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

## (RISER CRACKING OF COAL)

First Quarter Report
For the Period April 1 to June 30, 1976

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# TABLE OF CONTENTS

## LIST OF FIGURES



## LIST OF TABLES

# ABSTRACT

The starting date of this ERDA/FE contract to develop a processing system for coal conversion to synthetic motor fuels (Riser Cracking of Coal) was April 1, 1976.

During the first quarter of the project, the bench-scale unit design was finalized and the procurement of equipment began. Two areas in the pilot plant building at IGT are being prepared for this project: one to hold the bench-scale unit and another, larger bay to eventually house the PDU.

An experimental plan was outlined with emphasis on the first year of operation. This plan and the bench-scale unit design were presented to, and subsequently approved by, ERDA/FE.

An aerated feeding system was developed using a Plexiglas simulation. The range and consistency of the mass flow met our requirements. A high-pressure version will be built in steel for the bench unit.

A supply of North Dakota lignite was secured as the first feedstock through arrangements with the University of North Dakota.

## I. OBJECTIVE AND SCOPE OF WORK

The program objective is to maximize the production of gasoline constituents from coal using short residence time, hydrocracking techniques. The specific process to be developed is called "Riser Cracking of Coal." A bench-scale unit will be built to define basic design parameters, and, subsequently, a 100 lb/hr process development unit will be designed and operated.

## II. SUMMARY OF PROGRESS TO DATE

Task 1, the detailing of the bench-scale unit design and the experimental plan, has been completed and approved by ERDA/FE. A Task 1 report has been submitted.

All of the equipment required to build the basic bench-scale unit has been ordered. Installation of certain pieces of equipment has begun.

An aerated feeding system has been developed using a Plexiglas simulator.

Arrangements to obtain lignite feedstock have been made.

We believe that we are slightly ahead of schedule at this time, and we have shown our progress in Figure 1 as being about 3 weeks ahead of schedule.

7/76

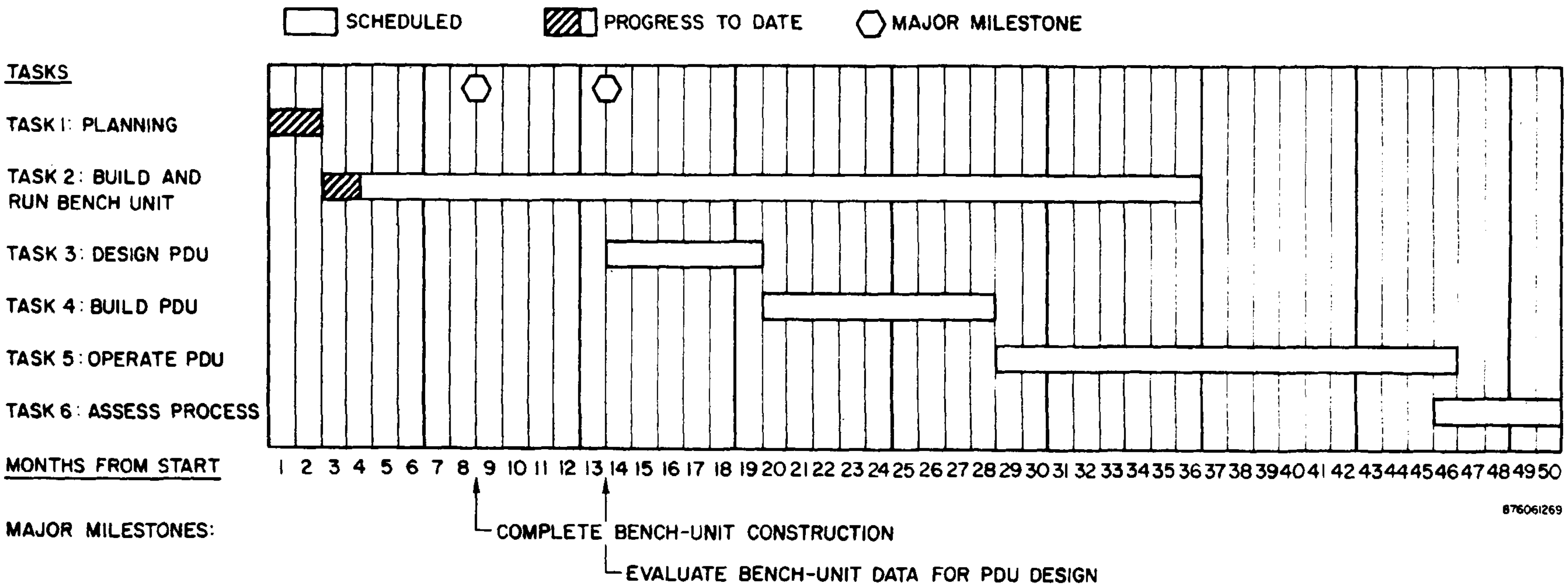


**Figure 1. PROGRESS CHART**

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## III. DETAILED DESCRIPTION OF TECHNICAL PROGRESS

### A. Task 1. Work Plan For Task 2

#### 1. Work Accomplished

##### a. Bench-Scale Unit Design

A detailed design of a high-pressure bench-scale hydrocracking system has been completed. The objective of the bench-scale unit is to simulate the flowing conditions of the riser cracking of coal. This design permits the accurate measurement of temperature, as well as control and variation of residence time in the reactor.

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

Coal Feed Rate: Up to 10 lb of coal per hour.

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

Alternative Carrier Streams:
1 — Hydrogen 100%
2 — Hydrogen and steam
3 — Hydrogen and carbon monoxide (Syngas)
4 — Syngas and steam

Pressures: 500 to 2000 psig.

Temperature: 900$^{c}$ to 1500$^{\circ}$F.

A flow diagram (Figure 2) shows the assembly of the individual components.

As a guide in the design of the various components of the system, a design operating pressure of 2000 psig was set as a minimum for the entire system. Of the various components, only the reactor furnace coil will approach the design allowable stress at 1500$^{\circ}$F operating temperature. At the severe conditions of 2000 psig and 1500$^{\circ}$F, Incoloy 800 furnace coil life will be limited to 1000 hours. At 1500 psig and 1500$^{\circ}$F, the coil life would be 10,000 hours.

###### 1) Coal Feed Hopper and Feeder

Coal will be ground, classified, and segregated in batches, which will be kept dry until ready for transfer to the feed hopper. The feed hopper has a capacity of 2/3 cu ft and is designed for pressurizing to 2100 psig for charging into the bench unit. It has a capacity of 20 lb of coal, assuming 30 lb/cu ft density in the hopper. A connection has been provided for nitrogen purging of the loaded hopper prior to charging. The feed hopper is designed for gravity feeding of the coal as uniformly as possible and at a continuous rate.

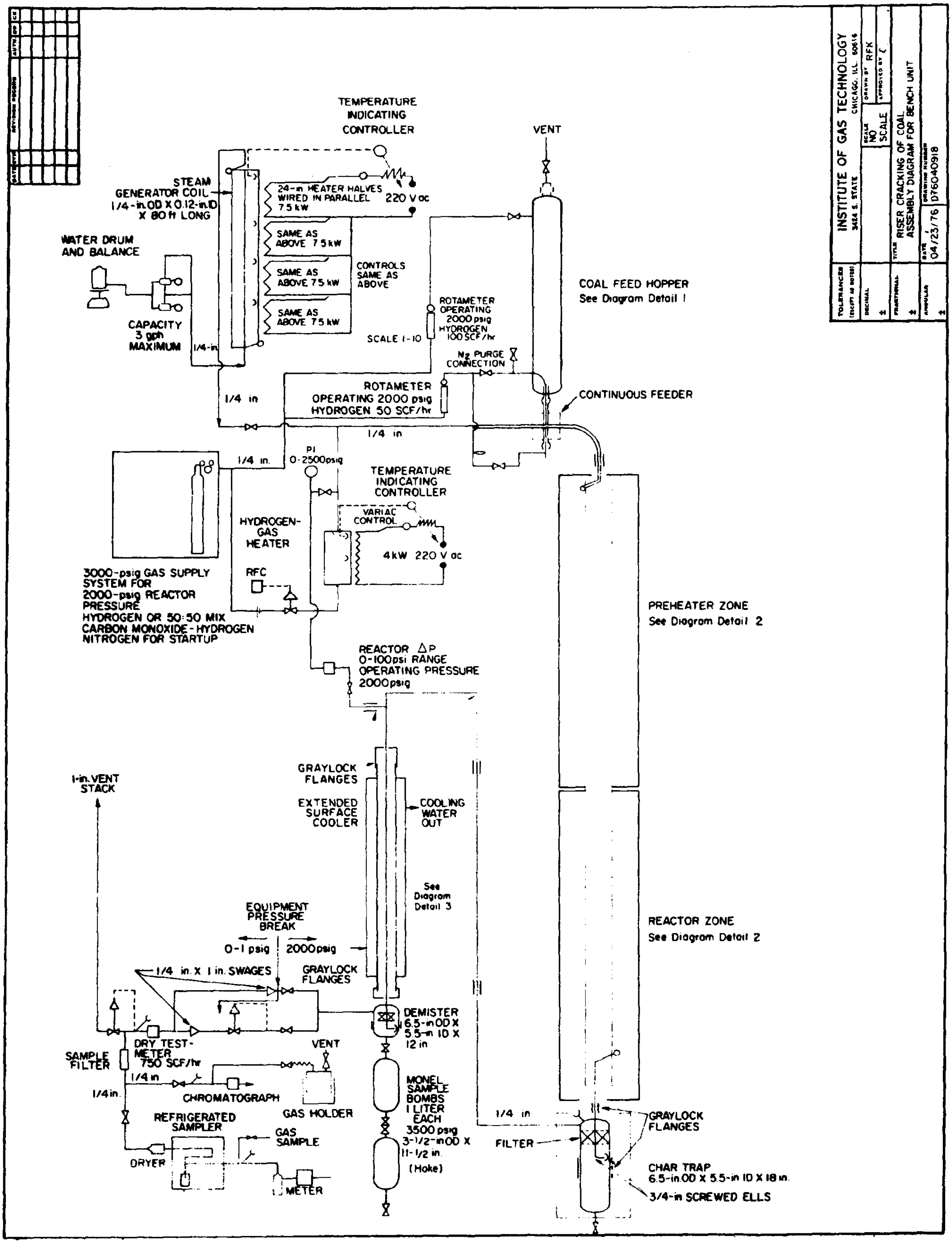


Figure 2. BENCH-SCALE UNIT FOR RISER CRACKING OF COAL

## 2) Carrier Gas Supply

Carrier gas will be supplied once-through by means of a 3000 psi storage system. Prior to a run it will be pressured up 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 will measure the water and raise its pressure, prior to vaporizing in a steam boiler, which 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 source of these gases is the carrier gas system.

A small heater has been provided on the carrier gas stream, mainly to keep this system warm and dry.

The details above are shown on the flow diagram of Figure 2.

## 3) Reactor Preheater

The main source of heat to the combined coal and carrier gas stream is a preheater coil located above the reactor coil and integrally enclosed 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.

## 4) Reactor Coil and Furnace

The reactor coil is designed to provide the range of flowing velocity of the carrier medium that would be obtained in a riser reactor. This range, 15 to 70 ft/s, may be obtained at a given pressure by varying the amount of carrier gas which accompanies the coal. The coal rate may also be varied to maintain a constant coal-to-gas ratio, where desired. Variation of coal rates and carrier rates for the fixed coil length and diameter provides a choice of residence time.

The 50-foot-long reactor coil will be made of Incoloy 800 tubing, 0.25-inch outside diameter by 0.12-inch inside diameter. Heat will be applied at many zones along the coil to permit changing the heating-rate curve. Thermocouples along the coil will monitor the temperature, employing installation techniques that have been successful in other high-temperature bench-scale work.

The coil is wound on a mandrel that serves to support it as well as the thermocouples. Each heating zone is independently monitored, and the monitored temperature is controlled.

5) Reactor Effluent Cooling System

The effluent of products, excess carrier-gas and unreacted char from the reactor coil will enter a char filter pot located immediately below the reactor. Effluent will be directed into the bottom section of the pot tangentially to the inside of the cylinder to effect separation of most of the char. The effluent, all vapor at this point, will then proceed upward through a filter for removal of remaining char fines. The upper part of the filter pot will be provided with a source of heat, so that natural heat loss will not result in tar condensation in this pot. The pot will be kept at about 1000°F and the gas residence time will be kept to a minimum by filling in the void space in the top of the vessel.

The filtered effluent will then proceed upward through an air cooler at a high enough velocity to preclude backflow of any condensate. This cooler will consist of a 12-foot length of 1/2-inch-OD extended surface condenser tube.

The partially-cooled effluent will then proceed down through a counter-flow water-cooled double-pipe exchanger. The inside tube through which the effluent flows is a 12-foot length of 1/2-inch OD extended surface exchanger tube identical with the air-cooled tube. Each tube provides 3.2 square feet of cooling surface with the water cooler being greatly more effective. The effluent is brought to a temperature of 300°F with the condensation of tar, oil and water in this cooler.

As shown on the flow diagram of Figure 2, the cooler drains downward into a gas-liquid separator located immediately below the cooler. The upper portion of this separator is identical, in construction, to the top of the char pot except that no heat is provided. The gases leave the top of the separator after passing through a vapor demister pad. The liquid from the bottom of the gas-liquid separator flows into a pair of one liter sample bombs, which serve the dual purpose of accumulators as well as containers for transporting the water-hydrocarbon sample to the laboratory for analyses.

6) Metering and Sampling System

The gas proceeds through a back-pressure regulator that holds the entire system pressure. It is metered at approximately atmospheric pressure in a dry test meter, and is then released to a vent stack. Small portions of the gas are filtered and routed through a continuous gravitometer to a gas holder for composite sampling. Another small portion of the gas from the meter proceeds through a dryer and to a refrigerated sampler. The gas portion from the refrigerated sampler is metered and sampled for analysis. The liquid condensed in the refrigerator sampler is measured and analyzed. The analyses of the refrigerated gas and liquid are combined in their observed ratio of production to provide an accurate composition of the main gas effluent stream.

The layout of the sampling system is depicted in the flow diagram of Figure 2.

b. Experimental Plan

1) First 15 Months

A program schedule for the first 15 months of work is shown in Figure 3. The areas of particular interest are: the completion of Task 1 (Planning), the beginning of Task 2 (Construction and Operation of a Bench-Scale Unit), and the beginning of Task 3 (Design of a Process Development Unit). Six months are allowed for the construction of the bench-scale unit, leaving about six months to perform experimental work relating to the design of the process development unit, or PDU. Because of the shortness of time, the initial experimental work will cover as much ground as possible, at the sacrifice of detail, which will be filled in later in the program.

While the bench-scale unit is being built, trials will be made to assess the operability of the equipment. In these trials, air, or nitrogen, will be used as the feed gas, and coke, or coal char, as the solid phase. The capability of making a good material balance will be demonstrated in these trials.

The principal variables to be investigated will 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 of these variables is shown in Figure 4. To complete such an array in a methodical manner would be time consuming. In order to develop as much information as possible to guide the design of the PDU, the number of experiments will be reduced in an ordered manner to confine experimental work to those regions where the expected liquid yields will be of sufficient quantity to be of interest. The path of experiments through arrays of pressure, temperature, residence time, particle size and temperature profile is shown in Figure 5, and is also summarized in Table 1.

The number of each experimental run is shown as the number within a box in an array starting with the number "1". The order has been arranged to favor ease of operation in the first runs and then the direction of expected decline in gas and liquid yields. Thus, the planned list of experiments might be shortened further as yields are found to be too low to justify completion of a series. At the end of the first phase of planned experiments (Milestone No. 1, Figure 3), the data will be examined and the effects of change in residence time evaluated. If possible, the number of operating pressures will be reduced to two.

In the next series of experiments, the effects of particle size will be explored. By reducing the particle size range from −150+200 to −325, the particle surface area per unit mass will approximately double, and the average particle diameter will be reduced by approximately one-half. Thus, mass transport across the gas film separating the particles from the bulk gas phase would be approximately doubled and the length of diffusion paths within particles greatly shortened. Superficially, particle heating would take place more quickly, leading to the more rapid release of volatile matter.

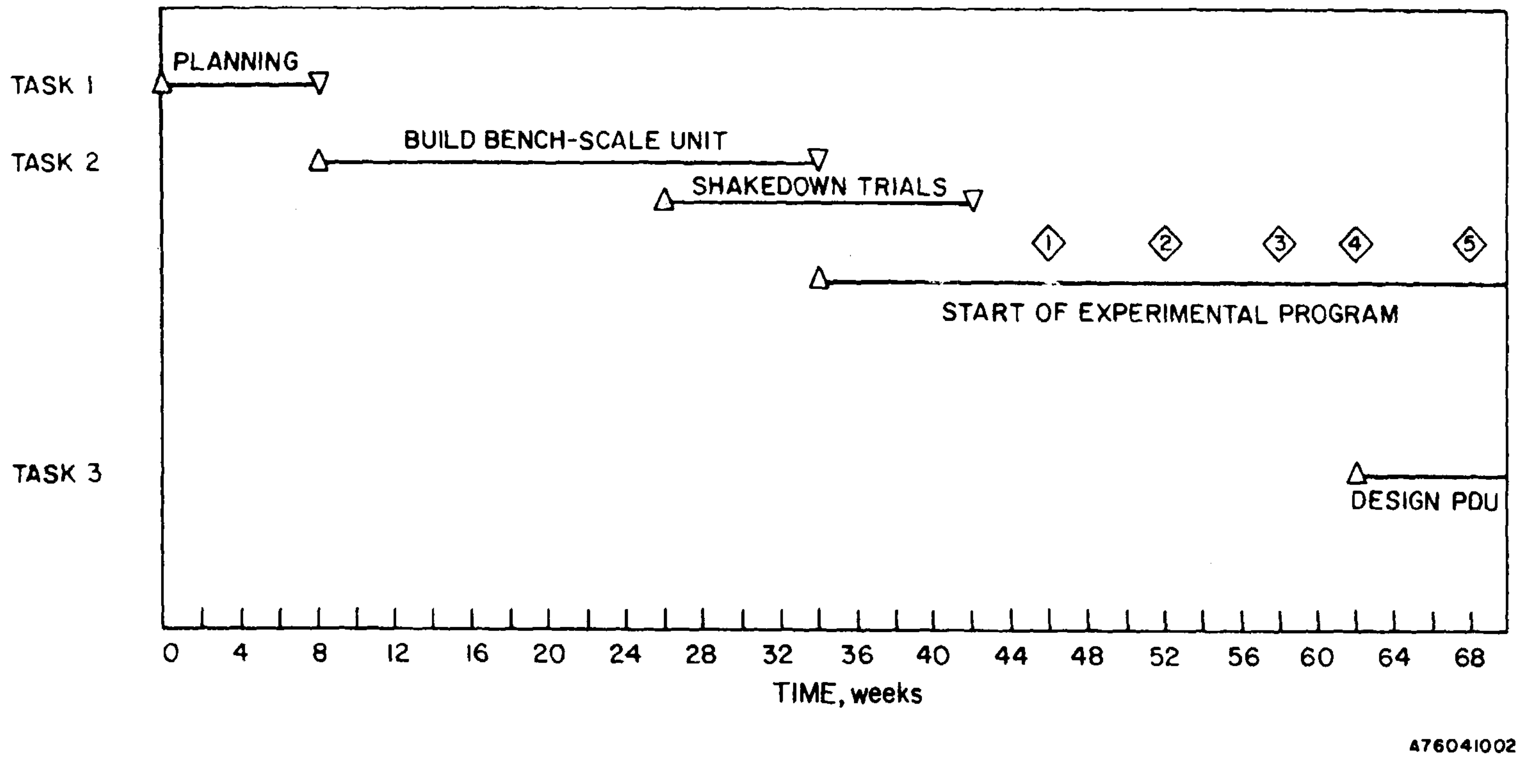


Figure 3. DECISION POINTS AND MILESTONES DURING FIRST YEAR OF EXPERIMENTAL WORK

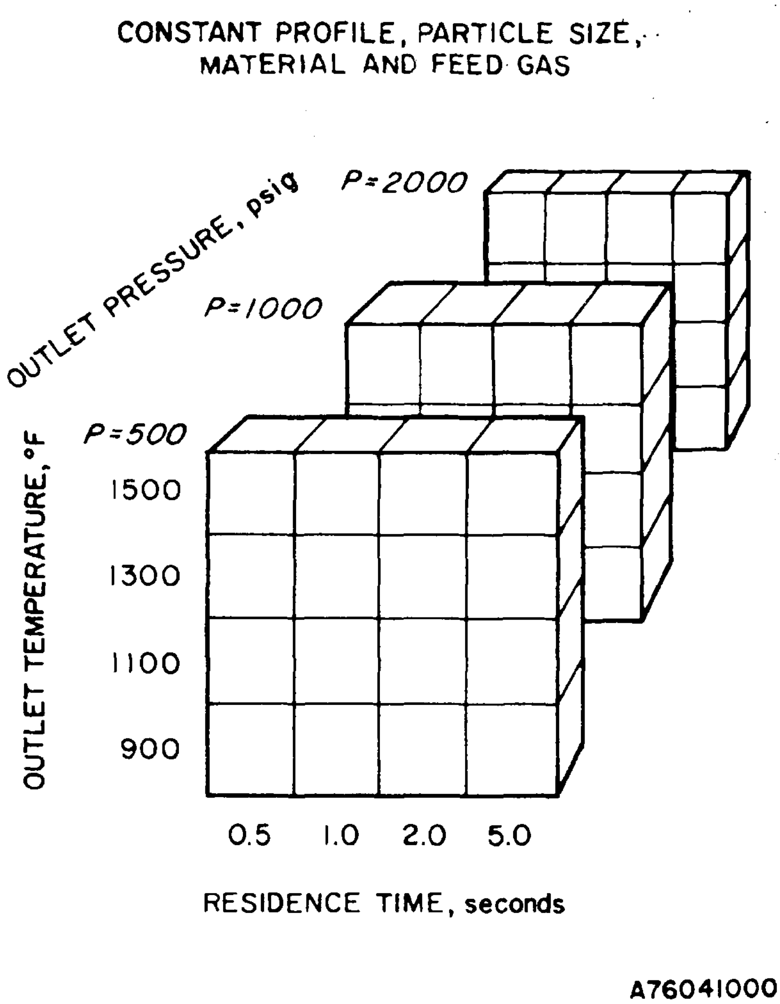


Figure 4. TYPICAL 4x4x3 ARRAY OF ORDERED EXPERIMENTS

7/76

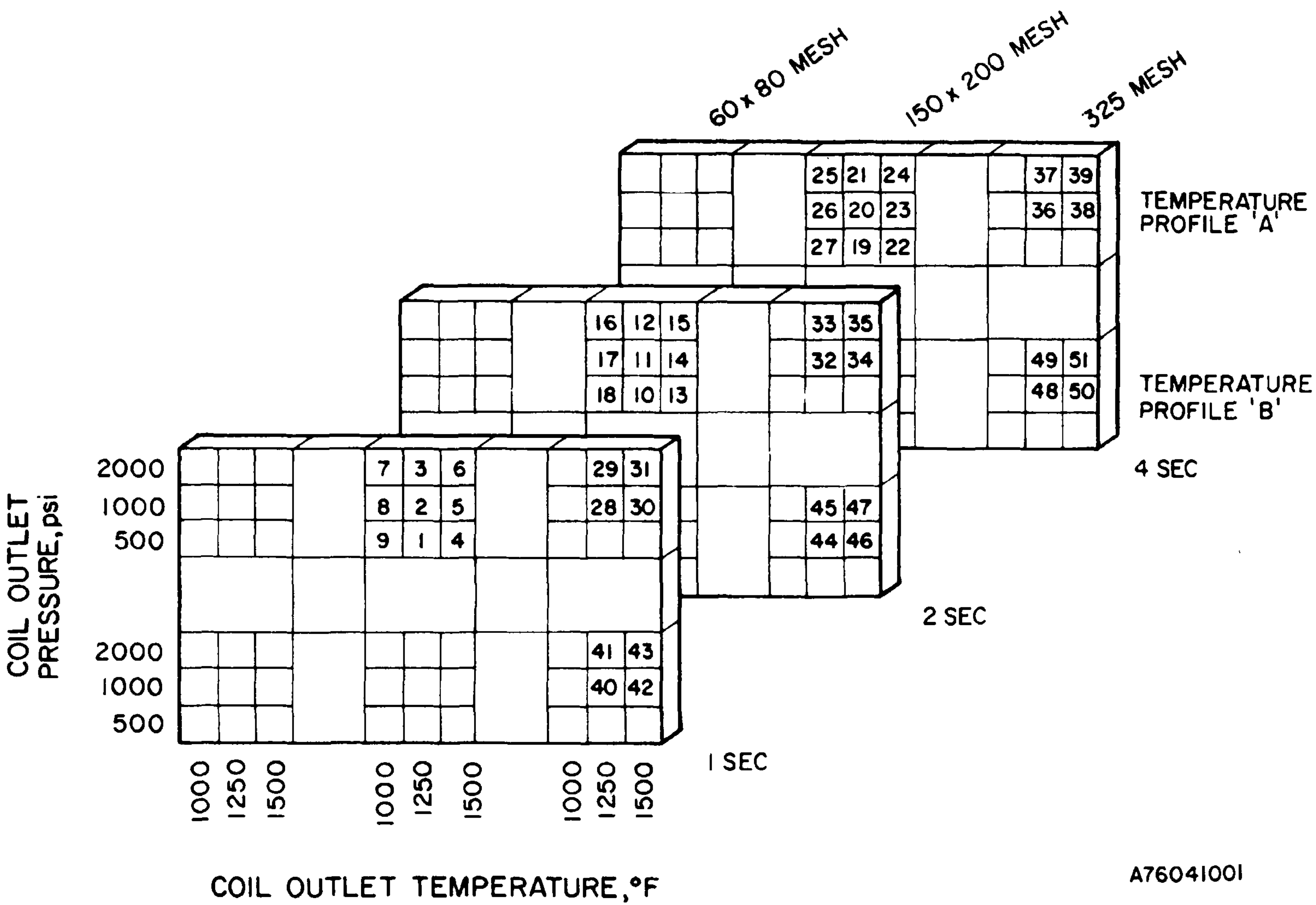


Figure 5. ORDERED PATH OF EXPERIMENTAL RUNS THROUGH PRESSURE-TEMPERATURE-RESIDENCE TIME-PARTICLE SIZE PROFILE ARRAYS

Table 1. RUNS FOR THE FIRST 15 MONTHS

| Runs* | Purpose |
|---|---|
| 1-27 | To evaluate the effects of coil outlet temperature, coil outlet pressure, and residence-time, holding the time-temperature history, feed-gas composition, and particle-size consist constant. In these runs, North Dakota lignite will be run with hydrogen. |
| | MILESTONE NO. 1: From analysis of the data, reduce the levels of operating pressure from three to two and determine the desirability of longer or shorter residence times. |
| 28-39 | Evaluate the effects of the increase in surface and reduction in particle radius obtained by using smaller particles. |
| | MILESTONE NO. 2: From analysis of the data determine if particle size reduction is beneficial. |
| 40-51 | Evaluate a second temperature profile at 1, 2, and 4 seconds residence time. |
| | MILESTONE NO. 3: From analysis of the data, determine if yields and distribution of products between gases, liquids, and solids can be markedly manipulated by change in temperature profile. |
| 52-64 | Make caking-coal trials; if initial runs are not successful, make trials in which coal ash is added to act as an adsorbent or antiblocking agent. |
| | MILESTONE NO. 4: From analysis of the data, determine what conditions, if any, will allow caking coals to be processed. Summarize and evaluate all data for PDU design. |
| 65-70 | Make trials using steam and hydrogen; carbon monoxide and hydrogen; and a mixture of steam, carbon monoxide, and hydrogen. |
| | MILESTONE NO. 5: From analysis of the data, assess the desirability of using syngas-like materials as feed gases. |
| 70-75 | If time permits, evaluate the use of catalytic materials added to the feed coal. The catalysis should be inexpensive. |

*The operating conditions for the first 51 runs are shown in Figure 5.

Pyrolysis reactions can be very roughly described as first order with respect to the amount of feedstock present, while hydrogenation reactions would be expected to be a function of the product of the amount of feedstock present and the hydrogen partial pressure. Changes in mass transport across the gas films surrounding the particles and changes in the heating rate of the particles may have some effect on the course of the pyrolysis and hydrogenation reactions. Examination of the distribution of products between gases, liquids, and solids should provide a basis for determining if change in particle size is an important effect and if there are potential benefits in using small particles.

The results of these runs would be examined (Milestone No. 2, Figure 3) and an assessment of particle size made. A change in residence time might also be considered at this time; to realize residence times of more than 5 seconds under all conditions will require a coil 100 ft or more in length.

After choosing a particle size for further work on the basis of the preceding experiments, the impact of changes in temperature profile on yield and product distribution will be explored. Upon completion of these experiments, three additional feed gases would be tested to explore the technical feasibility of using –

a. A mixture of 50 mole-percent hydrogen in steam,

b. A mixture of 50 mole-percent carbon monoxide in hydrogen,

c. A mixture of 50 mole-percent steam, 25 mole-percent carbon monoxide, and 25 mole-percent hydrogen.

The results of these runs will be assessed at Milestone No. 5. In addition, trials will be made using caking coal, with and without an adsorbent, or antiblocking agent, derived from coal ash, or other appropriate material (coal char, for example). The results of these trials will be evaluated at Milestone No. 4. In the final runs of the preliminary series, prior to the design of the PDU, some trials of catalysts will be made, if time permits.

### 2) Design of PDU

After approximately 12 to 15 months, the bench-scale unit will (hopefully) have been operated over a wide range of test conditions. The operating experience obtained in the early runs will be used to guide the choice of equipment for the PDU, particularly with respect to potential processing problems such as the collection of tar aerosols, uniform solids feeding, and the isolation of the products into solid, liquid, and gaseous fractions.

### 3) Continuing Studies in the Bench-Scale Unit

The continuing studies portion of the operation of the bench-scale unit will begin after the start of the PDU. The basic areas of investigation (Figure 6) will involve further investigations of the use of hydrogen, hydrogen/steam mixtures, synthesis gas and synthesis gas/steam mixtures as feed gases, and also the investigation of such variables as coal type, particle size, operating pressure, and the time-temperature history of the particles.

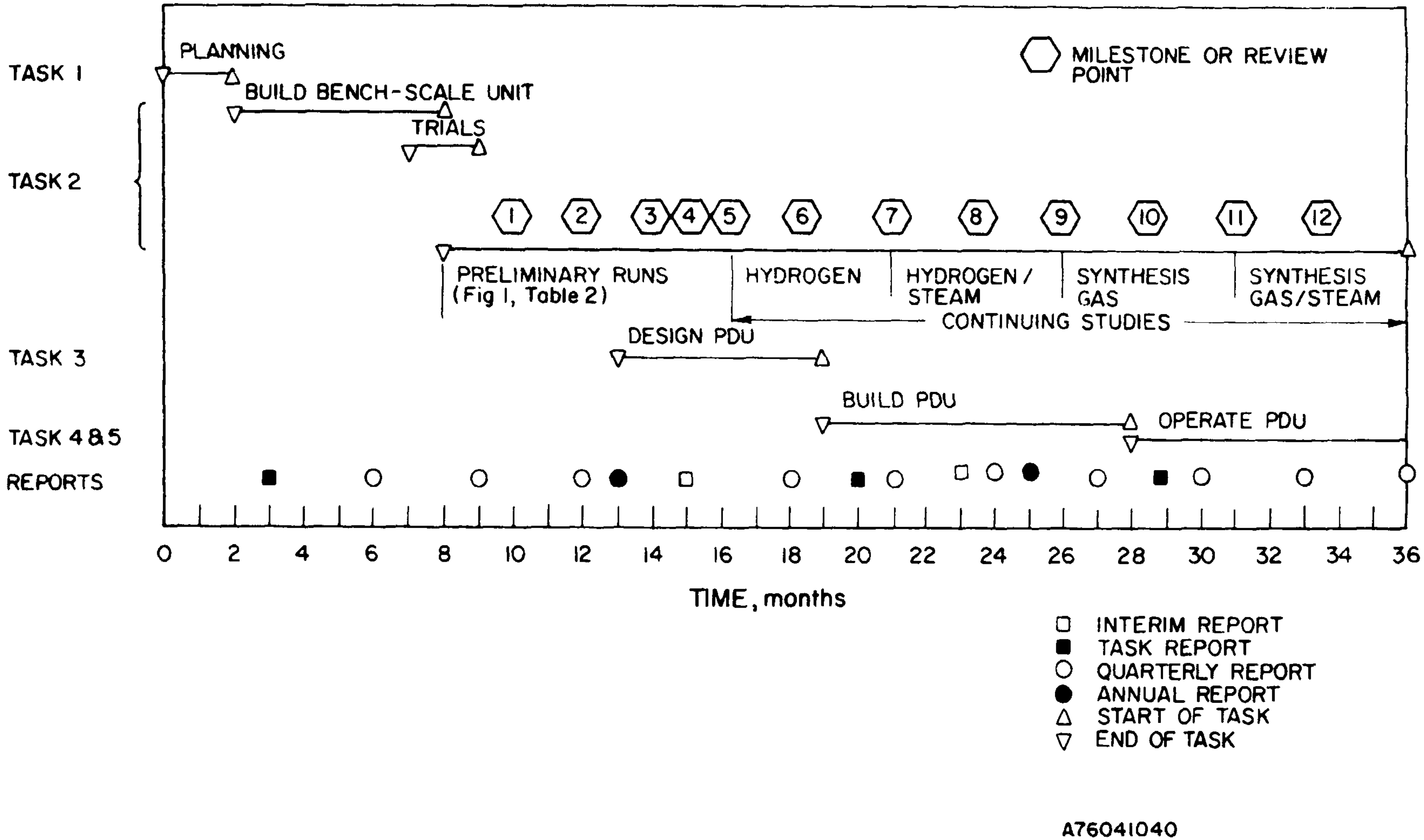


Figure 6. WORK PLAN FOR TASK 3

This portion of the experimental program would be reviewed periodically, as shown in Table 2, to assess the significance of results obtained. Further experiments would also be designed in these reviews. The basic experimental plans (Figure 4) would involve proceeding in an orderly manner through arrays of experiments in which pressure, temperature, and residence time would be the principal variables, holding coal type, particle size, feed-gas composition, and temperature-profile constant.

4) Basic Data

In operating the equipment, the following data would be taken for each run:

a. The amount of coal processed during the run, the proximate and ultimate analyses of the coal used in the run, and the particle size range.

b. The quantities of gas used to process the coal during the run, in the start-up and operation of the equipment. Transient conditions will be of short duration so that this information will convert to flow rates in a simple manner.

c. The quantity, composition, and specific gravity of the gases produced.

d. The weight and composition of any liquids produced; a minimum of a carbon-hydrogen analysis would be made, with identification of the species present being made when a more elaborate analysis seems justified. Any water will be separated from the liquids collected by appropriate means and reported as a part of the analysis of the liquids.

e. The quantity and composition of the solids residue; a minimum of a carbon-hydrogen analysis will be made. Where justified, both a proximate and ultimate analysis will be made.

Basic material balances will be made using this information, as well as elemental balances, particularly to explore the degree to which hydrogen is added to the products of the reactions.

In addition, operating information will be recorded, particularly the coil outlet pressure and temperature, and the variation in temperature over the length of the reactor tube, together with the control settings and observed values of all critical operating variables. This information will be used, where possible, to test mathematical models to explore the modeling of the conversions obtained.

Table 2. MILESTONES OR POINTS OF REVIEW FOR CONTINUING STUDIES IN BENCH-SCALE UNIT

| Milestone or Point of Review | |
|---|---|
| 6 | Review work with hydrogen and design experiments to complete work with hydrogen. |
| 7 | Review work with hydrogen and design experiments with mixtures of hydrogen and steam. |
| 8 | Review work with hydrogen/steam mixtures and design experiments to complete work with hydrogen and steam. |
| 9 | Review work with hydrogen/steam mixtures and design experiments with synthesis gas. |
| 10 | Review work with synthesis gas and design experiments to complete work with synthesis gas. |
| 11 | Review work with synthesis gas and design experiments with mixtures of synthesis gas and steam. |
| 12 | Review work with synthesis gas/steam mixtures and design experiments to complete work with synthesis gas and steam. |

In general, to be deemed a successful run, the equipment will have to have been operated over a sufficient length of time to bring the operation to a steady state; also, the shutdown must be voluntary; that is, much longer operating times must be technically possible. The quantities of feed materials must also be large enough so that serious error due to uncertainty of measurement will not result. In the turn-around processing, the equipment will be examined for evidence of coking or any other condition that might affect the operability of the equipment. Where deposition of tar on the interior surfaces of tubing downstream from the reactor is severe, it may be necessary to recover these materials by dissolving them in solvent and evaporating away the solvent to recover the material so deposited. This would be reported as a separate item in the material balance information.

2. Work Forecast

None. Task 1 has been completed.

B. Task 2. Build and Operate Bench-Scale Unit

1. Work Accomplished

a. Bench-Scale Unit Construction

All of the items required to construct the bench-scale unit have been ordered. The promised delivery times will permit us to stay on schedule.

We have elected to avoid the pulsating feed flow and the other difficulties associated with mechanical coal feeders, such as star feeders, by developing an aerated feeder. A Plexiglas model was constructed, whose configuration evolved to that shown in Figure 7. Its operation is described below. Our 5 to 15 lb/hr range has been obtained in this simulator and sufficient consistency in flow rate demonstrated. Some typical data are presented in Table 3. A high-pressure operational feed system will be built in stainless steel, based on the Plexiglas prototype.

1) Coal-Feeding System Operation

A cold-flow model of the solids feeding system (Figure 7) has been built and some preliminary tests made. In operation, the solids in the feed hopper move downward under the influence of gravity and a flow of gas introduced into the free space near the top of the hopper (Point 1) to replace the solids removed through the solids port. Additional gas is introduced near the bottom of the feed hopper through a hypodermic needle, which is aligned axially with the solids port. The solids flow rate is regulated principally through the control of the amounts of gases introduced at points 1 and 2.

Additional gas is introduced through a "Y" downstream from the solids port to regulate residence time in the coil and also to increase the gas velocity to prevent chocking and slugging. Some of the test results obtained are shown in Table 3. In these tests, the flow of solids from the coil exit was observed visually to be free of slugging and other gross irregularities in flow.

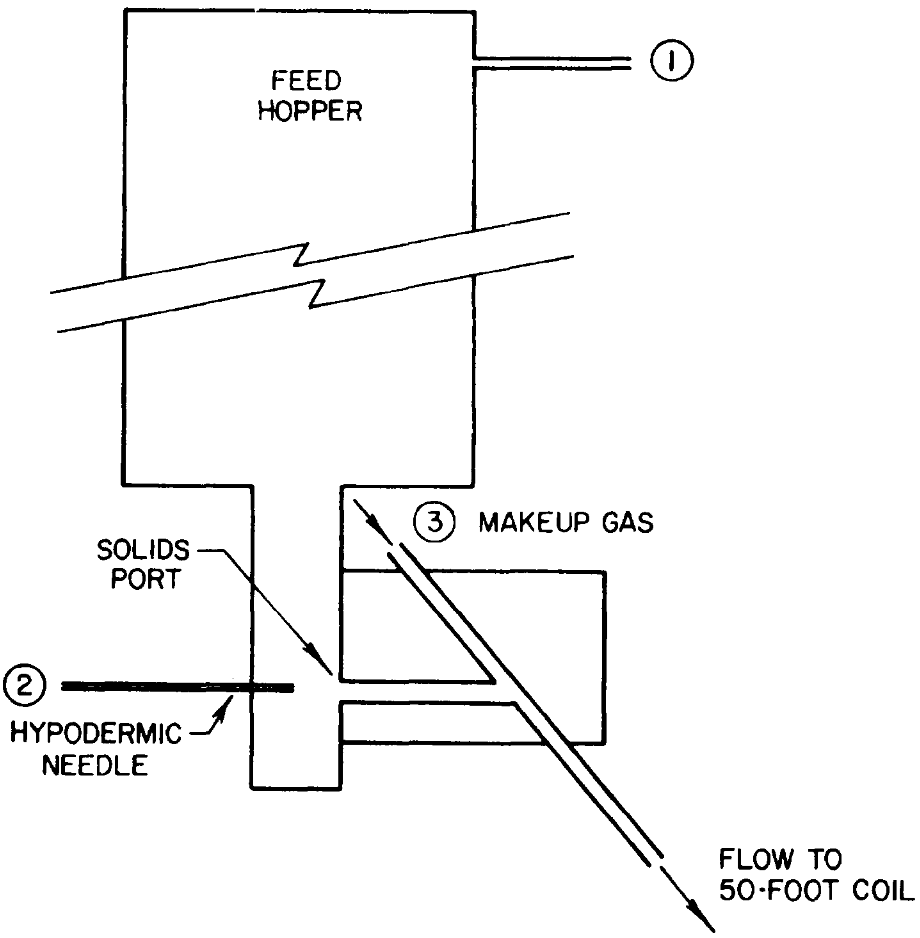


Figure 7. SKETCH OF COLD-FLOW MODEL OF COAL FEEDER

Table 3. PRELIMINARY DATA FROM COAL-FEEDING MODEL

| Particle Size Range | Superficial Velocity In Solids Port, ft/s | Superficial Velocity In Coil, ft/s | Coal/Feed Rate, lb/hr |
|---|---|---|---|
| 100 x 200 | 35 | 92 | 7.4 |
| 60 x 100 | 21 | 106 | 9.9 |
| 60 x 100 | 21 | 74 | 14.7 |

Work will be continued with the cold-flow model to study the effect of gas flow rates introduced at points 1, 2, and 3 on the solids flow rates observed at the exit of the 50-foot coil. Three particle size ranges will be used: 60x100, 100x200, and −325. A tentative design of the solids feeder to be used with the bench scale unit will be made from the results of these tests.

b. Coal Feedstock Procurement

The management of Project Lignite at the University of North Dakota has agreed to procure and send us a 2-ton shipment of North Dakota lignite for use as feedstock to the bench-scale unit.

The lignite will be from the North American Coal Company's Indianhead mine and, to the extent possible, it will be dried to 15-20% moisture, ground to −20 or −30 mesh and, without screening, loaded into plastic-lined drums for shipment.

From this material we hope to be able to screen out various size consists, such as 60x100, 100x200, etc.

2. Work Forecast

During the ensuing quarter we expect to build and test a high-pressure coal feeding system, and to completely assemble the bench-scale unit system, ready for shake-down tests.

## IV. CONCLUSIONS

The aerated coal feeding system can handle the range of feed rates required with sufficient consistency. A similar device should be built in steel for the bench-scale unit.

Gas residence time in the char catch-pot should be kept to a minimum to restrict continued reaction. For the same reason, the temperature at this point should be controlled as low as possible, but avoiding tars condensation.

Obtaining Incoloy tubing will not be a limiting factor in constructing the bench-scale unit.

# PATENT STATUS

The work performed during this period is not considered patentable.

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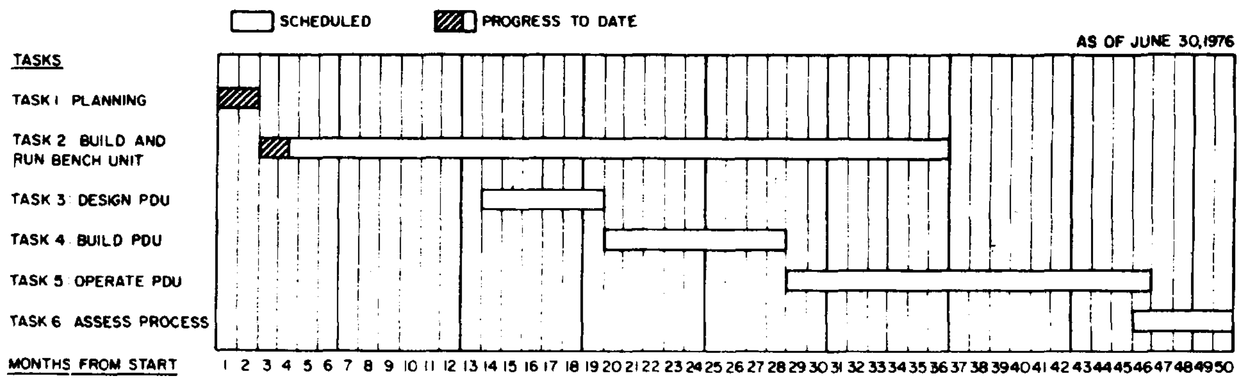
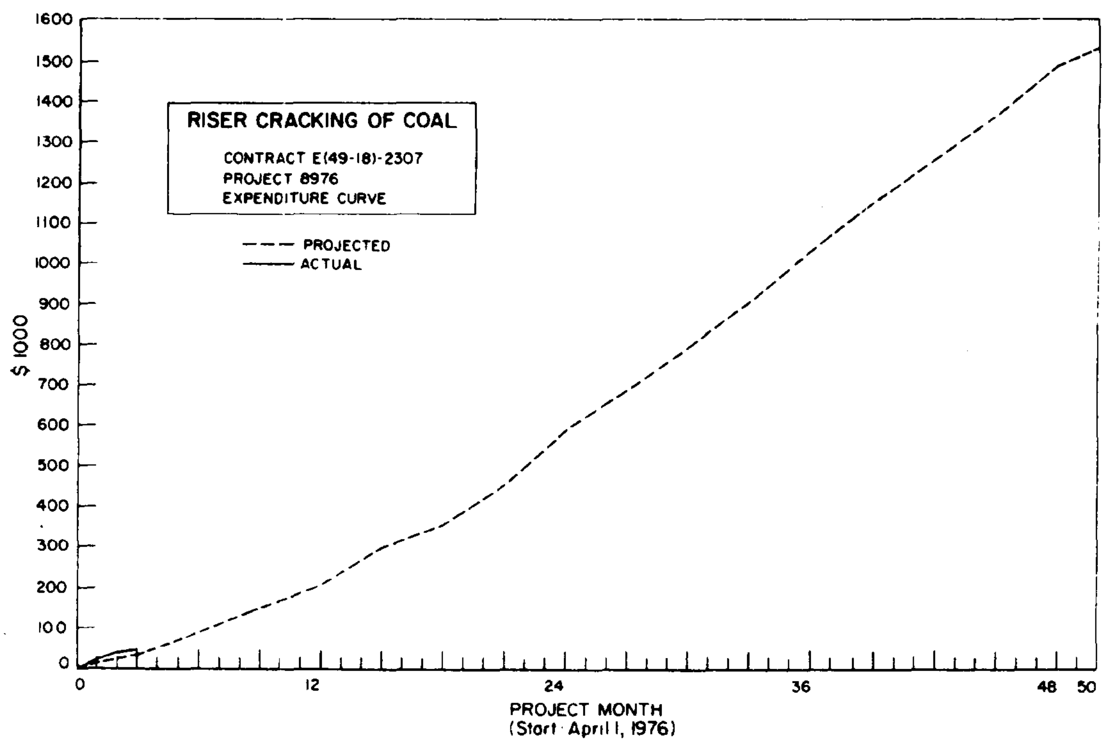
# APPENDIX A.

## Project Plan and Progress Report


SCHEDULED
PROGRESS TO DATE
AS OF JUNE 30,1976
TASKS
TASK 1 PLANNING
TASK 2 BUILD AND RUN BENCH UNIT
TASK 3 DESIGN PDU
TASK 4 BUILD PDU
TASK 5 OPERATE PDU
TASK 6 ASSESS PROCESS
MONTHS FROM START
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50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