SECTION 9. RESEARCH OPPORTUNITIES

The study of the literature and the discussions with research and development workers during field trips, have resulted in an excellent survey of the current "state of the art" of coal gasification. Techno-economic evaluation of the resultant data has indicated various areas wherein further research could lead to substantially improved coal gasification processes. These research opportunities have been summarized below in the following order: (a) synthesis gas processes, (b) fuel gas processes, (c) special gas processes, and (d) miscellaneous.

A. Production of Synthesis Gas

1. Two-stage Super-pressure Entrained Gasifier (Process 58): From the evaluation of potential gasification processes, the most promising route to improve the economies of synthesis gas processes appears to be by a combination of the favorable features of suspension gasification systems with those of fixed-bed or fluidized-bed systems, such as that proposed in the Two-stage Super-pressure Entrained Gasifier.(67) Evaluation of such a unit projected to full-scale commercial operation indicates that pipeline gas from coal in a 250 MM scf per day plant would cost about 5 cents less than by other synthesis gas processes evaluated.(68)

At first, experimental work on a laboratory scale is necessary for the exploration of the unknown factors of the reactions in the second stage of the process. Equipment suitable for these investigations is shown diagrammatically in Figure 9-1.

Next, data from these laboratory studies would be used to design an integrated continuous flow pilot plant, in which char is gasified in the first stage and fresh coal in the second stage. Such a pilot plant would then be operated to substantiate and further improve process equipment and design as well as to furnish data needed for evaluation and extrapolation of the total process to commercial scale.

2. Catalytic Steam Methanation Gasification (Process 65): Catalytic Steam Methanation Gasification (69) offers potential for the direct production of high Btu gas from coal. The results from the evaluation of the process as projected to full-scale commercial operation indicates that, if the process were successfully developed, the cost of high Btu pipeline gas would be significantly lower than that by other selected processes.(70)

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(67) See Process 58, Table 3-2, and Appendix 3.5.

(68) See Section 6.

(69) See Process 65, Table 3-2, and Appendix 3.5.

(70) See Section 6.

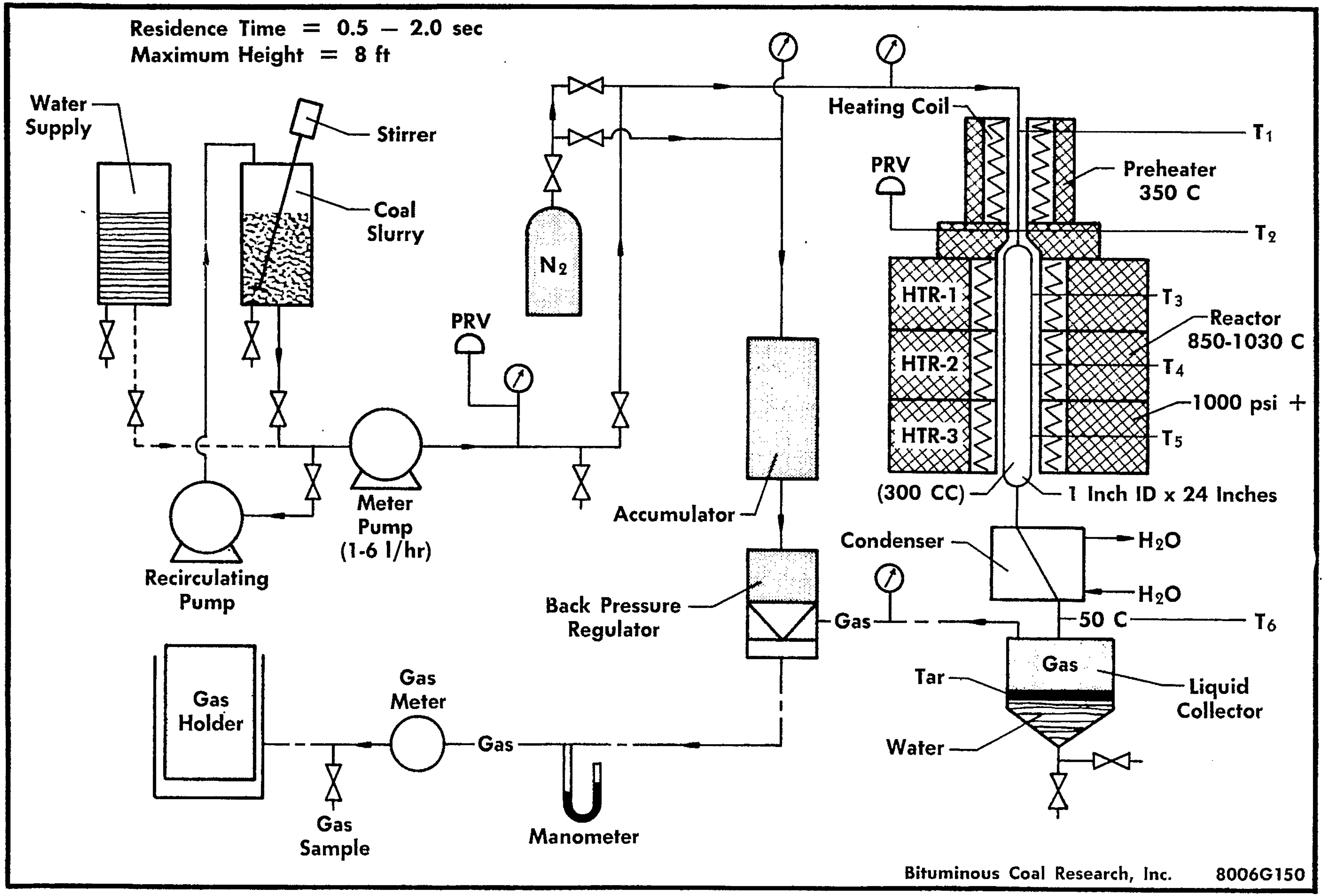

Figure 9-1 Schematic Diagram of Bench-scale Equipment for Two-stage Gasification Studies

In view of this potential, continued investigation of materials as catalysts for the steam methanation reaction is considered worthwhile.

Experimental work in simple bench-scale autoclaves would permit a screening of the catalytic properties of various elements and compounds as well as an evaluation of various methods of applying these materials. In such work, attention should be given, not only to catalysts that are impregnated on the coal, but also to catalysts that are applied in solid form of a different size than that of the coal.

Investigation of experimental conditions that will make the coal material mobile in presence of high pressure reactants is indicated, together with a study of catalysts that accelerate the gasification of the volatile matter of coal and of fixed carbon. Additives that increase the reactivity of the remaining carbon and catalysts that accelerate methane formation should be tested. The residence time required will indicate whether a fluidized bed of coal is needed, or whether such short duration as that obtained in entrained gasification is sufficient. Ultimately, the bench-scale investigations in autoclaves should be followed by experiments in continuous systems.

A diagram of bench-scale equipment proposed for such catalyst studies is given in Figure 9-2.

Successful development of catalysts for this reaction should also result in an additional decrease in the reaction temperature in the second stage in the proposed two-stage super-pressure gasification system, and thus an additional improvement in the overall process should be possible.

3. <u>Coal Composition</u>: The significance of coal petrography in areas of carbonization, combustion, preparation, and mining are well documented in the literature. The acceptance of coal petrography as an analytical tool was made possible through the establishment of specific correlations between coal composition and reflectance rank which enable the prediction of carbonization, combustion, mining, and preparation characteristics of coal.

However, information concerning the prediction and evaluation of the gasification potential of coal by microscopic means has not been developed.

In a research program directed toward the development of such information, coal, or fractions of coal, could be systematically analyzed with the use of the hot stage microscope in combination with a gas chromatograph to determine whether methane formation preferentially occurs with any particular coal maceral. As the coal samples are thermally treated, the evolved gases could be conveyed to a chromatograph for quantification. Should specific macerals exhibit exceptionally large yields of methane, attempts could then be made to concentrate, by standard methods, these highly desirable, methane forming coal entities for subsequent testing in the proposed gasification systems.

Thus, there is an excellent opportunity for making a significant contribution to the present coal gasification technology. A study primarily concerned with the construction of a basic research framework derived from microscopic information should result in a procedure for selecting those coals, or

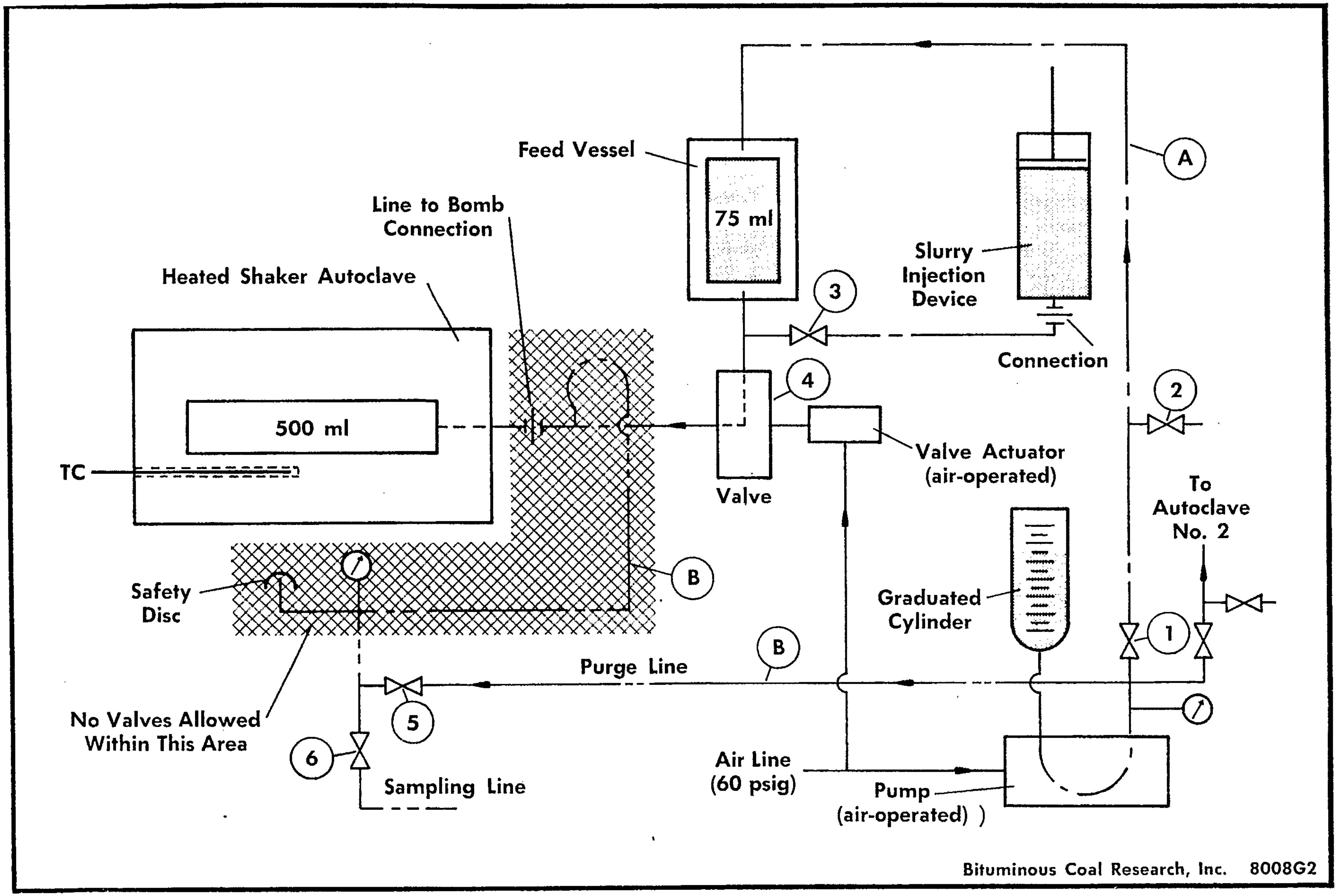

Figure 9-2 Flow Diagram for Catalytic Gasification Experiments in Laboratory Autoclave

fractions of coal, which inherently possess optimum gasification potential, particularly for a two-stage gasifier.

4. Determination and Prediction of Carbon Reactivity: Any successful system for complete gasification of coal requires a rapid, relatively complete reaction of the char residue produced in the initial gasification or devolatilization step. An increased reactivity of the residue would not only accelerate any possible carbon reactions, but also permit alteration of the environment to favor one reaction over another.

For example, carbon in the presence of steam and hydrogen react to decompose the steam and/or to form methane. Higher temperatures favor steam decomposition, while high pressures and low temperatures favor methane formation. If the carbon is relatively unreactive, lowering the reaction temperature to favor methane production would result in the production of some additional methane, but would leave large amounts of unreacted carbon. However, if the carbon is highly reactive, lowering the reaction temperature could result in significant methane production, and, in addition, since some of the $C + O_2 \rightarrow CO_2$ exotherm would be replaced by the $C + 2H_2 \rightarrow CH_4$ exotherm, less oxygen would be required.

Various factors have been reported as contributing to the reactivity of coal and char under gasification conditions; they include: source of coal, hydrogen content, surface area, degree of crystallinity, and incomplete graphitization. However, present information is too meager for an adequate understanding of this major process variable.

In the production of pipeline gas, increase of the amount of methane in the gas from the initial coal gasification step leads to a decrease in the cost of the final product gas. Thus, a reactivity as high as possible becomes not only desirable, but also essential to the successful development of practical gas generating systems.

The development of a procedure for measuring and predicting the reactivity of a given feedstock would thus represent a major contribution to gasification technology and practice. Such data would be valuable not only in the design of gasification equipment to handle a given feedstock, but also in the delineation of degree and kind of preparation and beneficiation of various feed materials that would be required for given equipment and operating conditions.

5. High Pressure Thermal Softening Properties of Coals: In the present evaluations, it has been assumed that Pittsburgh seam coal with its high swelling and caking properties could be used directly in fixed-bed and fluidized-bed gasifiers without any pretreatment. The information available on the effects of pressure on the plastic properties of coal is meager, and there is a need for bench-scale techniques for the measurement and prediction of the plasticity and swelling of any given coal at the high pressure of operation now being proposed.

Information on the effects of pressure, temperature, and atmosphere on the plasticity and swelling of coals under the conditions that exist in a coal gasification system would be of considerable value in the design of new and improved fluidized- and fixed-bed gasifiers.

A conceptual design of a high pressure coal plastometer is outlined in Appendix 9.1, together with suggested initial experiments.

6. Fixed-bed Pressure Gasification of Highly Swelling Coals: The Lurgi pressure gasification process, at first, used brown coal as feed. Brown coal retains its size well during the descent in the fuel bed. Later, it was found that slightly caking bituminous coals can also be gasified if a suitable stirrer is used in the part of the fuel bed in which the coal is in the plastic stage. The experience with such coals is that in this zone of plasticity of the fuel, a coke is formed with a size consist almost independent of that of the original coal. The question remained open for some time whether highly swelling coals can be appropriately gasified in a Lurgi gasifier.

At the initiative of the U.S. Bureau of Mines, a test was made at the commercial Lurgi plant in Dorsten, Germany, with 100 tons of a highly swelling American coal. This test was too short to give good performance data; however, it did indicate that gasification of this coal admixed with 20 percent ash posed no problem. The Lurgi company expressed the belief that for such a coal a somewhat longer gasifier shaft will be required.

Operation of a fixed-bed pressurized gasification pilot plant could make a valuable contribution to the solution of the problem of gasifying highly swelling coals in such units. The object of such a research program would be to obtain data for the performance when gasifying a highly swelling coal as compared with a nonswelling coal, with and without the addition of recycle ash, and preferably with an appropriate stirrer.

The successful development of procedures for the handling of highly caking coals in fixed-bed gasifiers would remove a major deterrent for the use of most American bituminous coals in such units.

7. Multi-stage Fixed-bed Gasifier: The only commercially available coal gasification process operating at elevated pressure is the Lurgi dry-ash process. To avoid melting of the ash, use of a high steam-oxygen ratio is required. This leads typically to a steam decomposition of only 25 percent and to an exit gas containing 42 percent water vapor. Since the total gas volume at the gas exit determines the capacity of the gasifier, an increase in throughput is possible if a higher steam decomposition can be attained using lower steam input. This is one of the reasons for the higher throughput of the slagging gasifier.

A similar high throughput can also be expected with dry-ash operation. If oxygen or oxygen-steam mixtures are added at one or more additional levels above the grate, the temperature of the gas at these points will be decreased below that near the grate because of the endothermal carbon-steam reaction. A proposed multi-stage unit designed to take advantage of these effects is shown schematically in Figure 9-3.

Assuming that steam decomposition in such a system is increased from 25 to 80 percent, an increase in coal throughput and gas production of 62 percent should be possible, together with steam addition being reduced by 68 percent. Also, since less steam must be heated to elevated temperatures, a decrease in oxygen consumption should be expected.

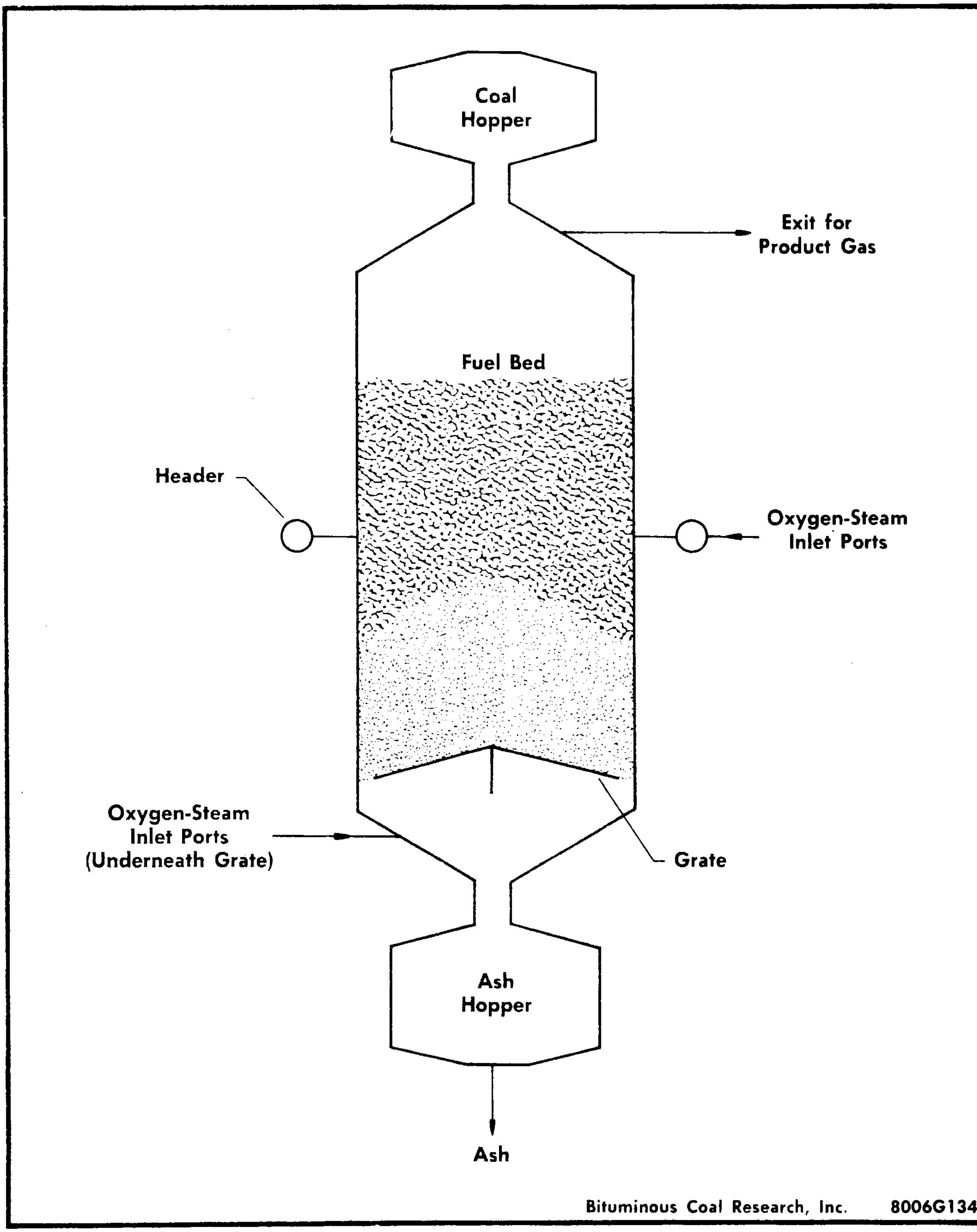

Figure 9-3 Diagram of Conceptual Multi-stage Fixed-bed Gasifier

This possibility appears worthwhile for further study in an existing pilot plant. An objective of such work would be to determine first the feasibility of attaining the suggested improvement with suitable nozzles, and second, the modification in bed height, if any, that may be needed to attain the proper gas exit temperature.

8. Coal Feeding Device: The cost of feeding coal into a pressure gasifier increases significantly with increases in pressure. It has been estimated that a positive displacement feeding device for pulverized coal at 70 atm pressure will reduce significantly the power requirement as well as the capital investment for gas compression.(71)

The first step in the further investigation of a new conceptual feeding device based on this principle (72) should be a detailed theoretical evaluation and model experiment program followed by a design study to develop a more accurate estimate of the cost. Then, if the costs are indicated to be favorable, a one cylinder experimental prototype unit should be built and tested with coal. For the design of an experimental unit, the services of a company with experience in the construction of special high pressure pumping units should be used. Data and results of this unit should then form the basis for design and construction of commercial size machines. Wherever possible, the experience (73) of others in the pumping of slurries of coal or ores in water should also be applied to the problem.

The successful development of such a coal feeding device would represent a major contribution not only to development of the Two-stage Super-pressure Entrained Gasifier, but also to high pressure gasifiers now being investigated by others, and to the pneumatic transport of fine coal in pipelines.

B. Production of Fuel Gas

1. Wellman-Galusha IFE Gas Producer: The literature survey and field trips have shown that basically there are two different gas producer systems with specific properties and applications:

(1) The one-stage producer, as built by Wellman-Galusha, is an efficient, low cost unit for the production of hot, raw producer gas from a great variety of coals including caking coals.(74) Because of the content of suspended matter, this gas cannot be used for operations that require: (a) dust free gas, or (b) continuous operation without shutdown for gas conduit cleaning at three to four month intervals. Cooling of the gas is also necessary if clean gas is needed; this increases the investment and operating cost considerably.

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(71) See Appendix 4.1.

(72) See Appendix 4.2.

(73) Constantini, R., "Feasibility of long distance solids pipelines," Mining Congr. J. 49 (1), 42-6 (1963).

(74) See Process 32, Table 3-1, and Appendix 3.5.

(2) The two-stage producer, as built by the International Furnace Equipment Company in England, produces a hot gas that does not have the limitations in use given for the hot raw gas from the one-stage producer.(75) Long operation of more than one year without conduit cleaning and contamination of the product to be heated has been proven. Cooling and cleaning of the gas is less costly than for gas from the one-stage producer.

The drawbacks of the two-stage producer are: (a) somewhat higher cost, and (b) inability to handle caking and swelling coals.

Discussions have indicated that designs might be found which would combine the insensitivity of the one-stage producer to coal caking with the greater cleanliness of gas from the two-stage producer. Development of such a producer would greatly extend the area in which producer gas can be used in competition with natural gas. Appropriate steps to initiate such a program appear worthwhile.

2. Traveling Grate Stoker Gasifier: Traveling grate cokers are being used to produce a small size "chemical" coke. The manufacture of high strength, large size metallurgical coke is under development. The coproduct of this process is steam and/or power and, thus, combination of a coal gasification plant with a power plant would be required. In present traveling grate and rotary kiln coking plants, the cost of the coke is much higher than the price of the coal used. A study whether sufficient cost reduction can be attained in an integrated large scale coking gasification power plant is advisable. Development work in this area is being undertaken by others to open the blast furnace coke market to these coking processes.

C. Production of Special Gases

In addition to the production of synthesis gas and conventional fuel gases, the gasification of coal can be a means for the production of other useful gases. In the present evaluations, consideration has been given to certain processes which have been brought to the attention of the Survey Group. They include the following:

(a) Complete gasification of char from the hydrogasification of coal to produce a steam iron reducing gas for manufacture of hydrogen required for the initial hydrogasification,

(b) Complete coal gasification combined with a gas turbine as the only means of power generation, and

(c) Partial coal gasification as part of a power plant cycle.

1. Two-stage Fluidized Super-pressure Gas Producer for Steam-iron Reduction Gas: An improved steam-iron process for the generation of hydrogen has been proposed for operation in conjunction with the hydrogasification of coal. Results from recent evaluations indicate that reductions in cost

(75) See Process 33, Table 3-1, and Appendix 3.5.

can be achieved by:

(a) Operating the steam-iron process at hydrogasification pressure,

(b) Production of producer gas also at hydrogasification pressure for use without cooling as the reducing gas in the steam iron-process, and

(c) Use of the char residue from hydrogasification of coal as the feedstock for the gas producer.

For the efficient operation of the steam iron process, a high ratio of $(CO + H_2)$ to $(CO_2 + H_2O)$ is required. Production of such a gas from the hydrogasification char is indicated to be possible.

To attain high carbon utilization in such a process, two-stage operation such as that of the proposed Two-stage Fluidized Super-pressure Gas Producer (76) will be needed.

For estimating the performance of the slagging first stage, data from the Ruhrgas Vortex gasifier and from pilot plants used in suspension gasification of coal at elevated pressure have been used.(77) Data needed for an estimate of the performance of the second stage, however, will have to be developed on an experimental program, beginning with bench-scale equipment. Operating data and results from these studies would then be used as a basis for design of a pilot plant with integrated first and second stage.

The successful development of an inexpensive process using air and char to produce the required feed gas for the steam-iron process would represent a major contribution to the development of the hydrogasification process and to the overall objective of developing practical gasification systems based on coal.

Thus, development of the needed information concerning the performance of the second stage of the two-stage fluidized super-pressure producer represents a real opportunity for further research and process development.

<u>2. Complete Coal Gasification for the Generation of a Gas Turbine Fuel</u>: Coal gasification produces a combustible gas which can be more readily purified and freed of dust. Such gas could be combusted with air and used to drive a turbine.

The gasification of coal in a fixed-bed, Lurgi type process appears to be suitable for this purpose.(78) Evaluation of the data obtained from Lurgi for the gasification of coal with air at elevated pressure in the present study, has indicated that the gas so produced is obtainable at a comparatively

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(76) See Process 46, Table 3-2, and Appendix 3.5.

(77) See Process 35, Table 3-1, and Appendix 3.5.

(78) See Process 37, Table 3-1, and Appendix 3.5.

low cost.(79) Further techno-economic evaluation of this process for the generation of power is suggested, since it promises to lead to economical power production in the 10,000 kw range. A fixed-bed process seems to be especially suitable for this purpose, since the product gas is obtained at a comparatively low temperature of about 1000 F where it can be freed of dust without the need for very complex equipment.

However, a study of coal gasification processes as a source of fuel for power generation should not be limited to the 10,000 kw size range nor to fixed-bed processes. It is indicated that progress in the development of low cost high efficiency gas turbines is rapid; thus, there is need for the evaluation of coal gasification processes using air for the generation of a gas turbine fuel taking into consideration the following items: (a) removal of dust from the gas, (b) removal of and recovery of hydrogen sulfide to minimize air pollution, and (c) investment and operating cost.

An area that is expected to be especially fruitful is the study of processes that permit removal of dust and hydrogen sulfide without the need of cooling of the gas.

3. Partial Gasification of Coal as Part of a Power Plant Cycle: The gas turbine using gaseous or liquid fuels has found wide application for the generation of power in such applications as gas compression. Use of gas turbines in the field of large, central base load power plants is receiving increased attention in both United States and abroad.

The combined use of gas and steam turbines in large coal-fired power plants represents another approach which promises an increase of about 4 percent in overall plant efficiency without an increase in investment cost. This would be accomplished by using gas obtained from the readily gasifiable portions of the coal as fuel for the gas turbine, and feeding the residual char, together with the gas turbine effluent, into the steam boiler.

Advantages of such an approach to power generation are discussed in further detail in Appendix 9.2 together with one suggested gas steam turbine combination based on a pressurized fluidized-bed gasifier.

Initially, a research program to establish the full potential of this approach to power generation should include the following:

(a) Development of material and heat balances based on data available for European plants,

(b) Development of flow sheets and cost estimates for the generation of gas by partial coal gasification under pressure to produce a gas turbine fuel by the most economical process,

(c) Evaluation of the merits of this process in power plant cycles in cooperation with equipment builders to indicate the most promising

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(79) See Section 8.A

direction for further research, and

(d) Delineation of a specific program of experimental work on that aspect of partial gasification appearing to be most promising.

D. Miscellaneous

1. Raw Gas Methanation Catalyst: The production of pipeline gas by coal gasification includes a methanation step. The present methanation catalyst is poisoned by sulfur compounds. Therefore, the synthesis gas, before entering the methanation step, must be cooled for removal of sulfur compounds, then reheated for the methanation reaction, and finally cooled again before entering the pipeline. The cooling and reheating equipment for a 250 MM scf per day pipeline gas plant cost about $2 million, or the equivalent to about 0.5 cent per MM Btu in the gas. This cost would be eliminated if a low cost methanation catalyst were available which would retain its activity in the presence of H_2S, H_2O, and CO_2 at the 1050 psi total pressure proposed for the super pressure gasification.

An even greater saving might be achieved by a methanation catalyst that methanates the raw gas and adjusts the shift reaction equilibrium as it methanates. This could mean an additional saving by the elimination of a separate shift reactor although the heat of reaction removed there would then appear and would have to be removed in the methanation reactor. Also, it may be remarked that the presently assumed cost for replacement of the Raney nickel methanation catalyst is 0.5 cent per MM Btu pipeline gas.

Tungsten based catalysts are known to be active for the methanation reaction. However, a low cost methanation catalyst that has high and long time activity in the presence of H_2S, H_2O, and CO_2 is not available. Development of such a catalyst appears promising and worthwhile.

2. Combined Gasifier-boiler for Control of Sulfur Oxides in Stack Gases: Present methods being developed for controlling sulfur oxides in stack gases are based on their removal from the final stack gas.(80) Total gasification of coal, especially pressure gasification, prior to combustion may offer a more economical method for sulfur oxide control--the sulfur would be present mainly as hydrogen sulfide and the gas volume would be significantly decreased.

It is suggested that pulverized coal be completely gasified under pressure in large plants using the principle of the atmospheric pressure 100 tons per day Ruhrgas Vortex unit. The tar-free gas so produced would be cooled by conventional boiler surface to a suitable temperature for fly ash and hydrogen sulfide removal. The resultant clean gas with 70 to 100 Btu/scf would then be burned in conventional gas-fired equipment. A suitable cycle is shown in Figure 9-4.

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(80) Frankenberg, T. T., "Removal of sulfur from fuels and products of combustion," ASME 64-WA/APC-2 (1964).

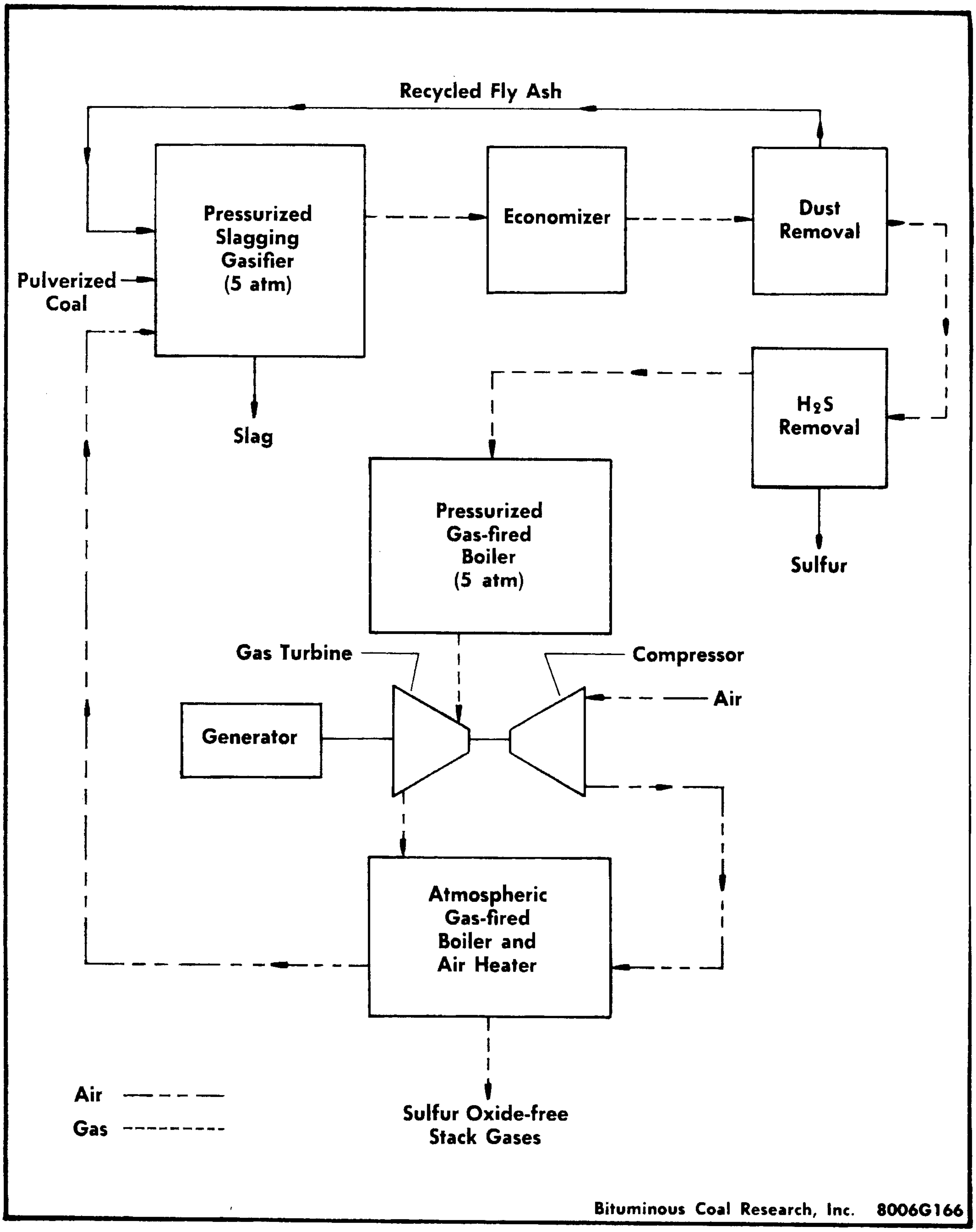

Figure 9-4 Combined Gasifier-boiler for Control of Sulfur Oxides in Stack Gases

Removal of hydrogen sulfide from gases is a well developed art; conventional techniques are available for removing hydrogen sulfide to a concentration of a few grains per 100 scf. Where gas scrubbing is involved, final dust removal might well be incorporated to eliminate an electrostatic precipitator. High temperature techniques are attractive.

Methods based on injection of inexpensive limestone or dolomite based products have been reported.(81) Also a process using hot iron oxide (82) and one based on catalytic conversion of hydrogen sulfide and sulfur dioxide to elemental sulfur (83) have been developed on a pilot scale.

The potential advantages of the proposed system are:

1. The gas volume to be processed is approximately 10 percent of that of the final flue gases.

2. Hydrogen sulfide should be more readily removed under pressure than sulfur oxides are at atmospheric pressure, and the technology is more fully developed.

3. The more efficient cycle based on gas turbines and pressurized boilers, and perhaps eventually MHD generators, should offset the cost of additional equipment.

4. Gas firing should allow a more efficient steam cycle than that of present coal firing practice. Coal ash corrosion, not thermal resistance, is the limiting factor on superheat and reheat temperatures in coal-fired units.

Establishment of the feasibility of this proposed method of combined power generation and sulfur removal would provide an attractive approach to a solution of one of the industry's major problems in air pollution control. For this, a techno-economic engineering evaluation and cost study in consultation with operators, designers, and manufacturers of steam electric power plants would be required.

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(81) Wicket, R., "Experiments for the sulfur removal before and after the burner to reduce SO_2 emission," Mitteilungen Der VG B 83, 74-82 (1963).

(82) Reeve, L., "Desulfurization of coke oven gas at Appleby-Frodingham," J. Inst. Fuel 31, 319-24 (1958).

(83) Audas, F. G., "A continuous process for the removal of hydrogen sulfide from industrial gases," Coke Gas 13, 229-34 (1951).