

RESEARCH AND DEVELOPMENT OF RAPID HYDROGENATION
FOR COAL CONVERSION TO SYNTHETIC MOTOR FUELS

(RISER CRACKING OF COAL)

Project 8976 Task Report: Task 3
Design of a Process Development Unit

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Date Published — May 1978

Prepared for the

UNITED STATES DEPARTMENT OF ENERGY

Under Contract No. EX-76-01-2307

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ABSTRACT

This report presents a detailed design of a process development unit (PDU) for the riser cracking of coal to produce fuel gases, high-octane motor gasoline blending stock, and fuel oil. At the maximum design capacity, 100 lb/hr of feed coal and 12 lb/hr of hydrogen can be processed. The riser reactor is not equipped with external heaters; the heat needed to accomplish the hydropyrolysis will be generated by the combustion of small quantities of oxygen or air injected into the riser reactor.

Because of the "balanced pressure" design, operating temperatures as high as 1800°F at 2000 psig can be readily obtained, allowing the PDU to be operated in an SNG production mode, as well as the lower temperature pyrolysis for the production of liquid fuels.

A. INTRODUCTION

The purpose of this report is to present a detailed design of a process development unit for the riser cracking of coal. This design has been predicated upon the results of experiments carried out in a bench-scale unit that was built and operated during the execution of Task 2; at present, more than 30 runs have been made. The results of the experimental work, which is ongoing, are summarized in Figure 1, which shows a uniform increase in the conversion of the North Dakota lignite feed with an increase in the severity of thermal treatment, and Figure 2, which shows the change in distribution of feed carbon between char, liquids, and gases with an increase in the severity of thermal treatment.

In Figure 1, the conversions are on the dry, ash free (DAF) basis. The severity of the thermal treatment is a function of temperature and residence time, and, over the range of experimental conditions explored thus far, both the conversion of the DAF lignite to products and the distribution of feed carbon among char, liquids, and gases appear to be practically independent of the manner in which the severity of the thermal treatment is developed. Over the range of experimental conditions used, the effect of operating pressure does not seem to be large, because the data taken at 2000 psig operating pressure (indicated by solid points in Figure 1) can be organized with the data taken at 1500 psig operating pressure, with all data showing the same degree of scatter about lines drawn through the data points.

1. Pyrolysis Mechanism

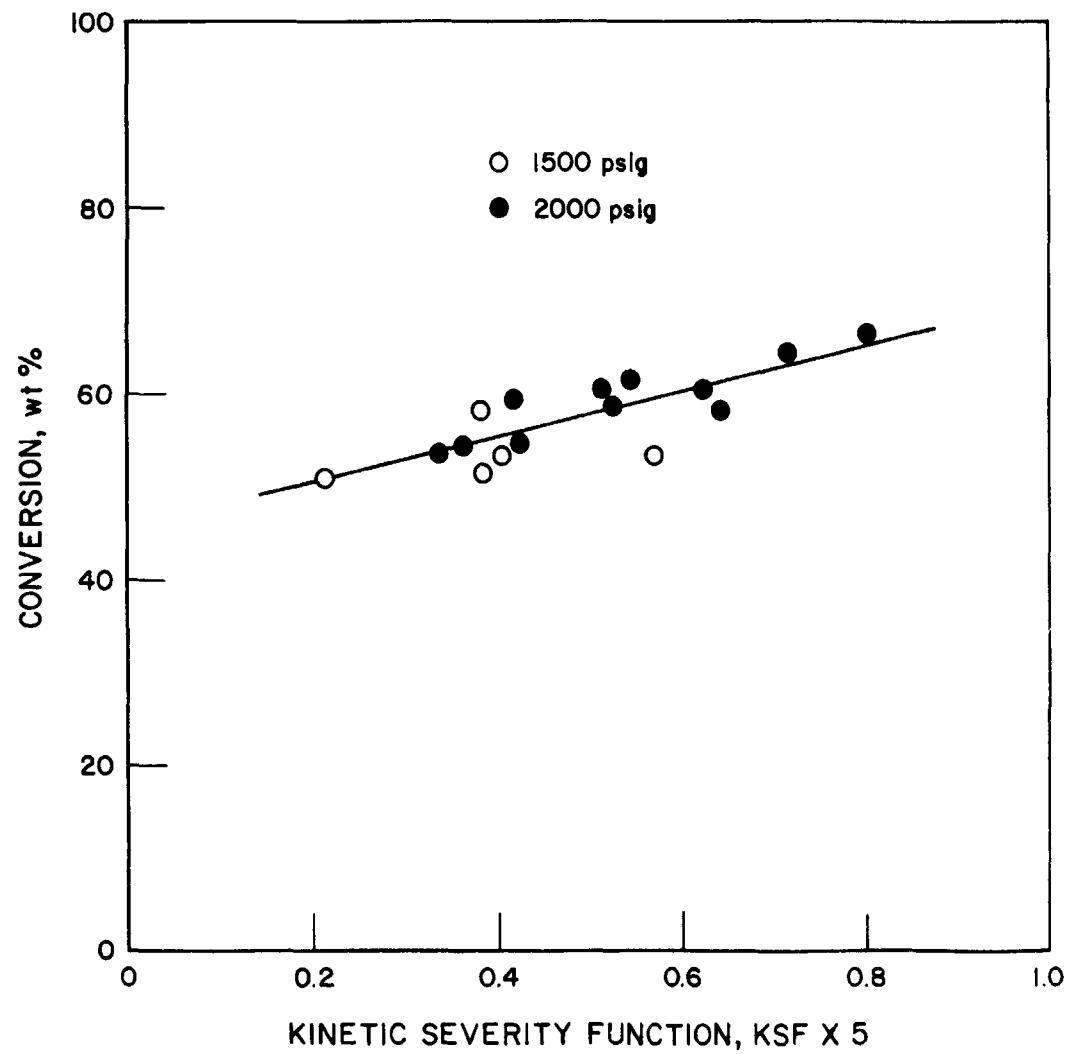
The mechanism of the pyrolysis of lignite in hydrogen at high pressure appears to follow the model proposed by Russel, Saville, and Greene (7) and also Johnson (5), who postulated that in the initial stages, depolymerization and decomposition reactions result essentially in a first order conversion of the feed carbonaceous solids to volatile species that flow out of the particles into the bulk gas phase. This flow is considerable, so there is little opportunity for hydrogen to approach the gas-solids interfaces by counter-diffusion to react directly with the carbonaceous solids.

In the bulk gas phase, however, the species liberated from the feed coal can react further with the hydrogen of the bulk gas phase. At high temperatures, molecular fragmentation would continue, and the free radicals produced would be stabilized by the large excess of hydrogen to form the most refractory species possible. Thus, alkylated aromatic species would be dealkylated to form benzene, methane, and to a lesser extent, ethane. At lower temperatures, the continued fragmentation would not be as pronounced, and alkylated species such as toluene, xylene, substituted naphthalenes, and phenol would survive the pyrolysis to appear among the final products. Detailed studies of the products of the hydropyrolysis of lignite in the bench-scale unit have shown the pyrolysis to follow this mechanism.

2. Overall PDU Concept

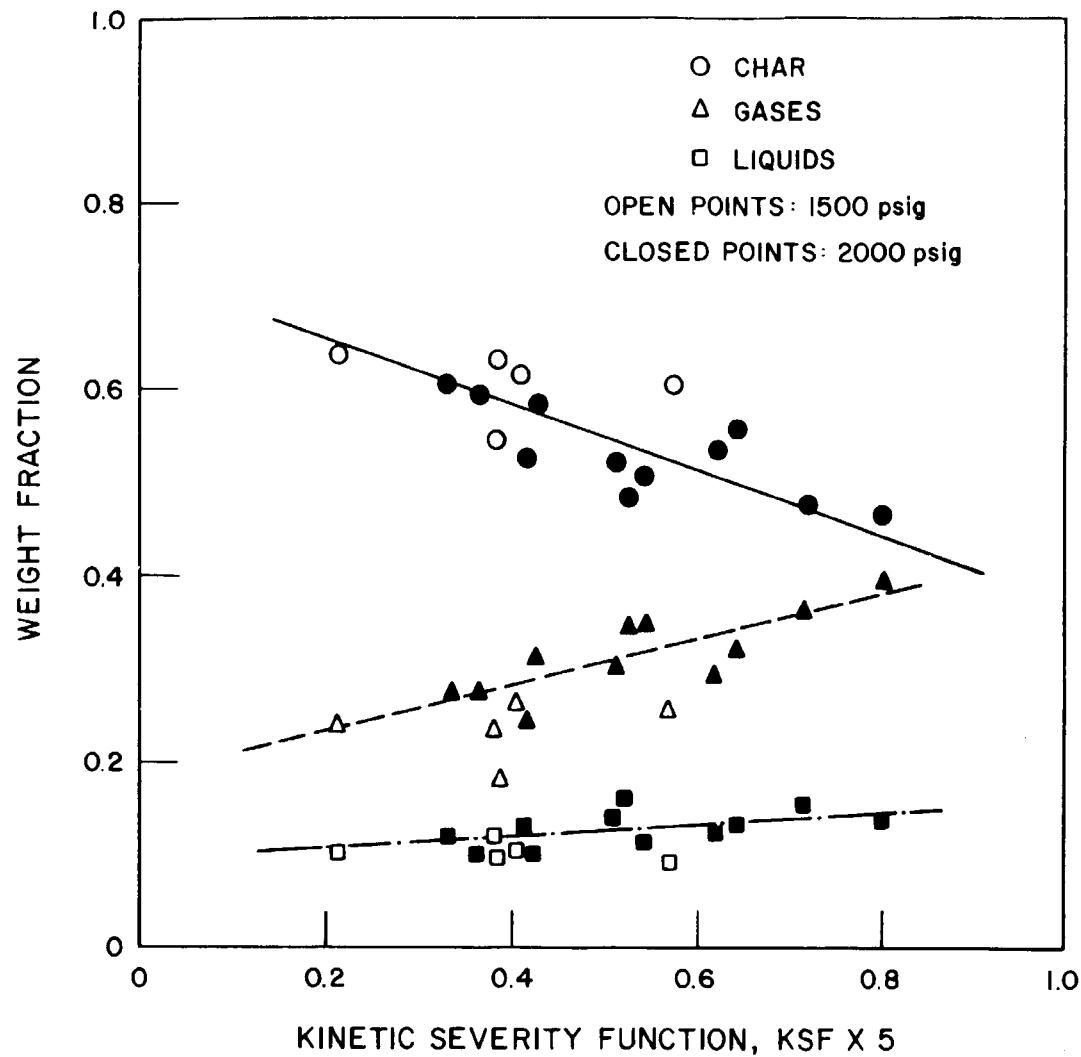
A simplified flow diagram showing the principal equipment of the proposed PDU is shown in Figure 3. In operation, feed coal charged to the feed hopper would be metered into the main carrier gas stream. The mixture of coal and hydrogen or other feed gas would then pass through a radiantly heated coil in

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Figure 1. CHANGE IN CONVERSION WITH INCREASINGLY SEVERE THERMAL TREATMENT, DAF BASIS



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Figure 2. CHANGE IN CARBON DISTRIBUTION WITH
INCREASINGLY SEVERE THERMAL TREATMENT

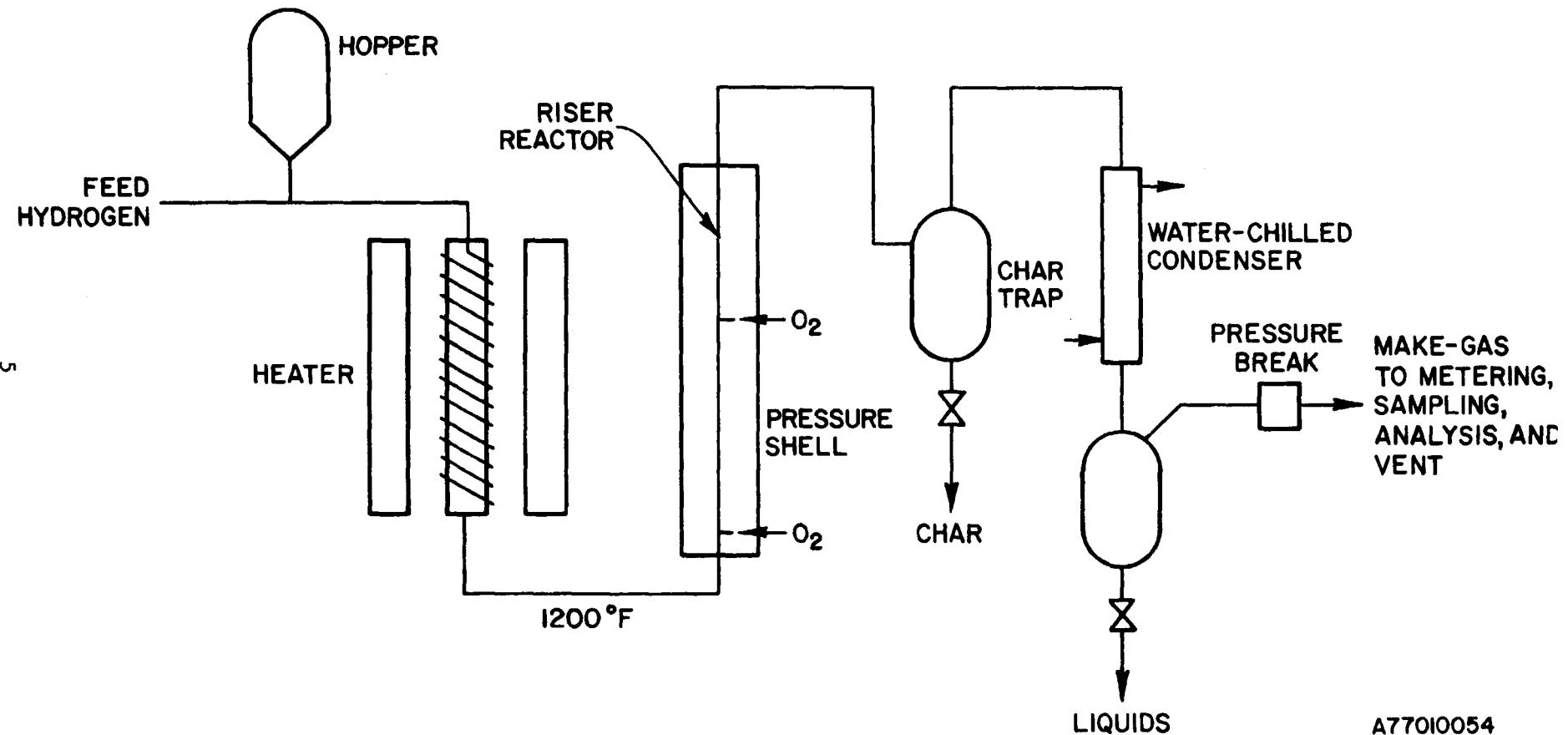


Figure 3. SIMPLIFIED FLOW DIAGRAM OF RISER REACTOR PDU

which the mixture would be heated to 1200°F. The preheated stream of coal and hydrogen would then pass into the bottom of the riser tube and move upwards. A short distance above the bottom of the riser tube, air or oxygen would be injected into the mixture of hydrogen and coal. The oxygen-hydrogen mixture would ignite on contact because the bulk stream temperature at the point of injection would be well above the ignition point. The amount of oxygen injected would be controlled so that the bulk temperature of the coal-hydrogen mixture is raised to no higher than 1400° to 1500°F. The combustion would take place in a region of turbulent mixing rather than in a discreet flame and would be completed within a few inches of riser tube length, so that the temperature changes would be essentially step changes. Two combustion stages are provided in the design so that the final outlet temperature can be attained in two steps. The riser tube is well insulated, so that the pyrolysis would essentially take place under adiabatic conditions.

Near the top of the riser tube, a slip joint is provided to take up the increase in the length of the riser tube due to thermal expansion. In operation, a small quantity of nitrogen from the pressure shell will be bled into the product stream to prevent reaction products from accumulating in the pressure shell, and also to partially cool the stream to stop chemical reactions and prevent rupture of the overhead line, which is not supported by a balanced pressure shell.

The reaction products would then be carried into the hot char trap where the spent char would be disentrained from the make gas, collecting as a dry powder in the bottom of the char trap. The effluent gases from the char trap would next be cooled by indirect heat exchange with cooling water to condense the liquids that would be collected in the liquid products accumulator, which would be maintained at room temperature. The stripped make-gas passing from the liquid products accumulator would then be reduced in pressure, dried and chilled to "wring out" remaining condensable hydrocarbons, sampled, analyzed, and vented in a manner very similar to that used in the present bench-scale unit.

3. Scope of Investigation

It is appropriate at this point to enumerate some of the processing concepts incorporated into the design of the PDU and to discuss how these processing concepts compare and complement other investigations in progress.

The proposed PDU equipment is relatively simple, is capable of high mass throughputs, and can be built on any scale using available construction techniques and materials. The feed to the riser reactor is heated to 1200°F, so that only small quantities of air or oxygen will be required to raise the bulk stream temperature the additional 200° to 300°F needed to accomplish the pyrolysis.

In the combustion zone, the feed coal particles will be exposed to very high temperatures and radiant heating by incandescent gases and particles caught in the eddies where the combustion is taking place. The temperature gradients in these regions would be large and could well exceed those obtained when the hydrogen is heated in a separate process step (6, 7, 8). The radiant character of the heat transfer would also tend to overcome the reduction in convective heat transfer caused by the outgassing of volatiles from the particles. This exposure to very high heating rates would be for very short time periods

and would be terminated by turbulent mixing that would equalize the temperature to the final bulk-stream temperature. In overall effect, a portion of the coal-hydrogen mixture would be heated to very high temperatures and then quenched to the final bulk-stream temperature in a very short time interval. We believe that this process concept is a logical extension of investigations currently in progress and would enlarge the technologies for converting coals and lignites to clean, convenient fuels by a significant degree.

In the proposed PDU, the operating mode could be shifted from conditions favoring the production of gaseous and liquid fuels to conditions favoring the production of gaseous fuels by raising the final bulk temperature of the coal-hydrogen mixture. We believe that this potential flexibility is an important attribute of the processing concept under investigation.

4. Feedstock Coals

The PDU design presented here would be expected to operate on lignites or non-caking Western coals without modification. Three methods might be used to allow the processing of caking coals: 1) pretreatments external to the hydropyrolysis process train, 2) modifications to the methods used to operate the process, or 3) modifications of the equipment used to operate the process.

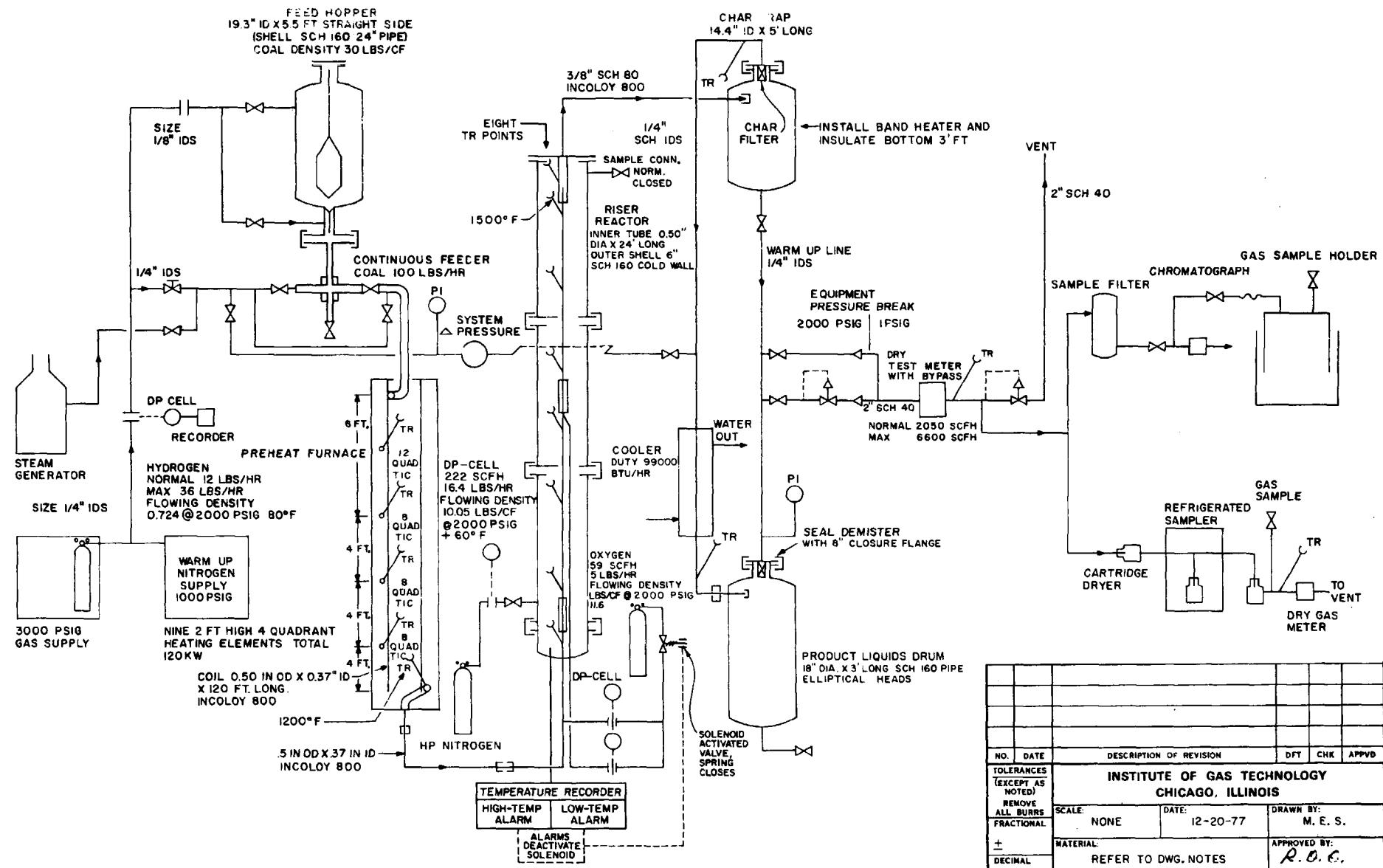
A number of pretreatment processes have been developed (1, 2, 4) that are intended for the reduction of sulfur in coals. These pretreatment processes also destroy the agglomerating properties of caking coals, and one (2) has been reported to actually enhance the reactivity of the treated coal. These processes are executed in aqueous slurries and could be adapted for use in hydropyrolysis by utilizing the slurry to bring the feed coal to the operating pressure of the process. A dewatering and drying step at pressure would be needed to prepare the feed coal for entrainment in the feed stream to the pyrolysis reactor. Any reduction in sulfur content of the feed coal obtained in the pretreater would lessen the need for gas cleaning in the subsequent combustion of the spent char to produce steam and synthesis gas.

Internal modifications of the process such as recycling char or shock heating the feed coal might also be used. The choice of method would be dependent upon the coal to be used and its specific caking properties. A more complete discussion of the many techniques that might be used are presented in a subsequent section of this report.

B. PDU DESIGN

In this section, detailed descriptions of the equipment to be used in the construction of the PDU (Figure 4) will be presented. The intended goal of this design is to construct equipment that will simulate the pressures, temperatures, and flow conditions in a riser reactor adapted for the hydropyrolysis of coal and lignite. Feed solids will be transported through the process in the dilute phase, and the riser reactor will operate as a lift-line operating in the dilute phase. The velocity of the gas will be maintained at values well above choking velocities so that choking and slugging are avoided.

A commercial riser reactor would normally be provided with an erosion-resistant refractory liner within a steel shell capable of withstanding the stresses imposed by the operating pressure of the system. This method of



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construction is not feasible on the small scale considered here, so an equivalent method of construction will be used. In the proposed construction, the riser will be contained in a pressurized shell so that only nominal stresses are developed in the walls of the riser tube itself. The space between the riser tube and the pressure shell will be filled with insulation to control heat loss from the system.

1. Basis for Design

The PDU will have a maximum throughput of 100 lb/hr, and this design is based on the operating conditions summarized in Table 1 below:

Table 1. DESIGN OPERATING CONDITIONS

Lignite Feed Rate, lb/hr	100
Hydrogen Feed Rate, lb/lb lignite	0.12
Riser Inlet Temperature, °F	1200
Riser Outlet Temperature, °F	1500
Operating Pressure, psig	2000

In the course of the experimental investigation, it will be desirable to vary both operating pressure and residence time; the effect of increasing residence time and reducing operating pressure on coal feed rate and total reactor throughput is summarized in Table 2. In Table 2, the hydrogen/coal ratio is assumed to be maintained at 0.12 lb/lb.

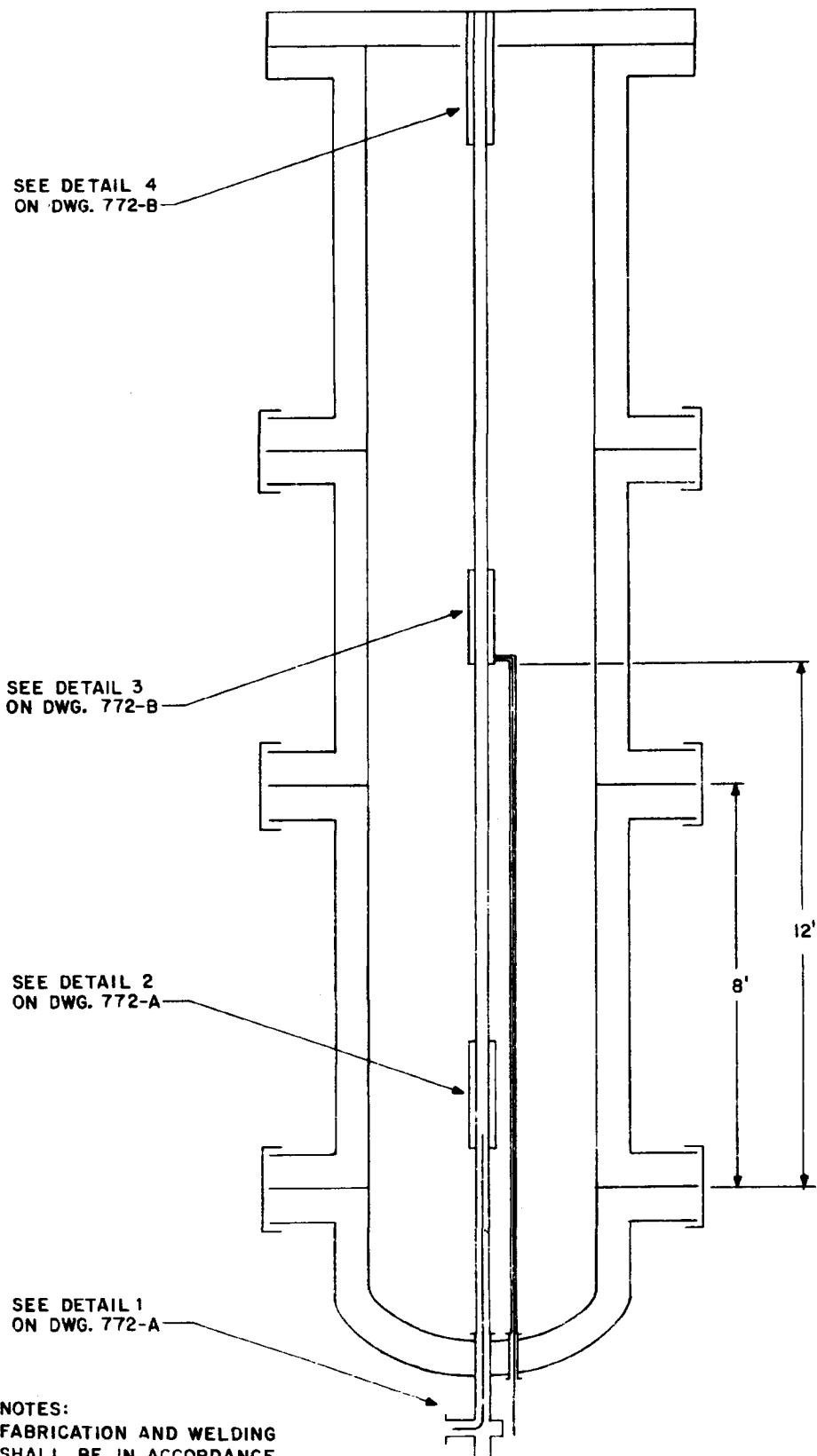
Table 2.
NOMINAL RESIDENCE TIMES AND THROUGHPUTS
FOR VARIOUS OPERATING CONDITIONS AT 1500°F

Residence* Time, s	Pressure, psig	Superficial Velocity, ft/s	Solids Feed Rate, lb/hr	Throughput, lb/hr-sq ft
1.0	2000	23.0	99.54	149,309
2.0	2000	11.5	49.77	74,655
1.0	1500	23.0	74.83	112,244
2.0	1500	11.5	37.42	56,122
1.0	1000	23.0	50.13	75,195
2.0	1000	11.5	25.07	37,597

* Based on superficial gas velocity at 1500°F.

2. Riser Reactor Design

An overall view of the riser reactor and pressure shell is shown in Figures 5A and 5B. The riser tube will be fabricated from 0.5-inch OD by 0.37-inch ID Incoloy 800 tubing enclosed in a pressurized shell. The outer shell will be built from three 8-foot sections of 6-inch schedule 160 steel pipe equipped with flanges. The outer shell will be capable of a maximum working pressure of 2580 psig, but will be limited by the rating of the 1500 psig flanges at 650°F. The space



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Figure 5A. RISER REACTOR

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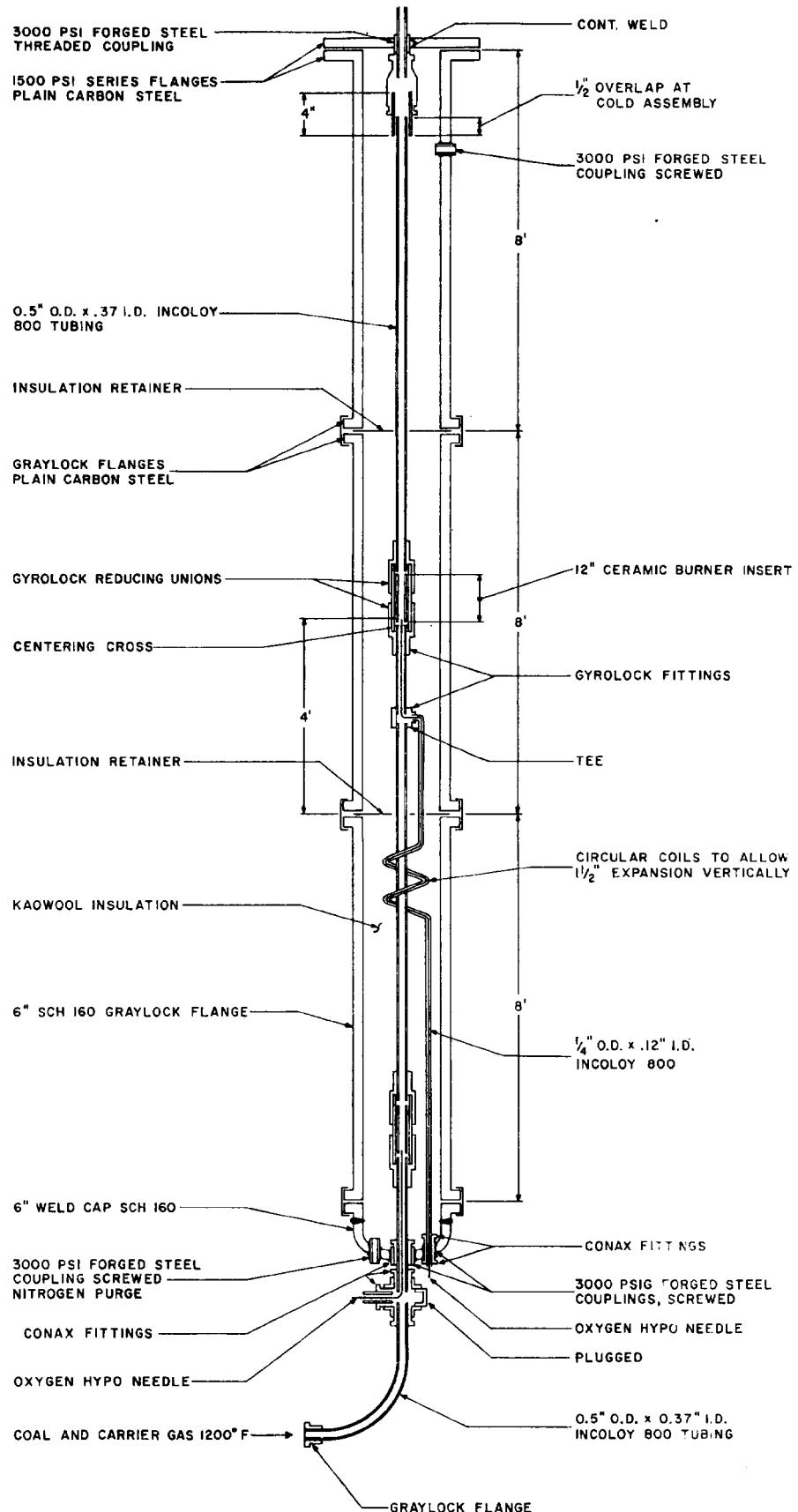


Figure 5B. RISER REACTOR

between the riser tube and the pressure shell will be filled with compacted Kaowool insulation to minimize convection currents within the pressure shell. High pressure nitrogen will be admitted to the high-pressure shell through a tap located in the lowest section of 6-inch pipe.

The riser tube and auxiliary connections will be made through a stationary bottom head that will support the entire reactor assembly (Figure 6). The bottom head will be attached to the upper section of the pressure shell by means of a Grayloc connection; both the riser tube entry and upper-combustion-zone oxygen line entry will be made using Conax fittings to avoid the difficulties of welding thin metal sections to the heavy pressure shell walls.

3. Lower Combustion Zone

A detailed sketch of the lower combustion zone is shown in Figure 6. The hypodermic needle for introducing the oxygen will be passed through the lower section of the riser tube to a point 8 inches above the entry point and will be centered in the riser tube by means of fixtures attached to the injection needle at several points. The metal tube forming the riser tube will be enlarged in the vicinity of the point of oxygen injection so that a ceramic insert can be positioned as shown. In the combustion zone, the ceramic insert will serve as the interior wall of the riser tube.

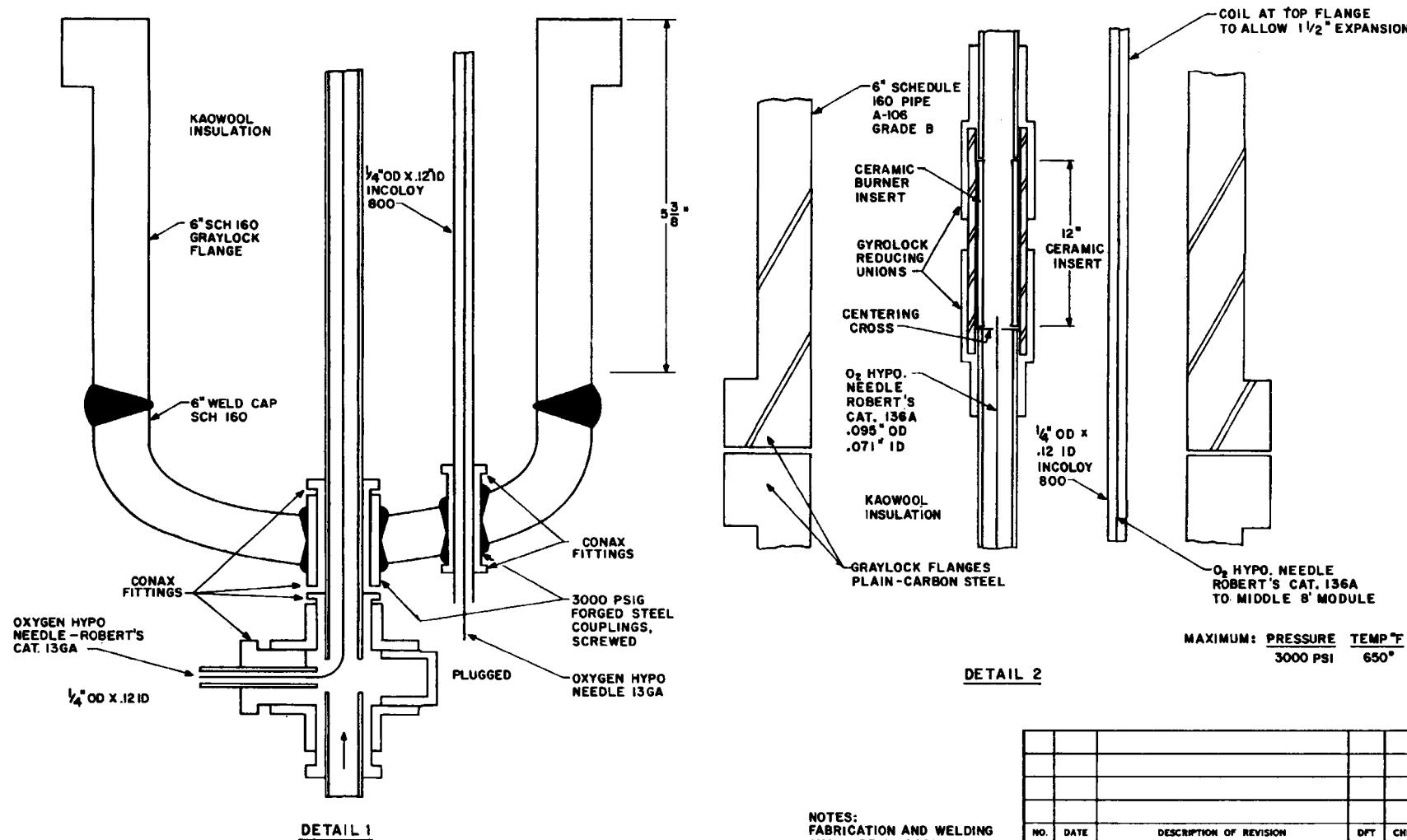
4. Upper Combustion Zone

A second combustion zone similar in construction is positioned 12 feet from the bottom (Figure 7). Both combustion zones are positioned near the flanges connecting sections of the outside pressure shell to facilitate construction of the apparatus. The tubing used to guide the oxygen needle from the bottom entry to the upper combustion zone will be wound into a coil having two turns to take up thermal expansion of the riser tube. The upper combustion zone will be fitted with a ceramic insert in a manner similar to the lower combustion zone.

Near the top of the pressure shell, the riser tube will be provided with a slip joint (Figure 7) to take up the thermal expansion of the riser tube assembly. In operation, a small bleed of high-pressure nitrogen will be maintained that will prevent reaction products from accumulating within the pressure shell. This bleed stream will also cool the riser effluent, stopping reaction and avoiding stress in the unsupported tubing downstream from the pressure shell.

5. Reactor Temperature Control

Temperature control will be accomplished using the bulk temperature of the gas stream downstream from the combustion zone as the feedback signal to control the quantity of oxygen or air burned. As depicted, control will be manual for both combustion zones. Using available calculation methods (3), it has been estimated that combustion of the oxygen will be completed within a few inches of the injection point. It is doubtful that a discreet flame will exist, but rather a turbulent combustion zone. The presence of the solids will afford some protection of the reactor wall by virtue of their opacity.



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Figure 6. RISER REACTOR - DETAILS 1 & 2

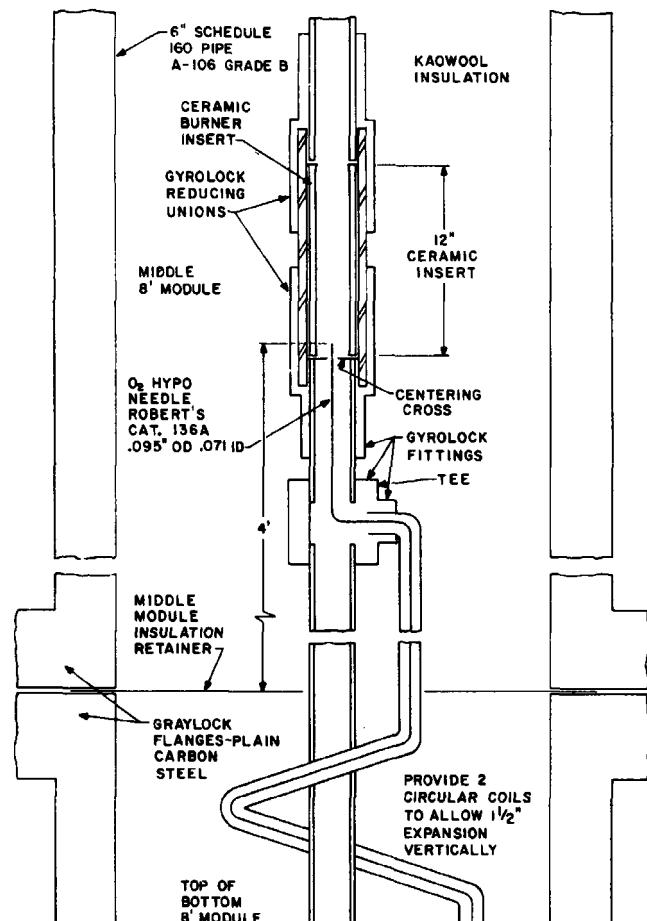
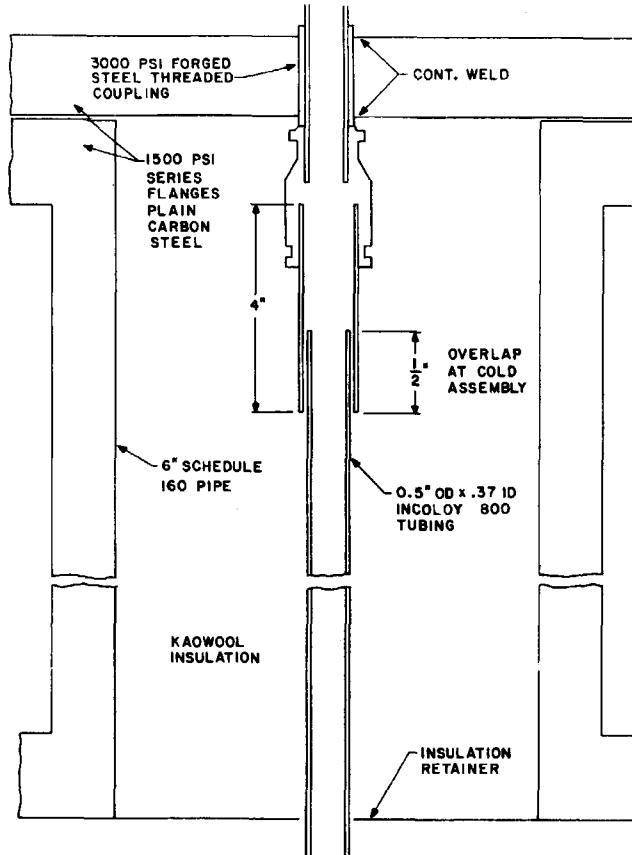


Figure 7. RISER REACTOR - DETAILS 3 & 4



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6. Coal Feed Hopper and Continuous Feeder

The coal feed hopper (Figure 8) has been sized to hold 500 pounds of coal and will be built from a section of 24-inch OD pipe 5-1/2 feet in length fitted with elliptical heads. The top head will be fitted with an 8-inch handhole for charging the hopper and introducing and positioning internals needed to control the flow of coal from the hopper. The coal will discharge from the hopper through a 4-inch nozzle fitted to the bottom head, flowing through a funnel fitted into the 4-inch nozzle into the 1.5-inch ID plenum chamber. As in both the cold flow model and the bench-scale unit, the feed coal will be carried down into the carrier gas stream by means of a small stream of gas introduced into the free space above the plenum through a tap on the side of the 4-inch nozzle. The mixing block in which the feed coal will be entrained in the main carrier gas stream will be machined from a single block of steel and it will be welded to the 1.5-inch plenum chamber which will, in turn, be attached to the 4-inch nozzle by means of a Grayloc fitting, allowing easy disassembly for maintenance and adjustment. After fabrication, the inside of the feed hopper will be coated to prevent corrosion and scale formation that might dislodge and plug the feed system.

7. Carrier Gas Supply

The carrier gas supply is designed to provide hydrogen at the rate of 2275 SCF/hr for from 3 to 4 hours at a supply pressure of 2100 psig. This will be partitioned as shown in Figure 5 into the main carrier gas stream and a small stream used for feed rate control. This small stream will entrain the coal in the feeder plenum chamber and carry it down into the main carrier gas stream.

8. Preheat Furnace for Reactor Feed System

The preheater furnace has been designed to heat a maximum of 100 lb/hr of coal and 12 lb/hr of hydrogen from ambient temperature to 1200°F. The heat duty for these conditions has been calculated to be 81,600 Btu per hour. The pre-heater will be built as a down-flowing 120-foot coil fabricated from 0.5-inch OD by 0.37-inch ID Incoloy 800 tubing wound on a 6-inch-diameter SS 304 mandrel 18 feet in length. The coil and mandrel will be enclosed in a furnace containing 9 sections of 24-inch high by 8-1/4-inch ID quadrants containing the heating elements. The controls and other details for the preheater furnace, which will require 120 kW to operate, are shown in Table 3.

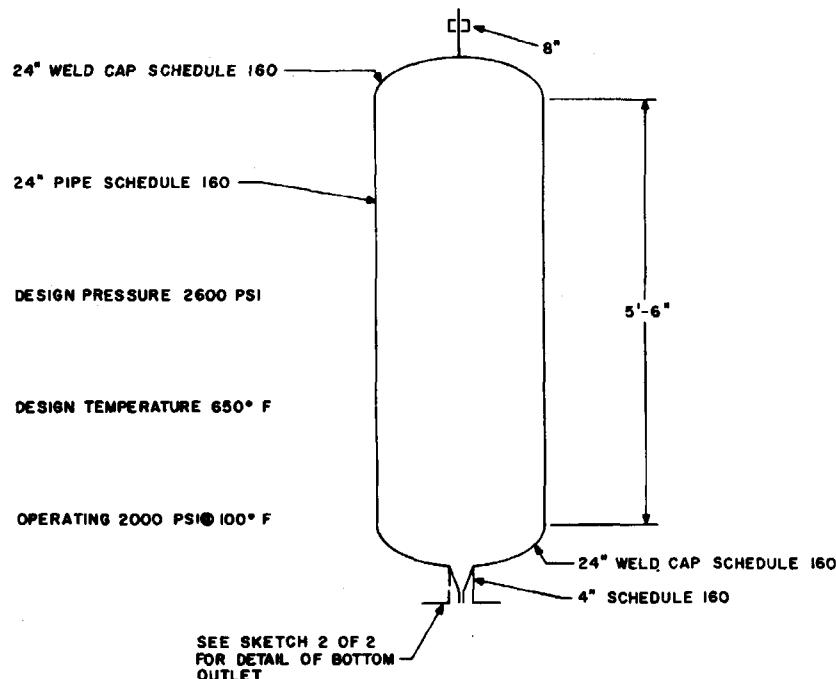
9. Unit Warm-Up Requirements

The equipment downstream from the preheater will be warmed to start-up temperatures by passing nitrogen at 1000 psig through the system with the pre-heater operating. The preheater will be operated at 100% over design duty to accommodate the warm-up load of 555 lb/hr nitrogen. Using this quantity of nitrogen, it is estimated that the PDU can be brought to appropriate start-up temperatures in about 3 hours.

10. Char Trap

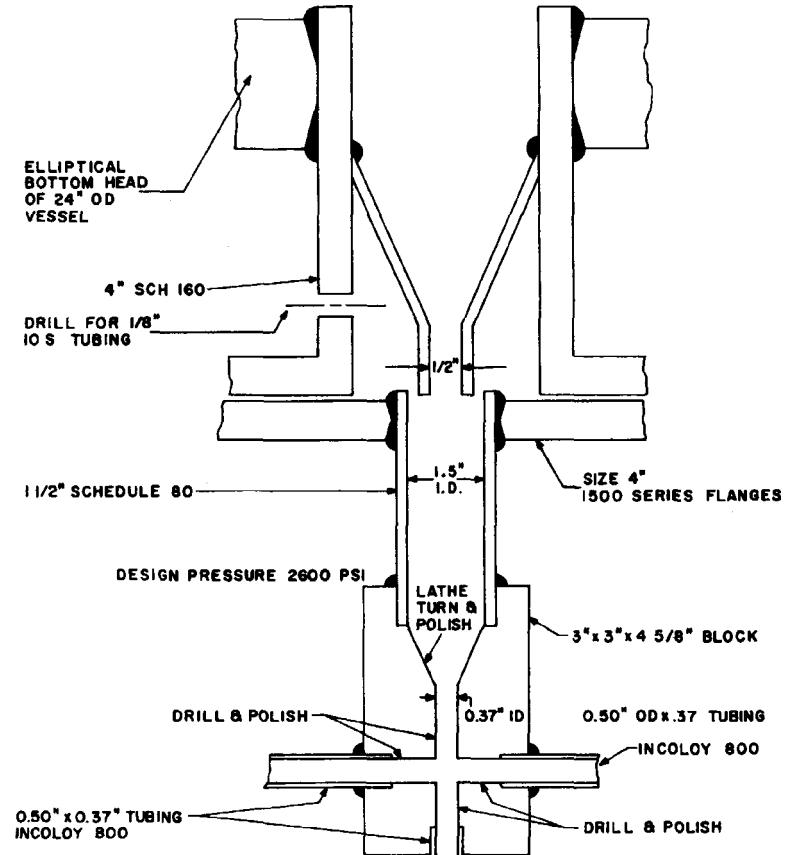
This vessel (Figure 9) has been sized to hold 250 pounds of spent char having an assumed density of 25 lb/cu ft and will be fabricated from a 5-foot length

FEED HOPPER
SKETCH 1 OF 2



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SKETCH 2 OF 2



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Figure 8. CONTINUOUS COAL FEEDER

Table 3. PREHEAT FURNACE ARRANGEMENT DETAILS

/1

Heating Elements - 4 Quadrants					Thermo-couple Location	Temp Recorder Points	Mandrel*		Coil**	
Item Number	Height, ft	Output, kilowatts	Controls Grouping	Item No.			Interval, ft	Accum. from Inlet, ft	Turns in Interval	Length in Interval, ft
1	2	13.4				5				
2	2	13.4	1,2&3	TIC-1	O	C	6		23	41
3	2	13.4			C	1	6			41
4	2	13.4					4		15	27
5	2	13.4	4 & 5	TIC-2	O	C	10			68
6	2	13.4					4		15	27
7	2	13.4	6 & 7	TIC-3	O	C	14			95
8	2	13.4					4		14	25
9	2	13.4	8 & 9	TIC-4	O	C	18			120
Total	18	120.6	4 Controls		4 Total	5 Total			67	120

* Mandrel Data: 6-inch OD X 0.12-inch thickness. Material 304 stainless steel.

** Coil Data: 0.5-inch OD X 0.37-inch ID X 120 feet long. Material Incoloy 800.

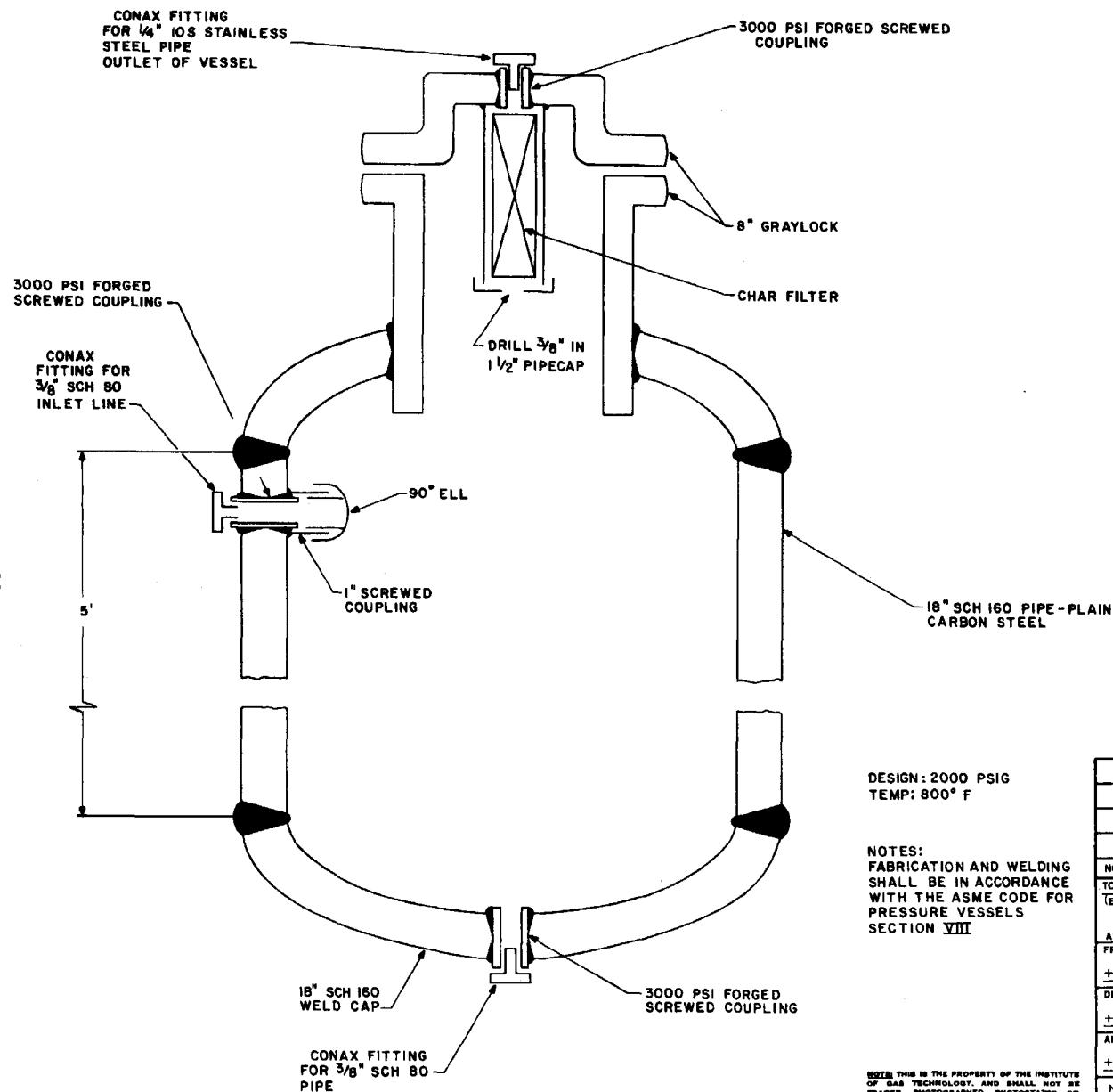


Figure 9. CHAR TRAP

DESIGN: 2000 PSIG
TEMP: 800° F

NOTES:
FABRICATION AND WELDING
SHALL BE IN ACCORDANCE
WITH THE ASME CODE FOR
PRESSURE VESSELS
SECTION VIII

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		DESCRIPTION OF REVISION			DFT	CHK	APPVD
INSTITUTE OF GAS TECHNOLOGY CHICAGO, ILLINOIS							
SCALE:	DATE:	12-20-77	DRAWN BY:	M. E. S.			
MATERIAL:	CARBON STEEL EXCEPT AS NOTED		APPROVED BY:	R. G. G.			
TITLE: PDU FOR RISER CRACKING OF COAL CHAR TRAP							
IGT INSTITUTE OF GAS TECHNOLOGY	PROJECT NO	DRAWING NO.	REV.				
	8976	773					

of 18-inch schedule 160 pipe equipped with elliptical heads. The top head will be fitted with an 8-inch handhole and cover plate. To hold the temperature of the char trap above the condensation point of the vapors, a circumferential band heater will be provided and the bottom two-thirds of the char trap will be insulated.

The inlet line to the char trap will pass through a Conax fitting positioned in the side of the vessel as shown and a 90° ell will be attached to the inside to impart a tangential motion to the gas. The gases emerging from the char trap will pass through a filter fitted to the cover plate to prevent particulates from collecting with the condensed liquids.

11. Product Cooler

Cooling of the make-gas will be accomplished in a coil of 1/8-inch SS (stainless steel) tubing wound on a mandrel and enclosed in a shell in which cooling water flows countercurrent to the make-gas.

12. Liquids Products Accumulator

This vessel (Figure 10) will be constructed of a 3-foot length of 18-inch schedule 160 pipe equipped with elliptical heads. The top head will be provided with an 8-inch nozzle and closure flange. A small demister will be located in this nozzle, and its top will be sealed by the closure flange.

13. Metering and Sampling System

The make-gas, stripped of condensable liquids emerging from the liquid products accumulator, will pass through a back-pressure regulator that holds the entire system pressure. The make-gas proceeds to a dry-test meter operating at slightly above atmospheric pressure. Approximately one-half of the stripped make-gas will be released through a low-pressure regulator to a vent stack. The remainder of the gas will be passed through a dryer and cold-trap in which the remaining condensable liquids are removed from the make-gas. This portion of the gas is warmed to room temperature and passed through a meter, allowing the quantity of liquids collected in the cold trap to be related to the quantity of gas carrying the condensable liquids.

A provision will be made to perform on-line chromatography on the make-gas passing the back-pressure regulator and on the stripped gases emerging from the cold-trap. A provision is also made to allow a small portion of the make-gas to be collected continuously to obtain a composite sample.

A P&I diagram for the PDU is shown in Figure 4.

C. HANDLING CAKING COALS

The following section is a continuation of an earlier section treating methods for handling caking coals. The discussion here will be with regard to a grass-roots commercial installation and also with regard to experimental work that might be carried out in the PDU.

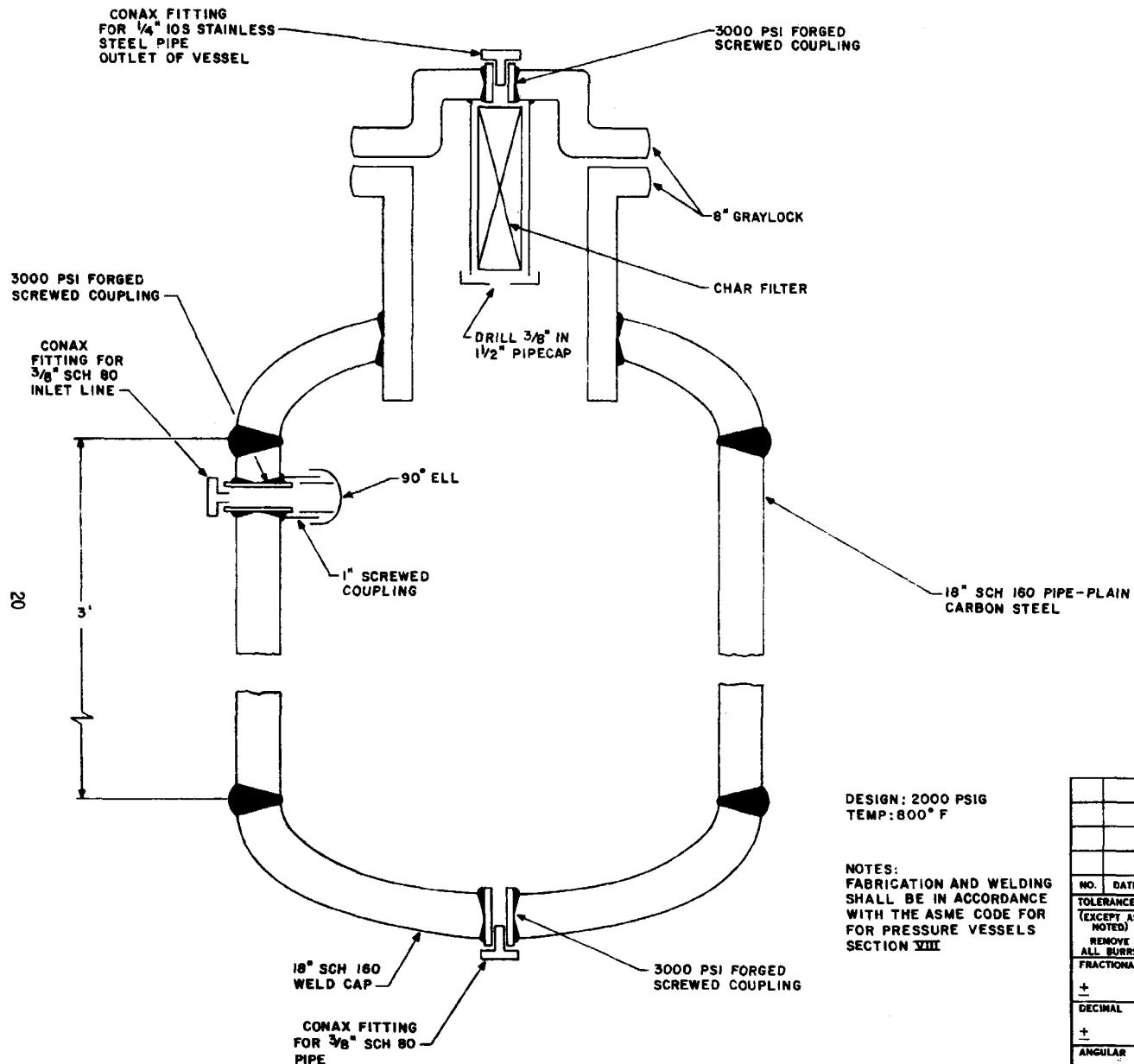


Figure 10. LIQUID PRODUCTS DRUM

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NO.	DATE	DESCRIPTION OF REVISION		DFT	CHK	APPVD
TOLERANCES (EXCEPT AS NOTED)		INSTITUTE OF GAS TECHNOLOGY CHICAGO, ILLINOIS				
REMOVE ALL BURRS		SCALE: NONE	DATE: 12-20-77	DRAWN BY: M. E. S.		
FRACTIONAL		MATERIAL: CARBON STEEL EXCEPT AS NOTED		APPROVED BY: R. O. O.		
DECIMAL		TITLE: PDU FOR RISER CRACKING OF COAL LIQUID PRODUCTS DRUM				
ANGULAR						
+						
—						
		PROJECT NO.	DRAWING NO.		REV.	
		8976	774			

1. Commercial Unit Considerations

In the IGT commercial unit design concept (Figure 11), the feed coal will be ground and slurried with an oil or solvent to allow the coal to be pumped into the high-pressure processing equipment. The slurring medium is removed from the coal in a fluidized-bed drying step. Many solvents could be used for this step. By selecting a solvent that is capable of extracting the portion of the coal responsible for the caking tendencies, it may be possible to remove or temporarily disengage this fraction of the coal in the drying step, thus obtaining a non-caking feed for the pyrolysis in the riser reactor.

The liquids obtained in the hydropyrolysis or fractions of the liquid products may be suitable for this step. The gasoline fraction will contain quantities of phenol, which has been found to be effective in reducing or eliminating the agglomerating tendencies of coals. Also, the naphthalene fraction could be partially hydrogenated to form a tetralin-rich oil that would convert a fraction of the coal to materials that would be vaporized in the slurry drying step. The slurring step could also be enlarged to include a time-tank in which the coal oil slurry would be held at an appropriate temperature to allow a portion of the coal to dissolve.

Water might also be used as the slurry medium. If this were done, the slurry dryer would operate at temperatures too low for the coal to form agglomerates. By moving the point at which the coal is introduced into the main carrier gas stream to the bottom of the riser, it may be possible to "shock heat" the coal with the preheated hydrogen, avoiding the plastic range.

2. PDU Alternatives

In the proposed commercial concept for the riser cracking of coal provision was made for the recycling of char from the 1800°F gasifier through the riser reactor and disengaging vessel back to the gasifier. The char recycle would be used primarily as a means of transferring heat from the gasifier to the feed streams at the bottom of the riser. Mixing char with the coal feed may also provide a means for transporting coal in the thermoplastic stage by providing a surface for the adherence of coal. Hot char would also heat the coal rapidly through the thermoplastic stage and scour the walls of the riser to prevent a build-up on the walls.

IGT has air-blown pretreating equipment, and the air-blown pretreatment may be of interest in processing mildly caking coals where the pretreatment would not be so severe as to seriously affect the susceptibility of the coal to the hydropyrolysis process.

Although it is not a part of the program outlined in the original proposal, a small-scale batch system could be built and operated to simulate the solvent pretreatment and slurry-drying steps for the commercial concept.

The notion of a solvent treatment could also be extended to simulate the direct charging of liquid slurry to the riser reactor. The coal slurry mixed with hydrogen moving through the furnace coil would be brought through the thermoplastic range in admixture with one of several possible solvents capable of altering the caking properties of the coal. Such a processing technique would require a high solvent-to-coal ratio, but this might be offset by improved liquid yields.

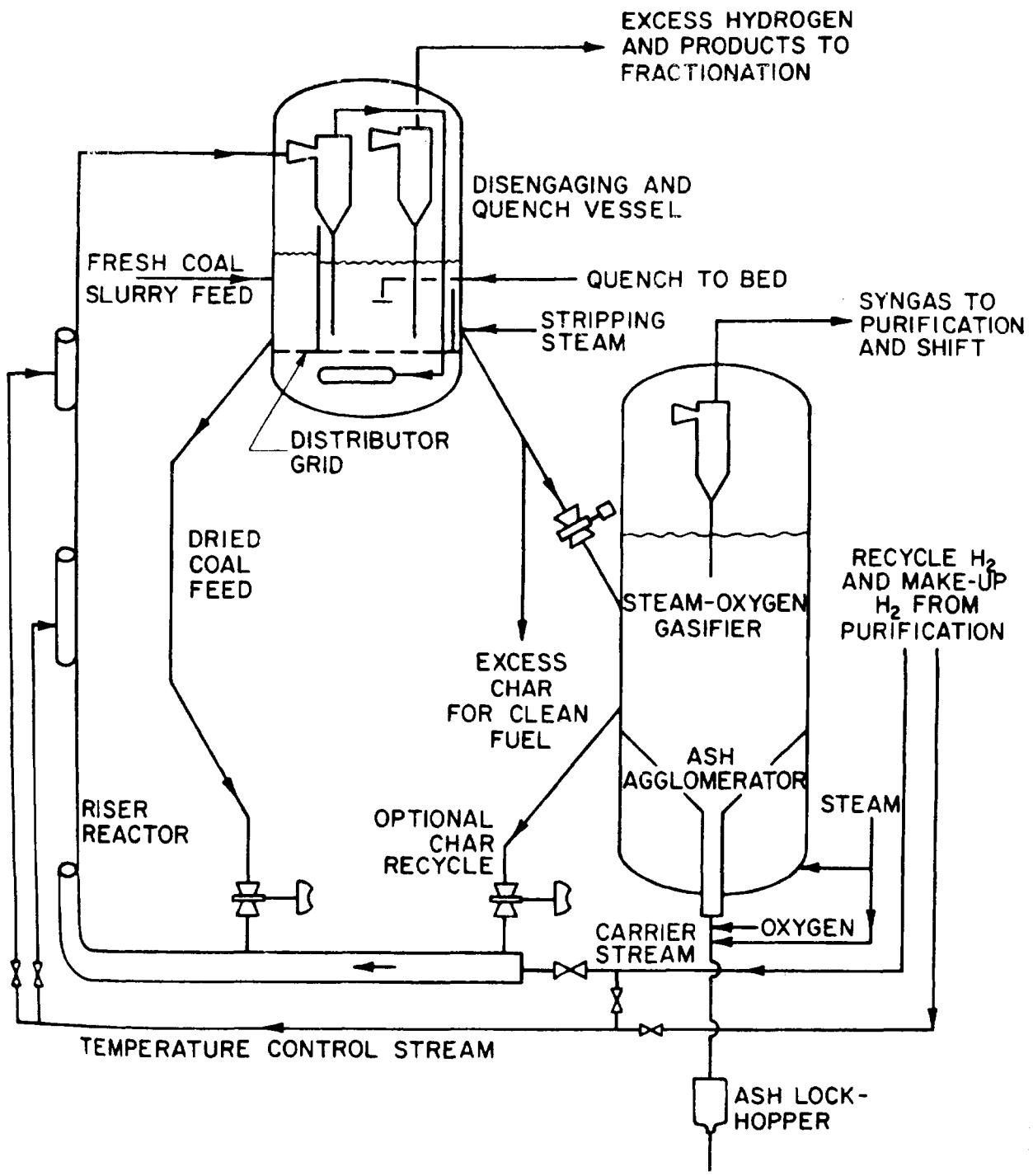


Figure 11. IGT COMMERCIAL RISER CRACKING CONCEPT

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