EVALUATION OF COAL CONVERSION PROCESSES TO PROVIDE CLEAN FUELS

Final Report

by

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ABSTRACT

A review is made of six general methods of coal utilization with elimination of the sulfur prior to or during combustion in an electric power generating plant:

 Fluidized Bed Combustion
 Coal Beneficiation
 Pyrolysis
 Coal Gasification
 Coal Dissolution and Liquefaction
 Insitu Combustion

The processes in each category were reviewed, analyzed and evaluated keeping in mind that they would have to be potentially better than stack gas cleaning processes to be considered as a viable alternative to the electric power industry. Critical process steps, where additional research must be done before the processes can be considered at the commercial stage of development, were identified.

A total of thirty-seven processes were reviewed: five in fluidized bed combustion, one in coal beneficiation, three in pyrolysis, twenty-two in coal gasification and six in coal dissolution and liquefaction. The advantages and disadvantages of the processes were identified. Additional processes are known to exist besides those reviewed. Many of these processes were not reviewed because of the severe time restrictions of the study rather than the merits of the
processes. Other processes were proprietary and not actively seeking outside funding.

Five topics, which are important to coal utilization although not actually coal conversion or utilization processes, are included in Part III. They include discussions of combined cycle systems, economics, retrofit capabilities, thermodynamics and coal slurry pipelines. These topics give perspective to the general subject of coal use.
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FOREWORD

The Task Force on Coal Utilization of the Electric Power Research Institute approached The University in March 1973 to assess the various processes which convert coal to clean fuels or utilize coal in environmentally acceptable ways for electric power generation. A preliminary proposal was submitted to Mr. Larry Simpkin on March 30, 1973 suggesting a 14-month study. After review and discussion, we were advised that EPRI's need was for a study to be completed by January 1974. Accordingly, the final proposal submitted on June 6, 1973, was for one-half the time and budget originally suggested.

The University of Michigan Team received oral reports on the stages of approval of the proposal and started preliminary studies on July 16, 1973. The Board of Directors of EPRI approved the proposal on August 15, 1973. The final contract agreement was signed by EPRI on October 24, 1973 and by The University on November 13, 1973. A news release, approved by EPRI, was issued by the University on August 24, 1973 and is given on page 390.

The seven-month study was very intensive; members of the team had teaching responsibilities for a substantial fraction of their time during the study in addition to the project work.
The results of the study of coal conversion processes were divided into two volumes: Part I and Parts II and III. In this way, the recommendations for support to the Electric Power Research Institute (Part I) are separate from the critical review and assessment of the coal utilization processes being developed in the nation (Part II).

Part I was prepared specifically to assist the Electric Power Research Institute management in the preparation of a long term research program in coal utilization on behalf of the electric power industry. Part I contains the choices and recommendations for research support by EPRI. Those processes which seem to have the best prerequisites for providing clean fuels from coal at the earliest dates were delineated. The bases for and the reasoning behind the choices are given.

Part II contains the process descriptions and general evaluations of some thirty-seven processes which are reviewed. The team of investigators reviewed and studied reports and research proposals for the processes. Personal visits were made to the organizations carrying out the research and development and to the sites where the experiments are being conducted at bench, process equipment development unit and pilot plant stages. The organizations were very cooperative in providing information and generous with their time in answering our questions. Several organizations provided further supporting information requested by telephone or letter after our visits.
The description of the processes is intended to give members of the electrical utility industry an overall understanding of processes and is not intended to transfer detailed technical knowledge in a thorough manner. References are cited for the more complete descriptions available to the team. It is from those references that the information given has been extracted. The understanding developed by the team and documented in Parts II and III of this report provided the basis for the evaluation rendered in Part I.

Part III contains several topics which are important to coal utilization although not actually coal conversion or utilization processes. These topics were added to give greater perspective to the general subject of coal use.

The authors for the several process descriptions or sections are listed. Dr. D.E. Briggs managed and edited the final report.

Donald L. Katz
February 1973
ACKNOWLEDGMENTS

We acknowledge the time and thoughtful comments freely
given by the many individuals in the organizations visited
during this project. We appreciate their help sincerely.

We were assisted in this work by the following graduate
students in chemical engineering:

Michael W. Britton
Andre W. Furtado
David E. Hammer
Gerald D. Holder
J. Andrew Stirling
Edward E. Timm

We hope their experience has been as interesting as ours.
PART II

CRITICAL REVIEW AND ASSESSMENT OF COAL UTILIZATION PROCESSES FOR CLEAN FUELS
The general methods for coal utilization with elimination of the sulfur prior to or during combustion in an electric power generating plant are:

- Fluidized Bed Combustion
- Coal Beneficiation
- Pyrolysis
- Coal Gasification
- Coal Dissolution and Liquefaction
- In situ Combustion
- Stack Gas Cleaning

These methods, except for stack gas cleaning, were reviewed. Stack gas cleaning was not within the scope of this study. The processes in each category were evaluated, however keeping in mind that they would have to be potentially better than stack gas cleaning processes to be considered as a viable alternative to the electric power industry.

A total of thirty-seven processes were reviewed: five in fluidized bed combustion, one in coal beneficiation, three in pyrolysis, twenty-two in coal gasification and six in coal dissolution and liquefaction. Additional processes
are known to be under development in some cases, but were not included because they were proprietary and not seeking funds from EPRI or there was insufficient time available to the team for acquisition, review and assessment of the necessary information.

As in all review studies, there is the possibility that the team member did not have all the information necessary for accurate analysis, or he may have misinterpreted the reported results. Thus, readers are urged to refer to the original references when questions arise.

The various categories are treated as separate stand-alone sections. The team members who prepared the individual sections are identified. Often other team members reviewed and suggested amplifications.

A tabulation of the conferences and interviews held during the course of this study is given on the next page.
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<tr>
<th>Organization</th>
<th>Conference Dates</th>
<th>Project Representatives</th>
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<td>Air Products</td>
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FLUIDIZED BED COMBUSTION

by

Donald L. Katz
Brymer Williams

Review and Assessment

Fluidized bed combustion R&D was started to obtain high heat transfer rates in boilers. The work on modular construction by Pope, Evans and Robbins has taken on the added burden of removing sulfur by fluidized limestone along with the coal. Other organizations working in the area include British Gas Council, Argonne, Esso and Westinghouse.

There is ample experience in the process industries to appreciate that fluidized bed combustors have lower volumes and higher heat transfer coefficients than combustion in conventional chambers of entrained solids. Fluidized bed boilers have been operated at atmospheric pressure and at pressures of 5-10 atmospheres to incorporate the boiler in a combined cycle system. Some of the reports available are listed (1-8), visits were made to Pope, Evans and Robbins and the British BCURA project in Leatherhead, Surrey, England.

THE 30 MW RIVESVILLE PLANT

A contract was let to OCR in July 1973 to Pope, Evans and Robbins to include Foster Wheeler for engineering and
construction for a 30 MW plant at atmospheric pressure. Combustion temperatures of 1500-1600°F and heat release rates of about 400,000 Btu/cu ft/hr are contemplated to give heat transfer rates larger than those of conventional pulverized coal boilers.

Figure 1 sketches the process flow. Air is preheated by exchange with stack gas. Part of the air conveys crushed coal (1/4" to 0) and limestone (8-10 mesh average) to the fluidized bed steam generator. The rest of the air enters through a distributor plate to support fluidization and combustion. Spent limestone and sulfate with a low unburned coal content are removed for disposal.

Coal fines and ash are elutriated to a cyclone, recovered, and combustion completed with 40 percent excess air in the carbon burn-up unit. There is also steam generation and/or superheating in this unit. Ash is recovered from all stack gas as indicated, including electrostatic precipitators.

Experimental work with a unit firing 600 lbs. coal/hr has been carried out by Pope, Evans and Robbins since 1967 at Alexandria, Virginia to provide the basis for the design (2,3).

The fluidized boiler has several advantages:

1) Efficient volumetric combustion reducing size and plant area needed and permitting shop fabrication.

2) High heat exchange rates permitting low combustion temperatures.

3) Reduced formation of nitrogen oxides.
4) Relatively low excess air, 5-10%.

5) Coal may contain up to 60-70% moisture after bed is operating.

6) Produces little or no alkali salts in flue gas.

7) Coal rich in pyrites can be utilized.

8) Handles high ash fuels, even with low softening temperatures. They anticipate no problems with high swelling indices but reported no indices for coals fired.

9) Full pulverization of coal not required.

HANDLING SPENT LIMESTONE

Essentially all the disadvantages of removing sulfur in a fluidized boiler with limestone center around the solids handling and/or chemical processing required to dispose of the sulfate-lime solids or to recover the sulfur and recycle the calcium oxide. The alternatives and problems will be discussed briefly.

Sulfur Removal in the fluidized bed is accomplished by the reactions:

\[ \text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2 \]  \hspace{1cm} (1)

\[ \text{CaO} + \text{SO}_2 + \frac{1}{2}\text{O}_2 \rightarrow \text{CaSO}_4 \]  \hspace{1cm} (2)

The issue is partly the degree of conversion of the CaO in a limestone particle before it is removed or the Ca/S ratio.

For a full stoichiometric ratio, all Ca going to sulfate, a ton of 4%S coal would require 215 lbs of limestone to produce 270 lbs of CaSO_4 for 90% sulfur removal. Experiences indicate that
only a fraction of this degree of conversion is accomplished at the desired fluidization condition. The assessment of the fluidized bed (1) used a 6/1 Ca/S ratio in the feed. At this ratio a ton of 4%S coal would require 6 times as much carbonate or 1290 lbs. and yield 602 lbs of CaO plus the 270 lbs of CaSO₄. Moreover the CaO would react with water exothermically if used as land fill and the Ca(OH)₂ product is water soluble. Therefore use of limestone with high Ca/S ratios give immense disposal problems, and chemical processing is required even for low Ca/S ratios. Ways might be found to separate and use the CaO or Ca(OH)₂ and/or CaSO₄ but a sizeable chemical plant would result.

Some indication of the improved Ca/S ratio which may be attained (using salt as an addition) is given by a short Bureau of Mines test on the PE&R unit in March 1973 (8). Here the CaCO₃ fed per hour as compared to coal fed give 1.5 and 2.0 as the Ca/S ratio. That is, the efficiency of limestone use was somewhat less than forty percent. These two tests were made to show that the stack gas met environmental standards, but did not include work to indicate the relative effectiveness of the heat transfer process. Other than this report, no experimental information was made available to us.

It is stated that CaSO₄ encapsulates the unused carbonate. It is doubtful that for atmospheric combustion units where unused Ca is in the form of CaO that sufficient protection of the environment could be provided from CaO solubility as
Ca(OH)$_2$ or even from CaSO$_4$ when the unprocessed waste is used as land fill.

Processes for regenerating the CaSO$_4$ have been studied by Esso (6), Argonne (5) and is described conceptually as being possible in a separate section of the boiler unit. Argonne's processes (5) involve regeneration with CO at 2000°F or converting CaSO$_4$ to CaS with CO at 1600-1700°F followed by reaction of CaS with CO$_2$ and H$_2$O at 1000-1300°F to release H$_2$S and yield the carbonate.

Esso has a 7 ton per day pilot unit under development for operation at pressures up to 10 atm. Their results obtained on a predecessor small pressurized unit is summarized as of February 28, 1971 (6).

PRESSURIZED FLUIDIZED COMBUSTION BOILERS

The use of pressure in a fluidized boiler has been investigated by Hoy (4) in England. His group is prepared to go to a 9 MW 75 psi unit with gas turbine expander, followed by a 250 MW plant at 150-225 psi.

From Esso, who have been testing at 10 atm with a view to regeneration, it is learned that dolomite is needed for pressurized boilers because of the CO$_2$ partial pressure. The MgCO$_3$·CaCO$_3$ transforms to MgSO$_4$·CaCO$_3$ or by heat alone to MgO·CaCO$_3$. The unreacted dolomite would likely be MgO·CaCO$_3$. For one atmosphere work with limestone, the unsulfated carbonate is in the form of CaO.
FUTURE PLANS

Pope, Evans and Robbins with Foster Wheeler have formed Fluidized Bed Combustion Company. This company together with Combustion Systems Limited, an off-shoot of the British efforts, propose to build and test an 18 MW pressurized boiler unit to operate at 90 psia followed by a larger unit (7).

CONCLUSIONS AND RECOMMENDATIONS

1. The fluidized boiler has great potential to give an economical and compact boiler. With limestone as a fluidized solid, sulfur can be removed during combustion.

2. Experimental work with the steam generator has overcome many operational problems indicating a high probability that larger units would be successful.

3. The short-comings lie in the limestone or dolomite requirements and the handling of the solid waste. Use of a once through basis increases the carbonate rock needs by a factor of at least 2 to 3 and some chemical processing of the waste is needed for atmospheric combustion units which give high CaO concentrations in the waste. Regeneration processes may be developed within ten years to recover elemental sulfur, but the plants would be substantial complex chemical installations as appendages to power generation.

4. Pursuit of larger pressurized fluidized boiler installations is recommended. They could be used with stack gas cleaning in pressurized systems as well as with dolomite in the fluidized bed to remove sulfur.
5. Chemical technology for disposing of sulfate lime or corresponding dolomite waste products should be developed to meet environmental restrictions.

6. Pursuit of R&D for regeneration of the CaSO$_4$ with sulfur recovery and/or use of the calcium sulfate such as in building products is advocated.


COAL BENEFICIATION
by
M. Rasin Tek

Review and Assessment

PHYSICAL BENEFICIATION

The physical cleaning of coal involves crushing, grinding, sizing, solid separation, washing, flotation in various combinations designed to reduce inorganic matter (1,2). A new process for removing pyritic sulfur by stage wise froth flotation and use of a chemical called Santathe was recently announced (3). Recently chemically induced breakage (comminution) using methanol and ammonia gained some interest through research conducted in Syracuse Research Laboratories (4). The coals subjected to physical treatment must satisfy the usual pulverized boiler feed and environmental sulfur emission requirements.

CHEMICAL BENEFICIATION

As compared to physical treatment described above, the chemical beneficiation goes a step further in the chemicals are used to remove the pyritic sulfur from the coal. For coals with low enough organic sulfur, this beneficiation is designed to make those coals suitable for meeting environmental standards. Chemical beneficiation is purported to remove up to 90-95% of pyritic sulfur and lose not more than 5% of the coal while current physical cleaning processes re-
move about 50% of pyritic sulfur and loose some 10% of the coal. The "Meyer's Process" developed by TRW has had bench scale (5 liter) extraction tests with ferric sulfate \( \text{Fe}_2 (\text{SO}_4)_3 \) as solvent on four typical Appalachian coals (5,6). Based on bench-scale and pre-pilot test data, engineering design and cost estimate studies have been under way for a process plant to treat 10,000 tons of coal per day (7).

The following information should be considered significant to assist in evaluating this route to provide clean solid fuel from coals:

1. The process uses -14 mesh top size so would normally be carried out in a site adjacent to plant utilizing the coal.

2. The physical size is unchanged and the moisture content of the proposed product is low enough to be fed directly into conventional coal burning plants.

3. TRW earlier reported that more than sufficient reserves exists of the coals low in organic but high in pyritic sulfur. They had indicated that 80 MM Tons/year of Appalachian production and 90 billion tons of Appalachian reserves would be released to power plants by chemical beneficitation. We found that their view and figures are further supported and justified by The Bureau of Mines (8), Dow Chemical Co. Engineers (9) and L. Lorenzi Jr. (10).

4. Engineers from both TRW and Dow Chemical Co. indicate that with substantiating data from the proposed
12 Ton/day pilot unit it would be possible to build and operate a demonstration plant using relatively conventional equipment within five years.

5. The concept of multi-vessel reactors came under discussion and scrutiny during the recent EPRI meeting in Atlanta. The concept of several reactors in parallel flow has been opted by both TRW and Dow design engineers. It is believed that the approach is sound and viable.

6. The overall thermal efficiency of TRW process, from coal unloaded from rail cars to coal loaded into unit trains has been calculated both by U of M Team based on latest Dow Flow Sheet and Dow Chemical Engineers to be about 87%.

7. The latest cost estimates by Dow Chemical Co. Engineers indicate a capital investment figure from dollars to kwatts to be $108/kw (9). Dow engineers further believe that the added operating cost for the chemically beneficiated coal calculated by U of M EPRI Team to be 40 to 60¢/MM Btu could eventually be reduced to about 25¢/MM Btu.

8. As compared to high temperature gasification or liquefaction plants, the chemical beneficiation is relatively simple, more conventional and uses existing technology. It is recommended that the Chemical Beneficiation Process (TRW Meyers) be given support through Pilot Plant Stage and further to larger scale, if it appears to match predictions.
TRW CO.--CHEMICAL DESULFURIZATION OF COAL

Crushed coal is treated with warm ferric sulfate solution in a large reactor where air or oxygen is also introduced. The chemical reaction transforms ferric sulfate to ferronsulfate sulfuric acid and releases elemental sulfur.

$$\text{FeS}_2 + 4.6\text{Fe}_2(\text{SO}_4)_3 + 4\text{H}_2\text{O} = 10.2\text{FeSO}_4 + 4.8 \text{H}_2\text{SO}_4 + 0.8 \text{S}$$

The liberated sulfur is removed by dissolution in a warm naptha bath. The solution is cooled permitting crystallization of sulfur filtrated out with naphtha heated and recirculated back to the sulfur extraction tank. The coal slurry from the sulfur extraction vessel goes to a washer filter and dryer.

The ferrous sulfate produced in the main reactor is regenerated into ferric sulfate "in situ" by air:

$$9.6\text{FeSO}_4 + 4.8\text{H}_2\text{SO}_4 + 2.4\text{O}_2 \rightarrow 4.8\text{Fe}_2(\text{SO}_4)_3 + 4.8\text{H}_2\text{O}$$

The excess ferric and ferrous sulfate generated are removed from the system. The flow sheet for the process is shown in Figure 1. The sulfate solution underflow from the reactor is split into two streams, one recycled back to the reactor the other to an evaporator to provide steam and wash water. The underflow from the evaporator is filtered with filtrate recycled back to the main reactor and excess solid sulfates removed from the system.

**Advantages:**

1. Low cost
Figure 1. Flow Sheet for TRW Meyers Process
2. No phase change involved
3. Potential for significant increase of acceptable reserves
4. Efficient in removal of pyritic sulfur (about 92% of pyrite sulfur is removed).

Problems, Limitations
1. Only applicable to certain coals
2. Currently not tested beyond bench scale
3. About 6.7% of the coal feed is lost as ash and coal fines.

Economics

Full economic analysis of the Meyers Process is given in detail as example calculations of our section on economics. The results are summarized below:

**Bases** 10,000 tons/day,

2.6 X 10" Btus of fuel per day heat input, approximately equal to a 250 MMCF/day high Btu gasification plant output.

**Operation** 24 hrs/day and 330 production days/year

10 days product inventory

3 days raw coal inventory

**Coal Feed** Lower Kittanning 14 mesh top size

**Chemical Analysis % Dry Basis**

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<tr>
<th>Component</th>
<th>%</th>
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<td>Ash</td>
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<tr>
<td>Organic sulfur</td>
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<td>Sulfate sulfur</td>
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<tr>
<td>Heating value</td>
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Coal Product
Chemical Analysis & Dry Basis

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<td>Sulfate sulfur</td>
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Heating value 12,900 Btu/lb.

Cost of Fuel Produced (Utilities Financing Method)

Input Parameters
Debt Equity Ratio 75%/25%
Per cent interest on debt 9.5%
Per cent return on equity 15%
Federal Income Tax Rate 48%

Total Capital Requirements

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<td>Total Plant Investment</td>
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<td>Interest during Construction</td>
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<td>Start-up costs (20% of gross from Operating Cost)</td>
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<td>Working Capital</td>
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Total Capital Required (TCR) 146.48
Initial Debt 75% of TCR 110.00
Initial Equity 25% of TCR 36.48
Operating Costs  Base load plant at 90% load factor

Coal (a) 30¢/MMBtu high heating value
       deep mined bituminous coal)  25.74

Other Raw Materials, Catalysts, Chemicals  17.00

Purchased Utilities
  Electric Power @ 0.9¢/KWH  .297
  Cooling Water  .119
  Sewerage @ 0.001 $/M Gal  .003
  Refuse disposal @ $ 0.50/ton

Labor
  a. Process labor @ $5.20/man hour  .593
  b. Maintenance labor (1.5%/year of Plant Invest.)  1.650
  c. Supervision (15% of operating and maint. labor)  .338
  d. Administration (60% of a,b,c, above)  1.550

Operating Supplies (30% of process operating labor)  .178

Maintenance Supplies (1.5%/year total Plant Invest.)  1.650

Local Taxes and Insurance (2.7%/year of T?P.I.)  2.960

TOTAL GROSS OPERATING COST  $MM  52.245

By products Credits

Sulfur @ $10.00/LT  1.0
Ammonia  -
Light Oil  -
Heavy Oil  -
Char  -

TOTAL NET OPERATING COST  $MM  51.245

The Table 1 shows the fuel cost calculation by year, Utilities Financing Method (M1).

The comparison of coal estimates given directly or computed indirectly by U of M EPRI Team, Dow Chemical Co,, and early economic study by TRW staff are summarized in Table II.
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<th>End of Year</th>
<th>(1) Rate Base TCR-Accrued Depr. @ Mid-Year</th>
<th>(2) Return on Rate Base 10.86% of (1)</th>
<th>(3) Return on Equity 15% of .25 of RB</th>
<th>(4) Fed. Inc. Tax $48 / 52 X 3</th>
<th>(5) Deprec. 0.05 (TCR-Work Cap)</th>
<th>(6) Total Net Op. Cost</th>
<th>(7) Total Fuel Rev. Reg. (2)+(4)+(5)+(6)</th>
<th>(8) Fuel Cost (7)/annual Feed €/MMBtu</th>
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### TABLE II Comparisons of Cost Estimates for Meyers Process

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<td>$/MM Btu</td>
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* The estimates by U of M EPRI Team are on the basis of Federal Power Commission Supply Task Force Utility Financing Method (II)
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2. Environmental Considerations in Future Energy Growth, U.S. Environmental Protection Agency, by Battelle Laboratories, Columbus, Ohio, April (1973).


4. Chemical Communion of Coal, Dr. Robert Aldrich, Life and Material Sciences Center, Syracuse University.


PYROLYSIS

by

Donald L. Katz

Review and Assessment

It is expected that in the years ahead coal refineries will spring up both at the mine and in the metropolitan areas to serve the fuel needs based on coal. The fuels that will come from such a plant will vary but are likely to include high and low Btu gases, low sulfur oils, char and/or low Btu gases. The high Btu gases are likely to be reformed to become SNG for pipelines, treated lower Btu gases could be used as turbine or boiler fuels. The liquids could well be light oil such as for turbine fuels, gasoline-based materials, diesel fuels and, of course, some heavier oils which might well be the fuel for utility boiler plants. Such multi-product plants are likely to reach the status of having high valued products and the so called by-products such as the heavy oils or low Btu gases which would normally become used in utility power plants. Several processes besides pyrolysis units may be included in these coal refineries.

Pyrolysis processes are designed to strip some liquids from coal without going to high pressures and are not primarily aimed at removing sulfur. They leave a large amount of char
with sulfur content like in the coal processed. The liquids are of high quality when hydrotreated to remove sulfur and a substantial fraction is likely to be too valuable to burn with boilers, but possibly economic for turbines. These processes have considerable technological experience, and when thought of as part of coal refineries could provide suitable clean fuels for utility plants. This section of our report deals primarily upon those processes involving pyrolysis, that is low temperature carbonization where the temperature decomposes the coal into gases, liquids and a char. 

**Pyrolysis Processes**

Three pyrolysis processes will be described: COED, TOSCOAL, and Garrett. COED has been under investigation for twelve years and currently makes, in a 36 ton/day pilot unit, high Btu gas and liquid products which are hydrogenated to low sulfur fuels. The third product is char. Currently the COED project proposes to install a gasifier for the char to demonstrate the advantages it has over coal.

The TOSCOAL process started with the conversion of oil shale to liquid and gaseous products, and has been adapted to coal conversion.

The Garrett flash pyrolysis process stresses rapid heating as a technique for pyrolysis coal. These processes will be discussed in turn.

It should be appreciated that coal refineries will include hydrogenation of liquids at higher pressures and possibly the
conversion of low Btu gas by the addition of hydrogen to high Btu gas. The oil from the pyrolysis unit requires hydrogenation to lower its viscosity, control its properties and remove the sulfur (desulfurization and reforming). It is quite possible that a COED plant would be placed beside a catalytic hydrogenation plant for coal in which the char from the COED plant would be used for generating hydrogen needed in the liquefaction plant. One could put together a whole series of processes which would have the advantage of making the most of the superior, or high priced products and therefore would have some "by-products" which could well be in large volume for the utility plant. Even chemicals such as benzene and other cyclic compounds could well come from coal as a skimming operation in which only a small portion of their products are removed but their value would be a significant contribution to the cost of the project. Even petrochemical plants could become adjunct to coal refineries like petrochemical plants are adjunct to oil refineries today.

It is appreciated that utility plants have some aversion to supporting such projects because of their feeling that the process really is not being developed for them and that they will get the low quality products anyway because refineries have no other place to sell it. Although there is a bit of truth in this view, if such plants can be developed to give lower cost fuels for utility plants, it would seem that EPRI should give encouragement to these coal refineries as one of the alternate routes for obtaining clean fuels and with relative ease as far as utility industry is concerned if they are sold to them in clean form across the fence.
A coal refinery adjacent to a utility plant might well provide the products and be co-owned as an integral part of a total project and that certain products from the coal refinery are designated for the utility plant such as desulfurized char, for a fluidized boiler unit. Should utility plants have use for multiple fuels, high Btu gas, liquids and low Btu gas or char, then captive pyrolysis plants could serve utilities directly.
Process Descriptions

FMC - COED

Project COED (Char-Oil-Energy-Development) has been under development at Princeton, New Jersey since 1962 under OCR sponsorship (1-5). It is a process for converting coal to produce char, oil and gas by reacting the coal in a multistage, fluidized bed, Figure 1. The pyrolysis-derived oil is being hydro-treated to produce a synthetic crude oil. The product gas is high Btu and can be reformed to a pipeline gas. The char product can be utilized as a fuel for power generation if the sulfur content is low enough, or gasified with sulfur removal. Project COGAS is concerned with processing the char and is discussed separately (6).

Dried crushed coal is treated in four fluidized bed stages at successively higher temperatures until a major fraction of the volatile matter of the coal is evolved. Heat for this pyrolysis is obtained by burning a portion of the char with oxygen in the last stage. Hot gases from the last stage then flow countercurrently to the coal and constitutes the fluidizing gas and heat supply for the third and second stages in order. Hot char from the fourth stage and the third stage is recycled to supplement the heat from the gases. The first stage fluidizing medium is supplied by burning a portion of the char or gas produced with air. Gas and oil are recovered by cooling and condensing the volatiles from the pyrolysis. A 36 tons of coal per day pilot plant has been in operation since August 1970. They desire to add a low Btu gasifier to operate with the char along with the pyrolysis unit and the hydrotreating of the oil.
Project COGAS is carried out under COGAS Development Co. (2), a joint venture of some 6 companies. They have a proprietary process for converting char to fuel gas which will be integrated with the pyrolysis - hydrotreating processes of COED to make both synthetic crude oil and pipeline gas.

Prepared by Donald L. Katz
OIL SHALE CORPORATION - TOSCOAL

The Tosco Pilot plant was visited, but not studied. No process results other than those published (7) were made available.

The Oil Shale Corporation process as developed for retorting oil shale and applied to coal is illustrated by Figure 2. The energy and pyrolysis is supplied by circulating hot ceramic balls which are about one-half inch in diameter. The balls are heated by air combustion of a fuel in an external circuit, so there is little contamination of pyrolysis gas by combustion products. The hot flue gas is used to preheat the coal which moves to the rotating pyrolysis drum where it is in contact with the heated balls. The gas, char, and balls are separated in a trommel. The char is cooled for storage or for shipping, the balls are recirculated in a ball elevator back to the ball heater, and the hot gaseous products are cooled and fractionated. Some or all of the gas can be burned in the ball heater. Supplemental fuel may also be necessary to operate the process. Tosco's experience includes the operation of

1. A one-ton per hour TOSCO II oil shale retort pilot plant at Rocky Flats, Colorado.

2. A 1000 ton per day semi-works oil shale plant at Parachute Creek, Colorado.

One coal, Wyodak from Gillette, Wyoming has been retorted in the pilot plant, and also in a 10 lb per hour continuous
Figure 2. Oil Shale Corporation TOSCOAL Pyrolysis-Type Fuel Gas from Coal Process
retort. The yields are reported to be close to those of a Fischer assay.

TOSCO's objectives are to produce some tar liquids of value, and a char of higher Btu content than raw western coal as a shippable product. Data for preliminary economic evaluation have been obtained, and char samples were sent to boiler manufacturers for testing. Yields are given in Table I, but no economic evaluations of either the products or the process were available for this study. The highest temperature cited 970°F may be close to the operating limits of the present process.

The benefits of the process are, then:

1. Improvement of the energy content (heating value) per unit weight of the char relative to the raw coal. A major share of this is simply drying of the particular coal.

2. Obtaining gas and liquid by-products. Part of these will have higher unit value than coal if there is a market.

Offsetting these benefits are the factors:

1. Although the heating value per unit weight of the char is higher than coal, the char is powdery, and the heating value per unit volume, then, may not be higher than raw coal unless some compacting is used. The char may be phrophoric.

2. The gas, or its thermal equivalent (and possibly part of the tar) is necessary to run the process. No process energy demands are reported.
TABLE I

TOSCO II RETORTING OF WYODAK COAL

Product Yields
(Lb/Ton of As-Mined Coal)

<table>
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<th>Retort Temperature</th>
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<td>C-8</td>
<td>C-2</td>
<td>C-3</td>
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<td>Char</td>
<td>1049.0</td>
<td>1011.7</td>
<td>968.7</td>
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<tr>
<td>Gas (C₃ and lighter) (SCF/ton)</td>
<td>119.0</td>
<td>156.7</td>
<td>126.0</td>
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<tr>
<td>Tar (C₄ and heavier) (gal/ton)</td>
<td>(1250.0)</td>
<td>(1777.0)</td>
<td>(1624.9)</td>
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<tr>
<td>Totals (lb)</td>
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<tr>
<td>Recovery (%)</td>
<td>99.2</td>
<td>100.7</td>
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There are some limitations of the process:

1. There is no desulfurizing treatment of the char. The process is handicapped unless the feed is one of those coals whose sulfur content is low enough to produce an acceptable char.

2. Caking coals cannot yet be handled, although the exact limit of the caking nature does not seem to have been established.

EQUIPMENT AND TECHNOLOGY

The TOSCO process was developed to handle oil shale which is about 85 to 90 percent ash. The pilot plant process equipment may, then not be optimally designed for processing coals of relatively much less ash. Much more pilot and some semi-works plant experience may be needed to give a basis for process comparisons. TOSCO has solved many problems in the development and operation of the 1000 ton/day semi-works plant. We have no knowledge of, nor information on, the difficulties and problems of scaling to full size, nor of the behavior and capacity of the equipment when processing coal rather than shale. The extent of coal preparation and grinding necessary has not been reported. We could not estimate processing costs, plant investments and net product yields from data available to us.

Prepared by Brymer Williams
Garrett Research and Development Co., Inc. has worked on a flash pyrolysis process for producing liquids and gases since 1969 (8, 9, 10). The process concept is based on the philosophy of partial gasification in which the direct yield of methane and other hydrocarbons is obtained by rapid pyrolysis of coal. It is conceptually similar to the Lurgi Ruhrgas Process (11, 12) in that hot recycled char provides heat for coal devolatilization.

Development work includes operation of a 5 lb/hr. continuous flow bench scale reactor and a 50 lb/hr. gasification pilot plant which has operated since January 1973. Pulverized coal is fed to a mixing chamber at approximately 50 psig where it is rapidly heated to 1600°F in less than two seconds with hot recycle char in entrained flow. This is shown conceptually in Figure 3. Pyrolysis gas is separated from the char in a series of cyclones. A portion of the char is withdrawn as product and the balance is partially combusted with air to provide hot recycle char which provides sensible heat for pyrolysis. The sensible heat contained in the combustion gas is recovered in further processing.

The objective of the rapid heating is to maximize the production of gas while producing tar oil and char. The product split on a western subbituminous coal, Big Horn, Wyoming, given in Table II is shown in Figure 4. The gas composition is given in Table III with the C_2^+ fraction being largely
Figure 3. Conceptual Schematic of Garrett Flash Pyrolysis Process
TABLE II

Composition of Big Horn, Wyoming Subbituminous Coal

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<td>H₂O</td>
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100.00

Heat of Combustion 9200 Btu/lb (HHV, as fed at 25°C)

TABLE III

Garrett Coal Gasification Process
Typical Gas Analysis at 1600°F
(Continuous Laboratory Reactor)

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<thead>
<tr>
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<tr>
<td>H₂</td>
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<tr>
<td>C₂⁺</td>
<td>14.4</td>
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100.0  

Molecular Weight 18.4

Heat of Combustion 646 Btu/scf (HHV at 25°C)
ethylene. The gas yield increases with temperature but temperature is limited to about 1750°F because of the low ash softening temperature. The tar properties are given in Table IV and the char properties in Table V. It should be pointed out that sulfur has been distributed into four streams: combustion gases, pyrolysis gases, tar oil and char. There is said to be 30-45% reduction in the sulfur level in the char on a pounds of sulfur per Btu basis.

Ignition characteristics of the char compare with those of coal in studies made. Pyrolysis chars have a tendency to be slightly dusty and pyrophoric.

Free swelling eastern bituminous coals would be more difficult to pyrolyze because of their tendency to be sticky on heating. It is possible they could be handled with greater char dilution of the feed coal and higher entrainment flow rates. The sulfur levels in the char would be much greater and desulfurization would be required in conjunction with its utilization.

The Lummus Co. was commissioned to conduct a study of a grass-roots 250 MM SCFD pipeline gas plant based on the Garrett process. Lummus did not see any particular equipment problems and concluded that by taking a 20¢/mm Btu credit for char, the Garrett process could produce pipeline gas at a cost 20-34¢/mm Btu less than by the Lurgi process.

Prepared by Dale E. Briggs
### TABLE IV

#### Typical Tar Properties at 1600°F

<table>
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<tr>
<th>Element</th>
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<tr>
<td>O</td>
<td>.8</td>
</tr>
<tr>
<td></td>
<td>100.0</td>
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</table>

Specific Gravity: 1.14 @ 100°C
Viscosity: 33 cp @ 100°C

### TABLE V

#### Typical Char Properties at 1600°F

<table>
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<th></th>
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</table>

Fischer Assay Tar: 0
Quinoline Insolubles: 100
Heat of Combustion: 11,700 Btu/lb (HHV)
Bibliography


COAL GASIFICATION

by

John E. Powers
Dale E. Briggs

Review and Assessment

Coal gasification implies the reaction of solid coal with air, oxygen, steam, carbon monoxide, hydrogen or mixtures of these gases to produce a gaseous product which can be used as an energy source. As compared to pyrolysis, coal gasification is directed toward total conversion of the carbon to gaseous products.

Coal gasification has a long history in this country. For many years air-blown, fixed-fuel-bed processes were used almost exclusively for complete gasification of coal or coke to town or producer gas. At one time, some 45 years ago, there were 14,000 such producer gas units in operation in the United States. When natural gas became available, the producers were phased out of existence because of the low cost and convenience of natural gas.

The need to produce synthetic natural gas and hydrogen for ammonia synthesis from coal, as aided by development of commercial oxygen plants, led to many new gasification concepts since 1940. Von Fredersdorff and Elliott (1) prepared an excellent review of the coal gasification literature up to 1962. Since 1962,
a substantial portion of work in coal gasification was either done by the U.S. Bureau of Mines or under sponsorship of the U.S. Office of Coal Research. The OCR Annual Report 1973 (2) lists the research and development reports on OCR sponsored projects since its establishment in 1960.

In the recent past, there has been substantial interest in production of environmentally acceptable low or intermediate Btu gases from high sulfur coals for utility use. These gases would replace fuel oil, natural gas and high sulfur coal where burned directly for steam generation.

The purpose of this section is to review the present coal gasification processes which could produce low or intermediate Btu gases for combustion in conventional boilers or combined cycle plants. An assessment is made of how these gasification processes might help to fill the electric power industry's need for clean fossil fuels.

GENERAL OBJECTIVES

An ideal coal gasification process is one in which coal can be converted to an environmentally acceptable gaseous fuel (synthesis gas) with minimum processing and maximum thermal efficiency. The EPA allowable emissions for new fossil-fuel fired steam generators are 1.2 lb SO$_2$, 0.2 lb NO$_2$, and 0.2 lb particulates per million Btu heat input (3). The gasification processes currently under development can satisfy these air pollution control requirements. All the processes have waste water streams and these streams must be treated before discharge.
In gasification, the sulfur compounds are almost all converted to hydrogen sulfide and the nitrogen compounds are mostly converted to ammonia by partial oxidation of the coal with oxygen (or air) and steam under reducing conditions. Hydrogen sulfide is considerably easier to remove from gases by scrubbing or chemical reaction than sulfur dioxide. The volume of the gas that must be processed is substantially less than that resulting from the complete combustion of coal with normal amounts of air. These are two advantages of coal gasification when compared to stack gas clean-up.

The composition of the synthesis gas is a function of the feed rates of oxygen (or air) and steam relative to the feed coal, the gasification pressure and temperature, and the flow of the feed and product gases relative to the feed coal. Most of the gasifiers under development in the AGA-OCR synthetic natural gas program are operated to maximize methane production in the gasifier. This usually involves two stages with transport and flow rate control of hot char (devolatilized coal) between stages. High pressures are used to facilitate methane formation, to reduce the volume of the gas being desulfurized and to eliminate compression to pipeline pressures.

The criteria for a coal gasification system for utility use are different from those for pipeline gas. Foremost, the gasifier must be reliable and the system must be able to follow the electric load variations during the day. The gasifier must meet environmental and process requirements for particulates, sulfur and nitrogen.
Particulate concentrations in fuel gases are not critical for conventional boilers, but should be considerably less than EPA requirements if the gas is to be used as a gas turbine fuel. According to Westinghouse (4), particulates in the range of 2-6 microns are most critical and should be reduced to concentrations less than 0.0005 gr/scf. In studies by Hoy (5), he concluded that there were no particular erosion problems from particulates impinging upon turbine blades, but they did tend to adhere to the blades as hard scale and severe damage could result if large pieces of scale let loose.

The heating value of the clean synthesis or fuel gas is important in several ways. Low (100-175 Btu/scf) and intermediate (250-400 Btu/scf) gases are less expensive to produce than pipeline quality gas (950-1000 Btu/scf). They are produced by air and oxygen blown gasifiers, respectively. Because of the greater volume of gas for a fixed amount of energy, low and intermediate gases must be used in the vicinity of the gasifier. Pipeline transportation for more than a few miles would not be economical in general.

The heating value of the fuel gas is important in retrofit applications. Some derating of boilers can be expected if a low Btu gas is used to replace natural gas or fuel oil. The amount of derating depends upon the boiler design. In some cases boiler modifications are possible which can minimize derating. When the heating value of the fuel gas is 300 Btu/scf or higher, derating becomes minimal.
Although there is an advantage in having some methane in the fuel gas from a Btu/scf basis, the combustion characteristics of carbon monoxide and hydrogen are superior to that of mixtures containing methane. The advantage of a somewhat smaller gas volume associated with the higher heating value of methane in reducing equipment sizes is not large. Gasifiers which have the potential to produce high methane yields are either complicated or they operate in a way that tar formation is a problem. Therefore, high methane yields in coal gasifiers for utility use are not critical and may not even be desirable.

Coal gasifiers designed and operated for utility use should not have to operate at pressures greater than about 300 psi. At this pressure sulfur removal is close to optimal and the clean fuel gas is at an acceptable pressure for combined cycle applications. Near atmospheric pressure coal gasifiers are satisfactory for retrofit applications, but equipment sizes are large. Such gasifiers do, however, eliminate the need for lock hoppers. Atmospheric pressure gasifiers can be used for combined cycle systems by cooling and removing particulates prior to compression and desulfurization.

Overall process thermodynamic efficiency is extremely important in comparing coal gasification systems. Air or oxygen and steam must be used. The energy required to compress or produce these feed streams must come from the coal used in the process and the amounts of these streams are
therefore important. Energy recovery from raw, hot gases which are being cooled for desulfurization, is also important.

COAL GASIFICATION PROCESSES

Coal gasification processes include all the steps from coal to clean fuel gas. A schematic representation of the process steps are shown in Figure 1 for production of both low and high Btu gas. There are many variations possible and deviations from the steps shown are common. Coal preparation, gas cleaning and sulfur removal are sub-processes which are reasonably well established. Although improvements are needed in each, the coal gasification step is critical and needs the greatest development.

Coal gasifiers can be characterized or classified in several ways. The two most important are: the coal flow relative to the gas flow, and the ash removal method. According to flow, gasifiers can be classified as fixed-bed, fluidized bed and entrained flow coal gasifiers. In fixed bed gasifiers, lump coal is fed at the top and the gases flow upward through the bed at rather low velocities. Even at low velocities fly ash and fine coal particles are entrained and carried over with the gas. Fluidized-bed gasifiers operate with crushed coal. The gases flow upward, thereby maintaining the bed of coal in an expanded and fluidized state. Fresh coal can be fed at the top, middle or bottom. Small particles tend to accumulate near the top of the bed and larger, more dense particles near the bottom. Fly ash and smaller coal
Figure 1. Schematic Representation of Processing Steps to Low and High Btu Gases
particles will be carried over at higher rates than in fixed bed gasifiers at comparable pressures and temperatures because the number of small particles is larger. The gas velocities need not be larger than for fixed bed gasifiers, but usually are. In entrained flow gasifiers, pulverized coal is carried along with the entraining gases. The gas flow can be either upward or downward.

The condition of the ash and ash removal method is closely associated with the maximum temperature within the gasifier. The mineral matter in coal has a softening point and a melting range depending upon the number and amounts of inorganic compounds present. At temperatures below roughly 1800°F, the mineral matter is dry. At temperatures somewhat higher, the ash becomes tacky and tends to agglomerate. As the temperature increases further the ash completely melts and the viscosity of the molten ash or slag decreases. Molten slag is usually free flowing at 2800-3200°F. In some cases fluxing agents are used to minimize the temperature required for free flow. Coal gasifiers can therefore be classified as dry bottom, agglomerating or slagging with respect to ash removal. Although most fixed bed gasifiers operate as dry bottom gasifiers, the ash may partially melt in the bed and then be cooled by the stream and air or oxygen entering at the bottom. The ash then leaves as a solid.

In application of the ash-agglomerating concept, control can be a problem because coals vary substantially in mineral matter, even in the same vein.
In general the average gasifier temperature is lowest in the fixed bed gasifier and increases in the order: fixed-bed, fluidized bed, ash-agglomerating, and ash slagging gasifiers. Reaction rates increase exponentially with temperature. This favors smaller reactors even when the gas volume is corrected for higher absolute temperatures. Tar cracking is also complete at high temperatures. This is a distinct advantage in gas cooling, desulfurization and waste water control.

Coal gasification in a molten bath of salt or iron is another method of converting coal to a gaseous product. It is generally done in one stage with the mineral matter and most of the sulfur retained in the molten bath. The process is unique since gasification and desulfurization take place in one stage. Unfortunately, a portion of the molten liquid or slag must be continuously withdrawn to remove the mineral matter and sulfur.

Gasifiers can also be classified as to pressure level, number of stages and the source of oxygen--air or oxygen blown. As compared to single stage gasifiers, higher methane yields are possible in two stage coal gasifiers and the amount of air or oxygen required is less.

Chemistry of Coal Gasification

The chemistry of coal gasification includes the reactions between coal, steam, oxygen and chemical compounds that can
form from these substances. It also includes the chemical equilibria between reacting species and products, as well as the chemical reaction kinetics. Von Fredersdorff and Elliot (1) have an excellent review of the subject.

To understand why gasifiers are operated the way they are, one must understand the basic objectives, constraints and thermodynamics of coal gasification. A partial listing includes:

1. The fuel gas should be produced with minimal loss of the energy in the coal.

2. The raw synthesis gas must be cooled to at least 300°F before desulfurization with commercial processes.

3. Tars and condensible oils in the raw synthesis gas are objectionable in terms of heat recovery, water clean-up and tar build-up in gas lines.

4. Steam decomposition should be substantially complete for good thermal efficiency.

5. Volatile ash can be a problem at temperatures above 1600-1800°F.

6. Smaller gas volumes reduce equipment sizes.

7. At elevated pressures, gas volumes are smaller and fluidization occurs at lower gas velocities. Methane yields are higher.

8. The use of oxygen in place of air reduces the gas volume. Depending upon the process, the use of oxygen may increase or decrease the total system cost.
9. The rates of chemical reactions increase exponentially with temperature until diffusion begins to control the rate. At or above 2000°F, rates are high, but often diffusion controlling except where gas velocities are high.

10. Chemical equilibrium is reached in short residence times above 2000°F and steam decomposition is substantially complete.

Steam is used in all coal gasifiers to convert sensible heat to fuel energy in the form of hydrogen by steam decomposition with carbon. In many coal gasifiers, hydrogen is the principal product. This is not the case for utility use, even though hydrogen is an acceptable fuel gas constituent.

Coal contains approximately 75-85% carbon and 5% hydrogen by weight (6) on an ash and moisture free basis. Low or intermediate Btu fuel gases contain 60-70% more hydrogen then the coal from which they are produced. Methane contains 25% by weight hydrogen or roughly 180% more hydrogen than the original coal.

The principal molecular species involved in coal gasification are C, CO, CO₂, CH₄, H₂, H₂O, and O₂. With four elements, four unique stoichiometric balances exist for these molecules. They can be conveniently represented as follows:

\[
\begin{align*}
    \text{C} + \text{O}_2 & = \text{CO}_2 \quad \text{exothermic} \quad (1) \\
    \text{C} + \text{H}_2\text{O} & = \text{CO} + \text{H}_2 \quad \text{endothermic} \quad (2) \\
    \text{C} + 2\text{H}_2 & = \text{CH}_4 \quad \text{exothermic} \quad (3) \\
    \text{C} + \text{CO}_2 & = 2\text{CO} \quad \text{endothermic} \quad (4)
\end{align*}
\]
Several other equations can be written which are combinations of the four above, such as Reactions (5) and (6). Such equations are useful in describing what happens in specific gasification steps. Reactions (7) and (8) are thermal cracking reactions.

\[
\begin{align*}
H_2 + \frac{1}{2} O_2 &= H_2O \\
CO + H_2O &= CO_2 + H_2 \\
C_{\frac{m}{4}}H_m &= \frac{m}{4} CH_4 + \frac{n-m}{4} C \\
C_{\frac{n}{2}}H_m + \frac{2n-m}{2} H_2 &= nCH_4
\end{align*}
\]

Two stage gasifiers are designed and operated to take advantage of the different equilibria that are possible at different pressures and temperatures. The equations above can be used to describe the reactions which lead to these equilibria. In two stage gasification, one stage is operated at high temperatures with coal or char together with oxygen and steam to produce heat and hydrogen for the second stage. The second stage operates as a coal devolatilizer.

Coals contain from 15-20% (medium volatile) to over 30% (high volatile) volatile matter (MAF) which is released during devolatilization. The gases consist mainly of hydrogen, carbon monoxide, methane and other hydrocarbons. Tar vapors and some incombustible gases are also released. The composition of the released volatile matter varies significantly with coal rank, with low rank coals containing a higher fraction of incombustible gases. The combustible gases constitute valuable gaseous fuel energy which is destroyed with accompanying carbon
deposition when the devolatilization step is carried out at high temperatures and long residence times. The equilibrium for reaction (4) shifts from the right to the left. Flash volatilization of fine coal particles is therefore best.

Thermal cracking of oils and tars to non-condensible fuel gas proceeds very rapidly at 1600-2000°F. Since coal is fed into a gasifier cold, the coal must be heated to these temperatures. Fine coal particles can be heated rapidly at short residence times to devolatilize the coal and crack the oils and tars. Immediate gas cooling eliminates carbon deposition.

Most of the heat for gasification comes by Reaction (1) in a single stage gasifier or in the partial combustion stage of a two-stage gasifier. The reaction is so rapid that it proceeds to completion with respect to oxygen consumption. Oxygen present in coal is rapidly converted to H₂O by Reaction (5) in high temperature devolatilization. The H₂O can be subsequently reduced back to H₂ by Reaction (2).

Reactions (1), (2), (4) and (6) are predominate in the partial combustion stage of a two-stage gasifier or in a single stage gasifier. Reactions (2), (3), (4), (7) and (8) occur in the devolatilization stage. The carbon gasification reactions (2) and (4) are never at equilibrium at exit conditions. Equilibrium for these reactions requires nearly 100% steam decomposition and negligible CO₂ content at temperatures above 2000°F and pressures from 1 to 20 atmospheres (1).
Reaction (4) is slower than (2) at equal reactant concentrations. In gasifiers operated to produce high methane yields, high hydrogen concentrations are used in the devolatilization stage to produce methane by Reaction (3).

The oxygen/steam ratio is an important gasifier variable since it affects the equilibrium gas composition and the enthalpy change for the carbon-oxygen-steam system. Von Fredersdorff and Elliot give several plots in Reference (1) which show the effects. The plots are helpful in understanding what happens in gasifier systems and the conditions which lead to high steam decomposition.

Processes in Development

The coal gasification processes reviewed in this study are listed in Table I and classified into types in Figure 2. Some of these are directed toward high methane yields and are in most cases supported in the AGA-OCR program. In recent years, some of these gasifiers have been modified in concept to be more appropriate for utility needs. Other gasifier systems have been developed specifically for utility use.

In addition to the processes listed in Table I, there is other work being done in coal gasification at universities and in industry. Some of the industrial work is proprietary. Processes not listed in Table I were omitted because their existence was not known or because of limited time. No process was omitted for lack of merit.
A summary development status of the gasifier systems is given in Table II. The table gives a time table for the various stages of development from bench scale to commercial scale.

Table III gives a summary review of typical gas compositions.

TABLE I
Coal Gasification Processes Reviewed

Lurgi
Koppers-Totzek
Winkler
Bituminous Coal Research--Bi-Gas
Combustion Engineering
Foster Wheeler
Atomics International - Molten Salt
M.W. Kellogg - Molten Salt
U.S. Bureau of Mines - Stirred Bed Gasifier
U.S. Bureau of Mines - Synthane
U.S. Bureau of Mines - Hydrane
Battelle - Ash Agglomerating Gasifier
IGT - Ash Agglomerating Gasifier
IGT - HYGAS
Westinghouse - Advanced Gasifier
Consolidation Coal - CO₂ Acceptor
Brigham Young - Entrained Bed
Texaco - Partial Oxidation Process
Shell - Partial Oxidation Process
Bituminous Coal Research - Fluidized Bed
Applied Technology Corp. - ATGAS
City College, City University, New York - Squires
Figure 2. Classification of Coal Gasifiers
TABLE II. Summary Development Status of Representative Coal Gasifier Systems

| PROCESS                          | TYPE  | NOTES  | SCALE          | SIZE T/D | 1960 | 1965 | 1970 | 1972 | 73  | 74  | 75  | 76  | 77  | 78  | 79  | 80  | 81  |
|---------------------------------|-------|--------|----------------|----------|------|------|------|------|-----|-----|-----|-----|-----|-----|-----|-----|
| Applied Technology (Algas)      | Gasif.| Bench  | Pilot          | ~        |      |      |      |      |     |     |     |     |     |     |     |     |
| Bureau of Mines (Stirred Bed)   | Gasif.| Pedu   | 10             |          |      |      |      |      |     |     |     |     |     |     |     |     |
| Battelle                        | Gasif.| Bench  | Pilot          | 2.5      |      |      |      |      |     |     |     |     |     |     |     |     |
| BCR B1-Gas                      | Gasif.| Bench  | Pedu Pilot     | 0.1      |      |      |      |      |     |     |     |     |     |     |     |     |
| Brigham Young Univ. of Utah     | Gasif.| Bench  | Pedu           | ~        |      |      |      |      |     |     |     |     |     |     |     |     |
| Consol Coal CO₂ Acceptor        | Gasif.| Bench  | Pilot          | 50       |      |      |      |      |     |     |     |     |     |     |     |     |
| Combustion Engineering          | Gasif.| (1)    | Pilot Pioneer  | 120      |      |      |      |      |     |     |     |     |     |     |     |     |
|                                |       |        |                | 2000     |      |      |      |      |     |     |     |     |     |     |     |     |
| Exxon                           | Gasif.| Bench  | Pioneer        | 0.5      |      |      |      |      |     |     |     |     |     |     |     |     |
|                                |       |        |                | 400      |      |      |      |      |     |     |     |     |     |     |     |     |
| Foster Wheeler (et al.)         | Gasif.| (1)    | Pioneer        | 1200     |      |      |      |      |     |     |     |     |     |     |     |     |
| Hydrane                         | Gasif.| Bench  | ~              |          |      |      |      |      |     |     |     |     |     |     |     |     |
TABLE II. Summary Development Status of Representative Coal Gasifier Systems (continued)

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- Firm Plans
- Proposal
- Design of Plant
- Construction of Plant
- Operation

NOTES: (1) Basic Data from BCR PEDU
(2) Koppers-Totzek and Lurgi Processes are Commercially Available
(3) Conceptual Design Underway Based on IGT DATA
(4) C.F. Braun is Evaluating Processes for High BTU Gas Production
TABLE III. Summary Review of Typical Product Compositions and Heating Values for Coal Gasifiers

**GASIFICATION PROCESSES**

**TYPICAL PRODUCT COMPOSITIONS AND HEATING VALUES**

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Heating Value Btu/scf Water-free basis

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1. Coal less than 1% sulfur
Identification of Problem Areas

In general, it is much less convenient to process a solid, if only to burn it, than either a liquid or a gas. With coal, solids handling problems are compounded by the facts that coal may swell up to 10 times its original volume when heated, will usually ooze sticky tars, and almost always contains significant amounts of moisture, ash, sulfur and nitrogen. In fact, coals contain many of the 92 natural elements.

The problems associated with just burning American caking coals have been essentially solved after many years of development. Even so, coal fired boilers are designed to burn a particular coal and a change in supply will often necessitate appreciable and expensive modifications of existing boilers.

It is now proposed that coal be processed by means other than total combustion to meet a variety of objectives. In particular, it is proposed to produce a low or intermediate Btu gas for use by the utility industry. Progress in this area can best be fostered by gaining an understanding of the problems which must be faced and either solved or circumvented before any such process can be put into successful operation.
The problem areas associated with coal gasification will be enumerated and the various individual proposed solutions described. The various processing steps are shown in Figure 3.

**Utilization of Coal Resources**

As coal becomes more and more expensive, it will be essential that it be used efficiently. The general problem of coal utilization is broken down into three areas of concern: use of all the coal prepared, use of caking coals and carbon utilization.

All coal crushing operations result in a size distribution of coal. Some coal fines are always produced. If successful operation of a gasifier depends on having relatively large lumps, as in the Lurgi gasifier, then the fines formed in coal preparation steps cannot be utilized without expensive preprocessing such as briquetting. It is preferrable to be able to feed coal without any preprocessing as with the Bureau of Mines stirred, moving bed gasifier.

Many gasifiers operate on pulverized coal. Although fines are not objectionable the capital cost and operating expenses of the equipment have to be considered. The power required to operate the pulverizers must be taken into account in calculating the thermal efficiency.

The vast majority of the coals found in the Eastern United States are caking coals, i.e., they swell when heated. As already indicated, such swelling is often substantial. In
Figure 3. Schematic Representation of Processing Steps and Processing Requirements for Coal Gasification
addition, during this period of swelling, tars ooze out and tend to make the coal particles agglomerate, reduce the effective area for reaction, and promote caking clinkers. In moving bed gasifiers, such as the Lurgi, operation may not be possible unless the coal has a swelling index of 3 or under. Naturally, for processing eastern coals it seems desirable to be able to feed all types of coal and caking coals in particular.

Carbon utilization is important in the thermal efficiency. In some processes, a significant portion of the carbon in the coal passes through unreacted. Even in conventional boilers, some unburned carbon appears in the ash resulting in a decrease in thermal efficiency. Carry-over of carbon as char fines is a problem in all gasification processes except for the molten salt or iron processes. It is a less serious problem for the fixed bed processes, becomes more of a problem in fluidized bed and reaches substantial proportions in entrained flow gasifiers. In the Bi-gas type gasifiers approximately 2/3 of the original coal is carried over as char. Fines must be recovered and returned to the partial combustion stage. As an extreme example, pyrolysis processes such as COED or TOSCOAL produce about one half as much char as feed coal and finding a suitable use of the char is a major problem.

**Getting Solid Coal into the Gasifier**

This problem area consists of two distinct steps. The first is raising the coal from atmospheric pressure to a
pressure above the gasifier operating pressure and the second is the physical feeding of coal into the gasifier.

If the operation is near atmospheric, the coal can be conveyed into the gasifier with steam and/or air or oxygen much as is done in conventional boilers. Alternatively, screw conveyors (similar to stokers) can be used.

The problems of introducing coal into a pressurized gasifier are much more severe. Lock hoppers have been used. In this case a gas at high pressure is merely introduced into a sealed hopper containing the coal and after pressure equilization is achieved, the coal is transferred from the hopper to the gasifier. Such transfer may result by gravity through a star wheel feeder (Lurgi), the solid coal may be conveyed with steam and/or air or oxygen (Bi-gas) or a screw conveyor may be used. Lock hoppers do work, but the gas compression and the loss of gas after the coal has been transferred to the gasifier are disadvantages (7).

Lock hoppers can be avoided by slurrying coal with water or coal oil and pumping the slurry to a high pressure feed tank or directly into the gasifier. Slurry feed has been used by the Institute of Gas Technology (8). Coal may be pumped in as a slurry in oil or water, but this results in a separation problem and a loss in thermal efficiency. Oil recovery at a subsequent step in the process is economically vital. This system does not appear to be free of technical problems at present.
Bituminous Coal Research is using a coal-water slurry feed system to pump coal to 1000 to 1500 psig for their Bi-Gas pilot plant gasifier. Texaco also uses a coal-water slurry pump in their gasifier system. A substantial amount of heat is needed to vaporize the water and there is little hope of recovering the energy.

Bituminous Coal Research has worked on the development of a piston feeder for feeding coals (9). Although excessive wear on certain parts existed, Koppers stated in an engineering evaluation (9) that the method should be tried in a large pilot plant.

**Introduction of Caking Coals**

Various schemes have been suggested for eliminating the problems associated with handling caking coals while they are swelling and oozing tars. These include pretreatment in either an oxidizing or reducing atmosphere and either dilution or dispersion to reduce the contact between coal particles during the caking period.

Most bituminous and especially high volatile C bituminous coals tend to swell, agglomerate and cake upon heating in the presence of hydrogen. In some coal gasifiers, such as the Lurgi gasifier, caking coals present a problem in coal utilization and ash removal. Caking coals may be treated by mild heating in the presence of steam and oxygen (10) or by the soaking of coal in solvents such as benzene (11).
There are three methods of mild oxidation pretreatment - fixed bed, free fall and fluidized bed. In the fixed bed method coal is contacted with steam at 800°F and 325 psig containing 1% oxygen by volume. The U.S. Bureau of Mines (12) converted strongly caking coal to a non-caking coal in about 2 seconds by passing preheated (to 660°F) 1/2 - 3/8 inch lump coal through steam containing 5.5 - 12.7% oxygen at 250-350 psig and 1040-1260°F. Fluidized bed treatment is generally unsatisfactory because it is difficult to control, the required residence times are long and excessive localized combustion occurs.

Approximately 1/4 of the volatile matter is lost in coal pretreatment. This is a serious loss in heat content and should be avoided. Many of the new gasifier designs are incorporating the pretreatment into the gasifier system. This permits all types of coals to be used with maximum thermal efficiency, since the volatile matter is not lost.

Dilution usually involves a solid phase, char, ash or limestone, as the dilutant. In the devolatilization stage of a fluidized bed gasifier the relatively inert solid is the fluidizing medium and the concentration of the fresh feed coal is only 1-3%. As a result, the fresh coal only comes into contact with the hot fluidizing medium which heats and devolatilizes the coal.
Pulverized coal is blown into conventional boilers so that the fine coal particles are effectively consumed in the combustion process before contacting another particle. This method of dispersion is carried over into the entrained flow gasifiers such as the Bi-Gas and Kopper-Totzek gasifiers.

Partial Combustion as a Means of Facilitating Sulfur Removal and Producing a Gaseous Product

A gasifier might be incorporated into a coal fueled power generating facility for one or more reasons. Partial combustion yields a mixture containing $\text{H}_2\text{S}$ rather than $\text{SO}_2$, and $\text{H}_2\text{S}$ is easier to remove from the gaseous products than $\text{SO}_2$. Partial combustion also produces a smaller amount of gas than total combustion so that the concentration of $\text{H}_2\text{S}$ following partial combustion is higher than $\text{SO}_2$ following complete combustion. Sulfur removal as $\text{H}_2\text{S}$ is further facilitated if the products of partial combustion are at elevated pressures.

It seems reasonable to expect that the thermal efficiency of a process involving partial combustion of coal will be increased if the number of hydrogen to carbon bonds that are broken are minimized. This has led to the concept of a devolatilization step. Such a step is similar to pyrolysis. In pyrolysis, the main product is a heavy tar oil with a hydrogen to carbon ratio of about 1.1 to 1.3 on a mole basis. In addition, some gas with a hydrogen to carbon ratio of about 3 or so is also formed.
In devolatilization, every attempt is made to eliminate tars and oils by cracking them to a gaseous product in the presence of steam. The gaseous product consists mainly of a mixture of CO and H\textsubscript{2} but the devolatilization step is often operated to produce as much methane as possible. When oils and tars are present water quenching of the gases is necessary before desulfurization. The loss of energy becomes substantial and should be avoided if possible. Although oils and tars can be skimmed off the condensate and returned to the gasifier, water treatment is necessary.

The devolatilization step can be represented in greatly simplified form by the endothermic chemical equation:

\[ 4\text{CH} = \text{CH}_4 + 3\text{C} \quad \text{(coal \ methane \ char)} \quad \text{(endothermic)} \quad (9) \]

The water shift reaction is also endothermic.

\[ \text{C} + \text{H}_2\text{O} = \text{CO} + \text{H}_2 \quad \text{(endothermic)} \quad (2) \]

In contrast, oxidation of carbon to yield either carbon monoxide or carbon dioxide generates heat.

\[ \text{C} + \frac{1}{2} \text{O}_2 = \text{CO} \quad \text{(exothermic)} \quad (10) \]
\[ \text{C} + \text{O}_2 = \text{CO}_2 \quad \text{(exothermic)} \quad (1) \]

There are several types of gasifiers or reactors that can be used to carry out these reactions. The petroleum and chemical industries have carried out reactions on rather large scale (cat cracking, for example) but these industries
attempt to avoid solids handling whenever possible. With coal, one is forced to deal with a solid phase. The four basic types of reactors that seem to be applicable are:

Fixed or slowly moving beds of solids
Extraired solids
Fluidized beds
Molten baths

Coal gasification units must be built such that the heat released by Reactions (1) and (10) is sufficient to support the reactions represented by Reactions (2) and (9). Whereas all existing, commercially proven, gasification processes (Lurgi and Koppers-Totzek are the more abundant) carry out all four processes in a single reactor, developments within the U.S. during the past decade or so—with very few exceptions—have been directed toward carrying out devolutilization [Reactions (2) and (9)] separate from combustion [Reactions (1) and (10)] with some provision for transfer of heat between the two zones. The discussion to follow will be divided on the basis of whether the devolutilization and combustion processes are carried out in either one or more regions.

Partial Combustion in a Single Unit. The oldest existing gasification processes are based on the technology of production of "town gas" and involve either a fixed bed or a slowly moving bed of coal. The Lurgi process is perhaps the best known of the existing commercially-proven processes and is carried out under pressure in one vessel. The design is such that solid chunks of coal form a bed that moves slowly downward by gravity against a flow of hot combustion products
passing upward. Thus, combustion takes place near the grate at the bottom of the bed and devolatilization occurs in the upper part of the bed. As a result of the countercurrent movement of coal solids and hot gases, the carbon utilization is very high and the gases leaving the gasifier are at moderate temperatures—1000°F or so. Whereas the relatively low temperature of the exiting gas is desirable from the point of view of energy efficiency, this same low temperature leads to a high yield of tars with undesirable consequences. The Lurgi process does serve to gasify non-caking coals under pressure in one unit. The Bureau of Mines Stirred Moving-Bed Reactor accomplishes the same task in about the same way but can be operated on run-of-mine caking coal. Both suffer from problems associated with scale-up.

Just as the utility industry has tended to shift from moving bed coal boilers with grates to units with entrainment of pulverized coal, recent developments in coal gasification have also shifted to processes involving entrained solids. The Koppers-Totzek is the existing, commercially-proven coal gasification process which utilizes entrained solids. In the Koppers-Totzek process, oxygen is used to partially oxidize coal in the presence of steam to produce a mixture of CO, H₂, CO₂ and H₂O. Essentially no hydrocarbons remain. In particular no tars remain. Thus, all the reactions appear to occur simultaneously in one stage. Coal, steam and oxygen enter, hot synthesis gas exits upward and molten ash leaves from the bottom. It is difficult to visualize a simpler process scheme—except for the fact that oxygen is used.
An attempt has been made to arrange a single fluidized bed so that flow of the coal in the bed is at least somewhat countercurrent to the flow of gas. In the Synthane process developed by the U.S. Bureau of Mines, a dense fluidized bed at 1100-1450°F is operated in the same vessel above a dilute fluidized bed at 1750-1850°F. The idea of two fluidized beds without physical separation sounds somewhat contradictory but apparently units have been operated.

Over the past several years M.W. Kellogg Company (13,14) and the Atomics International Division of Rockwell International (15), have gasified coal in a molten sodium carbonate bath at 1700-1800°F. Sodium carbonate has a catalytic effect on the reactions and promotes excellent heat transfer. Although primary emphasis is on partial combustion, methane formation is possible at high pressures. A substantial portion of the sulfur and mineral matter introduced with the coal is retained in the melt. Molten salt is continuously withdrawn to keep the ash and sulfur content below maximum allowable values and to regenerate sodium carbonate.

The ATGAS process carries out devolatilization and partial combustion in a single bath of molten iron. It is interesting to note that this process will apparently yield a net amount of iron in the process of producing low Btu gas.

**Partial Combustion in Two Separate Regions.** The majority of the gasification processes developed during the past decade or so in the U.S. generally have a separate devolatilization section (or even several devolatilization sections) and a
separate combustion section with some means of transferring heat between the two as discussed below.

1. Devolatilization with Elimination of Tars

Instead of devolatilization in the upper part of a moving bed of coal, many coal gasification processes under development incorporate separate devolatilization sections utilizing either entrained solids (similar to burning of pulverized coal), fluidized beds or molten baths.

The idea of devolatilizing coal by entraining it in pulverized form in a separate devolatilization region in hot combustion products was incorporated into the Bi-Gas type gasifier by Bituminous Coal Research (BCR). Several processes in various stages of development (including Combustion Engineering and Foster Wheeler, among others) utilize this basic approach to devolatilization. Surprisingly little actual data are available. In particular, the lower limit of temperature required to insure that no tars are present in the product gas is subject to some question.

Fluidized beds are not uncommon in chemical reactors and therefore such beds have been proposed for the chemical reactions involved in devolatilization and gasification. Consolidation Coal (CO₂ Acceptor) and Westinghouse-Bechtel (Advanced Gasifier) as well as IGT (HYGAS), the Bureau of Mines (Hydrane) and others are proponents of this approach. When the fluidizing media is limestone or dolomite, sulfur removal as CaS may be effected. Such beds would be operated at temperatures sufficiently high to insure cracking of tars. The
presence of large inventories of solids in the bed should prove helpful in eliminating tars but the dual objectives of eliminating tars and reducing sulfur as $H_2S$ may prove mutually inconsistent.

2. Combustion

The combustion region generally operates on the same principle as that of the devolutilizer; either entrained solids, fluidized bed or molten salt. The physical arrangement applied in the combustion zone is not necessarily identical to that of the devolutilizer but it seems to work out that way. The Hydrane process and the CCNY Mark II are two exceptions.

Combustion of recycle char fines and/or pulverized coal as entrained solids would seem to be very little different from the operation of conventional boiler units firing pulverized coal. For this reason, very little developmental work has been carried out on the combustion processes required to make two-unit entrained flow gasifiers operate successfully. However, it should be noted that the atmosphere in the combustion region of a gasifier is reducing whereas the atmosphere in a more conventional boiler is oxidizing. An entrained solids gasifier is proposed for the Bi-Gas process, which also utilizes entrained flow in the devolutilizer. Many developments, currently underway or proposed, make use of this principle.
Likewise, use of a fluidized bed in the combustion section of a gasifier should differ little from operation of a fluidized bed boiler. The primary difference is that the former is carried out in a reducing atmosphere and the latter in an oxidizing one. Of course, fluidized bed combustion has never been developed to the extent of conventional boilers firing pulverized coal as entrained solids. Westinghouse-Bechtel plan to utilize fluidized beds for both the combustion and devolatilization units as does the Consolidation Coal CO₂ Acceptor system and others. Both Westinghouse and Consolidation Coal plan to utilize the partial separation of solids of different densities which occur naturally in such beds to effect a partial separation of the fluidizing solid (limestone) from the ash.

3. Heat Transfer Between Combustion and Devolatilization Regions

When combustion and devolatilization is carried out in two different regions it is necessary to transfer the heat from the exothermic reactions of the former to satisfy the endothermic requirements of the latter. Such transfer is usually carried out by direct heat transfer involving either the gaseous products from the combustion region or solids or molten salts which are heated in the combustion zone. In one very unusual case, energy for the endothermic devolatilization reactions is supplied by application of an electric potential to a bed of carbon.

Many proposed gasification processes utilize the gaseous products from the combustion region to devolatilize the coal
in a separate devolutilizer zone, not unlike the Lurgi process which takes advantage of countercurrent flow of chunks of solid coal and hot combustion products. In contrast to Lurgi which produces a gaseous product at a relatively low temperature containing substantial amounts of tars, the separate devolutilization sections in most gasifiers under development are operated at temperatures that are high enough to eliminate tars. In the Bi-Gas process, for example, the devolutilization section exists in the same pressure vessel as the combustion zone and the hot gases from the latter pass through a restriction which separates the two zones. In the atmospheric process proposed by Combustion Engineering, the differentiation by zones is even less distinct with pulverized coal merely being injected into the hot gaseous products from the combustion zone in much the same manner as the coal is injected into the combustion zone. Even in some processes which utilize fluidized beds in both the devolutilizer and combustion zones - such as the Westinghouse-Bechtel Advanced Coal Gasification System - the principal heat transfer between the zones is a result of the fact that the hot gases from the combustion zone issue directly into the devolutilizer.

Several processes make use of solids heated in the combustion zone to transfer heat to the devolutilization zone. This has the advantage of permitting the use of air in the combustion zone without diluting the product gases with nitrogen. It has the disadvantage of producing two gaseous
streams, one normally containing $\text{SO}_2$ with lots of nitrogen and the other a relatively high Btu gas normally containing $\text{H}_2\text{S}$.

In the Bureau of Mines Coal Hydrogasification process, a synthesis gas is produced from the char resulting from hydrogenation of the coal. The synthesis gas is then shifted to obtain relatively pure hydrogen. "Grog" is the name given to the solids that are heated in the fluidized bed combustor and transferred to the synthesis gas producer to supply the heat required by the water shift reaction [Reaction (2)]. Grog is a mixture of ash and unburned carbon. In the TOSCOAL pyrolysis process, ceramic balls are heated in a combustion zone and used to supply the heat required for pyrolysis.

The $\text{CO}_2$ Acceptor system utilizes not only the sensible heat of limestone but relies on the reversible calcining reaction

$$\text{CaO} + \text{CO}_2 \rightleftharpoons \text{CaCO}_3$$  \hspace{1cm} \text{(exothermic)} \hspace{1cm} (11)

$$\text{CaCO}_3 \rightleftharpoons \text{CaO} + \text{CO}_2$$  \hspace{1cm} \text{(endothermic)} \hspace{1cm} (12)

The exothermic reaction occurs in the devolatilizer, not only supplying the energy required for devolatilization, but simultaneously reducing the $\text{CO}_2$ content of the product and increasing its Btu content. This latter factor is not nearly as important from the point of view of electric power generation as in the production of synthetic natural gas.

In the Electrochemical version of the HYGAS process (IGT) the energy required for the water shift reaction [Reaction (2)] was supplied by passing electrical current through a bed of
coal char. If electrical energy is generated with coal as the fuel, this process cannot compete with partial coal combustion as a heat source.

Removal of a Majority of the Ash

If one problem area were to be identified as making coal much less desirable as a fuel than natural gas and/or hydrocarbon liquids it would almost have to be the ash in coal. As labor costs go up and coal is mined by machines, more and more "dirt" is included with the coal in addition to its inherent ash content. Therefore coals with ash contents as high as 25-35% are not uncommon. The problems of removing the ash from the coal gasification reactor are further complicated if the gasifier is operated under pressure. Developmental work in this area has pretty much paralleled those in the utility industry and are conveniently classified as dry bottom, agglomerating or slagging.

The Lurgi slowly-moving bed gasifier sifts the coal ash through a grate located at the bottom of the gasifier and removes the ash through a lock hopper after quenching in water. When a fluidized bed is incorporated in the design of a gasifier, a partial separation of the ash and fluidizing medium occurs in the bed thereby permitting removal of an ash which is low in carbon relative to the major portion of the bed. Processes that produce a dry ash operate at relatively low temperatures.
As ash is heated from a lower to higher temperature, some softening takes place and the ash tends to stick together. In the self-agglomerating processes under development by IGT, CCNY and Battelle, the agglomerating action of the ash tends to produce larger sized ash particles. This facilitates separation of ash from unburned carbon fines, physical removal of the ash, and reduction of fly ash carry over. Whereas, one might hope that the gaseous product from a self-agglomerating gasifier after treatment by cyclones would be sufficiently free of fines to permit its use in a turbine without further reduction of the amount of particulates, the feasibility of this has never been demonstrated.

At even higher temperatures the ash melts (slags) and is removed from the gasifier as a liquid. The molten ash is generally quenched to form small, fractured, glass-like particles. This practice is not unlike that of existing boilers. In the Koppers-Totzek process, the amount of oxygen used is adjusted to insure that the gasifier operates at about 2800°F thereby insuring slagging conditions independent of the heating value and moisture content of the fuel. Other processes under development based on the Bi-Gas exploratory work operate the combustion zone under slagging conditions

**Removal of Sulfur**

Sulfur compounds in coal are converted in gasification to sulfur compounds which are fairly easily separated from the synthesis gas. This separation may take place in the
gasifier with sulfur reacting with limestone or sodium carbonate to form sulfide. In other instances the sulfur compounds are converted to $H_2S$ which is subsequently removed from the gas.

Removal of sulfur by reaction with limestone in a fluidized-bed gasifier is similar in some respects to the removal that occurs in a fluidized bed boiler and is different in other aspects. It is similar in that at least some removal is possible with this technique and that regeneration of the spent solids requires substantial development. Unslaked lime, CaO, is probably produced in both cases. Important differences are that CaS produced in the reducing atmosphere presents disposal problems at least equal to and probably in excess of those associated with the CaSO$_4$ produced in the oxidizing atmosphere of the fluidized bed boiler. Secondly, apparently removal of $H_2S$ by limestone is favored by increased temperatures whereas SO$_2$ removal in atmospheric pressure fluidized bed boilers must occur at temperatures at or below 1550-1600°F.

The Westinghouse-Bechtel advanced gasifier is designed to accomplish most if not all of the required removal of sulfur in a bed of fluidized bed of limestone. Some thought is being given both to subsequent treatment for removal of $H_2S$ if required and to treatment of the CaS for disposal purposes. The CO$_2$ acceptor system, similarly accomplishes partial removal of the $H_2S$ with limestone. Results on regeneration of the limestone in the combustor have not been particularly encouraging.
In gasifiers utilizing a molten bath, it is proposed that the sulfur will end up in the slag as it does in steel making. The ATGAS process is based on the use of a molten bath of iron with sulfur removal in a limestone slag. The Kellogg molten salt process makes use of molten sodium carbonate with the sulfur appearing as sodium sulfide in the melt. Slag desulfurization and salt regeneration complicate the processes.

The treatment of the products of combustion for removal of sulfur is facilitated not only by the fact that $H_2S$ is easier to absorb than $SO_2$ but also by the facts that the $H_2S$ is at higher concentrations than it would be as $SO_2$ in the stack gases and that such absorption is facilitated at elevated pressures. The process of $H_2S$ removal is complicated by the fact that it is generally desirable, from the point of view of thermodynamic efficiency and/or reduction of required, expensive heat exchange surface, to remove the $H_2S$ at elevated temperatures.

There are numerous commercial hydrogen sulfide and acid gas removal processes available. These processes fall into three broad categories:

- Absorption into a solvent
- Chemical conversion into another compound
- Adsorption on solids

The optimum process depends upon process requirements and conditions before and after desulfurization.
The majority of the acid gas processes are absorption systems. They are the most economical at present. Good absorber efficiency comes from operating at low temperatures and high pressures (100-2000 psia). Therefore, gas quenching and cleaning usually follows gasification and precedes absorption. Chemical absorption (as in one of the amines) is best at low acid gas partial pressures and physical absorption (as in Selexol—dimethyl ether of polyethylene glycol) is best when the partial pressures are high. Thermal efficiency is an important criterion in process selection. Extremely low H₂S concentrations are difficult to obtain by absorption alone. Solvent regeneration is required. The incremental cost benefit of operating at pressures greater than 400 psig is usually small.

Chemical conversion processes can operate at higher temperatures than absorption systems. This improves the overall gasification thermal efficiency. The reagents are solids and require regeneration. Typical reagents are zinc oxide (ZnO), ferric oxide (Fe₂O₃) at 600-650°F and calcium oxide (CaO), the active part of calcined dolomite. Good removal of H₂S is potentially possible with dolomite at 1450-1600°F. Work done by Consolidation Coal Company indicates that not all dolomites are effective and regeneration is not always complete. There have been reports that limestone or dolomite can suddenly lose its reactivity and regeneration is impossible. The reasons for this are not completely understood.
Solid adsorption involves the capture of $\text{H}_2\text{S}$ on the surface of a solid adsorbent such as iron sponge, molecular sieve type 13x or charcoal, usually at high pressures and low temperatures. Adsorption works best for low concentrations. It is used to further reduce $\text{H}_2\text{S}$ levels following an adsorber and at the same time remove traces of organic absorption solvents. Very low levels of both are obtainable.

Table IV lists some of the commercial absorption processes for $\text{H}_2\text{S}$ and CO$_2$ removal to satisfy pipeline gas specifications.

In the production of high Btu pipeline quality gas the principal desulfurization step will usually be located after the shift conversion where close to 90% of the CO$_2$ is removed along with the $\text{H}_2\text{S}$.

The rich hydrogen sulfide (and CO$_2$) produced from the absorber solvent regeneration step is recovered in a Claus unit. The $\text{H}_2\text{S}$ is burned with sufficient air in the presence of a bauxite catalyst to satisfy the stoichiometry of the Claus reactions

$$2\text{H}_2\text{S} + 3\text{O}_2 = 2\text{H}_2\text{O} + 2\text{SO}_2$$ (13)

$$2\text{H}_2\text{S} + \text{SO}_2 = 2\text{H}_2\text{O} + 3\text{S}_{x/\text{x}}$$ (14)

and sulfur is condensed out. Two or three reactors are required in series in addition to tail gas clean-up, often by a Beavon sulfur removal unit, to satisfy EPA emission standards.
TABLE IV

Summary of Commercial H₂S and Acid Gas Absorption Process

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<th>Absorbent</th>
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<td>600-1100</td>
</tr>
<tr>
<td>Sulfinol</td>
<td>Shell</td>
<td>Tetrahydrothiophene</td>
<td>~ 1000</td>
</tr>
</tbody>
</table>

* Will vary depending upon process and sulfur removal requirements.
The Consolidation Coal Company has experimented with a liquid-phase Claus reactor at 300°F to recover elemental sulfur by decomposition of $\text{H}_2\text{S}$ in the presence of $\text{CO}_2$. This is in a very early development stage in conjunction with their high temperature sulfur removal work.

**Gas Liquor Treatment**

Gas liquor is the sum of the aqueous streams condensed in the coal gasification and gas processing areas by quenching and scrubbing. They contain phenols, ammonia, carbon dioxide and hydrogen sulfide among others. Phenols can be removed and recovered by the Lurgi Phenosolvan Process. Other contaminants can be removed by heating and stripping.

**Removal of Particulates and Contaminants**

The preceding two sections contained a discussion of some of the problems associated with removal of the majority of the ash and reduction of sulfur in the effluent gas. In order for the product gas to be acceptable for almost any service it will be necessary to remove tars and eliminate fly ash either prior to, during or following subsequent combustion. Further, if the gas is to be used in a turbine, special attention must be given to chemical contaminants such as alkali metals, mercury and chlorides commonly found in coal. In fact, it is important that practical limits be established on the size and amount of particulates plus the nature and amounts of chemical contaminants in gases which serve as fuels in gas turbines.
Most of the gasifiers under development are designed to operate at temperatures high enough to eliminate tars and low enough to yield some of the lower hydrocarbons, principally methane, CH₄. The Lurgi process yields gas at a low temperature (~1000°F) that is loaded with tar. At present no attempt is made to use the sensible heat of this stream to generate steam because of the presence of both tar and fly ash. Instead, a water quench and clean up is utilized and the condensed tars are recycled to the gasifier.

Steam is generated in operation of the Koppers-Totzek gasifier. Special materials of construction are utilized because of the reducing atmosphere.

Several procedures have been suggested for removing all particulates down to sub-micron size. These range from advanced cyclones through impingement devices to the flowing sand filter under development at CCNY. The Westinghouse-Bechtel group has a man studying this subject under their OCR contract and any report should be of interest.

Several groups are very interested in the problems associated with the presence of alkali metals, mercury and other chemical contaminants in the gas produced from coal. There is some indication that the temperature of operation of pressurized fluidized bed boilers is more limited by the volatility of such contaminants than by sulfur removal with limestone.
Energy Efficiency

As the price of suitable fuels increases, more and more attention will be given to improving the thermal efficiency of processes used to generate electric power. It is indeed unfortunate that the premium fuels will apparently not be available for electric power production just at a time when development of the combined cycle gives promise of eventually achieving efficiencies approaching if not exceeding 50% during the next decade or so. (See Section entitled The Combined Cycle in Relation to Coal as a Fuel in Part III of this report).

The combined cycle efficiencies are markedly better than any presently attainable from conventional steam power plants fired either with low sulfur coal or utilizing a fluidized bed combustor, i.e., 36-38%. Use of stack gas cleaning will reduce these efficiencies to 33-37% - with the lower figure being the more probable.

Gasification processes have thermal efficiencies in the 65-72% range. Thus unusual circumstances would have to prevail to justify use of the gasifiers to produce fuel for conventional steam cycle power plants. Some retrofit situations might provide such justifications. In building new plants it appears that the thermal inefficiencies inherent in gasification processes might only be justified in comparison with stack gas cleaning or in combination with the higher thermal efficiency and lower capital costs of the
combined cycle. On this basis, as discussed in Part III, it is essential that the thermal efficiencies of actual and/or proposed gasification processes exceed 75%, before the coupling of a gasifier with a combined cycle can be justified.

A number of factors influence the thermal efficiency of a gasifier. Any large temperature swings – such as are often required to remove $\text{H}_2\text{S}$ – will result in a decrease in thermal efficiency. Similarly the water quench necessitated by tars in the gases from the Lurgi gasifier results in a seemingly unavoidable decrease in thermal efficiency. Even if waste heat can be used to generate steam, the necessity of removing $\text{H}_2\text{S}$ at low-temperatures will result in a reduction of efficiency. In addition it is important to recognize that anytime a reaction occurs at a reasonable rate the thermal efficiency suffers. In some cases thermal efficiency can be increased by heat exchange between streams but such exchange surface is usually very expensive.

CONCLUSIONS AND RECOMMENDATIONS

Commercial low-to-intermediate Btu coal gasification processes are currently available today through Lurgi and Koppers. They represent two diverse methods of gasification being fixed-bed and slagging entrained-flow gasifiers, respectively. The Lurgi gasifier is limited to non-caking coals at this time. The Winkler fluidized bed gasifier is also available. It is limited to non-caking coals. To date there has been no experience in coupling a gasifier system to a
conventional power plant or a combined cycle plant. Control experience for such systems is needed. Capital cost of the available intermediate Btu gasifier systems is believed to be about $170-$180/KW and the thermal efficiency is about 70%.

Coal gasification processes find their greatest opportunities in conjunction with combined cycle plants. The higher thermodynamic efficiency of such combined plants tend to offset the energy lost in gasification. It will be several years before the overall thermal efficiency matches that of a conventional plant.

Developments through the pilot plant scale in this country have evolved from the Lurgi and Koppers-Totzek gasifiers. The work is directed toward the capital cost reduction and the thermal efficiency improvements which are theoretically possible. It does not appear likely that commercial-size plants will be operating however before the 1980's based on these developments.

Low and intermediate Btu gasification processes must be located at or near the electric power plant because of the expense in transporting lower Btu gases. It is expected that coal gasifiers will have to follow load. Storage costs would be excessive except perhaps where underground storage is available. From an economic standpoint, base-load operations would be preferred since the gasification plant capacity must match the electric power generating capacity.

Coal gasification systems vary in complexity, start-up and shut down capability and turn-down. Unless the process
is simple, reliable and amenable to control, it will be difficult to couple to an electric power generation system. Turn-down of 50% for individual gasifiers seems likely for most systems. Environmental problems should be minimal.

The Combustion Engineering atmospheric two-stage slagging entrained flow gasifier concept is reasonable, simple, offers minimum development problems and is directed toward reducing or eliminating the oxygen requirements of a Koppers-Totzek gasifier. Support for this program is therefore recommended.

Foster Wheeler has proposed a demonstration size plant. Although proven equipment is employed where possible, it represents a bold effort. The gasifier would be conceptually similar to the Bi-Gas and Combustion Engineering gasifier and would operate at 500 psi.

Hydrogen production by a single stage slagging gasifier for hydrogen production is needed. A process which is economical and utilizes debris of liquefaction processes should be developed for the coal dissolution processes. A hydrogen generation unit should be incorporated into a dissolution process, with candidates recommended: Koppers-Totzek, Texaco or Shell gasifier processes.

The process analysis based on laboratory data shows that molten salt gasification and desulfurization has a possibility of low cost and simple construction. Salt regeneration is based on known technology although the ash is a definite complication. The process is worth support through the next step of development to evaluate its feasibility.
Process Descriptions

AMERICAN LURGI CORPORATION--LURGI PROCESS

The Lurgi process is one of the oldest and has been fairly common in Europe. The first commercial plant was erected in 1936 and 58 plants have been constructed since. Lurgi has also been a leader in the development of power plant systems in which a gasifier is used in conjunction with a turbo-expander or gas turbine, and a conventional boiler for power generation. It is one of the few processes to reach commercial use at present (16,17,18).

In the Lurgi process non-caking and weakly caking lump and/or briquetted coal (3/16 - 1-3/4 inch) are fed through a coal lock system at the top of the reactor into a water-jacketed gasifier as shown in Figure 4. Gasification takes place in a fixed bed configuration with oxygen or air and steam blowing up through a revolving grate. Continuous operation in gasifiers up to four meters in diameter is accomplished by removing dry ash through an ash lock. The temperature in the bed varies from 700° -800°F at the top to a high of about 2400°F two-thirds of the way down. The ash leaves the grate at 700°F and raw gases must be cleaned to remove dust and tar before desulfurization.

When operated at 300 psia and 600-1200°F with air, Lurgi gas will contain (dry basis) approximately 5% CH₄, 25% H₂ and 16% CO with a heating value of about 150 Btu/scf. Besides its use as a synthesis gas it also could be used in a combined cycle plant, as is presently being evaluated at STEAG's Lünen, Germany power plant.
The critical zone in the gasifier is the space above the distributor and at the top of the bed where coal is heated to devolatilizing temperatures. If caking or semi-caking coal is heated too rapidly in the presence of hydrogen, it will swell and form caking clinkers. When this occurs before solids pass the distributor and eventually caking clinkers will block the flow of coal.

The Lurgi gasifier seems to demand good control and close attention by operators. If the coal has caking tendencies, oxygen and steam rates must be controlled at levels which minimize or prevent caking. Subbituminous western coals are probably suitable, and some information may be forthcoming from tests on this type of coal at the Westfield, Scotland plant.

The main disadvantages of this gasifier, are, then:

1. Low thermal efficiency
2. Limitations on types of coal
3. Tar formation
4. Complicated, expensive construction
5. Sensitive operation required
6. Relatively low gasification rates, requiring high reaction volume and multiple units.

The essential advantage of the Lurgi unit is that its operability has been proven in service. Although this gasifier has been used in commercial plants, it is said that the design is essentially unchanged since the construction of the SASOL plant nearly twenty-five years ago. In view of the
number of organizations which have shown interest in the gasifier, experience with American coals is needed. The El Paso Natural Gas Company has proposed a development project to obtain information in the following areas:

1. Capacity. The effect of coal type on the throughput capacity has not been explored sufficiently. Appreciable savings in plant costs and in fabrication/delivery time could be realized by knowing which coals are best suited to this gasifier. There is a possibility that present designs may be conservative and that capacity increases may follow experience with operation.

2. Pressure. A development gasifier design is proposed at pressures about one-third greater than previous Lurgi experience. If comparable there are substantial savings realizable in reduction of the number of units required, and in possible increases in the methane content of the gas.

3. Mechanical improvements. There are potential improvements possible in the coal-lock system and in the coal distribution mechanisms, for example.

4. Coal size. Coupled with the mechanical modifications is the possibility that the design and operation can be changed to enlarge the size range to permit inclusion in the feed of sizes as small as 1-5 millimeters. This would substantially reduce the amount of briquetting now required.

5. Air blown gasification. It is desirable to explore the production of low Btu gas for use in combined cycles. Some experience may be available from current air blowing tests at Lünen.
6. By-products development. The disadvantage of tar formation might be turned around by exploring the operation to produce saleable by-products—light tar, naphtha, phenols, ammonia and so forth. Market development and liquids processing might be desirable.

7. Process optimization. Complete studies of the effects and interactions of parameters such as pressure, temperature, oxygen and steam flow ratio, bed depth, should be made.

8. Char as a feed stock. Several of the projected clean coal processes produce varying types of char. The success or value of these processes may depend upon use or disposal of char.

9. Control systems. There is a need to develop experience in control, particularly computer-control of this process with its multiple parallel units. There is a possibility that present gasifier designs are conservative and that significant improvements in throughput capacity can be realized by using the ability of modern control and information systems.

Prepared by Brymer Williams
KOPPERS CO., INC. - KOPPERS-TOTZEK COAL GASIFIER

The Koppers-Totzek gasifier is a single stage entrained flow, ash slagging gasifier which operates at a slight positive pressure. Based on the concept of Dr. Friedrich Totzek of H. Koppers, the first unit was built for the U.S. Bureau of Mines and tested at Louisana, Missouri (19). At present, there are 16 commercial plants either in operation or being built which use the gasifiers. In most of these plants hydrogen is produced for ammonia synthesis by coal gasification (20).

In the Koppers-Totzek gasifier, pulverized and dried coal is screw fed to the gasifier feed nozzles where it is entrained in oxygen and low pressure steam. By keeping the feed nozzle at moderate temperatures and by maintaining a high velocity through the nozzles, the coal is entrained in a somewhat dilute phase into the gasifier where the reactions of the coal, oxygen and steam occur. The character of the coal is not critical. It should work well on caking coals and on high ash coals. A schematic of the gasifier is shown in Figure 5.

Gasifiers are designed with either 2 or 4 feed nozzles operated in pairs spaced 180° apart. The nozzles are nominally horizontal although pointed down slightly.

The reaction temperature at the burner discharge is 3300-3500 F. At this temperature nearly all the carbon reacts instantaneously with oxygen and steam to produce carbon monoxide,
hydrogen and molten slag. No tars, condensable hydrocarbons
or phenols are formed. As a result of the endothermic reactions
and radiant heat absorption by the refractory walls of the
gasifier, the exit gas temperature is about 2750 F. Approximately
50% of the coal ash drops out as molten slag into the quench
tank below the gasifier and the remaining ash is carried out
of the gasifier as fine fly ash.

The gasifier outlet is equipped with water spray nozzles
to cool the gas temperature to below the ash fusion temperature
to prevent the ash from sticking to the tubes in the waste
heat boiler mounted above the gasifier.

The amount of water required is usually small although sig-
nificant cooling could be required if alkali metal vapor
corrosion of the waste heat boiler tubes becomes a problem.
This has not been a problem on the coals used.

Sufficient oxygen must be used to maintain satisfactory
ash fusion temperatures in the gasifier. At the temperatures
required, 11-12% of the carbon is converted to carbon dioxide.
When insufficient oxygen is used, indicated by lower carbon
dioxide concentrations, ash clinkers can form. Flux can be
added to the coal feed to adjust the ash fusion characteristics.
The cost of flux would have to be offset by the reduction in
oxygen costs. Molten slag is quenched in water below the
gasifier and the granular solid ash carried off to disposal.

The gases leave the spray section above the gasifier at
2200-2400 F. Part of the cooling comes from water evaporation and part from hydrogen formation.

The hot gases then pass into a waste heat boiler containing a radiant section and a convection section in series where the gases are cooled to about 350 F. Superheated, high pressure steam up to 1500 psig is produced in excess of total process requirements. There is more than enough steam to drive the air compressors for the oxygen plant and to supply steam for the gasifier.

After leaving the waste heat boiler at 350 F, the gases are cleaned and cooled to about 150 F in a high energy scrubbing system which reduces the entrained solids to 0.002-0.005 grains/scf.

Particulate-laden water from the gas cleaning and cooling system is piped to a clarifier. Sludge from the clarifier is pumped either to a filter or to the plant disposal area. The clean water overflows into a cooling tower and is recirculated through the gas washing system. Evaporation, windage and blow-down water losses at the cooling tower, plus moisture in clarifier sludge and in slag, necessitate the addition of a small quantity of makeup water to this system. If water is at a premium, air cooling may be used for certain applications and the cooling tower can be reduced in size to provide only the final trim in water temperature.

The cool, clean gas leaving the gas cleaning system contains hydrogen sulfide and other sulfur compounds which must be removed to meet gas specifications. The type of system
chosen depends upon the end use and pressure required of the product gas. For low pressures (up to 150 psig) and low Btu gas applications, there are chemical reaction processes, such as amine and carbonate systems. At higher pressures, the physical absorption processes, such as Rectisol, Purisol and Selexol, are used. The choice of the process is also dependent upon the desired purity of the product gas and the desired selectivity, with respect to the concentrations of carbon dioxide and sulfides.

The Kopper-Totzek gasifier is a very simple one stage piece of equipment and has a very high coal processing capability even at atmospheric pressure because of the high temperature. The gasifier has a steel shell with a water jacket to produce low-pressure process steam. The inside of the gasifier is lined with a thin chrome type refractory. The wall temperatures are maintained below the ash slagging temperature and slag tends to build up during operation and provides additional protection. In Coruna, Spain, the gasifiers have operated satisfactorily without any refractory lining with high ash lignites (21). The only moving parts associated with the gasifier system are screw feeders for solids handling and pumps for liquid circulation.

Start-up, shut-down and control are all simple operations. It takes about 30 minutes to start up a Koppers-Totzek gasifier from a cold condition. Supplementary fuel gas or oil
is used to heat up the gasifier and light off the coal-oxygen mixture. If the gasifier is hot, start-up time can be reduced to about 10 minutes (22).

Shut-down can be effected by stopping the flow of coal and oxygen. The gasifier is then immediately filled with steam or nitrogen to prevent flash back of synthesis gas into the gasifier. Hold-up in the system is very small so any change is seen almost immediately. As a safety precaution, oxygen flow is immediately stopped and the gasifier purged with either steam or nitrogen whenever the coal feed into the gasifier is interrupted.

Control is very simple. As long as the coal feed rate is uniform, the entrained flow nozzles provide uniform operations. A constant head of dry pulverized coal is required above the screw feeder. If the pressure build-up in the nozzle, the oxygen flow is stopped. The flow rates of oxygen and steam are regulated to give the proper operating conditions. Turn-down to about 60% of normal flow is possible with each pair of feed nozzles before operations become difficult to control. With a four-headed gasifier a turn-down to 30% is possible by shutting off one pair of nozzles completely.

A two-headed gasifier is capable of gasifying over 400 tons/day of coal and a four-headed gasifier is capable of gasifying 850 tons/day of coal. Carbon utilization is good. Data on a 35% ash coal showed only 0.56% carbon in the slag.
The gasifier can operate on up to 45% mineral matter in the coal. It can also operate on fuel oils containing large amounts of mineral matter by atomizing the oil to a fine mist with a gas (oxygen) atomizing nozzle. After ignition, the oil mists at first reacts with the admixed oxygen. The gaseous oxidation products and the non-oxidized oil secondarily react with steam which is also fed through the burner to produce hydrogen and carbon monoxide. A heavier steel lining is required with oil gasification because of the low ash content.

The overall thermal efficiency going from coal and oxygen to 350 F gases and steam is between 85 and 90% depending upon the coal. These numbers do not include the energy required to produce oxygen, energy losses in desulfurization and energy requirements for gas compression to pressures suitable for combined-cycle applications. When those requirements are included, the overall thermal efficiency for the production of a 295 Btu/sch (HHV) gas at 150 psig is estimated at 72%. Typical data are given in Table V.

The Koppers-Totzek gasifiers require a large amount of oxygen as shown in Table V. The cost of a captive oxygen plant to support a gasification plant is 1/2 the cost of the gasification plant. If air were to be used to replace oxygen, air preheat would be required to maintain slagging conditions in the gasifiers. The nitrogen in the air would nearly double the total gas flow rate. To accommodate the additional gas
### TABLE V

**Typical K-T Gasifier Data for U.S. Coals**

<table>
<thead>
<tr>
<th>Type of Coal</th>
<th>Western Coal</th>
<th>Illinois Coal</th>
<th>Eastern Coal</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>GASIFIER FEED</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dried Coal to Gasifier Analysis - Vol. %</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>56.76</td>
<td>61.94</td>
<td>69.88</td>
</tr>
<tr>
<td>H₂</td>
<td>4.24</td>
<td>4.36</td>
<td>4.90</td>
</tr>
<tr>
<td>N₂</td>
<td>1.01</td>
<td>0.97</td>
<td>1.37</td>
</tr>
<tr>
<td>S</td>
<td>0.67</td>
<td>4.88</td>
<td>1.08</td>
</tr>
<tr>
<td>O₂</td>
<td>13.18</td>
<td>6.73</td>
<td>7.05</td>
</tr>
<tr>
<td>Ash</td>
<td>22.14</td>
<td>19.12</td>
<td>13.72</td>
</tr>
<tr>
<td>Moisture</td>
<td>2.00</td>
<td>2.00</td>
<td>2.00</td>
</tr>
<tr>
<td>Gross Heating Value Btu/lb</td>
<td>9,888.</td>
<td>11,388.</td>
<td>12,696.</td>
</tr>
<tr>
<td>Oxygen-NT/NT Dried Coal</td>
<td>0.649</td>
<td>0.704</td>
<td>0.817</td>
</tr>
<tr>
<td>Purity-%</td>
<td>98.0</td>
<td>98.0</td>
<td>98.0</td>
</tr>
<tr>
<td>Process Steam-lbs/NT Dried Coal</td>
<td>272.9</td>
<td>541.3</td>
<td>587.4</td>
</tr>
<tr>
<td><strong>GASIFIER PRODUCTS</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Jacket Steam-lbs/NT Dried Coal</td>
<td>347.8</td>
<td>404.9</td>
<td>464.9</td>
</tr>
<tr>
<td>High Press. Steam-lbs/NT Dried Coal</td>
<td>2,147.1</td>
<td>2,292.2</td>
<td>3,023.6</td>
</tr>
<tr>
<td>Raw Gas Analysis (Dry Basis) Vol. %</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO</td>
<td>58.68</td>
<td>55.38</td>
<td>55.90</td>
</tr>
<tr>
<td>CO₂</td>
<td>7.04</td>
<td>7.04</td>
<td>7.18</td>
</tr>
<tr>
<td>H₂</td>
<td>32.86</td>
<td>34.62</td>
<td>35.39</td>
</tr>
<tr>
<td>N₂</td>
<td>1.12</td>
<td>1.01</td>
<td>1.14</td>
</tr>
<tr>
<td>H₂S</td>
<td>0.28</td>
<td>1.83</td>
<td>0.35</td>
</tr>
<tr>
<td>CÔS</td>
<td>0.02</td>
<td>0.12</td>
<td>0.04</td>
</tr>
<tr>
<td>TOTAL</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
</tr>
<tr>
<td>Gross Heating Value, Btu/SCF</td>
<td>295.1</td>
<td>290.2</td>
<td>294.4</td>
</tr>
<tr>
<td>Gas Make - SCF/NT Dried Coal</td>
<td>51,783.</td>
<td>59,489.</td>
<td>66,376.</td>
</tr>
<tr>
<td>Slag Make - NT/NT Dried Coal</td>
<td>0.22</td>
<td>0.190</td>
<td>0.138</td>
</tr>
<tr>
<td>Process Efficiency</td>
<td>88.2</td>
<td>85.0</td>
<td>90.3</td>
</tr>
<tr>
<td>Coal to Gas Efficiency</td>
<td>77.3</td>
<td>75.8</td>
<td>77.0</td>
</tr>
</tbody>
</table>
flow, either the number of gasifiers would be increased or the piping increased in size. The cost of air preheaters and additional gasifiers would be far in excess of the oxygen plant costs. Adequate steam is produced in the gasifiers to operate the oxygen plant.

The gas clean-up equipment can serve several gasifiers. The capital equipment cost of an oxygen plant and a gasification plant to produce a 290 Btu/scf gas at 150 psi for a combined cycle power plant fuel is estimated at $170/KW.

The Koppers-Totzek gasifier should be a very suitable gasifier for hydrogen production from the carbonaceous residues from coal liquefaction plants. The residue either dry or liquid, contains large amounts of mineral matter in addition to the carbon. There would be an advantage to gasifying a liquid since the gasifier could be operated at 150 psig or above by slurry pumping the feed into the gasifier. Lock hoppers would be needed for gasification of dry residues at such pressures. A liquid residue is possible if the mineral matter is separated from the dissolved coal extract by vacuum distillation. A dry residue will not contain enough carbon to satisfy the hydrogen requirements so supplementary coal is needed.

Prepared by Dale E. Briggs
WINKLER COAL GASIFIER (DAVY-POWER GAS, INC.)

The Winkler gasifier is a single stage fluidized bed gas generator which operates at atmospheric pressure with air or oxygen (23).

The Winkler gasifier was conceived in 1922 and the first commercial producer put into operation in 1926. Since then, 26 producers in 16 installations have been put into operation.

As shown in Figure 6, crushed coal, dried to 8% moisture or less, is screw fed into the gasifier near the bottom. Steam and oxygen keep the solids fluidized and well mixed. The temperature is maintained at 1500-1800°F which brings about cracking of tars and heavy hydrocarbons. As a result of fluidization, the larger and heavier particles fall down through the bed and pass into the ash discharge screw at the bottom of the generator.

The dust laden gases are cooled by heat exchange to produce process steam and are partially cleaned in a cyclone to produce a synthesis gas with a higher heating valve on a dry basis of 287 Btu/scf.

The commercial gasifiers operate on non-caking coals. Highly caking coals may be difficult to feed into the gasifier. Once the coal is dispersed into the char of ash caking problems are minimized.

The Winkler gasifier is rather large for its throughput
Figure 6. Winkler Coal Gasifier

- Crushed Coal
- Fluidized Bed Gasifier
- High Pressure Superheated Steam
- Water
- Air or O₂
- 350°F Synthesis Gas
- Cyclone
- Process Steam
- Ash

Temperature: 1500°F - 1800°F, 1 Atm.
since it operates at 1 atmosphere pressure and moderate temperatures.

Energy recovery from the hot synthesis gases improves thermal efficiency.

Prepared by Dale E. Briggs
BITUMINOUS COAL RESEARCH - BI-GAS

Work in this process began in 1965 and evolved into a two-stage super-pressure entrained flow gasifier (24,25,26). shown in Figure 7. Most of the research has been directed to the Stage 2 reactor, the technology for Stage 1 being fairly well established. Small autoclave studies on 5-gram charges (24) and on a 100 lb/hr internally-fired reactor (27) have resulted in the design by Koppers of a 120 tons/day pilot plant at Homer City, Pennsylvania which is scheduled to begin operations in 1974 (2). Work in the pilot plant will be directed toward optimizing methane yield and the flow patterns in the gasifier.

The pilot plant is being constructed to raise fresh fine coal (70% through 200 mesh) to the gasifier pressure of 1000 to 1500 psig by pumping a coal-water slurry to a coal feed hopper where the water is removed and the coal partially dried. This method was advocated by the Office of Coal Research as an alternative to lock hoppers. There is a substantial thermal penalty associated with coal drying. It was the opinion of Diehl (28) and others at BCR that lock hoppers are satisfactory up to 10 atmospheres but are not too satisfactory above.

Fresh coal is fed into the upper section (Stage 2) of the gasifier by entraining the coal in recycle gas and super heated steam.
Figure 7. Bituminous Coal Research Bi-Gas Coal Gasification System
Since the coal is blown into the gasifier in a dilute, entrained state, caking coals can be utilized without pretreatment.

Hot gases (carbon monoxide, hydrogen and water) at 2700°F from the lower section (Stage 1) provide heat for devolatilization of the coal and thermal cracking of oils and tars. As the feed coal devolatilizes, it expands into a low bulk density char with about 2/3 of the mass of the original coal. The synthesis gas and char leave Stage 2 at about 1700°F. At the high pressures and moderate temperatures the methane yield is high and the amount of oils and tars minimal.

The entrained char is separated from the synthesis gases in a cyclone separator. A 99% collection efficiency is expected. This is vital to realize high carbon utilization.

Char is collected in hoppers from where it is entrained in superheated steam and fed tangentially into the combustor (Stage 1) with oxygen to give vortex flow similar to cyclone slagging-bottom coal burning boilers. Stage 1 operates at about 2700°F under ash slagging conditions. The high carbon char is almost completely converted to carbon monoxide with a part of the heat released used to reduce steam to hydrogen.

The two-stage BCR gasifier requires less oxygen than the single stage Koppers-Totzek gasifier since roughly only 2/3 of the feed coal is partially oxidized in the combustion
stage. It is also hot, coming from Stage 2.

The molten slag is water quenched and collected in slag hoppers from which it is periodically removed.

A 380 Btu/scf gas is produced at system pressures in excess of 1000 psig when operating with oxygen (29). When operated at 300 psig with air, a gas is produced which may be desulfurized and cleaned to provide a 175 Btu/scf gas (30).

The BCR Bi-Gas gasifier was originally developed for the synthetic natural gas program. In the AGA-OCR Synthetic natural gas supported process, the operating pressures have usually been 1000 psig. There is no particular advantage in going higher and according to Siegel (31), ESSO R&D, a pressure of 500 psig may be adequate. For low Btu gas generation, system pressures of 200 psig are more than adequate. Diehl (28) and others at BCR indicated that 150 psig seems to be optimum for combined cycle applications.

The equipment is relatively simple with no moving parts. The gasifier stages and cyclone are lined with insulation and refractory to protect the metals from the high temperatures. Although they have been used in other gasifier applications, refractory and insulation integrity at high pressures and temperatures is always a potential problem.

Gasifier control is effected by regulating the amount of oxygen and steam to Stage 1 to maintain the proper ash slagging conditions. Since coals from the same vein can vary
substantially, ash slagging must be watched carefully. Some problems have been encountered in molten ash removal through small openings but this is not expected to be a problem in larger gasifiers. Char feed is regulated to maintain a constant level in the char feed hoppers. The coal and steam fed to Stage 2 must be regulated to control the exit temperature from the stage. In addition, the coal feed rate must be reasonably balanced to the char consumed in Stage 1, although some supplementary coal can be fed to Stage 1. Overall control of the system should prove satisfactory once operating experience is gained from pilot plant operation.

Turn-down of about 50% should be possible. The pilot plant operations will provide valuable information on turn-down for scale-up to commercial sizes.

Wen (10) estimated the overall thermal efficiency from coal to methane for a Bi-Gas gasifier system at 63.2%. A substantial heat loss occurs in cooling the hot raw synthesis gases for hydrogen sulfide removal.

Combustion Engineering and Foster Wheeler are also doing work on entrainment-type slagging coal gasifiers to produce low Btu gas with air. They are presently doing design studies. Development will be limited to pressures of 1 to 15 atmospheres and will be specifically aimed at power plant fuel use.
BCR and OCR commissioned a study, completed in 1971, in which an air-blown two-stage Bi-Gas type gasifier was used in conceptual designs of a 500-megawatt installation (30). Nominal operating pressure was 300 psig. The best design of the three is shown in Figure 8. Maximum energy recovery is practiced. This is especially important as coal increases in cost. The capital cost per installed kilowatt was $117 and the operating cost was 29.53 cents/Btu over the coal cost.

Prepared by Dale E. Briggs
Figure 8. Conceptual Design of a Air Blown Bi-Gas Process
Combustion Engineering is completing a twelve part design study concerning the development of an entrainment-type coal gasification process suitable for producing clean low-Btu gas for electric power generation. This work has been co-sponsored by the Office of Coal Research and Consolidated Edison Company of New York (2). As a result of this study Combustion Engineering recommends the design and construction of a five ton per hour atmospheric-pressure gasifier pilot plant at their Windsor laboratories, to be followed by a full-size demonstration plant on the Consolidated Edison system. An anticipated longer range development of this process is gasification and clean-up at ten atmospheres pressure and use of the gas in combined cycle generation schemes.

The atmospheric pressure design gasifier is described by Blaskowski and Kowucky(32) and is shown in Figure 9. The gasifier is comparable to a pulverized coal fired boiler, with similar fuel injection, gas flow patterns, and heat exchange surfaces. The principal differences are the two level firing arrangement, with recycled char being fired through the combustor nozzles of the lower zone and fresh coal, steam, and air being injected through the reductor nozzles and reacted in the upper zone. The two zone injection is based upon small scale experiments at Bituminous
Figure 9. Detailed Layout of the Combustion Engineering Atmospheric Pressure Coal Gasifier
Coal Research but the basic process of entrainment gasification has been in commercial operation for two decades in the Ruhrgas, Koppers-Totzek and duPont processes. The anticipated gasification temperature is 1600-1700 F. It is proposed to recover most of the sensible heat of the gases by cooling them to 300 F as they pass over the evaporator, economizer, process steam superheater, process steam evaporator, and liquid couple coils. As seen on the flow schematic, Figure 10, the cooled gas then passes through a char collector, raw gas scrubber, and H₂S absorber. Char is returned to the gasifier lower zone after it is pulverized and fired through the combustor nozzles. To improve thermodynamic efficiency the clean gas is reheated on its return from the clean-up system to the low-Btu gas fired boiler. Combustion Engineering favors the Stretford process to remove the sulfur and reduce it to elemental form. This process has been well proven in England in treating atmospheric pressure coke oven gas, town gas, and refinery gases. Ash is removed from the process in the form of liquid slag leaving the lower, or char combustion zone at 3000°F.

An evaluation of this proposed process indicated that the coal utilization is very good. Any coal suitable for pulverizing can be handled. The carbon consumption efficiency approaches that of a coal fired steam generator, i.e. 99+%. 
Figure 10. Combustion Engineering Atmospheric Pressure Gasification System
Air processing is based on proven equipment similar to conventional pulverized-coal fired steam generators, including primary air fans, forced and induced draft fans, and air preheaters. The power required for the air and flue gas handling is less than 4% of the turbine gross output. The problem of coal and char injection into the gasifier is the same as conventional pulverized-coal fired steam generators. Every coal burning utility has an intimate familiarity with the problems associated with pulverizers, entrained coal transport, burner maintenance, and flame control and stability. The use of steam as a coal carrier fluid and the operation under sub-stoichiometric combustion conditions are believed to be modest extensions of present technology.

The devolatilization of the coal takes place while the coal particles are suspended in the hot steam - flue gas stream. The rapid heating and wide dispersion of the particles should minimize any agglomerating or sticking tendency. The calculated temperature of the gases at the top of the gasifier, before heat exchange, is 1600-1700°F. This temperature is comparable to the 1800°F temperature of gases leaving the Synthane process and, according to Forney (33) volatilized tars and oils are present in the gases from the Synthane gasifier. These tars condense as the gas is cooled below the 800-1000°F level. Based on this we believe that the process steam boiler and liquid couple tubes will become encrusted with a tar-char layer, thereby reducing the heat transfer effectiveness and perhaps causing frequent
shut-down for cleaning. The experimental gasifier should be constructed so that these heat transfer surfaces may be readily removed and hot tar-char laden gas can be handled in the down-stream equipment.

The combustion of the recycled char occurs while the particles are entrained in the lower part of the gasifier. The oxidizing atmosphere and 3000 °F temperature is conducive to complete carbon utilization as well as liquid slag formation.

The slagging conditions in the combustion section permits a liquid slag tap and water quench. The atmospheric pressure within the gasifier -- combustor makes possible relatively large slag openings, mechanical accessibility, and visual observation of slag conditions.

Particulates are removed from the gasifier product gas by cyclone separators and water scrubbing. This equipment, operating at atmospheric pressure and low temperatures, is based on proven gas cleaning technology. Presence of condensing tars may seriously foul this equipment. Recycle of char and filter cake is an area which may require development for large scale, trouble-free operation.

The sulfur removal is based upon proven processes but involves considerable scale-up and possible equipment development. C-E believes the Stretford process appears to be the most economical for use with washed, atmospheric pressure product
gas. Gas-liquid contacting equipment is of very large size due to the low pressure. Indeed, the volume of gases to be scrubbed is about 60% of the volume of the gases going to the stack, and is comparable to stack gas scrubbers. More than 95% of the sulfur can be removed and collected in the elemental form.

The equipment required to gasify coal at atmospheric pressure is very large geometrically but involves few problems or unknowns in the way of extreme temperatures, pressures, gas tightness, thermal stress, etc. Much of the equipment for full size power plants (500 MW) would have to be field erected, necessarily forgoing the economics of shop fabrication. No unusual valve conditions or fabrication techniques are visualized. The materials of construction are conventional and similar to those employed in boiler and coke oven gas equipment.

Instrumentation and control of a large scale gasification process will probably require some development which would be done at the pilot plant stage. The problems and prospects of operation at less than design rating are unknown but it is probable that derating to 50% is feasible for each gasifier. The proposed 500 MW demonstration plant would have two gasifiers, giving a turn-down capability to 25% of rating. Complete shutdown every night is considered to be more difficult than a pulverized coal fired steam generator due to the added
complexity of the product gas scrubbing process.

The energy efficiency of the gasifier itself is calculated to be 93%, a value that is comparable to conventional boilers. The overall thermal efficiency, from coal to electrical output, is estimated to be 33.5%. This is almost identical to the national average thermal efficiency of coal fired plants. This efficiency is based upon many energy saving heat exchangers which may prove to foul, corrode, or be less effective than predicted. We believe that this gasifier system cannot achieve these efficiencies and that a rating of about 75% instead of 93% is more realistic.

This process has been conceptually designed and cost estimated. It is now ready for pilot plant design and construction at the 5 ton/hour size. The design phase would require up to two years with a peak effort of 45 persons. Ordering and construction could begin almost concurrently. In four years the pilot plant can be completed, shaken down, and operated for a sufficient time to be able to reliably scale up to full size (100 MW and larger) electric utility requirements. The proposed gasification process is based as much as possible on proven technology.

Prepared by Edward R. Lady
FOSTER WHEELER CORPORATION--PRESSURIZED COAL GASIFICATION

Foster Wheeler has completed the design and equipment specifications for a 50 ton/hr demonstration plant to gasify coal at 500 psig in an air-blown, entrainment-type gasifier (34). The design encompasses the production of 147 Btu/scf gas, sulfur and particulate removal, and use of the gas in a gas turbine and gas fired steam boiler. The project is co-funded by the Office of Coal Research, Northern States Power Company, Pittsburg & Midway Coal Mining Company, and Foster Wheeler Corporation. As a result of this design work, Foster Wheeler is prepared to initiate detailed construction design and equipment procurement so that the gasification system could be in operation within two years and the combined cycle power generation scheme would be in operation in 1977.

The 500 psig air blown, entrained flow gasifier design is based upon the small scale research and development work done by Bituminous Coal Research on the gasification portion of the process. As shown in Figure 11, the demonstration plant will feed dry pulverized coal at a rate of 50 ton/hr into the upper section of the high pressure gasifier through a lock hopper system. This coal is partially gasified by contact with steam and hot gases flowing from the lower section of the gasifier. The gases and partially gasified coal (char) flow from the top of the unit at 1800°F, 440 psig. Under these conditions Foster Wheeler believes no tars or heavy
Figure 11. Foster Wheeler Pressurized Gasifier
hydrocarbons will be present in the gas due to the very rapid heating of coal. Efficient recovery of the sensible heat of the gases should be possible. The char is removed in a high temperature cyclone separator and recycled to the lower portion of the gasifier where it reacts with air and steam at 2800°F. This provides the hot gas source for the upper portion of the gasifier and the ash is removed as liquid slag, which is subsequently quenched and removed from the system in a water slurry. The crude gas steam from the char separator is cooled from 1700°F to 200°F, in a series of heat exchangers used to generate steam and preheat air. The gas is then scrubbed, H₂S is removed by the Selexol process, and delivered as 147 Btu/scf clean gas at 350 psig. Initially this gas will be burned in an existing boiler and ultimately it will be used as the fuel in a combined cycle gas turbine-steam generating plant.

Evaluation of this proposed demonstration plant indicates that coal utilization should be complete under the slagging conditions which are expected to exist in the lower portion of the gasifier. Some carbon may be lost in the fines which are scrubbed out of the crude gas stream. Tests will be made with various coals, beginning with Illinois No. 6. Most coals regardless of free swelling index should be able to be gasified in this process.

A 33,000 hp air compressor is required to supply 68,000 scfm of air compressed to 500 psig. This is a conventional multi-stage centrifugal compressor with a steam turbine drive.
The air compressor and drive represents about 5% of the total plant investment. The 24,600 KW turbine is comparable to the 21,000 KW turbine to be powered by the steam from the waste heat boiler. Overall there is a multiplicity of air and gas compressors, steam and gas turbines which represents a very large amount of rotating mechanical equipment for the generation of 87 MW. This amount of compression equipment would be significantly decreased once the process is demonstrated by using the extra air compressed by the gas turbine compressor instead of expanding it to the atmosphere.

The coal feeding system is recognized by Foster Wheeler as one which needs development. The demonstration plant will use a lock hopper system, workable and proven, though expensive to build, operate and maintain. Hot char recycle and injection also poses serious problems. Again lock hoppers are proposed although a continuous flow system is much preferred.

Foster Wheeler believes devolatilization of the coal while suspended in the upper zone of the gasifier will take place without tar formation. This is uncertain and must be demonstrated. All types of coal should be usable although some injector development may be required. The recycle and combustion of the char in the lower portion of the gasifier at 3000°F and at 500 psig is an unproven technique. Serious potential problems may arise in refractory life, slag removal, flow and combustion stability. Safety interlocks on all streams are required. The transfer of thermal energy between the exothermic lower zone and the endothermic upper zone is
accomplished by the hot gases moving upward through the gasifier. Carry over of slag must be avoided, however. Slag and particle removal may be major problems. A liquid slag tap and water quench are proposed. Plugging of the slag hole may occur. Because this is at 500 psig pressure, visual observation and mechanical break-up of slag clinkers may be very difficult. Particulates are removed in the char cyclones and water scrubber. They may deposit on the various heat transfer surfaces and soot blowers should be considered.

Hydrogen sulfide is removed by a liquid scrubbing and subsequently converted to elemental sulfur for disposal. Since the crude gas is at 350 psig and scrubbing system is relatively compact compared to atmospheric pressure systems. A proven removal system, such as the Selexol process is one of the several which could be used. The gas treatment portion of the plant will rely on established technology and no attempt will be made to refine or optimize this portion of the plant.

Pressure equipment needed for the demonstration plant is large but most can be shop fabricated and shipped to the plant site. Sizes range up to 75 ft long, 11.5 ft diameter, and 100 tons per vessel. Such large equipment is not readily modified or replaced as is frequently necessary in a development program. Problems are anticipated in refractory life, flow patterns, thermal stresses, gas leakage, and especially with high temperature-high pressure valves. A gas tight valve suitable for handling particulate laden gases at 1800°F and 500 psig in 12 to 18 inch pipe size is needed. One of the
principal material problems is the refractory lining in the 3000°F, reducing atmosphere section of the slagging or lower zone of the gasifier.

Problems of instrumentation and control may be serious due to the low inventory of coal and char and the rapid throughput. Intermittent solids injection due to plugging of lines could be very harmful. The thick pressure shells, refractory linings, and extreme temperatures make instrumentation problems severe. Similar conditions have been successfully employed in ammonia synthesis, partial oxidation and other chemical processes. Turndown capability and start-stop operation for reduced or intermittent load service is completely unknown and the demonstration plant operating experience will help determine if entrained flow pressurized gasifiers can be used for this service.

Maximum energy efficiency is not the goal of this plant but rather to demonstrate full size gasification. However, the projected 82.2 MW net station output from 50 ton/hr of Illinois No. 6 coal (12,330 Btu/lb gross heating value) gives an overall thermal efficiency of about 23%. If credit is given for air compressed by the gas turbine, this is raised to 28.9%. Foster Wheeler estimates that this process can be improved by higher gas turbine inlet temperatures. If the thermal efficiency reaches 36.5%, and anticipated equipment costs are reduced in second generation plants this combined cycle, low Btu gasification process will be competitive with conventional coal fired boilers having SO₂ removal equipment costing $50/KW.
This project is a bold effort to effect a 1000 to 1 scale up of partial pilot plant data into a full size demonstration plant. Conservative and proven design and equipment is used where ever possible but a number of unknowns make this a relatively high risk venture. Foster Wheeler has completed a detailed preliminary design and is ready to initiate purchasing and fabrication within a few months. Initial operation of the gasifier portion is planned for two years after start, with the overall program requiring 5-1/2 years.

Prepared by Edward R. Lady
The Molten Salt Gasification process basically consists of oxidation of carbon to CO and partial pyrolysis and distillation of volatile matter in a bed of molten sodium carbonate, sulfide and sulfate (15).

**Chemistry**  
**Gasification Reaction Involves 2 Steps**

\[
\begin{align*}
\text{Na}_2 \text{SO}_4 + 4\text{C} & \rightarrow \text{Na}_2\text{S} + 4\text{CO} & \Delta H &= +224,000 \text{ Btu/lbm} \\
\text{Na}_2\text{S} + 2\text{O}_2 & \rightarrow \text{Na}_2\text{SO}_4 & \Delta H &= -414,000 \text{ Btu/lbm} \\
4\text{C} + 2\text{O}_2 & \rightarrow 4\text{CO} & \Delta H &= -190,000 \text{ Btu/lbm}
\end{align*}
\]

**Carbonate Regeneration**  
\[
\text{Na}_2\text{S} + \text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{Na}_2\text{CO}_3 + \text{H}_2\text{S}
\]

**Comments**  
Because the process takes place in a large mass of molten salt the gasifier temperature is effectively controlled, the ash gets easily trapped in the melt. Acid gases are reacted with the salt. The oxidation of C into CO is 5 times faster then conventional combustion. About 90% of the heating value of coal is retained in the gas produced.

**Advantages**

1. Ability to handle caking coals
2. Retention of sulfur in melt
3. Retention of ash in melt
4. Absence of tars (1750°F)
5. Relatively simple gasifier design
6. Absorption of halides and other trace impurities
7. Control of temperature below $\text{NO}_x$ formation levels
8. Potential adaptability to combined cycle operation

**Disadvantages**

1. Carbon losses in discharge melt (3.5%)
2. Melt loss
3. Alkali metal in overhead gas
4. Filtration Problems in ash removal from aqueous solution during regeneration

**Critical Areas for Further Evaluation**

1. Air and coal feed distribution
2. Size, size distribution of particulates, alkali metal melt entrainment
3. Control of heat release
4. Equipment for melt removal, piping, valving, etc.
5. Lack of data on completely integrated system

Figure 12 shows a schematic flow sequence of Atomic's Molten Salt Process in low Btu gas, combined cycle application.

Prepared by M. Rasin Tek
Figure 12. Flow Sheet for Molten Salt Gasifier
The process consists of gasification of coal in a bath of molten sodium carbonate with simultaneous injection of steam. Sodium carbonate has a strong catalytic effect on the basic steam-coal reaction permitting essentially complete gasification of coal at reduced temperature. Molten salt also acts as an excellent vehicle to supply heat to the coal undergoing gasification (13, 14).

The main items in process sequence are shown in Figure 13. According to latest concepts the gasifier is a single reactor to which sodium carbonate and coal are fed through lock hoppers via fluidized media by steam and oxygen.

Under increased pressure and with a pure oxygen atmosphere complete gasification is expected to take place at temperatures low enough to allow direct formation of methane in the gasifier. At the prevailing thermodynamics conditions 1200 psia @ 1700-1800°F all tar components from the coal are gasified. The major portion of preheated oxygen and steam are also added at the bottom of the gasifier.

The gasification reactions (primarily endothermic steam and coal) also supply some of the process heat required along with some coal combustion to CO₂.

The sulfur in the coal accumulates in the bath as sodium sulfide which in reaction with CO₂ and H₂O regenerates the sodium carbonate releasing sulfur in the form of H₂S in the gas phase.
The bleed stream from the gasifier is quenched in water to dissolve the sodium carbonate. The underflow from the dissolving tank goes to a filter where ash and carbon are separated and disposed. The dilute carbonate solution is further carbonated by CO₂ from purification into bicarbonate which is calcined into sodium carbonate and recycled back into the gasifier.

Advantages:

1. Ability to handle caking coals
2. High methane yield
3. Retention of ash in the melt
4. Retention of sulfur in the melt
5. Absence of tars
6. Simple gasifier design
7. Possible absorption of other trace impurities
8. Potential adaptability to combined cycle operation

Disadvantages:

1. Carbon and melt losses
2. Complex ash removal
3. Complex and inefficient carbonate regeneration
4. Needs oxygen and CO₂
5. Sodium carry over on overhead line from gasifier
Kellogg feels they would have sufficient information to conclude bench scale gasifier program by the end of the current year.

It is believed they are $3$ MM and 3 years away from a conclusive pilot program and perhaps $30$ MM and 5 years from a demonstration plant.

Prepared by M. Rasin Tek
U.S. BUREAU OF MINES - STIRRED-BED GASIFIER

Fixed-bed producers have been used to generate low to intermediate-Btu fuel gas for many decades. Sixty-five producers of all types have been described in a comprehensive survey made by Bituminous Coal Research (24). Most producers are limited to noncaking or mildly caking coals and are unsuitable for strong caking coals from the Pittsburgh seam or moderately strongly caking coal such as Illinois No. 6 seam. The inability of producers to handle caking coals and coal of all sizes is a severe limitation for use in the United States.

The Bureau of Mines in Morgantown, West Virginia, has operated an experimental producer which includes a stirring device to continuously agitate the fuel bed, thereby preventing agglomeration. The design has been described by McGee (35) and test results reported by Lewis (36). The stirred-bed producer shown in Figure 14 was operated at a nominal 100 psi, thus giving promise for use in combined cycle power plants. The test reactor is 3.5 feet diameter by 24 feet long and is characterized by a slowly rotating agitator which also has vertical oscillation so that all parts of the bed are broken up. Coal feed and ash removal is via lock hoppers. Gas produced from the gasifier contained approximately 21% CO, 16% H₂ and 3% CH₄, with a gross heating value of about 150 Btu/scf. These are normal values for air blown producer gas. The producer gas also contains tars which must be scrubbed out. The cold gas efficiency ranged from 62 to 72%, a value that
Figure 14. U.S. Bureau of Mines Stirred-Bed Coal Gasifier
may be raised to the 75% level with larger scale units. If high temperature \( \text{H}_2\text{S} \) removal means were available, if the tars were not condensed, and if the sensible heat of the 900-1200\(^\circ\)F gases could be utilized, the thermal efficiency would be significantly improved.

The experimental gasifier produced 9.4 Mscfh of 145-164 Btu/scf gas per square foot of grate area. It appears that a higher gasification rate may be possible. If one assumes a gasifier of 12 feet diameter (Lurgi size limit) and 12 Mscfh per square foot, then a single gasifier could produce 1.36 MMscfh of 150 Btu gas. Using a steam plant thermal efficiency of 33.5%, this represents the fuel supply for 20 MW of electrical generation. Using a combined cycle thermal efficiency of 40%, the result is 24 MW. It can be seen that 40 to 50 gasifiers would be required for a 500 MW power plant. Atmospheric pressure agitated gasifiers, such as the Wellman-Galusha units commercially available (37), are offered up to 10 feet diameter. These have about half the gas output of a pressurized producer of equal diameter.

Stirred-bed gasifier advantages:
1. All coals can be processed
2. Low-Btu gas is produced under pressure
3. Overnight banking appears feasible

Stirred-bed gasifier disadvantages:
1. Bed size is limited by mechanical considerations, similar to Lurgi gasifiers
2. Lock hopper and valves have high maintenance
3. Oils and tars must be removed from gas

4. A very large number of gasifiers would be required for a 500 MW electric generating unit.

In essence, the stirred-bed gas producer is a modification of a Lurgi gasifier so that strongly caking coals of wide size range can be handled. The unit at Morgantown should continue to be operated to gain further experience on problems associated with transport of low Btu gas containing tar and flyash and to provide a source of hot, tar laden gas for development of high temperature H₂S removal systems.

Prepared by Edward R. Lady
The Hydrane Process is a gasification of coal process in the early stage of development at the Bureau of Mines. It is based upon the reaction of raw unpretreated coal with hydrogen to form methane directly. The process has been described by Feldmann (38,39) and the schematic flow diagram is shown in Figure 15.

Pulverized coals of any rank are fed to the top of Stage 1 operating at 1000 psi and 1650°F. The coal devolatilizes while flowing in dilute phase suspension concurrently downward with hot methane and hydrogen rich gas from Stage 2. The coal falls into Stage 2 and the synthesis gases are removed and cooled. Partial gasification and methanation occurs in Stage 2 by contacting the Stage 1 char with nearly pure hydrogen at 1650°F. The heat released through methanation is removed by steam generation in Stage 2 heating coils. Char from Stage 2 is conveyed by steam into the gasifier for producing hydrogen. Heat for the gasifier is supplied by a hot grog (inert ceramic material) heated in a fluidized bed combustor.

Exit gases from Stage 1 contain about 75% methane (with a CO₂ removal step included), 15% H₂, and 4% CO on a dry basis.

Pilot plant work has been done on a 3-inch diameter reactor using a caking coal.
Figure 15. U.S. Bureau of Mines - Hydrane Process for Coal Gasification
Process Advantages:
1. High methane production directly from coal.
2. Purification and methanation requirements are less for high Btu gas generation.
3. Use air or small oxygen plant.
4. Processes all coal types.

Process Disadvantages:
1. Process involved and complicated.
2. Still requires shift conversion and gas purification.
3. Substantial development is needed to produce H$_2$ from char and steam.

The development work on this process at the Bureau of Mines has been inhibited by the lack of funding. Although the process may prove to be more efficient thermodynamically than the Synthane Process in the production of methane for pipeline gas, it lags the Synthane development by at least two years. Thus there is little reason to believe that this process can contribute to the near term clean energy requirements of the electric power industry.

Prepared by Edward R. Lady
U.S. BUREAU OF MINES - SYNTANE PROCESS

The Synthane Process is a gasification system developed by the Bureau of Mines for converting bituminous coal, sub-bituminous coal, and lignite into a satisfactory substitute for natural gas. The research results through mid-1972 are described by Forney and McGee (40).

Crushed coal passes through a lock hopper into a fluidized bed pretreater where the caking properties of raw coal are destroyed by contacting the coal with a steam-oxygen mixture at 800°F (40). The process is shown in Figure 16. Decaked coal from the pretreater enters the top of the fluidized bed reactor which contains two operating zones. Steam and oxygen enter at the bottom to effect fluidization.

The gasifier operates at 600-1000 psi. The top portion of the gasifier operates as a dense fluidized bed at 1100-1450°F and the lower portion as a dilute fluidized bed at 1750-1850°F. As in most gasifiers devolatilization and limited methanation takes place in the top portion and hydrogen formation in the lower section. Ash and unconverted coal or char are removed through a lock hopper. The char is burned to generate process steam.

The Synthane process requires a minimum of oxygen and approximately 1/3 of the coal is converted to char. Some studies have been conducted on the gasification of coal using air-steam in a fixed-bed reactor (fuel supported on grate).
Although the gas produced is of low Btu/scf, there is more efficient energy use.

Preliminary engineering studies of the Synthane process by M.W. Kellogg Company concluded that the process is feasible. Lummus Company designed a 75 tons/day pilot plant for producing pipeline quality gas. Rust Engineering Company is currently constructing the plant in Bruceton, Pennsylvania which is scheduled for completion in mid-1974.

**Process Advantages:**

1. Process flow and equipment are simple.
2. All coals can be processed.

**Process Disadvantages:**

1. Feeding system is still awkward requiring several large pressure vessels. High energy loss is encountered.
2. Need oxygen plant.
3. Methane concentration from gasifier is low.

The Synthane Process could be adapted to produce an intermediate heating value gas by omitting the shift and methanation steps, thereby lowering fuel cost for electric utility use. However, the use of oxygen and the high pressures involved will tend to make this a high cost fuel. The 75 ton/day pilot plant construction is underway and after operating data are available the electric utility industry will be able to assess the potential of this process towards meeting clean fuel needs. In this assessment consideration should be given
to production of lower heating value, and consequently lower cost, gas.

Prepared by Edward R. Lady
This design of an agglomerated-ash fluidized bed was developed by Battelle Columbus under sponsorship of Union Carbide (41-44). The reaction section, as designed for the process development unit is shown schematically in Figure 17. Pulverized coal is fed to the gasifier which will operate at pressures up to 100 psig. This is a steam gasifier in which the energy for the reaction is furnished by a steam of hot agglomerate flowing from the burner. The energy needed to heat this stream in the burner is supplied by the air-combustion of cycled char, coal, or other fuel. The temperature in the burner is controlled at a point near enough to the ash fusion point to promote the agglomeration phenomenon. The agglomerate stream recirculates between the two vessels, using steam as a lifting fluid. The technology is similar to the fluid catalytic cracking prevalent in the petroleum industry.

Some indicative laboratory data have been released. Figure 18 shows the relationship between temperature and the effectiveness of the phenomenon in collecting ash particles—that the agglomeration is more intense as the fusion point is approached. The burning efficiency has been determined, also as a function of temperature, Figure 19. Although other conditions are not disclosed, carbon can be fully utilized at temperatures which are compatible with effective agglomeration. Finally, stable fluidization can be obtained, Figure 20, at these same temperatures. The fact that this temperature
Figure 17. Battelle Memorial Institute Self Agglomerating Gasification Process
Figure 18. The Effect of Temperature and Bed Depth and Gas Velocity on Ash-Collection Efficiency
Figure 19. Combustion Efficiency as a Function of Temperature in the Fluidized-Bed Coal Burner
Figure 20. Stability of the Self-Agglomerating Fluidized-Bed Coal Burner as a Function of Temperature and Gas Velocity
dependence of stability has been observed in other fluidization processes is qualitative assurance that general fluidization experience is applicable to the analysis and design of fluidized agglomerate systems.

During the development of the gasification process, tests were made in which turbine blade materials were exposed to ash-containing streams \(^{(45)}\) in order to evaluate rates of erosion and existence of erosive conditions.

The Battelle process has some interesting characteristics:

1) The agglomerating phenomenon in the burner is a mechanism for capture of fly ash. The product gas may prove to be sufficiently free of erosive particulates so that expensive cleaning steps may be minimized before expansion in a power recovery turbine or before use in a combined cycle turbine.

2) The use of a stream of hot solids as a transfer medium avoids dilution of the product gas by combustion gas. Also, air may be used instead of oxygen without incurring the penalty of nitrogen dilution.

3) The gasifier should have a moderately good turn-down ratio. Like fluid units in petroleum refining, solids circulation could be maintained indefinitely with the gasification in an idling condition.

There is one operating characteristic which seems essential to the success of agglomerating processes. That is, the separation, or layering, in the fluidized beds of the agglomerated particles from the smaller unburned coal or char and ash. The maintenance of this same characteristic is also necessary
to the operation of the CO$_2$ Acceptor process. It is encouraging to know that this part of the acceptor process has been satisfactorily demonstrated in the 48 ton/day Rapid City plant. It seems reasonable to expect that the operators of the Battelle PDU will also be able to master this aspect of their process.

The Union Carbide/Battelle process is close to testing in a unit designed to operate at a pressure of 100 psi. A contract was awarded to Battelle by the Office of Coal Research in January 1973 for the construction and operation of a Process Development Unit to evaluate commercial feasibility. Chemical Construction Company was chosen as the engineering contractor to build a plant at Battelle's station in West Jefferson, Ohio with a design capacity of 25 tons per day of eastern bituminous coal. Gasifier and burner have been carefully designed. Figure 21 shows the block flow diagram of this plant which is expected to start-up in the Summer of 1974 unless delayed by shortages and fabrication schedules.

The burner and gasifier designs are based upon atmospheric experimental data which have not been publicly disclosed. Burner and agglomeration behavior have been studied in vessels up to one foot in diameter. The coupling of burner and gasifier has been studied on laboratory scale, and Battelle has additional experience with fluidization of other materials in this application. The design of the Process Development Unit, reactors and system were based upon these Battelle data
Figure 21. Block Diagram of Battelle Coal Gasification PDU
and was completed with the assistance of the C.F. Braun Company.

No cost estimates nor projected operating costs and efficiencies are yet available for this process. It is included in the group which the C.F. Braun Company is evaluating, under OCR/AGA sponsorship, to choose a process for the demonstration plant (2).

Prepared by Brymer Williams
A group of five professors at CCNY with Dr. Arthur M. Squires as Principal Investigator have been carrying out a varied and wide-ranging program toward improved techniques for gasifying coal during the past several years under sponsorship of the RANN division of NSF. Much of the effort is directed to longer term solution of the nation's energy needs and includes flowsheet and system studies of a "Coalplex" to produce pipeline quality gas, petrochemicals (benzene, toluene, and xylenes) and low-Btu gas for power generation by combined gas- and steam-turbine cycles. The studies of an "Energy Corridor" as a possible means of supplying energy in a variety of forms in urban areas also falls in this category of long-range studies and should also be of interest to the electric utility industry. The multifaceted aspects of this coordinated research were presented in an Industrial Briefing held January 16-17, 1974, at CCNY and have been dealt with in a number of presentations made at various technical meetings (46-49). Only those aspects which appear to be of the most immediate interest to the utility industry--the ash agglomerating fluidized bed gasifier and the cleaning of hot low-Btu gas--will be reviewed here.

It should be mentioned in passing that Squires and his group have come up with a number of "clever" experiments
which appear to have isolated critical factors for individual study from among the miriad of problems facing those who are attempting to work in the general area of gasification of coal to produce a low Btu gas. As such their efforts might well serve as a model for similar research at other universities.

AGGLOMERATING FLUIDIZED BED GASIFIERS

Development work is progressing on both a moving grate gasifier for "immediate" application (the Mark I gasifier) and a longer term development incorporating a gas fluidized bed (the Mark II). Both will be described in some detail in the sections to follow.

THE MARK I GASIFIER

The Mark I gasifier is essentially a revamp of the Ignifluid boiler developed by Albert Godel and Babcock-Atlantique that is operating today in Morocco, France, Scotland, and Korea (50-52) as illustrated in Figure 22.

Run-of-the-mine coal is used after being crushed to reasonable size. If the fluidized bed has a high ash inventory as is expected, it should be possible to use caking coal. Carbon utilization is expected to be high (greater than 99%) based on prior experience with Ignifluid boilers. Air is used and is entered so as to cool the main grate. The Mark I gasifier is an atmospheric unit designed primarily to
Figure 22. The Mark I Coal Gasifier
produce gas to retrofit existing gas-fired boilers and therefore physical entry of the coal should provide no special problems.

A single bed is utilized and therefore devolatilization and combustion occur simultaneously with heat transfer being facilitated by the fluidized ash bed. The major innovation is the moving grate which serves to remove the agglomerated ash-clunkers. The moving grate is cooled by recycled power gas. The work of Godel in developing the Ignifluid fluidized bed combustor has demonstrated that the operation of both fixed and moving grates can be reliable even at temperatures high enough to provide for agglomeration of ash to uniform, near spherical particles. Apparently the sticky ash sticks only to other ash—not to the grates and not to coal.

A major part of the fines carried up out of the fluidized bed are removed from the product gas by cyclones and recycled to the bed, again based on current practice with the Ignifluid units. Tars should not be a problem both because of the high temperature of operation (2200-2400°F) and the presence of solids. At the high temperature of operation of the bed, significant amounts of volative metals (Na, K, Hg, etc.) will undoubtedly appear in the product gases.

In situ removal of sulfur is not provided and such removal would require advanced developments because of the elevated temperatures required to bring about agglomeration of the ash. Although external removal of sulfur by any number of processes
might be utilized, such removal will be hampered by the fact that the unit operates at atmospheric pressure. Instead, Squires (48) recommends the Mark I gasifier for use with western low sulfur coal in the Southwest to retrofit utility and industrial boilers currently firing 4 trillion standard cubic feet of natural gas each year.

From an equipment standpoint the use of fixed and moving grates would be expected to cause problems but apparently these have been resolved by Ignifluid. Probably only large-scale experiments would demonstrate like reliability in a reducing atmosphere and such experiments have been proposed (48).

The energy efficiency of the unit will depend to a large extent on the feasibility of utilizing the sensible heat of the hot product gases and in the absence of tars and excessive fines. This would seem to pose no unusual problems other than those normally associated with hot reducing atmospheres.

THE MARK II GASIFIER

As illustrated in Figure 23 the Mark II gasifier attempts to utilize one section in which fines are devolatized (fast fluidized bed) above another section in which the larger particles are burned and ash agglomerated for ease of removal.

Besides the usual attendant problems of getting coal into a gasifier operated at pressure, there should be few problems with using run-of-the-mine coal of any swelling index crushed to less than 3/4 in, because the coal should be well dispersed in the fast fluidized bed and the residence time is low.
If agglomerates form, they will drop to the bottom and be burned. In accordance with the Godel principle, the utilization of coal should be high. Air (rather than oxygen) and steam are used as the fluidizing medium. One advantage claimed is the very small steam/air ratio required (0.033 moles of steam per mole of air vs. 0.49 for Lurgi). Further advantages are claimed if steam is utilized as the fluidizing medium in the fines return leg.

The fast fluidized bed is designed for devolatization as the fines are entrained and taken overhead and recovered by cyclones for recycle.

As indicated in Figure 23 the combustion of the larger coal particles and the recycle fines are carried out in a fluidized bed composed primarily of agglomerated ash at the bottom of the gasifier. If the carbon in the recycled fines were to be inert, a build-up of fines and subsequent overloading might occur. The problem might be more severe here than in the BCR gasifier which utilizes a higher temperature in the combustion zone.

The heat required by the devolatization reactions are supplied by the hot products of combustion from the ash-agglomerating fluidized bed as in many other gasifiers. Mixing of solids between the two sections of the gasifier provides for fantastic heat transfer.
One of the primary objectives of the CCNY Mark II gasifier is application of the Godel principle to bring about agglomeration of the ash particles in a dense fluidized bed with the attendant advantages to reduction of fly ash carry-over, high carbon utilization and ease of removal of the larger ash particles from the pressure vessel.

The agglomerating bed should ease the problem of particulate removal but any ash in the fines that are devolatilized will undoubtedly be carried over and some will pass through the cyclones necessitating further treatment before use as a fuel for gas turbines. The lower temperature of the fast fluidized bed section combined with the recycle of cooled solids from the overhead should serve to reduce the amount of volative metals in the power gas but further treatment will probably be required as discussed in the next section.

The high temperature of operation of the dense fluidized bed probably precludes the in situ removal of sulfur. Sulfur removal will be external to the gasifier and the Benfield system has been included in one study (46). Other possibilities for treatment of the sulfur at high temperatures are being considered as will be discussed in the next section.

One advantage claimed for the Mark II is its relatively small size. "A vessel with an inside diameter of 3.3 meters (about 10 ft) and a height of 15 meters (about 50 ft) will handle 3,000 tons per day of coal, and will provide power gas sufficient for 300 megawatts. This is a good match for
combined cycle equipment of the type the U.S. electricity industry is now building in about 20 locations" (48). As indicated in Figure 23 it is proposed to eliminate valving in the fines return line by utilizing a fluidized leg. There may be problems with proper distribution of air throughout the ash-agglomerating fluidized bed but developments currently underway in connection with fluidized bed combustion should be helpful. Removal of the agglomerated ash could also prove bothersome until the bugs are worked out but here again recent developments in connection with fluidized bed combustion should prove helpful.

CLEANING HOT LOW-BTU GAS

Several avenues of approach are being followed in an attempt to provide means of cleaning hot low-Btu gas so that it can be utilized more efficiently in gas turbines. Not only is the panel bed filter under development at CCNY but studies are also being made to follow up work previously carried out at the Bureau of Mines and elsewhere.

PANEL BED FILTER

The panel bed filter as illustrated in Figure 24 is an old idea [probably invented in 1877 for Deacon's chlorine process according to Squires]. Gas is cleansed by causing it to flow in a horizontal direction across a bed of granulated solids dispersed in a tall, narrow "panel". The bed may be
Figure 24. Schematic of a Panel Bed Filter
held in place, for example, by louvered walls that resemble venetian blinds. The original technique has been improved upon by the City College team by development of a procedure for discharging granular solid from the gas-entry side of the panel. Periodically, a sudden blast of gas at high pressure is directed against the rear side of the panel, dislodging a controlled amount of solid uniformly from the gas-entry side. With this technique, the flows of gas and treating solid can approach countercurrentricity as closely as may be required. This procedure also leads to a marked improvement in filtration efficiency, with efficiencies beyond 99.99% having been achieved. It is claimed that the panel bed is ready for scale-up to a size of about 30,000 cubic feet per minute and a commercial partner for development of a unit on this scale is sought.

SULFUR REMOVAL

In addition to the removal of fines as proven for the panel bed, it is necessary to provide for removal of $H_2S$ to meet current standards if coal containing more than 0.8% sulfur is to be burned. Some work on $H_2S$ removal with half-calcined dolomite has been encouraging and is being continued. Such work included regeneration by reaction with steam and carbon dioxide at atmospheric pressure (49).

The CCNY group has also done some exploratory work with the fly ash-iron oxide composite developed at the Bureau of
Mines, Morgantown, W. Virginia. Work is continuing at CCNY with both half-calcined dolomite and BOM composite.

Prepared by John E. Powers
The design of this gasifier is based on ash agglomeration phenomenon and its use in a fluidized bed (53, 54). Figure 25 is a simplified presentation of the essential elements of the reactor. It is a single stage unit with oxidation and steam-gasification occurring in a single fluidized bed. It is intended to operate at pressures up to 300 psi.

Ground coal is fed to the gasifier from a lock hopper. Coals with caking tendencies would be introduced first into a pretreater, held at 800°F and with an oxidizing atmosphere to eliminate caking. Heat release in this section is large, about one-half billion Btu per hour for a 7500 ton per day unit. The bed must have steam generating coils to remove this energy. Pretreated coal overflows to the gasifier section.

There is a temperature gradient in the gasifier. The temperature in the bottom zone must be high enough to permit agglomeration, that is, approximately 2000°F. The temperature in the gas or disperse phase above the fluidized bed may be as low as 1500°F. Residence times of the solid in the pretreater and the gasifier are 30 and 50 minutes, respectively. Residence time in the gas space above the bed is about 10-15 seconds. Analysis of devolutilization and cracking rates indicates that tar formation should not be a serious problem with the proposed combination of temperature profiles and gasifier geometry. Small particles of ash and unburned char, which are entrained overhead from the bed, are collected in
Figure 25. Institute of Gas Technology Ash-Agglomerating Gasifier
two-stage cyclones and returned to the gasifier bed. The ash is removed at the bottom of the bed. The bottom zone must be designed and operated so that agglomerated ash can grow to sizes which will separate from the bed by gravity and fall into a pool of boiling water below the air entrance. The proper design of this part of the reactor is a critical element in the success of the process. Proposed designs have been based upon several types of experience with fluid bed behavior and with due analysis of fluid dynamic considerations.

The "U-Gas" process has been tested and explored in a four-inch diameter unit. Since agglomeration is thought to be independent of pressure over possible operating ranges, the tests were made at atmospheric pressure. The Ralph M. Parsons Company is working with IGT to prepare a design and cost estimate of the higher pressure demonstration plant. The size of this has not been determined or announced, but it will be of a capacity in which large scale problems can be solved in the absence of small-scale difficulties. The size of 100 megawatts has been mentioned as a possibility.

IGT has prepared an economic appraisal of the process for three conditions: retrofit, separate and combined cycle plants. Table 1 summarizes the essential numbers for the three cases. It should be noted that this process includes (1) an 800°F sulfur removal unit using IGT's Meissner process which is under development, and (2) a gas turbine with a 2400°F inlet temperature which it is hoped will be available when the
"U-GAS" process is ready for full commercialization. Using these assumptions, the costs appearing in Table VI were calculated.

Prepared by Brymer Williams
<table>
<thead>
<tr>
<th>Application</th>
<th>Retrofit</th>
<th>Separate</th>
<th>Combined-Cycle</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal Feed Rate, ton/day</td>
<td>7346</td>
<td>7346</td>
<td>7346</td>
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<tr>
<td>Coal Cost, ¢/million Btu</td>
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<td>30</td>
<td>30</td>
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<tr>
<td>Plant Investment, $, million</td>
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<td>90</td>
<td>216</td>
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<tr>
<td>Process Overall Thermal Efficiency, %</td>
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<td>78</td>
<td>42</td>
</tr>
<tr>
<td>By-Products (assumed value)</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Sulfur, long ton ($10/LT)</td>
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<td>188</td>
<td>188</td>
</tr>
<tr>
<td>Steam, 1000 lb/hr ($0.90/1000 lb)</td>
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<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Power, MW (8 mills/KW hr)</td>
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<td>146</td>
<td>--</td>
</tr>
<tr>
<td>Net Product Output</td>
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</tr>
<tr>
<td>Low-Btu Gas, $10^9 Btu/day</td>
<td>129</td>
<td>129</td>
<td>--</td>
</tr>
<tr>
<td>Electricity, MW</td>
<td>--</td>
<td>--</td>
<td>942</td>
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<tr>
<td>Unit Price of Output*</td>
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<td>65.3</td>
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</table>

The Hygas Process is designed to produce pipeline quality gas through direct hydrogenation of coal at high pressures (55-60). Developmental work was begun by the Institute of Gas Technology (IGT) in 1946, but the majority of the work has been accomplished since 1964 under a 14 million dollar contract sponsored by the Office of Coal Research and the American Gas Association. At least 22 tests run have been made on a 75 ton/day pilot plant that was completed in 1971 at Chicago.

Figure 26 illustrates the Hygas process. Coal is sized and pretreated if necessary (caking coals are partially oxidized to prevent swelling) and then slurried with light recycle oil. The slurry is fed to the top of a four-stage reactor at 1000 psi. The first stage (1000-1500 psi, 600°F) flashes the light slurry oil leaving a dry coal to be fed to stages 2 and 3. The second and third stage of the reactor are the hydrogasification sections where the following reaction occurs.

\[
C + H_2 \xrightarrow{\text{High Pressure, High Temperature}} \text{CH}_4
\]

About 10-20% of the coals gasified in the first hydrogasification stage at 1100°F and 1000-1500 psi. The second hydrogasification stage accomplishes the remaining hydrogasification at 1600°F.

The fourth stage of the reactor is for producing hydrogen. Three methods currently exist for accomplishing this: the electrothermal process, the steam iron process and the
Figure 26. Institute of Gas Technology, HYGAS Coal Gasification Process
steam-oxygen process. The latter of these holds the most potential and is the one of primary concern at present. In the steam oxygen process, steam reacts with char to form hydrogen and carbon monoxide. Heat is provided for the reaction by combustion of a portion of the char with oxygen. By regulating the oxygen flow rate the temperature can be carefully controlled. The operating temperature is in the range of 1800-2000°F; bottom solids are discharged through a water slurry system.

Areas of accomplishment and/or concern:

1. Coal Feed - The slurry is able to carry a high percentage of solids (up to 45% by weight coal) and equipment has been developed to handle the transport problems (i.e. erosion) without significant wear. However, not all the recycle oil is yet recoverable and this must be accomplished for the system to be economical. Additionally, most coals will require pretreatment to prevent swelling, thus increasing the cost of the system.

2. Gasifier - The solids transport in the fluidized gasifier is still somewhat of a problem and is presently being studied.

3. Overall - Predicted efficiencies are 40% for the carbon and 70-75% on the heating value. However these efficiencies have not been achieved on pilot runs.

The pressure in the reactor is much too high (1000-1500 psi) for use in combined cycle. About 300 psi is the practical limit for most combined cycle turbines.
The equipment needs for this process are perhaps greater than those of any other process. The C.F. Braun Company calculated that 3 reactors, 22 feet in diameter and 250 feet tall would be required for a 250 billion Btu/day plant. Reactor weight would be 3000-4000 tons each.

Generally, IGT, has done a commendable job in developing the Hygas process. Although there are still some problem areas, these problems are defined and progress is being made toward their solution. Work on the process is quite advanced and commercialization is predicted as early as 1981. Unfortunately, the process is primarily applicable to production of high Btu pipeline gas and probably would not be efficient for use in combined cycle plants for the electric power industry.

Prepared by M. Rasin Tek and Gerald Holder
WESTINGHOUSE - ADVANCED GASIFIER

Work has been underway at Westinghouse for over 10 years starting with the use of coal in fuel cells. This is a real plus factor because both experimental and theoretical work with coal requires a "soaking period" which is lacking with many organizations which have entered the field only recently.

The proposal submitted to OCR ((61) by the Industry Team of Westinghouse, Public Service Indiana, Bechtel Corporation and Amax Coal Company is very well thought out and should be required reading for anyone entering the field of coal gasification. Every problem is addressed squarely and a solution proposed. Apparently it was felt that novel—rather than established—procedures were required in order to obtain funding. Unfortunately "novelty" translates to "unproven" in most cases. Therefore, with respect to gasification, the Westinghouse advanced gasifier concept incorporates a number of unproven ideas. The problems are well enumerated and very competent people are working to solve the problems that have been identified.

The Advanced Coal Gasification System for Electric Power Generation involves considerably much more than just a gasifier with associated $\text{H}_2\text{S}$ removal which will be discussed in detail below. The complete process involves a gas turbine using low Btu gas as fuel incorporated into a combined cycle with steam power generation. Many problems areas have been
identified. For example, experts are using various approaches
to attempt to determine apriori what level of particulate
removal will be required and how much carry over of alkali
metals will occur to provide data so vital in the development
of gas turbines which will require reblanding only every 5 years.
The group working together has exceptional expertise in areas
all the way from coal mining and processing and coal analyses
(AMAX and Westinghouse), materials of construction (including
insulation) (Bechtel), construction itself (Bechtel) particulate
removal and required level of such removal (Westinghouse),
control of alkali metal carry over and effect of such materials
on turbine blade life (Westinghouse), problems associated with
burning fuels in a gas turbine plus experience in building such
turbines, (Westinghouse), and operation of large scale power
generating facilities (Public Service Indiana). This combined
capability should be considered to be a plus factor.

The weakest link in the advanced Coal Gasification System
is probably the gasifier itself. Even this fact is recognized
in the proposal and the group is willing to accept any proven
gasifier at the time the large scale test of the combined
cycle is to be put into effect. The basic flow diagram for
the gasifier unit is shown in Figure 27. As discussed in
detail in the proposal to OCR a solution has been offered
for every one of the problem areas.

Any type of coal can be used. The sizing of the coal
fed to the system is not critical; crushed run-of-mine coal
can be used. A drying step is shown but this step is not
Figure 27. Westinghouse Multistage Fluidized Bed Process for the Total Gasification of Coal with Desulfurization for an Electric Power Plant
designed to change the caking characteristics of the coal but only to dry it. The carbon utilization should approach 100%.

Air must be compressed because the system is designed to operate at pressure. There seems to be no reason to use oxygen.

Lock hoppers are to be used to get the coal up to pressure. The coal is conveyed from the lock hopper to the dryer using hot recycle product gas which also serves to transport the dried coal from the dryer to the fluidized bed devolatilizer.

The dried coal is entered directly into the central draft tube of the devolatilizer. In this tube the coal feed and large quantities of recycled solids—char and lime—are carried upward by gases from the total gasifier flowing at velocities greater than 15 ft/sec. The recycle solids needed to dilute the coal feed and to temper the hot inlet gases flow downward in a fluidized bed surrounding the draft tube. These solids, flowing at rates up to 100 times the coal feed rate, effectively restrict the agglomeration of the coal feed as it devolatilizes and passes through the plastic sticky phase.

Air is entered with steam into the bottom of the total gasifier. These gases serve to fluidize the bed and there are no apparent problems with this aspect of the operation.

The devolatalization takes place in the fluidized-bed devolatalizer. This unit operates at 1500-1700°F and no tars contaminate the product fuel gas. This is probably true even
at these relatively low temperatures because the solids in
the fluidized bed should act to retain the tars until they
are further cracked to lower molecular weight compounds.

Partial combustion is carried out in a separate unit.
The hot char as recovered from the fuel gas issuing from the
devolatilizer is bed to the combustor.

Heat transfer between the devolatilizer and combustion zones
is provided by the hot gases from the combustor which are
entered into the draft tube of the devolatilizer with the dry
coal so that maximum devolatilization occurs in the draft tube
itself.

It is hoped that the fluidizing action in the bottom
leg of the combustor will serve to concentrate the agglomerating ash to facilitate ash removal and yet provide for high
carbon utilization. Whether or not a segregation does occur
remains to be seen.

Several problems will inevitably be associated with any
agglomerating unit. The ash will agglomerate properly only
over a fairly narrow temperature range which may vary somewhat
even for coal from the same mine. Therefore operation of such
a unit will probably require above-average operators. The ash
will be removed through lockhoppers.

Westinghouse recognized that removal of all particulates
of greater than 1 micron in size and at high temperature will
probably be required. They have 2-3 professionals working
essentially full time on attempting to provide a solution to
this critical problem. A number of possible solutions have been considered including special cyclone designs, impingement devices, etc. This is a problem common to all coal gasification processes for combined cycle application and Westinghouse will be making a sizable contribution if they provide a solution to even this one problem.

It is proposed to remove the sulfur as CaS using either lime (CaO) or limestone (CaCO₃). This poses several problems:

(1) The removal may not be sufficient to meet present or proposed standards when either lime or limestone is used as the fluidizing medium in a reducing atmosphere. Westinghouse has recognized this as a potential problem and are prepared to treat the low Btu gas that is produced to remove H₂S. If this is the case, a common problem is shared; there is no proven high temperature process for removal of H₂S.

(2) Lime is considerably more expensive than CaCO₃ and requires considerable amounts of energy to manufacture.

(3) It is proposed that the fluidizing action will also result in separation of the unburned char from the heavier CaS and CaO. This is required to obtain good carbon utilization and has yet to be proven. The recent results from the CO₂ acceptor pilot plant have been encouraging in this respect.

(4) The CaS will require further treatment before it can be properly disposed of. Regeneration of spent CaS will be difficult.
Taking the entire concept, many equipment problems remain to be solved not the least of which is the turbine itself. With respect to the gasifier there are many advanced ideas which remain to be proven including the draft tube concept in the application of feeding of coal to the devolatilizer. Valving will probably be required for control of the unit with all the attendant problems.

Materials of construction always limit engineering achievement. The problems here range from finding insulating materials to operate without erosion or corrosion over long periods of time at high temperatures in a reducing atmosphere to work with turbine blades. Someone has to find solutions and it might well be Westinghouse.

Control problems will undoubtedly exist especially when all elements of the complete power generating facility are assembled and load variations make themselves felt. However the high thermal mass of the fluidized beds should make the unit inherently stable.

Turn down could very well provide a severe challenge in that proper fluidizing velocities probably cover a fairly limited range. Undoubtedly multiple units would be required but start-up and shut down problems of even a single unit would not be insignificant. Their current thinking runs to operating to produce gas within fairly wide range of Btu criterion.

Energy efficiency will probably be reasonably high if suitable insulating materials can be found and if carbon utilization proves to be high. The incorporation of the
combined cycle concept will help but with gases leaving the devolatalizer at 1500-1700°F and requiring further processing to remove particulates (and perhaps even \( H_2S \)) it seems highly unlikely that the overall energy efficiency will exceed 40%.

The proposed process doesn't seem to pose any special safety or health problems. Of course, anytime you process mixtures containing CO and \( H_2 \) at elevated temperature and pressure you're asking for trouble.

At present the advanced coal gasification system is essentially conceptual in status although as noted in the proposal to OCR "it is composed of subsystems which have been successfully operated by others." A variety of model studies and other basic experiments have been performed in an attempt to provide data necessary to design a miniplant gasifier (1200 lb/hr). The actual unit was designed by Bechtel under the supervision of Westinghouse and is under construction near Pittsburgh. Completion is scheduled for about March 1974 and it is planned to get the unit operational and obtain sufficient data to design a larger plant by March 1976.

Beyond this, two routes have been proposed:

1) Design, construct and operate a 5 ton/hr unit, and after successful operation at this scale, to proceed with the commercial scale demonstration plant at PSI.

2) Jump immediately to the large scale unit upon successful completion and operation of the mini-plant.

If the first alternative is selected, the commercial unit will not be onstream until March 1979. If the second alternative is selected the opinion is expressed that the demonstration unit can be on stream by the beginning of 1978.
In summary, Westinghouse and their associates are faced with many difficult problems in attempting to develop their Advanced Coal Gasification System for Electric Power Generation. They are to be congratulated for undertaking such an extensive development program which includes all aspects.

It wouldn't be a complete surprise if Westinghouse and their associates solved the myriad of problems facing them before others even find out what their problems are, thereby perhaps providing the utility industry with a package solution as to how coal can be used as fuel for the combined cycle. Therefore, at the very least, EPRI should follow this development with special interest and encourage this group in every way possible in their quest to help the utility industry meet its challenge of the next decade or two.

Prepared by John E. Powers
CONSOLIDATION COAL COMPANY--CO$_2$ ACCEPTOR PROCESS

The CO$_2$ Acceptor Process has been under development since 1963 on a laboratory scale (62) and with the construction of a pilot plant in 1971 this process must be considered as one of the most developed of the not yet quite-commercial coal gasification processes. Stearns Roger built and operates the 50 ton/day pilot plant at Rapid City, South Dakota for the Consolidation Coal Company. The plant has been intermittently operating during the past year or so. Several short runs and starts have been made in this period using petroleum char rather than coal as the gasifier feedstock (63). These runs have not resulted in sustained gasifier operation for longer than a week or so, however, each has resulted in knowledge which has made the following runs more successful. Operation with lignite should begin in March 1974. Future plans include operation with some bituminous coals and different dolomites. Chemical composition of the ash may be a significant factor in the adaptability of this process.

Figure 28 is a process schematic of the CO$_2$ Acceptor Process. The unique feature of this process is the use of an "acceptor" which reacts exothermically with CO$_2$ to provide the heat necessary for gasification. This results in a product gas stream which is uncontaminated by the products of combustion and undiluted by nitrogen as compared to a more conventional air-using gasification system, for example, Lurgi. The CO$_2$ Acceptor Process uses the exothermic reaction between CaO
Figure 28. Consolidation Coal Company, Inc. CO₂ Acceptance Coal Gasification Process as Originally Built
and \(\text{CO}_2\) to provide the heat of gasification.

\[
\text{CaO} + \text{CO}_2 = \text{CaCO}_3
\]

The actual acceptor material used in the process is dolomite which is a combination of calcium and magnesium carbonates. The carbonated acceptor is reactivated with the release of \(\text{CO}_2\) upon heating in the regenerator to 1900°F. Operational difficulties have occurred in this section owing to reactions with sulfur compounds in the gas.

The gasifier in this process is operated at 150 psig and 1500°F. Char or coal feed is fed into the gasifier pressure vessel from lock hoppers. The feed is introduced near the bottom of the reaction bed which is kept fluidized with steam. The volatile material in the feed coal is cracked to \(\text{CH}_4, \text{H}_2, \) and \(\text{CO}_x\) during its passage through the gasifier bed. Calcined acceptor is fed into the vapor space above the gasifier bed where it "accepts" \(\text{CO}_2\) and releases heat. The carbonated acceptor then "shower" down through the bed due to its density, which is greater than coal, and is removed from the bottom of the bed along with ash and char. This can be a delicate process, but the operators have mastered the technique. Product gas is taken from the top of the gasifier and is routed through a gas scrubbing unit if sulfur compounds are present. Typical product from the gasifier contains \(\text{CH}_4, \text{CO}, \text{CO}_2\) and \(\text{H}_2\) and has a higher heating value of 420 Btu/scf.
The bottom stream from the gasifier consisting of carbonated acceptor, char and ash is lifted to the regenerator vessel by a pneumatic conveyor. The solids contained in the lift leg of the pneumatic conveyor serve as a gas seal between the gasifier and regenerator. In the regenerator a portion of the char from the gasifier is burned with air to produce the heat necessary to regenerate the acceptor. The bed in the regenerator is fluidized at a gas velocity sufficiently high to elutriate the char ash and acceptor fines. The flue gas taken overhead from the regenerator flows through a cyclone to remove particulate matter and is scrubbed before discharge in a wet scrubber. Regenerated acceptor is fed through another pneumatic conveyor to the gasifier. The ash and fines collected in the cyclone are removed through lock hoppers and slurried with water for transportation to the disposal site.

Several problems which are more operation-oriented than process oriented have limited the length of pilot plant runs, but these are successively attacked and solved. Several mechanical modifications have been made to improve operability. Major problems encountered were:

1. Failure of the refractory linings in both the gasifier and the regenerator

2. Failure of the pneumatic conveyors to provide seals between gasifier and regenerator

3. Plugging of the pneumatic conveyors

4. Failure of the steam distributors in the gasifier to properly distribute the fluidizing steam which resulted in
inability to maintain the char-acceptor interface at an acceptable level.

5. Failure of the cyclones to operate as intended

6. Corrosion when not expected.

Modifications may still be necessary to the pilot plant which should make it operable. Additional instrumentation is being developed to monitor the flow conditions in both the fluidized beds and the pneumatic conveyors so that corrective action may be taken before improper flow causes development of a condition which would terminate operation. Operation has shown that the devolatilizer up-stream of the gasifier is not necessary. Solution of the problems which have plagued the pilot plant is expected to lead to a demonstration run of sufficient duration to quantify acceptor deactivation and attrition problems. The problems encountered to date in operating this pilot plant should not lead the reader to a negative evaluation of the CO₂ Acceptor Process. These problems are typical of all pilot programs and it is the solutions developed for these problems which make construction of a full scale plant feasible. Most of the other gasification processes have yet to reach the pilot stage where mechanical and operating problems which are unique to them can be identified.

Prepared by Brymer Williams
The gasification work at this institution (64, 65) involves a different concept in reactor design from all others covered in this report. The gasifier is the entrained flow type, but the flow of gas and solid is downwards in contrast to the commonly used up-flow designs. This design permits much smaller residence times in the reactor zone than reported in other research efforts. The research has been conducted under a contract with Bituminous Coal Research with OCR funding.

The bench scale gasification tests covered the following ranges of

- Reactor size: 0.75 - 2 in I.D.
- Coal rate: 0.7 - 4.1 lb/hr
- Coal studied: Illinois, Utah
- Coal size: -200 mesh
- Residence times: 12-300 milliseconds
- Temperatures: 1200°-2500°F
- Pressure: Atmospheric
- Conversion of Carbon Percent to gas: 20-70
- Space rate: 13-400 lb/(ft³)(hr)
- Typical gas heating value: 360 Btu/scf

Oxygen-supported combustion is the source of heat for the gasification reaction. The oxygen is introduced at the top of the reactor, sometimes premixed with hydrogen. Pulverized
coal is also fed to the top of the reactor in a carrier gas. Hydrogen, steam, and nitrogen have been tested in this use. Extensive data have been obtained in this bench unit, possibly more than any other installation covered in this (EPRI) study. The data have been analyzed with unusual thoroughness and with due regard for reaction kinetics, product distributions, energy requirements, and limitations of equilibrium.

Using these results in conjunction with the BCR data from the two larger units, reactor and process system designs have been made for various configurations including, (1) a char recycle gasifier in which the ash is removed as a slag, (2) fuel gas recycle, (3) the use of an inert solid heat carrier and (4) a scaled-up 100 ton per hour (of coal) down-flow pressure gasifier. Investment and operating cost projections based upon these designs are favorable enough to justify the next step in the progression of experimental work.

Work is being continued. A pilot plant-capacity up to fifty pounds per hour of coal—should be in operation in 1974. Additional kinetic and product distribution data will be obtained, and various mechanical features will be studied. Previous work was performed within the Chemical Engineering Department of the University, continuing work will be conducted by the same group, but within the newly formed Eyring Research Institute of Brigham Young University
At this stage in the development of the process, this reactor and accompanying process design study are, certainly, not final. The use of the laboratory data as a basis for the conceptual design strongly indicates that continuation of this high-temperature, low-residence time reactor is fully justified.

The possible benefits are:

1. A drastic reduction of the reaction volume of gasifiers. Relative to existing Lurgi and Koppers units, a reduction of as much as 80 percent may be possible.

2. A higher turn-down ratio then is possible with fluid bed units, or up-flow entrained bed basifiers.

3. Production of intermediate heating value gas with good efficiency of coal conversion.

Prepared by Brymer Williams
TEXACO INC. - PARTIAL OXIDATION PROCESS

The Texaco partial oxidation gasifier shown in Figure 29 has been operated at pilot scale as a gas producer and commercially for the production of hydrogen for the synthesis of ammonia (1).

A water-coal slurry feed of 70% through 200 mesh coal is pumped through a preheater in which the water is vaporized and the mixture heated to 1000°F at a pressure of 225 psia. The steam-coal ratio is controlled by separating steam from the mixture in a cyclone separator before gasification.

The steam-coal mixture enters at the top of the gasifier and is fed through a nozzle either axially or tangentially. Preheated oxygen or air is fed through a separate water cooled nozzle to prevent oxygen impingement on the gasifier walls. The temperature of the gases in the reaction zone range from 2000 to 2500°F. At these conditions the mineral matter forms molten slag which flows down the gasifier walls and through a constricted opening at the bottom of the gasifier into quenching water. The solidified slag is removed as a finely divided suspension in water.

Throughputs of 300 lb. coal/cuft of reactor volume and oxygen consumption rates of 15,000 to 20,000 scf/ton of dry coal have been reported (1).

Air preheat can be accomplished in a pebble-bed heat exchanger in which alumina pebbles form a downward-moving
Figure 29. Texaco Partial Oxidation Coal Gasification Process
bed. Since the raw gases contain up to 15% ash, the operation of such an exchanger above the ash fusion temperature is questionable.

Hydrocarbon Research, Inc. has proposed using this gasifier to produce hydrogen from a vacuum bottoms stream in their H-Coal Process (66).
SHELL OIL - PARTIAL OXIDATION PROCESS

Shell is marketing a partial oxidation process for high sulfur, heavy residue. They claim to have more units in operation—all overseas—than Texaco. They also claim that utilities can obtain long term contracts for such fuels. Royal Dutch Shell has an exceptional pilot plant for partial oxidation of residual fuels in Amsterdam and plans to convert to work with coal. Such work will encompass a "whole new ball game" because of the ash in coal. They would welcome financial support of this undertaking.

Prepared by John E. Powers
BITUMINOUS COAL RESEARCH - FLUIDIZED BED

Bituminous Coal Research, Inc. is also developing a multiple fluidized bed coal gasification process for the production of low Btu gas from both caking and non-caking coals (2). This is shown in Figure 30.

Fuel gas will be the only product. Operating conditions will depend upon the end use of the fuel. Gasification with air and steam will yield a low Btu gas. With oxygen and steam, a higher Btu gas can be produced. Gasification with carbon dioxide will yield a carbon monoxide rich gas suitable for perhaps MHD power generation.

Laboratory data obtained included kinetic data for various chars that would be produced in Stage 1. The studies have led to the design of a 100 lb/hr process equipment development unit. The design was completed by Foster Wheeler and OCR has agreed to support the work (67).

Prepared by Dale E. Briggs
Figure 30. Bituminous Coal Research, Inc. Fluidized-Bed Gasifier System
The Two-Stage process shown in Figure 31 is a different type gasification process in which crushed coal and limestone are dissolved at about 2500°F in a mass of molten iron (68,69). The strong affinity which exists between the iron and sulfur, keeps the sulfur in solution as well as the coal's fixed carbon. The volatile matter in the coal cracks into carbon monoxide and hydrogen. The dissolved coal sulfur migrates to a lime-bearing slag floating on top of the iron and the sulfur is removed in the slag as calcium sulfide. The slag must be desulfurized and a portion of the slag returned to the gasifier to keep the slag alkaline and the sulfur content below 5%.

The dissolved carbon in the iron is gasified to additional carbon monoxide by the reaction of air injected through a lance immersed into the molten iron. The position of the lance is above where the coal is added. Although the developers refer to the process as a two-stage coal combustion, it is really two steps in one stage.

Because of the pyritic nature of coal, the process is a net producer of iron and is in many ways comparable to the attempts at continuous steel making.

To date only small scale studies have been conducted with a 23 to 30 inch combustor. The process produces a low Btu gas
Figure 31. Applied Technology Corporation Two Step Gasification System
in the range of 190 Btu/scf containing particulates which would have to be removed.

A review of the process by Battelle (70) indicated that work to date had not demonstrated three key aspects for successful operation.

The PATGAS process is similar to the Two-Stage process except that oxygen is used. The product gas would have a heating value of 315 Btu/sch. In the ATGAS process the PATGAS product is upgraded by methanation to a heating value of 940 Btu/scf.

Prepared by Dale E. Briggs


37. Wellman-Galusha Gas Equipment, Bulletin No. 147, McDowell Wellman Companies, Cleveland, Ohio, 16 pp.


70. Corder, W.C., H.W. Lowrie, W.T. Reid, C.E. Sims, T.W.
    Zegers, "A Technical Evaluation of the BS&D Coal Combustion
    Process to TVA", (Contract No. 72 P 33-12757) Battelle,
    May 19, 1972.
COAL DISSOLUTION AND LIQUEFACTION

by

Dale E. Briggs
David E. Hammer

Review and Assessment

Coal can be dissolved in solvents to produce liquid products. The first systematic study of the action of solvents on coal was made by de Marsilly in 1862 (1). Catalytic hydrogenation was investigated by Berguis (2) in 1911 using an impure iron oxide in a pasting oil to produce gases and oils. Solvent extraction of bituminous coal was initiated in 1927 by Drs. Pott and Broche (2). Sinnatt (1) completed an extensive study of solvent effects in 1933 and the U.S. Bureau of Mines (3) began a systematic study in 1936 of the hydrogenation and liquefaction of American coals and lignites.

Gasoline was produced commercially by catalytic hydrogenation of coal in 1927 in Germany. In 1942 Germany produced 32,000,000 barrels of aviation gasoline from coal.

The current work in coal dissolution can be sub-divided as shown in Figure 1. The British have experimented in recent years in dissolving coal in toluene above the critical pressure of toluene (4). High yields of low-molecular weight aromatic hydrocarbons are produced.
Figure 1. Classification of Coal Dissolution and Liquefaction Processes
The processes closest to commercial development are those which utilize recycle solvents derived from the coal itself. These dissolution processes can be carried out in three general ways to yield liquids.

Consolidation Coal Company (Consol) (5) and Exxon (6) dissolve 70-90% of the coal (MAF) in a hydrogen donor solvent at approximately 750°F and 400 psig. The mineral matter is separated from the coal extract and the extract hydrogenated in the presence of a catalyst to produce low sulfur oils and recycle hydrogen donor solvent.

Pittsburg and Midway Coal Mining Co. (PAMCO) (7) dissolves coal in the presence of a recycle solvent and hydrogen or a mixture of hydrogen and carbon monoxide at approximately 800°F and 1500 psig. In this process 90-95% of the coal (MAF) can be liquefied to produce approximately 1% S liquids which congeal below 300°F. If low sulfur oils are needed, subsequent processing is required.

Gulf Research and Development Corporation (8) and the U.S. Bureau of Mines (9) dissolve coal in a recycle solvent in a fixed bed reactor in the presence of hydrogen gas and a desulfurization catalyst. The operating conditions are approximately 800°F and 3500 psi. Under these conditions over 90% of the coal (MAF) is dissolved to produce low sulfur fuel oils. Depending upon the operating conditions high distillate yields are possible. These can be converted to gasoline, diesel fuel and jet fuel.
The Hydrocarbon Research, Inc. HRI (10) process is a coal dissolution process carried out in a ebullated bed reactor in the presence of hydrogen and a desulfurization catalyst at conditions comparable to the other catalytic processes and with comparable products. Although the catalytic processes are capable of dissolving and desulfurizing coal in a single reactor, the catalyst activity declines more quickly than it does in ordinary crude oil desulfurization because of the presence of the mineral matter in the coal.

GENERAL OBJECTIVES AND IDENTIFICATION OF AN IDEAL SYSTEM

An ideal liquid fuel process is one in which coal can be converted to liquids with minimum processing and maximum thermal efficiency. An Illinois No. 6 coal represented by the chemical formula \( \text{CH}_{0.795}^0 \text{N}_{0.013}^0 \text{O}_{0.108}^0 \text{S}_{0.018}^0 \) and containing 7.13% ash and 2.7% moisture can be converted by dissolution to a low sulfur fuel oil, represented by the chemical formula \( \text{CH}_{1.1}^0 \) by the addition of hydrogen. Ideally, only about 3.6 scf hydrogen/lb raw coal or 7200 scf/ton is required. The sulfur, nitrogen and solids levels in the final product should satisfy process and environmental requirements.
COAL DISSOLUTION PROCESSES

Coal dissolution or liquefaction processes are directed toward converting coal to liquid fuels with minimal production of gases and organic solid residues. All ranks of coal can be liquefied although some are clearly better than others. The liquid products vary both with the type or rank of coal liquefied and the process used. It is therefore as important to understand the nature and character of coal as it is to understand the processes.

From the work to date in coal liquefaction in the presence of hydrogen and a catalyst, it appears that western coals and lignites can be liquefied under less severe conditions to produce less viscous liquids but the hydrogen consumption per ton of coal is greater (8,10). Some western coals can also be liquefied without desulfurization catalyst to produce a liquid product at ambient conditions. The alkali metals in the mineral matter promote hydrogen transfer through a catalytic effect (11).

Chemistry of Coal Dissolution and Liquefaction

From its origin as woody material, peat proceeds through biochemical and chemical change from a low rank coal to a high rank coal with ever changing carbon to hydrogen and carbon to oxygen ratios. The ranks in order of increasing age are peat, brown coal, lignite, subbituminous, bituminous (low, medium and high volatile) semi-anthracite, anthracite and methanthracite.
Higher rank coals have a greater weight fraction carbon and generally have a higher heating value.

On a molecular level coal consists of aromatic lamellae. The individual lamellae consist of mono-, di-, tri- and perhaps tetra-cyclic aromatic monomers with mean molecular weights of 230-350 connected by methylene linkages into generally linear polymers with some cross linking through oxygen to carbon and sulfur to oxygen bonds. The coal polymer has some distribution of molecular weights, but an average of two or three thousand is probable (12). The extent of the cross linking is not considered to be too significant since coal will solubilize under conditions that should not cleave covalent bonds.

Through millions of years of chemical change, gases formed and diffused out of the coal to voids and fissures and accounts for high methane concentrations in coal mines. Free radicals were left in the coal. The immobility of the solids prevented reaction.

When coal is contacted with coal derived solvents, such as anthracene oil, and heated, the coal begins to swell, takes up solvent and goes into solution. As the temperature and time of contact increases the organic portion of the coal is nearly completely dissolved in the coal solvent. At the same time, the stable free radicals which had existed lose their stability and the number of free radicals increases due to molecular detachment at methylene linkages and at sulfur to
carbon or oxygen to carbon bonds which originally cross-linked the large polynuclear coal structure. If hydrogen can be transferred to the free radicals relatively low molecular weight molecules will form. If hydrogen cannot be transferred, the aromatic lamellae will repolymerize into significantly larger molecules---asphaltenes. Immediately after solubilization, there is a tendency for hydrogen to be transferred from certain molecules to others in moving toward chemical equilibrium (13). Molecules which lose hydrogen tend to polymerize into larger molecules. This tendency is minimized when adequate hydrogen is present.

It is quite clear from all the experimental results, that the ability to transfer hydrogen to the free radicals as they form is essential. This can be improved in two ways; by increasing the mass transfer conditions or the amount of hydrogen that can be transferred. In other words the rate of mass transfer and the driving force for mass transfer are both important.

Coal solubilization appears to be most rapid in aromatic and hydroaromatic ring compounds which have a boiling range of 570-610°F, the ability to transfer hydrogen, a relatively high dipole moment and ring stability (14). Most of the compounds are heterocyclic molecules frequently containing phenol rings.
Coal dissolution can take place with or without the presence of hydrogen gas and with or without the presence of a catalyst. The catalyst may be a commercial desulfurization catalyst or a catalytic effect may be obtained from the alkali metals in mineral matter found in the coal. The process variations and the process identifications are shown in Table I. In all the processes, it is essential that the process be able to produce enough solvent to recycle back to slurry and dissolve the coal.

**TABLE I**

Summary of Coal Dissolution Methods

<table>
<thead>
<tr>
<th>Without Hydrogen</th>
<th>With Hydrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td>Without Catalyst</td>
<td>Consol</td>
</tr>
<tr>
<td></td>
<td>Exxon</td>
</tr>
<tr>
<td>With Catalyst</td>
<td>Consol*</td>
</tr>
<tr>
<td></td>
<td>Exxon*</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*For coals for which the minerals act as a catalyst.

**Dissolution Processes in Development**

Table II lists the major dissolution processes under development and/or pilot plants under construction. In addition, coal liquids research is being supported by the Office of Coal Research at several universities (15). The Colorado School of Mines is studying "The Removal of Sulfur from Coal by Treatment with Hydrogen." The University of North Dakota
<table>
<thead>
<tr>
<th>Process</th>
<th>Developer</th>
<th>Location</th>
<th>Engineering Design</th>
<th>Present Development Size</th>
<th>Next Stage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Project Gasoline or Consol</td>
<td>Consolidation</td>
<td>Cresap, W. Va.</td>
<td>C.F. Nofsinger Co.</td>
<td>20 tons/day</td>
<td></td>
</tr>
<tr>
<td>Synthetic Fuel</td>
<td>Coal Company</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PAMCO Solvent Refined Coal-SRC</td>
<td>Pittsburg &amp; MidwayCoal Mining Co.</td>
<td>Merriam, Kansas</td>
<td>Pittsburg &amp; Midway</td>
<td>75 lb/day Bench Scale and 1 ton/day Process Development plant</td>
<td>Pilot plant under construction</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Tacoma, Wash.</td>
<td>Stearns-Roger</td>
<td>50 tons/day (construction scheduled for completion)</td>
<td></td>
</tr>
<tr>
<td>Solvent Refined Coal</td>
<td>Southern Services (PAMCO SRC process)</td>
<td>Wilsonville, Ala.</td>
<td>Catalytic, Inc.</td>
<td>6 tons/day (scheduled to operate on coal January 1974)</td>
<td></td>
</tr>
<tr>
<td>H-Coal</td>
<td>Hydrocarbon Research, Inc.</td>
<td>Trenton, N.J.</td>
<td>HRI</td>
<td>3 tons/day</td>
<td>700 tons/day</td>
</tr>
<tr>
<td>Synthoil</td>
<td>U.S. Bureau of Mines</td>
<td>Bruceton, Pa.</td>
<td>U.S. Bureau of Mines</td>
<td>1000 lbs/day</td>
<td>8 tons/day</td>
</tr>
<tr>
<td>Gulf Catalytic Coal Liquids</td>
<td>Gulf Research &amp; Development</td>
<td>Harmarville, Pa.</td>
<td>Gulf R&amp;D</td>
<td>120 lbs/day</td>
<td>1 ton/day</td>
</tr>
</tbody>
</table>
has a project on "Premium Fuels from Northern Great Plains Lignite-Project Lignite." The University of Utah has a project on "Intermediate Coal Hydrogenation Process."

The National Science Foundation (RANN) is supporting work at Auburn University, the University of Kentucky and the University of Michigan related to solvent refining of coal.

Exxon is doing work in coal liquefaction along the lines of the Consol CSF process but little information has been released.

Several conceptual commercial size plant designs have been completed in the past two or three years. Some of these are given in Table III. The thermal efficiencies reported for the plant designs vary from 63.5% to 73.9%. In some recent studies by HRI, a value of 73-75% looks possible (16).

The economic projections are subject to considerable uncertainty, but are probably comparable or somewhat more expensive than low Btu gasification on a $/KW basis.

A brief description of the major development processes can be found under Process Descriptions in this section. Particular attention was directed toward where the critical problem areas are and how they are treated. A summary of the major operating conditions and products is given in Table IV.
TABLE III. Conceptual Commercial Size Coal Liquefaction Plants

<table>
<thead>
<tr>
<th>Process</th>
<th>Engineering Design</th>
<th>Plant Size tons coal/day</th>
<th>Coals</th>
<th>Main Fuel Oil Products</th>
<th>Estimated Overall Thermal Efficiency %</th>
<th>Plant Capital Cost* Million $</th>
<th>Date of Study</th>
<th>MW Potential at 35% Efficiency</th>
<th>Cost $/MMBtu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Modified SCR</td>
<td>Ralph M. Parsons (17)</td>
<td>10,000</td>
<td>Illinois No. 6, 3.4%S</td>
<td>0.2%S 13.9*API</td>
<td>63.5</td>
<td>270</td>
<td>1973</td>
<td>620</td>
<td>435</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Pittsburg Seam 4.2%S</td>
<td>0.5%S -9.7*API</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Consol CSF</td>
<td>Foster-Wheeler (18)</td>
<td>20,000 (MF)</td>
<td></td>
<td>0.056%S 58*API</td>
<td>71-73.9</td>
<td>230</td>
<td>1972</td>
<td>1530</td>
<td>150</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.128%S 10.3*API</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>H-Coal</td>
<td>Hydrocarbon Research, Inc. (10)</td>
<td>25,000 (MF)</td>
<td>Illinois No. 6, 5%S</td>
<td>0.1%S 27*API</td>
<td>69.6</td>
<td>299</td>
<td>1973</td>
<td>1800</td>
<td>166</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.5%S -3.1*API</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gulf CCL</td>
<td>Gulf R&amp;D(8)</td>
<td>33,000</td>
<td>Wyodak 0.7%</td>
<td>&lt; 0.2% 39.3*API</td>
<td>**</td>
<td>445</td>
<td>1973</td>
<td>2000</td>
<td>222</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Big Horn 0.54%</td>
<td>&lt; 0.04%S 35.3*SPI</td>
<td>**</td>
<td>423</td>
<td>1973</td>
<td>2300**</td>
<td>184***</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.04%S 9*API</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Does not include interest during construction
** Not given
*** Estimated
(MF) Moisture Free
### TABLE IV. Coal Liquefaction Process Operating Conditions and Typical Products

<table>
<thead>
<tr>
<th>PROCESS</th>
<th>H-COAL</th>
<th>PARSONS MODIFIED PAMCO</th>
<th>PAMCO (G.SERV.)</th>
<th>BUR. OF MINES</th>
<th>GULF CCL</th>
<th>GULF CCL</th>
<th>CONSOL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen used in dissolution?</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Subsequent Extract Hydrogenation?</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Catalytic Dissol Approximate Reactor Temperature Reactor Pressure.</td>
<td>Yes 850°F</td>
<td>840°F</td>
<td>850°F</td>
<td>840°F</td>
<td>800°F</td>
<td>800°F</td>
<td>730°F</td>
</tr>
<tr>
<td></td>
<td>3000 psig</td>
<td>1200 psig</td>
<td>1500 psig</td>
<td>4000 psig</td>
<td>3000 psig</td>
<td>3000 psig</td>
<td>400 psig</td>
</tr>
<tr>
<td>Coal Sulfur, Wt. %</td>
<td>Ill. No. 6 5%</td>
<td>Ill. No. 6 3.38%</td>
<td>--</td>
<td>Kentucky 4.6%</td>
<td>Big Horn Subbit. 0.54%</td>
<td>Pittsburgh Seam (Bit.) 1.49%</td>
<td>Pittsburg Seam Coal 3.67%</td>
</tr>
<tr>
<td>Solvent to Coal Ratio (to slurry).</td>
<td>1:1(by Wt.)</td>
<td>2.0:1(by Wt.)</td>
<td>2:1(by Wt.)</td>
<td>1.22:1.0(by Wt.)</td>
<td>2.33:1.0(by Wt.)</td>
<td>91%</td>
<td>63%</td>
</tr>
<tr>
<td>Percent Coal Dissolved (MAP), Hydrogen Consumption Scf/ton Coal (MAP).</td>
<td>90%+</td>
<td>90%+</td>
<td>90%+</td>
<td>90%+</td>
<td>90%+</td>
<td>90%+</td>
<td>90%+</td>
</tr>
<tr>
<td>Solids Separation, Solids Content in Product.</td>
<td>15,300</td>
<td>12,600</td>
<td>82% by Wt. 7600</td>
<td>Filtration 0.23 Wt.%</td>
<td>Centrifuge 1.3 Wt.%</td>
<td>Hydroclones &amp; Filt. 0.02 Wt.%</td>
<td>Hydroclones &amp; Filt. 0.03 Wt. %</td>
</tr>
</tbody>
</table>

**Principal Products**

1. **Fuel**
   - **Yield bbl/ton** 1.73 bbl/ton
   - **API gravity** -3.1°API
   - **Viscosity**
   - **Sulfur, Wt. %** 0.5%
   - **Nitrogen, Wt. %**

2. **Fuel**
   - **Yield bbl/ton** 0.54bbl/ton
   - **Viscosity**
   - **Sulfur, Wt. %** <0.1%
   - **Nitrogen, Wt. %** 0.2%

*Exclusive of H₂ Production*
Identification of Problem Areas

The general processing steps in coal dissolution are shown in Figure 2. In general coal is crushed, dried and pulverized prior to producing a slurry with recycled solvent. The slurry is heated and the coal dissolved in the solvent. In most processes the dissolution step is carried out in the presence of hydrogen. The extract is cooled to remove hydrogen, hydrocarbon gases and hydrogen sulfide, and the liquid flashed to a low pressure to separate condensable vapors from the extract. Mineral matter and organic solids are next removed and used to produce hydrogen for the process. The solids free extract can then be hydrogenated to desulfurize the extract and improve the hydrogen transfer characteristics of the solvent or it can be separated into the recycle solvent and the major product without intermediate processing.

In reviewing the work being done to produce liquid fuels from coal, which can be used by the electric power industry, particular attention has been directed toward the identification of the critical problem areas. Process modifications to avoid serious operating problems have been and continue to be sought.

All the processes require energy to convert coal to low-sulfur liquid fuels. Process modifications to maximize energy conservation consistent with process economics are needed.
Figure 2. Processing Steps in Coal Dissolution and Liquefaction.
In the processing steps shown or indicated in Figure 2, the critical operations are:

(1) Slurry Pumping
(2) Preheater - Dissolver
(3) Pressure Let-down
(4) Solids - Extract Separation
(5) Energy Recovery
(6) Hydrogen Production and Carbon Utilization

There are minor problems and considerations in each processing step. Careful design and construction should eliminate most of them. Each processing step will be discussed in some detail.

**Coal Preparation**

No particular coal preprocessing is required. It would however be preferrable if as much mineral matter as practical be removed in coal cleaning operations at the mine as long as coal losses are not economically significant.

The raw coal is first crushed and then pulverized and dried in conjunction with pulverization. The particle size is not particularly critical and standard pulverizers are adequate. At Cresap, West Virginia, Consolidation Coal Co. used -14 mesh coal (19). Gulf Research and Development indicate -60 mesh, HRI, -80 mesh, Bureau of Mines, -100 mesh and Pittsburg and Midway Coal Mining Co., -200 mesh. The smaller particles have some advantages in slurrying, in preventing settling, and in enhancing the coal dissolution rate.
Coal-Solvent Slurrying

Prepared coal is slurried with recycle solvent to make a feed mixture which can be pumped through the preheater to the dissolver-reactor.

The viscosities at 167°F of slurries made up of a representative coal and solvent, at various solvent-to-coal ratios are given in Table V. A break in the curve between 55 and 63% indicates that an upper limit for coal concentration does exist. The solvent-to-coal ratio required is dictated by process operations, e.g., pressure drop in dissolution equipment, coking in solution preheaters, filtration rate, etc., and not necessarily by pumping capabilities.

TABLE V

Viscosity of a Representative Coal Slurry at Various Concentrations of Coal (7)

<table>
<thead>
<tr>
<th>Coal Concentration, Wt.%</th>
<th>Viscosity, cp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>4.5</td>
</tr>
<tr>
<td>21</td>
<td>11</td>
</tr>
<tr>
<td>25</td>
<td>18</td>
</tr>
<tr>
<td>33.5</td>
<td>36</td>
</tr>
<tr>
<td>35</td>
<td>50</td>
</tr>
<tr>
<td>40</td>
<td>90</td>
</tr>
<tr>
<td>45.5</td>
<td>260</td>
</tr>
<tr>
<td>50</td>
<td>540</td>
</tr>
<tr>
<td>55</td>
<td>1000</td>
</tr>
<tr>
<td>63</td>
<td>∞</td>
</tr>
</tbody>
</table>
As shown in Figure 3, slurry viscosity decreases substantially with temperature (19). This makes the slurry more pumpable and offers some advantages of heat recovery when the recycle solvent is hot. At higher temperatures, safety, pump maintenance and premature coal dissolution become matters of concern.

In the Stearns-Roger's solvent refined coal pilot plant design (20), the slurry tank was heated and equipped with a mixer. The slurry was recirculated by a centrifugal pump to the inlet of a reciprocating feed pump, with provision for returning the excess to the slurry heating vessel. Mixing and recirculation are needed to prevent settling.

From the Consolidation Coal Co. (19) work at Cresap, West Virginia, they believed a 1.5 solvent-coal ratio would be satisfactory for slurry preparation providing an additional 0.5 solvent fraction were added to the dissolver-reactor.

**Slurry Pumping**

There are three major pump applications:

1. Moderate temperature (up to 300°F), low head, and low pressure centrifugal pumps for solvent-coal recirculation.

2. Intermediate temperature (up to 500°F), high head (up to 4500 psi) reciprocating pumps for slurry feed.

3. High temperature (up to 850°F), low head and high pressure (up to 4500 psi) for solids-extract recirculation.
Figure 3. Viscosity Values for Fine Coal Particles in 0:1 and Water (19)
Centrifugal pumps for solvent-coal recirculation suffer from erosion. Equipment improvements are needed to improve reliability.

The dissolver-reactor feed pumps have been of the triplex plunger type. Depending upon the process, they must operate up to 400°F and 4500 psig. Extract solidification on the plunger rod causes erosive conditions which can destroy packing. At Cresap (19) a solution to the packing problem was developed by a redesign of the packing gland to provide two packed sections separated by a lantern ring to which flush liquid was injected.

At lower temperatures and pressures to 1200-1500 psi the triplex plunger pumps are satisfactory (21). Continued development is needed to improve long term reliability at higher temperatures and pressures.

The viscosity of the solvent-coal slurry increases significantly during dissolution and reaches a peak as the coal goes through the plastic stage. This is illustrated in Figure 4. The equipment must be designed to accommodate for this condition.

The Consol and HRI processes involve reactor recycle pumps. These pumps must operate at the reactor operating conditions. Although the head requirements are small, the operating conditions are severe. It is quite possible that reactor recycle pumps can be used advantageously in other processes to recover heat and to minimize processing requirements. Work is needed to improve reliability.
Figure 4. Viscosity Peaks from Bath Extraction of Kentucky No. 11 Coal (19)
Preheater and Dissolver-Reactor

The solvent-coal slurry is heated and the coal liquefied in the preheater and dissolver-reactor. Because of the nature of the dissolution process, the preheater and the dissolver-reactor should be considered together conceptually.

Preheater. The solvent-coal slurry is preheated from the slurry preparation and storage temperature of 100-300°F to the reactor inlet temperature of 650-750°F in a preheater. In the pilot plants or proposed demonstration plants the preheaters have been direct fired tubular furnace heaters. For a fully integrated plant, the fuel gases for the heater could come from the process itself as fuel gases are produced in liquefaction or in hydrogen production.

As the cost of energy continues to increase, the need for energy recovery becomes essential. A significant amount of the heat required for slurry preheat should be obtained by heat exchange with the extract from the reactor which must be cooled to separate the gases and condensible vapors from the liquid product.

Hydrogen, if used in dissolution, is injected prior to preheating. The amount injected is substantially more than consumed to insure an adequate partial pressure driving force. The hydrogen not consumed is recovered and recycled.

The hydrogen purity is not exceptionally critical. As the purity is reduced the total system pressure must be
increased to maintain the required hydrogen partial pressure. In certain cases as will be discussed later, a synthesis gas of hydrogen, steam and carbon monoxide is very satisfactory under certain conditions.

Dissolver-Reactor. The dissolver-reactor vessel has taken various forms in the processes advocated to date. These different forms have been directed toward taking the coal through the plastic stage under the best reaction conditions possible. Since coal contains oxygen and nitrogen in addition to organic and inorganic sulfur, attention must be paid toward the fuel oil product requirements. The choice of operating temperature, operating pressure and solvent also depend upon the products desired and process economics.

The amount of coal that is dissolved is important. The fraction of the coal dissolved must be consistent with the overall operating and thermal efficiency requirements of the process. The fraction of the coal that is not dissolved must generally be used to produce hydrogen for the process in order to realize good carbon utilization. Partially dissolved coals continue to dissolve and react after they leave the reactor.

The rate of dissolution depends upon the coal, coal particle size, solvent, temperature and general agitation. It is quite clear that the coal slurry should not be preheated beyond the point where any appreciable dissolution occurs.
When dissolution does begin, the slurry should be in the dissolver-reactor under the most favorable mass transfer conditions to insure hydrogenation. Sufficient heat release occurs in the dissolver to further raise the temperature. At 50-60 Btu/scf of hydrogen consumed, the temperature rise can approach as much as 150°F.

The types of reactions which occur depend upon temperature and time. The reactions which remove nitrogen, oxygen and sulfur from the coal are desirable but the hydrocarbon cracking reactions which reform the midrange molecular weight molecules into gases and asphaltenes are to be avoided if liquid fuels are desired. Reactors operated at high temperatures and low through-puts produce more gases and asphaltenes and use more hydrogen. Typical residence times are 4-60 minutes.

The three basic methods of coal dissolution will now be treated with regard to the dissolver-reactors.

(1) Coal Dissolution Without Hydrogen or Catalyst. In the Consolidation Coal Co. Consol Synthetic Fuel (CSF) Process a hydrogen donor solvent is used to dissolve the coal at 730-765°F and 400 psig. At these conditions hydrogen is transferred from the donor to the free radicals of the coal molecules as they detach from the solid or fracture. The solvent subsequently becomes deficient in its ability to transfer hydrogen and must be regenerated by hydrogenation.
The dissolution process is carried out in a stirred extractor vessel. Adequate stirring is essential in order to have the hydrogen donor solvent present when a chemical bond is broken. For a Pittsburgh Seam (Ireland Mine) bituminous coal, about 78% of the coal (MAF) is dissolved to produce about 75% extract, 3% gas and 22% residue. The extract would contain about 0.9% S depending upon the sulfur content of the coal and would be a liquid at ambient conditions.

Since hydrogen must be produced for rehydrogenation of the donor solvent, the 22% residue can be used to produce hydrogen by gasification and is therefore not necessarily a liability. In addition, some fuel gas is needed for preheating.

There is need however for second reactor to rehydrogenate all or a portion of the extract to produce adequate hydrogen donor solvent for recycle and to produce adequate fuel products as required. The extract hydrogenation reactor operates at 775-800°F and 4200 psig in the presence of a catalyst. Although the need for a second reactor is a disadvantage in cost, the operating conditions with a catalyst are considered more satisfactory since most of the mineral matter and dissolved carbon can be removed before feeding the extract to the reactor. Organometallic compounds, usually reported as TiO₂, which are soluble in the extract, do affect the catalyst.
Exxon is doing pilot work in coal liquefaction along the same basic concept as Consol. They believe this method gives them greater flexibility and control over the final products (6).

(2) Coal Dissolution with Hydrogen but Without Catalyst. In the Pittsburg and Midway Coal Mining Company (PAMCO) Solvent Refined Coal Process (SRC), a coal derived anthracene oil is used to dissolve the coal at about 800°F and 1000-1500 psig in the presence of hydrogen gas. The extract mechanism for hydrogen transfer to the coal is not clear. Some think it is through the hydrogen donor capacity of the solvent with the dissolved hydrogen serving to rehydrogenate the solvent and others think there is some direct reaction of dissolved hydrogen with the coal as dissolution and bond fracture occur.

The dissolution process is carried out in a tubular reactor with preheated coal slurry and hydrogen fed at the bottom. Agitation is enhanced by the hydrogen in the gas phase, although not to the same extent as would occur with a mixer.

In the continuous flow bench scale unit at Merriam, Kansas, Pittsburg and Midway Coal Mining Company (7) dissolved close to 90% of a Kentucky No. 11 coal (MAF) at 800°F and 1000 psig to convert 76% of the coal (MAF) to a solid product and 12% to gases. The extract from the SRC process is usually a solid (congealed liquid) at ambient temperatures.
It contains less than 0.1% mineral matter. The product has a potential advantage in terms of storage but may present problems in pulverizing and handling because of the low softening temperature. Burning characteristics will need evaluation.

The SRC process, processing a bituminous coal with recycle solvent and nominally pure hydrogen, will produce a more viscous extract and more gases than the Consol CSF process. Although the solvent is more capable of dissolving the coal, it is not as effective in transferring hydrogen to the free radicals of the coal molecules and many repolymerize to form larger molecules. The mass transfer conditions are also not as good. Higher temperatures favor gas formation through thermal cracking.

Over 60% of the organic sulfur is removed in the reactor. The pyritic sulfur can be removed subsequently when the mineral matter is separated from the extract.

The Southern Services pilot plant at Wilsonville, Alabama is nominally a SRC plant.

The SRC process is "non-catalytic" in the sense that a commercial catalyst is not used in the reactor. There is considerable experimental evidence, however, that the inorganic mineral matter in certain western lignites and subbituminous coals has a significant catalytic effect, especially sodium. While the catalytic effect is certainly not as great as that obtained from a fresh commercial hydrodesulfurization catalyst, the extract viscosity and sulfur content are both reduced.
Additional laboratory work at the Pittsburg and Midway Laboratory has revealed that the use of carbon monoxide and steam to replace part of the hydrogen promises to result in further process improvements\(^{(22)}\). This concept is based on the work at the U.S. Bureau of Mines \((23,24,25)\). The recent work of Pittsburg and Midway indicates that the use of carbon monoxide and steam was more effective than hydrogen alone for processing lignite and subbituminous coals. Mixtures of carbon monoxide, hydrogen and steam have been found to be about as effective as carbon monoxide and steam alone. The results for bituminous coal are not so clear.

The reaction of carbon monoxide and steam in the dissolvers forms hydrogen in excess of that used in the solution process. The mechanism appears to be through the formation of sodium formate as shown in the following equations.

\[
\text{NaHCO}_3 + \text{CO} + \text{H}_2\text{O} \rightarrow \text{NaCOOH} + \text{CO}_2 \quad (1)
\]

\[
\text{NaCOOH} + \text{H}_2\text{O} \rightarrow \text{NaHCO}_3 + \text{H}_2 \quad \text{Coal} \quad (2)
\]

with the net reaction being

\[
\text{CO} + \text{H}_2\text{O} \rightarrow \text{H}_2 + \text{CO}_2 \quad (3)
\]

(3) Coal Dissolution with Hydrogen and Catalyst. The work of HRI, Gulf R&D, and the U.S. Bureau of Mines has been directed towards dissolving coal in the presence of a desulfurization-hydrogenation catalyst and hydrogen to produce the desired
products in a single reactor. Micron and sub-micron size organic and inorganic particles present in the coal extract adsorb onto the catalyst to a certain extent. This makes the operating conditions for the catalyst more severe. There has been concern that catalyst activity will be difficult to maintain. HRI uses an ebullated bed reactor and Gulf R&D and the U.S. Bureau of Mines use a fixed catalyst bed.

The catalytic liquefaction processes operate at 800-850°F and 2500-4000 psig to produce a 0.19-0.4% sulfur fuel extract which can be separated into recycle solvent, a low sulfur distillate and a 0.3-0.5% sulfur residual fuel oil. All are capable of dissolving 90+% of the coal (MAF).

The advantages of catalytic assisted coal liquefaction are:

1. Less hydrogen is required than for non-catalytic operations when the product sulfur level is to be below 0.6% sulfur.

2. Less feed preheat is required because of the heat released in the desulfurization and hydrogenation.

3. Mass transfer conditions for hydrogenation of the coal free radicals are better and the extract molecular weight distribution is narrower.

4. Compared to non-catalytic dissolution, about a 50°F lower temperature is required to dissolve the same fraction of the coal with the same space velocity; or, at the same temperature, a higher space velocity can be used.

5. The amount of gases and heavy asphaltic materials produced is less and the amount of middle range oil is greater.
6. The product is more amenable to different types of mineral matter separation schemes because much of the lower viscosity extract can be taken as an overhead product in a vacuum distillation and the viscosity is more acceptable to filtration at moderate temperatures.

7. The process can easily produce an excess of recycle solvent.

8. Considerably more sulfur can be removed in one step at a slight increase in operating cost when compared to other one step processes.

The disadvantages of catalytic assisted coal liquefaction are:

1. Catalyst is required to operate in a liquid containing 3-10% solids in the micron-size range. Metals from the mineral matter tend to accumulate on the catalyst, thus decreasing activity (26).

2. Without regeneration catalyst requirements would be large. HRI found they needed 1 lb. catalyst per ton coal processed in their bench scale unit to maintain catalyst activity (27).

3. Catalyst regeneration would be required to be economical. Regeneration cost is about one-third the cost of fresh catalyst (27).

4. Regeneration catalyst approaches fresh catalyst activity but declines in activity about three times as rapidly (27).

HRI uses an ebullated bed reactor in which the upward passage of the solids, liquid and gaseous materials maintains the 1/32 - 1/16 inch catalyst in a fluidized state in the lower three-fourths of the reactor. The extract is withdrawn from the top of the reactor, normally free of catalyst. A fraction of the extract is recycled straight back to the reactor to reduce the solid coal concentration and to improve agitation.
The recirculation and internal circulation keeps the reactor temperature uniform and allows the inlet feed slurry temperature to be lower.

Catalyst can be withdrawn from and fed to the reactor while operating. This is a distinct advantage.

Gulf R&D and the U.S. Bureau of Mines have operated small vertical fixed bed reactors. The reactor can potentially be smaller than the ebullated-bed reactor but care must be taken to operate the reactor in such a way as to prevent plugging. The extract temperature rises along the bed length from the heats of reactions. There would be an advantage to recirculating hot extract to the reactor to minimize preheat, to reduce the potential plugging and to further increase mass transfer. The process must be shut down to replace catalyst.

Gulf R&D has done substantial work in catalyst development. The U.S. Bureau of Mines has done a substantial amount of commercial catalyst evaluation.

**Gas-extract Separation**

Hydrogen, hydrogen sulfide, ammonia and other gases are removed from the extract-gas mixture by cooling and phase separation. As mentioned previously, the future economics associated with thermal efficiency will likely dictate that the hot extract be used to partially preheat the coal slurry. The hydrogen is recovered from the gases and recycled to the preheater.
This process step does not appear to present any critical problems.

**Pressure Let-down**

There are one or two pressure let-down steps in all the liquefaction processes. The pressure is dropped from the reactor pressure to a lower pressure to remove condensible hydrocarbons prior to separating the solids from the extract and/or prior to distillation of the extract into the products needed. The work by Consol at Cresap and HRI both indicate a further need for reliable pressure let down equipment since malfunctioning of the systems did occur. As with pumps, mechanical components at severe operating conditions lead to problems.

Pressure let down systems can take various forms such as expansion through a controlled orifice, controlled volume let down (28), and turbine expansion or piston expansion with energy recovery. Innovative ideas are needed.

**Extract-Solids Separation**

Separation of the mineral matter and undissolved organic matter has been one of the unanswered critical problem areas. Several approaches are possible none of which are completely satisfactory. Some of the alternatives have been:

1. Filtration
2. Hydroclones
3. Centrifugation
(4) Evaporation-distillation
(5) Carbonization-coking
(6) Solvent precipitation and/or solvent washing

Filtration work has been conducted on bench and process development units by Pittsburg and Midway (7), Consol (29), HRI (10), the Bureau of Mines (11), and Gulf R&D (8). Most of the work has been directed toward rotary drum filtration through a precoated filter. Cake blinding by micron-size particles has been a problem. To obtain adequate filtration rates, the temperature must be high enough to keep the extract fluid. Depending on process conditions, most extract from the catalytic liquefaction processes is fluid enough to be filtered at temperatures as low as 100°F (8,11). HRI has been able to achieve filtration rates of 160 lb/hr-sq.ft. submerged surface at 400°F with a 30 psi pressure difference.

Since filtration is a mechanical process, it is to be avoided if possible.

Hydroclones, 2-3 inch diameter liquid cyclones, have been evaluated for at least partial separations. Consol did a substantial amount of evaluation work at Cresap (19, 29) and HRI (10) and Gulf (28) have included hydroclones in preliminary process designs. Their greatest potential use is to effect a partial separation. In such cases the low-solids overflow can be recycled back to the reactor and the
high solids underflow further processed. Because of the size required, hundreds of hydroclones must be used in parallel and this presents problems.

Centrifugation work to date has indicated that this method is not satisfactory (7,19,29). Some feel that a well-designed centrifuge could work, however.

Evaporation or distillation is a way of concentrating the solids in a high boiling residue extract. Since some distillation is always required to effect product separation and solvent recycle, this method looks fruitful. The less viscous extract from the catalytic processes lends itself better to distillation. By vacuum distillation, it is possible to take 80-90% of the solids-free extract as overhead distillate leaving the solids in a 800-900+ residue which can be gasified to produce hydrogen or synthesis gas. Entrained solids, carried over in the distillate, should not be a problem.

Carbonization or fluid coking was evaluated at Cresap (19) as a means of removing solvent from the solids and also to produce low ash char or coke from extract containing little or no mineral matter. The only advantage over distillation is that it leaves the residue dry.

Solvent precipitation of organic solids has been examined but is considered too expensive (10).
Solvent Regeneration

Recycle solvent is recovered from the extract generally by distillation. By blending certain cuts, it is possible to tailor the solvent to the end-products desired to a certain extent. It is essential that adequate recycle solvent be produced in order to maintain self-sufficient operations.

In the case of the hydrogen-donor solvents, it is necessary to hydrogenate at least the recycle solvent portion of the extract.

Energy Recovery

Energy recovery is essential in reducing the operating cost of coal dissolution processes. This becomes more important as the cost of coal increases.

The reactor effluent must be cooled to separate gases from the extract, extract must be cooled somewhat prior to filtration and the final liquid product must usually be cooled before storage. At the same time the recycle solvent must be hot for slurrying the coal, and the slurry must be heated to the reactor temperature. It is therefore important to include heat exchanger equipment to maximize the potential gain from energy recovery. As always there is a trade-off between added capital cost and reduced operating costs.
The 63.5% thermal efficiency in the Parsons' Demonstration Plant Design (17) reflects the absence of heat recovery equipment. When such equipment is included, it appears possible to increase the thermal efficiency up to 71-75% (16,18) depending upon the process and products. The less viscous products from catalytic liquefaction are more amenable to heat recovery in terms of process options and process heat exchange equipment.

Hydrogen Production

Hydrogen consumption has been identified as one of the major cost factors in coal liquefaction. The hydrogen or synthesis gas required in liquefaction must be produced by gasification of fresh coal or preferrably carbon containing residues from the liquefaction process. The residues can be either dry or liquid, although the liquid residue would have an advantage since it can be slurry pumped to the gasifier operating pressure. The bottoms from the vacuum distillation column containing mineral matter and undissolved carbon would be a satisfactory feed to a gasifier. It is also a way to maximize carbon utilization.

Compared to fresh coal, heavy liquid residues and dry carbonaceous wastes are fairly refractory and require higher temperatures for gasification.

Catalytic, Inc. (30) made a brief study of coal gasification for hydrogen production and estimated little or no
difference in hydrogen costs between Lurgi gasification at 430 psig and Kopper-Totzek gasification at 1 atmosphere. In a detailed study of the Lurgi, Kopper-Tolzek and Winkler gasifiers by the Indian Government (31) for the production of hydrogen for ammonia synthesis, the Koppers-Totzek process was recommended because of its simplicity of operation and construction and the general satisfaction expressed by the process users on a variety of feedstocks. Foster Wheeler (32) compared the Lurgi and Bi-Gas type gasifiers for hydrogen production from coal and concluded that the Bi-Gas type gasifier had the potential for lower costs.

For heavy liquid residues and dry carbonaceous wastes, the slagging type gasifier looks attractive because of the high temperature and potential to run with high concentrations of mineral matter. Since such feed stocks contain little volatile matter, the two stage Bi-Gas type gasifier offers no significant advantage over the single-stage Koppers-Totzek gasifier. Although, the commercial Koppers-Totzek gasifiers operate at atmospheric pressure, there is no reason why they could not operate at 150-300 psi on liquid feeds which are air/oxygen and steam atomized.

The Shell and Texaco partial oxidation gasifiers have operated commercially for years at such pressures with heavy oil feed stocks containing little or no solids. From the
combined experience of Koppers and Shell, a slagging single stage gasifier should be able to be built to operate on mixtures of liquids and solids.

There is a definite advantage to operating the gasifier at pressures in terms of gas clean-up, shift conversion of carbon monoxide and steam to hydrogen, and hydrogen compression. Desulfurization and gas purification do not appear to be problem areas, but attention is needed to effect energy recovery where possible from the gases prior to clean-up.

CONCLUSIONS AND RECOMMENDATIONS

The progress and development in coal dissolution or liquefaction has been studied with respect to producing low sulfur and nitrogen fuels for electric power generation. The liquid products range from distillates with less than 0.04% S and 0.2% N by weight to heavy fuel oils containing 0.5% S by weight. The solid (congealed liquids) product from a solvent refined coal process contains about 1% S by weight depending upon the sulfur content of the original coal. The products are storable which make them suitable for intermediate and peak shaving load applications. The capital costs estimates for coal dissolution plants shown in Table III can not be considered reliable or firm since they represent conceptual designs in a period of material shortages and inflation. They
do give some idea of the cost of coal dissolution relative to stack gas clean up and low-Btu gas generation. The overall process thermal efficiency, defined as the total energy available divided by the total energy required, falls in the range of 63-75% and is believed to be fairly reliable. It is quite clear that as the cost of coal increases, the thermal efficiency of conversion becomes increasingly important. With a tightly balanced and integrated material balance and energy recovery, it should be possible to reach 75% overall thermal efficiency.

Coal dissolution plants require several processing steps and would be considered complex. Control and operation should not be a problem because of the years of experience in petroleum processing. It is natural to expect that these plants will be operated by petroleum companies and the fuels sold for electric power generation.

As crude oil shortages persist, it is quite likely that coal liquefaction and coal pyrolysis plants will be built for gasoline, fuel oil and chemical feed stocks when scale-up data become available. Heavy residual oils from such plants should fill utility needs as they do today.

From our study we conclude that single step catalytic coal dissolution and desulfurization (and denitrogenization) offers the best potential to go to commercial scale at this time. The work of Hydrocarbon Research, Inc. in the development of the H-Coal process has made substantial progress in terms of commercial-size applications. It is therefore considered a worthy process for support in the area of coal liquefaction. Support should be directed toward construction and operation
of a large pilot plant or small demonstration plant to evaluate reactor scale-up and to evaluate other critical processing steps with equipment of near commercial-size.

Catalytic coal liquefaction offers two definite advantages. First, lower sulfur fuel oils (<0.5% sulfur) and distillates (<0.1% sulfur) can be produced at costs comparable to other single step processes. Secondly, the product is less viscous and lends itself to process variations which are less expensive, more thermally efficient and more reliable. Catalytic liquefaction should work on both eastern bituminous coals and western subbituminous coals and lignites.

Catalyst cost may represent some 3-5% of the operating costs depending upon the price of coal. Catalyst development is needed in: (1) selectivity for the products desired, (2) maintaining activity in the presence of gaseous and metallic poisons and (3) catalyst reactivation.

Recent developments by Pittsburg and Midway in the use of synthesis gas in place of hydrogen for coal liquefaction of western coals warrants consideration. This development offers potential savings in hydrogen production while producing a product of acceptable sulfur levels which is fluid enough to possibly eliminate filtration for solids removal. This type of work should be carried out at the Southern Services Wilsonville, Alabama pilot plant. The operating staff and the engineering back-up appear to be good.

Small scale work on fixed bed catalytic liquefaction by Gulf R&D and the U.S. Bureau of Mines looks attractive. There are some advantages to fixed bed reactors, but plugging problems
cannot be considered solved and small scale work should be pursued. It could well be appropriate to consider the installation of a fixed bed reactor in the Wilsonville pilot plant once the problems are better in hand.

It will be advisable to continue to make use of the pilot plant facilities that are currently available because of time, cost, and experience of operating personnel. With this in mind, demonstration plants should be designed with the maximum long range flexibility and adaptability.

Hydrogen production constitutes a substantial fraction of the capital investment and contributes heavily to the loss of overall process thermal efficiency. From the preliminary economic evaluations by HRI (10,16), it appears that the best way to produce hydrogen (from a cost and thermal efficiency standpoint) is by gasification of the vacuum distillation bottoms which contain the solids from coal dissolution. This gasification feed stock would be pumpable to a gasifier operating at pressure. HRI has included a Texaco partial oxidation gasifier in their designs. Process development and engineering is needed for gasifying liquids containing high levels of solids. The Koppers-Totzek gasifier works well on coals containing high mineral matter content and on liquids without solids. It could likely work well for liquids containing 20-40% solids or dried filter cake combined with coal.

Development work is needed at a commercial-size level on all the components parts of coal dissolution processes which contain moving parts such as pumps, control valves, filters
and pressure let-down valves. The experience at Cresap clearly indicates that process success is more related to mechanical operability than to realizing product yields and specifications. Mechanical components should be operated and evaluated at commercial-size, independent of the non-mechanical components of a process when possible. This would expedite the time to reach commercial scale. Specifically, a slurry test loop with a high pressure drop let-down system for high temperature operation should be commissioned.

As in all research and development programs, the progress in the various methods of coal dissolution must be evaluated regularly. Research management must be prepared to terminate support selectively, where projections are not satisfactory, or where, among competing processes the success of one eliminates the need to develop others.
HYDROCARBON RESEARCH, INC. - H-COAL

The H-Coal process (10) is a coal dissolution process carried out in an ebullated bed reactor in the presence of hydrogen and a desulfurization catalyst. The process has been operated effectively on two ranks of coal, Illinois No. 6 and Wyodak coals, at the bench and process equipment development unit scale (3 tons/hr. of coal). A schematic drawing of the process is given in Figure 5.

The H-Coal process for coal conversion to liquid products was invented by Hydrocarbon Research, Inc. (HRI) as a further application of the ebullated bed technology developed in the H-Oil process. H-Oil ebullated bed reactors have operated commercially since 1963.

The H-Coal process should be effective with all types of coal. It should be particularly useful on high volatile eastern coals containing high sulfur. These coals tend to cause problems in gasification and pyrolysis processes and require substantial stack gas clean-up if burned in a conventional boiler. The fuel oil requirements in the eastern states are also substantial.

In the H-Coal process coal is dried to 150-200°F, pulverized and slurried with coal derived solvent oil, then mixed with compressed make-up hydrogen and fed to the preheater and reactor. The heart of the process is the ebullated bed reactor containing catalyst. The coal is catalytically hydrogenated
Figure 5. H-Coal Process for Fuel Oil Production—Devolatilization Plant
as the dissolution occurs. Upward passage of the solid, liquid and gaseous materials maintains the catalyst in a fluidized state. The relative size of the catalyst and coal is such that only the extract, unconverted coal, mineral matter and gaseous products leave the reactor while the catalyst is retained therein. Catalyst can be added and withdrawn continuously to maintain a near constant catalyst activity. The reactor provides a simple means of controlling reactor temperature and a good contact between the reacting species and the catalyst.

Internal circulation is facilitated by a hot oil recycle pump which takes suction from the liquid phase at the top of the catalyst-oil disengaging space in the reactor. With this recycle, preheat to only 700°F is required for an 850°F reactor temperature. The heat release associated with hydrogenation is 50–60 Btu/scf hydrogen consumed. Operating pressure is about 3000 psig.

The reactor operating conditions depend upon the products desired from a particular type of coal. The principal independent variables are temperature, hydrogen partial pressure and liquid residence time in the reactor. A maximum liquid yield from an Illinois No. 6 coal is obtained at 850°F and 3000 psig.

The reactor product slurry is let-down at reactor temperature to the atmospheric pressure flash drum in which a portion of the lighter hydrocarbon liquids is flash-vaporized and fed to the atmospheric tower. The slurry material remaining after
the atmospheric flash is separated with hydroclones into a lower solids overhead stream for recycle solvent. About two-thirds of the solids can be removed quite easily. The higher solids underflow stream is filtered with a precoated rotary drum filter to remove the mineral matter and undissolved carbonaceous matter. The use of hydroclones make it possible to recycle solvent without processing the total reactor effluent in a distillation step. This provides a substantial savings in distillation but the hydroclones represent a potential problem because of the large number required. Filtration of a higher solids slurry is an advantage, however.

The filtrate can be further separated by vacuum flash or distillation to produce vacuum distillate and vacuum bottoms. By a process modification the high solids hydroclone underflow can be sent directly to a vacuum distillation column where the fuel oil product is taken off as an overhead product, and the mineral matter and undissolved carbonaceous residue can be removed as a high solids slurry in the vacuum bottoms. The fraction of the feed which can be taken overhead is related to the reactor operating conditions.

The choice of removing solids by filtration or by vacuum distillation depends to a certain extent on the method for producing hydrogen. With filtration, the carbon in the residue filter cake is not sufficient for hydrogen needs and some coal must be used in addition. For high pressure gasification, lock hoppers would be needed.
When the solids are removed by vacuum distillation, the entire product must be taken as an overhead product. This makes the distillation step more expensive but eliminates filtration. The solids which are collected in the vacuum bottoms can be pumped, atomized and gasified at pressure without additional coal. Lock hoppers are avoided.

The reactor vapors not condensed at ambient temperature are scrubbed with a medium volatility oil in the recycle gas scrubber. Light hydrocarbons are removed and the hydrogen concentration is increased in this scrubbing system before the gas is returned to the H-Coal reactor.

Most of the experimental work has been done on Illinois No. 6 and Wyodak coals. Typical coal analyses are given in Table VI and products in Table VII. The catalyst used in the H-Coal operations is a commercial hydrogenation catalyst. Bituminous coals require less hydrogen for liquefaction as seen in Table VII.

By increasing through-put by two and one-half times, a high portion of low sulfur fuel oil is produced instead of synthetic crude while maintaining high coal conversion and greatly reduced hydrogen consumption. This is shown in Table VII. The product is more viscous and contains more sulfur than the synthetic crude oil.

The H-Coal Process has been under active development by HRI since 1964. In the period prior to 1971, HRI first supported the research work itself, then on a contract with the Office of Coal Research, and finally a contract (about two
TABLE VI

Coal Analysis (As-Received) - H-Coal Study (10)

<table>
<thead>
<tr>
<th></th>
<th>ILLINOIS NO. 6</th>
<th>WYODAK</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture, Wt.%</td>
<td>17.5</td>
<td>30.4</td>
</tr>
<tr>
<td><strong>Proximate Analysis, Wt.% (Dry Basis)</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ash</td>
<td>9.9</td>
<td>7.9</td>
</tr>
<tr>
<td>Volatile Matter</td>
<td>42.0</td>
<td>44.1</td>
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<tr>
<td>Fixed Carbon</td>
<td>48.0</td>
<td>48.1</td>
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<tr>
<td><strong>Ultimate Analysis</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon</td>
<td>70.7</td>
<td>68.4</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>5.4</td>
<td>5.4</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>1.0</td>
<td>0.8</td>
</tr>
<tr>
<td>Sulfur</td>
<td>5.0</td>
<td>0.7</td>
</tr>
<tr>
<td>Oxygen (Difference)</td>
<td>8.1</td>
<td>16.9</td>
</tr>
<tr>
<td>Ash</td>
<td>9.9</td>
<td>7.9</td>
</tr>
<tr>
<td>Desired Product</td>
<td>Illinois Synthetic Crude</td>
<td>Illinois Low-Sulfur Fuel-Oil</td>
</tr>
<tr>
<td>---------------------------------</td>
<td>--------------------------</td>
<td>------------------------------</td>
</tr>
<tr>
<td>Normalized Product Distribution</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$C_1$-$C_3$ Hydrocarbons</td>
<td>10.7</td>
<td>5.4</td>
</tr>
<tr>
<td>$C_4$-$400^\circ$F Distillate</td>
<td>17.2</td>
<td>12.1</td>
</tr>
<tr>
<td>400-$650^\circ$F Distillate</td>
<td>28.2</td>
<td>19.3</td>
</tr>
<tr>
<td>650-$975^\circ$F Distillate</td>
<td>18.6</td>
<td>17.3</td>
</tr>
<tr>
<td>875$^\circ$F+ Residual Oil</td>
<td>10.0</td>
<td>29.5</td>
</tr>
<tr>
<td>Unreacted Ash-Free Coal</td>
<td>5.2</td>
<td>6.8</td>
</tr>
<tr>
<td>$H_2O, NH_3, H_2S, CO, CO_2$</td>
<td>15.0</td>
<td>12.8</td>
</tr>
<tr>
<td>Total (100.0 + $H_2$ Reacted)</td>
<td>104.9</td>
<td>103.2</td>
</tr>
<tr>
<td>Conversion, % M.A.F. Coal</td>
<td>94.8</td>
<td>93.2</td>
</tr>
<tr>
<td>Hydrogen Consumption, SCF/Ton</td>
<td>18,600</td>
<td>12,200</td>
</tr>
</tbody>
</table>

$C_4$ + Liquid

<table>
<thead>
<tr>
<th></th>
<th>Illinois Synthetic Crude</th>
<th>Illinois Low-Sulfur Fuel-Oil</th>
<th>Wyodak Synthetic Crude</th>
</tr>
</thead>
<tbody>
<tr>
<td>API Gravity</td>
<td>15.0</td>
<td>4.4</td>
<td>26.8</td>
</tr>
<tr>
<td>Hydrogen, Wt.%</td>
<td>9.48</td>
<td>8.43</td>
<td>10.54</td>
</tr>
<tr>
<td>Sulfur, Wt.%</td>
<td>0.19</td>
<td>0.43</td>
<td>0.16</td>
</tr>
<tr>
<td>Nitrogen, Wt.%</td>
<td>0.68</td>
<td>1.05</td>
<td>0.64</td>
</tr>
</tbody>
</table>
years) with Atlantic Richfield Company. The aggregate funds expended during this time were something more than $4,000,000.

Effective January 1, 1971 HRI entered into contracts for the purchase and sale of technology with six companies: Ashland Oil, Atlantic Richfield, Consolidation Coal (Continental Oil Company), Esso Research and Engineering (EXXON USA), Gulf Minerals Resources (Gulf Oil Company), and Sun Oil Company.

In the two-year period which terminated December 31, 1972 HRI spent an aggregate of $2,640,000. Of this total roughly 75-80% was spent at the Trenton, New Jersey laboratory. The balance was spent for engineering studies to evaluate the experimental work, to prepare conceptual designs and cost estimates for commercial plants, and to study the design and cost of a demonstration-scale unit. The experimental work includes extensive bench-scale studies as well as work in the process equipment development unit (PEDU) with an 8-inch reactor. It can process three tons of coal per day. The PEDU has operated for 44 days on Illinois No. 6 coal. In September 1973, the PEDU had over 150 total days of operation.

Three of the above six companies did not choose to continue to purchase technology in 1973 strictly for budget reasons. These three were Consolidation Coal, Gulf Mineral Resources and Sun Oil Company.

On January 1, 1973 HRI acquired a new purchaser, namely Standard Oil Company (Indiana). The program for the year 1973 is currently budgeted at a level of $1,240,000.

The next stage of developed proposed is a 700 ton/day demonstration plant.

Prepared by Dale E. Briggs
Initial process work began in 1962 under contract to the Office of Coal Research (7). Small batch (150 gms. of coal) and small continuous flow reactor (75 lbs/day) studies led to the design, construction, and operation of a 1 ton/day process equipment development plant. A 50 tons/day of coal pilot plant (33) is currently being built by Rust Engineering at Port Lewis, Tacoma, Washington under Office of Coal Research support. The plant which is scheduled for mechanical completion around March 1974 will be operated by Pittsburg and Midway. A schematic drawing of the pilot plant process is given in Figure 6. Hydrogen will not be generated from coal or extract residue but would have to be in a commercial plant.

Raw coal is pulverized (50-200 mesh) and mixed (1 part by weight coal and 2 to 3 parts solvent) with a recycle solvent having a boiling point range of about 550°-800°F. This solvent is nearly identical to the anthracene oil derived from coal tar which is used for starting the plant. The resulting slurry, together with hydrogen, is passed through a direct fired furnace preheater to a series of dissolvers. In the preheater and the dissolvers, the coal is "depolymerized" and the organic material almost completely dissolved (over 90% of the coal MAF) in the solvent at 850°F and 1050 psig. The undissolved carbon may be graphitic carbon inherent in the coal.
Figure 6. Pittsburg & Midway Coal Mining Company Solvent Refined Coal Process - SRC
The dissolvers consist of four vertical tubes in series with the flow of both liquid and gas upward through the dissolvers. Initially, solvent is taken up by the coal causing the coal to swell and the slurry viscosity to increase significantly. As the temperature and time of contact increases, the coal is nearly completely dissolved in the solvent. It is necessary to have hydrogen present to tie up the free radicals generated in the depolymerization. The presence of hydrogen is also important in converting the organic sulfur and some of the inorganic sulfur to hydrogen sulfide. The inorganic sulfur is removed with the mineral matter. This would leave about 1% sulfur compared to 4-6% in an Illinois coal feed. It would be expected that little nitrogen would be removed.

With up-flow of gas and liquid in the dissolvers, the dissolvers will act as particle size classifiers. The nominal velocity is 1.67 ft/min. The larger particles will have a tendency to settle and stay in the dissolvers until they are small enough to be carried over. A 3-phase (vapor-liquid-solid) slurry goes overhead.

Following the dissolver step, the excess hydrogen, plus hydrogen sulfide, carbon dioxide and light hydrocarbons produced in the reaction, are separated from the slurry of undissolved solids and coal solution. The gas is then scrubbed to remove $\text{H}_2\text{S}$ and $\text{CO}_2$. The excess hydrogen is recycled after removing some of the methane and other light hydrocarbons produced in the dissolver. To maintain hydrogen purity in the circulating gas, fresh hydrogen is added before recycling it.
to the dissolver. The hydrocarbon gas, CO₂ and H₂S (after conversion to elemental sulfur) are potential by-products of the process. The slurry from the dissolver goes to the mineral separation area where the solids are separated using either a rotary precoat filter or series of centrifuges. The extract from a non-catalytic liquefaction process tends to react further outside the reactor. There appears to be some hydrogen transfer with the formation of asphaltenes which make filtration more difficult. The effects are not critical for short time storage but are for long term storage.

Continuous pressurized filtration is planned for separating the extracted coal solution from the mineral and carbonized residue. This method was selected over hydroclones which have no moving parts and fewer operational difficulties. Unfortunately, typical solids overflow from hydroclones at Cresap averaged 12% solids (7% of which is mineral residue) and the underflow contained approximately 17% of the feed liquid (29). These figures make hydroclones unattractive when used as the only solids separation step.

Two Goslin-Birmingham Corporation rotary pressure precoat filters are being installed at Fort Lewis (33). One has an area of 40 square feet and the other has an area of 80 square feet. The extract liquid-mineral residue slurry will be cooled to 600°F and the pressure reduced to 150 psig for filtering. This is a compromise between ease of filtration and reliability of equipment. The conditions are
rather severe. Nitrogen will be used to maintain a pressurized gas atmosphere for the upper half of the rotary drum to force the extract through the precoat and filter medium. The drums are segmented to permit a wash and dry cycle as well as filtration.

The solids are washed with a light wash solvent in the filter or centrifuge and then dried. The dried solids (mineral residue) consist of about 50 to 70% inorganic material from the coal plus some undissolved carbon.

The solids-free coal solution goes to the solvent recovery area where the solvent is removed by vacuum flash separation, is further fractionated into a light hydrocarbon by-product stream, a wash solvent stream for recycle to the filter or centrifuge, and a process solvent stream for recycle to the dissolvers.

The vacuum residue congeals to a solid at about 300°F. When cooled to ambient temperature, it is an ash-free solid with a fairly low sulfur content. It is the major product of the process. The liquid can be spray-cooled or prilled to shiny beads which form a free-flowing solid bed, probably transportable by pneumatic means. This product can be remelted and burned as a liquid fuel or pulverized and burned as a solid fuel. Limited studies on pulverizing and burning the solid (congealed liquid) product indicate that substantial work must be done on large lots of product to test the acceptability of the product (34,35).
Recent work by Pittsburg and Midway (22,26) has been
directed to coal dissolution in the presence of synthesis
gas, a mixture of hydrogen, steam and carbon monoxide,
instead of pure hydrogen. By a catalytic effect, certain
metals in western coals produce hydrogen from steam and
carbon monoxide and at the same time produce a product which
is liquid at ambient temperatures and makes an attractive concept.
They propose to evaluate this further in the pilot plant at
Tacoma, Washington.

The Ralph M. Parsons Company under contract of the Office
of Coal Research completed the design of a demonstration
plant (17). The design constraints were specified by OCR
and include many of the recent P&M developments. Figure 7
is a schematic flow sheet of the process.

Prepared by Dale E. Briggs
Figure 7. Parsons—PAMCO Hybrid Demonstration Plant Schematic Flow Sheet
SOUTHERN SERVICES INC. - SOLVENT REFINED COAL

The Edison Electric Institute and the Southern Company system, an electric utility group operating in the Southeast, are jointly sponsoring a $6,000,000 research project leading to the development of a "clean" utility fuel from coal (37,38). The project involves construction and operation of a 6 tons/day of coal pilot plant for studying key steps in solvent refining of coal as originally developed by Pittsburg and Midway Coal Mining Company in their SRC process.

The cost of the pilot plant was $4,000,000 and the cost of one year of operation and related development work will be about $2,000,000. The Edison Electric Institute pledged $4,000,000 to support the project. Southern Services, Inc., an affiliate of the Southern Company, pledged $2,000,000 support, provided a site for the pilot plant, and is providing management to the project.

Under contract to Southern Services, Inc., Catalytic, Inc. designed and built the pilot plant and will operate the pilot plant for one year. Construction was completed in late August 1973 near Wilsonville, Alabama, about 35 miles southeast of Birmingham, and the SRC pilot plant was to begin operation on coal in early 1974.

In the solvent refined coal process shown in Figure 8, pulverized coal is mixed with about three parts of a solvent fraction that is generated internally in the process. This
recycled solvent fraction has a boiling range of about 550-850°F. Hydrogen is added to the slurry of coal and solvent and the mixture is preheated and fed into a single-stage reactor or dissolver. The reactor operates at a temperature of about 825-850°F, and a total of about 1000-2000 psig. Under these conditions, most of the carbon in the coal feed is dissolved during the residence time of approximately 30 minutes in the reactor. About 60% of the organic sulfur in the coal is converted to hydrogen sulfide. Substantially all the inorganic sulfur is removed later by filtration. The hydrogen consumption was given as 1-2% of the weight of the coal processed, but this may be on the low side.

The effluent from the reactor is passed to a high pressure receiver where the liquid and gas phases are separated. The liquid slurry is then subjected to a mineral separation step where the undissolved solids are removed. Filtration will be the method of mineral separation initially used in the pilot plant. Other techniques for mineral separation are also being considered for investigation. The filtrate from which the solids have been removed is flashed in a vacuum column. Process solvent is recovered overhead and recycled to slurry the coal feed. Bottoms from the vacuum column form the solvent refined coal product which solidifies at about 300°F. If the processing plant is adjacent to a power plant, the solvent refined coal may be handled and burned directly as a hot, viscous liquid. If not, the solvent refined coal must be solidified for subsequent
storage and handling. If the product is maintained as a liquid at about 300°F, the liquid composition tends to change. Both smaller and larger molecular weight molecules form with the larger molecules being asphalthic in nature. The process for product solidification that will be used in the initial pilot plant studies involves freezing the refined coal in a water bath from which it is moved by a conveyor. Other methods of product solidification, such as prilling and flaking, will also be studied. Solidification is a likely problem area.

In the pilot plant, the gas phase from the high pressure receiver will be scrubbed with a 20% caustic solution to remove the hydrogen sulfide produced in the reactor. The waste caustic solution will be disposed of by a contract waste hauler. In a commercial plant, one of many commercial absorption processes would be used and the sulfur would be recovered as elemental sulfur.

The solid residue from the filter is discharged to a rotary dryer. The filter residue contains a substantial quantity of wash solvent which is recovered and recycled back to the process. The dried solids, which contain most of the pyritic sulfur, other ash-forming materials, and some undissolved carbon from the coal feed will be hauled from the pilot plant to disposal. It has been suggested that the solid residue may have some value, but investigations of its use will not be included in the project.
The properties of the solvent refined coal product are reasonably independent of the raw coal feed, which should offer advantages in both design and operation of coal-fired facilities. The ash content of the product is reduced to about 0.1%, and sulfur is reduced to less than 1%. On western lignites and subbituminous coals, sulfur levels as low as 0.3% may be possible. The solvent-refined coal has a higher heating value than raw coal, about 16,000 Btu/lb compared with 8000-13,000 Btu/lb. The ability to use any raw coal feed leads to easing fuel procurement problems. The high value and quality of the fuel product leads to potential savings in fuel transportation and storage facilities, and perhaps, to a reduction in furnace size.

Because the refined coal is essentially free of ash, pulverizers may be smaller and require less power and less maintenance. Pulverization tests will have to be carried out, although, to establish if the solvent refined coal can be pulverized without getting sticky. The need for ash handling and ash storage facilities, precipitators and ash ponds will be minimized.

Because the sulfur content of solvent refined coal is low, lower preheater temperatures may be possible, leading to higher plant efficiencies and less preheater maintenance. Without this refined fuel, scrubbers and high stacks will be required at most coal-fired plants, and some credits may be taken due to elimination of such requirements when clean fuel is assured.
The solvent refining process promises to be less expensive than some competing processes for converting raw coal into a clean solid or liquid fuel because, by comparison, it requires (1) less hydrogen consumption, and this requirement for hydrogen may be further reduced by replacement with carbon monoxide and steam or synthesis gas at even less cost; (2) potentially less capital and operating costs as a result of less severe operating conditions; and (3) no catalyst. On the other hand the sulfur levels are considerably higher than for the catalytic dissolution processes and the product is a solid (congealed liquid) rather than a pumpable liquid at ambient temperatures.

One of the more important evaluations of the program will be to test the storage, handling, pulverizing and combustion characteristics of the solid SRC product. The limited tests to date have been inconclusive (34,35).

Prepared by Dale E. Briggs
Gulf R&D has been working on coal for about six years somewhat in parallel with residual oil desulfurization. Their work has been directed toward fixed bed coal liquefaction. They have considerable experience in catalyst development and they believe they have made progress in catalysts for coal liquefaction.

To date the Gulf R&D work has been on a small pilot unit with a feed rate of 120 lbs/day (8). A conceptual commercial catalytic coal liquids plant is shown in Figure 9. Feed coal is ground and slurried with a solvent generated by recycling a portion of the product. The recycle solvent can be obtained in several ways as discussed in the other processes. The choice depends somewhat on the coal and quality of the product required. The feed slurry is combined with hydrogen, heated and passed through a specially designed fixed bed reactor which is supposed to minimize plugging. From the reactor the product goes to a gas-liquid separator where hydrogen is recovered for recycle. The liquid product goes to solids separation. Gulf has experimented with hydroclones to separate the liquid reactor effluent into a low solids overflow steam which is recycled for slurrying with the feed coal and a high solids underflow product which is sent to a solids removal process such as filtration or vacuum distillation. The product would be taken off as filtrate or distillate, respectively.
Figure 9. Gulf R&D Catalytic Coal Liquids Process
In the initial work, anthracene oil was used as a solvent and the unit operated without solvent recycle. Recently, two hydroclones in series have been used to separate the reactor product into a low solids recycle solvent and a high solids stream. The high solids stream is easily filtered on a rotary drum filter to produce the final product because of the low viscosity. A six week long run was completed successfully with no reactor plugging. During a 10 minute power failure, plugging in the preheater occurred. When the preheater was replaced the unit was restarted with no further difficulty and no apparent loss in catalyst activity.

Results of runs on the coals listed in Table VIII are given in Table IX. A material balance for hydroclone recycle runs are given in Figure 10.

Coking the filter cake to recover some of the heating value has been examined. This concept would have to be evaluated in terms of the overall process economies. Hydrogen production is obviously a part of it.

Gulf is currently considering building a 1 ton/day pilot plant to provide engineering data for the design of a 500 tons/day semi-works unit. The rationale behind building a plant this size is based on:

(1) Detailed process and mechanical information for commercial plant design, including operating and maintenance experience on commercial scale equipment, could be developed.

(2) The reactor, which is the heart of the process would be of sufficient size for thorough evaluation and little risk would be involved in further scale-up to commercial reactor size.
**TABLE VIII**

Typical Analyses of Coals (8)

<table>
<thead>
<tr>
<th>RANK</th>
<th>GLENHAROLD MINE</th>
<th>BIG HORN</th>
<th>KENTUCKY #9 &amp; #11*</th>
<th>PITTSBURG SEAM</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>LIGNITE</td>
<td>SUBBITUMINOUS</td>
<td>BITUMINOUS</td>
<td>BITUMINOUS</td>
</tr>
<tr>
<td>PROXIMATE ANALYSIS,</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WT. %</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MOISTURE</td>
<td>32.1</td>
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<td>VOLATILE MATTER</td>
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<td>FIXED CARBON</td>
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<tr>
<td>ASH</td>
<td>7.4</td>
<td>3.4</td>
<td>16.2</td>
<td>8.1</td>
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<tr>
<td>CHEMICAL ANALYSIS,</td>
<td></td>
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<td></td>
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<tr>
<td>WT. % (MOISTURE FREE BASIS)</td>
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<tr>
<td>CARBON</td>
<td>65.44</td>
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<td>76.84</td>
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<tr>
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<td>4.60</td>
<td>5.06</td>
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<td>0.40</td>
<td>1.46</td>
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<td>0.54</td>
<td>4.63</td>
<td>1.49</td>
</tr>
<tr>
<td>ASH</td>
<td>10.90</td>
<td>4.39</td>
<td>15.00</td>
<td>8.28</td>
</tr>
<tr>
<td>TOTAL</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
</tr>
</tbody>
</table>

**HEAT OF COMBUSTION (GROSS)**

AS RECEIVED, BTU/LB 7,420 8,730 11,590 13,510

*NOT TYPICAL: CHOSEN FOR HIGH SULFUR CONTENT*
Table IX
Comparison of Catalytic Liquefaction Runs Using Various Coals (8)

<table>
<thead>
<tr>
<th>RANK</th>
<th>GLENHAROLD MINE</th>
<th>.BIGR.NORN</th>
<th>KENTUCKY #9 &amp; #11</th>
<th>PITTSBURG SEAM</th>
</tr>
</thead>
<tbody>
<tr>
<td>LIGNITE</td>
<td>40</td>
<td>30</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>SUBBITUMINOUS</td>
<td>30</td>
<td>30</td>
<td>BITUMINOUS</td>
<td>BITUMINOUS</td>
</tr>
</tbody>
</table>

RESULT SUMMARY

LIQUEFACTION, % (a) 95 91 92 90

FILTRATE ANALYSIS

| GRAVITY, °API | 1.4 | 5.1 | 6.7 | 1.2 |
| VISCOSITY, CS at 210° | 2.5 | 2.0 | 3.5 | 4.3 |
| HYDROGEN, WT% | 7.65 | 8.22 | 7.56 | 7.85 |
| NITROGEN, WT% | 0.60 | 0.41 | 0.73 | 0.63 |
| OXYGEN, WT% | 1.01 | 0.75 | 1.18 | 1.00 |
| SULFUR, WT% | 0.07 | <0.05 | 0.19 | 0.11 |

---

(a) % LIQUEFACTION = \( \frac{\text{LB MAF COAL FED} - \text{LB MAF UNDISSOLVED COAL IN SLURRY PRODUCT}}{\text{LB MAF COAL FED}} \times 100 \)
Figure 10. Process Material Balance for Bituminous and Subbituminous Coals (8)
(3) A 500 ton per day plant can be built with commercially available equipment.

(4) The plant would be able to supply enough product for field tests in power plant furnaces and diesel engines.

(5) The plant will be big enough to be incorporated later into a commercial operation, if desired.

Prepared by Dale E. Briggs
U.S. BUREAU OF MINES - SYNTHOIL

The U.S. Bureau of Mines has been operating a 5- to 10-lb/hr catalytic coal liquefaction process unit for five years to produce low sulfur oil in a process called Synthoil (9,39,40). A schematic drawing of the unit is shown in Figure 11.

Five coals of different grades have been processed in the 5 lb/hr unit and have demonstrated the applicability of the process and have added to the knowledge of coal dissolution. All the products had less than 0.3% sulfur even from coals containing about 5% sulfur.

A slurry of recycle solvent and coal is mixed, preheated and conveyed through the reactor with recycled hydrogen and a small amount of makeup hydrogen. The reactor in the 5 lb/hr unit is a folded 5/16-inch ID by 68 ft long tube packed with 1/8-inch pellets of a commercial catalyst as used in desulfurization of petroleum derivatives (cobalt molybdate on silica-activated alumina catalyst). The reactor is usually operated at 840°F and 2000-4000 psig.

Effluent gases are separated from the extract in the high pressure receivers. The hydrogen sulfide and ammonia are removed and the hydrogen recycled. After pressure let-down the extract oil is either centrifuged or filtered to remove mineral matter and undissolved organic matter. Over 90% of the organic coal substance is dissolved. The product oil flows freely at room temperature and is filterable at moderate temperatures.
The key to the long-term operability with the fixed catalyst bed is the turbulent flow of hydrogen which promotes excellent mass transfer for dissolution and chemical reaction and at the same time allows the coal to pass through the sticky plastic stage without adhering to or plugging the bed. In the system the intent is to just do enough hydrogenation to remove the sulfur and to take whatever amount of liquefaction that occurs. The hydrogenation is mild because of the short residence times and little gas and asphalt-like components are formed. This reduces the amount of hydrogen needed which is a major operating expense. The hydrogen consumption is about 3000 scf/bbl product or about 9000 scf/ton coal.

The fixed bed reactor has the potential to be smaller in size than the ebullated-bed reactor and to produce an excellent product with less hydrogen consumption. There are certain disadvantages. The amount of hydrogen recycle (compression) is high, the reactor must be shut-down to replace catalyst and there is always some danger of plugging. This problem has occurred in their experimental work. Titanium, as an organometallic compound, is a potential catalyst poison, but this is a problem with all desulfurization catalysts.

Since the product oil has a comparatively low viscosity, several options are open to separating the solids from the oil.
The operating temperature range is somewhat limited. At low temperatures, the liquid viscosity is too high and the pressure drop through the reactor excessive. At high temperatures, the liquid viscosity and pressure drop are lower but the amount of gas produced becomes excessive and hydrogen consumption is greater. By increasing the solvent to coal ratio, the operating temperature can be lowered somewhat. There are several economic and engineering trade-offs.

Scale-up considerations are important. The Bureau of Mines, has recently completed a 1/2 ton/day pilot plant. The reactor is 1-inch ID x 15 ft long and packed with 1/8 x 1/8-inch catalyst pellets. Three other lengths can be installed as folded reactors (11). They propose to go to a 4-inch ID reactor next followed by a 16-inch ID. Flow distribution and channeling could be a problem in coal dissolution.

They have evaluated some 85 catalysts and have concluded that it is easier to desulfurize coal than oil. Once dissolved the individual molecules are smaller and more reactive. In other studies, they found about a 20% decay in activity after 42 days of operation. The catalyst was still good for desulfurization but not as effective in liquefaction. Catalyst regeneration work is important. If the approximately $1.00/lb catalyst is replaced every six months, the cost is high. Some reactivation is possible. They have demonstrated an increase from 80% to 90% activity by burn off.
Results and conditions of two 30-day runs in the 5 lb/hr unit are given in Tables X and XI. The yield on a Middle Kittanning (Ohio) bituminous coal was 3 barrels product oil per ton of coal (MAF). The oil contained 0.31% sulfur and flowed at ambient temperature. Hydrogen consumption was 3000 scf/bbl of product or 9000 scf/ton of coal (MAF). A Kentucky coal containing 4.6% sulfur was converted to a 0.19% sulfur oil with a yield of 3 bbl/ton of coal (MAF). A 0.5% sulfur Wyoming coal (8,000 Btu/lb) was converted into a 0.2% sulfur oil (15,000 Btu/lb) with a yield of 2.5 bbl/ton of coal (MAF). Hydrogen consumption was 6000 scf/bbl. Product yields do not reflect the fact that hydrogen must be produced from coal or a portion of the product.

Prepared by Dale E. Briggs
### TABLE X

Hydrodesulfurization of Kentucky Coal (39)

**Experimental Conditions:**
- Reactor, catalyst, temperature and liquid feed throughput as in Figure 11.
- Slurry feed: 45 coal + 55 recycle oil
- Hydrogen recycle rate: 125 scfh
- Pressure: 4,000 psig

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfur in feed coal, wt. pct.</td>
<td>4.6</td>
</tr>
<tr>
<td>Sulfur in recycle oil (product oil), wt pct</td>
<td>0.19</td>
</tr>
<tr>
<td>Yield: bbl oil/ton coal maf</td>
<td>3.0</td>
</tr>
<tr>
<td>Hydrogen consumption, scf/bbl</td>
<td>3000.0</td>
</tr>
</tbody>
</table>

**Solvent Analysis of Product Oil, Wt Pct**

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<table>
<thead>
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</thead>
<tbody>
<tr>
<td>Oil (pentane-soluble)</td>
<td>79.5</td>
</tr>
<tr>
<td>Asphaltene</td>
<td>17.4</td>
</tr>
<tr>
<td>Organic benzene insolubles</td>
<td>2.1</td>
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<tr>
<td>Ash</td>
<td>1.0</td>
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</tbody>
</table>

**Elemental Analysis of Product Oil (Ash-Free), Wt Pct**

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon</td>
<td>89.9</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>9.2</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>0.6</td>
</tr>
<tr>
<td>Sulfur</td>
<td>0.19</td>
</tr>
<tr>
<td>Viscosity of product oil, SSF at 180°F</td>
<td>21-31</td>
</tr>
<tr>
<td>Calorific value of product oil, Btu/lb.</td>
<td>17,700</td>
</tr>
</tbody>
</table>
### TABLE XI

Results of Hydrodesulfurization of Middle Kittanning #6 Seam Coal in Recycle Oil (39)

**Experimental Conditions**
- Reactor: 5/16-inch is × 68 ft long
- Catalyst: silica-promoted cobalt molybdate
- Temperature: 450°C
- Pressure: 2,000 psi
- Liquid feed: 30 coal + 70 recycle oil
- Liquid feed throughput: 140 lb/hr/ft³ reactor volume
- Gas recycle rate: 500 scfh

### A. Gross Results

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
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<tbody>
<tr>
<td>Sulfur in feed coal, st pct</td>
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</tr>
<tr>
<td>Sulfur in centrifuged oil, wt pct</td>
<td>0.31</td>
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<tr>
<td>Yield, bbl oil/ton coal maf</td>
<td>3.0</td>
</tr>
<tr>
<td>Hydrogen consumption, scf/bbl</td>
<td>3000.0</td>
</tr>
</tbody>
</table>

### B. Analyses of Centrifuged Product Oil

#### Solvent Analysis, Wt Pct

<table>
<thead>
<tr>
<th>Component</th>
<th>Value</th>
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<tbody>
<tr>
<td>Organic benzene insolubles</td>
<td>11.6</td>
</tr>
<tr>
<td>Ash</td>
<td>1.3</td>
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<tr>
<td>Asphaltene</td>
<td>24.4</td>
</tr>
<tr>
<td>Oils</td>
<td>62.7</td>
</tr>
</tbody>
</table>

#### Elemental Analysis (Ash-Free Basis), Wt Pct

<table>
<thead>
<tr>
<th>Element</th>
<th>Value</th>
</tr>
</thead>
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<td>Carbon</td>
<td>89.6</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>7.6</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>0.9</td>
</tr>
<tr>
<td>Sulfur</td>
<td>0.31</td>
</tr>
<tr>
<td>Oxygen (by difference)</td>
<td>1.6</td>
</tr>
</tbody>
</table>

Range of specific gravity, 60°/60°F: 1.126

Range of viscosity, SSF at 180°F: 75-204

Calorific value, Btu/lb: 16,840

### C. Analyses of Residue (Centrifuge Cake)

#### Solvent Analysis, Wt Pct

<table>
<thead>
<tr>
<th>Component</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Organic benzene insolubles</td>
<td>33.1</td>
</tr>
<tr>
<td>Ash</td>
<td>27.7</td>
</tr>
<tr>
<td>Asphaltene</td>
<td>9.0</td>
</tr>
<tr>
<td>Oil</td>
<td>30.2</td>
</tr>
<tr>
<td>Sulfur content, wt pct</td>
<td>2.10</td>
</tr>
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</table>
This process is the result of development started by Consolidation Coal Company and Standard Oil Company of Ohio in the late 1950's. The original work was done on a bench scale unit at Library, Pennsylvania. Later a 75 B/D pilot unit was constructed at Cresap, West Virginia for the Office of Coal Research (5,19,29).

Pittsburgh seam coal is slurried with a donor solvent and heated to about 750°F at about 400 psia to dissolve around 80% of the coal (MAF) as shown in Figure 12. This is strictly solubilization with no gaseous hydrogen added to the reactor. The resulting slurry was passed through hydroclones or other solids removal units to concentrate the solids in the underflow. The overflow extract is hydrogenated over a CoMo catalyst at 800-850°F and 3000-4200 psia. The hydrogenation is complicated by the fact that the hydroclones are relatively inefficient and the solids remaining in the extract tend to plug fixed beds of catalyst. The solids are particularly troublesome because much of them are in the micron-size range, so some even pass through filters. The organo-metallic compounds, such as those of titanium, are nominally soluble in the extract and tend to accumulate on and deactivate the catalyst. Continuous hydrogenation of extract to a 0.2% sulfur product in ebulliated bed type reactors was accomplished in a 0.1 ton/day reactor by Consol and in a 3 tons/day H-oil reactor by HRI. Because of many mechanical problems,
Figure 12. Consolidation Coal Co. CONSOL Synthetic Fuel (SCF) Process
comparable data were not obtained in the limited operating period on the 13 tons/day extract hydrogenation plant at Cresap. After hydrogenation, a fraction of the product is taken out as the recycle solvent and fed to the slurry section. This hydrogenation enhances the hydrogen donor characteristics of the solvent.

To minimize the effect of solids in extract hydrogenation, rotary precoat filtration with solvent wash was also tested. The difficulties encountered in filtration were essentially the same as for the SRC process. Pressurized operation at high temperature is difficult mechanically. Unless the filter cake is to be coked to recover solvent, large amounts of gas may have to be compressed and recycled to dry the filter cake and to recover the solvent. Filter cake could be gasified with supplemental coal to produce hydrogen. Again, solids removal may present significant problems in commercial operations.

The operation of the pilot plant encountered many difficulties and program extensions were required to solve them. Considerable credit is due to plant staff for overcoming these problems. Much experience was gained in understanding and solving the severe mechanical problems. The identification of critical operating problems and a partial solution to many made the project worthwhile.

The National Academy of Engineering reviewed the process for OCR (41) and concluded that the process was technically
feasible. Foster Wheeler concluded likewise in an engineering evaluation of the process (18) and included some economics which were updated in the Consol final report (42). A summary analysis from the report is given in Table XII. Hydrogen production was to be by a Bi-Gas gasification step. As seen from the Table XII, hydrogen production constitutes about 40% of the capital investment.

The Office of Coal Research plans to reactivate the Cresap plant to test process modifications and to evaluate component process equipment used in all of the coal dissolution processes (43).

Prepared by Dale E. Briggs


<table>
<thead>
<tr>
<th><strong>A. Thermal Performance</strong></th>
<th><strong>Extraction</strong></th>
<th><strong>Hydrogenation</strong></th>
<th><strong>H₂ Mgf</strong></th>
<th><strong>Combined</strong></th>
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<tbody>
<tr>
<td>(10⁶ Btu/hr)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Input</strong></td>
<td></td>
<td></td>
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<td></td>
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<tr>
<td>Coal -Pittsburg Seam</td>
<td>21,000</td>
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<td>-</td>
<td>21,100</td>
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<td>H₂</td>
<td>258 (1)</td>
<td>4,162</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Hydro Feed</td>
<td>-</td>
<td>14,370</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Char</td>
<td>-</td>
<td></td>
<td>4,900</td>
<td>-</td>
</tr>
<tr>
<td>Process Input</td>
<td>21,358</td>
<td>18,532</td>
<td>4,900</td>
<td>21,100</td>
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<tr>
<td><strong>Fuel</strong></td>
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<td></td>
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<tr>
<td>Process Furnaces</td>
<td>1,652</td>
<td>395</td>
<td>-</td>
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<td>Steam Generation</td>
<td>-436</td>
<td>-133</td>
<td>2,338</td>
<td>1,769</td>
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<tr>
<td>Power Generation</td>
<td>196</td>
<td>89</td>
<td>326</td>
<td>611</td>
</tr>
<tr>
<td>Total Fuel</td>
<td>1,412</td>
<td>351</td>
<td>2,664</td>
<td>4,427</td>
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<tr>
<td><strong>TOTAL INPUT</strong></td>
<td>22,770</td>
<td>18,883</td>
<td>7,564</td>
<td>25,527</td>
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<tr>
<td><strong>Output</strong></td>
<td></td>
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<tr>
<td>Low Btu Gas</td>
<td>636</td>
<td></td>
<td>-</td>
<td>636</td>
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<tr>
<td>Hi Btu Gas</td>
<td>356</td>
<td>2,736</td>
<td>71</td>
<td>3,163</td>
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<tr>
<td>Light Oil &amp; Dist.</td>
<td>160</td>
<td>2,466</td>
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<td>2,626</td>
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<td>Heavy Oil</td>
<td>270</td>
<td>8,650</td>
<td>-</td>
<td>8,920</td>
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<td>-</td>
<td>3,500</td>
<td>-</td>
<td>3,500</td>
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<tr>
<td>Fuel Output</td>
<td>1,422</td>
<td>17,352</td>
<td>71</td>
<td>18,845</td>
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<td><strong>Intermediates</strong></td>
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<tr>
<td>H₂</td>
<td>-</td>
<td>258 (1)</td>
<td>4,162</td>
<td>-</td>
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<tr>
<td>Hydro Feed</td>
<td>14,370</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Char</td>
<td>4,900</td>
<td></td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td><strong>TOTAL OUTPUT</strong></td>
<td>20,692</td>
<td>17,610</td>
<td>4,233</td>
<td>18,845</td>
</tr>
<tr>
<td>Output/Input, %</td>
<td>91.0</td>
<td>95.0</td>
<td>56.0</td>
<td>73.9</td>
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<td><strong>Processing Costs, MM $/yr</strong></td>
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<td>Operating Costs (ex fuels)</td>
<td>6.50</td>
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<td>Capital Charges</td>
<td>9.25</td>
<td>10.05</td>
<td>13.18</td>
<td>32.48</td>
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</table>

1) Via donor solvent transfer
Bibliography


INSITU GASIFICATION OF COAL

by

Donald L. Katz

The concept of converting coal into fuel gases in the ground or "insitu" is an old idea. Elder (1) gives a historical treatment of underground gasification of coal including the Russian experiences. They have conducted by far the most field operations for using the gas beneficially.

Currently a test is in progress at Hanna Wyoming by a Bureau of Mines field test. Figure 1 shows the cross section of the Hanna underground gasification site and Table 1, the heating value of the composite gas produced from the wells. Here the coal seam is 30 feet thick and lies at a distance of 370-430 feet from the surface. Air has been injected into a well and the coal ignited by firing with a propane fuel initially. The combustion of the coal by the oxygen yields gases at wells to which it is driven from the injection well and such gases are sampled and the composition measured. Example composition of these gases are given on Table 1 reproduced from (3). It should be noted that leakage of injected air took place up to 90% at one point because of the lack of seal around the various wells drilled into the coal. In September and October while injecting 1-1.2 million cubic feet of air per day gas production was maintained from 1.7 - 2.4 million cubic feet per day.

297
Figure 1. Section of Insitu Gasification Experiment, Hanna, Wyoming (3)
TABLE I

Typical Gas Analyses of Gas from Hanna

In situ Gasification Experiment (3)

<table>
<thead>
<tr>
<th>Date</th>
<th>June 4</th>
<th>June 8</th>
<th>June 17</th>
<th>July 5</th>
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<tbody>
<tr>
<td>H₂</td>
<td>8.8</td>
<td>8.2</td>
<td>7.8</td>
<td>12.6</td>
</tr>
<tr>
<td>O₂</td>
<td>9.7</td>
<td>1.0</td>
<td>.9</td>
<td>1.0</td>
</tr>
<tr>
<td>N₂</td>
<td>68.4</td>
<td>50.7</td>
<td>53.9</td>
<td>54.8</td>
</tr>
<tr>
<td>CH₄</td>
<td>4.3</td>
<td>16.5</td>
<td>11.9</td>
<td>6.6</td>
</tr>
<tr>
<td>CO</td>
<td>3.1</td>
<td>4.3</td>
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<td>.2</td>
</tr>
<tr>
<td>C₂</td>
<td>.5</td>
<td>1.6</td>
<td>1.4</td>
<td>1.0</td>
</tr>
<tr>
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<td>22.2</td>
<td>21.8</td>
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<tr>
<td>Other</td>
<td>.1</td>
<td>.5</td>
<td>.7</td>
<td>2.0</td>
</tr>
</tbody>
</table>

| Rate scf/min | 163 | 174 | 255 | 152 |
| Btu/scf     | 94  | 249 | 189 | 150 |
A.D. Little delivered a report in December of 1971 (2) which gave an appraisal of underground coal gasification and noted that the Russian experiments and others world wide had been discontinued as of 1965. Generally the problems noted are:

1) the ability of the fluids to be conducted through the coal bed if air or oxygen are injected in one well and the fuel gases are expected to go to another well. 2) When the coal has been burned and only the ash remains it is expected that the roof will collapse and subsidence of the earth is likely to take place. 3) Beds lack uniformity which permits injecting the air or oxygen and having a smooth burning front take place between injection well and producing well. Accordingly air may bypass other fuel gas and mix as they come out, thereby loosing the value of the injected air and diluting the product. 4) The main economic cost is the compression of the air with minor costs for drilling of wells for shallow deposits.

The non-uniformity of coal beds is worthy of evaluation. Tests have indicated that fractures represent the insitu permeability, while small units such as a cubic inch are of very low permeability. The distribution of the conduit system--major channels and micro interstices--would be helpful in designing an insitu combustion system.

A U.S. patent by Vaughn is cited on Table II in which explosives are used to crumble the coal before processing (8). Water is injected at the appropriate stage to react with hot
TABLE II U.S. Patent on Insitu Gasification of Coal Utilizing Non-Hypersensitive Explosives

No. 3,734,180 (May 22, 1973) V.W. Rhoades, Cities Service Oil Company

202 Overburden
210 Injection Well
213 Compressor
214 Fuel Gas and/or oxidizer
220 Production Well
228 Crumbled Coal
226 Combustion Front
227 Charred Residual
224 Fuel Gas Product
225 Coal Tar Products

[57] ABSTRACT

Two or more wells are drilled into a coal seam. The wells are completed so as to isolate all other strata from the coal seam and a radially extended horizontal fracture is directed by introduction of a non-hypersensitive explosive under hydraulic fracturing conditions so as to connect the wells communally. The explosive is ignited so that a horizontally and vertically directed fracture network is formed within the coal system. A combustion front is ignited and propagated through the fractured network to produce combustible gases and coal tar liquids.

3 Claims, 2 Drawing Figures
char. It is believed that there may be many patents granted or pending on gasification of coal and oil shale like Reference (8,9).

**LONG RANGE**

It would seem that most likely use of insitu combustion would be for coal seams just below the 300 foot depth level at which surface or strip mining may take place and for seams which are thin to the extent that underground mining would not be advantageous. The cost of the wells would be low because of the shallow depths and subsidence would be nominal because relatively thin seams are involved. The method has the advantage of using oil field technology and avoids disturbance of the environment unduly.

**INSITU COMBUSTION AS OIL RECOVERY MECHANISM**

Considerable effort has been made over the years to use insitu combustion or thermal recovery procedures for crude oil (4,5,6). Here field experiments have been conducted in which air or oxygen are injected and a burning front proceeds from the injection well to the producing wells. Several of these experiments have been successful technically but few, if any, at the prices of the crude oil prior to 1973 have been economic successes. Again the chief cost is the compression of the large volumes of air required to carry out the combustion process.

Efforts have been made at Shale Oil Recovery in a similar manner but their success was limited because of the relatively low permeability of the shale. In both insitu combustion of
shale or coal, the concept of fracturing in a horizontal layer if possible, placing an explosive in the fracture gap, and exploding the wafer of explosive seems worth pursuing. The fragmented layer would provide a burning zone and give a conduit for oxidant injection and product gas production. Although there are indications such procedures may be developed in the years ahead no demonstration has been published or announced (7).

CONCLUSION

It may be concluded that the above mentioned problems and lack of success after much effort would indicate that at least a decade of effort is required before any viable method can be developed which would assist the utility industry. Small scale efforts to study such processes would seem reasonable. For example studies of permeability of coal or lignite relative to their ability to conduct gases through the beds would seem to be important measurements. Even threshold pressure measurements on coals should be made to find the gas pressure that is required to displace the water from the pores. Permeabilities, porosities and other reservoir engineering type factors measured through core holes (insitu) or on cores taken from coal beds are suggested.

The ability of the caprock above the coal and of the under-lying rock to contain the gases especially after they become heated is important. Also the influence of subsidence on gas containment should be studied.

Work with explosives insitu for "crumbling" coal layers at 300-500 feet deep would be helpful with coring afterward
to test the outcome of the explosive is a first step in evaluating the explosive, with combustion pilot studies for "exploded" layers only after some success has been found by coring.

Bibliography


PART III

RELATED TOPICS IN COAL UTILIZATION
A group of activities carried out by the project team cannot be characterized as coal utilization processes. These topics seemed to be of sufficient importance to be discussed separately in Part III as they relate to the general subject of coal use. The topics include combined cycle systems, retrofit capabilities, thermodynamics and coal slurry pipelines.
THE COMBINED CYCLE IN RELATION TO COAL AS A FUEL

by

John E. Powers
Edward R. Lady

In the burning of clean fossil fuels--natural gas, natural gas liquids, and liquids obtained from crude oil--to generate electric power, the advantages of the combined gas-steam turbine cycle have provided great incentive for its development. The net operating capability of combined cycle plants under construction or on order is 6047 MW according to the 54th Semi-Annual Electric Power Survey of the Edison Electric Institute. Over half this capacity is for Southern California Edison Company. As it appears that it may be necessary to replace oil and gas with coal not only in new plants but even by retrofitting old ones, it is essential to attempt to evaluate the effects that substitution of coal as fuel will have on the future development of the combined cycle.

WHAT AND WHY OF THE COMBINED CYCLE

As supercritical steam cycle generating units were installed and found to have problems with somewhat lower availability factors, it became evident that the energy efficiency of steam cycles had reached a plateau of about 36-38%. At the same time, modified aircraft gas turbines were being utilized for peak loads. Whereas the unit capital costs for gas turbines were low,
the maintenance costs were high and fuel costs even higher because of the required use of premium fuels and the poor thermal efficiency, reflecting a large energy loss in the hot exhaust gases.

In an attempt to take advantage of the potential for the higher thermal efficiency resulting from the relatively high inlet temperatures to gas turbines and adapt them to intermediate and/or base load situations, the hot exhaust gases were utilized to generate steam in a more or less conventional steam cycle. This combination of a gas turbine and a steam cycle, the "combined cycle" yielded higher thermal efficiencies with a reduction in capital cost per kilowatt of generating capacity.

RECENT DEVELOPMENTS

Such a development necessitated more than adaptation of existing aircraft engines because gas turbines for intermediate or base load use must meet basic requirements different from those in aircraft or for peak shaving applications. Whereas it is reasonable and perhaps economical to require complete inspections and overhaul of gas turbines used in commercial aircraft every 2000 hours or so, and current practice with stationary gas turbines used in electric power generation necessitates reblading after about 10,000 hours of operation, it is not uncommon for steam turbines to operate for 25,000-50,000 hours between major overhauls—which does not include reblading. Another difference is that weight is of little consequence in connection
with power production. Perhaps the most important design difference is that gas turbines for electric utility power production are designed for use at one fixed set of conditions over relatively long periods of time at high efficiencies whereas aircraft engines, of necessity, must operate at reasonable efficiencies over extreme ranges of temperature and pressure at the inlet.

GAS TURBINES REQUIRE A "CLEAN" FUEL

Whether one is dealing with aircraft service or electrical power generation, gas turbines require a clean fuel. Principally, the amount and size of solid particles and liquid droplets which may enter the turbine are both extremely small—and no one knows just how low these must be. Secondly, many of the elements whose chemical compounds are harmful to turbine blades are present in coal. Attempts to burn pulverized coal directly in turbine combustors were unsuccessful. Further research in this direction should probably be directed toward pressurized, fluidized bed combustion. Ideally, if coal is to be used in connection with gas turbines in a combined cycle, the coal must first be converted to a clean gaseous or liquid fuel.

WHAT ALTERNATIVE METHODS WILL YIELD A CLEAN FUEL FROM COAL?

The major thrust of the report to EPRI, of which this manuscript is a part, is directed toward answering the above question. In brief, clean liquid fuel can be produced either by pyrolysis,
solvent refining, or liquefaction (catalytic or noncatalytic). A variety of gasification schemes have been proposed. Each of these complete systems operates at a thermal efficiency of less than 100%--and very considerably less. Therefore, attention must be directed toward the thermal efficiency of the process required to obtain clean fuel from coal as well as the efficiency of the combined cycle in planning future developments.

WHAT ARE THE CURRENT AND PROJECTED THERMAL EFFICIENCIES OF COMBINED CYCLE PLANTS?

Hottel and Howard in New Energy Technology present a graph of Turbine Inlet Gas Temperature plotted against the Year. This graph is reproduced as Figure 1 with permission. Several additions have been made as explained below:

According to a reputable source (1), power generating plants in construction since 1971 and currently operating, have established gas inlet temperatures of 1850°F as indicated by a cross, "+". This appears right on the curve drawn in 1970 or 1971.

Present projections call for a combined cycle with a gas inlet temperature of 2050°F utilizing air cooled blades to be in operation in early 1975. This is shown as a circled cross on Figure 1. Future improvements in allowable operating temperatures involving gas cooled turbine blades are projected at the rate of 65°F per year as indicated by a dashed line. An increase in gas inlet temperature will certainly result in some increase in thermal efficiency. However, the picture is somewhat clouded by the inefficiencies associated with air cooling of the blades.
As with most problems associated with improvements suggested by thermodynamic considerations, the desirability of increasing the temperature of operations is hindered by the necessity of finding materials to withstand continued exposure to such conditions. There is some hope that silicon nitride turbine blades might soon prove practical and Westinghouse plans to have a large-scale gas turbine without cooling of the silicon nitride blades in operation by 1978. This point is plotted as an "X" on Figure 1. Future increases in operating temperatures are projected (rather arbitrarily) as a center line drawn with a slope of 65°F per year. General Electric has a similar development underway as reported in the business section of the New York Times recently.

From a practical point of view, gas inlet temperature is only important with respect to the thermal efficiency of the combined cycle. Several factors in addition to gas inlet temperatures serve to establish the thermal efficiency of the combined cycle so that it is difficult to establish thermal efficiencies of cycles involving gas turbines under development. However, Table I contains some values believed to be reasonable for simple combined cycles. The combined efficiency is a function of the steam conditions as well as the gas turbine inlet temperature. Brown Boveri is constructing a combined cycle, dual steam pressure plant at Geertruidenberg of PNEM for which the efficiency is projected to be 44%, with a turbine inlet of 1742°F (2). This is a somewhat complex cycle which includes much heat exchange surface.
TABLE I

<table>
<thead>
<tr>
<th>Inlet Gas Temperature, °F</th>
<th>Year</th>
<th>Combined Cycle with Gas Turbine Blades Cooled with Air %</th>
<th>Uncooled Si-N, %</th>
<th>Required of Process to Provide Clean Fuel for Turbine from Coal, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>1850</td>
<td>1973</td>
<td>41</td>
<td>--</td>
<td>90</td>
</tr>
<tr>
<td>2050</td>
<td>1975</td>
<td>43</td>
<td>--</td>
<td>86</td>
</tr>
<tr>
<td>2250</td>
<td>1978</td>
<td>45</td>
<td>--</td>
<td>82</td>
</tr>
<tr>
<td>2500</td>
<td>1978</td>
<td>--</td>
<td>49</td>
<td>75</td>
</tr>
<tr>
<td>2550</td>
<td>1981</td>
<td>47</td>
<td>--</td>
<td>78</td>
</tr>
</tbody>
</table>

*Based on an overall thermal efficiency of 37% for either a conventional steam cycle burning a clean coal or a steam cycle with fluidized bed combustion.

Table I gives the best estimates that could be made with the available data and time. It would certainly seem reasonable for EPRI to attempt to obtain better estimates although it should be recognized that it will be impossible to obtain exact numbers and, perhaps, difficult to obtain numbers that are significantly better than those presented above. To those who argue for even higher gas inlet temperatures based on data from military or commercial jet aircraft engines, we point out again that gas turbines for intermediate or base load use are really quite different from aircraft engines.

HOW DOES THE THERMAL EFFICIENCY OF PROCESSES TO PRODUCE CLEAN GASEOUS OR LIQUID FUELS FROM COAL TIE IN WITH THE COMBINED CYCLE?

The thermal efficiency of processes to produce clean gaseous and liquid fuels from coal have been estimated to be in the range from 50-80%. Firm figures based on actual continued plant
operation are simply not available.* However, it is very simple to calculate the thermal efficiencies that would be required for such processes such that the overall efficiency when coupled with present and proposed combined cycles will at least equal that available from the standard steam cycle utilizing limestone in fluidized bed combustion for sulfur clean up. The thermal efficiency for the latter is taken to be 37%. Based on this figure, the required efficiency of processes to provide clean gaseous or liquid fuels can be easily calculated according to:

\[
\text{Required Efficiency of Coal Conversion Process} = \frac{37\%}{\text{Thermal Efficiency of Combined Cycle}} \quad (1)
\]

The values of required thermal efficiency of processes to provide clean fuels from coal calculated from Equation (1) are listed in the right-hand column of Table I.

Some tentative conclusions can be drawn based on the figures presented in Table I:

1. The thermal efficiency of state-of-the-art gasifiers (Lurgi and Koppers-Totzek) is about 70% \((3)\) when proper account is given to steam requirements for auxiliary processes, etc. Therefore, in accordance with Equation (1) and as summarized in Table I, unless gas turbine inlet temperatures approach 3000°F, current gasification technology in conjunction with the combined

*There is good indication that operating gasifiers have thermal efficiencies with percentage efficiencies in the high 60's \((3)\).
cycle is not competitive on the basis of thermal efficiency with fluidized bed combustion and a steam cycle.

(2) If a gasification process with a thermal efficiency of 82% can be developed by 1978 at which time a gas turbine with cooled blades might be available for operation with a gas inlet temperature of 2250°F, the thermal efficiency of the coupled gasification process and combined cycle will just equal the thermal efficiency of atmospheric pressure, fluidized-bed boiler, steam cycle generating plants presently planned or under construction, i.e., 37%.

(3) If uncooled silicon nitride gas turbines can be made operational by 1978 with an inlet temperature of 2500°F, the thermal efficiency required of any gasification process to be used to provide clean fuel for a combined cycle must exceed 75% to be competitive based on thermal efficiency alone.

(4) The cost of coal processing plants are such that there is little doubt that the capital cost of such plants coupled with a combined cycle will be in excess of that of a steam cycle incorporating either a conventional boiler burning clean coal or a fluidized bed combustor. Therefore, it seems obvious that the anticipated thermal efficiency of coal processing plants must exceed 75% to justify extensive expenditures in development of coal processing plants on the part of the utility industry.
WHAT RESEARCH SHOULD BE DONE?

Interpretation of Table I together with a knowledge and/or understanding of some of the limitations of coal gasification and/or liquefaction processes seems to give strong indication that combined cycle operation based on the indirect use of coal as a fuel will require significant technological developments. The present severe limitation that the bed temperature in atmospheric fluidized bed boilers should not exceed 1600°F if sulfur is to be removed in the bed, dictates that the thermal efficiency of an atmospheric fluidized bed coupled with a combined cycle will probably not be increased above 40%. An increase from 37% to 40% hardly justifies substantial expenditures for research and development. On the other hand, removal of sulfur in pressurized fluidized bed boilers is apparently effective at temperatures up to 1750°F as established experimentally by Hoy in England. Operation at such elevated temperatures produces other problems. Some money should be spent on attempts to gain additional knowledge relative to the temperature limitations and to increase the temperature of operation of fluidized beds of limestone especially in connection with operation at pressure.

If gasification processes to produce a clean, low Btu gaseous fuel are to be developed, the problem of particulate
and $\text{H}_2\text{S}$ removal at high temperature must be studied. If the hot gas from the gasifier is usable in a gas turbine without quenching, substantial thermal economy can be achieved, thereby partially off-setting the energy losses in the gasifier. At present there seems to be no practical way to utilize the hot gasifier product gas and therefore this would appear to be a fruitful area for research—if coal gasification proves to be of interest to the utility industry.
Bibliography


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ECONOMIC EVALUATION
by
M. Rasin Tek

ECONOMIC EVALUATIONS OF COAL CONVERSION PROCESSES
TO PROVIDE CLEAN FUELS FROM COAL

In evaluating the several routes and processes leading from coal to clean fuels for electric power generation it becomes essential that a common basis be used in order to make relevant, fair and valid comparisons. The bases used during this project have resulted from modifications, wherever warranted, of the basis adopted in the supply - Technical Advisory Task Force - Synthetic Gas-Coal Report of Federal Power Commission.

Investments required for all of the processes must be based on a common point in time (mid 1973) then escalated through the years 1975, 1980, 1985, 1990. It is reasonable to assume escalation costs of 4% per year for 1973-75, 3.5% per year for 1975-80 and 3% per year thereafter.

Plant Capacities

The plant capacities used as basis during this study are those with heating value of product equivalent to the output of standardized 250 MMCF/D SNG plants with heat output of 2.5 X 10⁶ Btu/day.

Coal Costs

The basis used for coal costs were 30¢/MMBtu for the bituminous coal and 15¢/MMBtu for the subbituminous coal.
By-product Credits

Reasonable by-product credits must be allowed for sulfur ammonia, oil and char.

OVERALL ECONOMIC COMPARISONS BETWEEN SOLID LIQUID AND GASEOUS FUELS FROM COAL

The Table I gives an overview of the major processes for which economic analyses were made during this study and some figures obtained from the literature. The two major quantities listed for solid liquid and gaseous fuels from coal are total capital required in MM$ or $/BID, and the average selling price of fuel between now and 1990 as computed by AGA-FPC recommended accounting techniques. For the purposes of EPRI these two overall economic criteria may readily be translated in terms of $/KW-h and cents/KW by making the necessary efficiency assumption to translate the Btu permitted by the fuel to KW generated at the power plant.

The following section is a tabular presentation of the items involved in determination of total plant investment required, of annual operating cost, and finally the selling price of the fuel by AGA-FPC Public Utility and discounted cash flow accounting procedure.

Quick estimating methods and bases for auxiliary onsite investments required such as oxygen plant, Claus sulfur recovery, particulate recovery, etc. have been also included at the end. These materials have been taken from the Final Report of Supply Technical Advisory Task Force - Synthetic Gas - Coal, Federal Power Commission. A detailed example of complete economic analysis is given at the end.
**TABLE I**

OVERALL ECONOMIC COMPARISONS BETWEEN SOLID LIQUID AND GASEOUS FUEL FROM COAL

<table>
<thead>
<tr>
<th>SOLIDS</th>
<th>LIQUIDS</th>
<th>GASES</th>
</tr>
</thead>
<tbody>
<tr>
<td>TRW Liquefaction</td>
<td>Liquefaction</td>
<td>Overall</td>
</tr>
<tr>
<td>146 (M.R. Tek)</td>
<td>225***</td>
<td>232.1</td>
</tr>
<tr>
<td>108 (Dow Chemical)</td>
<td>(325 at 1980)***</td>
<td>181.1</td>
</tr>
<tr>
<td>7500**</td>
<td>75-115</td>
<td>270</td>
</tr>
<tr>
<td>11000**</td>
<td>(115 @ 1990)</td>
<td>230</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Western Bituminous Coal</th>
<th>Low Btu Mollen Salt</th>
<th>HYGAS</th>
<th>High Btu LURGI</th>
<th>SNG Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>70-90</td>
<td>250</td>
<td>145</td>
<td>97-116</td>
<td>(1)</td>
</tr>
<tr>
<td>1.15 $/Btu</td>
<td>110</td>
<td>125</td>
<td>(80 @ 1990)</td>
<td>11</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Western Coal</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>60-80</td>
<td>(321)</td>
</tr>
</tbody>
</table>

Basis Coal Costs 30¢/MM Btu bituminous coals 15¢/MM Btu subbituminous Western Coals


CALCULATION OF TOTAL CAPITAL REQUIREMENT PROCESS

1. All onsite plant sections
   Coal Storage
   Coal Preparation
   Coal Gasification
   Waste Heat Recovery
   Gas Purification (particulate removal scrubbing)
   Fresh Water Treatins
   Compression Facilities
   Cooling Towers
   Power Generation and Distribution
   Steam Generation
   Waste Disposal Facilities
   Office
   Shop
   Control System
   Oxygen Plant
   Hydrogen Plant
   Other on site plant sections

   SUBTOTAL

2. Contractors overhead and profit
   Engineering and Design costs

   PLANT INVESTMENT SUBTOTAL
3. Project Contingency

(In the absence of detail below use 15% of subtotal plant Investment)

Allowances for Results of a Detailed Design

a. New railroad spur to plant site
b. New roadways to plant site
c. New power transmission to plant site
d. New pipeline for water to plant site
e. New slurry pipeline if applicable
f. New unitrain if applicable
g. Contingency site problems:
   Prilling
   Grading
h. Additional clean up facilities foe efficient water gas etc.
i. Additional equipment for start up shut down stand by facilities
j. Additional safety equipment
k. Additional equipment for process or mechanical emergencies
l. Standby equipment for reliability
m. Additional spare equipment
n. Contingency for missed cost estimates
o. Premium labor for construction
p. Contingency for low labor productivity and strikes

TOTAL PROJECT CONTINGENCY
4. Process Development Contingency

(In the absence of detail below use 7% of Plant Investment Subtotal)

Detailed Allowances for Process Development Contingency

a. Lower gasification rates

b. Lower product yields from reactors

c. Lower thermal efficiency

d. Lower distillation contacting performance

e. Need for more sophisticated reactor, gasifier intervals (plates, grids, distributions, downcomers, cyclones, nozzles, turbulence promoters, etc...)

f. Cost escalation in feed or ash removal systems

g. Need for additional coal treatment

h. Unexpected fine removal problems

i. Unexpectedly high sulfur

j. Unexpected heat transfer, drying problems

TOTAL PROCESS DEV. CONTINGENCY

Sum of the items 1 through 4 above gives

5. Total Plant Investment

6. Interest during construction [Interest Rate X Total Plant Investment X 1.875 construction period]

7. Start-up costs [20% of total gross operating cost]

8. Working Capital

a. Coal Inventory (for 60 days at designate)

b. Materials and supplies 019% of Total Plant Investment

c. Net receivables @ 1/24 of annual gas revenue @ $1,00/MM Btu
BASIS FOR CALCULATING GROSS AND NET OPERATING COST

Base Load Plants @ 90% Load Factor

1. Coal (30¢/MM Btu high heating value, deep mined Bituminous coals and 15¢/MM Btu surface mined subbituminous coals)

2. Other Raw Materials

3. Catalysts and Chemicals

4. Purchased Utilities
   a. Electric Power @ 0.9¢/KWH
   b. Raw Water @ 30¢/M Gal (delivered)
   c. Cooling Water

5. Labor
   a. Process Labor [(Men/s shift) X 8304 Man-hours/year
      X 5.20 $/Man hour] (1)
   b. Maintenance Labor (1.5%/year of total Plant Investment)
   c. Supervision (15% of operating and maintenance labor)
   d. Administration & General Overhead
      60% of total items a,b,c, above.

6. Supplies
   Operating Supplies (30% of process operating labor)
   Maintenance Supplies (1.5%/year of Total Plant Investment)

7. Local Taxes and Insurance
   (2.7%/year of Total Plant Investment)

   TOTAL GROSS OPERATING COST/YEAR

8. By Product Credits
   Sulfur (10.00 $/LT)
   Ammonia (25.0 $/ST)
   Light Oil (where applicable) @ 15¢/gas
   Heavy Oil (Tars 30¢/MM Btu)
   Char (90% of Coal Cost)

(1) Based on 1973 figures
TOTAL COSTS

TOTAL NET OPERATING COST / YEAR

PROCEDURE FOR CALCULATING THE COST OF FUEL - UTILITIES FINANCING METHOD

BASIS

1. 20 year project life
2. Depreciation 20 year straight line, 5%/year

ESSENTIAL INPUT PARAMETERS

3. Debt/Equity Ratio for the mix of total Capital Requirement
4. Percent Interest on debt
5. Percent Return on Equity
6. Federal Income Tax Rate

DERIVED PARAMETERS

7. Rate Base = Total Capital Required - less accrued depreciation including 1/2 depreciation for current year

8. Percent Return on Rate Base = Fraction Debt X Percent Interest + Fraction Equity X Percent Return on Equity

CASH FLOW CALCULATION

9. Return on Rate Base X Fraction Return on Rate Base
10. Return on Equity - Fraction Equity X Rate Base X Fraction Return on Equity
11. Federal Income Tax = Return on Equity X Fraction Tax Rate/(1-fv. tax rate)
12. Depreciation = 0.05 X (Total Capital Requirement - Working Capital)
13. Total Cash flow revenue required = Item 9 + Item 11 + Item 12 + Total Net Operating Cost
14. In given year = Total Cash Revenue required (project life)/
20 X Annual Production

**Average Fuel Cost Equation** (UTILITY FINANCING METHOD)

By Utility Financing Method

**Basis** 20 year project life

5%/year straight line depreciation on (Total Capital
Required less working capital)

**Average Cost of Fuel**

\[
\frac{aN + 0.05(C-W) + 0.005 [p + \frac{48}{52} (1-d)r] C + W}{G}
\]

where \( p = (d)i + (1-d)r \)

\( a \) = a dimensionless parameter describing escalation of operating

cost during the project life. If from plant start up through

project completion date there is no escalation use \( a = 1.0 \).

If escalation is to be considered on the operating costs
during the project life it is recommended to use \( a = 1.3726 \)

for 1975 and, \( a = 1.3435 \) for 1980 and beyond.

\( C \) = Total Capital Required, MM$

\( W \) = Working Capital, MM$

\( p \) = Return on rate base %/year

\( a \) = Fraction debt

\( r \) = Return on Equity %/year

\( G \) = Annual production, MMM Btu/year, Trillion Btu/year

\( i \) = interest on debt %/year
PROCEDURE FOR CALCULATING COST OF FUEL

DISCOUNTED CASH FLOW METHOD (PRIVATE INVESTOR FINANCING)

BASIS

1. 25 year project life
2. 16 year sum of the year's digits depreciation on total investment
3. 100% equity capital

ESSENTIAL INPUT PARAMETERS

4. Discounted cash flow return rate
5. Federal Income Tax rate

PRINCIPAL COST ITEMS

6. Total Plant Investment and Working Capital are treated as capital costs at the completion of start up.
7. Return on Investment during construction (TPI X DCF rate X 1.875 years) is treated as capital cost at the completion of start up.

FUEL COST

8. In D.C.F. method a single value for fuel cost is calculated for the desired DCF return over the life of the project.

DISCOUNTED CASH FLOW

To determine the discounted cash flow a table is prepared where the gas revenue, start-up and operating costs, depreciation, taxable income, net income and investment costs are listed for each year of the project. A discount factor is calculated for every year. Depreciation plus net income after F.I.T. investment for each particular year is multiplied by the discount factor for that year to compute the necessary discounted cash flow.
FUEL COST EQUATION DCF METHOD

On the basis of 25 year project life, 16 years "sum-of-the-years" digits depreciation, 100% equity capital, 12% DCF return rate and 48% Federal Income Tax rate, the following equation has been developed for the cost of the fuel:

\[
\text{Fuel Cost @ 12% D.C.F. return} = \frac{aN + 0.2353I \times 0.1275S + 0.2308W}{G}
\]

\(a\) is a dimensionless parameter describing escalation of operating cost during the project life.

If there is no escalation from start-up through project completion date \(a = 1.0\). If escalation is to be considered on the operating costs during the project life, it is recommended (1) that \(a = 1.2651\) for 1975 and \(a = 1.2422\) for 1980 and beyond.

\(N = \) Total Net Operating Cost in First Year, MM$/year

\(I = \) Total Plant Investment, MM$

\(S = \) Start-up Costs, MM$

\(W = \) Working Capital, MM$

\(G = \) Annual Fuel Production, MMMM Btus/year, Trillion Btus/year

The values of I, S, W and N above must be adjusted to reflect the actual costs for the start-up completion date. The synthetic gas coal task force\(^1\) has recommended to calculate the total plant investment, start-up costs, working capital and total net operating cost for a base year (say 1973) then escalate them to the cases of interest. Total plant investment to be escalated to 2 years prior to start-up completion; Start-up costs, working capital and total net operating costs to start-up completion. The rates of escalation selected were 4% during 1971-1975, 3.5% during 1976-1980, and 3% after 1980.

COST ESTIMATING AUXILIARY ONSITE INVESTMENTS

a. **Oxygen Manufacture**

On site oxygen plant investment may be calculated from the following data (1)

1. Maximum capacity per train: 2000 ST/SD (Short Tons/Stream Day)
2. Discharge pressure 0.5 to 5.0 psig
3. Product oxygen compression to be estimated separately

Oxygen Plant Investment MM$ (Mid 1971) = \frac{\text{Capacity, ST/SD}^a}{1500} \times 7.4 \quad (1)

In Eq (1) a is investment capacity slope.

if capacity <1500 ST/SD a = 0.55
if 1500 ST/SD < capacity < 2000 ST/SD a = 0.80

b. **Claus Sulfur Recovery**

The Claus sulfur recovery plant to handle SH₂ and SO₂ streams from gas fortification and SO₂ produced in Wellman-Lord scrubbing step can be quick estimated from Table 1 below.

<table>
<thead>
<tr>
<th>Mole % SH₂+SO₂ in feed gas</th>
<th>Investment MM$, Mid 1971</th>
<th>Investment Capacity Slope &lt;100T/D</th>
<th>Investment Capacity Slope &gt;100T/D</th>
<th>Maxproduct S° per train ST/SD</th>
<th>% Sulfur Recovery</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>1.45</td>
<td>0.6</td>
<td>0.8</td>
<td>100</td>
<td>84</td>
</tr>
<tr>
<td>10</td>
<td>1.20</td>
<td></td>
<td></td>
<td>200</td>
<td>89</td>
</tr>
<tr>
<td>20</td>
<td>.95</td>
<td></td>
<td></td>
<td>350</td>
<td>91</td>
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<td>50</td>
<td>.75</td>
<td></td>
<td></td>
<td>700</td>
<td>93</td>
</tr>
<tr>
<td>100</td>
<td>.65</td>
<td></td>
<td>&gt;1000</td>
<td></td>
<td>95</td>
</tr>
</tbody>
</table>

c. **Estimating Investment Costs for Wellman-Lord SO₂ Recovery**

In order to meet the atmospheric pollution regulations
as of 1971 SO$_2$ recovery is required on the following streams:

1. Claus Tail Gases after incineration

2. CO$_2$/SH$_2$ streams from second stage of 2-stage scrubbing systems if SO$_2$ level after incineration is larger 150 vppm

3. Combustion flue gases where SO$_2$ level is 7500 vppm (corresponding to 0.7% by wt. sulfur in fuel)

If, for the above streams Wellman-Lord process is used for SO$_2$ scrubbing then the required investment for Wellman-Lord process units can be quickly estimated from Table II below.

<table>
<thead>
<tr>
<th>Table II</th>
<th>Investment Required for Wellman's Lord SO$_2$ Scrubbing</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base Capacity</td>
<td>120,000 SCFM Feed Gas</td>
</tr>
<tr>
<td></td>
<td>2,700 lbs/hr Feed SO$_2$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Investment MM$</th>
<th>Investment-Capacity Slope</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas Rate Related</td>
<td>0.85</td>
</tr>
<tr>
<td>Sulfur Rate Related</td>
<td>1.95</td>
</tr>
<tr>
<td>TOTAL</td>
<td>2.80</td>
</tr>
</tbody>
</table>

d. **Investment Costs for Process Water Treatment**

Contaminants in process waters such as dissolved phenols, ammonia, hydrogen cyanide, hydrocarbons, etc. must be to particular process, estimated in quantity so that precise calculations may be made for the cost of their removal. The various processes available for the removal of these contaminants cannot be generalized at this point.

A rough guideline for investment required for treatment of process and other miscellaneous water streams was given in the "Final Report of the Supply Technical Advisory Task Force Synthetic Gas-Coal" as follows.

<table>
<thead>
<tr>
<th>Investment MM$ Mid 1971</th>
</tr>
</thead>
<tbody>
<tr>
<td>Process Water</td>
</tr>
<tr>
<td>Miscellaneous Water streams</td>
</tr>
<tr>
<td>TOTAL</td>
</tr>
</tbody>
</table>
a. Investment Costs for Particulate Removal

For typical coal gasification processes the investment required for removal of particulates resulting from coal handling, crushing, conveying etc. was estimated in the F.P.C. report above for typical 250 MMCF/D plant to be 2 MM$. This investment figure however is to reduce the particulate emissions to environmentally acceptable levels via, cyclones, scrubbers, draft fans etc....

It does not represent investment for more sophisticated levels of particulate removal as dictated by erosion, corrosion, other problems, the low Btu gases would encounter in combined power generation.

Example

Economic Analysis TRW Chemical Beneficiation of Coal Meyers Process

Basis

10,000 Tons/Day

\[ 10^4 \times 2 \times 10^3 = 2 \times 10^7 \text{lb}_m \text{ of coal per day} \]

\[ 15 \times 2000 \text{ lbs} \quad 390 \times 10^6 \text{ Btus} \]

\[ \text{Btus/lb}_{\text{coal}} = \frac{3.9 \times 10^8}{15 \times 2000} = 13000 \text{ Btu/lb}. \]

Btu's/day

\[ 13000 \times 2 \times 10^7 = 2.6 \times 10^{11} \]

Which compares with a 250 MMCF/Day

or \[ 2.5 \times 10^8 \times 10^3 = 2.5 \times 10^{11} \text{ Btu/Day gasification flame.} \]

Costs analysis for coal preparation and chemical desulfurization n
Basis for Design

10,000 T/D of product coal
including inert components and 5% moisture and fuel coal

operation 24 hrs/day
330 production days/year
10 days product inventory
3 days raw coal inventory

Calculation of Total Capital Requirement Process . . . .

1. All on site plant sections
   Coal Storage MM$
   Coal Preparation S- product drying compacting shipping 16
   Coal Processing Equipment 38
   Waste Heat Recovery
   Gas Purification (particulate removal scrubbing)
   Site Development and Land Costs 4
   Fresh Water Treating
   Compression Facilities
   Cooling Towers
   Power Generation and Distribution
   Steam Generation
   Waste Disposal Facilities
   Utilities, Buildings 22
   Office
   Shop
   Control System
Oxygen Plant
Hydrogen Plant
Other on site plant sections
Special lining, tankage

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>SUBTOTAL</td>
<td>90</td>
</tr>
</tbody>
</table>

2. Contractors Overhead and Profit (included in the above)

Engineering and Design costs (included in the above)

Plant Investment Subtotal 90

3. Project Contingency

(In the absence of detail below use 15% of subtotal plant investment)

Allowances for results of a Detailed Design
a. New railroad spur to plant site
b. New roadways to plant site
c. New power transmission to plant site
d. New pipeline for water to plant site
e. New slurry pipeline if applicable
f. New unitrain if applicable
g. Contingency site problems:
   Piling
   Grading
h. Additional clean up facilities for effluent water gas etc.
i. Additional equipment for start up shut down stand by facilities
j. Additional safety equipment
k. Additional equipment for process on mechanical emergencies
l. Standby equipment for reliability
m. Additional space equipment
n. Contingency for missed costs estimates
o. Premium labor for construction
p. Contingency for low labor productivity and strikes

Total Project Contingency 13.5

4. Process Development Contingency

(In the absence of detail below use 7% of plant investment subtotal)

6.3

Detailed Allowances for Process Development Contingency

a. Lower gasification rates
b. Lower product yields from reactors
c. Lower thermal efficiency
d. Lower distillation contacting performance
e. Need for more sophisticated reactor-gasifier internals (plates, grids, distributors, downcomers, cyclones nozzles, turbulence fromolers, etc...)
f. Costs escalation in feed or ash removal systems
g. Need for additional coal healment
h. Unexpected fine removal problems
i. Unexpectedly high sulfur
j. Unexpected heat transfer, drying problems

Total Process Dev. Contingency 6.3
Sum of the items 1 through 4 above gives

5. **Total Plant Investments** 109.8

6. Interest during construction \[ = \text{Interest Rate} \times \text{Total Plant Investment} \times 1.875 \text{ construction Period} \]
   I. Rate 9.5% 19.56

7. Start-up costs \[20\% \text{ of total gross operating costs}\]

8. Working capital
   a. Coal inventory \(\text{for 60 days at design rate}\) 4.680
   b. Materials and supplies \(0.9\% \text{ of Total plant investment}\) .988
   c. Net receivables \(\@ 1/24\) of annual gas reserve \(@ \$1.00/\text{MMBtu}\) 1.0

Total W.C. 6.67

Details

8a. Btus equivalent of coal per day \(2.6 \times 10^{11}\) using 30¢ per MM Btu Bituminous Eastern Coal

Detail cost of Coal \(0.30 \times 2.6 \times 10^{11} = 0.78 \times 10^5 \) S \(\times 10^6\)

60 day inventory \(60 \times 0.78 \times 10^5 = 4.68 \text{ MM}$

8c. Estimated at 28¢/\(10^6\) Btu

\[
\frac{1}{24} \times 0.28 \times 2.6 \times 10^{11} \times 330.0 = 10^6\]$ \(\times 10^6\) = 1 MM$

Basis for calculating gross and net operating cost

Base load plants \@ 90\% load factor

1. Coal (30¢/MM Btu high heating value, deep mined bituminous coals and 15¢/MM Btu surface mined subbituminous coals 25.74
2. Other Raw Materials detailed list Page T6 17.00

3. Catalysts and Chemicals

4. Purchased Utilities
   a. Electrical Power @ 0.9¢/KWH 0.297
   b. Raw water @ 30¢/MGal (delivered) 0.119
   c. Cooling Water 20° temperature rise 0.003
   d. Sewerage @ 0.001 $/MGal 0.003
   e. Refuse disposal @ $0.50/ton 0.167

5. Labor
   a. Process labor [(Men/shifts) X 8304 Man-hours/year
      X 5.20 $/Man hour] (l) .593
   b. Maintenance Labor (1.5%/year of total plant invest-
      ment) 1.650
   c. Supervision (15% of operating and maintenance labor) .338
   d. Administration and General Overhead 60% of total
      items a,b,c, above 1.550

Raw Materials
Oxygen 99.5% pure @ $17/ton

\[
\text{Animal Coal} \quad \frac{3.0400}{10^6} \times \frac{17}{10} \times \frac{10000}{2,400}
\]

\[
\text{Oxygen} \quad \frac{3.04}{102.4} \times 17 \text{ MM}$
\]

Solvent Naptha
\[
@ 200 \text{ lb/hr} \times \frac{10}{2.4} \times 24 = 20000 \text{ lbs/Day}
\]

\[
\text{cost} @ $45/ST \quad \frac{4500}{2000} = 2.25¢ \text{ lb.}
\]

\[
\text{Annual cost of solvent} \quad 20,000 \times 330 \times 2.25/10^6 = 14.85 \text{ $MM}
\]

(1) Based on 1973 Figures.
Utilities

Electric Power $1000 \frac{10}{2.4} = 4166.7$ KW.

Annual cost @ $0.012$/KWH.

$$\frac{4166.7 \times 0.012 \times 24 \times 330}{1 \times 10^6} = 0.396 \text{ MM$/year}$$

@ 0.9 0.009 0.297 MM$/year

Cooling water 2000 GPM $\times \frac{10}{2.4} = 8333.3$ gpm

Annual cost delivered @ flat @ 30¢/MGal/

$$\frac{8333.3 \times 1440 \times 330 \times 0.30}{1000 \times 1 \times 10^6} = 1.188$$

1.188 x 0.001

.30

= .003.96

Annual Labor Cost 3.3 operating positions $\times \frac{10}{2.4}$

13.75 positions operating

Process Labor 13.75 x 8304 $\times \frac{5.20}{10^6} = .593 \text{ MM$}$

Operating and maintenance labor

1.650 + .593 = 2.243

Supervision 15% x 2.243 = .338

Operating and maintenance and supervision 2.581

Administration and general overhead 60% x 2.581 = 1.550 MM$
Credits

Total coal handled per year

\[ \frac{0.8 \times 10}{2.4} = 3.33 \text{ MMT/year} \]

Refuse generated \[ 3.33 \times 0.067 = 0.223 \text{ MMTons of ash &} \]
\[ 3.33 \times 0.036 \times 0.92 = 0.110 \text{ MMTons of Sulfur} \]
\[ 0.333 \text{ MMTons/refuse} \]

Refuse disposal cost @ $0.50/Ton

\[ 0.333 \times 10^6 \times 0.5 = 0.1665 \text{ MM$} \]

6. Supplies

Operating supplies (30% of process operating labor) \[ 0.178 \]

Maintenance supplies (1.5%/year of total plant investment) \[ 1.650 \]

7. Local Taxes and Insurance (2.7%/year of total plant investment) \[ 2.960 \]

Total Gross Operating Cost/year \[ $\text{MM} \quad 52.245 \]

8. By products Credits

Sulfur (10.00 $/LT) \[ 1.0 \]

Ammonia (25.0 $/ST) \[ 0.0 \]

Light Oil (Where applicable) @ 15¢/Gal \[ 0.0 \]

Heavy Oil (Tars 30¢/MM Btu) \[ 0.0 \]

Char (90% of coal cost) \[ 1.000 \]

Total Credits \[ 1.0 \]

Total Net Operating Cost/Year \[ 51.245 \]
<table>
<thead>
<tr>
<th>End of Year</th>
<th>Rate Base TCR-Accrued Depr. @ Mid-Year</th>
<th>Return on Rate Base 10.86% of (1)</th>
<th>Return on Equity 15% of .25 of RB</th>
<th>Fed. Inc. Tax 48/52 X 3</th>
<th>Deprec. 0.05 (TCR-Work Cap)</th>
<th>Total Net Op. Cost</th>
<th>Total Fuel Rev. Reg. (2)+(4)+(5)+(6)</th>
<th>Fuel Cost (7)/annual Feed $/MMBtu</th>
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<tbody>
<tr>
<td>1</td>
<td>142,980</td>
<td>15,500</td>
<td>5350</td>
<td>4950</td>
<td>7000</td>
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<td>10,966</td>
<td>3787</td>
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<td>69,100</td>
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<td>72,980</td>
<td>7,926</td>
<td>2736</td>
<td>2525</td>
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<td>51,245</td>
<td>68,696</td>
<td>80.1</td>
</tr>
<tr>
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<td>7,165</td>
<td>2474</td>
<td>2284</td>
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<td>66,692</td>
<td>77.7</td>
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<tr>
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<td>5,645</td>
<td>1949</td>
<td>1799</td>
<td>7000</td>
<td>51,245</td>
<td>65,689</td>
<td>76.6</td>
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<tr>
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<td>44,980</td>
<td>4,885</td>
<td>1686</td>
<td>1556</td>
<td>7000</td>
<td>51,245</td>
<td>64,686</td>
<td>75.4</td>
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<td>37,980</td>
<td>4,125</td>
<td>1424</td>
<td>1315</td>
<td>7000</td>
<td>51,245</td>
<td>63,685</td>
<td>74.2</td>
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<tr>
<td>17</td>
<td>30,980</td>
<td>3,364</td>
<td>1162</td>
<td>1072</td>
<td>7000</td>
<td>51,245</td>
<td>62,681</td>
<td>73.1</td>
</tr>
<tr>
<td>18</td>
<td>23,980</td>
<td>2,604</td>
<td>899</td>
<td>830</td>
<td>7000</td>
<td>51,245</td>
<td>61,679</td>
<td>71.9</td>
</tr>
</tbody>
</table>
SUMMARY ECONOMIC EVALUATION OF MEYERS (TRW) PROCESS

COAL FEED

LOWER KITTANING COAL 14 MESH TOPSIZE

Chemical Analysis % Dry Basis

Component

<table>
<thead>
<tr>
<th>Component</th>
<th>% Dry Basis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ash</td>
<td>20.7</td>
</tr>
<tr>
<td>Pyritic Sulfur</td>
<td>3.6</td>
</tr>
<tr>
<td>Organic Sulfur</td>
<td>0.6</td>
</tr>
<tr>
<td>Sulfate Sulfur</td>
<td>Nil</td>
</tr>
</tbody>
</table>

Heating Value 12,300 Btu/lb

RATE 10,000 Tons/Day

~ 260 Billion Btu/day

COAL PRODUCT

Chemical Analysis % Dry Basis

<table>
<thead>
<tr>
<th>Component</th>
<th>% Dry Basis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ash</td>
<td>14</td>
</tr>
<tr>
<td>Pyritic Sulfur</td>
<td>0.3</td>
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<td>Organic Sulfur</td>
<td>0.6</td>
</tr>
<tr>
<td>Sulfate Sulfur</td>
<td>Nil</td>
</tr>
</tbody>
</table>

Heating Value 12,900 Btu/lb

COST OF FUEL BY UTILITIES FINANCING METHOD

Input Parameters

Debt/Equity Ratio: 75%/25%
Per cent Interest on Debt = 9.5%
Per cent Return on Equity = 15%
Federal Income Tax Rate = 48%
Total Capital Requirement

<table>
<thead>
<tr>
<th>Description</th>
<th>Cost (MM$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Plant Investment</td>
<td>109.80</td>
</tr>
<tr>
<td>Interest During Construction</td>
<td>19.56</td>
</tr>
<tr>
<td>Start-up Costs (20% of Gross Operating Cost)</td>
<td>10.45</td>
</tr>
<tr>
<td>Working Capital</td>
<td>6.67</td>
</tr>
</tbody>
</table>

TOTAL CAPITAL REQUIRED (T.C.R.) = 146.48 MM$

Initial Debt 75% of TCR = 110.00
Initial Equity 25% of TCR = 36.48

Annual Fuel Production Rate

Plant Capacity 10,000 T/SD

Annual Fuel Production ≥ 2.6 x 10⁶ x 3.3 x 10² (1)
= 8.58 x 10¹³ Btus/year
= 85,800 Billion Btu/year

Per cent return on Rate Base: 0.75 X 9.5% + 0.25 X 15% = 10.86%

(1) 330 Stream Days per year
CAPABILITY OF EXISTING ELECTRIC GENERATING UNITS TO USE CLEAN FUELS DERIVED FROM COAL

by

Edward R. Lady

The processing of coal into a clean, low-sulfur fuel yields a combustible product that is delivered as a solid, liquid, or gas. Although specific combustion tests must be carried out, there is little doubt that such fuels can be utilized in steam generating equipment or combustion powered prime movers that are designed to handle the particular fuels to be used. With these fuels new plants will be able to meet the air quality standards required by statute.

The existing coal fired electric generating units ultimately will have to reduce their sulfur dioxide and particulate emissions according to present law. The average sulfur content of the coal burned by the utilities in 1970 was 2.58 percent by weight (1), a content considerably in excess of the approximately 0.6 percent sulfur coal needed to reduce sulfur dioxide emissions to the required level. Some boilers are being modified to burn low-sulfur, high ash western coal to meet pollution standards. The majority of existing coal fired plants will have to utilize eastern, high sulfur coal or shut down for the lack of fuel. This section of the report considers some of the problems and potentialities of utilizing clean, coal-derived fuels in power plants currently in operation.
EXISTING FOSSIL FUELED GENERATION CAPACITY

The projected growth of the installed generating capacity of the United States is shown in Table I. Although the projected share of the fossil-fueled steam generation capacity decreases from 77 percent in 1970 to 44 percent in 1990, the Federal Power Commission projections (2) indicate that 382,000 MW of new fossil fuel plants will be added during these two decades, while only 85,000 MW of old plant capacity will be retired. Thus, of the 259,000 MW fossil capacity units operating in 1970, 174,000 MW will see service throughout the period 1970-1990 and must be considered as candidate units for retrofitting to utilize clean fuel or install stack gas scrubbing. The units scheduled to be retired during this twenty year time period (1970-1990) do not have sufficient useful life to justify large capital investments required by the various clean fuel processes. A few of these latter units have been selected to be used as the fuel consuming section of clean-fuel demonstration plants. Such demonstration plants, to be placed into service in the 1975-1985 period, cannot be operated economically due to their small (50-100 MW) size and will be retired from service within five to ten years, thus matching the remaining life of the older steam plants connected to them. Let us examine the 174,000 MW of relatively new fossil steam plants and determine the portion which burns high sulfur coal.
### TABLE I

**Installed and Projected Electrical Generation Capacity of United States, in 1000 MW**

<table>
<thead>
<tr>
<th></th>
<th>1970</th>
<th>1990</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fossil - Fueled Steam</td>
<td>259</td>
<td>577</td>
</tr>
<tr>
<td>Hydro and Nuclear</td>
<td>81</td>
<td>683</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>340</td>
<td>1260</td>
</tr>
</tbody>
</table>

The 1970 fossil fuel breakdown by type of fuel was 55.6 percent coal, 15.3 percent oil and 29.1 percent gas (2). Assuming that generating units fueled by coal, oil or gas had about the same capacity factor, this indicates that plants representing some 97,000 MW burned coal. In addition, during the period 1965-1970 there were 226 boilers, supplying 16,250 MW of generation capacity, that had been converted from coal to oil firing (3). Seventy-seven percent (12,500 MW) of these converted boiler are re-convertible to coal.

The coal-fired generating plants in the Mountain Region, Figure 1, use coal averaging 0.65 percent sulfur and therefore need not be included in plants which may require low sulfur fuel unless local regulations specify less than 1.2 pounds sulfur dioxide per million Btu. However, this region represents only 8000 MW of capacity and may be off-set against possible reconversions of East Coast plants from oil to coal. Therefore, the potential of the market for
Figure 1. Major Geographic Divisions of the United States
retrofit of existing plants burning high sulfur coal to a clean fuel is 90,000 - 100,000 MW of generation capacity.

FUEL CONVERSION CONSIDERATIONS

Boilers may be designed to burn coal, oil or gas, either singly or in various combinations. A boiler designed for a given fuel can be converted to use a different fuel through burner replacement, adjustment in the heat transfer surface and possible changes in the forced draft, induced draft, and recirculation fan speeds. Each boiler must be examined on a case by case basis by the original manufacturer, a process that requires several weeks of engineering per boiler. It is agreed, however, that such conversions are possible and in the eight year period, 1965-1972, 398 boilers were converted from coal to oil. Table II shows the annual rate these boilers were converted. It is estimated that another 40 boilers were converted from coal to gas during this period.

The combustion of oil or natural gas results in a higher heat release rate per unit volume than the combustion of pulverized coal. Because of this, boilers designed for oil or gas are smaller in volume and conversion to coal firing would be impractical. As discussed above the reverse conversion is feasible and has been accomplished many times. The general considerations in converting coal fired boilers to the various clean fuels derived from coal are tabulated in Table III.
<table>
<thead>
<tr>
<th>Year</th>
<th>Number of Boilers Converted from Coal to Oil</th>
<th>Total Nameplate Capacity of Generators Associated with Boilers Converted from Coal to Oil, Megawatts</th>
<th>Associated Generator Nameplate Capacity of Boilers Re-Convertible to Coal, Megawatts</th>
<th>Associated Generator Nameplate Capacity of Boilers Not Re-Convertible to Coal, Megawatts</th>
</tr>
</thead>
<tbody>
<tr>
<td>1965</td>
<td>9</td>
<td>651.9</td>
<td>35.5</td>
<td>616.4</td>
</tr>
<tr>
<td>1966</td>
<td>16</td>
<td>1,411.0</td>
<td>375.0</td>
<td>1,036.0</td>
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<tr>
<td>1967</td>
<td>36</td>
<td>1,703.9</td>
<td>1,267.7</td>
<td>436.2</td>
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<tr>
<td>1968</td>
<td>51</td>
<td>2,295.3</td>
<td>1,427.4</td>
<td>867.9</td>
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<tr>
<td>1969</td>
<td>51</td>
<td>5,589.4</td>
<td>5,048.6</td>
<td>540.8</td>
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<td>1970</td>
<td>63</td>
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<td>87</td>
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<td>1972</td>
<td>85</td>
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</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td><strong>398</strong></td>
<td><strong>28,785.1</strong></td>
<td>22,703.9</td>
<td>6,081.2</td>
</tr>
</tbody>
</table>

Percent of Total

100.0 78.9 21.1

1/ Table taken from Reference (3).

2/ The electric utilities estimated the total cost of re-conversion at $106.5 million.
<table>
<thead>
<tr>
<th>Present Fuel</th>
<th>Replacement Fuel</th>
<th>Considerations Involved</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal</td>
<td>Solvent Refined Coal</td>
<td>a. Pulverizers may be too hot and cause SRC to become sticky</td>
</tr>
<tr>
<td></td>
<td></td>
<td>b. Negligible ash problem</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c. Additional combustion tests required to determine flame characteristics</td>
</tr>
<tr>
<td></td>
<td></td>
<td>d. Long term storage characteristic unknown</td>
</tr>
<tr>
<td>Coal</td>
<td>Liquefied Coal</td>
<td>a. Eliminates solid coal handling</td>
</tr>
<tr>
<td></td>
<td></td>
<td>b. New burners and atomizers required</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c. Special heating of fuel may be required</td>
</tr>
<tr>
<td></td>
<td></td>
<td>d. Storage tanks required</td>
</tr>
<tr>
<td></td>
<td></td>
<td>e. Similar to coal-oil conversions</td>
</tr>
<tr>
<td>Coal</td>
<td>High Btu Gas</td>
<td>a. New burners, gas piping</td>
</tr>
<tr>
<td></td>
<td></td>
<td>b. Precipitators not required</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c. Superheater and reheater banks may have to be modified</td>
</tr>
<tr>
<td></td>
<td></td>
<td>d. Eliminates coal and ash handling</td>
</tr>
<tr>
<td></td>
<td></td>
<td>e. Similar to coal-natural gas conversions</td>
</tr>
<tr>
<td>Coal</td>
<td>Low Btu Gas</td>
<td>a. Ductwork and burners must be added</td>
</tr>
<tr>
<td></td>
<td></td>
<td>b. Induced draft fan changes</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c. Superheater and reheater banks may need adjustment</td>
</tr>
<tr>
<td></td>
<td></td>
<td>d. Furnace volume probably sufficient</td>
</tr>
<tr>
<td></td>
<td></td>
<td>e. No ash, particulate problem</td>
</tr>
</tbody>
</table>
Such fuel conversions as listed in Table III require varying lengths of time to complete but it is extremely unlikely that the replacement fuel could be supplied more quickly than the time to complete the boiler conversion. The boiler conversion costs vary with the installation but will probably average more than the $4.67 per KW cost reported (3) to reconvert oil-fired boilers back to the original coal firing.

CONVERSION CASE STUDY

A study (4) has been made by a major boiler manufacturer to determine the modifications necessary to a modern 500 MW coal-fired boiler so that low Btu gas can be burned. The boiler was placed into operation in 1969 and operated some thirty months. It was then converted to No. 6 oil firing by adding steam atomized oil guns, gas recirculation through the hopper bottom and removing superheater surface. The coal to oil conversion cost was $16.60/KW, not including charges for loss of capacity during conversion.

Although the boiler is now oil fired, it may be considered a representative of a coal fired boiler since coal was the design fuel. The references study is based upon changing from oil to gas of 127 Btu/scf higher heating value. The changes necessary to burn this gas consist of:
a. New windbox internals
b. New gas ducts
c. New induced draft fans and drives
d. Gas recirculation not required

The oil-to-gas conversion cost estimate is $5.60/KW. Boiler outage time for conversion is 6 to 8 weeks.

It is seen that the conversion from coal to oil to gas or coal to gas can be done quickly and at relatively low cost. This conversion does not include the time and expense of the coal gasifier. This was estimated to cost $138.60/KW based on an atomospheric pressure gasifier design of the boiler manufacturer. This large capital cost of the gasifier increases the energy cost from this 500 MW unit by 5.9 mills/KWHR, based on a 51.5% capacity factor and 20% fixed charge rate.

CONCLUSIONS

In the United States there are several hundred relatively new boilers designed for and firing high sulfur Eastern coal. These could be modified to burn any of the clean fuels from coal that are likely to be developed. However, in addition to this 100,000 MW of convertible capacity, there will be added during the 1970-1990 period some 382,000 MW of new fossil-fuel steam plants. It is believed that half of the new fossil steam plants must rely on coal as the fuel source, or some 190,000 MW of new capacity will need clean fuel from coal or stack gas scrubbing.
Many factors lead to the belief that it will be new plants, rather than existing plants, that will utilize clean fuels derived from coal. Among these are:

a. There will be twice as much new plant capacity
b. Integration of coal processing and fuel consumption must be carried out for maximum economy

c. Coal processing should be continuous, 24 hours per day, to reduce capital charges. New generation plants are more suitable for continuous base-load operation.

d. Coal processing should be on a large scale, equivalent to 1000 MW generation capacity. Most older plants are smaller.

e. Existing plants have many site limitations which would prevent on-site coal processing

f. New plants designed to handle the clean fuel will avoid the cost and capacity loss of retrofitting existing plants.

In summary it is recommended that a few existing steam generation units be used as demonstration plants for the new generation of clean fuels but that full scale use of these fuels be directed toward the new plants as they are designed and constructed.
Bibliography


THERMODYNAMIC ANALYSES

by

John E. Powers
Andre W. Furtado

The projected needs of the utility industry are such that coal—low-grade sulfurous coal—must be reconsidered to be the primary source of energy for power generation not only at present but especially in the immediate future. Most of the coal burning plants presently in operation will have to be modified in some way so that degradation of the environment is not worsened. Many power generating stations now fueled with natural gas and petroleum fluids will have to be converted—or reconverted—to coal. New plants abuilding, in design or planned, will have to burn coal.

Many alternative solutions have been proposed to permit the utility industry to utilize the existing coal reserves in environmentally acceptable ways. It seems likely that no one solution will solve all of the problems of the utility industry. However, since the magnitude of the problem is so huge and the time scale which reflects needs is continually shrinking, it is essential that as many factors as possible be taken into account when considering alternative paths and processes. One factor that will be of ever increasing importance is the overall efficiency of generating electrical power from coal. In the utility industry this is succinctly expressed in terms of the Heat Rate, i.e., the Btu of heating value of the fuel per KW hr of net electrical power generation. The value for modern power plants is about 8500-9000 Btu/KW hr.
This method of expressing the Heat Rate has a parallel formulation in terms of the first and second laws of thermodynamics (as will be developed later) for power plants as currently operated. However as changes in methods of utility plant operation are brought about—as they most certainly will be—it is essential that the basis for thermodynamic analyses be understood so that this powerful analytical tool can be applied more generally.

Therefore the purposes of this section are to:

(1) Set forth a model that should be generally applicable to all processes whereby coal is used as the fuel in the generation of electric power.

(2) Illustrate the application of both the first and second laws of thermodynamics to both conventional boiler plants and to more complicated plants involving extensive processing of the coal and/or effluents.

(3) Describe some calculations that have been carried out to illustrate the required thermodynamic calculations.

A GENERAL MODEL FOR THE UTILITY INDUSTRY

No model will be realistic in its applications without recognition of the magnitude of the tasks facing the utility industry as outlined in the introduction to this section. Therefore the following restrictions are incorporated in the model:
(1) Coal is the fuel to be used. In addition, the coal is taken to be as provided by mother nature--i.e., "in place." That's the only kind that is available for purchase now and in the forseeable future.

(2) Nothing can be purchased for use by the utility on a continuing basis that is not in abundant supply in nature. The only raw materials permitted for large scale purchase and use are:
   
   (a) Limestone
   
   (b) Air
   
   (c) Water

   All other materials used in the process must either have long life (boilers, turbines, condensers, etc.) or be capable of being almost completely regenerated (diethanol amime, methanol, etc.) so that relatively small amounts are actually consumed.

(3) All effluents from the plant must be disposed of in an environmentally acceptable manner.

The system selected for thermodynamic analyses is represented in Figure 1:

![Diagram of a power generating station with labeled inputs and outputs.]

Figure 1. Generalized Power Generating Station
Whereas it is relatively simple to draw a box and designate the box as the system, it is essential that each point at which material or energy crosses the boundary be carefully identified. Consider first the inlet streams. It has already been decided that the system boundaries include the coal in the ground. This choice has several consequences:

(1) The coal (and associated ash material as mined) is at ambient temperature.

(2) The energy requirements necessary to mine, clean, and otherwise prepare the coal (crushing, grinding, screening, briqueting, etc.) must be taken into account in determining the net electrical power generation of the entire complex.

(3) Transportation of the coal must be taken into consideration. If fuels other than coal are required for transportation they must be factored in. If other fuels are required for light-off or to sustain combustion these must also be accounted for.

Unless there is evidence to the contrary it will be assumed that all other raw materials (water, air, and limestone) are also available to the plant as found in nature. Therefore energy requirements associated with winning these materials from nature and transporting them to the site will have to be taken into account. Unless otherwise justified it will be assumed that these raw materials are available at ambient temperature. Exceptions might include geothermal or other such natural sources.
of hot water. Of course pumping and/or required treatment of such waters would be charged against the process.

Consider now the various wastes emanating from the plant. It is self evident that every attempt should be made to produce such "wastes" in forms usable to society. Alas, the amounts are of such magnitudes that it is difficult for society to make use of them. However this should be the goal not only for the direct benefit of society but also for the economic benefit of the utility. More on this aspect later.

The symbol \( W \) is used to represent the net electrical power generation of the station. (There may be very exceptional cases where mechanical energy is transferred out of the system and this would be included as \( W \).) \( EQ \) represents all transfer of heat energy between system and surroundings, and here is where a liberal and very useful view of the "surroundings" is taken in establishing the boundaries of the system. Consider, for example, the combustion products which issue from practically all such plants. In general they are at temperatures in excess of 300°F. However as they mix with the air the temperature is reduced by mixing or heat transfer or what have you. The important thing is that there is no practical way of gaining useful work by reducing the temperature much below 300°F unless the \( SO_2 \) is removed. Therefore the system boundaries are taken such that the combustion products have attained ambient temperature.
If similar considerations are applied to other gaseous wastes and to the liquid and solid wastes, it will be seen that it is logical and consistent to consider that sufficient heat is transferred to cool all of these products to ambient temperature and that effectively, all of this heat is transferred at ambient temperature, i.e., a system is selected that is large enough to justify these assumptions. The same reasoning applies to heat losses from the boiler and turbine; the system is chosen to include the air surrounding these devices so that heat is transferred at ambient temperature. Exceptions will be made if the proponents of a process can establish that the temperature in excess of ambient is important to some other process. This will be very unusual.

Of course most of the heat transferred in any power generating station is transferred to water in condensers. If this is done without effective utilization of the heat for any useful purpose, it can be considered that the heat is effectively transferred at ambient temperature.

Two streams labeled "Process Steam" are placed alongside \( \Sigma Q \) in Figure 1. In some large industries (refineries generally) plus large universities, power plants serve the primary function of providing steam to heat buildings and find it economically attractive to first generate electrical power for their own use or for sale. It would seem apparent that such use of "waste heat" provides great potential on a national basis for conservation of fuel for home heating. This subject will also be addressed in some detail later.
In summary, the model is formulated so that in most cases all materials and energy (other than electrical energy and process steam) are accepted from nature and returned to nature at ambient temperature. The net electric power is the major objective and further consideration must be given as to how the process steam is taken into account.

APPLICATION OF THE MATERIAL BALANCE CONSTRAINTS AND THE FIRST LAW OF THERMODYNAMICS

Application of a steady-state model to the utility industry is certainly of questionable validity in view of the very significant changes in load that are indeed experienced over relatively short periods of time. However, in order to obtain some reasonable answers in a short period of time, it is almost mandatory to accept a period of observation (accounting period) long enough so that unsteady state effects are assumed to be negligible. For example if an intermediate load plant is to operate for 8 hours per day it seems reasonable to work with the total power generated during that period, the total coal used, etc. Under these conditions the energy required to heat up the boilers, etc., is taken into account even though these factors are indeed insignificant. Such an assumption may not be valid for peak-load plants but these are not of principal concern at this time.

In addition to assuming that pseudo-steady-state conditions prevail, it is also reasonable to assume that potential and kinetic energy effects will be negligible for the model proposed. Therefore the material balance and first law of
thermodynamics for the model can be written as follows:

\[ \Sigma M_{RM} + M_{PSi} = \Sigma M_w + M_{PSO} \]  \hspace{1cm} (1)

where \( M_{RM} \) refers to raw materials entering the system, 
\( M_w \) refers to waste streams leaving the system, and 
\( M_{PSi} \) and \( M_{PSO} \) refer to the process steam entering and 
leaving the system, respectively.

Similarly, the first law of thermodynamics can be written (1) as:

\[ \Sigma H_w - \Sigma H_{RM} + [H_{PSO} - H_{PSi}] = \Sigma Q - W \]  \hspace{1cm} (2)

where the symbol \( H \) is used to represent the enthalpy of a 
total stream and the subscripts are those identified in 
connection with Equation (1). \( \Sigma Q \) and \( W \) were identified in 
the previous section.

Under the conditions set forth in the model it is a 
relatively simple procedure to determine the specific enthalpies 
of each stream. For example, given the pressure, temperature 
and/or quality of the process steam the specific enthalpy, \( H \), 
can be obtained from steam tables. Thus Equation (2) can be 
rewritten as:

\[ \Sigma M_{H_w} - \Sigma M_{H_{RM}} + [M_{PSO}H_{PSO} - M_{PSi}H_{PSi}] = \Sigma Q - W \]  \hspace{1cm} (3)
In making calculations of this type it is found to be convenient to take the enthalpy of any compound in a stream to be equal to its heat of formation at that temperature and pressure relative to the elements at some base conditions. In most cases the enthalpy of the total stream can then be found by assuming ideal mixing of the component. Only in extreme cases (such as large amounts of liquid wastes) will it be necessary to take into account the non-ideality of mixtures.

So how are these equations to be applied?

(1) Unless a complete material balance as represented by Equation (1) is provided, one can't tell whether even the most basic and fundamental factors have been taken into account. Indeed, it is essential to extend the simple balance to individual molecular and atomic species to insure that stoichiometric constraints are met, i.e., that the law of conservation of mass is not violated.

(2) Once the amount, composition, temperature, and pressure of each stream is identified, it is a challenging but nevertheless fairly straightforward matter to determine a value for the specific enthalpy of that stream. The net amount of electrical energy must be either measured (as in the case of analysis of an operating plant) or estimated by some valid procedure (i.e., the proposed procedures for obtaining electrical energy cannot be in violation of the second law of thermodynamics).
Under these conditions one can calculate $\Sigma Q$ using Equations (2) or (3). Of course one would expect the number obtained to be approximately equal to the amount of heat transferred in the condenser when the temperature of the exiting flue gas and "heat losses" are properly identified.

The question of how this relates directly to the current and proposed methods of analysis of power plants is delayed until after discussion of the second law in the next section.

APPLICATION OF THE SECOND LAW OF THERMODYNAMICS

Ask five mechanical engineers, five chemical engineers, five chemists, and five physicists to write down the most general statement of the second law of thermodynamics that they can think of and you can rest assured that you will get at least 4 very different statements and more probably closer to 20 than 4. Therefore we will accept the general statement contained in a recent undergraduate text(1). In slightly abbreviated form it is presented in terms of a balance made on entropy analogous to those made on mass [Equation (1)] and on energy [Equations (2) and (3)] i.e., under steady-state conditions

$$\Sigma M_{w^*w} - \Sigma M_{RM} = \left[ M_{PSO} S_{PSO} - M_{PSi} S_{PSi} \right] = \frac{\Sigma Q}{T_b} + S_p \quad (4)$$

where $S$ refers to specific entropy
$T_b$ is the temperature at which heat energy crosses the boundaries of the system, and

$S_p$ is the entropy production.

In this context a very complete and general statement of the second law is:

(1) For any actual process the value of $S_p$ is always greater than zero, i.e.,

$$S_p > 0 \ [\text{All actual processes}] \quad (5)$$

(2) Only in the case of conceptual perfection does $S_p$ approach zero, i.e., a thermodynamically ideal process, i.e., a reversible process, is defined by

$$S_p = 0 \ [\text{Definition of a reversible process}] \quad (6)$$

So how can this be applied to aid in evaluating processes which produce electrical power?

If one can evaluate the specific entropy, $S$, for each component of a stream and further estimate the resulting entropy of the entire stream taking into account the effects of either ideal or non-ideal mixing, numerical values can be assigned to most of the terms in Equation (4). Without going into detail (2), it is noted that the third law of thermodynamics provides a convenient basis for such calculations in permitting one to assign values of absolute entropies
to compounds at any temperature and pressure. This is similar in many respects to identifying the enthalpy of compounds in terms of heats of formation in application of the first law [Equation (3)].

With this knowledge based on the most fundamental of thermodynamic data, most of the terms in Equation (4) can be assigned numerical values. Further, if, as has been specified in the general model, all heat transfer occurs at ambient (dump) temperatures, \( T_D \) such that

\[
\frac{\varepsilon^F_T}{T_b} = \frac{\varepsilon^F}{T_D}
\]  

(7)

then the entropy produced by any actual or proposed process can be calculated. In comparing actual processes, it can be assumed that the one with the lowest rate of entropy production (on comparable bases) has advantages over the one with the higher rate of entropy production. This point will be clarified in a later section.

Again we return to the question "How does this relate to the thermal efficiency of power generating stations as currently calculated?" An attempt will be made to explain this in the next section.

ENERGY EFFICIENCY OF CONVENTIONAL POWER PLANTS USING COAL

Most power plants do not produce significant amounts of process steam and therefore Equations (1), (3), and (4) reduce to
\[ \Sigma M_W - \Sigma M_{RM} = 0 \] (8)

\[ M_{W,W} - M_{RM,RM} = \Sigma Q - W \] (9)

\[ M_{S,W} - M_{RM,RM} = \frac{\Sigma Q}{T_D} + S_p \] (10)

In conventional power plants limestone would not be used nor would it appear as a solid waste.

As pointed out previously, Equation (8) (and similar expressions as applied to individual atomic species) can be used to insure that matter is conserved, Equation (9) can be applied to calculate the net amount of heat transferred (wasted) and Equation (10) can be used to calculate \( S_p \), the entropy production, i.e., a direct measure of the degradation of energy. However, entropy production is sort of a foreign concept and we might attempt to make it more understandable. To do this it has been found convenient to compare the actual (or proposed actual) process with a completely idealized, conceptual process. Consider two alternative processes as illustrated in Figure 2.

![Figure 2a. Actual Process to Generate Electrical Power.](image1)

![Figure 2b. Idealized Process to Generate Electrical Power.](image2)
Note that in the actual process, $S_p > 0$, as required by the second law of thermodynamics [Equation (5)] while in the idealized conceptual process (Figure 2b) $S_p = 0$ as permitted by Equation (6). Note also that the subscript $A$ is used to identify amounts of heat and work transfer in the Actual process and $I$ serves a similar purpose for the Idealized process.

Now there are an infinite number of idealized processes that could serve to convert coal, air, and water to electrical energy with concurrent production of wastes and thermal pollution, $\Sigma Q$. For purposes of direct comparison it seems reasonable to stipulate that the ideal process uses amounts of raw materials identical to the actual process, produces amounts of wastes identical to those of the actual process, and that the raw materials and wastes for both processes have identical compositions, temperatures, and pressures. As a direct result, the left-hand side of Equations (9) and (10) have identical numerical values for both the actual and the idealized processes; only those terms on the right are different. Thus it follows that

$$\Sigma Q_A - W_A = \Sigma Q_I - W_I$$  \hspace{1cm} (11)

and

$$\frac{\Sigma Q_A}{T_D} + S_p = \frac{\Sigma Q_I}{T_D}$$  \hspace{1cm} (12)
Naturally $S_p = 0$ for the idealized process as stated previously.

In order to emphasize the difference between the actual and idealized process it is convenient to define differences in heat transfer, $\Delta Q$, and differences in work generation, $\Delta W$ as follows:

\[
\Delta Q \equiv \Sigma Q_I - \Sigma Q_A
\]  
\[ (13) \]

\[
\Delta W \equiv W_I - W_A
\]  
\[ (14) \]

Combination of Equations (11), (13), and (14) with cancellation of terms yields

\[
\Delta W = \Delta Q
\]  
\[ (15) \]

Similarly combination of Equations (12) and (13) yields:

\[
S_p = \frac{\Delta Q}{T_D}
\]  
\[ (16) \]

so that

\[
\Delta W \equiv W_I - W_A
\]  
\[ (17) \]

\[
\Delta W = \Delta Q = T_D S_p
\]  
\[ (18) \]

$\Delta W$ is the difference between the amount of work (electrical energy) that might conceivably have been obtained from the fuel,
and the amount that was actually obtained, \( W_A \), and is therefore sometimes referred to as the lost work, \( W_L \). Thus, from Equation (18) we see that the entropy production can be converted to the amount of lost work by multiplying by the ambient (dump) temperature, \( T_D \).

One might reasonably be interested in the ratio of the actual work, \( W_A \), to that which might have been obtained from the same fuel, \( W_I \). Equations (9), (10), and (17) can be combined to yield an expression in terms of "availability" of energy.

\[
\sum M_W (H_W - T_D S_W) - \sum M_{RM} (H_{RM} - T_D S_{RM}) = - W_I
\]  

(19)

where the combination of thermodynamics properties and dump temperature, \( T_D, H - T_D S \), is commonly referred to as availability.

Therefore the ratio, \( W_A/W_I \), is given by

\[
\frac{W_A}{W_I} = \frac{W_A}{\sum M_W (H - T_D S_W) - \sum M_{RM} (H_{RM} - T_D S_{RM})}
\]  

(20)

How does this compare to the more commonly utilized thermal efficiency? This expression is defined by

\[
\text{Thermal efficiency} = \frac{W_A}{\text{Heating value of fuels.}}
\]  

(21)

If the heating value if taken as the net heating value, i.e., corresponding to water vapor rather than liquid water
as product, at 25°C (77°F) it can be shown that this is equivalent to

\[
\text{Thermal efficiency} = \frac{\sum W_A}{\sum M_{w} H_w - \sum M_{RM} H_{RM}} \tag{22}
\]

where the enthalpies are identified in terms of heats of formation.

When comparing Equation (22) with Equation (20), it will be seen that the thermal efficiencies is sort of related to the thermodynamic efficiency, \( \frac{W_A}{W_I} \), but only sort of.

WHAT ABOUT COMPARISON AND/OR CHARACTERIZATION OF MORE COMPLICATED PROCESSES?

In the preceding section, only the most simplified case in power generation was considered and only a similarity was found between thermal efficiency and thermodynamic efficiency. What happens if process steam is generated in the process? How should that be taken into account? What if limestone is fed to the plant and large amounts of ash containing gypsum and lime are carted away for disposal? The complete answers to these problems are much more complicated on a practical basis than on a thermodynamic one as will be considered in some detail in a later section. No attempt will be made to answer the question with regard to calculation of thermal efficiency because this would probably require development of an entirely new expertise. There are no problems associated with the calculation of thermodynamic efficiency. Carrying through similar manipulations but retaining the
terms associated with process steam yields

\[ W \equiv \dot{W} = T_D S_P \]

\[ = T_D \left\{ \sum_{W-W} M_{W} S_{W} - \sum_{RM-RM} M_{RM} S_{RM} + [M_{PSO} S_{PSO} - M_{PSi} S_{PSi}] - Q_{A} \right\} \]  

and

\[ \frac{W_A}{W_I} = \sum_{W} (H_{W} - T_D S_{W}) - \sum_{RM} (H_{RM} - T_D S_{RM}) + [M_{PSO} (H_{PSO} - T_D S_{PSO})] \\
- [M_{PSi} (H_{PSi} - T_D S_{PSi})] \]  

These equations can be applied to calculate the lost work and/or the thermodynamic efficiency of any process in which electrical power is generated.

**SHOULD A FOURTH LAW BE FORMULATED?**

Consider two alternatives procedures by which electrical power is generated and homes and/or office buildings in the surrounding area are heated. If the utility alone is considered as the system it can be shown that application of the first and second laws of thermodynamics will yield no significant distinction as to whether the heat transferred so as to condense the steam from the turbine is rejected as thermal pollution or used to heat the homes and/or office buildings
in the surrounding area. It seems obvious that much fuel would be saved and thermal pollution would be reduced if the condensing steam were used for some heating. This will definitely show up in the thermodynamic analysis if the homes and/or office building are included in the system of analysis and the existing method of analysis should be modified to include such a possibility. If the utilities were forced to provide such heating on a basis that is more wide spread than currently practiced, the savings in the nations prime energy, natural gas and fuel oil, would be very significant and the utility industry would double their income--at least in the winter--with very little capital expenditure.

SAMPLE CALCULATION FOR FLUIDIZED BED COMBUSTION PROCESS

Figure 3 illustrates a scaled-down version of a power plant employing a fluidized bed combustion chamber and using CaCO$_3$ to remove the majority (90%) of the sulfur in the solid waste. The original design specifications were provided by the Foster Wheeler Corporation (3). The proximate, ultimate, and ash analyses for the coal used in the design are indicated below:

a) Proximate Analysis (wt%)

<table>
<thead>
<tr>
<th>Component</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture</td>
<td>3.3</td>
</tr>
<tr>
<td>Volatile Matter</td>
<td>39.5</td>
</tr>
<tr>
<td>Fixed Carbon</td>
<td>48.7</td>
</tr>
<tr>
<td>Ash</td>
<td>8.5</td>
</tr>
</tbody>
</table>
Figure 3. Schematic of process used to illustrate the calculation of the thermodynamic efficiency.
b) Ultimate Analysis (wt%)  
(Moisture and Ash Free)  
\[
\begin{align*}
C &= 75.91 \\
H &= 6.28 \\
O &= 11.72 \\
N &= 1.495 \\
S &= 4.58
\end{align*}
\]

c) Ash Analysis (wt%)  
\[
\begin{align*}
\text{SiO}_2 &= 45.3 \\
\text{Al}_2\text{O}_3 &= 26.7 \\
\text{Fe}_2\text{O}_3 &= 28.0
\end{align*}
\]

Note: Although the ash actually contained ten oxides, the above three oxides constituted over 94% of the ash. As a first approximation, the ash analysis was normalized and expressed in terms of the three oxides above.

d) Heating value of coal = 13,000 Btu/lb.

The flue gas analysis is as follows:
\[
\begin{align*}
\text{CO}_2 &= .1462 \text{ (mole \%)} \\
\text{H}_2\text{O} &= .07419 \\
\text{SO}_2 &= .00032 \\
\text{N}_2 &= .75138 \\
\text{O}_2 &= .02784
\end{align*}
\]

STOICHIOMETRIC DEFINITION OF THE ORGANIC MATERIAL IN COAL

The chemical composition of coal is complex and undefinable and therefore deserves special attention. In particular, the ultimate analysis (moisture and ash free basis) is assumed
to define a hypothetical pure component for the carbonaceous material in coal, and together with the proximate analysis and ash analysis permits the mole fraction of all components to be specified as required.

The basis chosen for the stoichiometric definition of the organic material in coal is 1 gm atom of C.

Gm Atoms of C per 100 gms of coal analyzed = $75.91/12.01 = 6.32$.

Gm Atoms of H per 100 gms of coal analyzed = $6.28/1.00 = 6.28$. Therefore, Gm Atoms of H/Gm Atom of C = $6.28/6.32 = 0.986$.

By performing similar calculations given the O, N, and S content in the ultimate analysis we obtain the stoichiometric constitution of the organic combustible material in coal as

$$C_{1.0}H_{0.986}O_{0.116}N_{0.017}S_{0.023}$$

The molecular wt. of this compound is calculated as 15.82.

DEFINITION OF THE COMPOSITION AND RELEVANT THERMODYNAMIC PROPERTIES OF COAL

i) Calculation of the composition of the coal

The coal is now assumed to contain $H_2O$, $C_{1.0}H_{0.986}O_{0.116}N_{0.017}S_{0.023}$, $SiO_2$, $Al_2O_3$, and $Fe_2O_3$. Given the ultimate and the ash analysis and the molecular weights of all constituent components, the coal composition is calculated as:
$H_2O$ 0.0313 (mole fraction)
$C_{1.986}^1H_{0.116}^1N_{0.017}^1S_{0.023}^1$ 0.9519
$SiO_2$ 0.0109
$Al_2O_3$ 0.0038
$Fe_2O_3$ 0.0021

ii) Calculation of the Heating Value of the Coal

If the heating value of the coal is not supplied in the data, then it is calculated using Dulong's Law

$$\text{Heating Value} = [6.543 \cdot H + 424.62] \left[ \frac{C}{3} + \frac{H}{2} - \frac{O}{8} - \frac{S}{8} \right]$$

where the symbols stand for the wt. fraction of the appropriate element on a dry ash free basis.

$$= \left[6.543 \cdot \frac{(6.28)}{(1-0.118)} + 424.62\right] \left[\frac{75.91}{3} + \frac{6.28}{2} - \frac{11.72}{8} - \frac{4.58}{8} \right]$$

$$\left[\frac{1}{1-0.118}\right]$$

where 0.118 is the combined weight fraction of water and ash as determined from the ultimate analysis

$$= 14088 \text{ Btu/lb}$$

In this particular case we note that the calculated heating value overestimates the measured value of 13000 Btu/lb by 8.3 percent.

iii) Calculation of the Heat of Formation of Coal

The energy balance requires us to specify the enthalpy of each stream indicated in Figure 3. In this example the
enthalpies are specified from heat of formation data for each component in its natural state at 77°F. Such direct information is unavailable for coal, but the heat of formation of the organic material in coal can be indirectly calculated from the heating value using the formula (4)

\[-\Delta H_f = -\Delta H_C - N_C(\Delta H_{f_{CO2}}) - N_H(\Delta H_{f_{H2O}})/2-N_S(\Delta H_{f_{SO2}})\]

where \(N_C, N_H,\) and \(N_S\) represent the number of C, H, and S atoms in the formula for the hypothetical pure carbonaceous compound that has been selected to represent the coal. In essence the separate heats of combustion of the elements constituting the organic compounds minus the heat of combustion of the organic compound is used to calculate its heat of formation.

\[
\text{Ht. value} = \frac{13000 \text{ Btus}}{454 \text{ gms}} \times 0.252 \text{Kcal} \times 15.82 \text{ gms} = 109.87 \frac{\text{Kcal}}{\text{gm mole}}
\]

Therefore,

\[
(-\Delta H_f)_{C_1H_{x}O_{y}N_{z}S} = -109.87-(1)(-94.05)-\left(\frac{0.986}{2}\right)(-68.3)-(0.023)(-71.00)
\]

\[
= 19.45 \text{ Kcal/gm mole}
\]

The heats of formation of \(\text{CO}_2, \text{H}_2\text{O},\) and \(\text{SO}_2\) at 298°F used in the calculation are summarized in Table I which includes values for the other species involved in the analysis.
<table>
<thead>
<tr>
<th>Species</th>
<th>( \Delta H_f ) 298°K (Kcal/gm mole)</th>
<th>( S_{298°K} ) (cal/gm mole/°K)</th>
<th>M.W.</th>
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</thead>
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<td>( \text{CO}_2 ) (g)</td>
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<td>( \text{O}_2 ) (g)</td>
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<td>( \text{SO}_2 ) (g)</td>
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<td>12.01</td>
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<tr>
<td>( \text{CaCO}_3 ) (s)</td>
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<td>21.20</td>
<td>100.09</td>
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<td>-342.4</td>
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<td>56.08</td>
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<td>( \text{SiO}_2 ) (s)</td>
<td>-217.6</td>
<td>9.9</td>
<td>60.09</td>
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<tr>
<td>( \text{Al}_2\text{O}_3 ) (s)</td>
<td>-399.6</td>
<td>12.2</td>
<td>101.96</td>
</tr>
<tr>
<td>( \text{Fe}_2\text{O}_3 ) (s)</td>
<td>-196.5</td>
<td>21.5</td>
<td>196.2</td>
</tr>
</tbody>
</table>

iv) Calculation of the Absolute Entropy of the Organic Material in Coal at 298°K

The specification of the entropy of coal is a problem. The examination of the literature (7) indicates that for pure branched chain polycyclic compounds the entropy per C atom varies from 3.8 to 2.6 cal/mole°K/C atom as the number of C atoms varies from 11 to 25. As the carbonaceous material in coal is assumed to consist of a high mol. wt. compound of similar nature, an assumed value of 100 for the number of C
atoms produced an $S_{298^\circ K}$ value of 1.37 cal/mole/$^\circ$K/C atom on extrapolation.

Therefore \[
(S_{298^\circ K})_{C_1H_{OxN_yS_z}} = 1.37 \text{ cal/gm mole/$^\circ$K}
\]

v) **Calculation of the Properties of the Coal Stream**

Having calculated the coal composition in mole fractions in Part (i), the average molecular wt. of the coal is calculated as \[
MW = (0.0313)(18.02)+(0.9519)(15.82)+(0.0109)(60.09) \\
+(0.0038)(101.96)+(0.0021)(196.2) = 17.08
\]

The heat of formation of the mixture of water, organic compound and ash is calculated assuming ideal mixing

\[
-(\Delta H)_f^{298^\circ K} = (0.0313)(-68.3)+(0.9519)(-19.46)+(0.0109)(-217.6) \\
+(0.0038)(-399.6)+(0.0021)(-196.5) = 24.97 \text{ Kcal/gm mole}
\]

The absolute entropy at $298^\circ K$ is also calculated using ideal mixing

\[
S_{298^\circ K} = (0.0313)(45.11)+(0.9519)(1.37)+(0.0109)(9.9)+(0.0038) \\
(12.2)+(0.0021)(21.5) = 2.91 \text{ cal/gm mole/$^\circ$K}
\]

The stream properties as calculated by the computer program are summarized in Table II.

**MATERIAL BALANCE CHECKS**

i) **Check on Overall Material Balance (1 hour basis)**

\[
\text{Material in} = (F_1)(MW)_1 + (F_2)(MW)_2 + (F_3)(MW)_3
\]

where \(F\) stands for the flowrate in gm moles/hr and the subscript stands for the stream identification number. From the results of Table III we obtain:
Material in = (0.4031x10^5)(17.08)+(0.2582x10^6)(28.85)  
+ (0.1629x10^4)(100.09) = 0.8301x10^7 gms/hr  

Material out = (\dot{F}_4) (MW)_4 + (\dot{F}_5)(MW)_5 + (\dot{F}_6)(MW)_6 + (\dot{F}_7)(MW)_7  
= (0.2719x10^6)(29.73)+(0.2366x10^3)(1201) + (0.5976x10^3)  
(86.32)+(0.1704x10^4)(92.91) = 0.8298x10^7 gms/hr.  

% Error in overall material balance = \frac{(0.8298-0.8301)}{(0.8298+0.8301)} \times 100 = .036\%  

ii) Stoichiometric Balance Check (1 hour basis)  

For the case of a C balance we obtain  

\text{Gm atoms of C in} = (\dot{F}_i) C + (\dot{F}_c) 3  

where \( (\dot{F}_c)_i = (\dot{F}_i) \sum_{j=1} x_j (n_c)_j  

x_j \text{ is the mole fraction of substance j in stream i}  

(n_c)_j \text{ is the number of atoms of C per mole of substance j}  

\text{Gm atoms of C in} = (.4031x10^5)(0.952)(1.0)+(0.1629x10^4)  
(1.0)(1.0) = 0.400x10^5 gm atoms/hr  

\text{Gm atoms of C out} = (\dot{F}_c)_4 + (\dot{F}_c)_5  
= (0.2719x10^6)(0.1462)(1.0)+(0.2366x10^3)(1.0)(1.0)  
= 0.400x10^5 gm atoms/hr  

Similar stoichiometric balance checks were made for H, O, N, S, and Ca. The results are summarized in Table II below.
TABLE II

Material Balance Summary

<table>
<thead>
<tr>
<th>Elements</th>
<th>Gm Atoms/Hr In</th>
<th>Gm Atoms/Hr Out</th>
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<td>S</td>
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<td>Ca</td>
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</table>

ENERGY BALANCE - CALCULATION OF Q

The application of the energy balance to the model of Figure 3 yields the heat released \( -Q \) in Kcal/hr as

\[
-Q = \left( \frac{\dot{H}_1}{1} F_1 + \frac{\dot{H}_2}{2} F_2 + \frac{\dot{H}_3}{3} F_3 \right) - \left( \frac{\dot{H}_4}{4} F_4 + \frac{\dot{H}_5}{5} F_5 + \frac{\dot{H}_6}{6} F_6 \right) - W
\]

where \( \dot{H}_i \) is the specific enthalpy of stream \( i \) in Kcal/gm mole and \( W \) is the work done in Kcal/hr. Using values from Table II we obtain

\[
-Q = \left[ (-24.97)(0.4031\times10^5) + (0.00)(0.2582\times10^6) + (-288.4)(0.1629\times10^4) \right] - \left[ (-18.06)(0.2719\times10^6) + (0.00)(.2366\times10^3) + (0256.1)(.5976\times10^3) \right]
- 1769.24 k. watts \left( \frac{1}{1.1626\times10^3} \right) \text{Kcal/hrs/kwatts}
= + .24742\times10^7 \text{ Kcal/hr}
\]
ENTROPY BALANCE

The application of the entropy balance to the model of Figure 3 yields the entropy production $\dot{S}_p$ in cal/gm mole/$^\circ$K/hr. as

$$\dot{S}_p = (S_1 F_1 + S_2 F_2 + S_3 F_3) - (S_4 F_4 + S_5 F_5 + S_6 F_6) - Q/T_D$$

where $S_i$ is the absolute specific entropy of stream i in cal/gm mole/$^\circ$K and $T_D$ is the dump temperature assumed to be 298$^\circ$K in this particular case.

$$= [(2.91)(0.4031\times10^5)+(46.44)(0.2582\times10^6)+(21.20)(0.1629\times10^4)]$$

$$- [(46.58)(0.2719\times10^6)+(1.37)(0.2366\times10^3)+(11.85)(0.5976\times10^3)]$$

$$+ (16.68)(0.1704\times10^4)] + 0.24742\times10^7/298.15$$

$$= 0.8854\times10^4 \text{ Kcal/hr/$^\circ$K}$$

Lost work = $l\omega = T_D \dot{S}_p = W_I - W_A$

$$= (298.15)(0.8854\times10^4)$$

$$= 0.2640\times10^7 \text{ Kcal/hr}$$

CALCULATION OF EFFICIENCIES

Thermodynamic Efficiency

$$= \frac{W_A}{(W_A + l\omega)} = \frac{0.1521\times10^7 \text{ Kcal/hr}}{(0.1521\times10^7 + 0.2640\times10^7)}$$

$$= 0.3655$$

Thermal Efficiency

$$= \frac{W_A}{(\text{Heating Value})(\text{Coal Flowrate})}$$

$$= \frac{0.1521\times10^7 \text{ Kcal/hr}}{(109.97 \text{ Kcal/hr})(0.4031\times10^5 \text{ gm moles/hr})}$$

$$= 0.3436$$
The thermodynamic efficiency is seen to have a slightly higher value that that of the more commonly used thermal efficiency for the combustion reaction considered. This situation occurs because the entropy change $\Delta S$ for the combustion reaction is negative and proper credit is given in the calculation of the thermodynamic efficiency in contrast to the thermal efficiency which is restricted to the examination of the heat effects involved in the reaction.
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*See Figure 3 for identification*


BLACK MESA COAL SLURRY PIPELINES

by

Brymer Williams
Donald L. Katz

The Black Mesa coal slurry pipeline is mentioned because of the experience with continuous pumping of coal slurries. It extends 273 miles from near Kayenta, Arizona in the Navaho reservation to the Mohave Power Plant on the Colorado River just below Davis Dam. The plant is on the Arizona side of the river and is owned primarily by the Southern California Edison of Los Angeles but has three other partners in the operation of the two 790 megawatt thermal generating units. Coal, strip-mined by Peabody Coal Company is delivered by truck to the Black Mesa coal preparation plant. There it is crushed and slurried for entrance to the pipeline. There are three pumping stations along the 273 mile route across Arizona in addition to the entrance station. Pressures as high as one thousand pounds per square inch are used and the pipe is graded according to the pressure level which varies with the terrain. The entire system was built for about $35 million. It was engineered and built by Bechtel Inc. using data obtained from Consolidation Coal Co. plus information from additional tests with the Consolidation pipeline in Northeastern Ohio. There have been no serious interruptions with operation of the line. Twice the line stopped and it was necessary to locate the ensuing short plug and remove it, but this was done within 48 hours in each instance.
Approximately 660 tons per hour of coal are transported through an 18 inch diameter pipeline as an aqueous slurry of 46.4 weight per cent solids. Figure 1 shows the coal grinding and slurry preparation circuit. A three step crushing and grinding process prepares the coal to pass a 14 mesh screen, with a considerable fraction of -320 mesh. The design and operation know-how are proprietary, but the following items are noted as being of importance or of interest:

1. The slurry concentration is set within narrow limits, to retain flow characteristics while minimizing water to be handled at the power plant end. Limits are 44-48 weight percent solids, but are actually held at 46.5±0.1%

2. The size range of -14+-325 mesh is a compromise between pipeline and power plant requirements.

3. Slurry pumping is by Wilfley centrifugal pumps for low heads (up to 100 psi) and Wilson-Snyder positive displacement pumps for pipeline injection, up to 1500 psi. There are two parallel pumps in service at any one time, each pumping 2100 gallons of slurry per minute.

The service record of the pumps is reportedly excellent, but with a schedule of replacement parts. To ensure 100 percent on-stream time a third, standby, pump is installed for each service.
Figure 1. Schematic Flow Sheet for Coal Preparation Plant of Black Mesa Pipeline Company
4. The operators have developed effective techniques for restarting the pipeline after unavoidable shutdowns caused by such problems as power failures.

5. As an extra precaution, the slurry circles the plant in a 1000 foot loop where its behavior is monitored before leaving the plant limits.

Bibliography

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Aude, T.C., N.T. Cowper, T.L. Thompson and E.J. Wasp, (Bechtel Corp.), Chemical Engineering, June 28, 1971
ANN ARBOR—A team of University of Michigan engineers has undertaken an intensive, seven-month study to determine which of the existing methods for converting coal to clean fuels are the most likely to prove commercially feasible in the near future.

"The Board of Directors of the Electric Power Research Institute (EPRI) has approved a budget of $150,000 and contract negotiations are in progress," said project coordinator Donald L. Katz, the Alfred Holmes White University Professor of Chemical Engineering at U-M.

"Our goal is to recommend to EPRI those processes whose development warrants acceleration through the Institute's support," Prof. Katz said. EPRI is a national effort by the electric power industry, both public and private, to sponsor energy research of common interest and importance.

Joining Katz in the project are Dale E. Briggs, John E. Powers and M. Rasim Tek, professors of chemical engineering; Brymer Williams, professor of chemical and metallurgical engineering; Edward R. Lady, professor of mechanical engineering, all at U-M. Walter E. Lobo, independent consulting engineer, will be project consultant.

"Over the next few decades, clean fuels derived from coal will play a key role in helping electric utility companies handle the ever increasing demand for energy," Katz explained, adding that "clean" coal is that which has had most of the sulfur, a serious pollutant, removed.

"However, coal can be converted to clean fuels by either gasification, liquefaction, or solvent extraction," he continued, "and there are currently ten major coal gasification processes and several for liquefaction and solvent extraction."

(more)
In view of the steadily mounting energy crisis, the U-M chemical engineer pointed out, EPRI cannot afford either the time or the money required to see that every coal conversion process be fully explored.

"By analyzing and comparing the different processes, we hope to assist EPRI in focusing and intensifying its research so that the best processes can be realized in time to help alleviate the energy dilemma," Katz emphasized.

He said that all conversion processes which satisfy the environmental standards established by the Environmental Protection Agency will be considered. He said a comprehensive method for evaluating the different processes will be developed by the U-M team.

"We will be interested in such things as the percentage of energy remaining after cleaning, the difficulty in the physical handling of the fuel, the simplicity of the equipment required, the type of waste left after cleaning and problems associated with its disposal, the overall economics, and other factors," he observed.

Initially, the group will examine reports describing the processes and review papers on the subject prepared by the EPRI Task Force of Utilization of Coal. But it also plans to conduct extensive interviews with engineers and those managing the organizations sponsoring or offering coal conversion processes to the industry.

"The engineers on the project have considerable experience in industry," Katz noted, "and when it comes to handling thermodynamics, rate processes, or other technical aspects of these various processes, they will be fully capable of putting the information gathered into proper perspective." The final report prepared by the group for EPRI will be presented in January 1974.

Commenting on the project, David V. Ragone, dean of the U-M College of Engineering, said, "Nuclear power is not yet ready to assume the major portion of power production. And shortages of natural gas and limited domestic crude oil supply have put a new emphasis on coal. Therefore, I am highly pleased that our engineers are participating in a project that ultimately could help our country meet its energy needs through the environmentally sound consumption of coal."