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

(RISER CRACKING OF COAL)

Annual Report
For the Period April 1, 1976, to March 31, 1977

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EXECUTIVE SUMMARY

This is the first annual report for ERDA Project FE-2307 "Riser Cracking of Coal" under Contract No. E(49-18)-2307. The objective of the program is to develop a noncatalytic process for the rapid hydrogenation of lignites and coals in a short residence-time entrained-flow reactor at high temperatures and pressures for the production of feedstock for synthetic motor fuels. Operating temperatures in the range of 900° to 1500°F and operating pressures up to 2000 psig will be used; solids residence time will be varied from 1 to 10 seconds. Both a bench-scale unit (5-10 lb/hr) and a process development unit (50-100 lb/hr) will be used in this program.

The development program is based on a commercial concept in which the use of a short residence-time riser reactor, of the type used in contemporary catalytic cracking, is extended to the conversion of coals and lignites to gaseous and liquid products by reaction with gases such as hydrogen, synthesis gas, or mixtures of carbon monoxide and steam. The gases also carry the feed coal through the riser reactor. A maximized production of high-octane gasoline constituents (C_4-400°F boiling range including BTX) is an important aspect of this investigation. Light gases (C_1-C_3) will also be produced and will contain substantial proportions of methane and other light hydrocarbons that could be used either for fuel or for petrochemicals feedstock. Spent char would be used for synthesis gas or hydrogen production for use in the riser reactor.

The process is expected to have more favorable economics than other known processes — such as the Bergius or Fischer-Tropsch processes — because of the high space-velocity (volume efficiency) of the reactor and also because less hydrogen would be consumed. The distillate fuel yields are expected to be lower than those of the Bergius process, but gas and gasoline yields are expected to be greater.

IGT Program

The IGT program has been divided into various tasks as follows:

Task 1. Planning
Task 2. Building and operating a bench-scale unit
Task 3. Designing a process development unit (PDU)
Task 4. Building a PDU
Task 5. Operating a PDU
Task 6. Technical and economic evaluation of work with the bench-scale unit and PDU.

The first task has been completed, and the bench-scale unit has been built and is presently being operated to study the effects of the basic process variables of temperature, pressure, gas composition, and residence time. At this writing, six runs have been completed (Table ES-1).

The processing concept under investigation is noncatalytic, but some work with catalysts is planned, at least in the bench-scale unit.

In Task 3, data obtained from the bench-scale unit will be used to design a process development unit (PDU). The PDU design and construction (Task 4) are scheduled to be started during the first year of bench-scale work. In operations with the PDU (Task 5), emphasis will be placed on 1) simulating a viable commercial operation and 2) obtaining material balances and other information needed for further scale-up. In Task 6, a technical and economic evaluation will be made of short residence-time coal hydrocracking.

Work Accomplished

In the first quarter of the project year a plan of experimental work to be performed in the bench-scale unit was prepared and submitted, together with the design of the bench-scale unit; these comprised the Task 1 report. Procurement and fabrication of the experimental apparatus was started upon ERDA/FE approval of these plans.

Feeding of solids into small-diameter transport lines with mechanical devices did not appear feasible, so development of a solids feeder was started concurrently with the construction of the bench-scale equipment. A low-pressure cold-flow model was used to develop an aerated solids-feeding system that was subsequently demonstrated using 60x100, 100x200, and 200x325 mesh lignite. Correlations were developed to describe the time-rate of delivery of lignite from the feeder in terms of the volume of gas flowing through the feeder. A high-pressure version of this prototype was incorporated into the bench-scale unit.

Operations with the bench-scale unit were started in the last quarter of the year using North Dakota lignite as feedstock; six runs have been completed. Operating conditions and important results for these runs are summarized in Table ES-1. The first two runs were largely to test the operation of the equipment; conversion of carbon to liquid products was too low to be of interest, so complete data were not taken. In making Run P-2, a malfunction in the temperature-control system caused two sections of the coil heater furnace to burn out, at the same time melting a section of the coil reactor. The damage was repaired and a new coil reactor fabricated using Incoloy 800.

The next runs were more successful, and good material balances were obtained. The actual performance of the equipment was largely as planned. The char, separated from the make-gas at high pressure and 450°F was dry, and subsequent steam distillation at atmospheric pressure has shown that only negligible amounts of light oils remain absorbed on the char.

Some problems were encountered in separating water from the hydrocarbon liquids; at present, the liquid phase is acidified with hydrochloric acid, and then sodium chloride is dissolved in the aqueous phase to increase the specific gravity and float the hydrocarbon phase. The water phase is then removed from the hydrocarbon phase by means of a separatory funnel. The odors of both ammonia and hydrogen sulfide have been detected when handling the liquid products, although neither of these constituents has been detected in the gas analyses.

Table ES-1. SUMMARY OF OPERATING CONDITIONS AND RESULTS OF RUNS IN THE BENCH-SCALE UNIT

Run Number	P-1	P-2	P-3	P-4	P-5	P-6
Date	12-17-76	1-21-77	2-2-77	2-15-77	2-25-77	3-23-77
Coil Outlet, °F	1000	1200	1200	1400	1500	1400
System Outlet, psig	500	1000	1000	1000	1000	1500
Residence Time, s	1.3	2.1	1.3	2.2	2.0	2.1
H_2/Coal Ratio, lb/lb	0.091	0.074	0.611	0.079	0.079	1.69
Feed Wt Loss, %	--	--	15.8	44.9	51.4	61.7
Carbon Conversion, %	--	--	11.2	29.4	31.0	51.4
Oil Yield, wt % of solids feed	nil	1.9	4.8	5.0	3.5	(7.0) est.
Benzene in Make Oil, wt % of oil	--	--	--	9.5	47.8	--
Run Length, min	20	15	60	55	35	60
Overall Material Balance, wt % recovery	--	64.2	97.4	99.6	99.4	80.2
Shutdown*	E	P	V	E	P	V
Methane Yield, wt % of solids feed	--	--	1.0	5.3	7.5	5.8

* E = solids feed exhausted.
P = coil plugged.
V = voluntary.

Some trends were apparent from the data. As would be expected, higher carbon conversion is favored by increases in both hydrogen partial pressure and temperature. Examination of gas analyses for dense-phase runs (Runs P-4 and P-5) has shown that the actual hydrogen content of the make-gas is 50 to 60 mole percent, compared with the hydrogen content of the make-gas in dilute-phase runs (Runs P-3 and P-6), which was more than 97 mole percent. The large decrease in hydrogen content in dense-phase operation is due to the presence of water vapor and other reaction products.

Other differences in make-gas composition were observed; at relatively low temperatures (1200°F and 1000 psig, Run P-3) small quantities of alkanes and olefins were detected including propylene, butenes, and pentenes. At 1500°F and 1000 psig the amount of light hydrocarbons was greatly suppressed, with methane predominating as the chief nonaromatic hydrocarbon.

In raising the reaction temperature from 1400° to 1500°F at the 1000 psig level (Runs P-4 and P-5) a reduction in liquid yield but an increase in BTX yield was observed.

The reason for the poor material balance in Run P-6 (the most dilute run) is not known; the work was carefully done, and in this particular case about 100 grams of material cannot be accounted for. There is reason to suspect, however, that the aromatic light oils may be adsorbing on sample lines or sample bomb walls, so that the reported gas analyses tend to underestimate the quantities of hydrocarbons present in the make-gas.

Some operational difficulties have resulted in several aborted runs that were not numbered. These difficulties have chiefly involved bridging of the feed hopper and plugging of the coil reactor. Turnaround and work-up time at present is two to three days if no repairs or equipment changes are needed. Cleaning the coil reactor can be time consuming, especially if it has been plugged, since much jury-rigging and *ad hoc* procedures are required to dislodge and burn out the material that forms the plug.

Future Work

When the immediate set of preliminary runs is completed, we plan to revise the solids-feeding system to improve the reliability of the feeder and also to revise the equipment to heat the carrier gas. This will increase the gas velocity at the coil entrance from 5 ft/s to values at which gas-solids flow is more stable. In operations thus far, cold feed-gas has been used, and it has been possible to heat the feed gas and coal to operating temperature in the first 25 feet of coil length; the last 50 feet of coil have been operated isothermally. At the highest temperatures used (1500°F), plugs have formed that are believed to be formed from coking on the coil walls. It should be possible to reduce the tendency to plug by lowering the heating rate and lengthening the residence time.

No major difficulty is anticipated in operating the 1/8-inch ID bench-scale reactor on lignite, but improvements in its operability will be made in order to progress with the planned experimental program.

Our future work will concentrate heavily on the effects of the hydrogen to coal ratio and the temperature level of the hydrogen. These variables are expected to have a very significant effect on both yields and overall economics.

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ABSTRACT

During the first year of the "Riser Cracking of Coal" program the bench-scale coil reactor was designed, built, and put into operation. A detailed experimental plan made for the bench-scale unit was presented in a Task I report.

The 50-foot bench-scale reactor is being operated isothermally with cold hydrogen and coal feed, heated together in a 20-foot induction coil that is integral with the reactor. Six experimental runs have been completed. The highest reaction severity level attempted has been 1400°F at 1500 psig; a carbon conversion of 51.4 weight percent was attained. The highest reaction temperature reached was 1500°F (at 1000 psig). The benzene concentration was 47.8% in the make-oil from this run, but the oil yield itself was lower than at the 1400°F level at 1000 psig. The highest oil yield attained at this point was 11 weight percent on carbon (7 weight percent on wet lignite feed). Residence times have been about 2 seconds, and good material balances were obtained in 3 of the 4 most recent runs.

The reactor has proven operable on lignite, but some revisions are planned to improve the coal feeding and to increase the temperature of the feed hydrogen.

PROGRAM OBJECTIVE AND SCOPE

The objective of the research and development program described in this report is to develop the technology for short residence-time processing of coal in an entrained-flow gas-solids reaction system for the production of light hydrocarbon gases, aromatic liquids, and spent char. This work is being performed for the U. S. Energy Research and Development Administration under Contract No. E(49-18)-2307, "Research and Development of Rapid Hydrogenation for Coal Conversion to Synthetic Motor Fuels (Riser Cracking of Coal)."

In the program, the rapid gas-phase hydrogenation of coal is being investigated as a method for producing high-octane blending gasoline as a principal product. The scope of the investigation includes the design, construction, and operation of both a bench-scale unit (5-10 lb/hr) and a process development unit (50-100 lb/hr).

Previous investigations have shown that coal can be converted to gases and liquids by pyrolysis in a reducing atmosphere in both uncatalyzed systems[2,3,4,9,10,12] and catalyzed systems.[5,13,14,15] These investigations have demonstrated that short residence-time pyrolysis processes are at least technically feasible. The economic aspects of liquid-phase liquefaction of coal in processes similar to the Bergius process have also been investigated,[1,6] and initial studies at IGT have shown that more favorable economics can be expected of a hydropyrolysis process.

The experimental investigation being conducted in this program is oriented towards establishing a process that can be scaled up to commercial size in terms of current materials, equipment, and technology, compared with current investigations of the chemistry of coal hydropyrolysis. The work being done here is expected to complement the work of other investigators and — hopefully — to shorten the time needed to define the technical and economic aspects of a large-scale application.

Work in the bench-scale unit will be performed at operating temperatures up to 1500°F and operating pressures up to 2000 psig. Residence times will be varied from 1 to 10 seconds. The data from the bench-scale unit will be used to guide the design of the process development unit. Not all the subsystems needed to make a self-contained process plant will be incorporated into the PDU, since the principal task is the development of the riser reactor. Upstream and downstream conditions will be accommodated in a suitable manner.

PROGRAM PLAN

The work of this program is divided into six tasks scheduled over a period of 50 months (Figure 1) as shown below:

Task 1: Project planning and design of a bench-scale unit (Project months 1-2)

Task 2: Build and operate a bench-scale unit (Project months 3-36)

Task 3: Evaluate bench-scale unit data and design a process development unit (Project months 14-19)

Task 4: Build the PDU (Project months 20-28)

Task 5: Operate the PDU (Project months 29-46)

Task 6: Assess the process and report findings.

Task 1 and the first 10 months of the planned 34-month Task 2 comprise the work of the first project year. Continuation of Task 2, all of Task 3, and initiation of Task 4 comprise the work of the second project year.

Bench-Scale Work

In the first two tasks of the program, a bench-scale unit has been designed and built and is currently being operated to determine how reactor temperature, temperature profile, pressure, and residence time affect rapid hydrogenation of coals. These data will be used to design the process development unit; the data obtained from the bench-scale unit, while incomplete, will hopefully define the relationships of coal hydrogenation with enough clarity to permit the scale-up.

The bench-scale unit will be operated at temperatures up to 1500°F and pressures up to 2000 psig, and solids residence times will be varied from 1 to 10 seconds. An important attribute of the bench-scale unit will be the ability to regulate temperature profile methods for processing caking coals.

PDU-Scale Work

In Tasks 3, 4, and 5, a PDU will be designed, built, and operated using the data from the bench-scale unit as a guide in the design. Operations in the PDU will stress the simulation of the operation of a viable commercial operation. Finally, in Task 6, a technical and economic aspect of the rapid hydrogenation process will be assessed.

Summary of Achievements

During the first project year, the principal accomplishments have included the following:

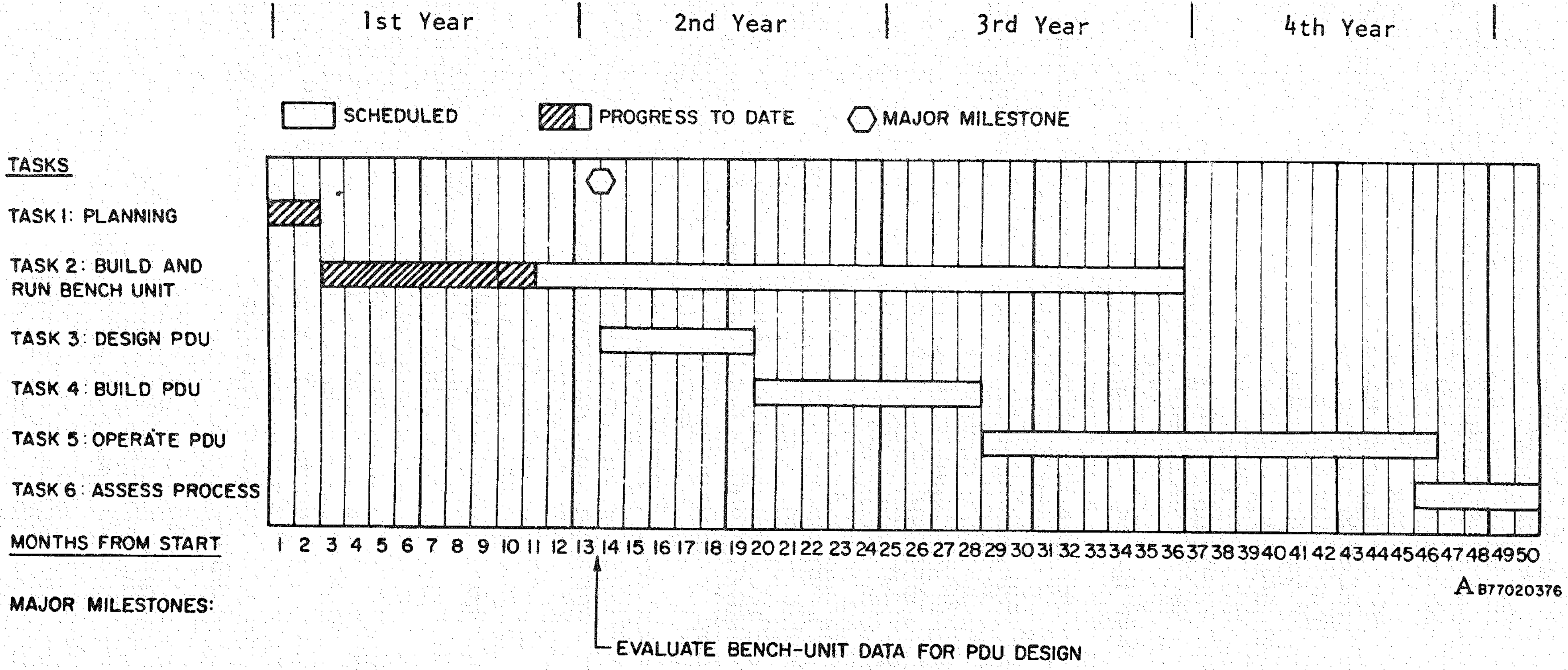

Figure 1. PROGRESS CHART

1. Completion of Task 1: Project Planning
2. Completion and approval of a bench-scale reactor design
3. Construction of a bench-scale reactor
4. Reactor shakedown and initiation of bench-scale testing
5. Six preliminary runs
6. Development of a solids feeder.

TECHNICAL PROGRAM

Task 2: Program Description

The scope of Task 2 in this program is to build and operate a bench-scale unit. The experimental program is divided into two sections; the first period of 15 months is to provide data for the design of the PDU. The second period involves the continuation of the bench-scale experimental work to explore particular areas in depth and, generally, to fill in details omitted in the first period of the investigation.

The principal variables to be investigated include coil outlet pressure, coil outlet temperature, residence time, temperature profile or time-temperature history, particle size, coal type, and feed-gas composition. An array showing blocks of experiments involving these variables is shown in Figure 2.

Originally we had expected to explore the 500 to 2000 psig reaction pressure range and temperatures from 900° to 1500°F. With some experience on the bench-unit, however, it has become clear that we will be concentrating our efforts on pressure levels above 1000 psig and temperatures in the 1400° to 1500°F range.

Our first runs have been given the designation "P" for preliminary and in this series we are not preheating the hydrogen feed. The coal and hydrogen are being heated together in the induction section of the reaction coil (first 20 feet) at a rate of several hundred degrees per second. This procedure is being followed both for expediency and practicability – it is obviously a simpler way of operating. The extent of improvement gained by preheating the hydrogen feed needs to be demonstrated in a quantitative way, and we hope to do this by running with hotter hydrogen in the near future. A separate coil within the furnace will be used to preheat the hydrogen.

Yet another practical expedient we are following in this first series of runs is that the lignite has not been completely dried and contains 10 to 15% moisture as it might in plant practice. The effect of this will be explored also in due course.

The importance of the hydrogen-to-coal ratio has become more apparent since the start of the project. We had elected to operate with a very low hydrogen concentration of 12 weight percent while others in this same area of research are operating with more than 100 weight percent hydrogen. The effect of this variable on yield structure and operability must be clearly understood because of the profound effect it has on the overall economics of the process. Thus both the hydrogen temperature level and the hydrogen-to-coal feed ratio will receive considerable attention in our research for the immediate future.

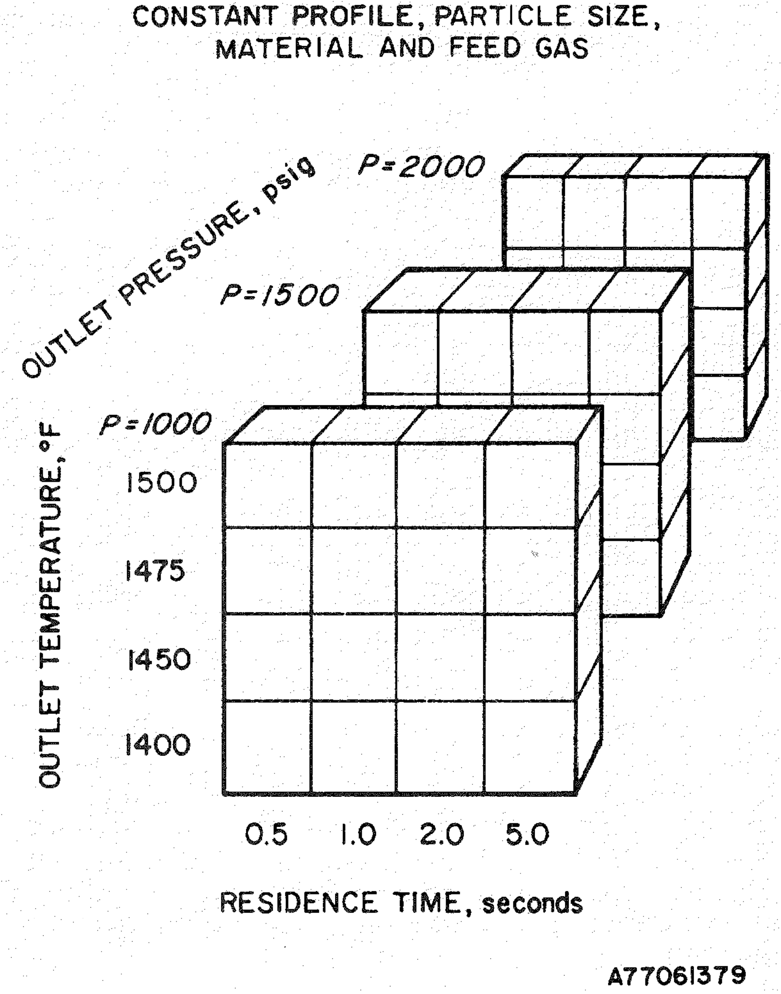

Figure 2. TYPICAL 4x4x3 ARRAY OF ORDERED EXPERIMENTS

Bench-Scale Unit Design

The bench-scale reactor is designed to simulate the flowing conditions of riser cracking of coal. This design allows measurement of reaction temperature, as well as control and variation of residence time in the reactor; it is, therefore, possible to explore residence time, temperature, and pressure.

The conditions for which the bench unit has been designed are as follows:

Coal Feed Rate: up to 10 lbs of coal per hour

Carrier Hydrogen Rate: from 15 ft/s (12 wt % hydrogen) to 70 ft/s.

Alternate Carrier Streams:
1. Hydrogen 100%
2. Hydrogen and Steam
3. Hydrogen and Carbon Monoxide (Syngas)
4. Syngas and Steam

Pressures: 500 psig up to 2000 psig

Temperature: 900°F up to 1500°F.

A flow diagram (Figure 3) shows the assembly of the individual components. Of the various components, only the reactor furnace coil approaches the design allowable stress at 1500°F. At the severe conditions of 2000 psig and 1500°F, the Incoloy 800 reactor coil life is limited to 1000 hours. At 1500 psig and 1500°F, coil life is 10,000 hours.

The feed hopper is a two-thirds cubic foot vessel designed for pressurizing to 2100 psig for charging into the bench unit. It has a capacity of 20 pounds of coal, assuming 30 lb/cu ft density in the hopper, and is designed for gravity feeding of the coal at a continuous rate. A connection has been provided for nitrogen purging of the loaded hopper prior to the start of charging.

Carrier gas is supplied once-through by means of a 3000 psi storage system. Prior to a run it is pressurized with hydrogen or a suitable mixture of hydrogen and carbon monoxide.

For the runs employing steam, a water tank and scale have been provided. An adjustable rate-proportioning pump measures the water and raises its pressure prior to vaporizing in a steam boiler that is an 80 foot coil of pipe inside a stand of four 2-foot-high resistance heating furnaces. The carrier gas system is provided with rotameters to measure a controlled flow of pressurizing gas to the feed hopper and aeration gas to the coal feeder.*

The main source of heat to the combined coal and carrier-gas stream is a preheater coil located above the reactor coil in the reactor furnace. This preheater coil has been provided to assure that the entire reactor coil heating system is available for temperature control of the 50-foot reactor coil.

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* A fixed restriction is being used at the time of writing.

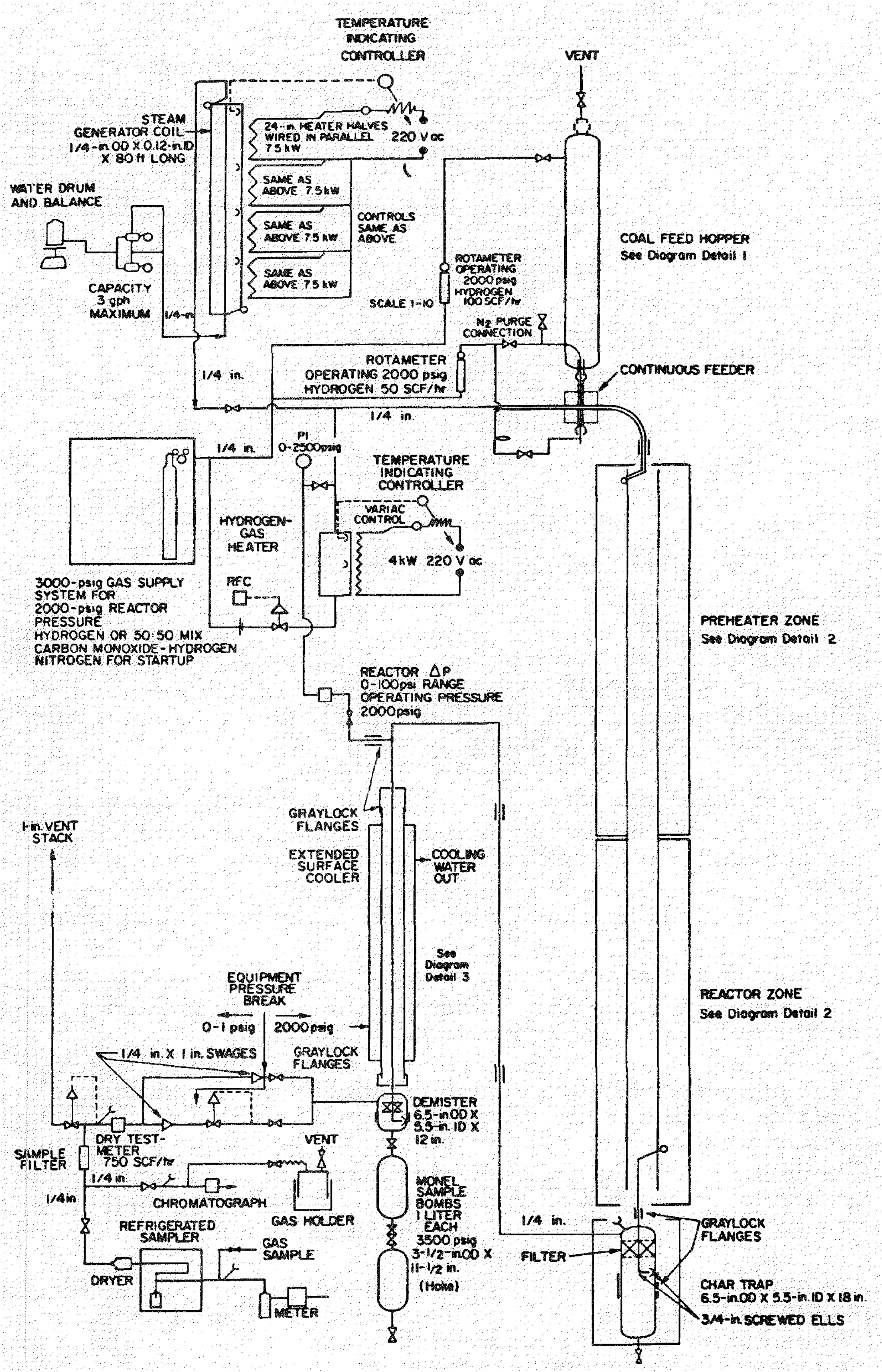

Figure 3. BENCH-SCALE UNIT FOR RISER CRACKING OF COAL

The 50-foot-long reactor coil is made of Incoloy 800 tubing, 0.25-inch outside diameter by 0.12-inch inside diameter. Heat is applied at many zones along the coil to permit changing the heating-rate curve. Thermocouples along the coil monitor the temperature.

The coil is wound on a mandrel that serves to support the coil and the thermocouples. Table 1 and Figure 4 show the orientation of the coils and thermocouples in the furnace and the number and length of coils in each of the various heating zones. Each heating zone is independently monitored, and the temperature is controlled.

The effluent products enter the char trap located immediately below the reactor, where most of the spent char is disengaged. The gases then move through a filter for removal of the remaining char fines. The upper part of the char trap is heated so that convective heat losses will not cause condensation of tars in the char trap and exit lines.

The effluent then proceeds through a cooler that drains downward into a gas-liquid separator where the liquid products are separated from the gases. A demister pad is provided to disengage any liquid droplets. The collected liquids flow downward into two 1-liter sample bombs. The make-gas is reduced to nearly atmospheric pressure after which it is metered with a dry-test meter, sampled, and vented. A portion of the gas from the meter proceeds through a dryer and cold trap.

The gas proceeds through a back-pressure regulator that holds the entire system pressure, is metered at approximately atmospheric pressure in a dry-test meter, and is released to a vent stack.

Another small portion of the gas from the meter proceeds through a dryer and to a refrigerated sampler;* from the refrigerated sampler it is metered and sampled for analysis. The liquid condensed in the refrigerated sampler is measured and analyzed.

Figure 5 is a photograph of the bench-scale unit, and Figure 6 shows the control panel.

Operating Procedure

Preparation of the Apparatus

1. Assemble the feed hopper and flush the coil with nitrogen to insure that the coil is free of any particles that might adhere to the walls and cause plugging. Then assemble the char catchpot and high-pressure receiver to the reactor and condenser. Test for leaks by pressurizing with nitrogen.

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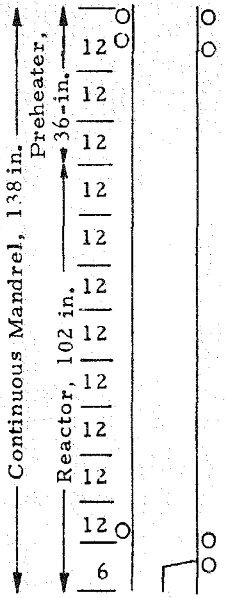
* This refrigeration system is currently being revised to operate on the total gas stream.

Table 1. COIL ORIENTATION AND MANDREL DETAILS FOR BENCH-SCALE UNIT FURNACE (Diagram Detail 2)



Heater Zone Height, in.	Number of Coils in Zone	Feet of Coil in Zone	Accumulated Feet of Coil From Inlet	Temperature Element No.	Heater Bottom Elevation From Bottom of Mandrel, in.	Heater Number for Temperature-Indicating Recorder and Temperature Recorder
				14[*]		
12	7.5	7.9	7.9	1	126	1
12	7.5	7.9	15.8	2	114	2
12	7.5	7.9	23.7	3	102	3
12	5.47	5.8	29.5	4	90	4
12	5.47	5.8	35.3	5	78	5
12	5.47	5.9	41.2	6	66	6
12	5.47	5.8	47.0	7	54	7
12	5.47	5.8	52.8	8	42	8
12	5.47	5.9	58.7	9	30	9
12	5.47	5.8	64.5	10	18	10
12	5.47	5.8	70.3	11 13[†]	6	11
6	Straight	0.5	70.8	12	0	12 (Outlet Temperature)

Mandrel Data:

3-3/4-in. OD x 3.5-in. ID x 11-1/2 ft Long. Material: 304SS

Coil Data:

	Preheater	Reactor in.	Total
Height of Wound Coil	36	96	132
Height of Straight Coil	0	6	6
Pitch (Distance Between Centerline of Adjacent Coils)	1.6	2.2	--

Tubing: 1/4-in. OD x 0.12-in ID x 71 ft Long. Material: Incoloy 800

[*] Temperature recording only.

[†] Temperature recording only; 4 in. above outlet.

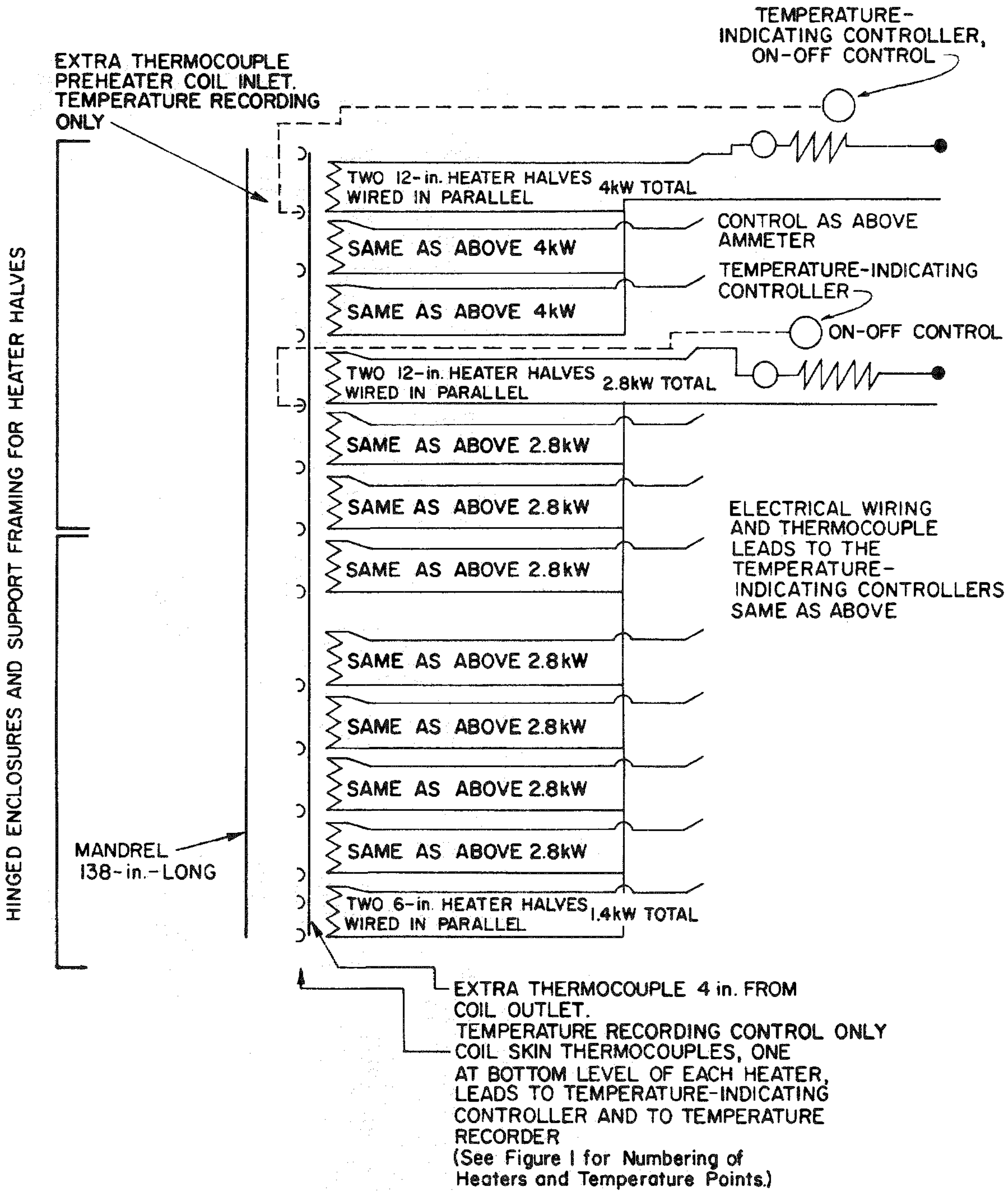


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Figure 4. ELECTRICAL AND INSTRUMENTATION PLANS FOR BENCH-SCALE UNIT FURNACE (Diagram Detail 2)

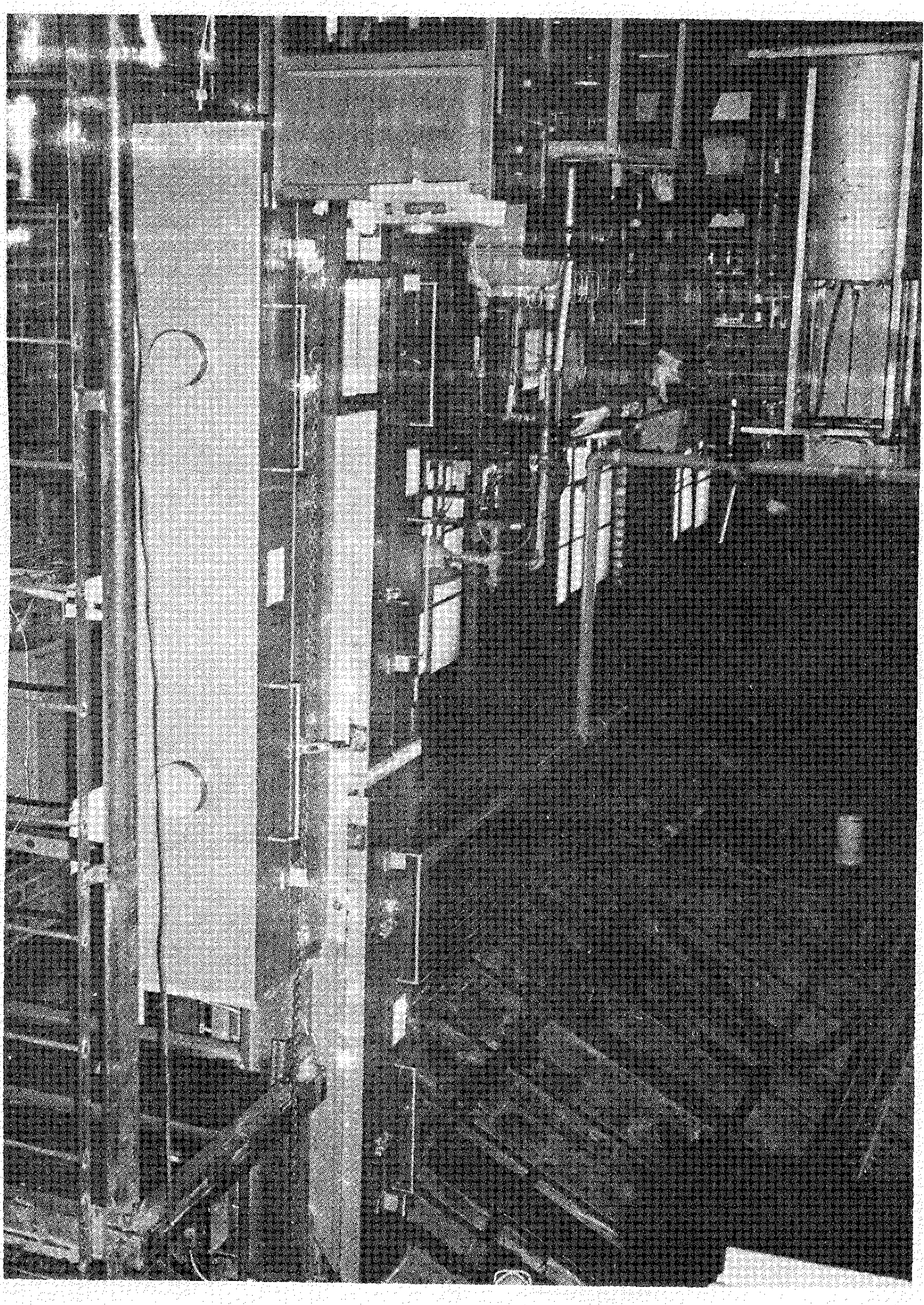

Figure 5. OVERALL VIEW OF THE BENCH-SCALE UNIT

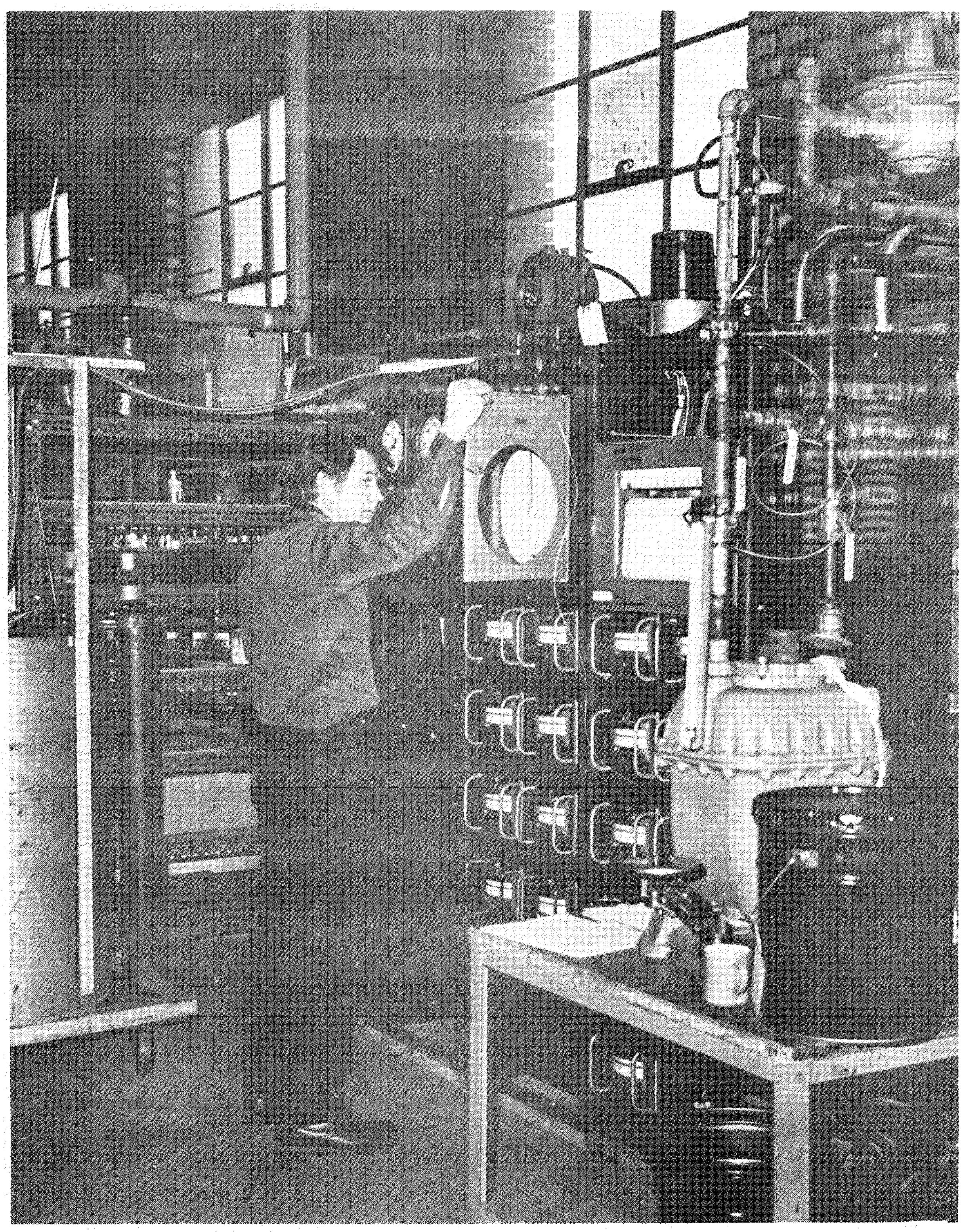

Figure 6. BENCH-SCALE UNIT CONTROL PANEL

2. Open the feed hopper and charge a weighed quantity of coal to the hopper. The charge of coal will have been extracted from a larger quantity of feed by riffling so that the charge is representative of the stock of feed coal. The feed coal is to be stored in sealed containers to protect the coal from exposure to air and change in moisture content.

3. If the coil heaters are not on idle, turn them on at this time and set the controls at 1200°F. Also, heat the char catchpot to 450°F to prevent condensation in the catchpot.

4. Empty the gas holder and position the valves so that gases from the reactor are vented. The gas sampling train is to be closed. Bring the refrigerated coolant system and gas chromatograph (when used) to standby conditions. Bring the feed-gas heater and steam generator (when used) to operating temperature. Load the steam generator feedwater reservoir and set the feedwater pump to deliver the desired rate of feedwater to the steam generator.

Start-Up

1. Start the flow of feed gas through the equipment, and bring the coil outlet pressure to the desired operating level. Adjust the flow rates to their operating values. Check the feed rate of non-condensable gases by determining the flow rate through the main gas-meter and correcting this observed flow rate to the inlet temperature and pressure.

2. Set the coil heaters to establish the temperature profile to be used during the run. Allow the equipment to run until a steady-state operation is established. Remove condensed steam from the high-pressure receiver by cracking the needle valve located at the bottom of the receiver. Drain the high-pressure receiver immediately before the start of solids feeding.

3. Start the flow of solids; at the same time, mark the temperature recorder chart, and close the by-pass around the main meter. (A few minutes before the start of solids feeding, the by-pass should be opened to stop the meter, and the meter reading recorded; closing the by-pass will restart the meter.) The manipulation of the equipment controls requires less than 1 minute.

4. Determine the flow rate of gases through the main meter every 10 minutes or other predetermined interval; also monitor the pressure differential across the reactor to discern the onset of a plugging condition requiring a shutdown.

5. Monitor the temperature profile by observing the temperatures shown on the temperature recorder. The upset caused by solids feeding is brief, with the temperatures returning to the control settings within a few minutes.

6. After about 10 minutes of operation, if it is apparent that the equipment is operating in a satisfactory manner, begin passing a slip stream through the gas-sampling train. Take samples of make-gas downstream from the main meter and downstream from the chiller in the gas sampling train.

7. A few minutes before the predetermined time interval for feeding solids has ended, close the gas-sampling train. Record the final meter reading and determine the amount of gas passed through the gas meter.

Emergency Shutdown

1. If the coil plugs, turn the feed gas off; then depressurize the system, and set the coil heater controls on 1000°F.

Normal Shutdown

1. Terminate the feeding of solids; at the same time, determine the amount of gas passed through the meter during the period of solids feeding. Close the upper valve on the high-pressure receiver.

2. Depressurize the system and purge with nitrogen. Set the coil heaters on "idle".

Turnaround Processing

1. Empty the coal-feed hopper and determine the quantity of coal used during the run.

2. Remove the high-pressure receiver and reserve for further processing. (This will involve expanding the compressed gases through an analytical train followed by a weighing and analysis of the liquids remaining after depressurization.)

3. Remove the char catchpot, determine the weight of the residue, and sample for analysis.

4. Remove the liquids collected in the gas-sampling train, determine the weight of liquids collected, and sample for analysis.

5. Sample the gas collected in the gas holder for analysis by mass spectrometer.

6. Check log books and data sheets for completeness, and check all samples for proper labeling.

7. Inspect the equipment for damage and tar deposits; where indicated, clean the tar deposits from the equipment by washing with solvent. Save the solvent washings, and recover the tars by distilling off the solvent. Determine the weight of tars recovered for inclusion in the material balances.

Coal Feeder Development

Feeding of solids into small-diameter transport lines with mechanical devices (such as star feeders or screw feeders) did not appear feasible because of bridging and sticking in the constriction leading into the transport line. The approach taken to resolve this problem was to aerate the solids and meter them into the main transport line by entrained flow through a small-diameter orifice leading to the main transport line. A flow diagram of the apparatus used in the development work is shown in Figure 7, and Figure 8 is a photograph of the apparatus.

Coal to be metered through the cold-flow model was placed in the feed hopper, which consisted of a Plexiglas tube 4-in. ID by 36-in. long. A polyethylene funnel was fitted inside the tube, near the bottom, to channel the flow of coal into a plenum chamber; there, it was entrained in a flow of gas moving through the plenum chamber into the main transport line through a 1/8-in.-diameter orifice. The main transport line was also 1/8-in. in diameter and led to a 50-foot coil of 1/8-in. ID copper tubing. The coil discharged into an open 5-gallon pail, where the feed coal was disentrained and recovered by settling. Time-rate of delivery was measured by holding a tared receptacle under the coil discharge for a measured time interval; the weight of coal collected was then determined, and the time-rate of delivery was calculated.

The gases to the apparatus were metered by means of rotameters, and mercury-in-glass manometers were used to measure both the pressure at the entrance to the plenum chamber and the pressure drop from the plenum chamber to the main transport line.

Only two gas streams were used to meter and transport the coal through the system. These streams were a carrier gas stream to maintain the solids in pneumatic transport and regulate residence time in the coil, and a metering stream, which passed through the plenum chamber (where the coal dropping through the funnel was entrained) and carried through the exit port into the main transport line. When operating, particular care was necessary to keep tramp material – such as scales of rust – out of the system. The sized fractions of coal used in the tests were screened frequently to remove foreign material.

In early work with the feeder, some problems with blocking and bridging were encountered; these were found to be due to charges of static electricity forming on the particles and also to the natural ability of the coal particles to lock together. The substitution of Teflon for Plexiglas in the exit section of the plenum chamber was found to aggravate problems with static electricity considerably, especially when coal moved rapidly over the Teflon surface. Typically, coal discharged from the coil was observed to migrate back and cling to the exterior metal surface of the coil, much like iron filings clinging to a magnet. The Teflon part was replaced with polyethylene, which did not cause excessive charges of static electricity to build up on the coal particles.

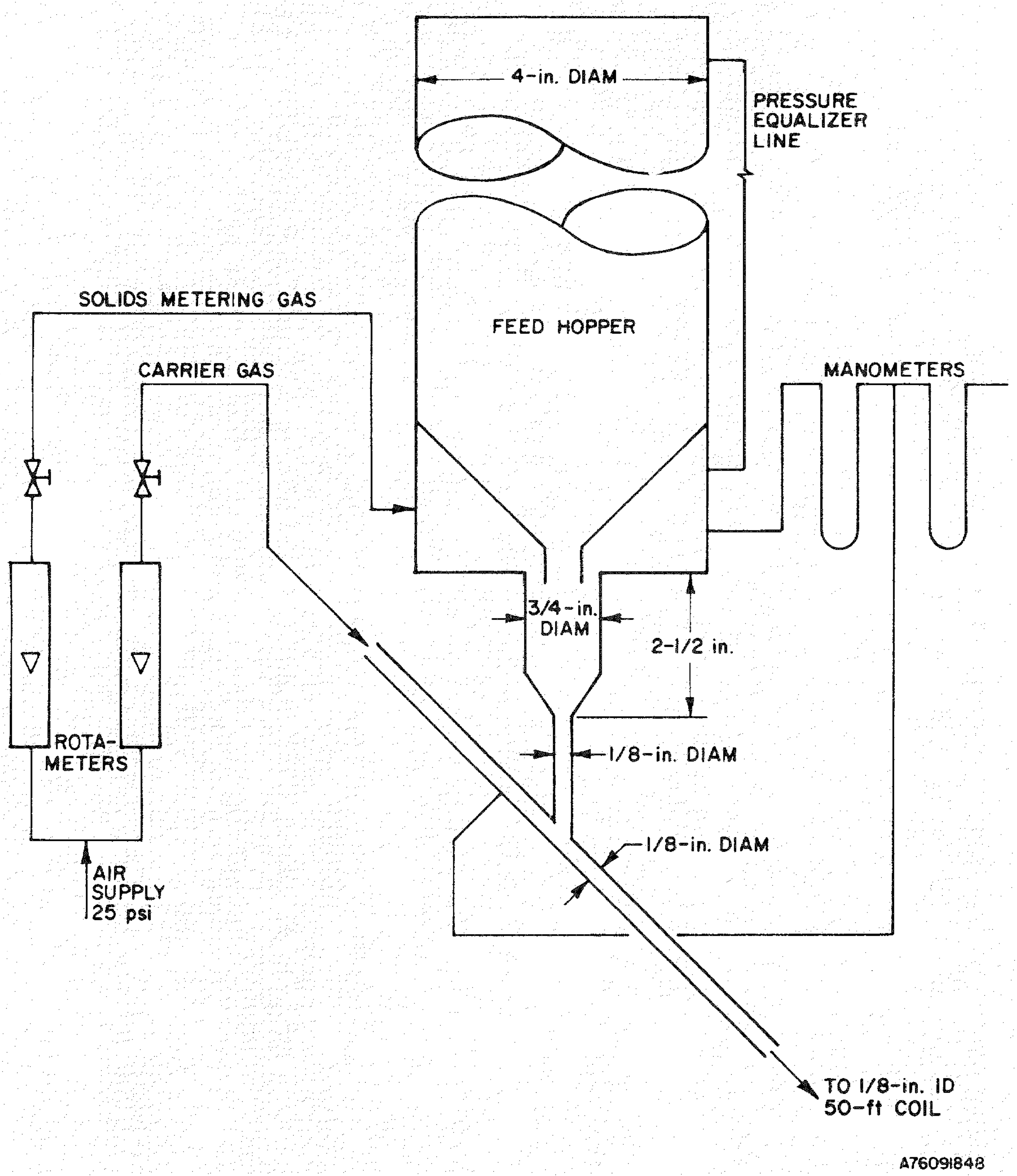

Figure 7. MODIFIED SOLIDS FEEDING SYSTEM

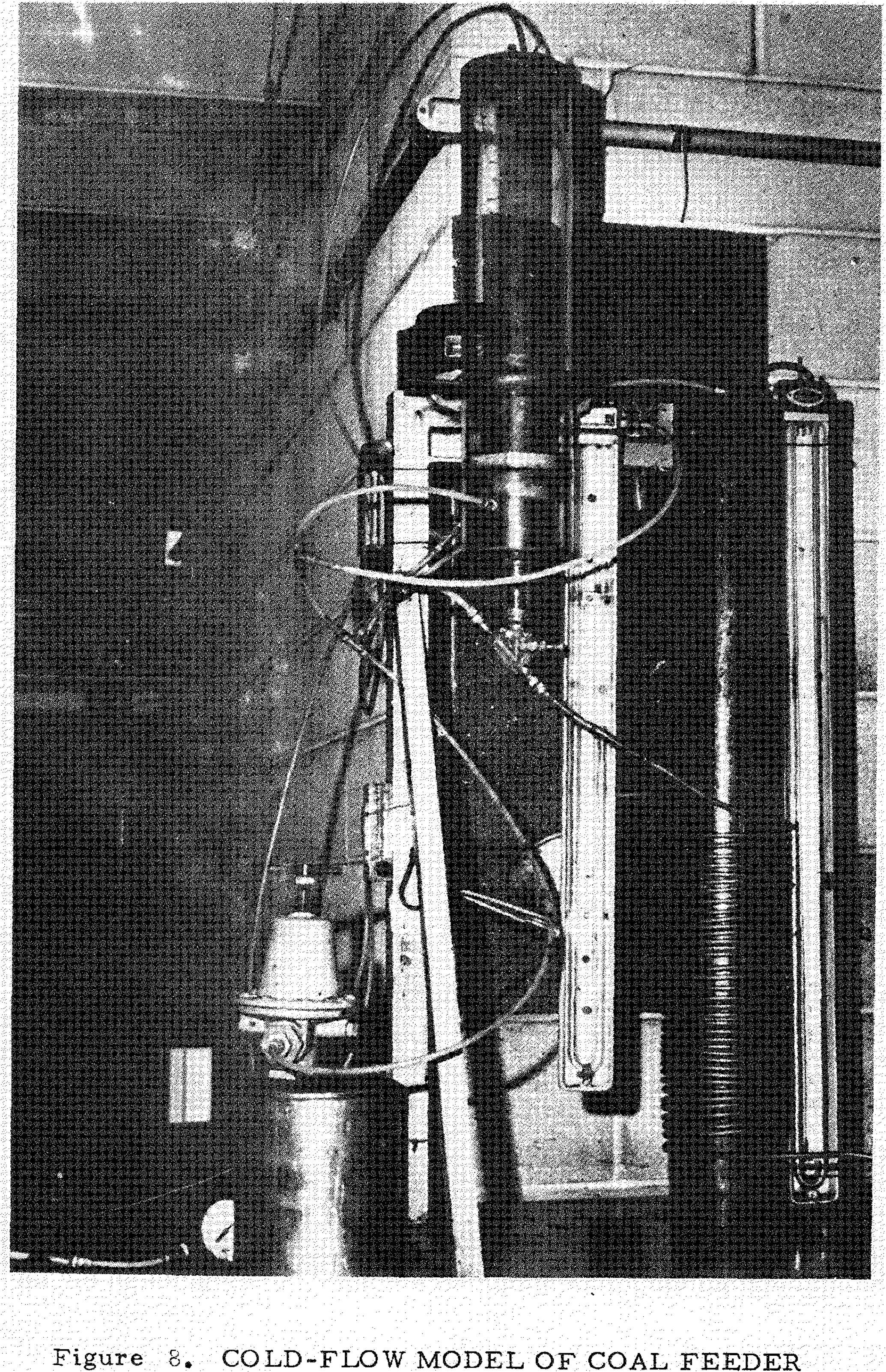

Figure 8. COLD-FLOW MODEL OF COAL FEEDER

The test coal was treated with an antistatic agent to further reduce blocking and sticking. To do this, approximately 4 pounds of sized coal was tumbled in a closed container with a sheet of "Bounce*" at room temperature; flow characteristics of the coal were greatly improved by the treatment. The angle of repose was reduced, and the tendency to cling to the hopper walls was diminished. Reproducibility of testing also improved.

The development work was continued with three particle-size ranges of coal and lignite: 60x100, 100x200, and 200x325 mesh. The 60x100 mesh material was free flowing, but it was necessary to attach a mechanical vibrator† to the feed hopper when working with 100x200 and 200x325-mesh materials. Antistatic treatment improved the processing of the 100x200-mesh material, but had no effect on the 200x325-mesh material. At low carrier-gas velocities, solids flow would degenerate to stick-slip flow at high solids-feed rates; the condition would occur at carrier-gas velocities of less than 10 ft/s and at solids-feed rates of more than 10 lb/hr.

The time-rate of delivery of solids into the main transport line was correlated with the square root of the superficial gas velocity in the feeder exit port. Data taken with 60x100 and 100x200-mesh lignite over a wide range of flow conditions are summarized in Figure 9, and data for 200x325-mesh lignite are summarized in Figure 10. The solid lines through the data are least squares fits of the data to the equation

$$\text{lb/hr} = a + b\sqrt{u_s} \qquad (4)$$

where u_s is the superficial velocity in the exit port at flow conditions. In the figures, the superficial velocity has been converted to actual cubic feet per hour through the feeder exit port.

The reproducibility of testing is shown in Table 2 for 200x325-mesh lignite. In general, the variation in solids-flow rate under a given set of test conditions was found to be only a few percent.

The contribution of the solids to coil-pressure drop is shown in Figure 11 for 100x200-mesh lignite, and in Figures 12 and 13 for 200x325-mesh lignite. At constant carrier-gas flow rate, the data can be fitted with straight lines that have very nearly the same slope. The design of tests could then be made by calculating the system-pressure drop for the solids-flow rate desired from correlations of data at the desired carrier-gas flow rate. The steady-state flow-rate of gas through the feeder plenum chamber would be calculated from the correlations between solids-flow rate and superficial gas velocity in the feeder exit port. Within wide limits, the solids-flow rate can be varied independently of the carrier-gas flow rate.

This cannot be extrapolated to the operation of the bench-scale unit in a simple manner because operation of the bench-scale unit is complicated by increases in gas velocity in the coil reactor from the increase in temperature and the increase in gas volume due to chemical reaction.

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[*] Proprietary product of Proctor and Gamble Manufacturing Co. The amount used was negligible.

[†] A 1.8 hp motor turning at 1550 rpm with a cam attached to the shaft was used.

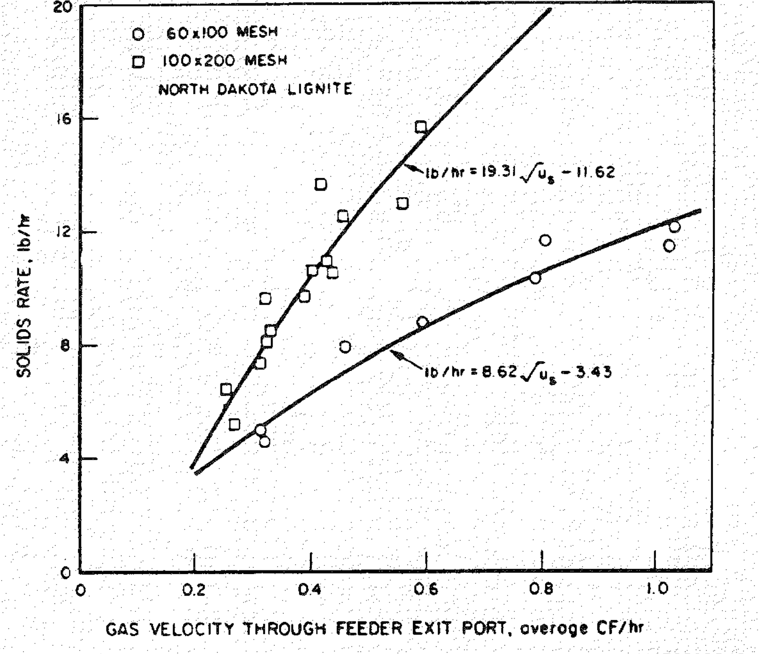

Figure 9. CHANGE IN SOLIDS FLOW RATE WITH SUPERFICIAL GAS VELOCITY IN THE FEEDER EXIT PORT

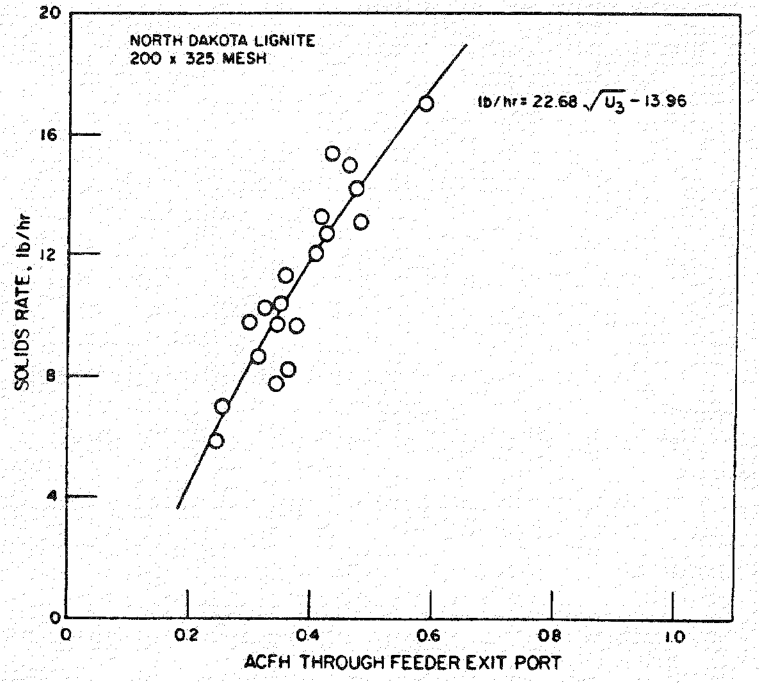

Figure 10. CHANGE IN SOLIDS FLOW RATE WITH OVERALL GAS VELOCITY IN THE FEEDER EXIT PORT

Table 2. REPLICATIONS OF TESTS USING 200 x 325 MESH NORTH DAKOTA LIGNITE

Time Elapsed, min	Solids Flow Rate, lb/hr					
0	-	-	-	-	-	-
5	9.97	9.74	9.79	11.48	11.42	11.39
10	9.79	9.68	9.72	11.34	11.56	11.43
15	9.80	9.71	9.81	11.48	11.42	11.48
20	9.91	9.70	9.71	11.38	11.49	11.35
25	9.78	9.67				
Average Solids Rate, lb/hr	9.85	9.70	9.76	11.42	11.47	11.41
Total System Pressure Drop, in. Hg	14.0	14.0	14.0	16.0	16.0	16.0
Maximum Variation in Solids Flow, wt %	±1.0	±0.4	±0.5	±0.6	±0.6	±0.6
Coil Inlet Velocity, ft/s	21.7	21.8	21.8	21.0	20.9	20.9
Coil Outlet Velocity, ft/s	31.8	32.0	32.0	32.3	32.1	32.1
Average Coil Velocity, ft/s	26.8	26.9	26.9	26.6	26.5	26.5
Average Residence Time, s	1.87	1.86	1.86	1.88	1.89	1.89
Solids in Gas, wt %	93.1	93.0	93.0	94.0	94.0	94.0

The data shown in Figures 11, 12,and 13 suggest that the system-pressure drop for gas-solids flow through coiled tubes can be expressed as the sum of the pressure drop for gas flow (alone) and a contribution to the system pressure due to the presence of solids in the gas stream or

$$\Delta p = \Delta p_g + \Delta p_s \tag{5}$$

The energy balance for gas flowing in a short length of tubing in compressible flow can be written[8]

$$\frac{dp}{\rho_g} + d\left(\frac{u_o^2}{2g_c}\right) + \frac{f u_o^2}{2g_c r_h}\, dL = 0 \tag{6}$$

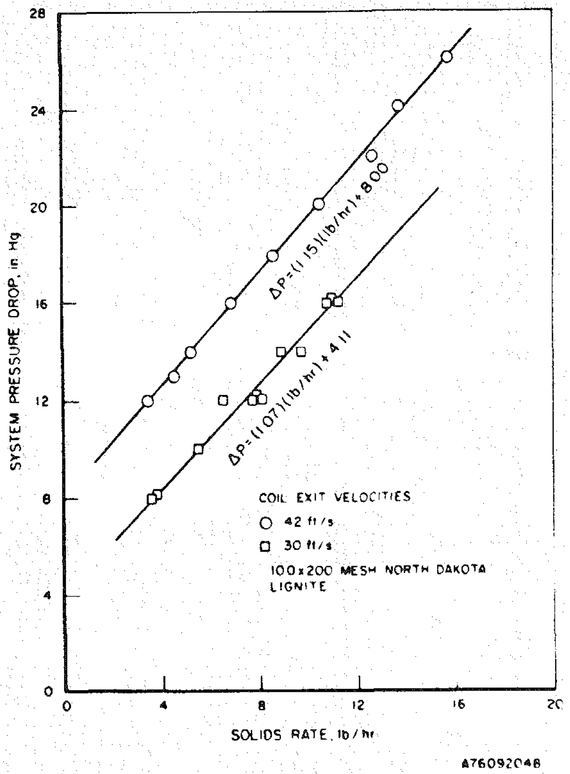

Figure 11. INCREASE IN SYSTEM PRESSURE DROP WITH SOLIDS FLOW RATE (100x200 Mesh North Dakota Lignite)

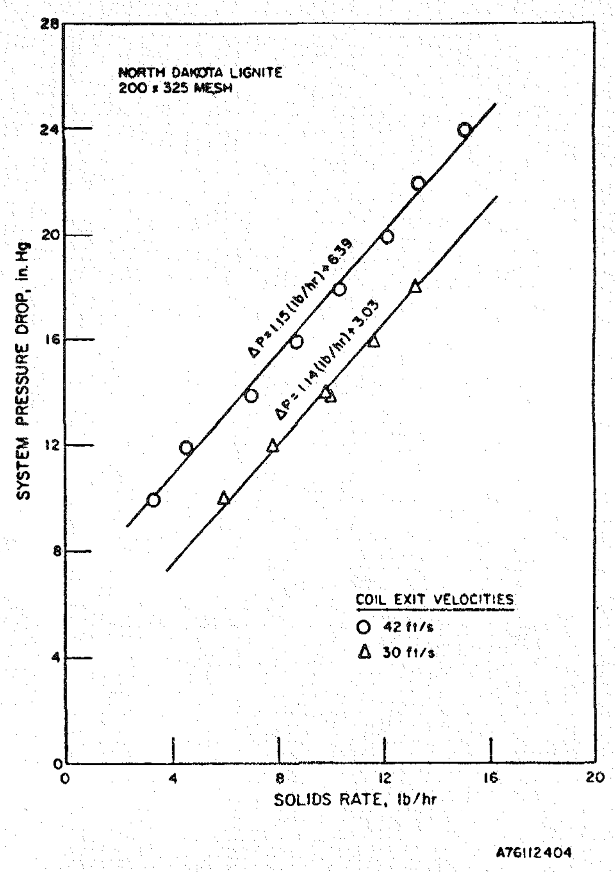

Figure 12. INCREASE IN SYSTEM PRESSURE DROP WITH SOLIDS FLOW RATE (200x325 Mesh North Dakota Lignite)

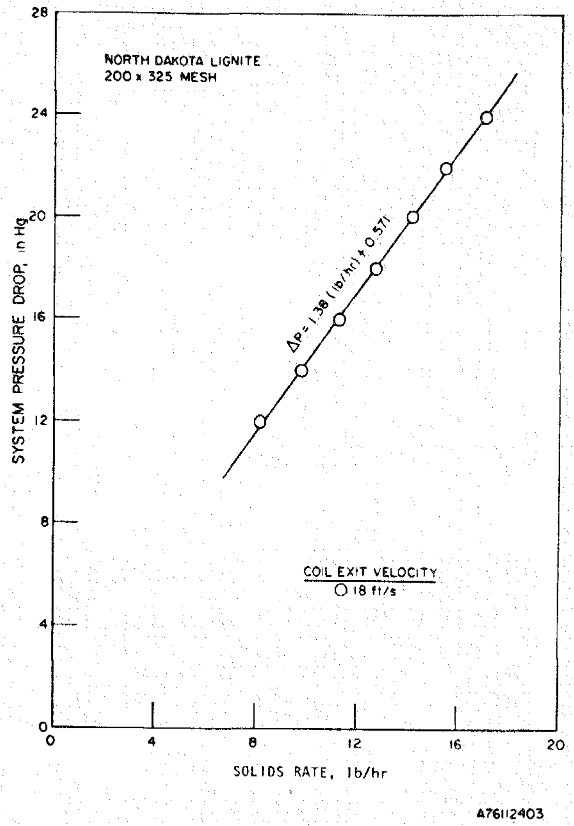

Figure 13. INCREASE IN SYSTEM PRESSURE DROP WITH SOLIDS FLOW RATE AT LOW VELOCITY (200x325 Mesh North Dakota Lignite)

Because ρ_g and u_o are functions of pressure, they can be expressed as functions of pressure through the ideal gas law for the isothermal case. When this is done, Equation 6 can be integrated over the length of the tube, obtaining

$$p_a^2 - p_b^2 = \frac{2RT}{M}\left[\frac{G^2 fL}{2g_c r_h} - \frac{G^2}{g_c}\ln\frac{(\rho_{ga})}{\rho_{gb}}\right] \tag{7}$$

Using methods given in Section 5 of Perry's Handbook,[11] this expression can be used to evaluate pressure drop through coiled tubes.

The results of calculations made in this manner for the cold-flow model of the coal feeder are summarized in Table 3, in which the expected pressure drop is compared with the observed pressure drop for several flow rates. Neither the calculated nor the observed pressure drop agree well with the values that would be expected by extrapolating the curves shown in Figures 11, 12, and 13 to zero solids-flow rate. An attempt was made to estimate the contribution of solids present in the gas solids stream using the expression

$$\Delta p_s = \frac{\pi}{2}\, f_p\, \frac{\rho_g u_o^2}{2g_c}\left(\frac{\rho_s}{\rho_g}\right)^{1/2}\frac{L}{d_t}\left(\frac{G_s}{G}\right) \tag{8}$$

suggested by Kunii and Levenspiel[7], but the estimates formed underestimated the pressure drop actually observed. A more accurate model might be obtained by modifying Equation 8 to account for two effects. These are the effects of changing pressure on superficial gas velocity, and the pressure drop accompanied by the energy transport involved in the continued acceleration of the solid particles toward the axis of the coil.

Summary of Cold-Flow Modeling

Development work with a Plexiglas cold-flow model of the solids feeder has shown that uniform and reproducible solids-feed rates can be obtained with 60x100-, 100x200- and 200x325-mesh lignite. In operation, the cold flow can be controlled by controlling the pressure drop through the system. Attempts to model the system-pressure drop from first principles were not successful.

Reactor Operations

Feed Material

A 2-ton shipment of lignite was obtained from North American Coal Company's Indianhead mine for use in the project. This lignite was ground in a hammer mill by Project Lignite personnel at the University of North Dakota, and repackaged in plastic-lined drums under a nitrogen blanket prior to shipment to IGT. The screen analysis of a ground lignite sample, supplied by Project Lignite personnel, is shown in Table 4.

Table 3. CALCULATED AND OBSERVED PRESSURE DROP THROUGH THE COLD-FLOW PLEXIGLAS FEEDER

(Critical Reynolds Number = 6648)

Rotameter Setting	CFH	Inlet Velocity, ft/s	Reynolds Number	Calculated Pressure Drop, psi	Observed Pressure Drop, psi
16	30.84	47.05	5454	5.41	5.04
14	27.25	43.17	4819	4.51	4.44
12	23.65	39.16	4183	3.78	3.62
10	19.58	33.43	3463	2.92	2.95
8	16.46	29.16	2911	2.33	2.29
6	13.38	24.56	2268	1.86	1.78
4	9.66	18.51	1709	1.17	1.20
2	5.27	11.18	932	0.48	0.47

Table 4. LABORATORY REPORT OF SIEVE ANALYSIS OF GROUND NORTH DAKOTA LIGNITE

U.S. Screen Mesh	Retained on Sieve, wt %	Cumulative Retained, wt %
65	6.6	6.6
100	7.5	14.1
150	9.9	24.0
170	5.8	29.8
230	16.3	46.1
325	29.9	76.0
Pan	24.0	100.0

Preliminary Testing

In making shakedown tests, nitrogen and air were passed through the system. During the shakedown tests, a flake of scale that had formed on the feed-hopper walls was dislodged, entered the coil, and jammed, partially blocking gas flow through the system. The flake was removed (with difficulty) by back-flushing the coil with high-pressure nitrogen. To remove the scale, the feed hopper was filled with sand and fluidized for several hours; after the treatment, the interior of the hopper was coated with an epoxy resin to stabilize the surface. Using the calculation method described earlier, expected values for system-pressure drop were calculated for several flow conditions and were compared with the observed pressure drop. The results of these tests are summarized in Table 5. Good agreement was obtained, showing that the coil was free of blockage. No solids were used in these tests.

Table 5. SYSTEM PRESSURE DROP UNDER VARIOUS OPERATING CONDITIONS (Critical Reynolds Number = 6511), USING NITROGEN AT ROOM TEMPERATURE

System Outlet Pressure, psig	Ambient CF/hr	System Outlet Velocity, ft/s	Reynolds Number	Calculated System Pressure Drop, psi	Observed System Pressure Drop, psi
0	45.3	160.2	9,658	31	29
0	25.4	89.7	5,406	14	14
0	55.8	197.4	11,099	37	39
125	72.6	27.1	15,478	15	25
200	187.3	45.4	39,939	49	50
200	177.8	43.1	37,910	45	50
425	327.0	38.7	70,537	69	75
488	150.3	15.6	30,722	16	13

Calculation Study

A calculation study explored the expected flow regimes (laminar or turbulent as measured by Reynolds number) in the bench-scale unit. Flow through coils is stabilized by the curvature of the coil so that the change from laminar to turbulent flow takes place at higher Reynolds numbers than for straight conduits. Methods for calculating the Reynolds number at which the transition takes place are given in Perry's Handbook.[11] Flow in the bench-scale unit is expected to be laminar for pure hydrogen at the temperatures, pressures, and velocities of interest (Figure 14). For nitrogen or feed gases having a density similar to nitrogen (Figure 15), flow is expected to be turbulent.

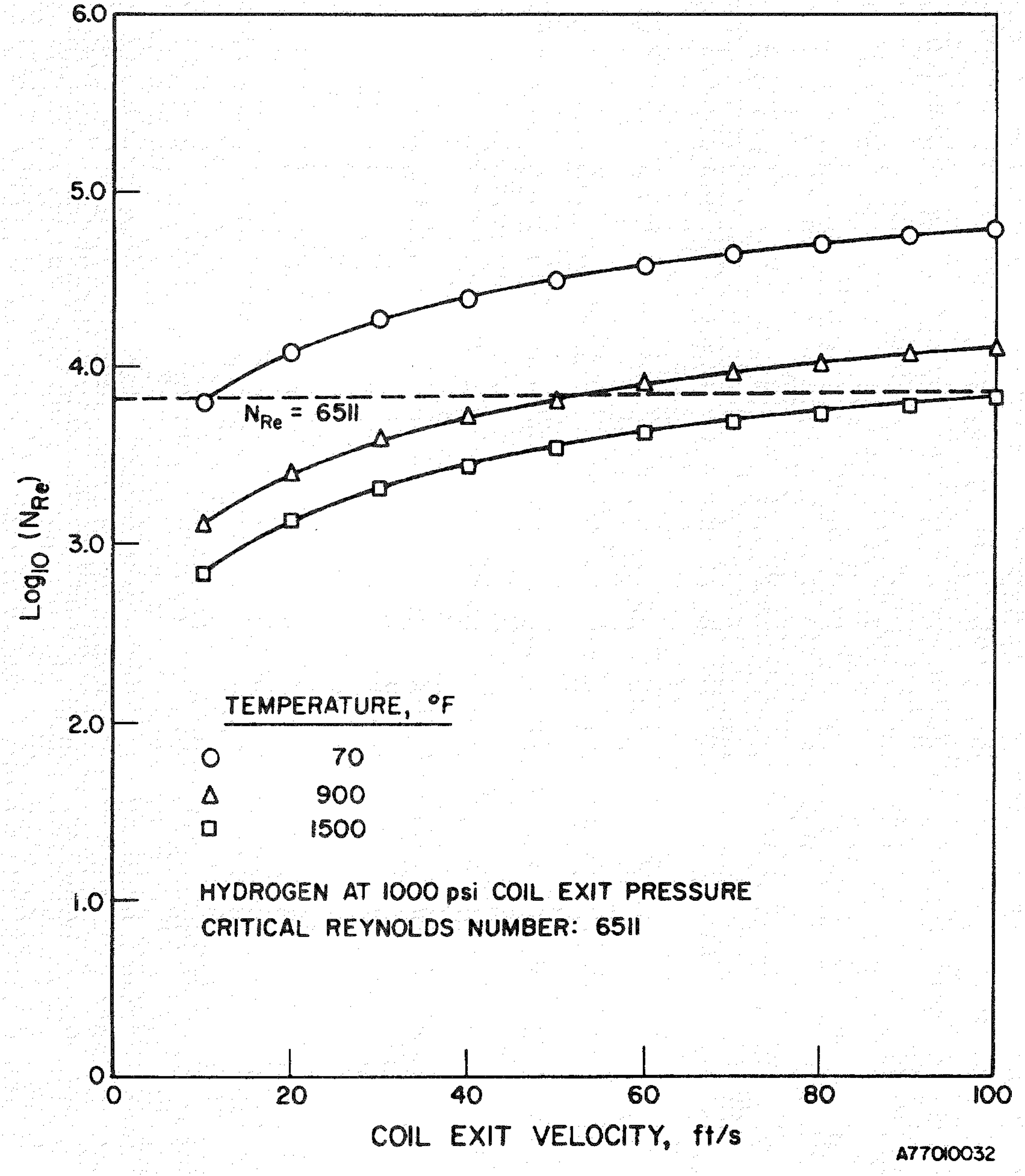

Figure 14. CHANGE IN REYNOLDS NUMBER WITH COIL EXIT VELOCITY FOR HYDROGEN AT 70°, 900°, AND 1500°F

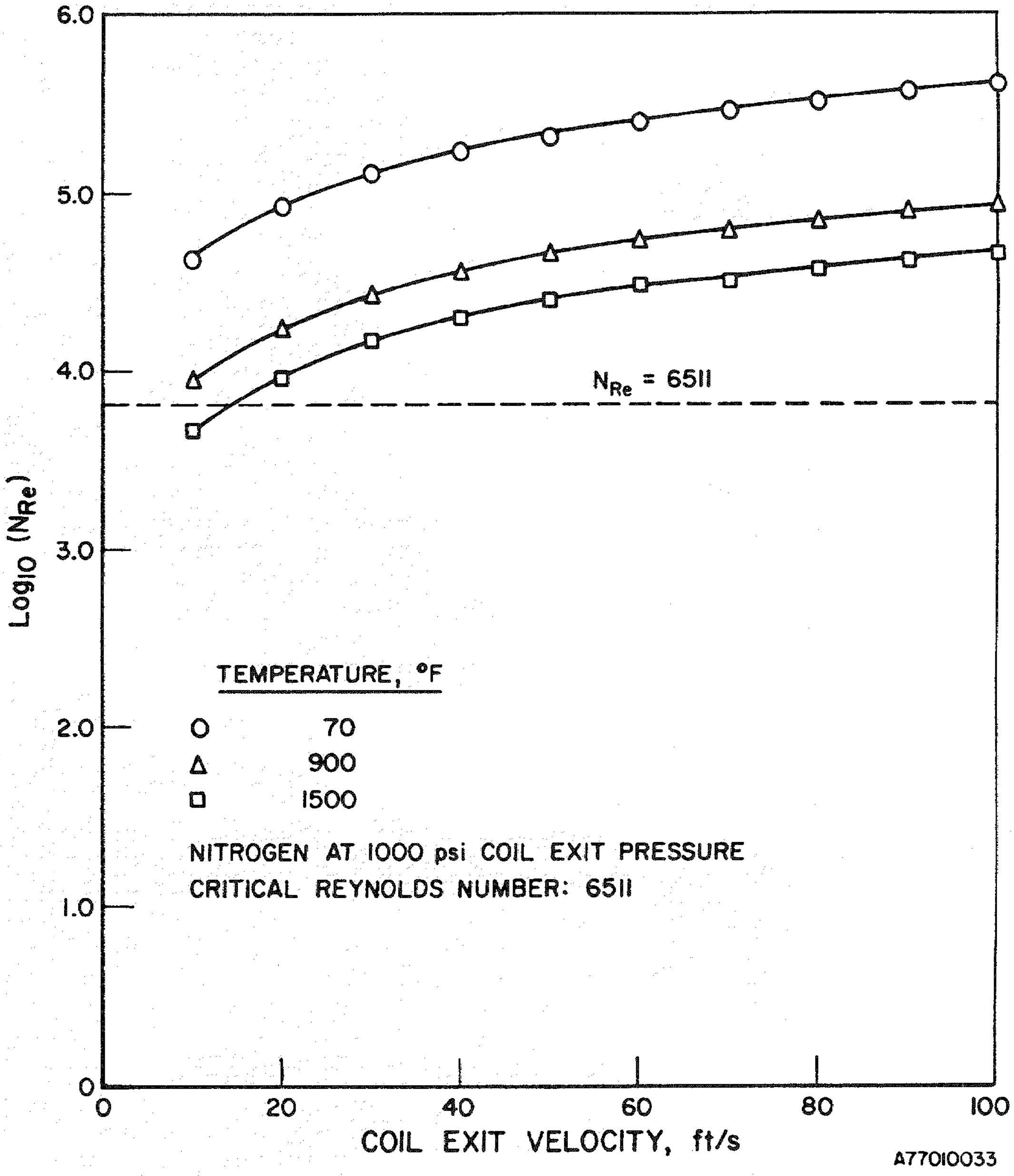

Figure 15. CHANGE IN REYNOLDS NUMBER WITH COIL EXIT VELOCITY FOR NITROGEN AT 70°, 900°, AND 1500°F

Start-Up

A list of the runs made thus far is shown in Table 6. The purposes of these runs were largely to test the operation of the equipment, uncover deficiencies, and make a broad excursion into the effects of temperature, pressure, and solids level in the feed stream. In the first preliminary run (P-1), the system outlet pressure was set at 500 psi, and the coil outlet temperature was maintained at 1000°F. This run was ended when the coil plugged after approximately 20 minutes of operation. An insignificant amount of liquid product was collected. The material balance was poor; no further analysis of the data was attempted when it became apparent that a good closure would not be obtained in the material balance.

January Testing

An attempt was made to operate the coil isothermally at 1200°F and a system outlet pressure of 1000 psig. During heat-up, a malfunction in the temperature control system occurred causing sections of the furnace to overheat. Two heater elements then burned out, at the same time melting sections of the coil. Examination of the wiring showed that no error had been made in assembling the equipment. The overheating was traced to drafts of cold air entering the furnace through the aperture formed by the closure of the two halves of the furnace. These drafts played over the thermocouples and caused them to read low. Calculations also showed that expansion of the mandrel supporting the coil would be sufficient to lower some of the thermocouples away from the heater sections that they were intended to control. This was corrected by raising the mandrel 1-1/2 in. in the furnace. The damaged heater sections were rebuilt, and the coil was repaired by replacing the damaged section with new tubing. The furnace was sealed with tape to prevent drafts from outside the furnace from playing on the thermocouples and causing them to read low.

After the repairs were completed, the equipment was readied for a run (P-2) in which the coil would be operated isothermally at 1200°F and a system outlet pressure of 1000 psig. The system was pressurized with hydrogen, and the feed rate was set to deliver 2.43 cubic feet of hydrogen per hour to the system, corresponding to a coil inlet velocity of 9.0 ft/s and a coil outlet velocity of 28.3 ft/s. Solids feeding was started, and the system-pressure drop increased to approximately 38 psi, at which point it became stable. After approximately 20 minutes, the pressure drop across the system increased to 50 psi, showing the coil to be plugged. The supply pressure was increased from 1050 psig to 1100 psig; the block cleared momentarily and then reformed. The supply pressure was increased a second time to 1200 psig, but the plug could not be dislodged, and the run was terminated. During the run, an on-line gas sample was taken after 10 minutes of operation (Table 7).

After the run, the composite sample in the gas holder was sampled for analysis, and the gas holder was emptied. The system was then depressurized slowly, taking a portion of the gas into the gas holder through the cold trap; a sample of this was sent for analysis. The reactor was then allowed to cool to room temperature. When the system was being depressurized, feed lignite was observed to be carried into instrument lines, plugging the differential pressure gage. A portion of the feed lignite may also have been lost through the feed-hopper vent line.

Table 6. LIST OF RUNS MADE

Run Date	Objective	Results
P-1 12-17-76	Operate isothermally at 1000°F and 500 psig system outlet pressure	Partially successful; bridge in feed hopper stopped solids feeding after 20 min of operation
P-2 1-21-77	Operate isothermally at 1200°F and 1000 psig system outlet pressure	Partially successful; coil plugged after 15 min of operation
P-3 2- 2-77	Operate at 1200°F and 1000 psig system outlet pressure; replication of P-2	Successful, run ended voluntarily
P-4 2-15-77	Operate isothermally at 1400°F and 1000 psig system outlet pressure	Successful, run ended when feed supply of lignite was exhausted
P-5 2-25-77	Operate isothermally at 1500°F and 1000 psig system outlet pressure	Partially successful, coil plugged after 35 min of operation
P-6 3-23-77	Operate isothermally at 1400°F and 1500 psig system outlet pressure	Successful
P-7	Operate isothermally at 1500°F and 1500 psig system outlet pressure	Plugged

Table 7. GAS COMPOSITION OBSERVED IN OPERATING AT A COIL EXIT TEMPERATURE OF 1200°F AND SYSTEM OUTLET PRESSURE OF 1000 psig

Component	On-Line Operating Sample	Side-Stream After Freeze-Out	Gases Released in Depressurizing
	mol %		
Nitrogen	0.4	4.3	1.2
Carbon Monoxide	2.4	1.6	1.4
Carbon Dioxide	4.5	1.5	2.2
Hydrogen	87.5	91.0	92.4
Methane	3.5	2.1	1.9
Ethane	0.77	0.23	0.41
Propane	0.30	0.09	0.15
i-Butane	0.07	0.02	0.06
Ethylene	0.19	0.10	0.12
Propylene	0.21	0.07	0.11
Butenes	0.12	0	0.06

The information for the gross material balance was then obtained; 1.27 pounds of char were recovered together with 0.36 pounds of liquids. By difference, 3.42 pounds of lignite were removed from the hopper during the run and depressurization step. Calculation showed that 0.56 pounds of hydrocarbon gases and carbon oxides was present in the make-gas and in the gas vented when the system was depressurized. The poor material balance is suspected to be due to loss of feed lignite into the instrument lines and the atmosphere through the feed hopper vent.

An attempt was made to repeat the run; the plug in the coil (near the outlet) was removed, and the coil was repaired. The system was then pressure tested and heated to operating conditions. The coil plugged after only a few minutes of operation, ending the run. The first plug is believed to have been caused by tars condensing near the coil outlet (the char trap was not heated in this run), and the second plug appears to have been caused by packing of feed lignite in the supply lines during pressure testing.

February Testing

During February, a new coil was fabricated using Incoloy 800 to allow runs at higher pressures and temperatures than were possible with the original coil fabricated from chromium-molybdenum alloy steel. The temperature controllers were equipped with thermocouple break-protection circuits to prevent overheating in the event of a thermocouple failure. Some minor equipment repairs were made, and the mechanical vibrator for the feed hopper was replaced with a pneumatic vibrator. Some cold-flow tests were also made to test the operation of the feeder.

In Run P-3, the reactor was operated isothermally at 1200°F and a 1000 psig system outlet pressure. The equipment performed in a satisfactory manner and shut-down was voluntary. A good material balance was obtained (97.4%), but conversion was low, with the weight loss in the feed being only 15.8%.

In Run P-4, the reactor was operated isothermally at 1400°F and a 1000 psig system outlet pressure. The results of the run are summarized in Tables 8 and 9. The overall material balance showed that there were no large losses from the system. The make-gas was found to contain carbon oxides and light hydrocarbons in addition to small quantities of benzene and toluene. The quantity of benzene present in the make-gas would correspond roughly to the amount expected for benzene in equilibrium with liquid benzene at room temperature and system pressure. The product distribution (Table 11) — between char, water, light gases, carbon oxides, and hydrocarbon liquids — showed that water much in excess of that present in the feed lignite appears in the products. By calculation, this would correspond to approximately half of the oxygen present in the feed lignite, with the remainder of the oxygen appearing as carbon oxides. (See Table 10.)

The liquids recovered from Run P-4 were placed in a sealed container, and sodium chloride added to increase the density of the water phase and float the hydrocarbon phase. The hydrocarbon phase was then separated from the water phase by means of a separatory funnel, after which the hydrocarbon phase was weighed and sampled for analysis (Table 12). The char was examined for readsorbed material by distillation with steam at atmospheric pressure. Traces of a light oil appeared in the condensate, but, as in all cases so far, the weight loss of the char subjected to the steam distillation was negligible, showing that reaction products are not being readsorbed on the char in the char trap.

Two additional runs were attempted in February, but bridges formed in the feed hopper that prevented solids from passing into the reaction system. This appeared to be due to the inability of the pneumatic vibrator to shake the heavy feed hopper to keep the solids flowing freely into the feeder plenum chamber.

Table 8. OPERATING CONDITIONS FOR RUNS MADE

Variable	P-2	P-3	P-4
System Outlet Pressure, psig	1000	1000	1000
Coil Outlet Temperature, °F	1200	1200	1400
Residence Time, s	2.1	1.3	1.9
Lignite Feed Rate, lb/hr	7.2	2.1	7.2
Solids in Gas Stream, %	90.5	62.1	92.7
Liquid Recovered, % of feed lignite	nil	1.9	4.8

Table 9. MAKE-GAS COMPOSITIONS OBSERVED DURING RUN P-4

Time, min	10	20	30	40	50
Composition, mol %					
Nitrogen	0.2	0.0	0.0	1.0	0.5
Carbon Monoxide	2.8	5.7	5.3	5.9	6.1
Carbon Dioxide	1.9	3.4	3.4	4.2	4.6
Hydrogen	88.1	77.6	77.8	73.3	72.9
Methane	5.4	10.3	10.5	12.0	12.2
Ethane	1.3	2.4	2.3	2.8	2.8
Propane	0.12	0.25	0.27	0.33	0.36
Butanes	0.01	0.02	0.03	0.03	0.04
Ethylene	0.05	0.09	0.08	0.15	0.09
Propylene	0.02	0.03	0.04	0.06	0.06
Butene	0.0	0.01	0.01	0.01	0.01
Benzene	0.08	0.14	0.23	0.17	0.23
Toluene	0.02	0.03	0.05	0.05	0.08
Total	100.00	99.97	100.01	100.00	99.97

Table 10. OXYGEN BALANCE FOR RUN P-4

Source	Oxygen, g
Free Moisture in Lignite	249
Oxygen in Lignite	593
	842
Oxygen in CO_x	230
Water	634
	864

Table 11. DISTRIBUTION OF PRODUCTS FROM RUN P-4

	g	Feed Lignite, wt %
Hydrocarbon Liquids, Including BTX From Make Gas	150	5.0
Light Gases	287	9.6
Carbon Oxides	315	10.6
Char	1639	55.1
Water	713	24.0
Total	3104	104.3

Table 12. ANALYSIS OF HYDROCARBON LIQUIDS RECOVERED DURING RUNS P-4 and P-5

Run	P-4	P-5
Component, wt %	Wt %	Wt %
Unidentified	43.8	15.4
Methyl Naphthalene	3.7	1.3
Dicyclopentadiene	0.5	0.2
Naphthalene	9.9	20.6
Indene	1.3	0.5
Xylene	4.5	0.5
Aromatic C-9's	1.2	0.3
Toluene	11.9	11.6
Ethyl Benzene	0.8	0.6
Styrene	0.2	0.1
Benzene	9.5	47.8
Cyclopentadiene	3.4	1.1
IBP:	169°F	176°F
EP:	760°F	632°F
Recovery:	92 vol %	79 vol %

March Testing

During March, two runs aborted when the coil plugged at the start of solids feeding. Examination of the coil showed an abnormally high pressure drop for gas flow without solids, indicating a partial block. The coil was then heated to 1500°F, and air was passed through the coil to burn off any coke. The coil was then allowed to cool to room temperature and scoured with –60 mesh sand blown through the coil. Following this treatment, the pressure drop through the coil was normal.

Some additional tests were made to examine the manner in which the gas stream was partitioning between the feeder plenum chamber and the main transport line; the main problem with the feeder was found to be imparting enough vibrational energy to the feed hopper to maintain free flow into the plenum chamber. A modification of the vibrator mount was installed that, by visual observation of the discharge from the feed hopper, was capable of keeping the plenum chamber flooded with feed lignite. Some scale was observed to fall from the feed hopper, so an epoxy liner was applied to completely coat the interior of the hopper and stabilize the surface.

Two runs were then made using hydrogen-to-coal ratios of 0.079 (Run P-5) and 1.7 (Run P-6). The data from these runs permit the comparison of "dense phase" processing with "dilute phase" processing. The carbon conversion in the dilute phase was on the order of 50%, but the chemical analyses needed to calculate the actual conversion have not been completed, so a complete analysis of this run cannot be presented at this time.

In dense-phase processing, sufficient analytical information has been obtained to compare the results of an isothermal coil temperature of 1400°F and a 1000 psig system outlet pressure with an isothermal coil temperature of 1500°F and a 1000 psig system outlet pressure (Runs P-4 and P-5). The operating conditions, material balances, and other information for these two runs are shown in Tables 13, 14 and 15. At 1500°F, the total conversion of feed carbon to gas and liquid products was somewhat higher than at 1400°F, but the increase is manifested in an increase in carbon oxides and light gases; net liquids production actually decreased slightly, although the benzene yield increased.

The analyses of the feed lignite and spent char for these runs (Table 15) show that the potential weight loss (obtained by adding the volatile matter and moisture in the spent char to the observed weight loss in the feed) is greater than the weight loss that would occur through simple devolatilization of the feed lignite. Clearly, the products observed are more than those of a simple carbonization of the feed. The carbon/hydrogen weight ratio in the spent char is approximately twice the carbon/hydrogen ratio of the feed lignite, showing that a net hydrogen depletion occurred in spite of the high hydrogen pressure.

Table 13. OPERATING DATA, OVERALL MATERIAL BALANCES, AND CARBON DISTRIBUTION AMONG PRODUCTS FOR RUNS P-4 AND P-5

Run	P-4	P-5
System Outlet Pressure, psig	1000	1000
Coil Outlet Temperature, °F	1400	1500
Residence Time, s	2.2	2.0
Lignite Feed Rate, lb/hr	6.6	6.2
Solids in Feed Gas, wt %	92.6	92.7
Run Length, min	60	35
Overall Material Balances, g		
Lignite Feed	2976	1652
Hydrogen	236	131
Total In	3212	1783
Hydrogen	157	106
Light Gases (C_1-C_4)	287	172
Carbon Oxides	315	217
Hydrocarbon Liquids	150	57
Water	713	418
Char	1639	803
Total Out	3261	1773
Carbon Balance, %	102.6	97.9
Hydrogen Balance, %	96.8	94.3
Ash Balance, %	89.8	97.2
Overall Mass Balance, %	101.5	99.4
Distribution of Carbon Among Products, wt %		
Liquids (85% carbon)	7.97	6.00
Carbon Oxides	7.51	9.35
Light Gases (C_1-C_4)	13.90	16.32
Char	70.60	68.33

Table 14 ANALYSIS OF FEED LIGNITE AND SPENT CHAR FOR RUNS P-4 AND P-5

Run	P-4	P-5
Feed Lignite, wt %		
Moisture	9.4	15.3
Volatile Matter	36.6	34.2
Ash	12.4	11.8
Fixed Carbon	41.6	38.7
Carbon (dry basis)	59.3	59.0
Hydrogen	3.89	3.85
Ash	13.67	13.96
Spent Char, wt %		
Moisture	0.2	0.3
Volatile Matter	12.8	11.0
Ash	20.2	23.6
Fixed Carbon	66.8	65.1
Carbon (dry basis)	71.6	69.0
Hydrogen	2.42	2.12
Ash	20.24	23.64

Table 15. AVERAGE MAKE-GAS COMPOSITIONS FOR RUNS P-4 AND P-5

Run	P-4	P-5
Component	mol %	
Carbon Monoxide	3.79	3.72
Carbon Dioxide	2.57	2.39
Hydrogen	57.20	58.80
Methane	7.40	7.44
Ethane	1.70	1.48
Propane	0.20	0.03
Butane	0.02	0
Ethylene	0.07	0.04
Propylene	0.03	0
Butylene	0.01	0
Benzene	0.12	0.05
Toluene	0.04	0
Xylene	0	0
Steam	26.84	26.04
	99.99	99.99

The compositions of the make-gas for Runs P-4 and P-5 are shown in Table 15. The water content of the make-gas was calculated by adding the moles of water produced during the run to the total moles of make-gas collected. Two important effects can be seen in the composition of the make-gas: the increase in temperature from 1400° to 1500°F was accompanied by a reduction in olefins, and the presence of steam and carbon oxides caused a large reduction in the effective hydrogen pressure.

The analyses of the liquids collected during Runs P-4 and P-5 are shown in Table 13. Both products would be classified as "high boilers." The liquids from Run P-4 are comprised of about 30% BTX constituents and large amounts of high-boiling aromatic compounds. The liquids from Run P-5 are over 60% BTX constituents; this increase is largely due to an increase in the coil outlet temperature of from 1400° to 1500°F; other processing conditions remained approximately the same.

Before making further runs, some revisions will be made in the equipment, as shown in Figure 16. By these changes, the make-gas will be dried and then passed through a dry ice/isopropanol freeze-out train to remove light oils (BTX) before metering and sampling. The feed-gas metering system will also be enlarged to allow separate control of the gas used to regulate the flow of solids from the feed hopper into the carrier gas stream.

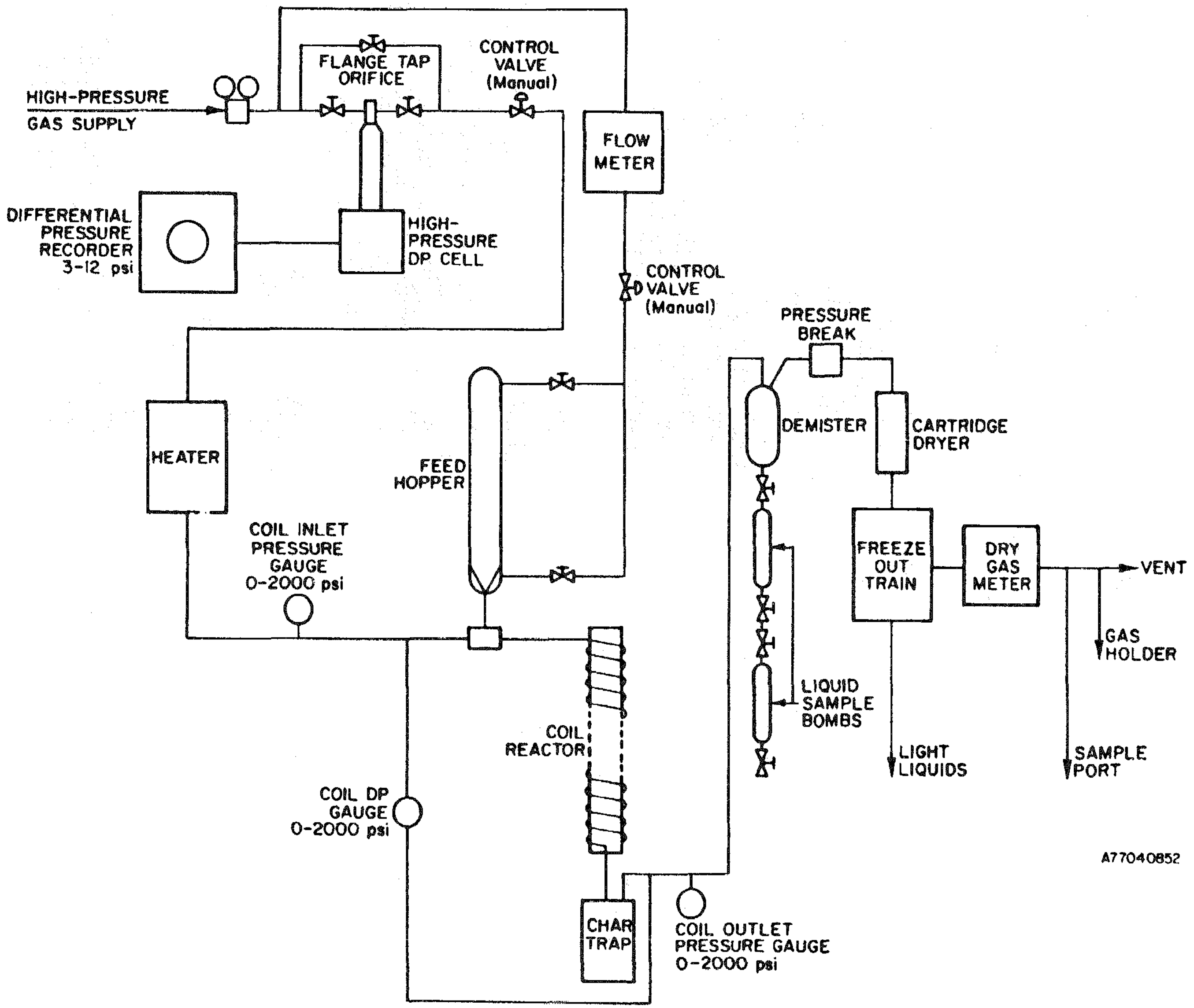

Figure 16. REVISED EQUIPMENT LAYOUT

SUMMARY

Introduction

This is the first annual report for ERDA Project FE-2307 "Riser Cracking of Coal" under Contract No. E(49-18)-2307. The objective of the program is to develop a noncatalytic process for the rapid hydrogenation of lignites and coals in a short residence-time entrained-flow reactor at high temperatures and pressures for the production of feedstock for synthetic motor fuels. Operating temperatures in the range of 900° to 1500°F and operating pressures up to 2000 psig are being used; solids residence time is being varied from 1 to 10 seconds. Both a bench-scale unit (5-10 lb/hr) and a process development unit (50-100 lb/hr) will be used in this program.

The development program is based on a commercial concept in which the use of a short residence-time riser reactor, of the type used in contemporary catalytic cracking, is extended to the conversion of coals and lignites to gaseous and liquid products by reaction with gases such as hydrogen, synthesis gas, or mixtures of carbon monoxide and steam. The gases also carry the feed coal through the riser reactor. A maximized production of high-octane gasoline constituents (C_4-400°F boiling range including BTX) is an important aspect of this investigation. Light gases (C_1-C_3) will also be produced and will contain substantial proportions of methane and other light hydrocarbons that could be used either for fuel or for petrochemicals feedstock. Spent char would be used for synthesis gas or hydrogen production for use in the riser reactor.

The process is expected to have more favorable economics than other known processes — such as the Bergius or Fischer-Tropsch processes — because of the high space-velocity (volume efficiency) of the reactor and also because less hydrogen would be consumed. The distillate fuel yields are expected to be lower than those of the Bergius process, but gas and gasoline yields are expected to be greater.

IGT Operations and Results

The IGT program has been divided into six tasks as follows:

Task 1. Planning
Task 2. Building and operating a bench-scale unit
Task 3. Designing a PDU
Task 4. Building a PDU
Task 5. Operating a PDU
Task 6. Technical and economic evaluation of work with the bench-scale unit and PDU

The first task has been completed, and the bench-scale unit has been built and is presently being operated to study the effects of the basic process variables of temperature, pressure, gas composition, and residence time. At this writing, six runs have been completed (Table ES-1).

The processing concept under investigation is noncatalytic, but some work with catalysts is planned, at least in the bench-scale unit.

In Task 3, data obtained from the bench-scale unit will be used to design a process development unit (PDU). The PDU design and construction are scheduled to be started during the second project year. In operations with the PDU (Task 5), emphasis will be placed on 1) simulating a viable commercial operation and 2) obtaining material balances and other information needed for further scale-up. In Task 6, a technical and economic evaluation will be made of short residence-time coal hydrocracking.

Operations with the bench-scale unit were started in the last quarter of the year using North Dakota lignite as feedstock; six runs have been completed. Operating conditions and important results for these runs are summarized in Table ES-1.

Some problems were encountered in separating water from the hydrocarbon liquids; at present, the liquid phase is acidified with hydrochloric acid, and then sodium chloride is dissolved in the aqueous phase to increase the specific gravity and float the hydrocarbon phase. The water phase is then removed from the hydrocarbon phase by means of a separatory funnel. The odors of both ammonia and hydrogen sulfide have been detected when handling the liquid products, although neither of these constituents has been detected in the gas analyses.

Trends apparent from the data were as follows:

- Higher carbon conversion is favored by increases in both hydrogen partial pressure and temperature.
- Examination of gas analyses for dense-phase runs has shown that the actual hydrogen content of the make-gas is 50 to 60 mole percent, compared with the hydrogen content of the make-gas in dilute-phase runs which was more than 97 mole percent. The large decrease in hydrogen content in dense-phase operation is due to the presence of water vapor and other reaction products.
- At the relatively low temperature of 1200°F and 1000 psig, small quantities of alkanes and olefins were detected including propylene, butenes, and pentenes.
- At 1500°F and 1000 psig, the amount of light hydrocarbons was greatly suppressed, with methane predominating as the chief non-aromatic hydrocarbon.
- In raising the reaction temperature from 1400° to 1500°F at the 1000 psig level, a reduction in liquid yield with an increase in BTX yield was observed.
- There is reason to suspect that the aromatic light oils may be adsorbing on sample lines or sample bomb walls, so that the reported gas analyses tend to underestimate the quantities of hydrocarbons present in the make-gas.

Future Work

After finishing the present series of runs, the solids-feeding system will be revised to improve the reliability of the feeder. The equipment to heat the carrier gas will also be modified; this will increase the gas velocity at the coil entrance from 5 ft/s to values at which gas-solids flow is more stable. In operations thus far, cold feed gas has been used, and it has been possible to heat the feed gas and coal to operating temperature in the first 25 feet of coil length; the last 50 feet of coil have been operated isothermally. At the highest temperatures used (1500°F), plugs have formed that are believed to be formed from coking on the coil walls. It may be possible to reduce the tendency to plug by lowering the heating rate and lengthening the residence time. To accomplish this, a longer coil will be installed.

Conclusions

No major difficulty is anticipated in operating the 1/8-inch ID bench-scale reactor on lignite and in achieving project goals.

LITERATURE CITED

1. U. S. Bureau of Mines, "The Bureau of Mines Synthetic Liquid Fuels Program, 1944-1955. Part 1.—Oil from Coal," Report of Investigation 5506, (1959).

2. Coates, R. L., Chen, C. L. and Pope, B. J., "Coal Gasification in a Low Pressure, Low Residence Time, Entrained Flow Reactor," ACS Div. Fuel Chem. Prepr. 18, No. 1, 165-81 (1973).

3. Fallon, P. and Steinberg, M., "Coal Liquefaction by Rapid Gas Phase Hydrogenation." Paper presented at the 169th National Meeting of the American Chemical Society, Philadelphia, April 6-11, 1975.

4. Fallon, P. and Steinberg, M., "Design and Preliminary Results of the BNL 1 lb/hr Coal Flash Hydropyrolysis Experiment." Paper presented at the 173rd National Meeting of the American Chemical Society, New Orleans, March 20-25, 1977.

5. Hill, G. R. and Wood, R. E., "Rapid Conversion of Hydrogen-Coal Slurries to Liquid Fuels in Small Diameter Tubes." Paper presented at the 164th National Meeting of the American Chemical Society, New York, August 27-September 1, 1972.

6. Hirst, L. L. et al., "Estimated Plant and Operating Costs for Producing Gasoline by Coal Hydrogenation," U. S. Bureau of Mines Report of Investigation 4564, (1949).

7. Kunii, D. and Levenspiel, O., Fluidization Engineering, New York: John Wiley & Sons, 1969.

8. McCabe, W. L. and Smith, J. C., Unit Operations of Chemical Engineering, New York: McGraw-Hill Book Company, 1963.

9. Oberg, C. L. et al., "Coal Liquefaction Under High-Mass Flux and Short Residence Time Conditions," ACS Div. Fuel Chem. Prepr. 22, No. 2, 185-96 (1977).

10. Pelofsky, A. H., Greene, M. I., and Ladelfa, C. J., "Short Residence Time (SRT) Coal Hydropyrolysis," ACS Div. Fuel Chem. Prepr. 21, No. 5, 154-69 (1976).

11. Perry, J. H. ed. Chemical Engineer's Handbook, 4th ed., New York: McGraw-Hill Book Company, 1963.

12. Rosen, B. H., Pelofsky, A. H. and Greene, M. I., "Coal Hydrogenation to Produce Liquids," U. S. Patent 3,960,700 (1976) June 1.

13. Schroeder, W. C., "Hydrogenation of Coal," U. S. Patent 3,030,297 (1962) April 17.

14. Schroeder, W. C., "Hydrogenation of Coal," U. S. Patent 3,152,063 (1964) October 6.

15. Wiser, W. H., "Hydrogenation of Western Coal in Dilute Phase Under Medium Pressures for Production of Liquids and Gases," Final Report, ERDA Contract FE(49-18)-1200, (1975).

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