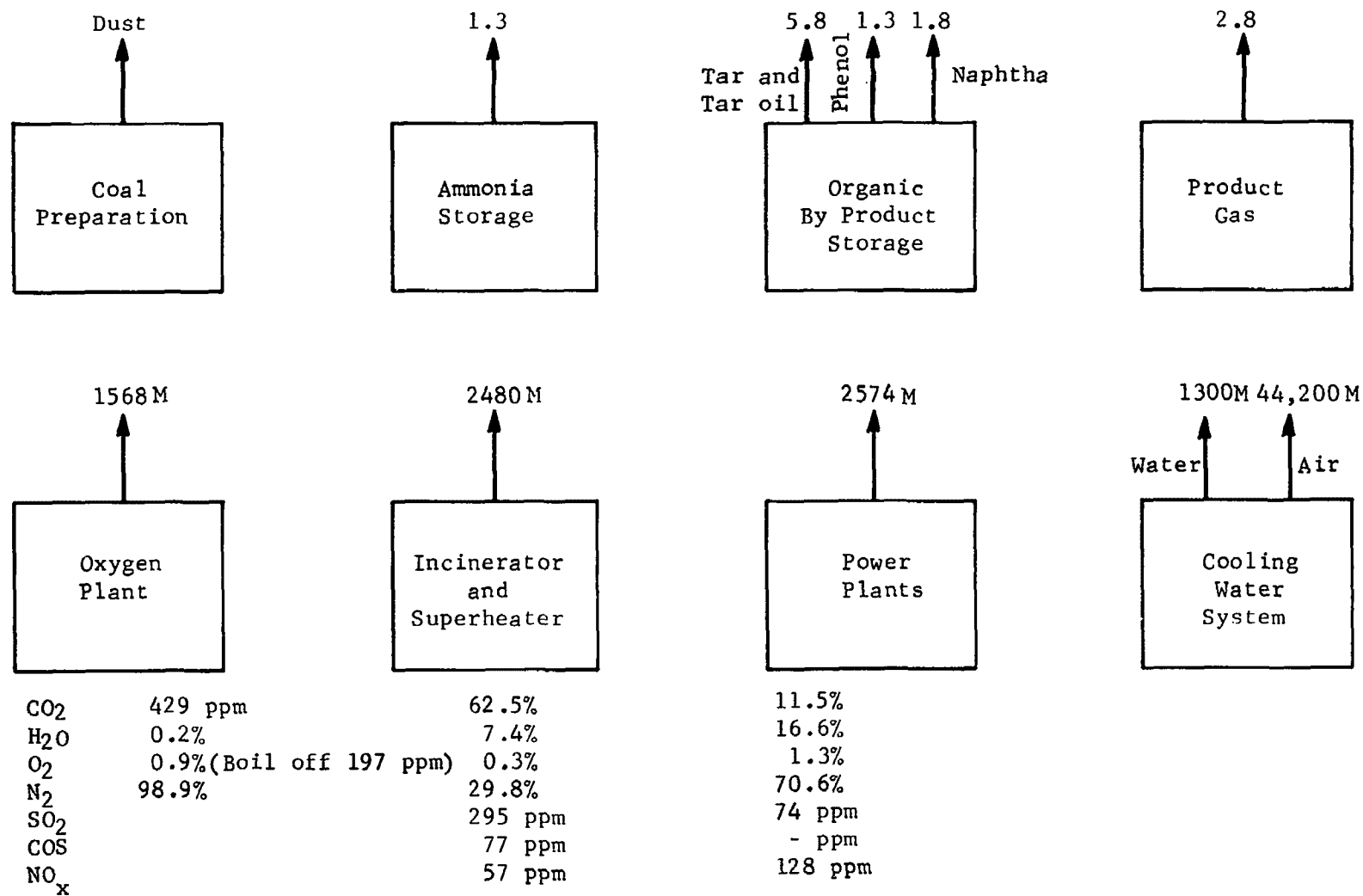
Oxides of nitrogen (NO_x) are produced in fuel combustion processes using air as the oxidizer. At flame temperatures, the combination of atmospheric oxygen and nitrogen results in the formation of nitric oxide (NO). The rate of NO formation and decomposition is highly temperature-dependent. Organic nitrogen compounds present in the fuel provide another source of NO in combustion processes. Based on experimental evidence, the role of fuel nitrogen appears to vary from being the dominant source of NO at low combustion temperatures to being of lesser importance at high temperatures. Recent experimental evidence (28) indicates that in pulverized coal combustion, over 90% of the NO_x is produced by chemically combined nitrogen in the fuel.

The sources of the oxides of nitrogen in the Lurgi dry ash gasification plant are the superheater/incinerator, the power plant and associated equipment such as the fuel heater, and gas turbines. Approximately 176 lb/hr of NO_x are emitted from the superheater/incinerator. This quantity of NO_x meets the regulation promulgated by EPA on December 23, 1971 for new fossil fuel fired steam generating units of more than 250 MM Btu/hr heat input (29). The standard is 0.2 lb MM Btu of heat input when gas fired (2 hour average). Similarly the power equipment produces 537 lb/hr of NO_x measured as NO_2 . Substantial reductions in NO_x can be accomplished by combustion modification techniques described by Bartok et al. for boilers (30) and by Shaw for gas turbines (31). The NO_x effluent from power equipment also meets the 0.2 lb NO_2 MM Btu EPA standard.

Figure 3

AIR EFFLUENTS

(in pounds per hour)



Estimates were also made of the quantities of oxide of nitrogen that would be emitted if the power plant of the gasification complex were to use coal directly rather than gasifying it and using the gaseous fuels. These calculations were done primarily to estimate the increase in thermal efficiency if coal were used directly. This is more fully described in Section 3. If coal were burned for power, the estimated amount of NO_x expected would be on the order of 1,000 lb/hr. Most of it would be due to the conversion of the nitrogen contained in the coal (28). Roughly, this quantity would meet the EPA standard of 0.7 lb/MM Btu heat input for coal firing. The only other source of oxides of nitrogen in the plant would occur in the case of a plant upset where a large portion of product fuel would be burned in the plant flare system.

2.1.2 Sulfur Emissions

The SO_x emissions coming out of the boiler, gas turbines, and other combustion equipment will be on the order of 0.16 lb SO_2 /MM Btu. This amount is well within the tolerance allowed for steam generating plants. On the other hand, the amount of sulfur oxides being emitted out of the incinerator/superheater stack will be 1.82 lb SO_2 /MM Btu. This amount is in excess of the EPA standards for even a coal burning plant which has a standard of 1.2 lb/MM Btu (30). It is not clear at this time whether emission standards in coal gasification plants should be based on the heating value of the coal or that of the gas. In the same vein the allowed emission levels for gas are more restrictive than those for coal. By combining the heat input into the gasifier and boiler, and combining the sulfur output from both stacks, the emission level becomes 0.55 lb SO_2 /MM Btu. This number is less than half the national standard for coal fired utility power plants. As mentioned previously, an additional stage in the Stretford Plant would reduce the SO_2 emissions from the superheater incinerator stack to a level comparable to that coming out of the boiler stack. The latest design of the plant (6) claims about 99% sulfur removal in the Stretford process. If this efficiency is achievable then one stage of Stretford would be adequate.

The oxides of sulfur emissions, if Navajo coal is used to fire the power plant and superheater, would be over 100 M lb SO_2 /d. This would result in an emission level of 1.42 lb SO_2 /MM Btu. This amount is above the national standard of 1.2. Thus, in order to use coal, a desulfurization technique would have to be used to clean the flue gas. An alternate approach to reduce SO_2 emissions would be to burn a smaller amount of coal in the power plant and make up the difference in heat requirement with gas coming out of the purification system. Since the heating value of the gas from purification is 415 Btu/scf, about 28 MM scfd or about 10% of the total production would be required in the power plant to give 1.2 lb SO_2 /MM Btu. Alternatively, the by-product tar, tar oil naphtha, and naphtha can be burned to reduce the pollution from coal burning alone. The overall gasification efficiency would also improve this way. The economics associated with using the liquid by-products as fuel as opposed to their sale as chemical raw materials must be considered.

It is interesting to compare the flue gas composition predicted for coal combustion in the boiler with that which has been observed for similar types of coal in the Four Corners Plant of Arizona Public Service Co. (32). The predicted flue gas composition are 13.8% CO₂, 281 ppm CO, 3.4% O₂, 73.2% N₂, 9.3% H₂, 537 ppm NO₂, and 652 ppm SO₂. The emission levels of the Four Corners Plant were approximately the same for the same amount of oxygen in the flue gas. The Four Corners plant effluent levels were 741 ppm NO_x and 788 ppm SO₂. There is an approximate 20% variation between the produced emissions and those reported by Crawford (32). This difference can easily be accounted for by variations in the coal.

Arrangement should be made to replace the raw product gas in the lock hoppers with nitrogen or CO₂ before filling them with coal in order to prevent the escape of raw product gas containing H₂S to the atmosphere. The raw gas can be incinerated without increasing the SO₂ emissions significantly, or can be compressed and returned to the main gas stream.

2.1.3 Particulates Emissions

The particulate composition from coal combustion generally consists of about 40% silica, 30% alumina, and 10% iron oxide (33). The size distribution of these emissions is on the order of 90% less than 100 microns, and 30% less than 10 microns for pulverized fuel furnaces (33). The levels of particulate emissions from all stoker type boilers, other than spread stokers, are on the order of 5 lb/MM Btu (uncontrolled) (33). The actual level of particulate emissions is subject to wide fluctuation depending on ash content of the coal, heating value of the coal, and method and rate of burning the coal.

The present Lurgi plant is designed to minimize particulate emissions. The power plant and incineration/superheater sections fire a gaseous fuel, and therefore, few particulates are emitted. The only other potential source of particulate emissions is associated with the solids handling areas of the plant. The coal grinding and screening operations should therefore be enclosed. The coal piles should be protected from the wind. This is generally accomplished by orienting the piles in order to minimize wind pick up, or by erecting wind barriers. Coal leading and dumping operations also generate dust. In order to minimize particulate emissions one must anticipate potential dust and particulate sources. Wet scrubber dust collectors should be installed in the screening and briquetting plants to eliminate dust and fumes. Dust suppression sprays should be used as required at all coal transfer points. Similarly, dust collection/suppression facilities should be added to all coal storage bunkers and ash locks. Major roads and parking areas should be hard surfaced to suppress dust. Unpaved areas should be sprayed periodically to reduce dust. All piles should be oriented properly to keep dust levels down.

2.1.4 Other Pollutants

A number of miscellaneous air pollutants are also expected to be emitted in very low concentrations. Among these are carbon monoxide, hydrocarbons, ammonia, and hydrogen fluoride. Carbon monoxide generally results from inefficiencies in the combustion process. The level of carbon monoxide is not expected to exceed 0.02 lb/MM Btu (34). Hydrocarbons are emitted to the atmosphere due to incomplete combustion and from leaks in hydrocarbon by-product transfer and storage. The level of hydrocarbons emitted due to incomplete combustion is not expected to exceed 0.007 lb/MM Btu measured as methane (34). The emissions of ammonia to the atmosphere will be associated with the effluent water treatment process that is discussed in Section 2.2.1. Hydrogen fluoride is generated from the trace of fluorine, probably as an inorganic compound, found in the coal. The hydrogen fluoride is expected to follow ammonia into the aqueous waste stream. Very little HF is expected to go into the atmosphere. Hydrogen fluoride will therefore be discussed along with water pollution.

Large quantities of water vapor will also be emitted from this plant. Water per se is not a pollutant but can cause some environmental problems during certain parts of the year when the water might be conducive to fog formation or through its reaction with other emissions such as SO₂. In the winter these large quantities of water might condense and cause icing problems. The power plant flue gas is expected to produce about 273 M lb/hr of water. The incinerator emits about 89 M lb/hr of water. Another 2 MM lb/hr of water are lost through evaporation, venting, drift losses, etc. Drift losses will carry along any trace materials present, while venting and water evaporation can lead to loss of volatile compounds.

In addition to the hydrocarbon emissions from incomplete combustion, there are numerous sources associated with transportation and storage of products. Leakage of hydrocarbons through heat exchange equipment leads to emissions from cooling towers. Hydrocarbon emissions are found near the seals of moving equipment such as pumps and compressors. Valves generally leak a small amount of hydrocarbons. A major source of hydrocarbon emissions is associated with by-product storage. Estimates were made of the emissions in this Lurgi design using API suggested methods (35) due to leakage and storage. The emission rates are:

Crude Phenol	1.3 lb/hr.
Tar Oil Naphtha	2.3
Tar	3.5
Naphtha	1.8
Methanol	1.4
Ammonia	1.3
Product Gases	2.8

2.1.5 Trace Elements

Pollution by toxic metals and their potential health effects are rapidly causing public and governmental concern. Even at trace levels, certain of these metals have received a great deal of attention in the popular press.

In accordance with the Clean Air Act Amendments of 1970, the Environmental Protection Agency has listed mercury and beryllium as hazardous metallic pollutants. On March 30, 1973, the EPA set national emission standards for asbestos, beryllium, and mercury, the first three air pollutants designated hazardous to health. In addition to these pollutants, other elements about which there may be concern include: Cd, As, V, Mn, Ni, Sb, Cr, Zn, Cu, Pb, Se, B, F, Li, Ag, Sn and Ba.

In addition to the metals present as elements or inorganic compounds, trace stack gas constituents may also be in the form of organometallic compounds. Finally, organic compounds of the heavy, condensed ring aromatic type that are either present in the fuel or that may be formed in the course of the process, can also contribute to the emission of trace pollutants in fuel conversion.

The exact fate of the trace elements present in the coal during the gasification process may vary with the operating conditions and also with the ratio of trace elements present in any one stream. Calcium, for example, may be present in the ash in minor or major amounts, and its amount relative to the sulfur present has a major effect on the form in which calcium, sulfur, and oxygen appear in the final ash emitted. Similar interactions are known or suspected for certain potentially hazardous elements in the list such as arsenic and selenium. In this case, the presence of a large or small amount of one potentially toxic element may substantially affect the amount of another potentially toxic element emitted to the atmosphere or retained in the ash from the gasifier. The balance between alkali and alkaline earth elements, and trace elements whose oxides are acidic, is also expected to be particularly important in this connection.

The emission levels of trace elements from Navajo coal are very difficult to anticipate. In general, one would expect that most of the trace elements would be retained in the ash and thus disposed of back in the mines. Some of the more volatile trace elements, such as mercury, selenium, and others could conceivably go overhead and end up in the water stream. Some of these trace elements can be adsorbed on particulate matter and be removed with particulates. Alternatively, these materials could also be retained as adsorbed matter on the surface of the various processing vessels associated with gas treating. The range level of trace elements that can be produced in the Lurgi plant is listed in Table 9 on the following page.

Table 9

RANGE OF TRACE ELEMENTS

in lb/hr

<u>Trace Elements</u>	<u>Minimum</u>	<u>Maximum</u>
Antimony	0.65	2.6
Arsenic	0.22	6.5
Bismuth	0.00	0.43
Boron	130	324
Bromine	0.86	0.9
Cadmium	0.43	0.86
Fluorine	432	1690
Galium	1.1	17
Germanium	0.13	1.1
Lead	3.0	8.6
Mercury	0.43	0.76
Nickel	6.5	65
Selenium	0.17	0.45
Zinc	2.4	58
Total	578	2212

As can be seen from Table 9, 91 to 97% of all the trace elements can be accounted for by boron and fluorine. No directly relevant study has been made of the fate of trace elements in a Lurgi gasification plant. One is therefore forced to rely on the data of other experimental studies regarding the fate of trace elements. Two recent studies, one using samples from the Hygas bench scale pilot plant (36) and the other of the TVA Allen Plant (37) indicate that sampling, as well as chemical and analytical procedures, are major obstacles for accurate material balances for trace elements. Table 10 indicates the percent disappearance (removal from the remaining solids) of some of the trace elements after various steps in the process. Note that between 85 and 97% of the mercury is not accounted for in these two plants. In a similar manner, selenium, arsenic, and lead could not be accounted for. A recent study on the levels of airborne beryllium due to coal combustion (38) indicated that a maximum of 16% of the beryllium in the coal could be accounted for in the fly ash. The level of beryllium one mile from the Hayden Power Plant where the test was conducted, was a factor of two to four higher than normal background. It was concluded from this study that the rise in background beryllium was unquestionably due to the Hayden Power Plant.

Table 10

PERCENT DISAPPEARANCE OF TRACE ELEMENTS

	<u>Hygas Bench Scale (36)</u>			<u>Sum</u>	<u>TVA Allen Plant (37)</u>		
	<u>Preheater</u>	<u>Hydrogasifier</u>	<u>Electrothermal</u>		<u>Slag Tank</u>	<u>Precipitator</u>	
	<u>430°C</u> <u>1 atm</u>	<u>650°C</u> <u>74 atm</u>	<u>1000°C</u> <u>74 atm</u>			<u>Eff.(1)</u>	<u>Unaccounted(2)</u>
Hg	30	48	19	97	87	?	85
Se	41	21	12	74	71	60	58
As	22	25	18	65	97	95	64
Te	36	18	9	63	--	--	--
Pb	25	19	19	63	99	98	51
Cd	24	23	14	61	65	96	-8.5
Sb	13	7	13	33	97	96	71
V	-9	18	21	30	70	99	24
Ni	8	8	8	24	58	92	39
Be	-9	7	21	19	67	98	69
Cr	-13	7	7	1	40	98	31

(1) Efficiency of trace element collection.

(2) Difference between trace element quantity entering with coal and that accounted by the precipitator and slag tank.

A series of studies using different sized burners were reported by Schultz et al. (39). The studies indicated that the maximum emission of mercury was 50% of that contained in the coal if the pyrite fraction was removed prior to firing. Lead and cadmium were accounted for to a larger extent than mercury in the fly ash. Roughly 30 to 40% of these elements were not accounted for and were presumed to be emitted with the gaseous effluent. Schultz also pointed out the need to exercise great experimental care in doing trace element analysis since the handling procedures could add to the concentration of trace elements.

Another recent study (40) found mercury levels of 0.1 to 0.7 ppm by weight in the coal supplied to a 5.5×10^6 lb/hr steam generator. This study employed aromatic stripping voltammetry, plasma emission spectroscopy, and neutron activation methods for Hg analysis. Mercury balances obtained by analyzing the coal, bottom, hopper, fly ash, flue gas, and water leaving the plant were deficient by as much as 50% but averaged within 10% for the study. This study emphasized the need for reliable sampling techniques, and concluded that 90% of the mercury in the coal fired is emitted as vapor.

Trace elements can cause operational problems, even if properly contained from an environmental point of view. Janeson (41) recently reported that alkali metal compounds from gasification of coal tend to cause hot corrosion and fouling problems in gas turbines. The study concluded that chlorine present in the coal promotes alkali release by forming alkali chlorides. The chlorides react with sulfur compounds at gas turbine combustion temperatures to form sulfate deposits.

A recent study has given some indication that fine grinding followed by selective oil agglomeration can significantly reduce the level of trace metals in feed coal (42). Elements that are organically bound to the coal tend to remain with the feed coal stream. Thus, barium, beryllium (43), boron, germanium (44), mercury (45), selenium, titanium, and zirconium tend to remain with the agglomerated product. Clearly, additional studies are essential to delineate the fate of the trace elements. Parallel studies are needed to define more clearly what the maximum allowable levels should be in order not to create an environmental hazard where none exists. In view of the relatively large number of Lurgi plants in world wide operation, it would be highly desirable to determine the distribution of trace elements in the various parts of the process in operating plants.

2.2 Water Pollution

The handling of the process and cooling water stream can represent one of the major pollution problems in an SNG plant. For economic and other reasons many gasification plants are seriously considering recycling all process water to extinction. The SNG plant water treatment systems will have to be designed specifically for each plant. No one process will be universally applicable. The variety of coal sources and gasifier operating conditions will differentiate the aqueous wastes in the various processes under development.

Water treatment technology has been historically divided into primary, secondary, and tertiary treatment. Primary treatment is usually done first and is designed to remove much of the suspended solids and BOD. The conventional operations in primary treatment, sometimes called clarification, are coagulation, flocculation and sedimentation. Secondary or biochemical treatment oxidizes dissolved organic material to reduce BOD by about 90%. Tertiary treatment involves treatment of pollutants with lower BOD. The operations involved in tertiary treatment have, in general, not been used commercially for more than 5 years. The processes included in tertiary treatment are listed in Table 11 (46).

The Lurgi plant is designed for zero water effluents. Thus, all the pollution that can be carried by the water will be retained at the plant site. Overall water balance is given in Table 8. Roughly 80% of the total water make-up comes from the river and only about 5% of the total water consumed leaves the plant as part of the wet ash and in the by-product ammonia solution. Essentially all of the organic by-products are removed through various stages in the process (obviously some trace amounts remain). Finally, the soluble phenols fraction is removed in the Phenolsolvan process (26). Inorganic by-products such as ammonia, hydrogen sulfide, and hydrogen cyanide are treated in fairly conventional sour water treatment processing schemes. Ammonia is steam stripped from the liquor and condensed as an aqueous solution of 24.1 wt. % ammonia. This solution is stored and ultimately sold for its by-product value. Carbon dioxide and hydrogen sulfide are collected from a deacidifier column and are sent through the Rectisol process to the Stretford Plant. The liquid and solid by-products and effluents for this Lurgi plant are summarized in Figure 4.

It might be desirable to have additional storage capacity in the effluent water treatment section to provide hold-up in case of a process upset. There is danger that the levels of phenol or ammonia would be excessive for the biological activity level present in the biox units. Thus, the microorganism population might be exterminated and it could take time to reestablish adequate activity (47). Another procedure for treating such a stream would be to use a tertiary water treatment technique, which should be available on a stand-by basis prior to mixing it into the normal biox feed stream. For best results, the feed stream composition to the biox units should be kept as constant as possible.

Table 11

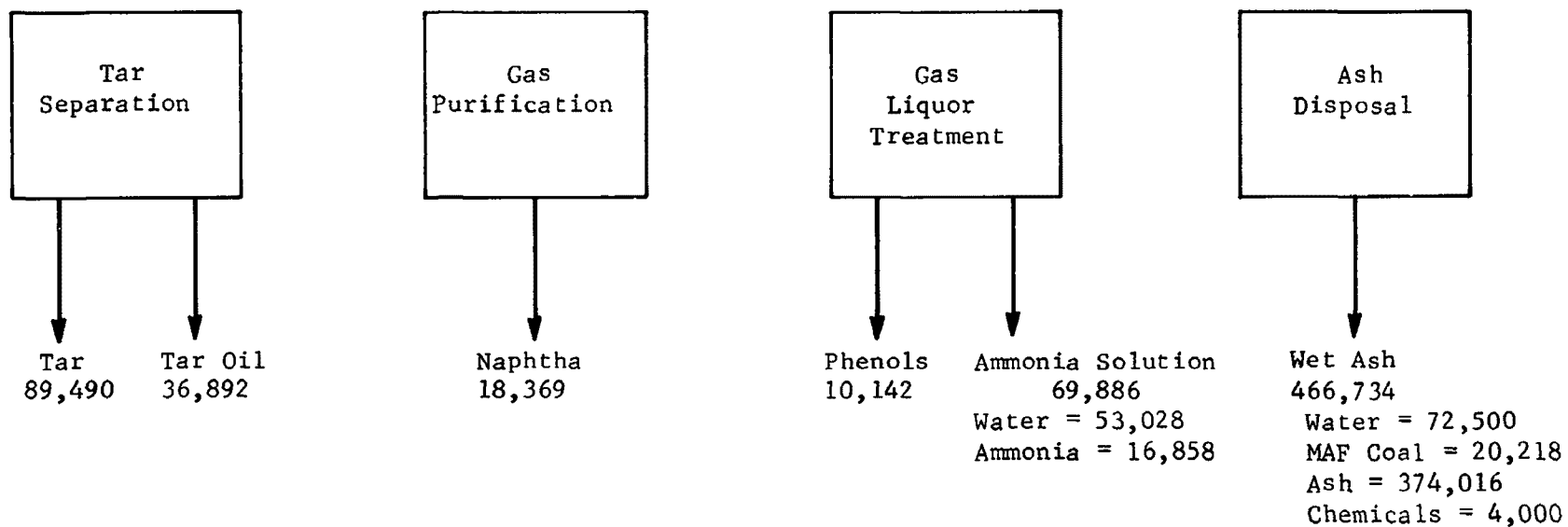
TERTIARY WASTE TREATMENT TECHNOLOGY

<u>Technology</u>	<u>Potential Usage</u>
1. Biological - Carbon Adsorption	Biological Effluent Polishing
2. Carbon Adsorption	Soluble Organics
3. Ozone Oxidation	Taste and Odor Control and Destruction of Other Refractories
4. Evaporation	Organic and Inorganic Separation
5. Ion Exchange	Selected Organic and Inorganic Constituents
6. Reverse Osmosis	Inorganic and Organic Molecules Separation from Water
7. Dialysis	Inorganic and Organic Molecules Separation from Water
8. Precipitation	Phosphate and Metals Removal

Figure 4

LIQUID AND SOLID BY-PRODUCTS AND EFFLUENTS

(in pounds per hour)



2.2.1 Ammonia

Since no process water is returned to the river or to any other water resource, the water treatment methods necessarily relate to purifying the water to process quality. The ammonia that is treated is the residue remaining after by-product ammonia has been removed from the gas liquor treatment section. Trace quantities of ammonia also come from the API separator and from the sanitary sewer sewage system into the effluent water treatment section. Approximately 100 ppm ammonia comes in free and 950 ppm comes in as fixed ammonia. The ammonia is treated first in an aeration basin followed by a settling basin and then through an aeration/settling polishing unit. The effluent from the system contains less than 5 ppm ammonia measured as amines and is sent back to the cooling tower sump. It is interesting that Kostenbader and Flecksteiner (48) indicate that fixed ammonia may not be readily removed by biological treatment and that free ammonia may be removed into the air.

The sour water stripper used to recover ammonia has to be designed to treat certain feed impurities which could cause pollution problems. The major factor in obtaining proper stripper operation is the pH of the feed stream. Impurities such as Cl^- , oil, phenols, mercaptans, cyanides, thiocyanates, and polysulfides can affect stripper capacity and corrode the materials of construction as well as contaminate the products. Oil can cause reboiler fouling and foaming in the tower. If the oil is stripped with the H_2S it could produce a black sulfur product which has a poor sale value. Most of the other impurities are potentially corrosive to the materials of construction.

2.2.2 Phenols

The source of phenol in the water is similar to that of ammonia. It comes from the gas liquor treatment section. The residual concentration of phenol in the water depends on the efficiency of the Phenosolvan process. It is estimated that 500 parts per million phenol enter the effluent water treatment section (biological degradation) and are processed through two stages of aeration and settling ponds. The effluent water contains less than 3 parts per million of phenol and is sent to the cooling tower sump.

2.2.3 Other Aqueous Pollutants

The other aqueous pollutants that are treated by the biological treatment section include fatty acids, BOD_5 , and suspended solids. The fatty acid concentration which starts out at about 1750 ppm (acidic acid) is reduced to less than 9 ppm. The BOD concentration which starts out at 2500 ppm is reduced down to 75 ppm. Suspended solids which are negligible in the inlet stream increase to about 5 parts per million. As mentioned previously, the effluent stream from the biological treatment section (effluent water treatment) is sent to the cooling tower sump.

Pollutants that are not accounted for quantitatively in the water phase include hydrogen cyanide and hydrogen fluoride. The quantities of hydrogen cyanide that are expected to be produced in coal gasification depend on gasification temperature and pressure. At the Lurgi gasification conditions some HCN is expected to be produced and can pass through the SNG system (49). HCN comes in contact with water at a number of points. In the production of metallurgical coke, roughly one percent of the coal nitrogen is converted to HCN. It appears that HCN is produced by the secondary reaction of ammonia with carbon in the reactor. It has been shown that HCN formation is a function of ammonia partial pressure, contact time, and pressure (50). Increased partial pressure of steam suppresses the production of HCN. Hydrogen cyanide will follow the fate of the hydrogen sulfide and is removed in the Stretford process. These quantities of HCN might end up in the water stream. If so, they might have to be treated separately since they can be very detrimental to the biological activity of the effluent water treatment section, especially if levels fluctuate.

Hydrogen fluoride, because of its high reactivity, is expected to react with the calcium oxide, silica, or alumina in ash and ultimately be disposed of with the ash. Any hydrogen fluoride that ends up in the water stream will probably be neutralized by basic minerals that are present there. Small amounts of calcium oxide can be added to neutralize the hydrogen fluoride.

Some coal dust will invariably end up in the waste water stream. Dust from the coal pile as well as dust which is washed in water sprays from the screening operations is carried in the water stream and ultimately ends up in the evaporation pond. It is very difficult to quantify this stream.

The water stream may contain traces of organic materials that are carcinogenic and which are not readily removed by biological treatment. (Only about 90% of the total organic carbon is removed by biological action.) These materials could enter the environment in the water spray from the cooling towers.

Other sources of aqueous pollution such as the chemicals used for regenerating the demineralizers system, will most likely end up in the ash quench and removal section and be ultimately carted back to the mine. The resulting slurry will contain leachable materials. Some solid materials and solid inorganic compounds will end up in the effluent water stream from the Stretford process due to leakage. Quantities are small but disposal may be difficult.

2.2.4 Water Quality Plan

Raw river water is treated conventionally to up-grade its quality to that of boiler feed water. It is filtered, treated with lime, and demineralized. This high purity water is used for steam production as well as for cooling the air and oxygen gasifiers. The biggest source of water consumption is through evaporation and drift losses. These account for approximately 2/3 of the total amount of water lost. The other third is lost by reaction in the gasification steps. Other important considerations for the water treatment part of the plant include a lined evaporation pond which is used to handle aqueous wastes which are not feasible to recycle. In effect, these wastes are stored in the pond for the life time of the project. Oily waste water is treated with an API oil separator and the effluent is sent to the biox units. The lime sludge from the raw water treatment system is sent to the evaporation pond and concentrated there.

In order to conserve water, air cooling is used to dissipate waste heat and thus conserve water. Similarly, cooling tower circulation water will be recycled as much as practicable. The blowdown stream from the cooling tower is sent to the evaporation pond. This stream contains a number of chemicals which are needed to prevent corrosion and the build-up of micro-organisms in the cooling tower. Sludge from the effluent treatment biox units is sent to the mine with the ash. A separate drainage system in the area is used so as not to mix the water resulting from rain and other sources with plant waste streams. Similarly, storm water is diverted to prevent overloading the biox treatment section.

2.3 Solids

There are three major sources of solid wastes that must be considered in the Lurgi plan. These are: ash from the coal, sludge from the biox effluent water treatment section, and chemicals and catalysis that are used in the process and in water treating. Dust from the coal pile has been discussed under air pollutants.

2.3.1 Ash

The total quantities of ash that are expected to be produced from gasification are 314,000 lb/hr from the oxygen gasifier, and 80,200 lb/hr from the air gasifier. The ash contains the equivalent of about 5.4 wt % DAF coal. Thus, 0.05 wt % sulfur on a dry basis is contained in the ash. The two sources of ash are mixed with demineralizer blowdown water resulting in 466,700 lb/hr of wet ash which is sent back to the mine for burial. The burial site for the ash should be such that no trace metals are leached from the ash into the water system. Good quantitative data is lacking in this area although one study (52) has shown that large quantities of minor

elements from spent shale are leachable. One substance in the coal ash that might cause some environmental problems associated with leaching is boron oxide, B_2O_3 . Boron oxide is generally non-toxic in low concentrations, and is in fact a necessary plant nutrient (51). The effect of B_2O_3 at higher concentrations around the vicinity where the ash is buried is not known and should be investigated.

2.3.2 Chemicals

All chemical effluents will be contained in water process streams or in the evaporation ponds. The cooling tower water treatment system will use such chemicals as alum, chlorine, sulfuric acid, sodium hydroxide, ferric chloride, calcium carbonate, corrosion inhibitors, and polymers. The raw water treatment chemical wastes consist mostly of lime sludge which make the water associated with the ash alkaline and thus fix most of the acid wastes. Other sources of solid wastes include catalysts from both the shift and methanation reactors. In general, these catalyst beds are expected to last from 2 to 3 years. A small fraction of the catalyst bed is expected to be replaced yearly with fresh catalyst in order to maintain sufficient catalytic activity. The Stretford solution provides another source of solid wastes. (The reason for replacing the Stretford solution is the limits on the concentration of solids (21).) The general method of operation is to maintain a concentration of 25 wt % solids in the solution, and as the concentration increases a fraction of the solution is blown down. If the concentration ever reached 40 wt % then the whole solution is replaced.

2.3.3 Trace Elements

Some of the trace elements present in the coal are highly toxic (53). For example, lead and arsenic are well known poisons that have caused accidental deaths in industry. Mercury is the most volatile of the trace constituents and is known to cause nerve damage and possibly death. The fate of these trace elements is not known in the gasification plant. Probably the largest fraction of the trace elements will end up with the ash. More volatile elements will be quenched in the tar separation section, thus ending up in the gas liquor system. The likelihood of any of these trace elements becoming part of the synthetic natural gas is very small.

A number of recent studies have indicated that large fractions of trace elements do not end up in the ash (See Section 2.1.5). Unfortunately good material balances were not achieved in all these cases. In order to really determine the fate of trace elements it is essential to do a complete study in which full material balances can be accomplished. Table 10 lists the results of two such studies. Note that the percentages listed in the table indicate the amount of trace constituents that were not accounted for. A negative number indicates that more of the trace element was recovered than was put in.

2.4 Noise

Although most of the coal gasification plants are expected to be in remote areas near coal mines, noise pollution may be a problem. Noise has been found excessive in a plant producing SNG from low molecular weight hydrocarbons (54). Noise control plans should comply with the 1971 Occupational Safety and Health Act (55) and noise control measures should be designed into the system prior to construction (56). Once construction is under way it becomes more difficult to control noise in installed equipment. Gas fired turbines should be enclosed and air and exhaust systems should be properly muffled. Sound absorbing insulation should be placed on piping and equipment as needed while sound absorbing walls and panels should be used in buildings in which size reduction and screening operations take place. The incinerator and boiler should include modern design concepts which reduce combustion noise substantially.

3. THERMAL EFFICIENCY

The overall plant thermal efficiency is an important technical parameter in any fuel conversion process. It explains the quantities of environmentally less acceptable fuels that have to be used to produce environmentally acceptable ones. The heating value of fuel that is consumed must necessarily end up in the atmosphere as a waste product. The thermal efficiency for the Lurgi dry ash gasification process has been calculated in two manners. The first is for the indicated design (6) in which electrical power and steam for plant use are produced from the burning of fuel gas. The second is for a design that assumes that electricity and steam for plant use are produced from direct coal combustion.

As can be seen from Tables 12 and 13 there is only a slight gain in efficiency in burning coal as opposed to producing a fuel gas. This difference might be even less, for some of the fuel gas is used in a combined cycle operation to drive a gas turbine and part is used in a fuel gas expander. This benefit is partially balanced by the air compression necessary for the fuel gas case. Also, no energy debit was taken for flue gas desulfurization in the coal case. The effects of these changes on the overall conclusions are minor.

In order to realistically assess the thermal efficiency, all the by-products were included as part of the effluent stream according to their heating value. Thermal efficiency for producing SNG is 52.9% in the fuel gas case. If one adds the heating value of the tar oils, then the thermal efficiency goes up to 63.1%. If naphtha is included, the efficiency becomes 64.8%, and when crude phenol is added the efficiency becomes 65.5%. The values for adding ammonia and sulfur are also included in Table 12 but are not believed to be realistically useable values. In the case of the coal fired boiler (Table 13) the thermal efficiency, including all of the by-products, adds up to 67.3% and thus the potential advantage of burning the coal directly is only 0.7%. On the other hand, if the objective is to produce SNG then the potential advantage in thermal efficiency is 2.2%.

It should be mentioned that 5 different methods of generating steam and power for the Lurgi gasification process were investigated by Stearns-Roger (6). They concluded that the fuel gas combined cycle technique described in this report was as economical and efficient as any of the other four. They felt that this system was less complex and more reliable than a coal burning unit using flue gas desulfurization. They studied the following cases: low Btu gas fired turbines with heat recovery boilers, low Btu gas fired boilers, medium Btu gas fired turbines with heat recovery boilers, coal tar and tar oil fired boilers with medium Btu gasifier turbines, and tar and coal fired boilers with steam turbine drives.

Table 12

OVERALL THERMAL EFFICIENCY USING FUEL GAS FIRED BOILER

<u>IN</u>	Mass Rate <u>M Lb/Hr.</u>	Heat Rate (HHV) <u>MM BTU/Hr</u>	Cummulative Thermal Efficiency <u>Percent</u>
Coal to Oxygen Gasifier	1722	15280	--
Coal to Air Gasifier	440	3900	--
TOTAL	2162	19180	--
<u>OUT</u>			
Substitute Natural Gas	461	10142	52.9
Tar	89.5	1387	60.1
Tar Oil	36.9	572	63.1
Naphtha	18.4	318	64.8
Crude Phenol	10.1	141	65.5
Ammonia	16.9	164	66.3
Sulfur	12.3	49	66.6

Table 13

OVERALL THERMAL EFFICIENCY USING COAL FIRED BOILER

	<u>Mass Rate</u> <u>M Lb/Hr.</u>	<u>Heat Rate (HHV)</u> <u>MM BTU/Hr</u>	<u>Cummulative</u> <u>Thermal</u> <u>Efficiency,</u> <u>Percent</u>
<u>IN</u>			
Coal to Oxygen Gasifier	1722	15278	--
Coal to Boiler	353	3132	--
Total	2075	18410	--
<u>OUT</u>			
SNG	461	10142	55.1
Tar	67.6	1048	60.8
Tar Oil	36.9	572	63.9
Naphtha	18.4	318	65.6
Phenol	8.7	122	66.3
NH ₃	14.5	141	67.0
S	10.0	40	67.3

It should be pointed out that the thermal efficiency calculated in Tables 12 and 13 are somewhat overstated since the higher heating value of the by-products would not be fully recovered. Part of the heating value would have to be used in achieving water vaporization. The possibility of using coal and liquid by-products to fire the utility boilers could present an alternative. Some, but not all of the undersized coal could be used, thus minimizing the amount of briquetting that would be required. The liquid fuel by-products could also be used in the superheated boiler in order to reduce the sulfur emissions from that unit.

It should be pointed out that the products spectrum of the gasification complex can be shifted depending on demand. Thus, if substitute natural gas is the most desirable product, all the other hydrocarbon liquids could be recycled through the gasifier to increase the yield of SNG. Naturally, there would then be a sizeable debit in overall thermal efficiency, although the efficiency to SNG production would be increased. If the carbon containing by-products are gasified then the overall thermal efficiency would be 59.6% (282 MM scfd SNG), and 61.3% (278 MM scfd SNG) for fuel gas and coal fired power plant respectively. If the carbon containing by-products are fired as fuel in the power plant then the thermal efficiency would be 60.0% and 62.0% for fuel gas and coal respectively.

4. PROCESS ALTERNATIVES

The present design of the Lurgi Dry Ash Gasification Process was examined to assess its pollution potential and to estimate its thermal efficiency. In this section, discussion will center around the potential process improvements which will further optimize the pollution control aspects of the process. This section of the report is subdivided into three parts. The first part evaluates small modifications involving simple design changes to improve pollution control. The second part evaluates certain process improvements which might require some development work. The last part assesses technology needs which might require considerable research and development.

4.1 Engineering Modifications

The Lurgi design evaluated in this report is based on the specific design by Stearns-Roger for the El Paso Natural Gas Company (6). The designer makes full use of the present state-of-the-art in minimizing environmental problems. No major engineering modifications are apparent which will significantly improve the pollution aspects of this design. One of the engineering areas which might require some additional consideration is the acid gas treatment section. This design uses a Linde-Lurgi Rectisol system which is an extremely efficient method of removing acid gases, but is a very high power consumer. In principle, this type of acid gas treatment system should be able to separate the carbon dioxide from the hydrogen sulfide. It is not clear from this design why the two acid gases are not separated, but are sent jointly to the sulfur recovery plant. Two other benefits of the Rectisol system are that naphtha can be separated from the crude gas stream and that methanol also acts as a dryer before final SNG compression.

Two potential alternatives present themselves in lieu of this type of acid gas treatment. First, the Rectisol plant could be redesigned to separate the hydrogen sulfide from the carbon dioxide in order to increase the concentration of H_2S and use a cheaper sulfur recovery process such as a Claus Plant. The Claus Plant would of course need some flue gas treatment facilities. Since Stretford is being used as the sulfur recovery process one could use an alternative acid gas treatment process such as the promoted hot potassium carbonate (57). In addition to a hot potash acid gas treatment section one would also need a dryer to dry the final pipeline SNG stream.

In this design, the cooling water requirements have been minimized by using air cooling as much as possible. Also, production of a low Btu gas using a combined cycle for power generation has allowed that portion of the design to be used very efficiently. A relatively small item in the design involves the use of water scrubbing in areas where coal dust can become a problem. Other techniques for reducing the quantities of coal dust in the area should be considered and might indeed be necessary. For example, electrostatic precipitation or back filtration might be preferable.

4.2 Process Improvements

A number of process alternatives can be discussed which will improve the overall plant efficiency. These improvements, on the other hand, might not optimize the economics of the process. For example, using all the by-products except sulfur and ammonia as fuel for the power plant and the superheater/incinerator combination with about 135,000 lb/hr of coal, would increase the SNG production thermal efficiency to approximately 62% and would meet all the air pollution standards. Alternatively, coal could be used in the plant boilers but flue gas desulfurization and dust removal would be required to meet the environmental standards. Some additional efficiency could be gained by using the coal fines to fuel the boiler since the briquetting plant would not be needed. The third possibility would be to use the coal fines in a slagging type gasifier (58) to produce the low Btu gas needed to fire the power equipment. Thus, the coal fines are utilized in producing a fuel gas.

A second processing improvement which would help reduce the amount of sulfur emitted from the plant would be to use a carbonyl sulfide hydrolysis step. This could be done either prior to the acid gas treatment section (as for example, in the by-pass stream around the shift reactor), or prior to the sulfur plant, since carbonyl sulfide tends to go through a Stretford unit unreacted.

The possibilities of some slightly higher gasification pressures should also be considered. Higher pressure gasification would tend to reduce the oxygen requirements and the ultimate compression debit, thus improving thermal efficiency. Similarly, if the steam input into the gasifier could be reduced by going to somewhat higher conversion, the thermal efficiency would also be improved. Some process improvements are also possible in the area of methanation. There is research going on at this time to optimize a fluid bed methanation reactor (59). This would allow for better heat transfer between the catalyst and the water cooling tubes, and would save energy on gas recycle. Thus, the efficiency and effectiveness of methanation would be improved.

4.3 Technology Needs

One of the principle objectives of the present study is to anticipate potential pollution problems, thus calling attention to any technology gap that might exist. Research and development programs can then be instituted to meet the particular anticipated needs prior to commercialization. In the present Lurgi design a carbonyl sulfide hydrolysis section would be desirable since sulfur emissions from

the superheater/incinerator stack could be reduced by over 1/3. A high temperature raw gas treatment reactor would be very desirable to minimize particulates and potential sulfur corrosion. In terms of thermal efficiency, a high temperature acid gas removal would increase thermal efficiency (60). The need to go from a relatively hot gas down to temperatures below 0°F would thus be avoided.

Highly selective acid gas separation processes would be very desirable since they would reduce the volume of gas that has to go to the sulfur treatment plant and this would reduce the size of the sulfur treatment plant (61).

A high pressure gasification process that could utilize coal fines would remove the necessity for briquetting the fines. This could improve thermal efficiency.

One of the areas of research and development in which information is most lacking is the one that deals with the fate of trace elements. It would seem essential to do complete material balances of trace elements around all the gasification pilot plants that are under development. Thorough studies of analytical techniques as well as sampling techniques are required before the fate of the trace elements can be adequately determined. Similarly, the ash from all the gasification pilot plants should be studied in order to determine its leachability under a variety of conditions that simulate extremes in mine burial. The ability to dispose of the ash in the mine will probably be a function of ash stability.

Other areas where information is lacking include composition of dust and fumes from coal storage, analysis of water run-off from coal storage, and composition of effluents in vapors from evaporation ponds, cooling towers and vents.

5. GLOSSARY AND CONVERSION FACTORS

<u>Abbreviation</u>	<u>Definition</u>
acfm	actual cubic feet per minute
atm	atmosphere - unit of pressure
biox	biological oxidation
BOD	biochemical oxygen demand
Btu	British thermal unit
cal	calorie, thermochemical
cfm	cubic feet per minute
d	day
DAF	dry ash free (usually coal)
°C	degree Celsius (Centigrade)
°F	degree Fahrenheit
°K	degree Kelvin
°R	degree Rankin
ft	foot
gpm	gallons per minute
g	gram
HHV	higher heating value
hr	hour
in	inch
Kcal	kilocalorie
KW	kilowatt
MW	megawatt
MM	million
mol	mole
lb	pound
ppm	parts per million
psi	pounds per square inch
psia	pounds per square inch absolute
psig	pounds per square inch gauge
ROM	run of mine coal
scfm	standard cubic feet per minute (60°F, 14.7 psia)
sec	second
SNG	synthetic natural gas
M	thousand
W	watt

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APPENDIX I

Material Balance For A Lurgi
Dry Ash SNG Gasification Plant

The mass flow rates and stream compositions are given in Table 14. The flow rates were taken from the El Paso FPC application (6) and are presented here with minor modification. The material balance points are numbered according to Figure 1. The Figure is repeated in this appendix for convenience.

Figure 1

PROCESS FLOW DIAGRAM FOR LURGI DRY ASH GASIFICATION PROCESS

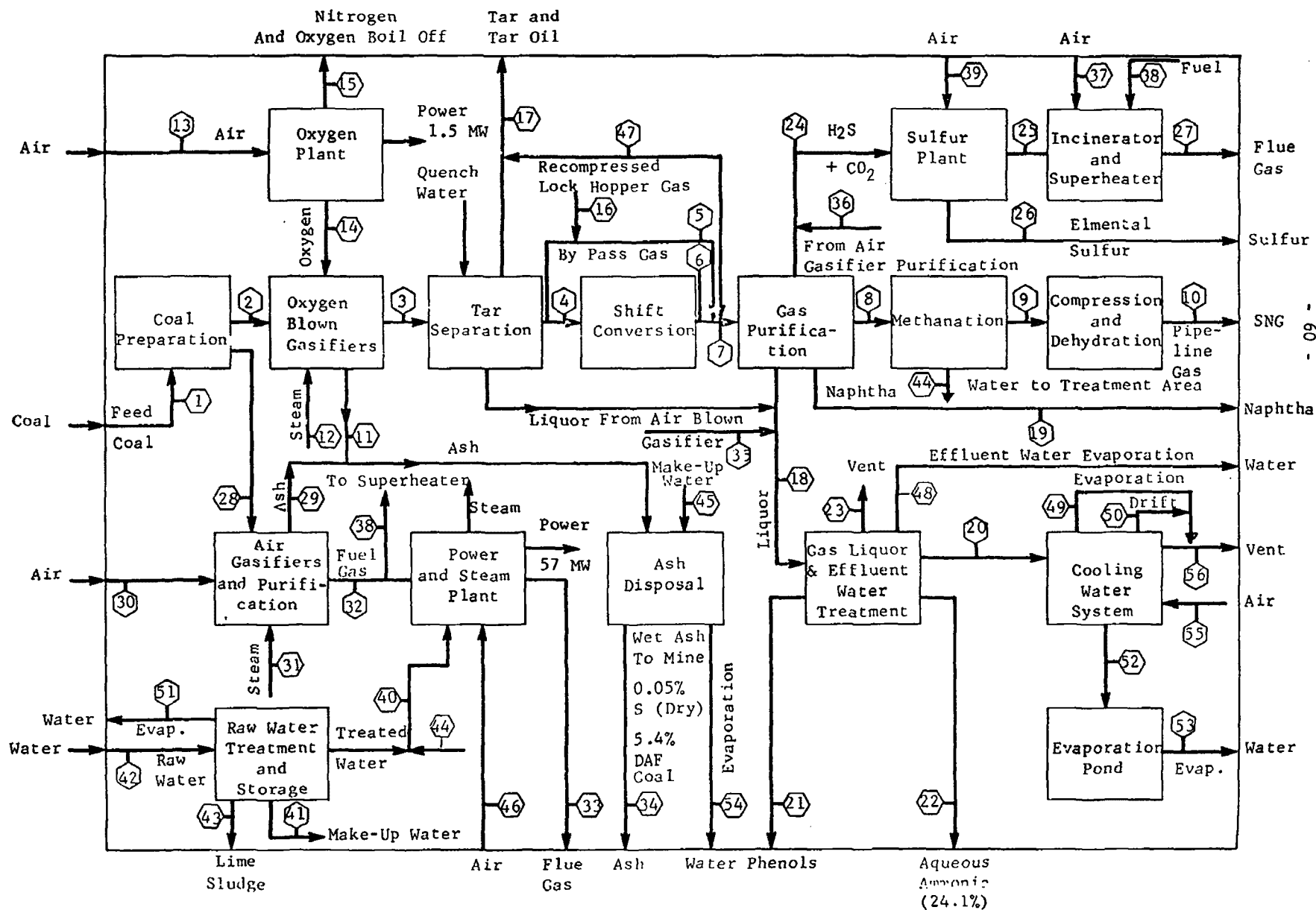


Table 14 (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

MATERIAL BALANCE, LB/HR

Stream Number	51	52	53	54	55	56
Stream Identification	Evaporation From <u>Raw Water</u>	<u>Blow Down</u>	Evaporation From <u>Pond</u>	Evaporation From <u>Ash Quench</u>	<u>Air To Cooling Tower</u>	<u>Air From Cooling Tower</u>
<u>MW</u>						
CO	28.01					
CO ₂	44.01					
H ₂	2.02					
CH ₄	16.04					
C ₂ H ₄	28.05					
C ₂ H ₆	30.07					
H ₂ S	34.08					
COS	60.07					
SO ₂	64.06					
NH ₃	17.03					
N ₂ + Ar	28.00				33,901,400	33,901,400
O ₂	32.00				10,298,600	10,298,600
NO ₂	46.00					
TOTAL DRY GAS					44,200,000	44,200,000
Water	72,560	105,100	363,000	79,100	243,300	1,300,000
Coal MAF						
Ash						
Sulfur						
Naphtha						
Tar Oil Naphtha						
Tar						
Crude Phenol						
TOTAL	72,560	105,100	363,000	79,100	44,443,300	45,500,000

Table 14

LURGI DRY ASH GASIFICATION PROCESSMATERIAL BALANCE, LB/HR

Stream Number	1	2	3	4	5	6
<u>Stream Identification</u>	<u>ROM Coal</u>	<u>Coal to O₂ Gasifier</u>	<u>Raw Product Gas</u>	<u>Shift Feed Gas</u>	<u>Crude Gas to Gas Cooling</u>	<u>Shift Product Gas</u>
	<u>MW</u>					
CO	28.01		535,315	257,821	300,723	58,754
CO ₂	44.01		1,243,519	598,910	752,660	911,971
H ₂	2.02		76,665	36,924	43,001	51,287
CH ₄	16.04		173,954	83,780	100,069	83,780
C ₂ H ₄	28.05		10,973	5,285	6,954	5,285
C ₂ H ₆	30.07		17,937	8,639	10,852	8,639
H ₂ S	34.08		9,960	4,821	6,196	4,821
COS	60.07		220	107	138	107
SO ₂	64.06					
NH ₃	17.03		10,952	5,275	6,157	5,275
N ₂ + Ar	28.00					
O ₂	32.00					
NO ₂	46.00					
TOTAL DRY GAS			2,079,495	1,001,562	1,226,750	1,129,919
Water	356,525	283,975	1,287,646	620,154	710,772	464,526
Coal MAF	1,431,694	1,140,354				
Ash	374,016	297,906				
Sulfur						
Naphtha			18,369	8,847	9,522	8,847
Tar Oil Naphtha			36,892	17,768	19,124	17,768
Tar			27,058	13,032	14,026	
Crude Phenol			4,365	2,102	2,263	
TOTAL	2,162,235	1,722,235	3,453,825	1,663,465	1,982,547	1,621,060

Table 14 (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

MATERIAL BALANCE, LB/HR

Stream Number		7	8	9	10	11	12
<u>Stream Identification</u>	<u>MW</u>	<u>Gas to Purification</u>	<u>Methanation Feed</u>	<u>Methanation Product</u>	<u>SNG</u>	<u>Ash From O₂ Gasifier</u>	<u>Steam</u>
CO	28.01	359,578	351,965	924	924		
CO ₂	44.01	1,625,166	200,378	115,451	24,338		
H ₂	2.02	94,279	93,384	417	417		
CH ₄	16.04	183,836	181,969	430,130	432,714		
C ₂ H ₄	28.05	12,238	5,666				
C ₂ H ₆	30.07	19,471	9,096				
H ₂ S	34.08	11,017	--				
COS	60.07	245					
SO ₂	64.06						
NH ₃	17.03						
N ₂ + Ar	28.00	11,431	11,183	11,183	11,183		
O ₂	32.00						
NO ₂	46.00						
TOTAL DRY GAS		2,317,261	853,641	558,106	460,576		
Water		3,061		1,594			1,762,170
Coal MAF						16,104	
Ash						297,906	
Sulfur							
Naphtha		18,369					
Tar Oil Naphtha							
Tar							
Crude Phenol							
TOTAL		2,338,691	853,641	559,700	460,576	314,010	1,762,170

Table 14 (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

MATERIAL BALANCE, LB/HR

Stream Number		13	14	15	16	17	18
		Air Feed To Oxygen Production	Oxygen	Nitrogen Waste	Lock Hopper Gas	Tar By-Product	Gas Liquor
<u>Stream Identification</u>	<u>MW</u>						
CO	28.01				23,229		
CO ₂	44.01				108,051		
H ₂	2.02				3,260		
CH ₄	16.04				9,895		
C ₂ H ₄	28.05				1,265		
C ₂ H ₆	30.07				1,555		
H ₂ S	34.08				1,057		
COS	60.07				25		
SO ₂	64.06						
NH ₃	17.03				480		
N ₂ + Ar	28.00	1,559,334	10,227	1,549,107			
O ₂	32.00	473,717	458,240	15,832			
NO ₂	46.00						
TOTAL DRY GAS		2,033,051	468,467	1,565,980	148,818		30,140
Water		11,190	--	1,590	43,290		1,490,699
Coal MAF							
Ash							
Sulfur							
Naphtha							
Tar Oil Naphtha						36,892	
Tar						89,490	
Crude Phenol							
TOTAL		2,044,241	468,467	1,567,570	192,108	126,382	

Table 14. (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

		<u>MATERIAL BALANCE, LB/HR</u>					
Stream Number		19	20	21	22	23	24
		Naphtha	Treated	Phenol	Ammonia	Gas Liquor	Sulfur Plant
<u>Stream Identification</u>		<u>Product</u>	<u>Water</u>	<u>Product</u>	<u>Solution</u>	<u>Vent To</u>	<u>Feed</u>
	<u>MW</u>		<u>To Re-Use</u>			<u>Incineration</u>	
CO	28.01						2,056
CO ₂	44.01					8,450	1,695,595
H ₂	2.02						164
CH ₄	16.04						4,111
C ₂ H ₄	28.05						5,686
C ₂ H ₆	30.07						9,445
H ₂ S	34.08						13,571
COS	60.07						306
SO ₂	64.06						
NH ₃	17.03				16,858	860	
N ₂ + Ar	28.00						1,176
O ₂	32.00						
NO ₂	46.00						
TOTAL DRY GAS						9,310	1,732,110
Water			1,406,124		53,028	35	1,896
Coal MAF							
Ash							
Sulfur							
Naphtha		18,369					
Tar Oil Naphtha							
Tar							
Crude Phenol				10,142			
TOTAL		18,369	1,406,124	10,142	69,886	9,345	1,734,006

Table 14 (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

MATERIAL BALANCE, LB/HR

Stream Number		25	26	27	28	29	30
		Sulfur Plant Effluent	By-Product Sulfur	Superheater Flue Gas	Coal To Air Gasifier	Ash From Air Gasifier	Air To Air Gasifier
<u>Stream Identification</u>	<u>MW</u>						
CO	28.01	2,056					
CO ₂	44.01	1,695,595		1,828,643			
H ₂	2.02	164					
CH ₄	16.04	4,111					
C ₂ H ₄	28.05	5,686					
C ₂ H ₆	30.07	9,445					
H ₂ S	34.08	632					
COS	60.07	306		306			
SO ₂	64.06			1,258			
NH ₃	17.03						
N ₂ + Ar	28.00	21,814		554,425			447,241
O ₂	32.00			6,212			135,805
NO ₂	46.00			176			
TOTAL DRY GAS		1,739,809		2,391,020			583,046
Water		8,977		88,806	72,550		3,209
Coal MAF					291,340	4,114	
Ash					76,110	76,110	
Sulfur			12,161				
Naphtha							
Tar Oil Naphtha							
Tar							
Crude Phenol							
TOTAL		1,748,786	12,161	2,479,826	440,000	80,224	586,255

Table 14 (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

MATERIAL BALANCE, LB/HR

Stream Number		31	32	33	34	35	36	37
		Steam To	Clean	Power	Wet Ash	Gas Liquor	Acid Gas	Air To
<u>Stream Identification</u>		<u>Air Gasifier</u>	<u>Fuel Gas</u>	<u>Plant Flue</u>	<u>To Mine</u>	<u>From Air Gasifier</u>	<u>From Fuel Gas</u>	<u>Superheater</u>
	MW							
CO	28.01		188,076				398	
CO ₂	44.01		78,703	460,211			187,487	
H ₂	2.02		17,826				45	
CH ₄	16.04		36,730				91	
C ₂ H ₄	28.05		2,797				25	
C ₂ H ₆	30.07		4,516				27	
H ₂ S	34.08		259				2,554	
COS	60.07		6				61	
SO ₂	64.06			432				
NH ₃	17.03							
N ₂ + Ar	28.00		445,126	1,801,185			1,040	471,699
O ₂	32.00			39,132				144,381
NO ₂	46.00			537				
TOTAL DRY GAS			774,039	2,301,497			191,728	616,080
Water		311,960	45,027	272,668	72,500		1,896	3,391
Coal MAF					20,218			
Ash					374,016			
Sulfur								
Naphtha								
Tar Oil Naphtha			10,806					
Tar								
Crude Phenol								
TOTAL		311,960	829,872	2,574,165	466,734	213,165	193,624	619,471

Table 14 (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

MATERIAL BALANCE, LB/HR

Stream Number		38	39	40	41	42	43	44
		Fuel Gas To Superheater	Sulfur Plant Air	Treated Water To Steam	Treated Water Non-Steam	Raw Water	Lime Sludge	Methanation Water Product
<u>Stream Identification</u>	<u>MW</u>							
CO	28.01	25,766						
CO ₂	44.01	10,782						
H ₂	2.02	2,442						
CH ₄	16.04	5,032						
C ₂ H ₄	28.05	383						
C ₂ H ₆	30.07	619						
H ₂ S	34.08	36						
COS	60.07							
SO ₂	64.06							
NH ₃	17.03							
N ₂ + Ar	28.00	60,982	20,597					
O ₂	32.00		6,212					
NO ₂	46.00							
TOTAL DRY GAS		106,043	26,809					
Water		6,170	148	2,248,000	337,770	2,455,000	90,072	294,235
Coal MAF								
Ash								
Sulfur								
Naphtha								
Tar Oil Naphtha								
Tar								
Crude Phenol								
TOTAL		112,213	26,957	2,248,000	337,770	2,455,000	90,072	294,235

Table 14 (Cont'd)

LURGI DRY ASH GASIFICATION PROCESS

MATERIAL BALANCE, LB/HR

Stream Number	45	46	47	48	49	50
<u>Stream Identification</u>	<u>Water To</u> <u>Ash Disposal</u>	<u>Air To</u> <u>Boilers</u>	<u>Tar Oil</u> <u>Naphtha</u> <u>Product</u>	<u>Evaporation</u> <u>From Biox</u>	<u>Evaporation</u> <u>From</u> <u>Cooling Tower</u>	<u>Drift</u> <u>Loss From</u> <u>Cooling Tower</u>
	<u>MW</u>					
CO	28.01					
CO ₂	44.01					
H ₂	2.02					
CH ₄	16.04					
C ₂ H ₄	28.05					
C ₂ H ₆	30.07					
H ₂ S	34.08					
COS	60.07					
SO ₂	64.06					
NH ₃	17.03					
N ₂ + Ar	28.00	1,417,243				
O ₂	32.00	440,747				
NO ₂	46.00					
TOTAL DRY GAS		1,857,990				
Water	72,500	10,226		87,570	1,170,000	130,000
Coal MAF						
Ash						
Sulfur						
Naphtha			36,892			
Tar Oil Naphtha						
Tar						
Crude Phenol						
TOTAL	72,500	1,868,216	36,892	87,570	1,170,000	130,000

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16. ABSTRACT <p>The report gives results of a review of the Lurgi Dry Ash Gasification Process for high-Btu gas, from the standpoint of its potential for affecting the environment. The review included a process analysis of the process. Waste stream compositions were calculated for a 250 million scfd substitute natural gas plant using a subbituminous coal. Thus, the quantities of solid, liquid, and gaseous pollutants were estimated, where possible. Thermal efficiency was calculated for various process alternatives. A number of process modifications were suggested which would reduce pollution and/or increase thermal efficiency. The report includes an assessment of technology needs to control pollution.</p>		
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