

Figure 18. The Effect of Temperature and Bed Depth and Gas Velocity on Ash-Collection Efficiency

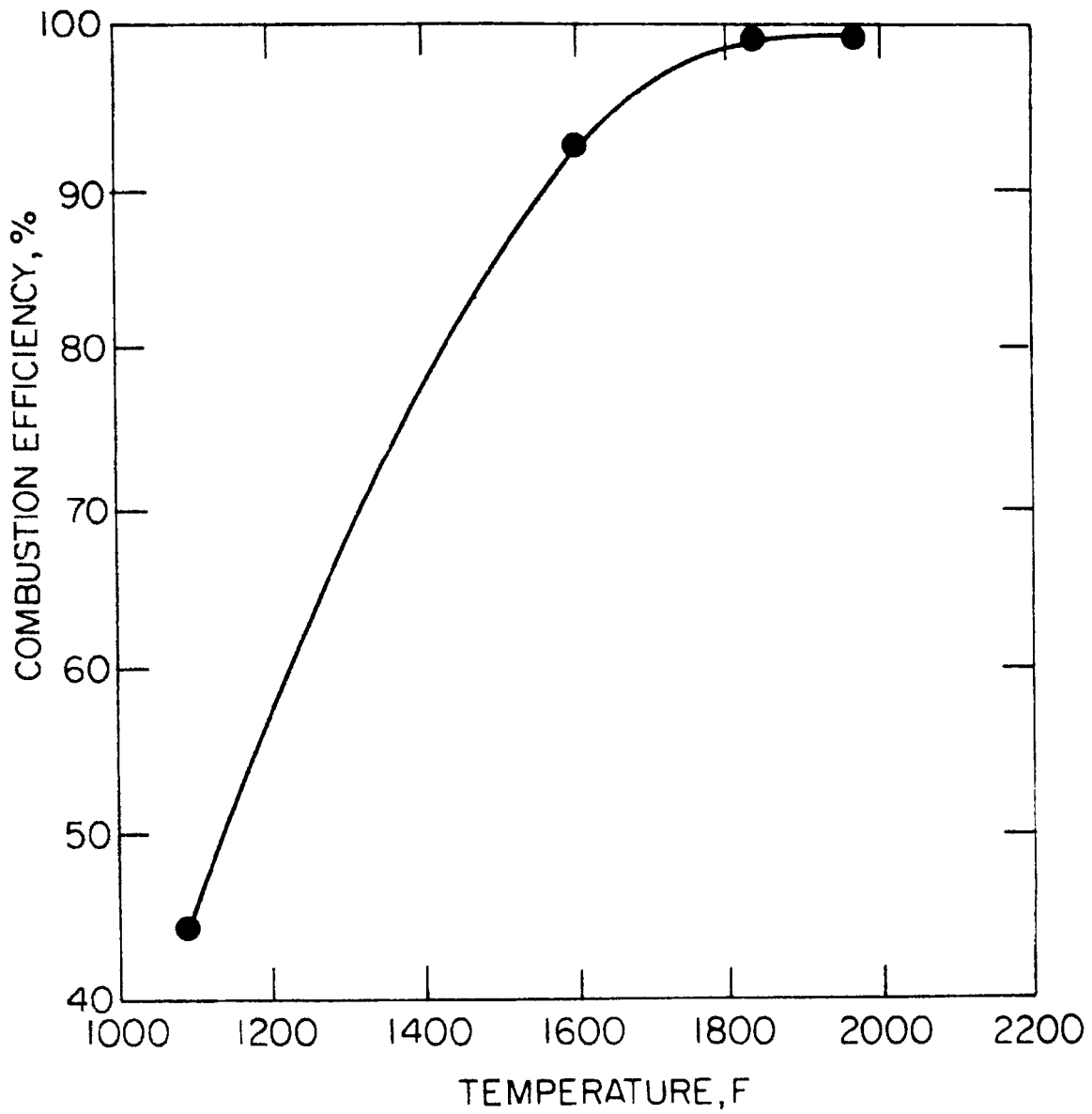


Figure 19. Combustion Efficiency as a Function of Temperature in the Fluidized-Bed Coal Burner

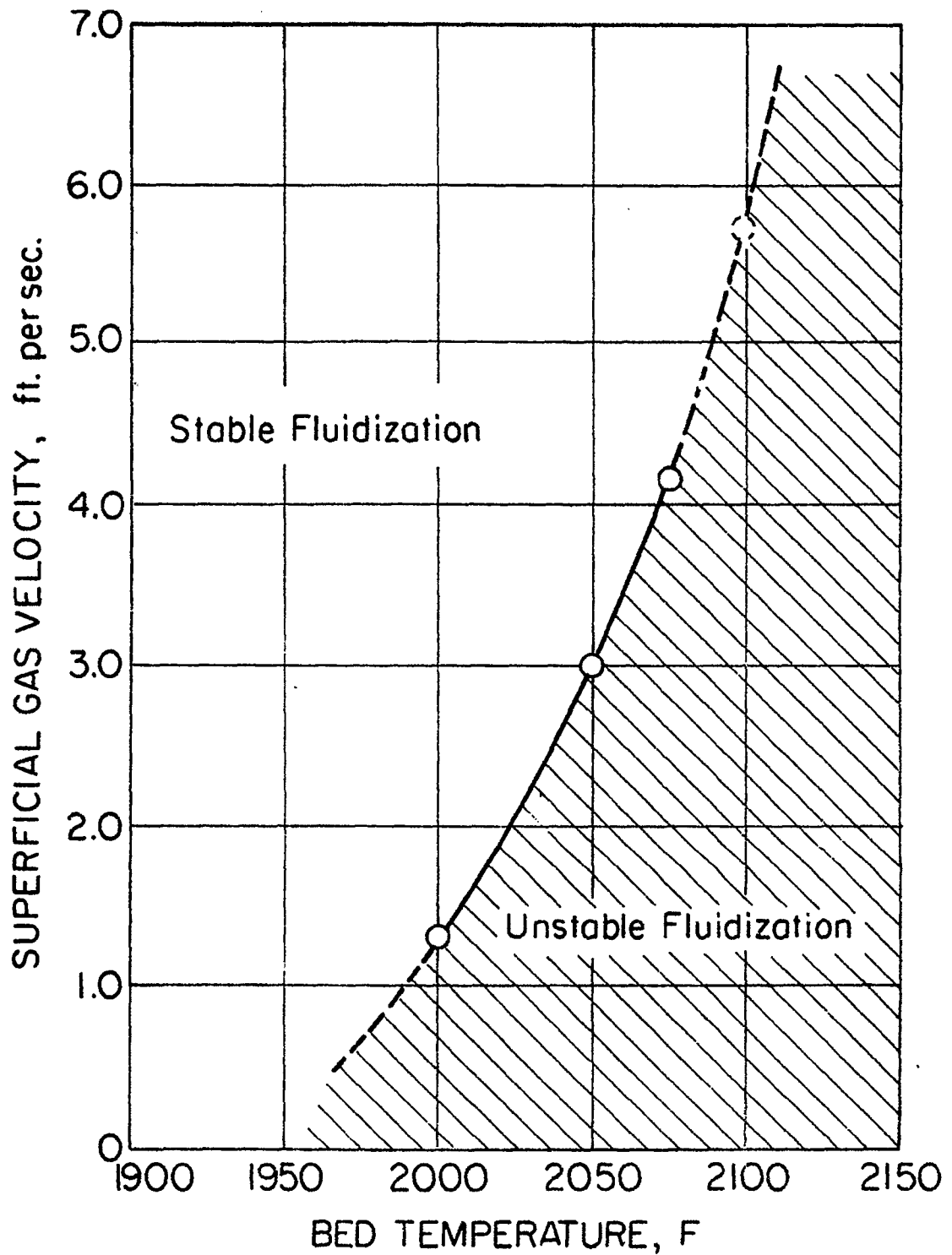


Figure 20. Stability of the Self-Agglomerating Fluidized-Bed Coal Burner as a Function of Temperature and Gas Velocity

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<sub>2</sub> 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

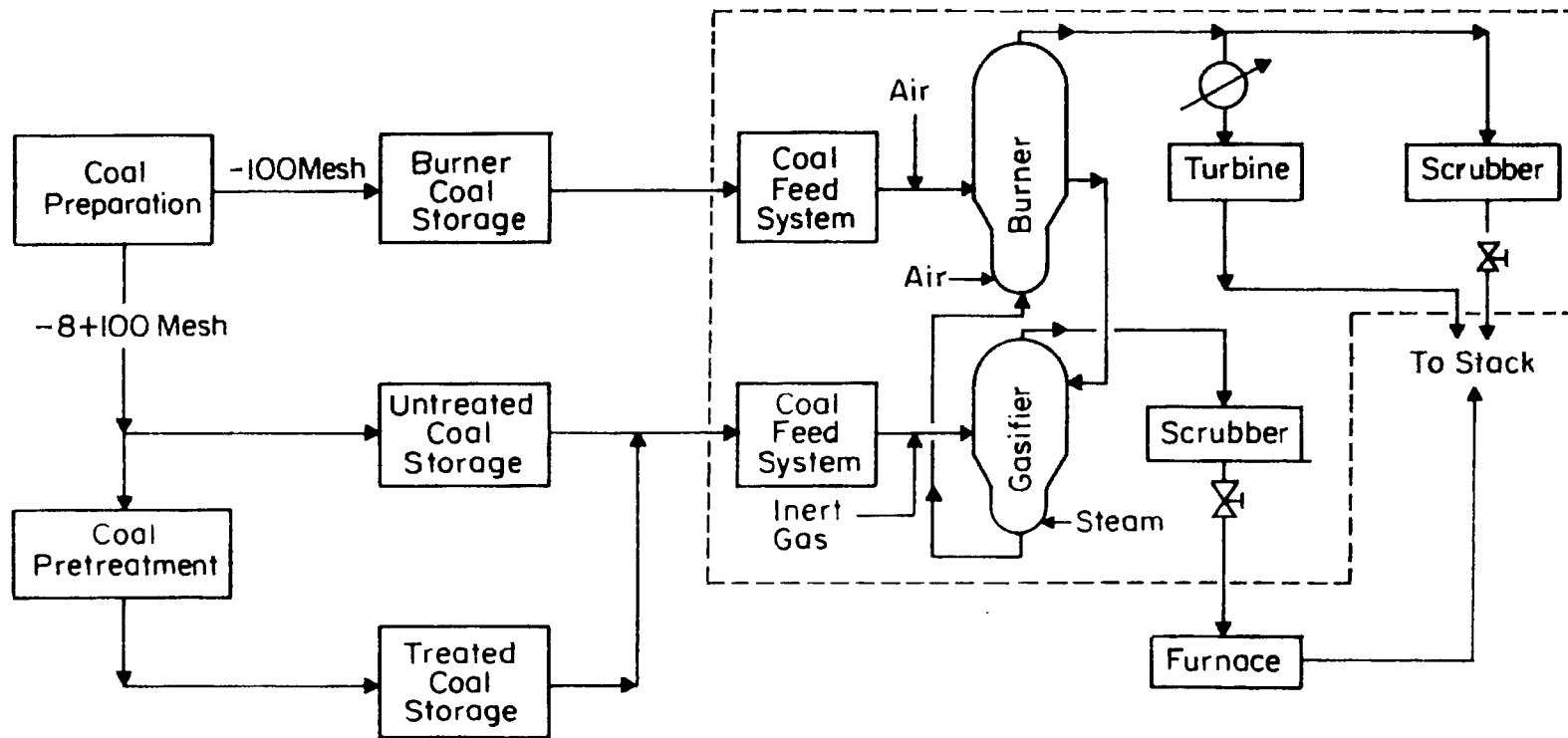


Figure 21. Block Diagram of Battelle Coal Gasification PEDU

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

## THE CITY COLLEGE OF THE CITY UNIVERSITY OF NEW YORK - SQUIRES

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 myriad 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

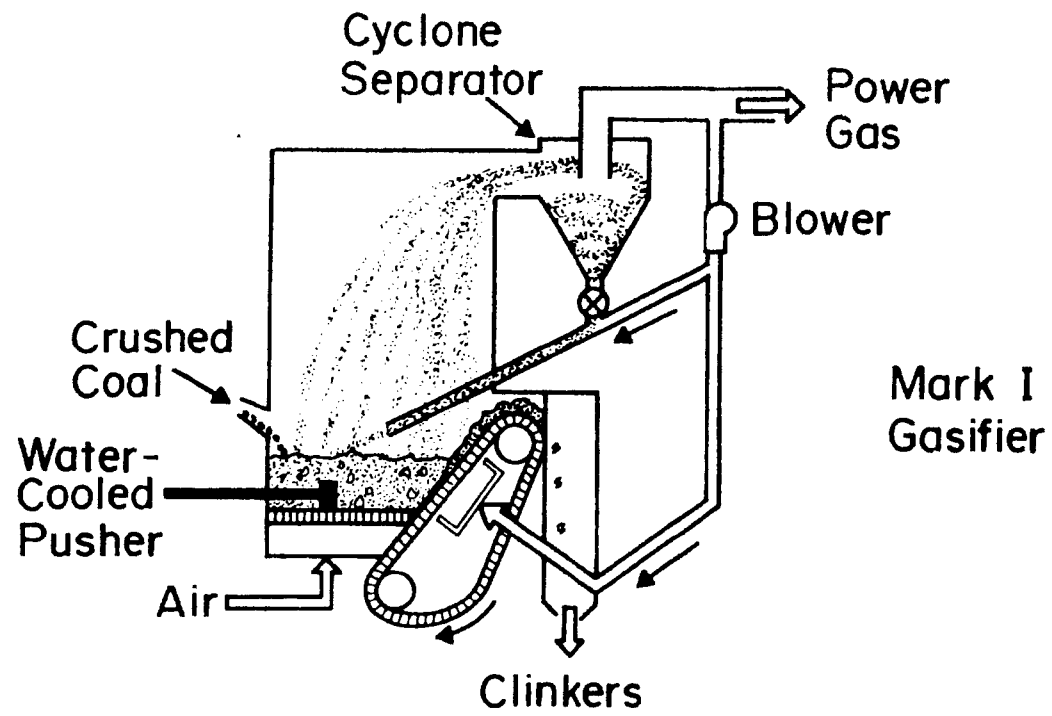


Figure 22. The Mark I Coal Gasifier

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 volatile 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 devolatilized (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.

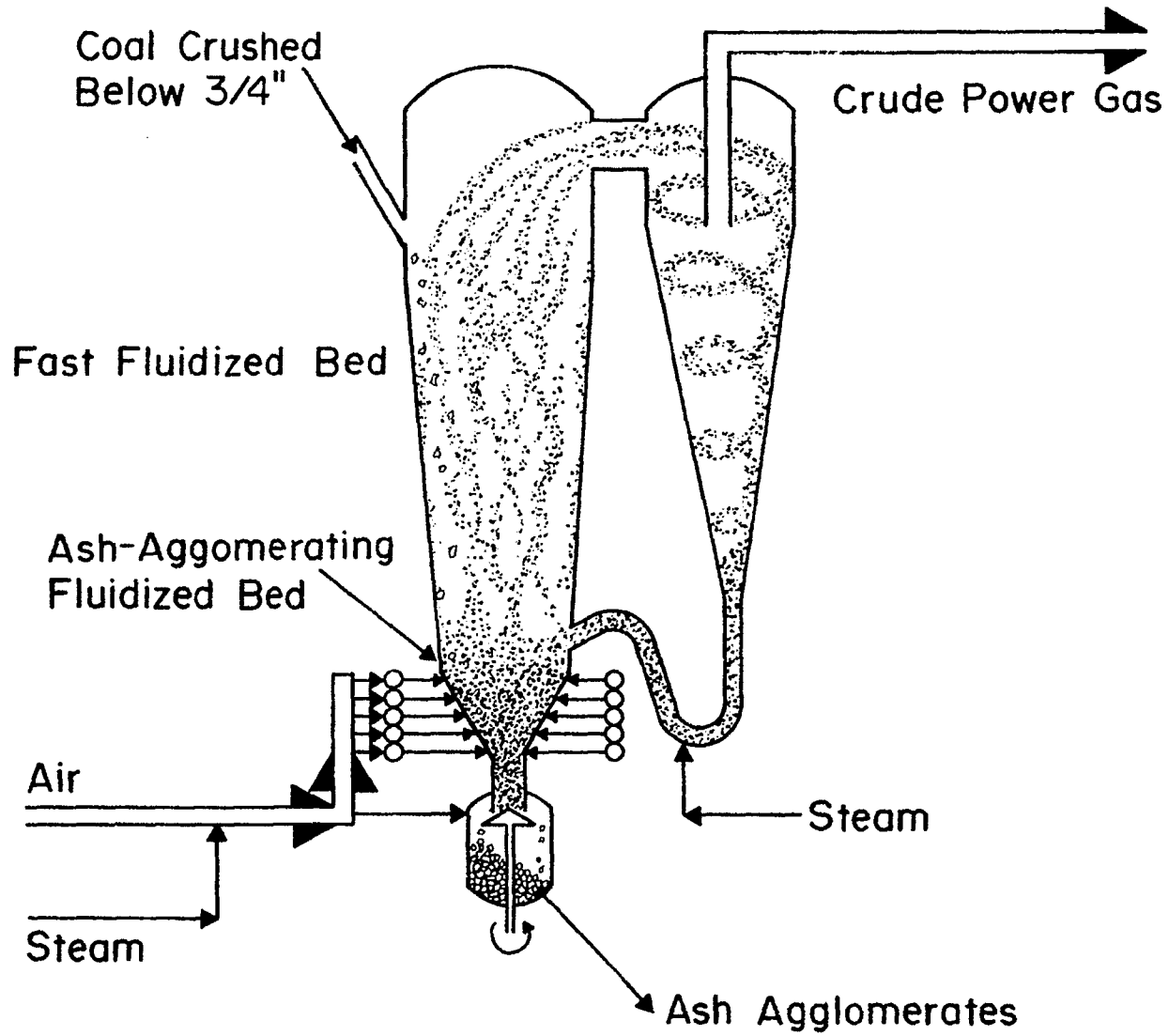


Figure 23. The Mark II Coal Gasifier

If agglomerates form, they will drop to the bottom and be burned. In accordance with the Gelding 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 devolatilization 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 devolatilization 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 volatile 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



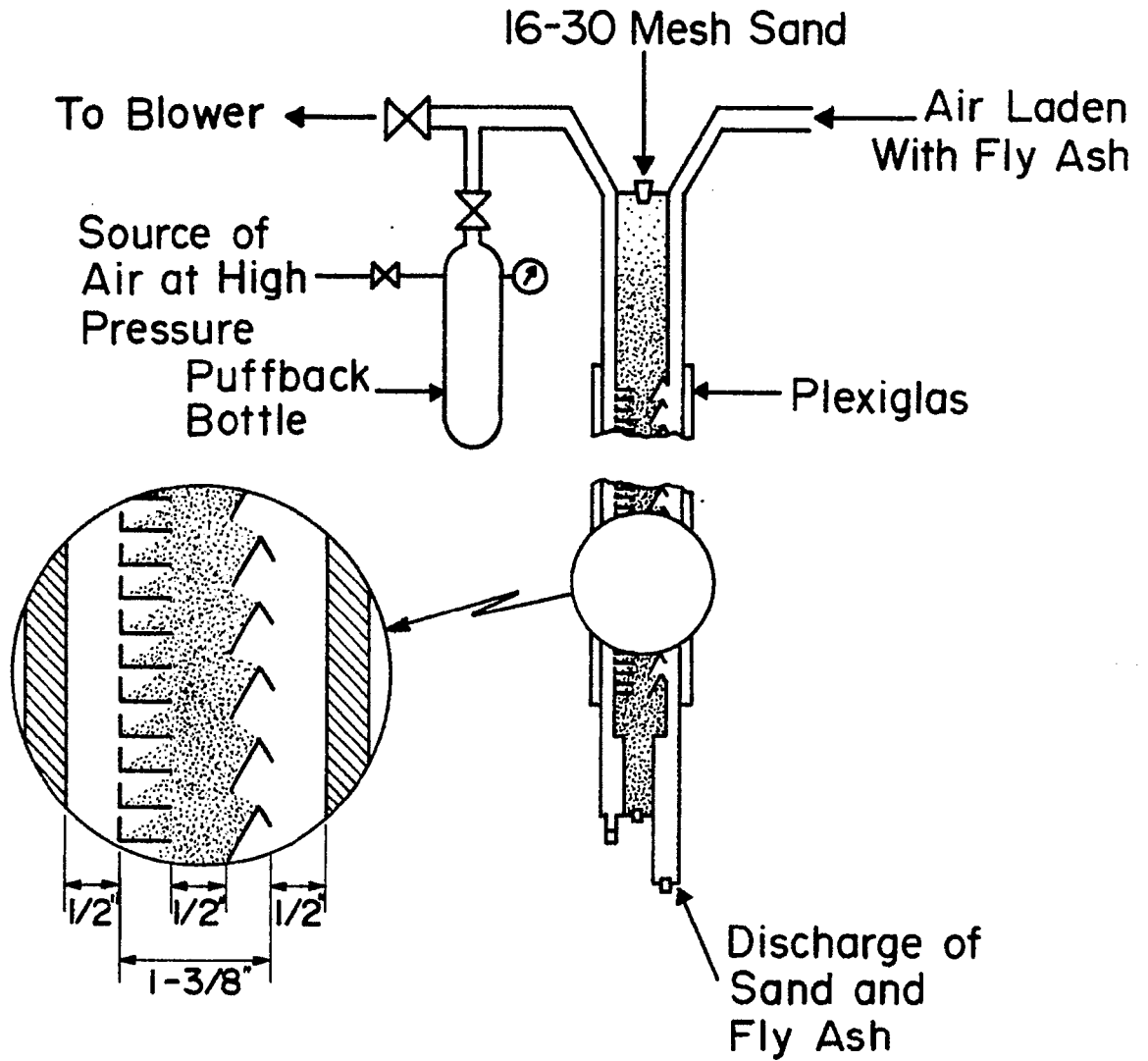


Figure 24. Schematic of a Panel Bed Filter

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

INSTITUTE OF GAS TECHNOLOGY - U-GAS<sup>TM</sup> PROCESS

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 devolatilization 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

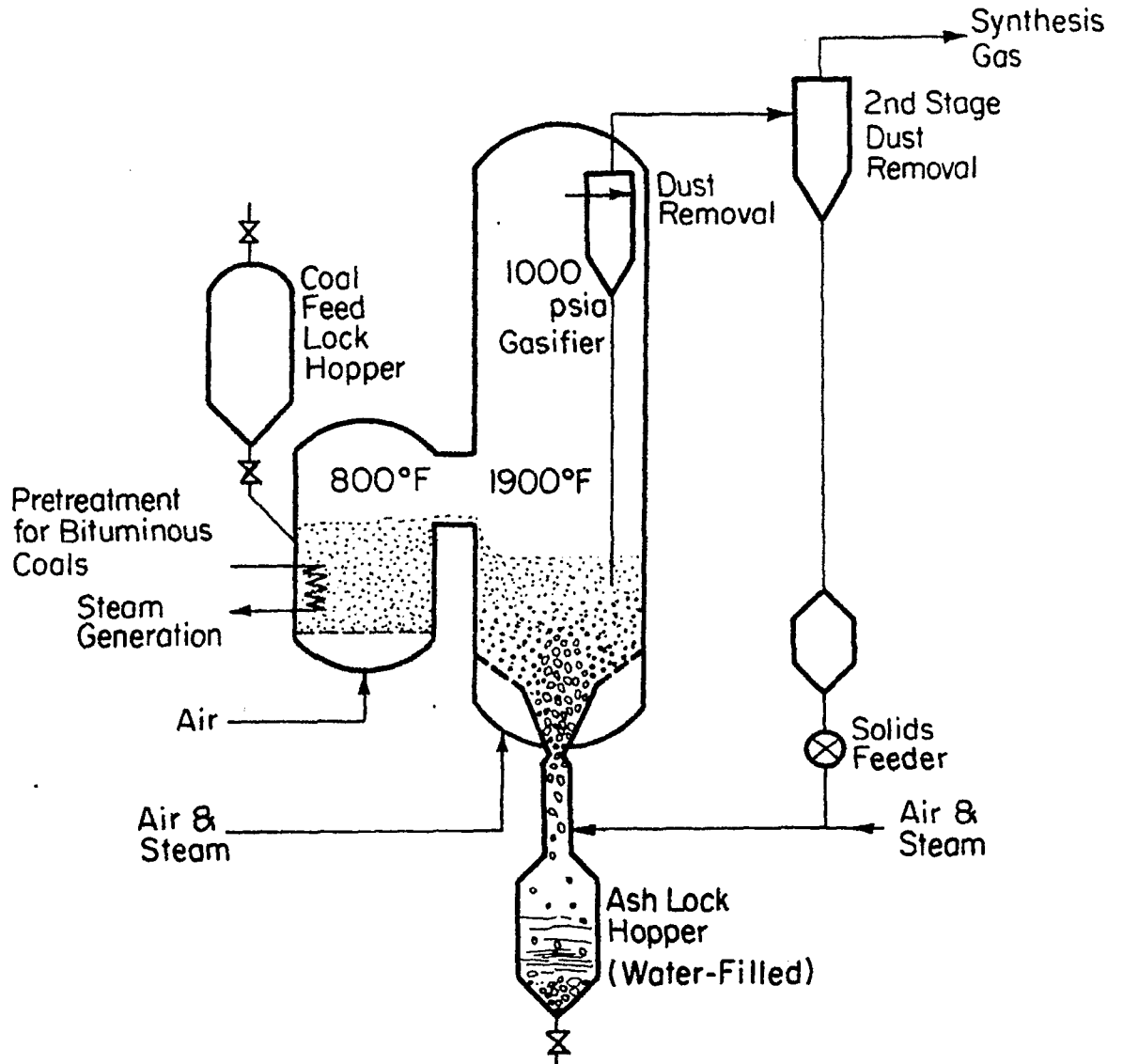


Figure 25. Institute of Gas Technology Ash-Agglomerating Gasifier

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 VI. SUMMARY OF ECONOMICS FOR EACH OF THREE U-GAS™  
CLEAN POWER PLANT PROPOSALS

	Application		
	<u>Retrofit</u>	<u>Separate</u>	<u>Combined-Cycle</u>
Coal Feed Rate, ton/day	7346	7346	7346
Coal Cost, ¢/million Btu	30	30	30
Plant Investment, \$, million	65	90	216
Process Overall Thermal Efficiency, %	85	78	42
By-Products (assumed value)			
Sulfur, long ton (\$10/LT)	188	188	188
Steam, 1000 lb/hr (\$0.90/1000 lb)	721	--	--
Power, MW (8 mills/KW hr)	22	146	--
Net Product Output			
Low-Btu Gas, 10 <sup>9</sup> Btu/day	129	129	--
Electricity, MW	--	--	942
Unit Price of Output*			
Low-Btu Gas, 90% load factor ¢/million Btu	61.2	65.3	--
Electricity, 80% load factor mills/KW hr	--	--	9.0

176

\*20-year average price determined by A.G.A.-OCR Utility Accounting for process plants.  
Early 1973 Economics



## INSTITUTE OF GAS TECHNOLOGY - HYGAS PROCESS

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.



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

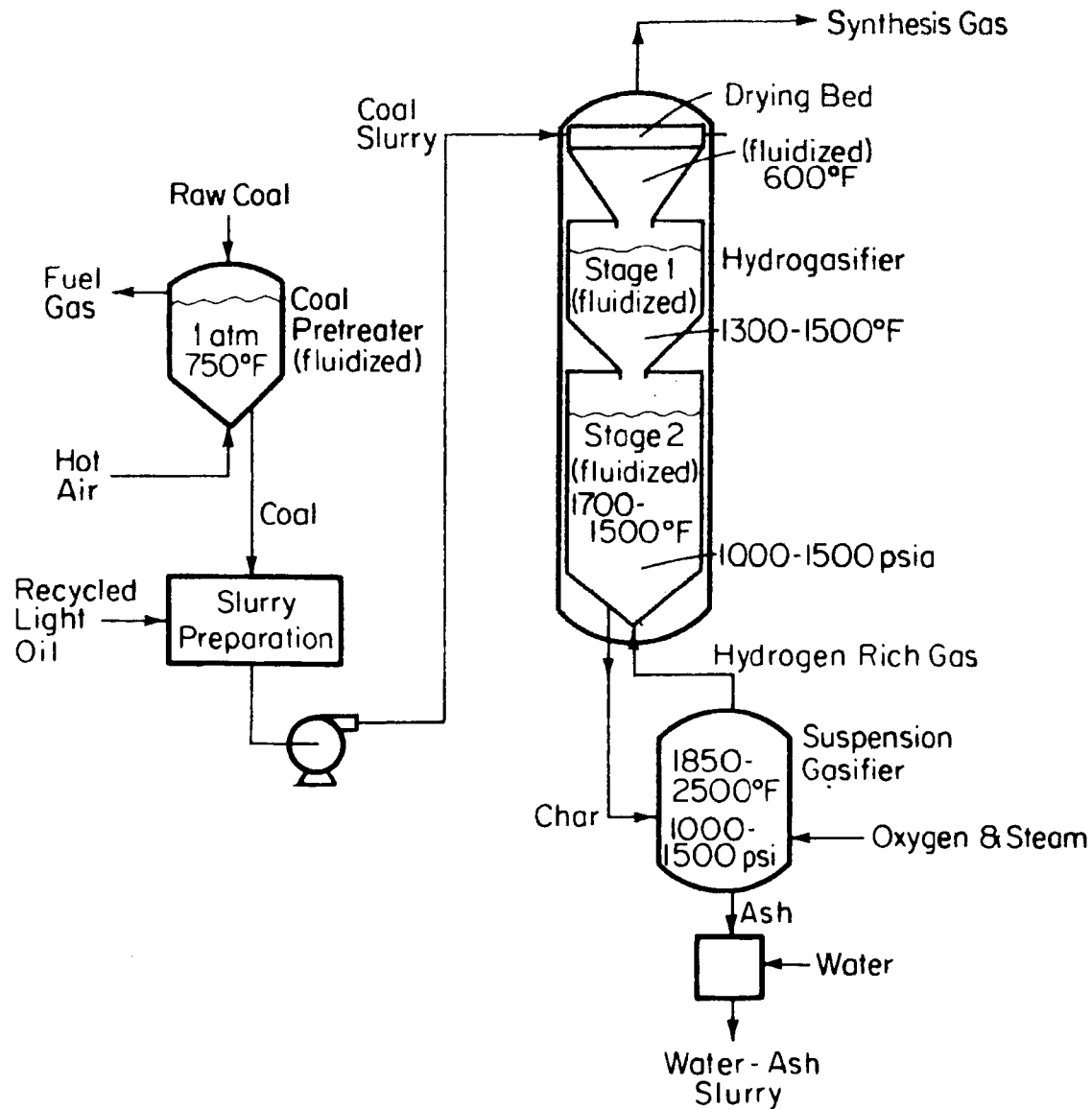


Figure 26. Institute of Gas Technology, HYGAS Coal Gasification Process

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  $H_2S$  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 reblading 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

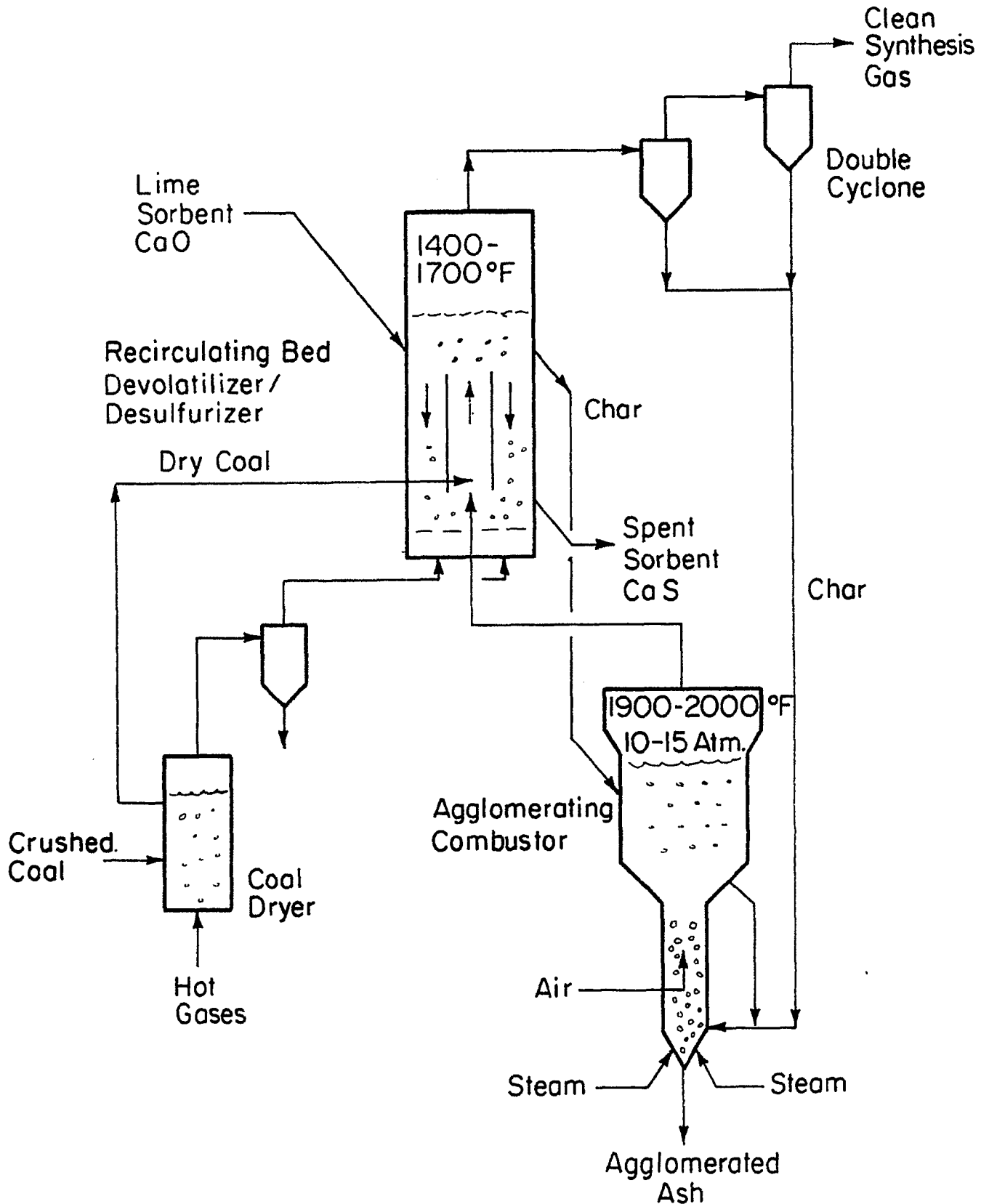


Figure 27. Westinghouse Multistage Fluidized Bed Process for the Total Gasification of Coal with Desulfurization for an Electric Power Plant

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 devolatilization takes place in the fluidized-bed devolatilizer. 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 (Ca CO<sub>3</sub>). 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 H<sub>2</sub>S. If this is the case, a common problem is shared; there is no proven high temperature process for removal of H<sub>2</sub>S.

(2) Lime is considerably more expensive than Ca CO<sub>3</sub> 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 CO<sub>2</sub> 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<sub>2</sub>S) 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<sub>2</sub> 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<sub>2</sub> ACCEPTOR PROCESS

The CO<sub>2</sub> 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<sub>2</sub> Acceptor Process. The unique feature of this process is the use of an "acceptor" which reacts exothermically with CO<sub>2</sub> 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<sub>2</sub> Acceptor Process uses the exothermic reaction between CaO

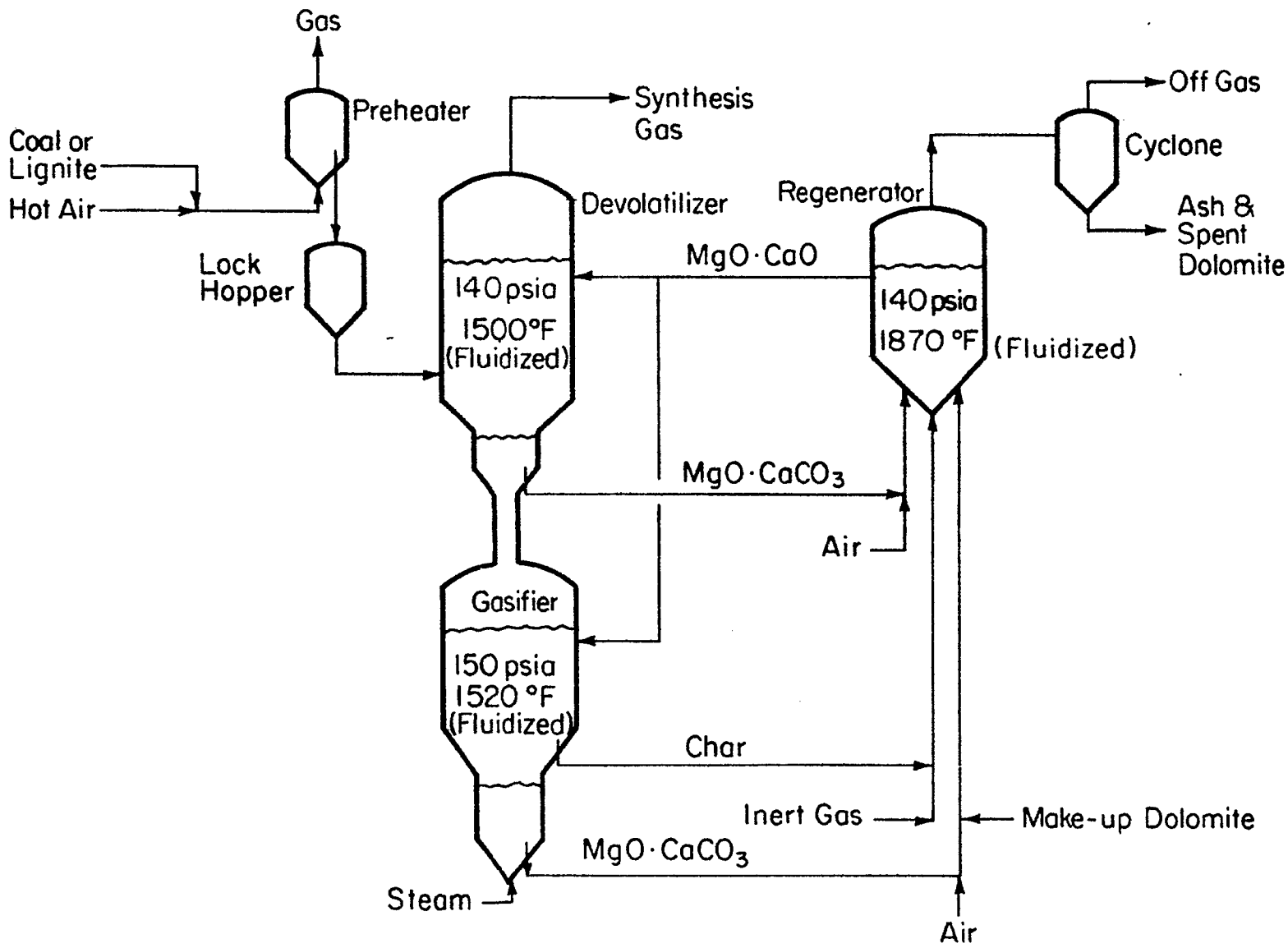
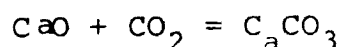


Figure 28. Consolidation Coal Company, Inc. CO<sub>2</sub> Acceptor Coal Gasification Process as Originally Built

and  $\text{CO}_2$  to provide the heat of gasification.



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  $1900^\circ\text{F}$ . 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  $1500^\circ\text{F}$ . 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 "showers" 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 CO<sub>2</sub> 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

## BRIGHAM YOUNG UNIVERSITY - ENTRAINED BED GASIFIER

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/(ft <sup>3</sup> ) (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 than 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

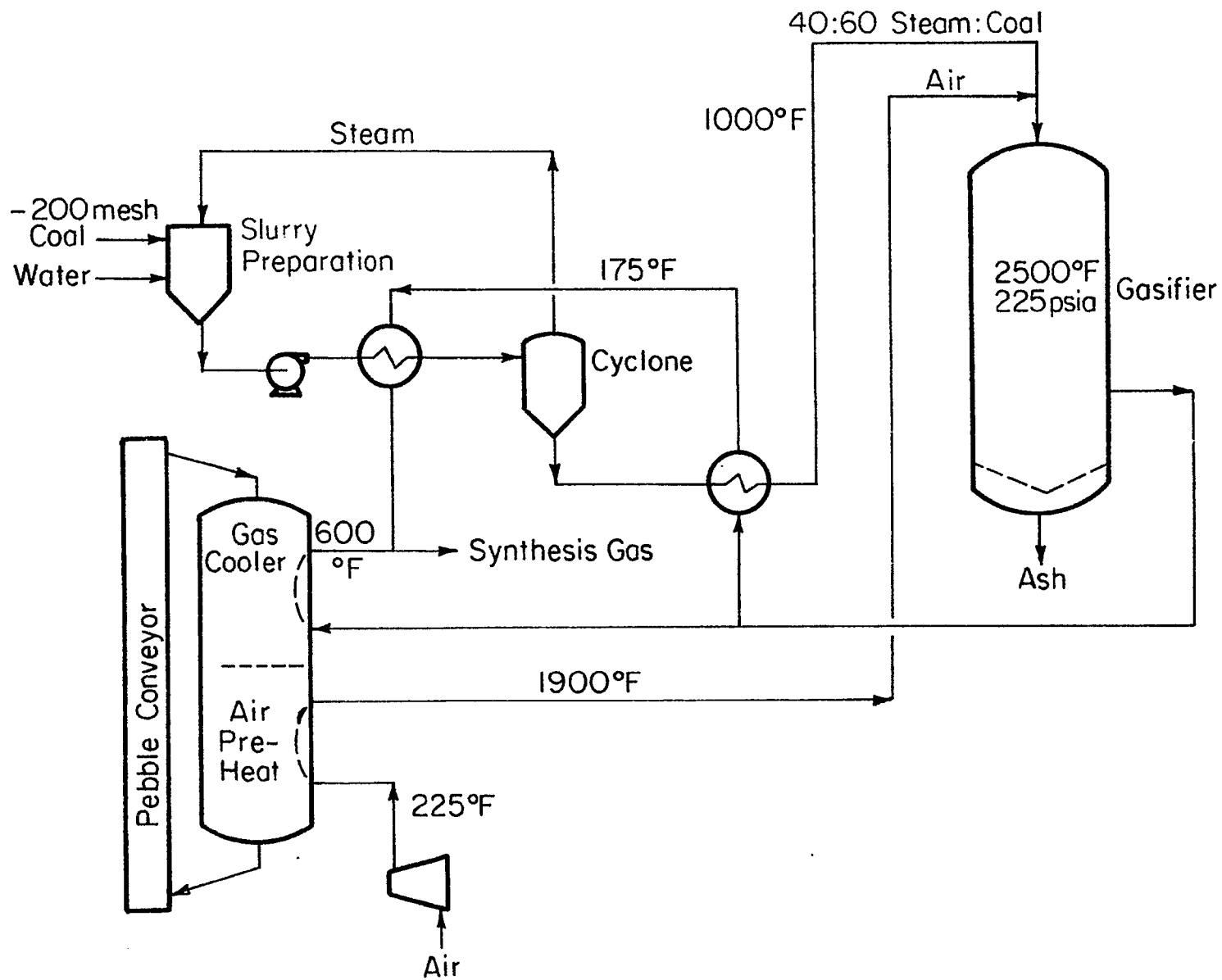


Figure 29. Texaco Partial Oxidation Coal Gasification Process

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).

Prepared by Dale E. Briggs



## 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

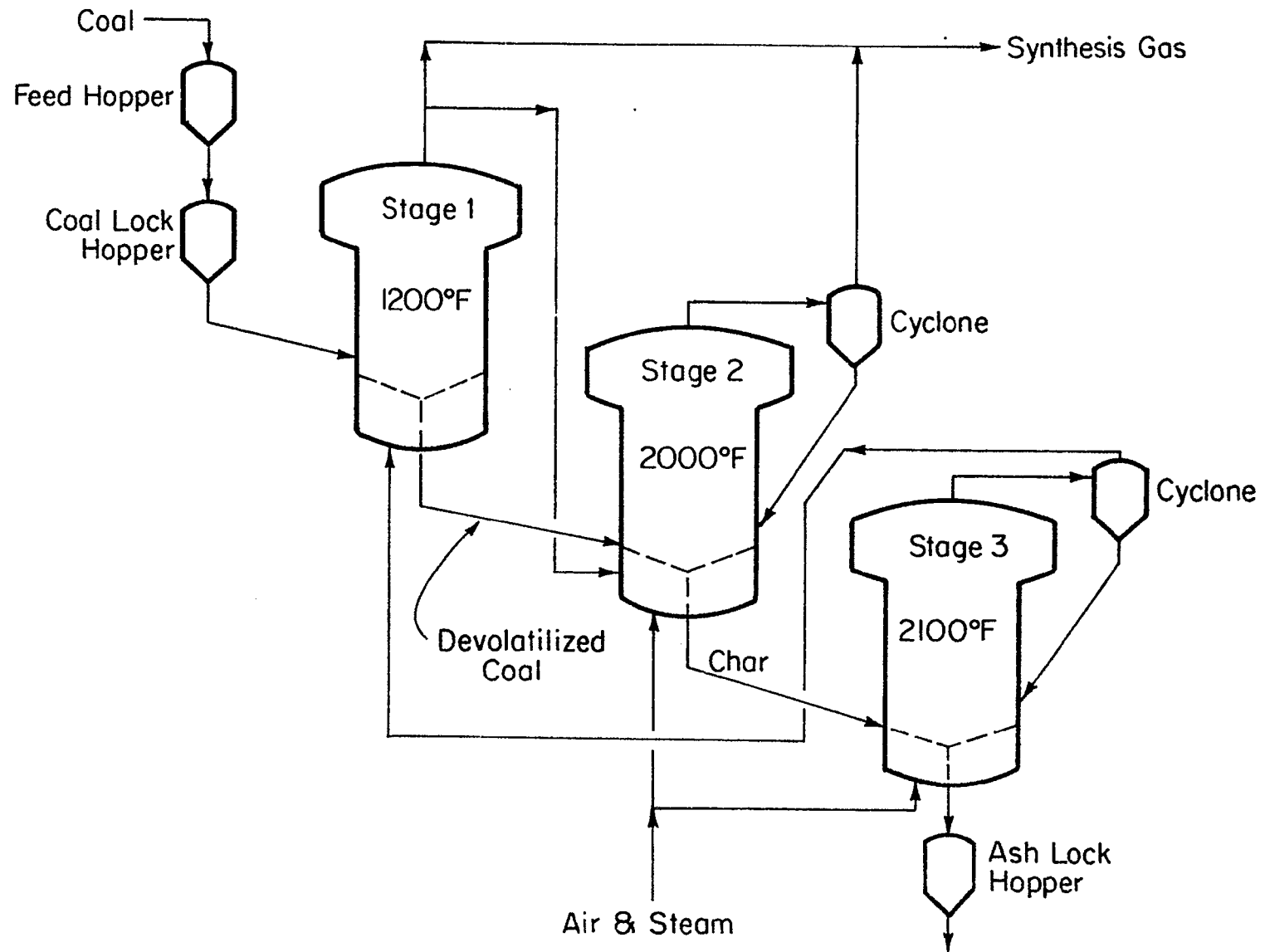


Figure 30. Bituminous Coal Research, Inc. Fluidized-Bed Gasifier System

APPLIED TECHNOLOGY CORPORATION - TWO STAGE COAL COMBUSTION PROCESS,  
PATGAS, ATGAS

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

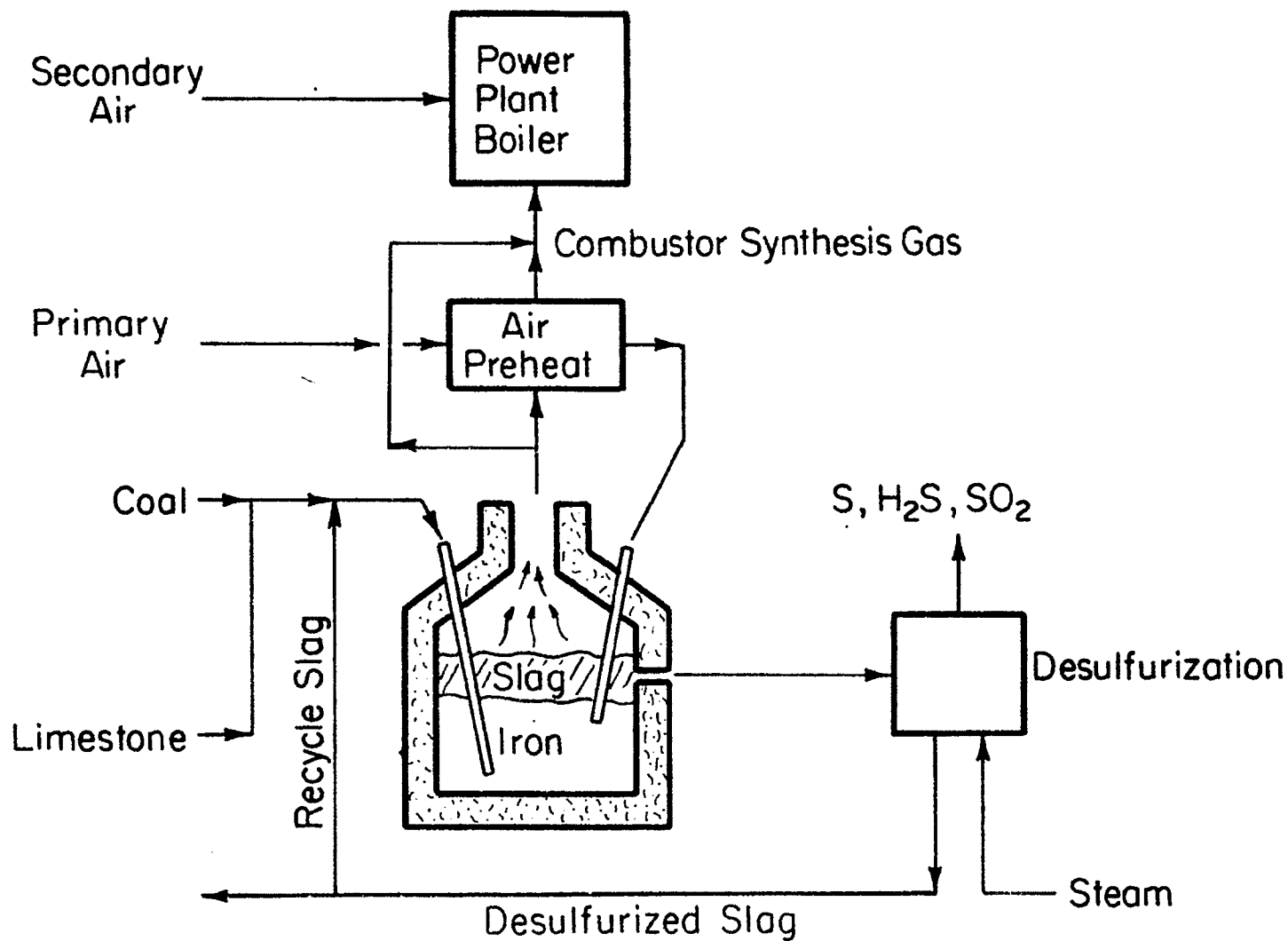


Figure 31. Applied Technology Corporation Two Step Gasification System

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

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# 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.

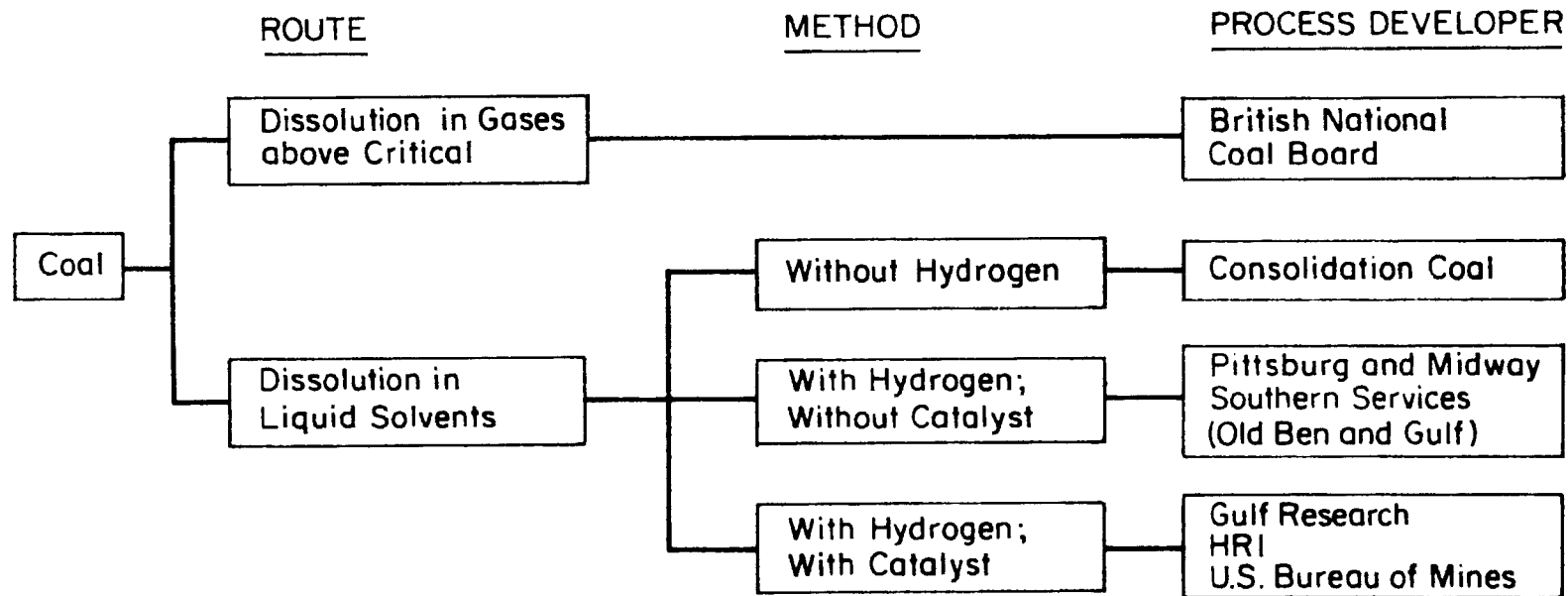


Figure 1. Classification of Coal Dissolution and Liquefaction Processes

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  $\text{CH}_{0.795}\text{N}_{0.013}\text{O}_{0.108}\text{S}_{0.018}$  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  $\text{CH}_{1.1}$ , 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

	Without Hydrogen	With Hydrogen
Without Catalyst	Consol Exxon	PAMCO Southern Services
With Catalyst	Consol* Exxon*	Gulf R&D HRI U.S. Bureau of Mines

\*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 II. Present Coal Dissolution Processes and Development Programs

<u>Process</u>	<u>Developer</u>	<u>Location</u>	<u>Engineering Design</u>	<u>Present Development Size</u>	<u>Next Stage</u>
Project Gasoline or Consol Synthetic Fuel	Consolidation Coal Company	Cresap, W. Va.	C.F. Nofsinger Co.	20 tons/day	
PAMCO Solvent Refined Coal-SRC	Pittsburg & Midway Coal Mining Co.	Merriam, Kansas	Pittsburg & Midway	75 lb/day Bench Scale and 1 ton/day Process Development plant	Pilot plant under construction
		Tacoma, Wash.	Stearns-Roger	50 tons/day construction scheduled for completion)	
Solvent Refined Coal	Southern Services (PAMCO SRC process)	Wilsonville, Ala.	Catalytic, Inc.	6 tons/day (scheduled to operate on coal January 1974)	
H-Coal	Hydrocarbon Research, Inc.	Trenton, N.J.	HRI	3 tons/day	700 tons/day
Synthoil	U.S. Bureau of Mines	Brecceton, Pa.	U.S. Bureau of Mines	1000 lbs/day	8 tons/day
Gulf Catalytic Coal Liquids	Gulf Research & Development	Harmarville, Pa.	Gulf R&D	120 lbs/day	1 ton/day

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

<u>Process</u>	<u>Engineering Design</u>	<u>Plant Size tons coal/day</u>	<u>Coals</u>	<u>Main Fuel Oil Products</u>	<u>Estimated Overall Thermal Efficiency %</u>	<u>Plant Capital Cost* Million \$</u>	<u>Date of Study</u>	<u>MW Potential at 35% Efficiency</u>	<u>Cost \$/KW</u>
Modified SCR	Ralph M. Parsons (17)	10,000	Illinois No. 6, 3.4%S	0.2%S 13.9°API 0.5%S -9.7°API	63.5	270	1973	620	435
Consol CSF	Foster-Wheeler (18)	20,000 (MF)	Pittsburg Seam 4.2%S	0.056%S 58°API 0.128%S 10.3°API	71-73.9	230	1972	1530	150
H-Coal	Hydrocarbon Research, Inc. (10)	25,000 (MF)	Illinois No. 6, 5%S	0.1%S 27°API 0.5%S -3.1°API	69.6	299	1973	1800	166.
		35,211	Wyodak 0.7%	< 0.2% 39.3°API	**	445	1973	2000	222.
Gulf CCL	Gulf R&D(8)	33,000	Big Horn 0.54%	< 0.04%S 35.3°SPI 0.04%S 9°API	**	423	1973	2300**	184.***

\* Does not include interest during construction

\*\* Not given

\*\*\* Estimated

(MF) Moisture Free

TABLE IV. Coal Liquefaction Process Operating Conditions and Typical Products

PROCESS	H-COAL	PARSONS MODIFIED PAMCO	PAMCO (S.SERV.)	BUR. OF MINES	GULF CCL	GULF CCL	CONSOL
Hydrogen used in dissolution?	Yes	Yes	Yes	Yes	Yes	Yes	No
Subsequent Extract Hydrogenation?	No	Yes	No	No	No	No	Yes
Catalytic Dissol. Approximate Reactor Temperature.	Yes 850°	No 840°F	No 850°F	Yes 840°F	Yes 800°F	Yes 800°F	No 730°F
Reactor Pressure.	3000 psig	1200 psig	1500 psig	4 000 psig	3000 psig	3000 psig	400 psig
Coal Sulfur, Wt.%	111. No.6 5%	111. No.6 3.38%	-- 5%	Kentucky 4.6%	Big Horn Subbit. 0.54%	Pittsburg Seam (Bit.) 1.49%	Pittsburg Seam Coal 3.67%
Solvent to Coal Ratio (to slurry).	1:1(by Wt.)	2.0:1(by Wt.)	2:1(by Wt.)	1.22:1.0(by Wt.)	2.33:1.0(by Wt.)	2.33:1.0(by Wt.)	2:1 (by Wt.)
Percent Coal Dissolved (MAF).	90%+	90%+	90%+	90%+	91%	90%	63%
Hydrogen Consumption Scf/ton Coal (MAF).	15,300	12,600	142% by Wt. 7600	9000	22,800	17,500	16,300
Solids Separation, Solids Content in Product.	Hydroclones and/or filtration	Filtration	Filtration 0.23 Wt.%	Centrifuge 1.3 Wt.%	Hydroclones & Filt. 0.02 Wt.%	Hydroclones & Filt. 0.03 Wt. %	Hydroclones
Principal Products							
1. Fuel	Fuel Oil	Residual Fuel Oil	Solvent Refined Coal 1116 lb/ton*	Fuel Oil	Filtrate Fuel Oil	Filtrate Fuel Oil	Fuel Oil
Yield bbl/ton	1.73 bbl/ton	1.43bbl/ton		3 bbl/MAF ton*	2.3 bbl/ton	3.6 bbl/ton*	1.52 bbl/ton coal
API gravity	-3.1°API	-9.7°API	60/60	Sp Gr=1.12-1.14	9.0°API	1.2° API	10.3°API
Viscosity				Visc = 75-204 SSF@ 180°F	7.1 CS @100°F	4.3 CS @210°F	
Sulfur, Wt.%	0.5%	<0.5%	<1.2%	0.31%	0.04%	0.11	.128%
Nitrogen, Wt.%				0.9%	0.40%		
2. Fuel	Naphtha	Distillate Fuel Oil			Light Ends	Light Ends	Naphtha
API gravity	38.4°API	13.9°API	60/60		35.3°API	0.45 bbl/ton*	58.0°API
Yield bbl/ton	0.54bbl/ton	0.71 bbl/ton			0.9 bbl/ton*		0.52 bbl/ton
Viscosity					1.2 CS @100°F		
Sulfur, Wt.%	<0.1%	0.2%			0.04%		.056%
Nitrogen, Wt.%					0.19%		

\*Exclusive of H<sub>2</sub> 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.

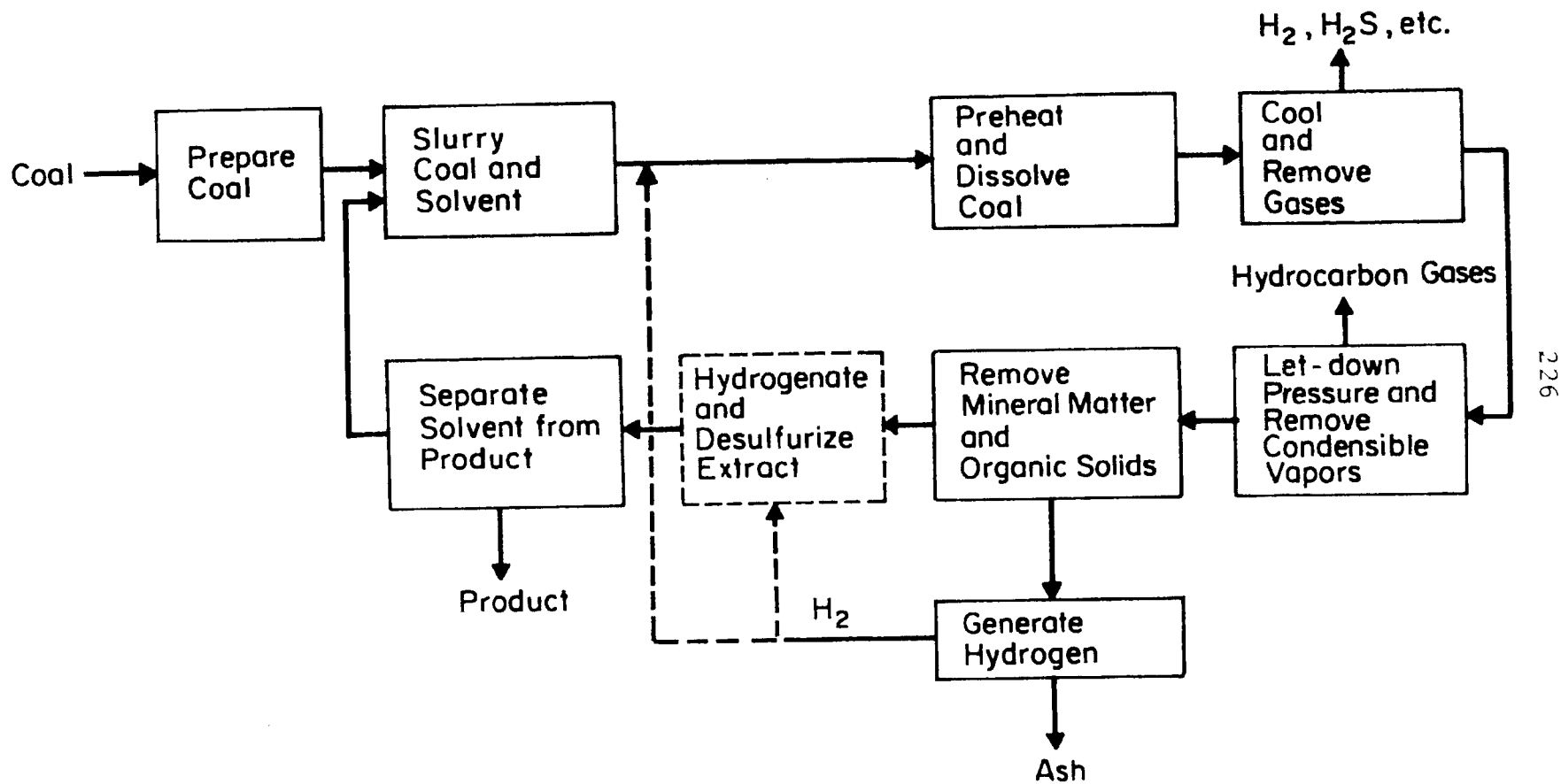


Figure 2. Processing Steps in Coal Dissolution and Liquefaction

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 preferable 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 slurrifying, 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)

Coal Concentration, Wt. %	Viscosity, cp.
0	4.5
21	11
25	18
33.5	36
35	50
40	90
45.5	260
50	540
55	1000
63	∞

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.

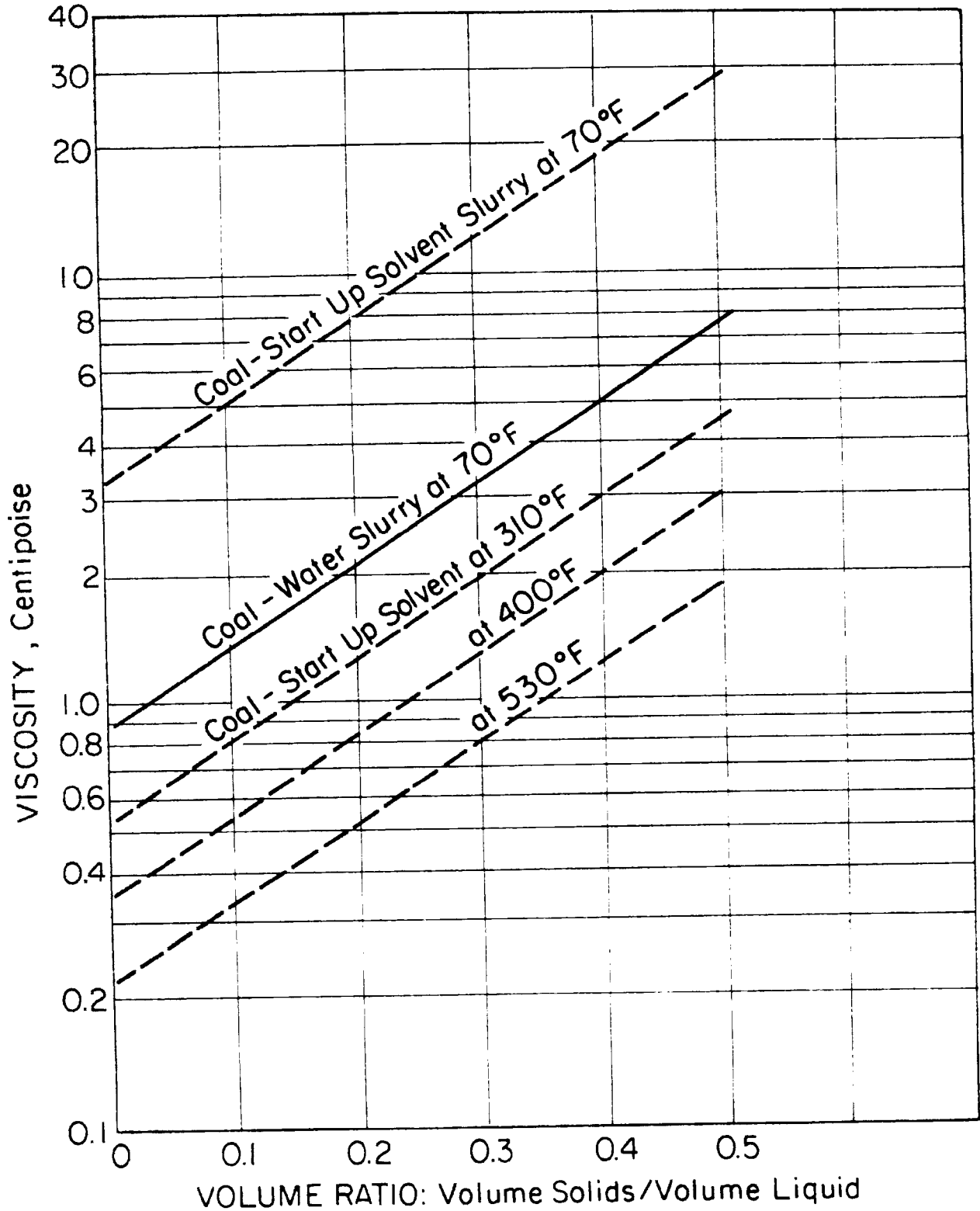


Figure 3. Viscosity Values for Fine Coal Particles in 0:1 and Water (19)

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.

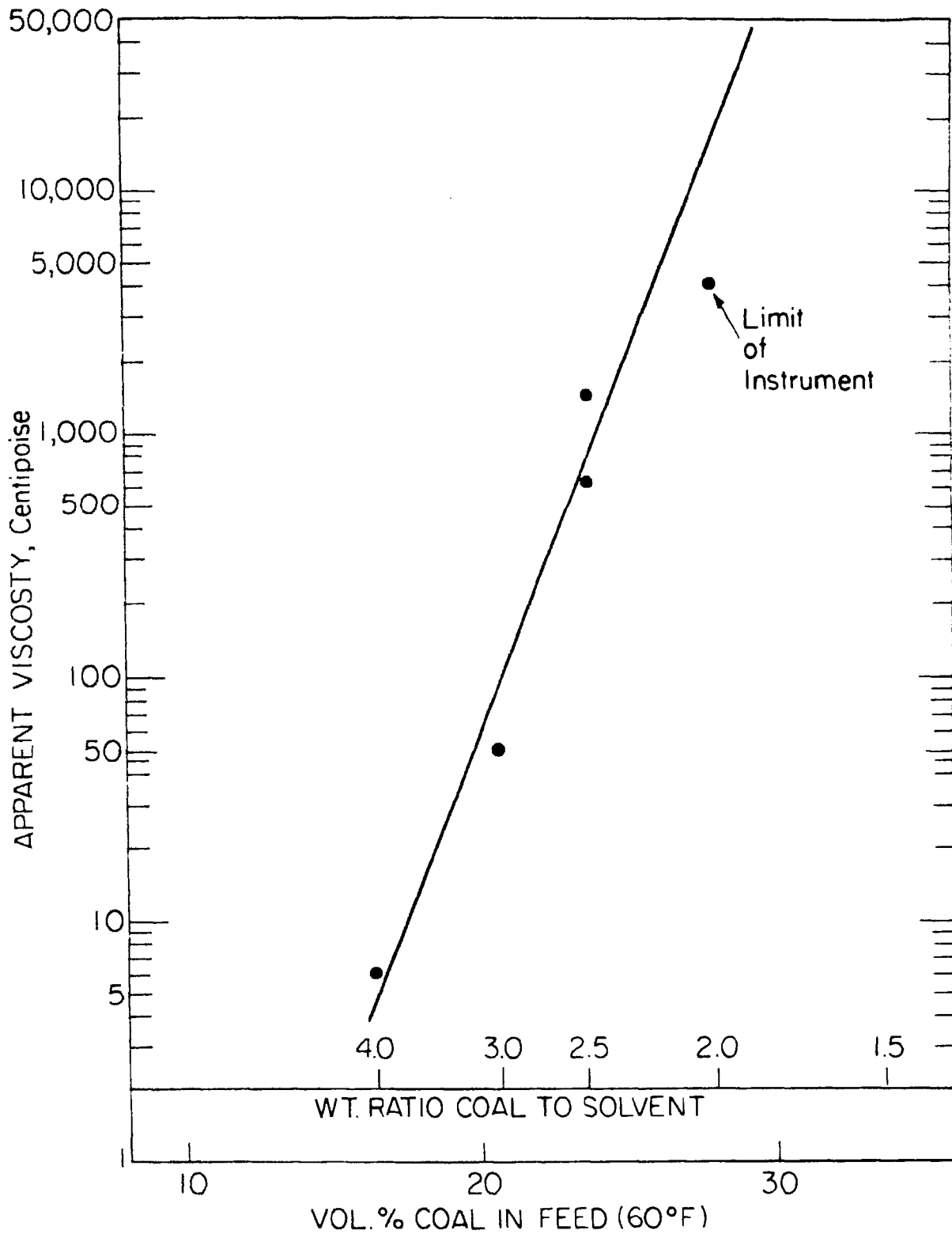


Figure 4. Viscosity Peaks from Bath Extraction of Kentucky No. 11 Coal (19)



### 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 condensable 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  $TiO_2$ , 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.

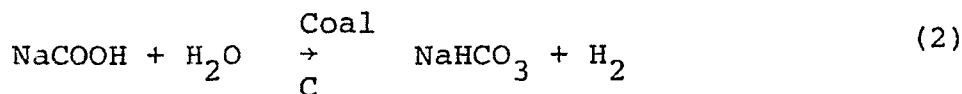
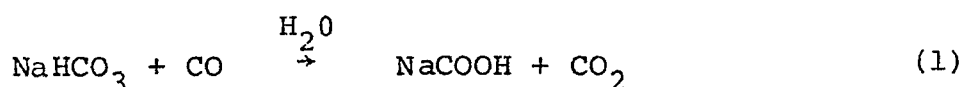
Over 60% of the organic sulfur is removed in the reactor. The pyritic sulfur can be removed subsequently when the mineral matter is separated from the extract.

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

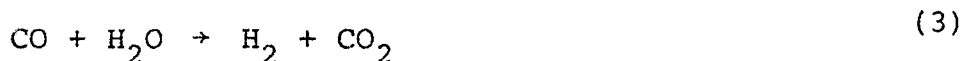
The SRC process is "non-catalytic" in the sense that a commercial catalyst is not used in the reactor. There is considerable experimental evidence, however, that the inorganic mineral matter in certain western lignites and subbituminous coals has a significant catalytic effect, especially sodium. While 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 Pittsburgh 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 Pittsburgh 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.



with the net reaction being



(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 condensable 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 slurring 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 preferably 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 stand point) 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.

## Process Descriptions

## 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

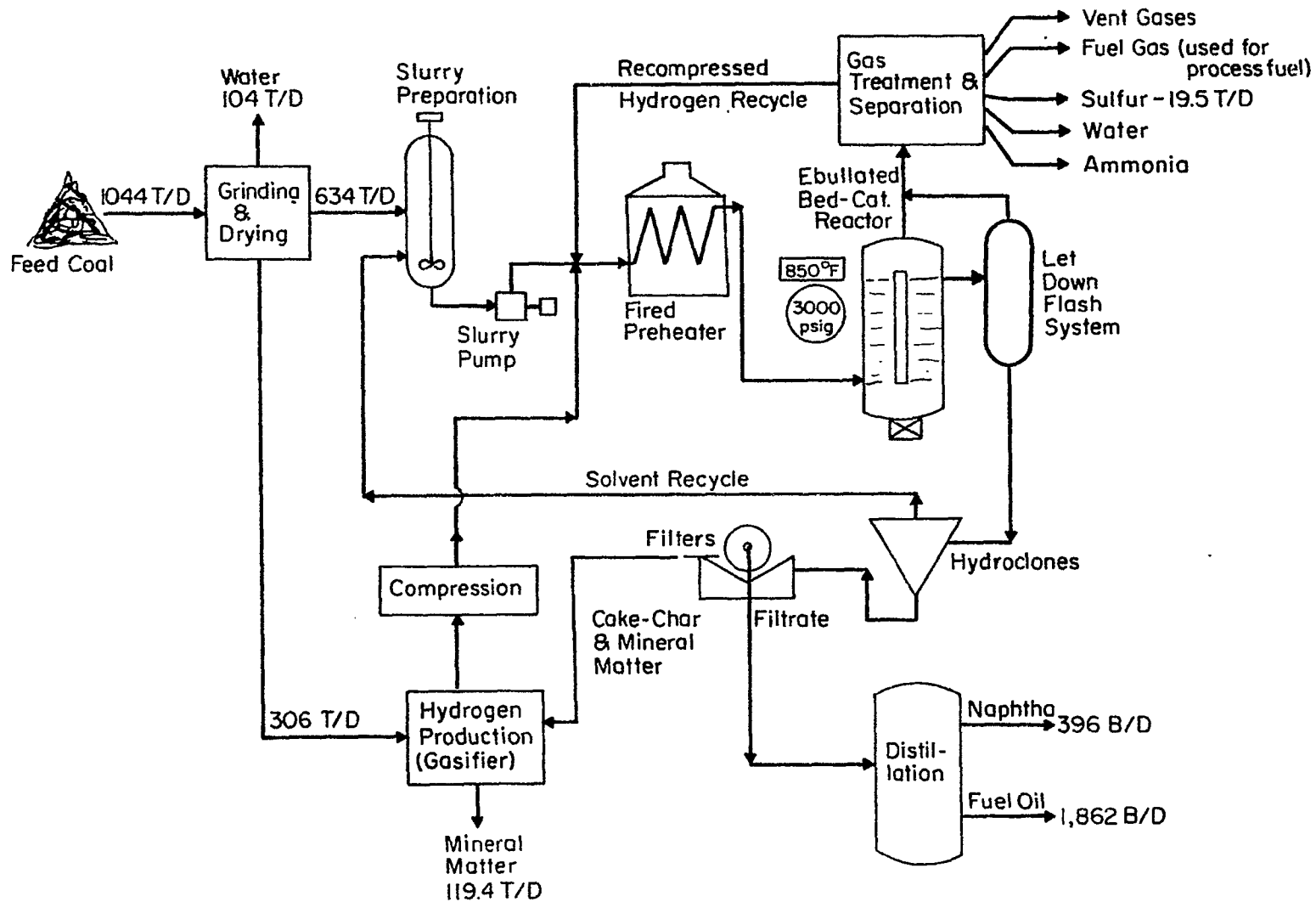


Figure 5. H-Coal Process for Fuel Oil Production--Devolatilization Plant

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)

	<u>ILLINOIS NO. 6</u>	<u>WYODAK</u>
Moisture, Wt.%	17.5	30.4
<u>Proximate Analysis, Wt.% (Dry Basis)</u>		
Ash	9.9	7.9
Volatile Matter	42.0	44.1
Fixed Carbon	48.0	48.1
<u>Ultimate Analysis</u>		
Carbon	70.7	68.4
Hydrogen	5.4	5.4
Nitrogen	1.0	0.8
Sulfur	5.0	0.7
Oxygen (Difference)	8.1	16.9
Ash	9.9	7.9

TABLE VII

Coal Hydrogenation Results (Wt. % of M.A.F. Coal)-H-Coal Study (11)

Coal Desired Product	<u>Illinois</u>		<u>Wyodak</u>
	<u>Synthetic Crude</u>	<u>Low-Sulfur Fuel-Oil</u>	<u>Synthetic Crude</u>
<u>Normalized Product Distribution</u>			
C <sub>1</sub> -C <sub>3</sub> Hydrocarbons	10.7	5.4	10.2
C <sub>4</sub> -400°F Distillate	17.2	12.1	26.1
400-650°F Distillate	28.2	19.3	19.8
650-975°F Distillate	18.6	17.3	6.5
875°F+ Residual Oil	10.0	29.5	11.1
Unreacted Ash-Free Coal	5.2	6.8	9.8
H <sub>2</sub> O, NH <sub>3</sub> , H <sub>2</sub> S, CO, CO <sub>2</sub>	<u>15.0</u>	12.8	<u>22.7</u>
Total (100.0 + H <sub>2</sub> Reacted)	104.9	103.2	106.2
Conversion, % M.A.F. Coal	94.8	93.2	90.2
Hydrogen Consumption, SCF/Ton	18,600	12,200	23,600
C <sub>4</sub> + Liquid			
API Gravity	15.0	4.4	26.8
Hydrogen, Wt. %	9.48	8.43	10.54
Sulfur, Wt. %	0.19	0.43	0.16
Nitrogen, Wt. %	0.68	1.05	0.64

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.

PITTSBURG AND MIDWAY COAL MINING CO. - PAMCO SOLVENT REFINED  
COAL PROCESS (SRC)

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 Fort 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.

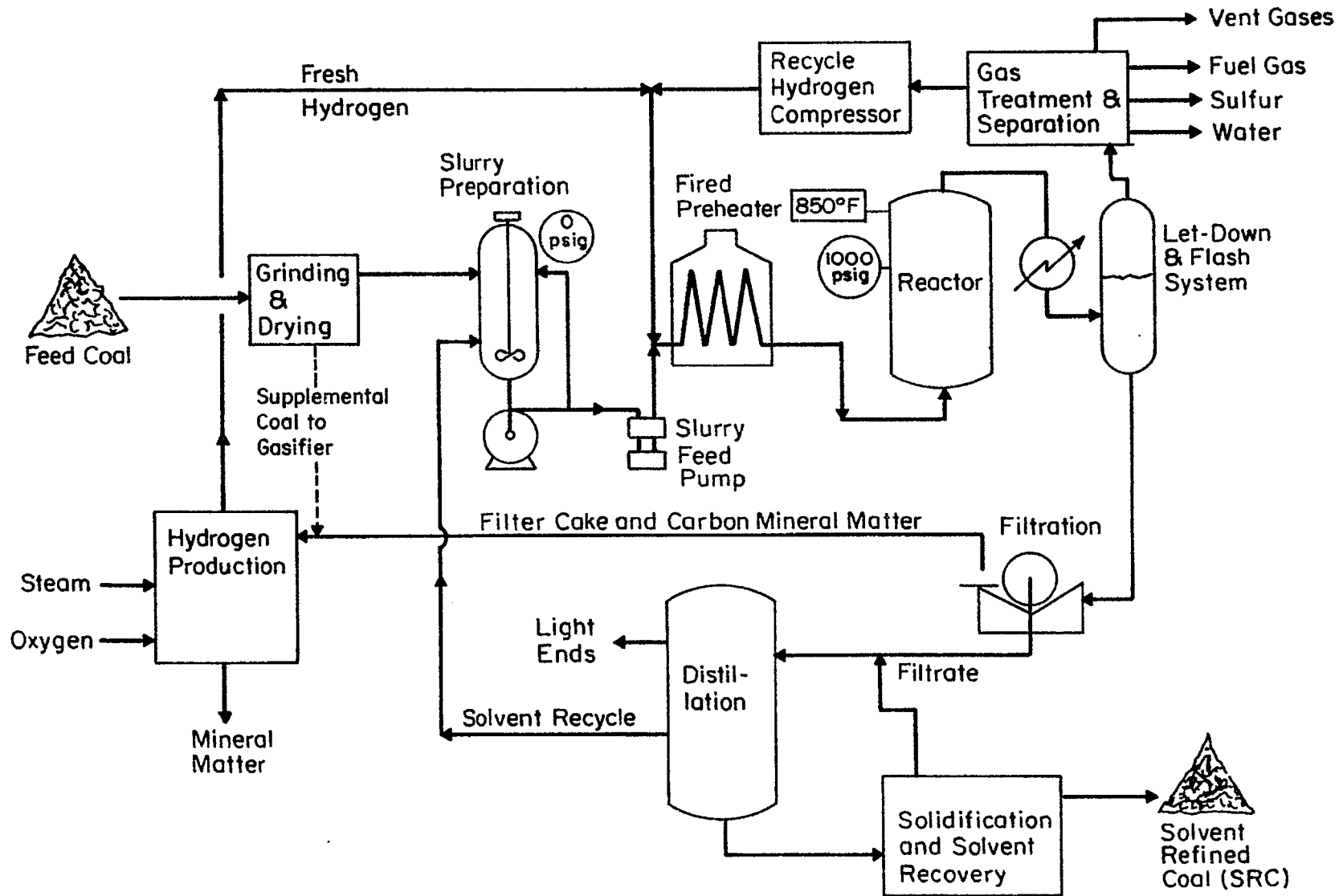


Figure 6. Pittsburgh & Midway Coal Mining Company Solvent Refined Coal Process - SRC

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  $H_2S$  and  $CO_2$ . 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,  $\text{CO}_2$  and  $\text{H}_2\text{S}$  (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  $600^\circ\text{F}$  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

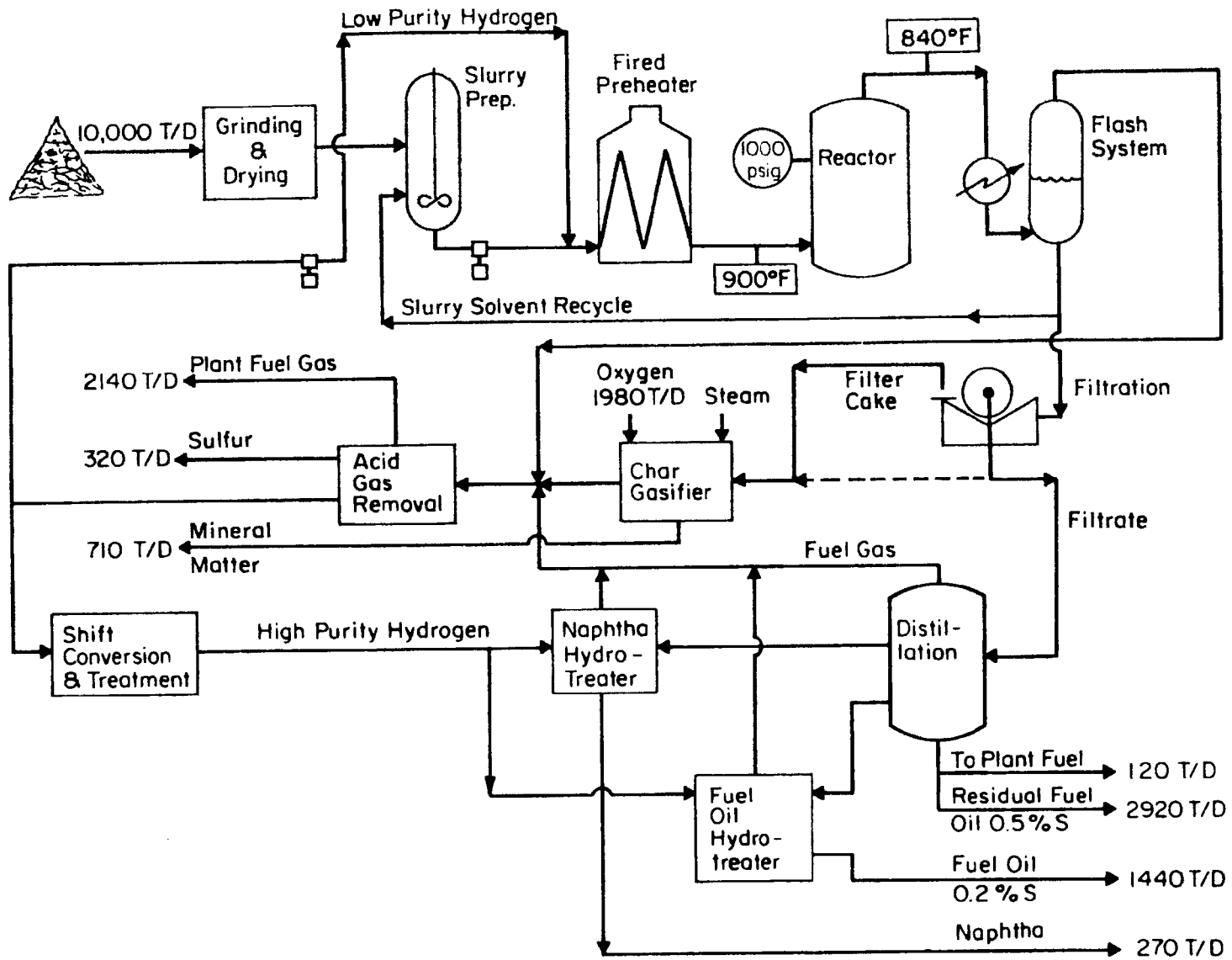


Figure 7. Parsons--PAMCO Hybrid Demonstration Plant Schematic Flow Sheet

## 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

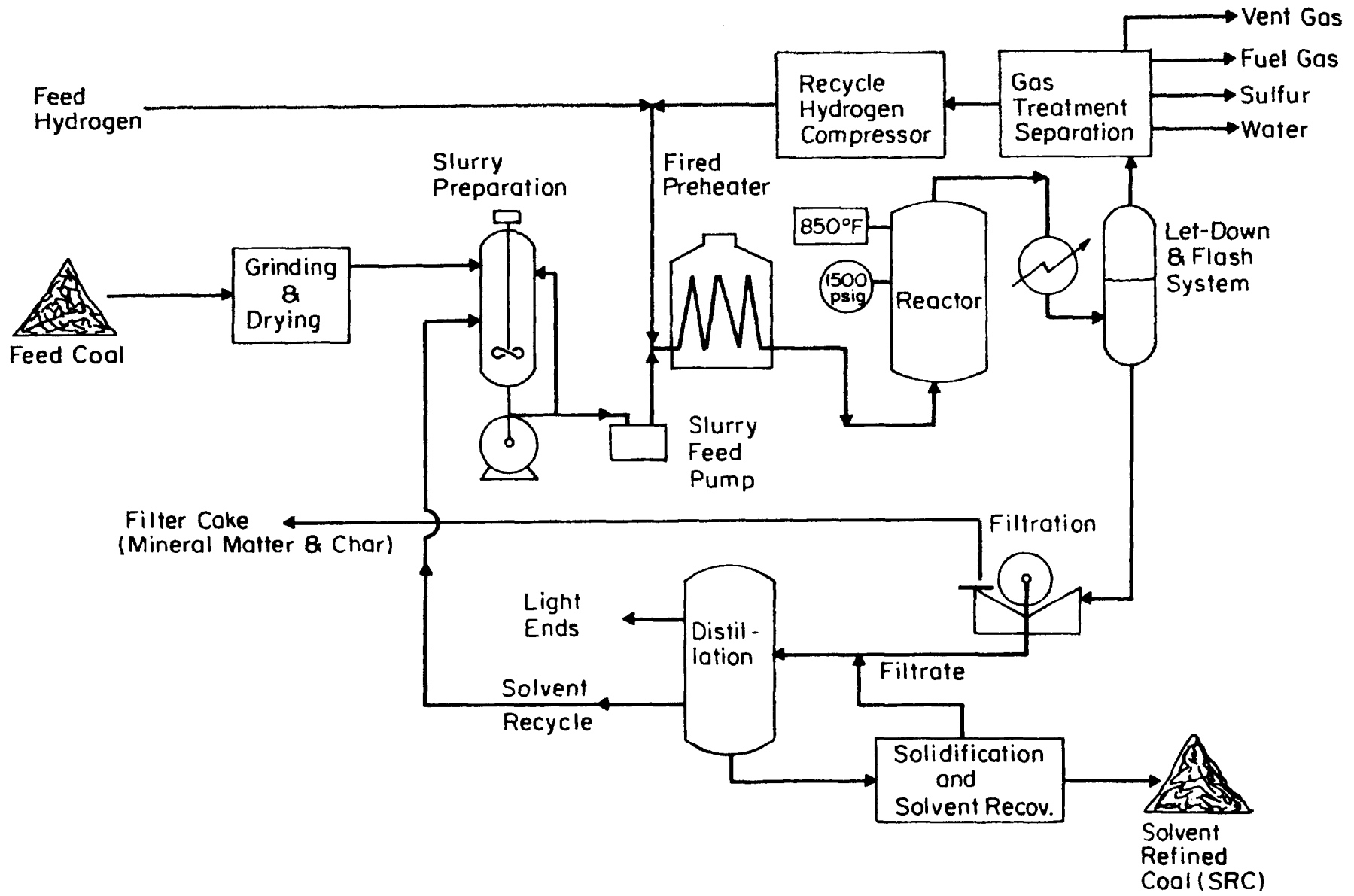


Figure 8. Schematic Flow Sheet of the Southern Services, Inc. Pilot Plant

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 asphaltic 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 RESEARCH AND DEVELOPMENT - GULF CATALYTIC COAL LIQUIDS

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 stream which is recycled for slurring 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.

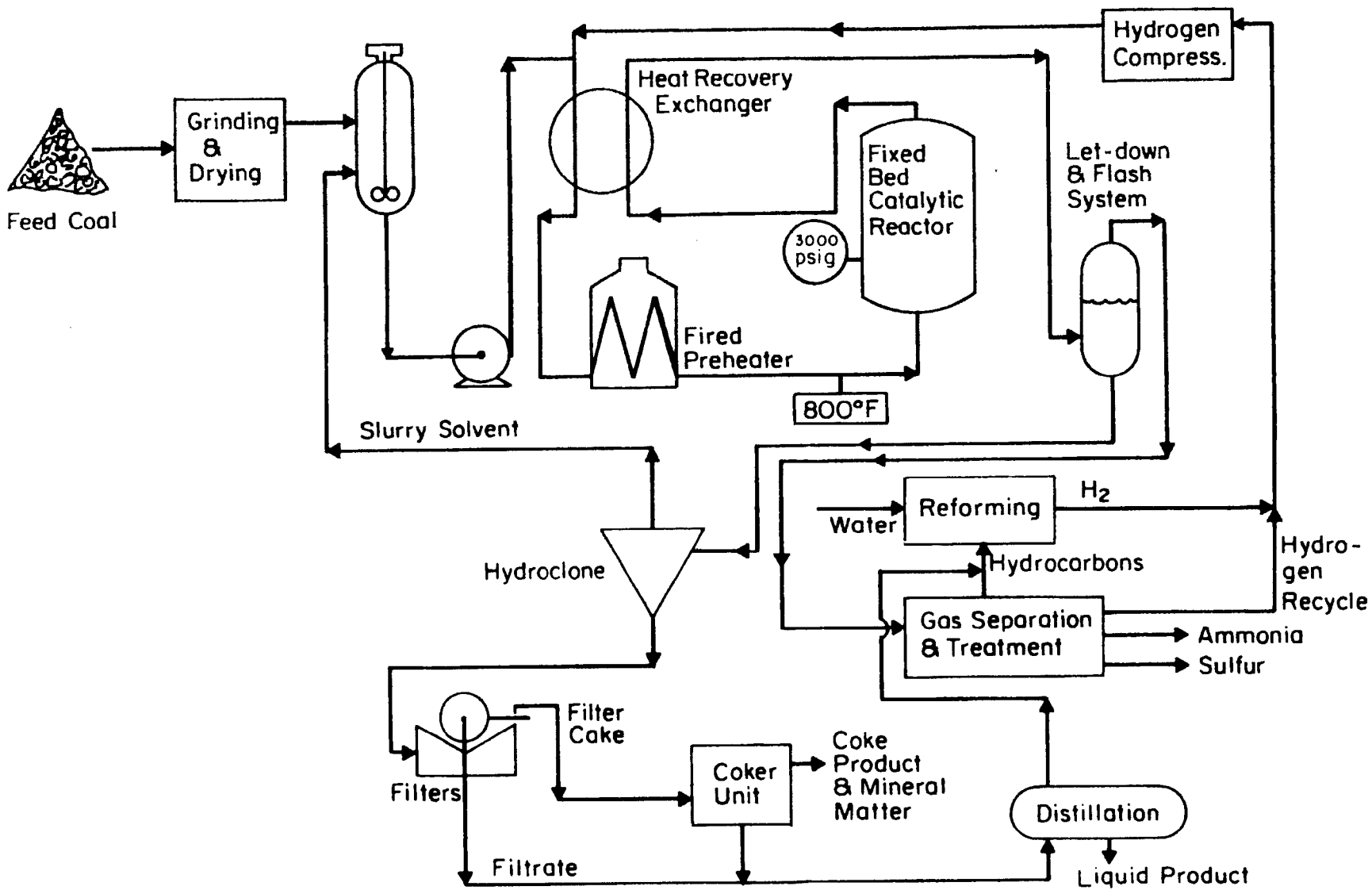


Figure 9. Gulf R&D Catalytic Coal Liquids Process

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)

RANK	<u>GLENHAROLD MINE</u>	<u>BIG HORN</u>	<u>KENTUCKY #9 &amp; #11*</u>	<u>PITTSBURG SEAM</u>
	LIGNITE	SUBBITUMINOUS	BITUMINOUS	BITUMINOUS
<u>PROXIMATE ANALYSIS,</u> WT. %				
MOISTURE	32.1	22.0	7.2	2.9
VOLATILE MATTER	43.0	33.2	32.3	33.1
FIXED CARBON	17.5	41.4	44.3	55.9
ASH	7.4	3.4	16.2	8.1
 <u>CHEMICAL ANALYSIS,</u> WT % (MOISTURE FREE BASIS)				
CARBON	65.44	69.34	68.53	76.84
HYDROGEN	4.49	4.60	4.60	5.06
NITROGEN	1.01	1.23	1.42	1.61
OXYGEN (DIFF)	17.39	19.90	5.82	6.72
SULFUR INORGANIC		0.14	3.17	0.66
ORGANIC		<u>0.40</u>	<u>1.46</u>	<u>0.83</u>
TOTAL	0.77	0.54	4.63	1.49
ASH	10.90	4.39	15.00	8.28
TOTAL	<u>100.00</u>	<u>100.00</u>	<u>100.00</u>	<u>100.00</u>
 <u>HEAT OF COMBUSTION (GROSS)</u> <u>AS RECEIVED, BTU/LB</u>				
	7,420	8,730	11,590	13,510

278

\* NOT TYPICAL: CHOSEN FOR HIGH SULFUR CONTENT

Table IX  
Comparison of Catalytic Liquefaction Runs Using Various Coals (8)

<u>RANK</u>	<u>GLENHAROLD MINE</u>	<u>BIGHORN</u>	<u>KENTUCKY #9 &amp; #11</u>	<u>PITTSBURG SEAM</u>
	LIGNITE	SUBBITUMINOUS	BITUMINOUS	BITUMINOUS
COAL SLURRY FEED CONCENTRATION, WT%	40	30	30	30
<u>RESULT SUMMARY</u>				
LIQUEFACTION, % (a)	95	91	92	90
<u>FILTRATE ANALYSIS</u>				
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

-----

$$(a) \% \text{ LIQUEFACTION} = \frac{\text{LB MAF COAL FED} - \text{LB MAF UNDISSOLVED COAL IN SLURRY PRODUCT}}{\text{LB MAF COAL FED}} \times 100$$

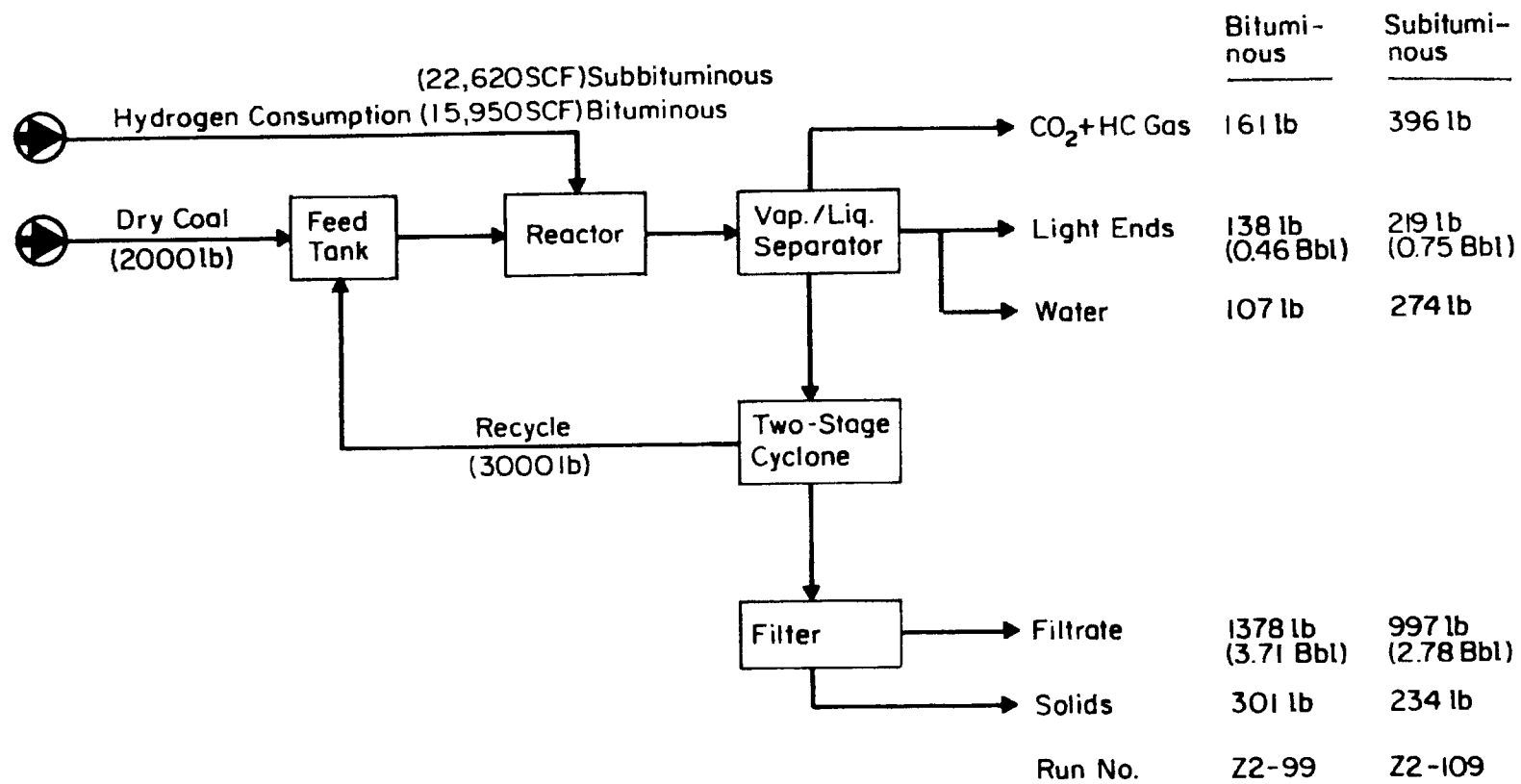


Figure 10. Process Material Balance for Bituminous and Subbituminous Coals (8)



(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.

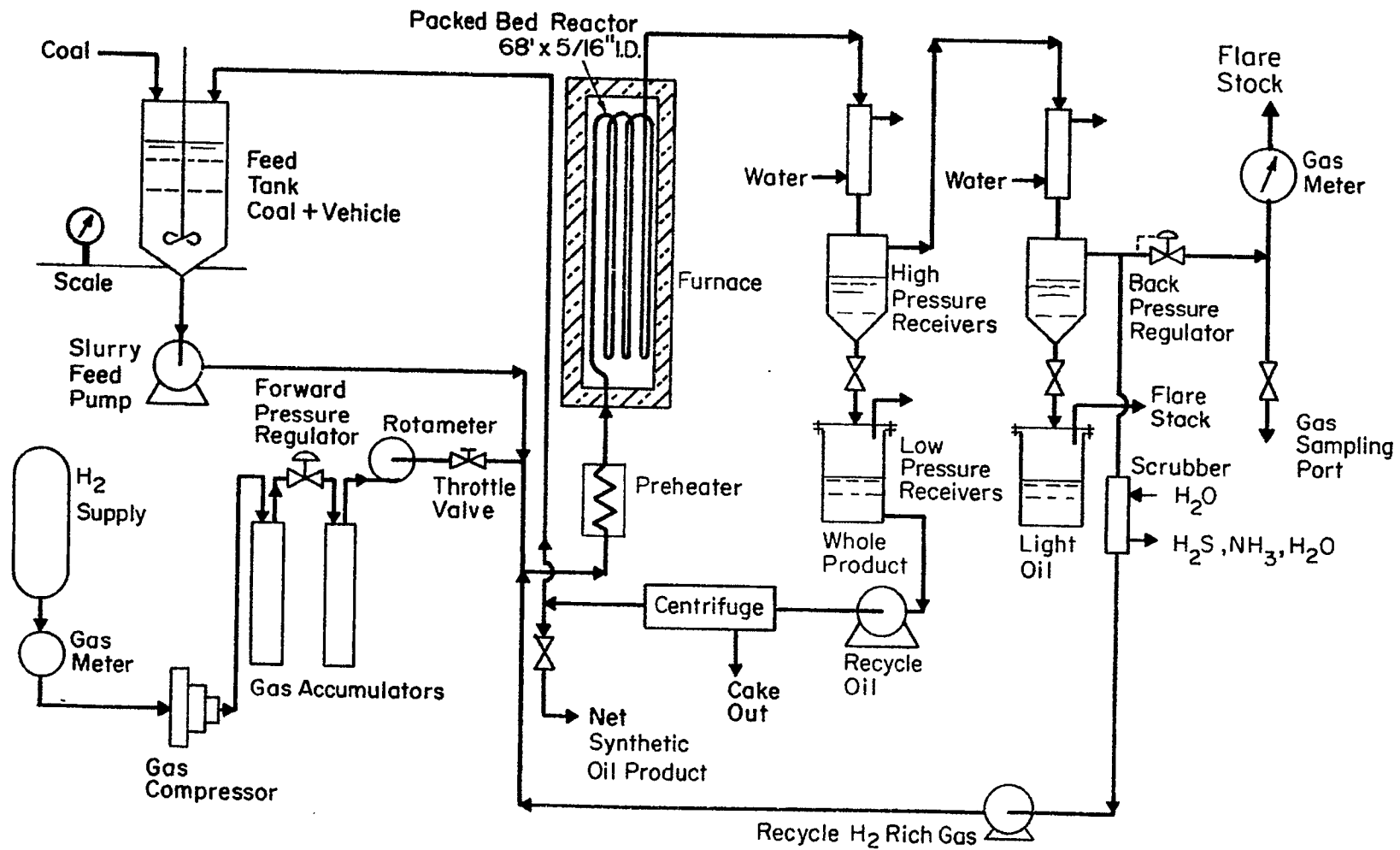


Figure 11. Schematic Drawing of the U.S. Bureau of Mines Synthoil Process Unit

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

Sulfur in feed coal, wt. pct. . . . .	4.6
Sulfur in recycle oil (product oil), wt pct . . . . .	0.19
Yield: bbl oil/ton coal maf . . . . .	3.0
Hydrogen consumption, scf/bbl . . . . .	3000.0
<u>Solvent Analysis of Product Oil, Wt Pct</u>	
Oil ( pentane-soluble) . . . . .	79.5
Asphaltene . . . . .	17.4
Organic benzene insolubles . . . . .	2.1
Ash . . . . .	1.0
<u>Elemental Analysis of Product Oil (Ash-Free), Wt Pct</u>	
Carbon . . . . .	89.9
Hydrogen . . . . .	9.2
Nitrogen . . . . .	0.6
Sulfur . . . . .	0.19
Viscosity of product oil, SSF at 180°F . . . . .	21-31
Calorific value of product oil, Btu/lb. . . . .	17,700

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/ft<sup>3</sup> reactor volume  
Gas recycle rate: 500 scfh

A. Gross Results

Sulfur in feed coal, st pct . . . . .	3.00
Sulfur in centrifuged oil, wt pct . . . . .	0.31
Yield, bbl oil/ton coal maf . . . . .	3.0
Hydrogen consumption, scf/bbl . . . . .	3000.0

B. Analyses of Centrifuged Product OilSolvent Analysis, Wt Pct

Organic benzene insolubles . . . . .	11.6
Abh . . . . .	1.3
Asphaltene . . . . .	24.4
Oils . . . . .	62.7

Elemental Analysis (Ash-Free Basis), Wt Pct

Carbon . . . . .	89.6
Hydrogen . . . . .	7.6
Nitrogen . . . . .	0.9
Sulfur . . . . .	0.31
Oxygen (by difference) . . . . .	1.6
Range of specific gravity, 60°/60°F . . . . .	1.126
	1.141
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

Organic benzene insolubles . . . . .	33.1
Ash . . . . .	27.7
Asphaltenes. . . . .	9.0
Oil . . . . .	30.2
Sulfur content, wt pct . . . . .	2.10



## CONSOLIDATION COAL COMPANY - CONSOL SYNTHETIC FUEL (CSF) PROCESS

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).

Pittsburg 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 accululate on and deactivate the catalyst. Continuous hydrogenation of extract to a 0.2% sulfur product in ebullated 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,

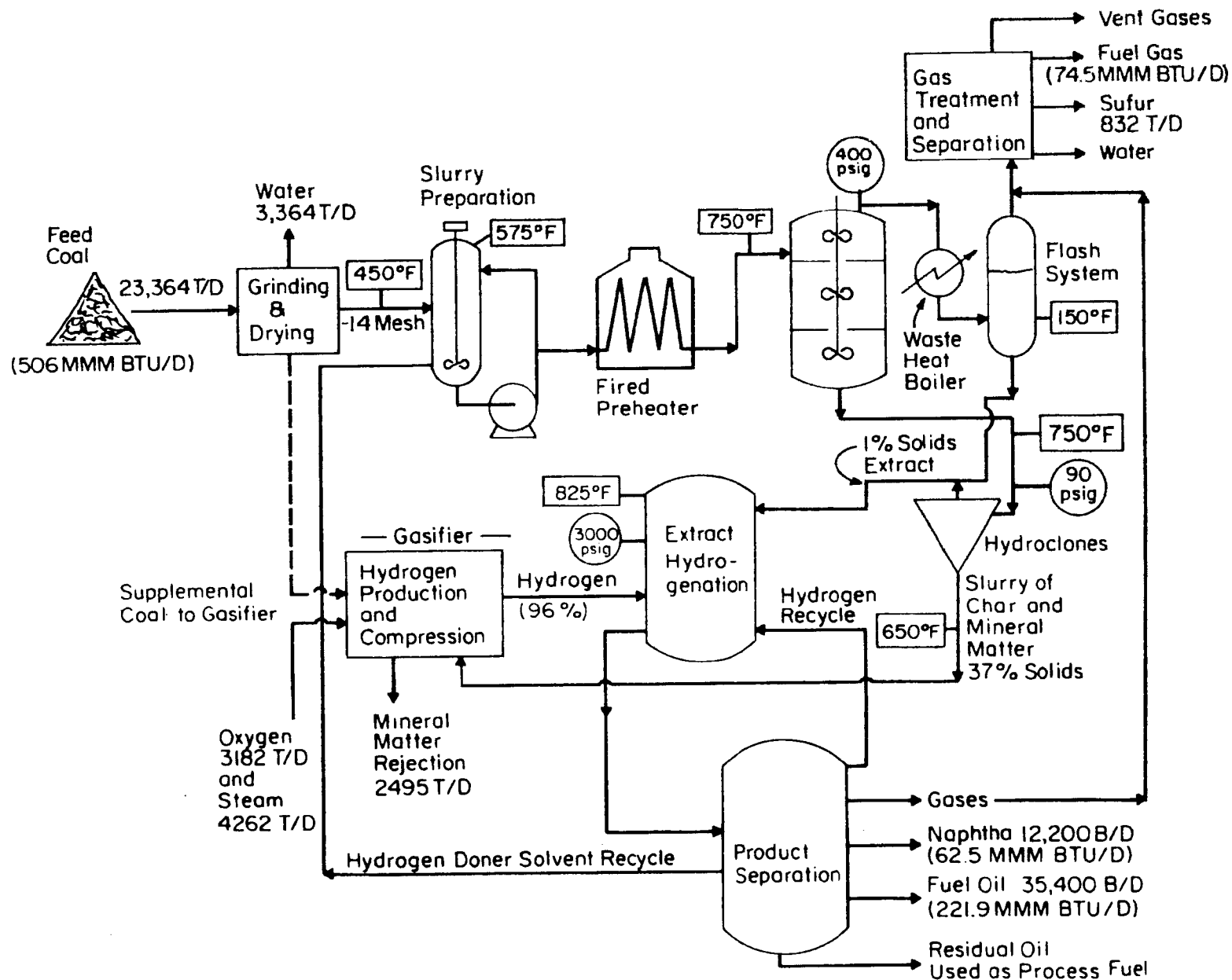


Figure 12. Consolidation Coal Co. CONSOL Synthetic Fuel (SCF) Process

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 XII

## Analysis of Liquefaction Process (Foster Wheeler Case A) (42)

	<u>Extraction</u>	<u>Hydrogenation</u>	<u>H<sub>2</sub> Mgf</u>	<u>Combined</u>
<u>A. Thermal Performance</u>				
<u>(10<sup>6</sup> Btu/hr)</u>				
<u>Input</u>				
Coal -Pittsburg Seam	21,000	-	-	21,100
H <sub>2</sub>	258 (1)	14,162	-	-
Hydro Feed	-	14,370	-	-
Char	-	-	4,900	-
Process Input	21,358	18,532	4,900	21,100
<u>Fuel</u>				
Process Furnaces	1,652	395	-	2,047
Steam Generation	-436	-133	2,338	1,769
Power Generation	196	89	326	611
Total Fuel	1,412	351	2,664	4,427
TOTAL INPUT	22,770	18,883	7,564	25,527
<u>Output</u>				
Low Btu Gas	636	-	-	636
Hi Btu Gas	356	2,736	71	3,163
Light Oil & Dist.	160	2,466	-	2,626
Heavy Oil	270	8,650	-	8,920
Hydro Residue	-	3,500	-	3,500
Fuel Output	1,422	17,352	71	18,845
<u>Intermediates</u>				
H <sub>2</sub>	-	258 (1)	4,162	-
Hydro Feed	14,370	-	-	-
Char	4,900	-	-	-
TOTAL OUTPUT	20,692	17,610	4,233	18,845
Output/Input, %	91.0	95.0	56.0	73.9
<u>B. Investment, MM \$</u>	72.3	78.5	103.1	253.9
<u>C. Processing Costs, MM \$/yr</u>				
Operating Costs (ex fuels)	6.50	10.20	11.30	28.00
Capital Charges	9.25	10.05	13.18	32.48

(1) Via donor solvent transfer

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## 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.

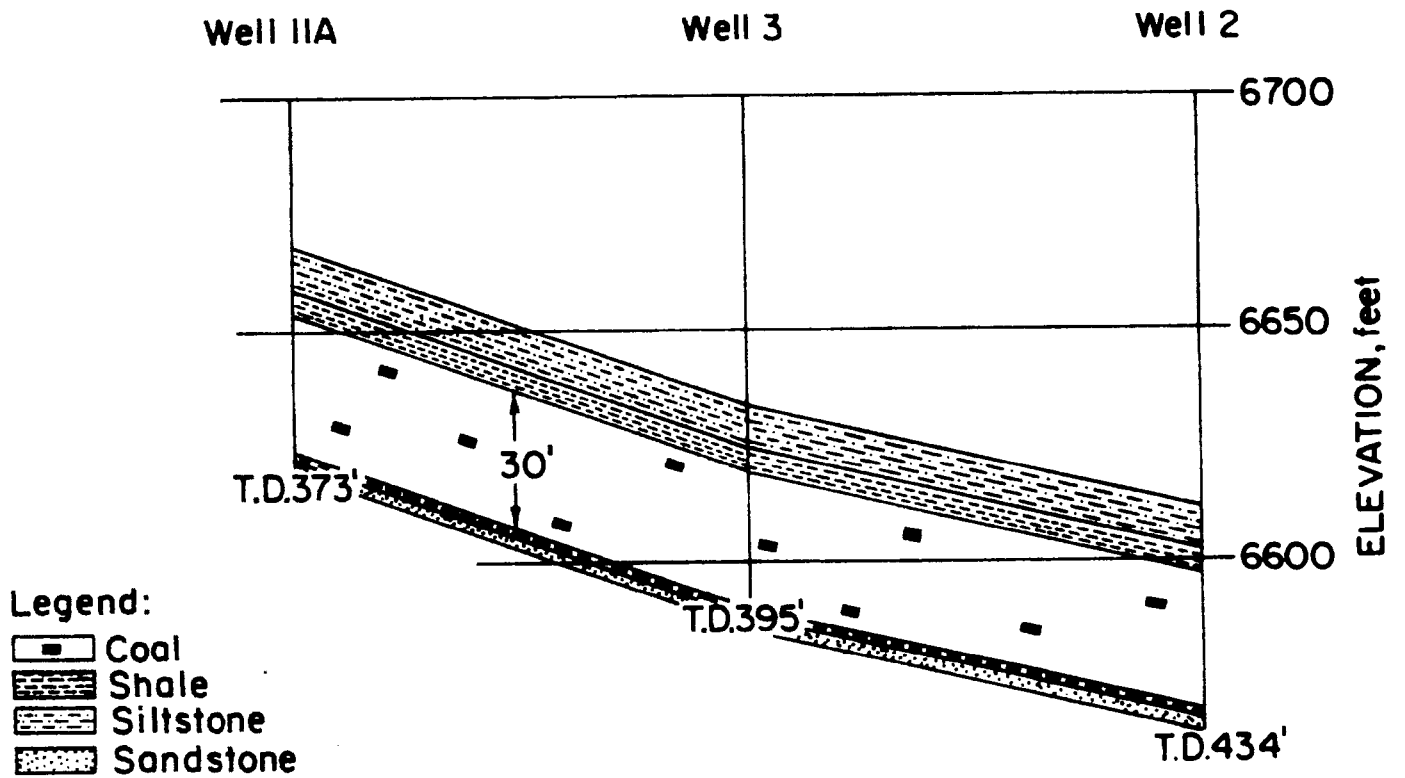


Figure 1. Section of Insitu Gasification Experiment, Hanna, Wyoming (3)

TABLE I

Typical Gas Analyses of Gas from Hanna  
In situ Gasification Experiment (3)

	Date			
	June 4	June 8	June 17	July 5
	Mole Percent			
H <sub>2</sub>	8.8	8.2	7.8	12.6
O <sub>2</sub>	9.7	1.0	.9	1.0
N <sub>2</sub>	68.4	50.7	53.9	54.8
CH <sub>4</sub>	4.3	16.5	11.9	6.6
CO	3.1	4.3	1.2	.2
C <sub>2</sub>	.5	1.6	1.4	1.0
CO <sub>2</sub>	5.0	17.1	22.2	21.8
Other	.1	.5	.7	2.0
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 losing 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.

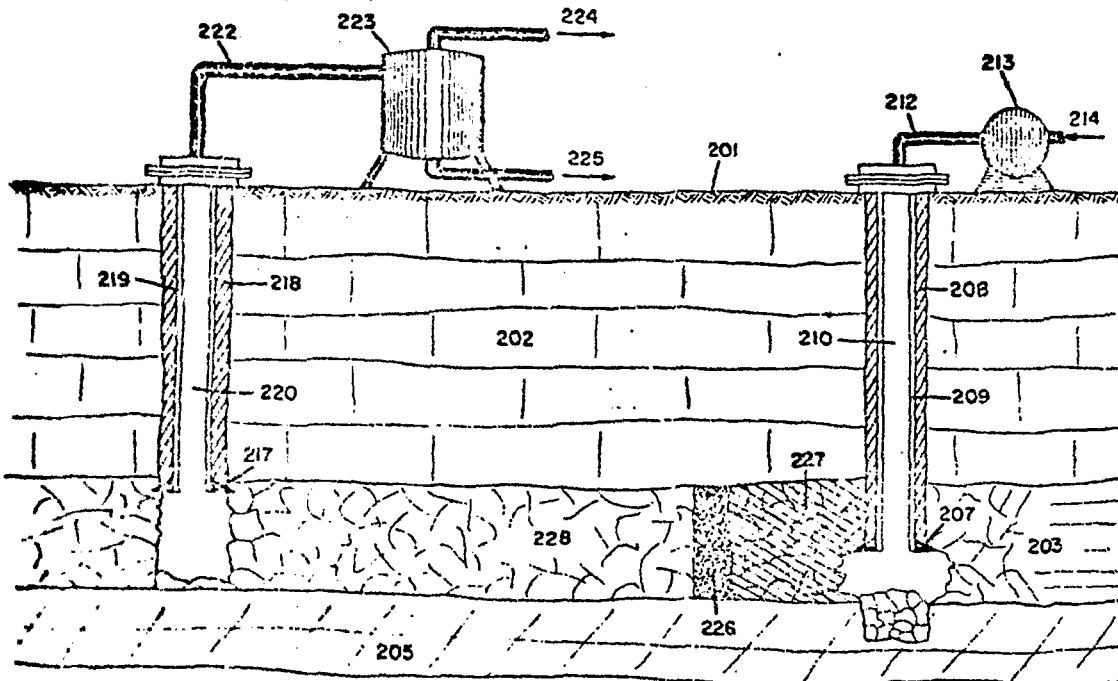
The non-uniformity of coal beds is worthy of evaluation. Tests have indicated that fractures represent the insitu permeability, while small units such as a cubic inch are of very low permeability. The distribution of the conduit system--major channels and micro interstices--would be helpful in designing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

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- 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 communitively. 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 underlying 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.

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