BITUMINOUS COAL RESEARCH, INC.
SPONSORED RESEARCH PROGRAM

GAS GENERATOR RESEARCH AND DEVELOPMENT

Progress Report No. 6

(BCR Report L-457)

I. INTRODUCTION

This report summarizes progress achieved during the ninety-eighth month of work on the general program, "Gas Generator Research and Development," being conducted by Bituminous Coal Research, Inc., for the Office of Coal Research. The program which was initiated under Contract No. 14-01-0001-324 December 20, 1963, was transferred to Contract No. 14-32-0001-1207 on August 19, 1971. Thus, this report represents the sixth report of progress under the new prime Contract.

The overall objective of the program continues to be to develop processes for gasifying coal to produce fuel gas and high-Btu pipeline gas.

Laboratory-scale coal gasification experimentation is to be continued together with process and equipment development. With the aid of engineering subcontractor(s), a multipurpose research pilot plant facility is to be designed, constructed, and test operated.

A. Work Schedule

Work on the project is being conducted according to a schedule reflecting the program outlined under the new prime contract. This schedule was shown in Figure 1, page 2, Progress Report No. 1.

B. Monthly Progress Charts

Monthly progress charts reflecting proposed rate of effort and expenditures are shown in Appendixes A-1 and A-2.

II. PHASE II PROGRESS ACHIEVED DURING MONTH ENDING FEBRUARY 25, 1972

A. Laboratory-scale Process Studies

1. Coal Composition and Beneficiation Studies, and Laboratory Pyrolysis of Coal (R. G. Moses, R. D. Saltsman, and J. E. Noll): The summary report covering the work performed since September 1970 will be mailed during the week of March 6, 1972.

2. Fluidized-bed Gasification (E. K. Diehl and J. T. Stewart): Work on the two-phase project continued during the month of February with a proposed design for a three-stage fluidized-bed coal gasification PEDU being submitted to OCR. Laboratory investigations began on a fluidized-bed batch reactor.

a. Fluidized-bed PEDU: A meeting with OCR was held on February 16, 1972, to discuss the PEDU program. BCR went to the meeting to seek approval of the alternate PEDU design as discussed in last month's progress report. BCR proposed to add an additional gasifier in series to the gasifier described in the September 30, 1971, Koppers Specification and to add the necessary gas washing system. The purpose of this two-gasifier design was to simulate the first and second stages and, in turn, the second and third stages of the commercial gasification concept proposed by OCR during a January 20, 1972, visit to BCR.

The idea of using one or two gasifiers to simulate, in turn, each stage of a three-stage gasification process was deemed unsatisfactory by OCR. Further, the PEDU design presented by BCR did not accurately reflect the process currently envisioned by OCR as the one warranting study in the time frame under consideration. The estimated constructed cost of the PEDU, as presented, was judged by OCR to be in excess of the amount necessary to conduct the required research. Following this discussion, BCR asked for additional time to reexamine the PEDU concept.

With the aid of Koppers' engineers, a preliminary design based on the flow concept shown in Figure 95 was prepared and presented to OCR in a letter dated February 25, 1972. The following criteria were used as the design bases:

1. The program objective is to study the gasification of coal, with the only product being a clean, low-Btu fuel gas, rich in carbon monoxide.

2. Oxidizing gas is oxygen, with carbon dioxide as the moderator.

3. Nominal throughput is 100 pounds of coal per hour.

4. Gasification is conducted in series with regard to flow of coal and char, and with gas flow directed to transfer heat and destroy tars and oils.

5. Equipment and instrumentation are considered to be minimal for the conduct of an experimental program.

Item (5) above can be expanded to point out the following important observations:

1. In addition to the three fluidized-bed reactors, only those vessels considered absolutely essential for the operation of the unit are included.

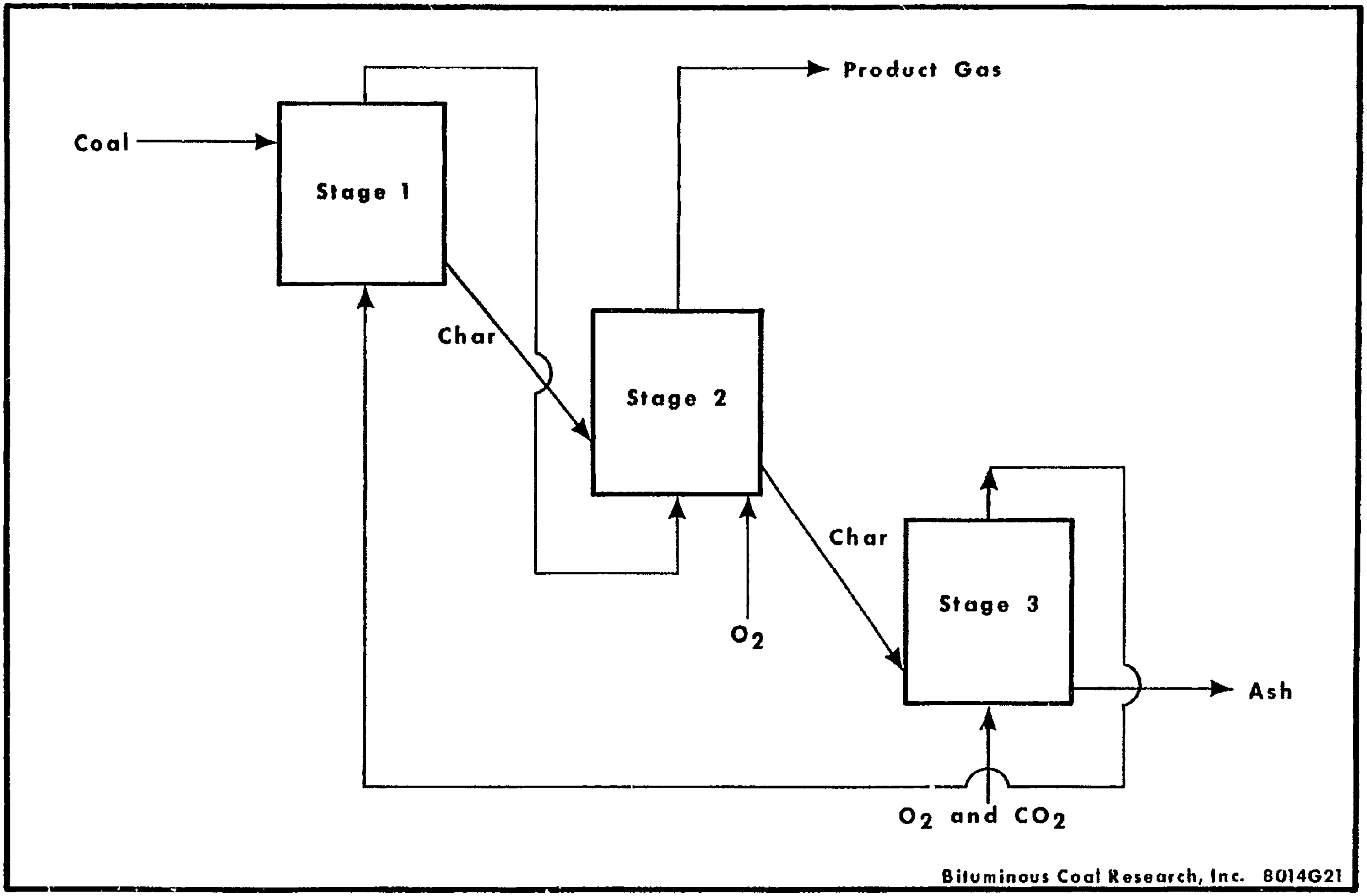

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Figure 95. Schematic Flow Diagram of Three-stage Fluidized-bed Coal Gasification Concept for Producing Low-Btu Fuel Gas

2. By excluding electric heating of the reactors, which for three reactors would represent an appreciable cost for the controls and equipment, experimental flexibility has been sacrificed. Generation of heat for the system is now solely determined by the carbon-oxygen reaction, and experimental altering of the oxygen-carbon ratio is limited.

3. While three-bed operation is anticipated, initial study will have to be centered on the Stage 1 reactor which will receive raw coal. Thus, it is necessary to supply a simulated Stage 3 gas to this vessel. Further, provision must be made for directing gas from this vessel through a cleanup system in order to meet local pollution control codes.

4. In keeping instrumentation to a minimum, capability to acquire and process data by means of the computer that would be available has been deleted.

The total constructed cost of the proposed PEDU is no less than the previously estimated cost for a single-bed unit to study char gasification. Nevertheless, it is believed that the present system provides the minimum amount of process equipment and instrumentation consistent with the objectives of the program. While gasifier design has been simplified, the inclusion of three gasifiers in the system requires additional piping, controls, and instrumentation.

b. Laboratory Investigations: The results of the char reactivity tests in the TGA were used to develop rate equations for the reaction of various chars with steam and carbon dioxide. To test the applicability of these derived rate equations to fluidized-bed reactor design, a laboratory-scale fluidized-bed batch reactor has been constructed.

Figure 96 is a photograph of the reactor and allied equipment. The reactor was constructed from a piece of 1-1/4-inch schedule 160, type 316, stainless steel pipe. The overall length is 20-1/4 inches, including a sight glass mounted on top of the reactor. Reactor pressure is set between 1 and 5 atmospheres and maintained by a Masoneilan Model 77-7 back-pressure regulator. Temperature is measured by means of a chromel-alumel thermocouple immersed in the bed. Temperatures up to 1100 C can be achieved by placing the reactor in the Marshall furnace that is part of the TGA apparatus described in earlier reports.

Twenty-gram batch samples of char are placed in the reactor and brought to temperature and pressure in an inert atmosphere. The reaction is then allowed to proceed at a controlled reacting gas partial pressure for some predetermined time. The reacting gas is then turned off and the char is subsequently weighed to determine the mass of carbon gasified. Enough data should be collected for discussion by the next report period.

c. Future Work: Additional design and detail engineering for the fluidized-bed PEDU will continue if approval is granted by OCR.

The laboratory-scale batch reactor studies will continue.

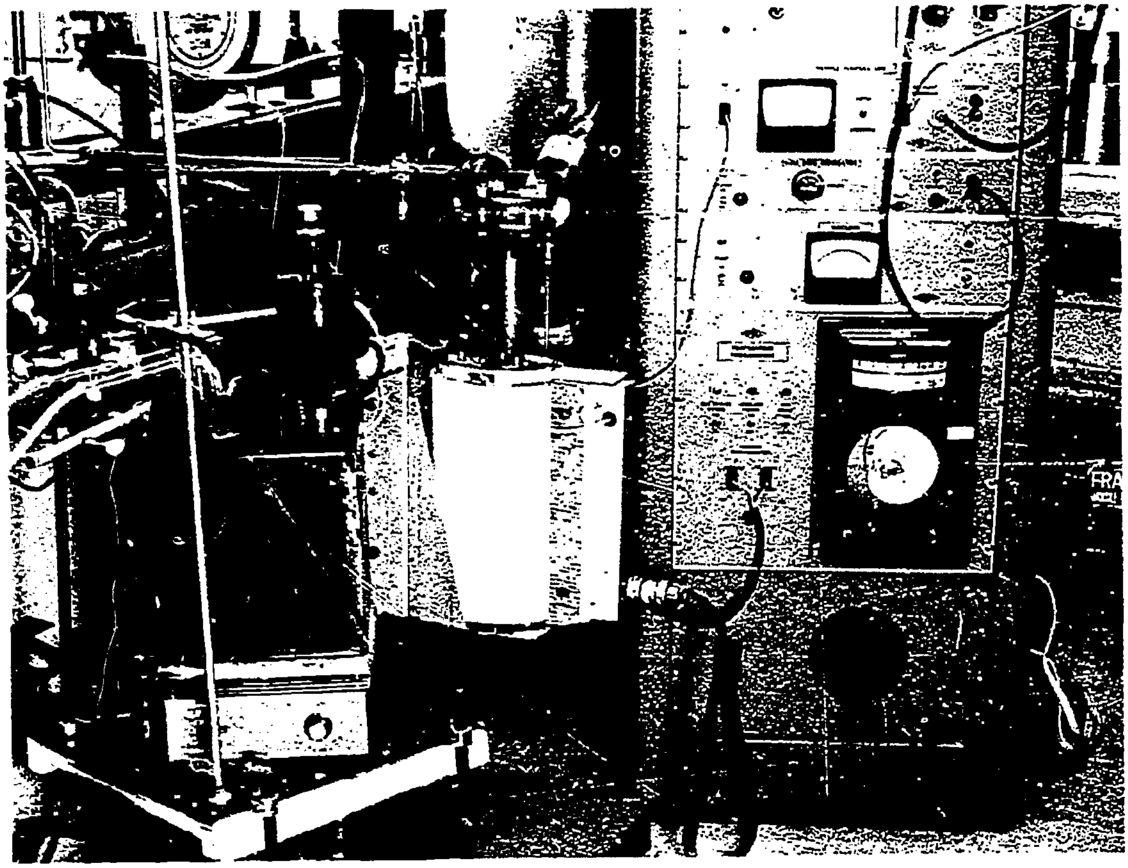

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Figure 96. Laboratory Scale Fluidized-bed Batch Reactor

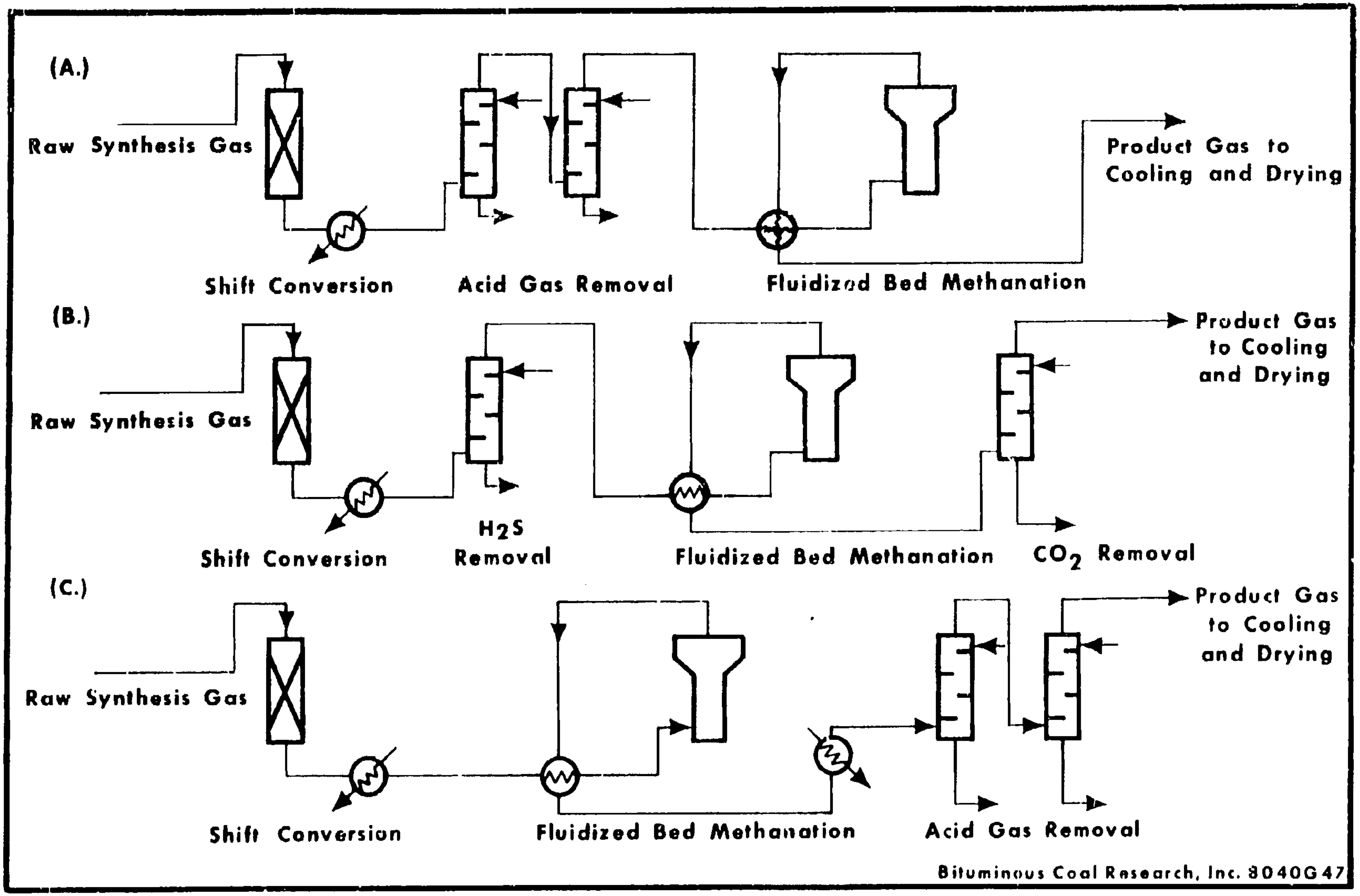

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Figure 97. BI-GAS Gas Processing Systems

3. Gas Processing (M. S. Graboski): Work continued in the area of gas processing methanation studies in accordance with the time schedule shown in Figure 82, Progress Report No. 5. This report summarizes progress achieved in the bench-scale and PEDU gas processing programs during the month of February.

a. Bench-scale Studies: The purpose of the bench-scale program is to investigate methanation catalysts under conditions imposed by the BI-GAS process. These include high carbon monoxide concentrations, high pressure, and a 3/1 hydrogen to carbon monoxide ratio.

Three processing schemes are currently under investigation. These are summarized in Figure 97. Scheme A reflects current planning where methanation follows shift conversion and acid gas removal; Scheme B considers hydrogen sulfide removal before and carbon dioxide removal after methanation; and Scheme C shows both hydrogen sulfide and carbon dioxide removal after methanation. Bench-scale tests are being conducted to determine which scheme is optimal for the BI-GAS process.

(1) BSM Tests: During the month, four BSM tests, Nos. 52 through 55, were conducted using Lot 2903 molybdenum oxide catalyst. The previous bench-scale tests had indicated activity and effect of temperature on reaction rates at high carbon monoxide partial pressures. Impetus has now been directed toward product distribution studies at high conversion levels in a fluidized-bed system.

The bench-scale apparatus used in these tests is shown in Figure 98. The current BSM system has been modified to handle a single pass, 1.25 inch ID fluidized-bed reactor. The reactor itself is equipped with two porous plate discs to contain the catalyst and is externally heated to supplement the gas preheat. While the methanation reaction is very exothermic, little problem with temperature control is present in the reactor. Thermocouples are located in the bed just above the distributor and at a bed depth of about 7 inches.

(a) BSM Test 52: The purpose of BSM Test 52 was to investigate carbon monoxide conversion over 2903 molybdenum oxide catalyst in a fluidized-bed unit under Case A (Figure 97) conditions. In Test 52, 73 grams of catalyst were charged to the unit and synthesis gas of the following composition was used:

Component	Volume, Percent
Hydrogen	61.62
Carbon monoxide	20.48
Methane	15.71
Nitrogen	2.11
Ethane	0.08

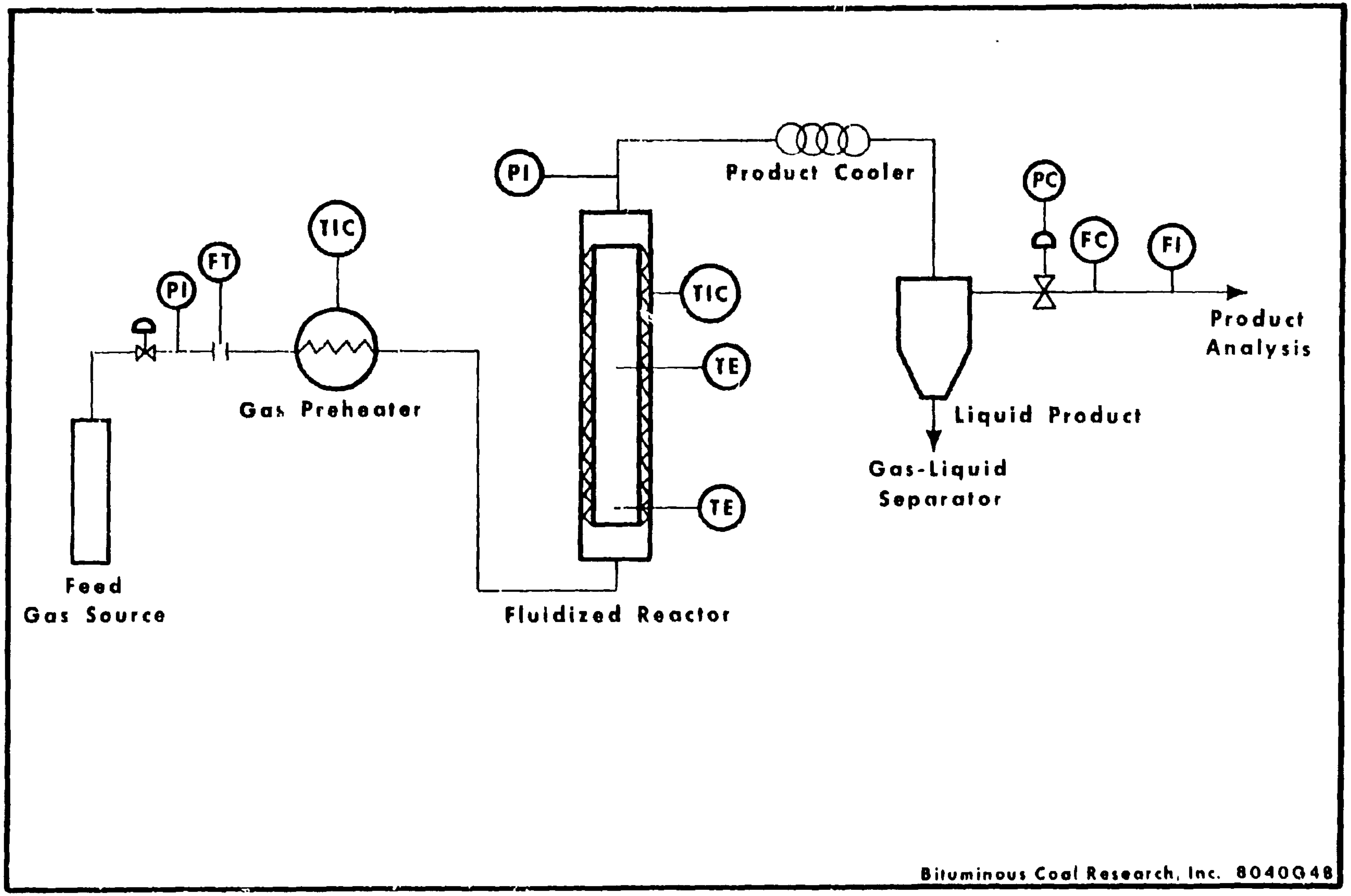


Figure 98. Bench-scale Fluidized-bed Methanation Apparatus

The test pressure selected was 1000 psig and test temperatures varied from 700 F to 1000 F.

Physical Properties of Catalyst: Physical property data for 2903 catalyst were measured in air. The results were:

Minimum fluidization velocity in air, U'_{mf} = 0.006 fps

Bulk density at minimum fluidization, ρ_B = 38 lb/cu ft

Thus for Test 52, the minimum fluidization velocity as scaled by the ERGUN equation was

$$U_{mf} = U'_{mf} \times \frac{\mu_{air}}{\mu_{gas}} = \frac{(0.006)(0.018)}{0.0125} = 0.004 \text{ fps}$$

Where μ = gas viscosity

U_{mf} = minimum fluidization velocity in synthesis gas

The unexpanded bed height for Test 52 was 15.0 cm.

Test Conditions and Notes: During Test 52, four product samples were collected representing three different temperatures. These were approximately 700 F, 800 F, and 900 F, respectively. However, difficulty was encountered in maintaining an isothermal reactor due to a lack of heat tracing at the distributor area of the reactor. Characteristically, the temperature difference ranged from 150 F to 220 F throughout the reactor. The system pressure throughout the test varied from 1000 to 1018 psig. Product flow rate was about 10 to 20 scfh, representing reactor velocities in the range of 0.01 to 0.03 fps.

Results for Test 52: The material balance data for Test 52 were computed based on normalized carbon balance and the outlet reactor flow rate. These data for Periods 1 through 4 are summarized in Table 74. Table 75 presents an overall summary of results for Test 52.

The data as analyzed in Table 75 show the effects of temperature on the product distribution, conversion, and shift ratio for Test 52. As the temperature is increased, the useful conversion of $(CO + H_2)$ increases rapidly, but the conversion to ethane is decreased. In each case, the shift constant is about 100 to 150 F lower in temperature than the reactor outlet.

(b) BSM Test 53: BSM Test 53 was conducted with 125 g of Lot 2903 molybdenum oxide catalyst at pressures and temperatures comparable to those used in Test 52 and under Case A conditions. A distributor area heater was installed to provide more isothermal conditions.

TABLE 74. DATA AND RESULTS FOR BSM TEST 52, PERIOD 1,
CONDUCTED AT 615 to 755 F AND 1000 PSIG
MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

Component	Feed mole percent	Feed g moles/hr	Product mole percent	Product g moles/hr
Carbon Monoxide	20.48	6.52	18.07	5.53
Carbon Dioxide		-	1.47	0.45
Hydrogen	61.62	19.61	60.18	18.41
Nitrogen	2.11	0.67	2.27	0.69
Methane	15.71	5.00	17.72	5.43
Ethane	0.08	0.02	0.25	0.08
Water				0.10
Total	100.00	31.82	100.00	30.69
Total Moles Carbon		11.56		11.57
Total Moles Hydrogen		59.34		59.22
Total Moles Oxygen		6.52		6.53

B. Conversion Data

	g moles/hr
CO → Products	0.99
H_2 → Products	1.20
Reactants → H_2O	0.22
CO → CH_4	0.43
CO → C_2H_6	0.12
CO → CO_2	0.45
α, Percent	8.2
β, Percent	12.2
K shift (experimental)	15.0
K shift (theoretical at outlet)	11.5

TABLE 74. DATA AND RESULTS FOR BSM TEST 52, PERIOD 2,
CONDUCTED AT 710 to 870 F AND 1018 PSIG
MOLYDBENUM OXIDE CATALYST NO. 2903

A. Material Balance

	Feed		Product	
Component	mole percent	g moles/hr	mole percent	g moles/hr
Carbon Monoxide	20.48	4.03	8.03	1.26
Carbon Dioxide			6.96	1.09
Hydrogen	61.62	12.11	52.62	8.25
Nitrogen	2.11	0.42	2.73	0.43
Methane	15.71	3.09	28.72	4.50
Ethane	0.08	0.02	0.94	0.15
Water				0.84 *
Total	100.00	19.67	100.00	15.68
Total Moles Carbon		7.16		7.15
Total Moles Hydrogen		36.70		37.08
Total Moles Oxygen		4.03		4.28

B. Conversion Data

			g moles/hr
CO	→	Products	2.77
H_2	→	Products	3.86
Reactants	→	H_2O	0.84
CO	→	CH_4	1.41
CO	→	C_2H_6	0.26
CO	→	CO_2	1.09
α, Percent			40.6
β, Percent			8.5
K shift (experimental) **			12.3
K shift (theoretical at outlet)			6.4

* Theoretical water is 0.58 g moles
** Theoretical water used for calculation

TABLE 74. DATA AND RESULTS FOR BSM TEST 52, PERIOD 3, CONDUCTED AT 780 to 1010 F AND 1012 PSIG MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

Component	Feed mole percent	Feed g moles/hr	Product mole percent	Product g moles/hr
Carbon Monoxide	20.48	5.62	1.71	0.27
Carbon Dioxide			7.13	1.12
Hydrogen	61.62	16.92	32.95	5.15
Nitrogen	2.11	0.58	3.73	0.58
Methane	15.71	4.31	54.00	8.45
Ethane	0.08	0.02	0.48	0.08
Water				3.11
Total	100.00	27.45	100.00	18.76
Total Moles Carbon		9.97		10.00
Total Moles Hydrogen		51.20		50.80
Total Moles Oxygen		5.62		5.62

B. Conversion Data

	g moles/hr
CO → Products	5.35
H_2 → Products	11.77
Reactants → H_2O	3.11
CO → CH_4	4.14
CO → C_2H_6	0.12
CO → CO_2	1.12
α, percent	75.3
β, percent	1.4
K shift (experimental)	6.9
K shift (theoretical at outlet)	3.6

TABLE 74. DATA AND RESULTS FOR BSM TEST 52, PERIOD 4, CONDUCTED AT 785 to 1005 F AND 1012 PSIG MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

Component	Feed mole percent	Feed g moles/hr	Product mole percent	Product g moles/hr
Carbon Monoxide	20.48	5.49	1.71	0.26
Carbon Dioxide			7.07	1.07
Hydrogen	61.62	16.51	32.58	4.95
Nitrogen	2.11	0.56	3.74	0.57
Methane	15.71	4.21	54.46	8.27
Ethane	0.08	0.02	0.44	0.07
Water				3.11
Total	100.00	26.79	100.00	18.30
Total Moles Carbon		9.70		9.74
Total Moles Hydrogen		49.98		49.62
Total Moles Oxygen		5.49		5.51

B. Conversion Data

	g moles/hr
CO → Products	5.23
H_2 → Products	11.56
Reactants → H_2O	3.11
CO → CH_4	4.06
CO → C_2H_6	0.10
CO → CO_2	1.07
α, percent	75.2
β, percent	1.2
K shift (experimental)	6.55
K shift (theoretical at outlet)	3.6

TABLE 75. SUMMARY OF RESULTS FOR BSM TEST 52. MOLYBDENUM OXIDE CATALYST NO. 2903

Period	Temp, F	Pressure, psig	Space Velocity[1]	U/U_{mf} Inlet	U/U_{mf} Outlet	α[2]	β[3]	K_s[4]	τ, sec[5]
1	610-755	1000	6300	6.5	7.0	8.2	12.2	15.0	24
2	710-870	1018	3900	4.3	4.0	40.6	8.5	12.3	37
3	780-1010	1012	5400	6.2	5.2	75.3	1.4	6.9	28
4	785-1005	1012	5300	6.2	5.0	75.2	1.2	6.6	28

[1] Standard volumes/volume catalyst/hr at inlet conditions

[2] α, useful conversion, 100 x (moles ($CO + H_2$) converted to hydrocarbons)/(total moles ($CO + H_2$) fed)

[3] β, selectivity, moles ethane produced/total moles hydrocarbons produced

[4] K_s, shift constant, $(P_{CO_2})(P_{H_2})/(P_{CO})(P_{H_2O})$ at outlet

[5] τ, residence time assuming 30% bed expansion and $U = (U_{in} + U_{out})/2$

In Test 53, the molybdenum oxide catalyst was fluidized with a synthesis gas of the following composition:

Component	Volume, Percent
Hydrogen	61.33
Carbon monoxide	20.64
Methane	15.84
Nitrogen	2.12
Ethane	0.07

The catalyst physical properties used for calculations were the same as those used in Test 52, namely, $U_{mf} = 0.004$ fps and $\rho_B =$ 38 lb/cu ft.

Test Conditions: Tests were made to attempt to reproduce Test 52 data in an isothermal reactor. To this end, with a system pressure of 1000 psig to 1018 psig, the temperature was varied from 870 F to 970 F. The low temperature point in Test 52 was not reinvestigated because of the low conversion achieved.

Results for Test 53: Material balance data for Test 53 were based on normalized carbon balance and the outlet flow rate. Data are summarized in Table 76. A summary of test results is given in Table 77.

Data for Test 53 show relatively high useful conversions over the molybdenum oxide catalyst. For Periods 1 through 3, gaseous hydrocarbon production accounted for 75 to 77 percent consumption of $(CO + H_2)$. Ethane selectivities were low due to the high temperature. A trace of propane was found in the product gas, and oil was apparent in the product water.

Fluidization velocities were fairly high in Test 53, and the system could possibly have slugged. Residence times for the gas were higher than for Test 52.

It appears from these test results that a pseudo equilibrium has been reached with respect to the hydrogenation reaction. The very high steam partial pressure at the reactor outlet in Test 53, 160 to 180 psi, has probably caused suppression of hydrogenation by the mechanism:

$$\Gamma_{Hydrogenation} = \frac{\Gamma_{waterfree}}{1 + K\left(P_{H_2O}\right)} \qquad (1)$$

Where Γ = reaction rate

K = constant

$\left(P_{H_2O}\right)$ = partial pressure of water

TABLE 76. DATA AND RESULTS FOR BSM TEST 53, PERIOD 1
CONDUCTED AT 870 F and 1010 PSIG
MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

	Feed		Product Gas	
Component	mole percent	g moles/hr	mole percent	g moles/hr
Carbon Monoxide	20.64	5.45	1.70	0.26
Carbon Dioxide			6.95	1.05
Hydrogen	61.33	16.19	32.75	4.94
Nitrogen	2.12	0.56	3.74	0.56
Methane	15.84	4.18	54.20	8.17
Ethane	0.07	0.02	0.66	0.10
Water				2.86
Total	100.00	26.40	100.00	17.94
Total Moles Carbon		9.67		9.68
Total Moles Hydrogen		49.22		48.88
Total Moles Oxygen		5.45		5.22

B. Conversion Data

	g moles/hr
CO → Products	5.19
H_2 → Products	11.25
Reactants → H_2O	2.86
CO → CH_4	3.99
CO → C_2H_6	0.16
CO → CO_2	1.05
α, Percent	76.9
β, Percent	2.0
K shift (experimental)	7.0
K shift (theoretical at outlet)	6.4

TABLE 76. DATA AND RESULTS FOR BSM TEST 53, PERIOD 2, CONDUCTED AT 875 F AND 1010 PSIG MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

Component	Feed mole percent	Feed g moles/hr	Product mole percent	Product g moles/hr
Carbon Monoxide	20.64	5.31	1.72	0.25
Carbon Dioxide			7.21	1.06
Hydrogen	61.33	15.79	33.06	4.88
Nitrogen	2.12	0.54	3.73	0.55
Methane	15.84	4.08	53.63	7.92
Ethane	0.07	0.02	0.65	0.10
Water				2.88
Total	100.00	25.74	100.00	17.64
Total Moles Carbon		9.43		9.43
Total Moles Hydrogen		48.02		47.80
Total Moles Oxygen		5.31		5.25

B. Conversion Data

	g moles/hr
CO → Products	5.06
H_2 → Products	10.91
Reactants → H_2O	2.88
CO → CH_4	3.84
CO → C_2H_6	0.16
CO → CO_2	1.06
α, Percent	75.4
β, Percent	2.1
K shift (experimental)	7.2
K shift (theoretical at outlet)	6.4

TABLE 76. DATA AND RESULTS FOR BSM TEST 53, PERIOD 3, CONDUCTED AT 970 F AND 1010 PSIG MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

Component	Feed		Product	
	mole percent	g moles/hr	mole percent	g moles/hr
Carbon Monoxide	20.64	5.22	2.09	0.29
Carbon Dioxide			6.32	0.89
Hydrogen	61.33	15.50	30.97	4.36
Nitrogen	2.12	0.54	3.89	0.55
Methane	15.84	4.00	56.16	7.91
Ethane	0.07	0.02	0.57	0.08
Water				3.06
Total	100.00	25.28	100.00	17.14
Total Moles Carbon		9.26		9.25
Total Moles Hydrogen		47.12		46.96
Total Moles Oxygen		5.22		5.13

B. Conversion Data

	g moles/hr
CO → Products	4.93
H_2 → Products	11.14
Reactants → H_2O	3.06
CO → CH_4	3.91
CO → C_2H_5	0.12
CO → CO_2	0.89
α, Percent	77.4
β, Percent	1.5
K shift (experimental)	4.4
K shift (theoretical at outlet)	4.2

TABLE 77. SUMMARY OF RESULTS FOR BSM TEST 53. MOLYBDENUM OXIDE CATALYST NO. 2903

Period	Temp, F	Pressure, psig	Space Velocity[1]	U/U_{mf} Inlet	U/U_{mf} Outlet	α[2]	β[3]	K_s[4]	τ, sec[5]
1	870	1010	3300	6.5	4.5	76.9	2.0	7.0	50.9
2	875	1010	2950	6.5	4.5	75.4	2.1	7.2	50.9
3	970	1010	2900	6.7	4.7	77.4	1.5	4.4	48.8

[1] Standard volumes/volume catalyst/hr at inlet conditions

[2] α, useful conversion, 100 x (moles ($CO + H_2$) converted to hydrocarbons)/(total moles ($CO + H_2$) fed)

[3] β, selectivity, moles ethane produced/total moles hydrocarbons produced

[4] K_s, shift constant, $(P_{CO_2})(P_{H_2})/(P_{CO})(P_{H_2O})$ at outlet

[5] τ, residence time assuming 30% bed expansion and $U = (U_{in} + U_{out})/2$

As the steam partial pressure increases, the hydrogenation rate falls. BSM tests indicated that this effect on cobalt-molybdenum catalysts reduces the activity by one half. The same trend on molybdenum oxide should be true, although to what extent is unknown.

Conversions throughout Test 53 compare well with Test 52 results even though Test 52 was operated at slightly shorter residence times and comparable exit shift constants. However, the approach to shift equilibrium was much closer in Test 53 than in Test 52. Comparison of results for the two tests is impossible because of the temperature gradient in Test 52.

(c) BSM Test 54: BSM Test 54 was conducted with 125 g of Lot 2903 molybdenum oxide catalyst. The purpose of the study was to determine conversion with a simulated 1/1 recycle gas in a small (1.25-inch) fluidized reactor system operated under Case A conditions. The apparatus used was described in detail in sections reporting Tests 52 and 53.

In Test 54, the following gas feed was used:

Component	Volume, Percent
Hydrogen	31.31
Carbon monoxide	10.27
Methane	53.94
Nitrogen	4.36
Ethane	0.12

Catalyst 2903 has a minimum fluidization velocity of 0.004 fps at reactor conditions and a poured bulk density of 38 lb/cu ft.

Test Conditions: Tests were made in the isothermal reactor bed at 865 F and 700 and 1000 psig. In all cases, the velocity of the gas was held constant; the space velocity was thus lower in the 700-pound pressure test run. Reactor gas velocity was in the range of 3 to 4.5 times minimum fluidizing velocity.

Results for Test 54: Material balance data for Test 54 are summarized in Table 78. These results are based on the outlet flow rate and a normalized carbon balance.

Data for Periods 1 through 4 show a definite trend with respect to error in the hydrogen balance. As the sample period number increases, the hydrogen imbalance decreases. In Period 3, the oxygen balance is slightly high indicating that the product water weight is in error. Review of the reactor system showed two areas where hydrogen could accumulate and thus mix with the feed gas by diffusion, resulting in a variable composition feed with respect to time. Thus the total balances were altered to reflect additional hydrogen in the feed gas at the sampling times. This was not a problem in Tests 52 and 53 where the feed gas was lower in molecular weight and contained 60 percent hydrogen.

TABLE 78. DATA AND RESULTS FOR BSM TEST 54, PERIOD 1, CONDUCTED AT 865 F AND 1015 PSIG MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

Component	Feed mole percent	Feed g moles/hr	Product mole percent	Product g moles/hr
Carbon Monoxide	10.27	1.73	0.83	0.12
Carbon Dioxide		0.00	2.39	0.33
Hydrogen	31.31	5.28	18.95	2.65
Nitrogen	4.36	0.73	5.36	0.75
Methane	53.94	9.09	70.91	9.91
Ethane	0.12	0.02	1.06	0.15
Propane	0.00	0.00	0.50	0.07
Water		0.00		1.01
Total	100.00	16.85	100.00	13.98
Total Moles Carbon		10.86		10.87
Total Moles Hydrogen		47.04		48.42
Total Moles Oxygen		1.73		1.79

B. Conversion Data

	Raw Data g moles/hr	H_2 Normalized g moles/hr
CO → Products	1.61	1.61
H_2 → Products	2.63	3.32
Reactants → H_2O	1.01	1.01
CO → CH_4	0.82	0.82
CO → C_2H_6	0.26	0.26
CO → C_3H_8	0.21	0.21
CO → CO_2	0.33	0.33
α, Percent	69.8	63.5
β, Percent	19.6	19.6
K shift (experimental)	6.6	6.6
K shift (theoretical at outlet)	7.2	7.2

TABLE 78. DATA AND RESULTS FOR BSM TEST 54, PERIOD 2,
CONDUCTED AT 865 F AND 1015 PSIG
MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

Component	Feed mole percent	Feed g moles/hr	Product mole percent	Product g moles/hr
Carbon Monoxide	10.27	1.93	0.97	0.15
Carbon Dioxide	0.00		2.57	0.40
Hydrogen	31.31	5.89	18.39	2.85
Nitrogen	4.36	0.82	5.39	0.84
Methane	53.94	10.15	71.14	11.03
Ethane	0.12	0.02	1.04	0.16
Propane	0.00		0.50	0.08
Water	0.00		0.00	1.01
Total	100.00	18.81	100.00	16.52
Total Moles Carbon		12.12		12.14
Total Moles Hydrogen		52.50		53.44
Total Moles Oxygen		1.93		1.96

B. Conversion Data

	Raw Date g moles/hr	H_2 Normalized g moles/hr
CO → Products	1.78	1.78
H_2 → Products	3.04	3.51
Reactants → H_2O	1.01	1.01
CO → CH_4	0.88	0.88
CO → C_2H_6	0.28	0.28
CO → C_3H_8	0.24	0.24
CO → CO_2	0.40	0.40
α, Percent	67.7	63.8
η, Percent	20.0	20.0
K shift (experimental)	6.6	6.6
K shift (theoretical at outlet)	7.5	7.5

TABLE 78. DATA AND RESULTS FOR BSM TEST 54, PERIOD 3, CONDUCTED AT 865 F AND 1015 PSIG MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

	Feed		Product	
Component	mole percent	g moles/hr	mole percent	g moles/hr
Carbon Monoxide	10.27	1.86	1.05	0.16
Carbon Dioxide	0.00	0.00	2.69	0.40
Hydrogen	31.31	5.67	17.71	2.62
Nitrogen	4.36	0.79	5.38	0.80
Methane	53.94	9.76	71.65	10.59
Ethane	0.12	0.02	1.02	0.15
Propane	0.00	0.00	0.50	0.07
Water	0.00	0.00	0.00	1.01
Total	100.00	18.10	100.00	15.80
Total Moles Carbon		11.66		11.66
Total Moles Hydrogen		50.50		51.08
Total Moles Oxygen		1.86		1.97

B. Conversion Data

	Raw Data g moles/hr	H_2 Normalized g moles/hr
CO → Products	1.70	1.70
H_2 → Products	3.05	3.34
Reactants → H_2O	1.01	1.01
CO → CH_4	0.83	0.83
CO → C_2H_6	0.26	0.26
CO → C_3H_8	0.21	0.21
CO → CO_2	0.40	0.40
α, Percent	65.5	63.1
β, Percent	19.4	19.4
K shift (experimental)	6.6	
K shift (theoretical at outlet)	7.2	

TABLE 78. DATA AND RESULTS FOR BSM TEST 54, PERIOD 1, CONDUCTED AT 865 F AND 705 PSIG MOLYBDENUM OXIDE CATALYST NO. 2903

A. Material Balance

	Feed		Product	
Component	mole percent	g moles/hr	mole percent	g moles/hr
Carbon Monoxide	10.27	1.37	1.11	0.12
Carbon Dioxide			2.77	0.30
Hydrogen	31.31	4.17	17.86	1.95
Nitrogen	4.36	0.58	5.38	0.59
Methane	53.94	7.19	71.39	7.79
Ethane	0.12	0.02	0.99	0.11
Propane			0.50	0.05
Water	0.00			0.84
Total	100.00	13.33	100.00	11.75
Total Moles Carbon		8.60		8.58
Total Moles Hydrogen		37.22		37.80
Total Moles Oxygen		1.37		1.56

B. Conversion Data

			g moles/hr
CO	→	Products	1.25
H_2	→	Products	2.22
Reactants	→	H_2O	0.84
CO	→	CH_4	0.60
CO	→	C_2H_6	0.18
CO	→	C_3H_8	0.15
CO	→	CO_2	0.30
α, Percent			63.8
β, Percent			19.0
K shift (experimental)			6.6
K shift (theoretical at outlet)			7.8

The data for Test 54 are summarized in Table 79. Results for Test 54 indicate significant conversion to ethane and propane even at the relatively high temperature of 865 F. The useful conversion and selectivity remained constant over the four periods. The decrease in pressure during Period 4 was compensated for by increased residence time resulting in the same conversion percentage.

The significant propane conversion is interesting. At comparable temperatures and a 20 percent carbon monoxide feed, much less propane was apparent. The longer gas residence time and higher methane partial pressures could explain this fact. In any event, higher hydrocarbons are desirable since they increase the quality of the product gas.

The useful conversion of Test 54 is lower than for Tests 52 and 53. This may be attributed to several reasons. First, the Test 53 reactor was probably slugging due to the higher U/U_{mf} ratio. In a slugging system, all gas sees some catalyst and hence, higher conversion is achieved. A second possible reason is the fact that the carbon monoxide partial pressure in Test 54 was lower than in Test 53. While the reaction is zero order at high partial pressures of carbon monoxide, it becomes first order at low pressures. Thus, the Test 54 gas might have been in the first order region for a longer period than that for Test 53, and conversion could thus have been reduced.

(d) BSM Test 55: Data reduction for Test 55 is not completed. Results will be reported next month.

(2) Catalyst Life Tests: During the past month and a half, life testing of BCR Lot 2904, chromic oxide methanation catalyst, has been conducted. Results for this study, as currently available, are reported here. In addition, a new life tester has been designed and is currently in the construction stage. This unit improves the flexibility of the life test system by providing the ability to test four catalysts simultaneously.

Test 2904: Catalyst Life Test 2904 has been underway for 800 hours. The conversion data during this time are indicated in Figure 99. It is readily evident that the useful conversion, α, over the chrome catalyst achieved a nearly stable value after about 200 hours of operation. Slight deactivation may have been occurring after that time; however, the conversion data to 450 hours do not indicate this as a definite trend. At about 500 hours, a power failure occurred. The duration was such that the catalyst was subjected, under synthesis gas flow, to a slow cooling and shock heating cycle. It is almost certain that the temperature fell close to ambient before power was regained. Since the temperature controller is not on a programmed heating cycle, the bed would thus have been returned to 800 F rapidly (15 minutes to 30 minutes).

Catalyst activity was reduced by the cycle, but conversion was reestablished after several hundred more hours of operation. A possible mechanism for this occurrence is the formation of wax or heavy oil at colder temperatures during the power failure, resulting in the plugging of catalyst pores. After sufficient time under high temperature and hydrogen, the wax or

TABLE 79. SUMMARY OF RESULTS FOR BSM TEST 54. MOLYBDENUM OXIDE CATALYST NO. 2903

Period	Temp, F	Pressure, psig	Space Velocity[1]	U/U_{mf} Inlet	U/U_{mf} Outlet	α[2]	β[3]	K_s[4]	τ, sec[5]
1	865	1015	4830	4.4	3.5	63.5	19.6	7.2	70
2	865	1015	4400	4.8	4.1	63.8	20.0	7.5	64
3	865	1015	4620	4.6	3.9	63.1	19.4	7.2	66
4	865	705	6360	3.3	2.9	63.8	19.0	7.8	90

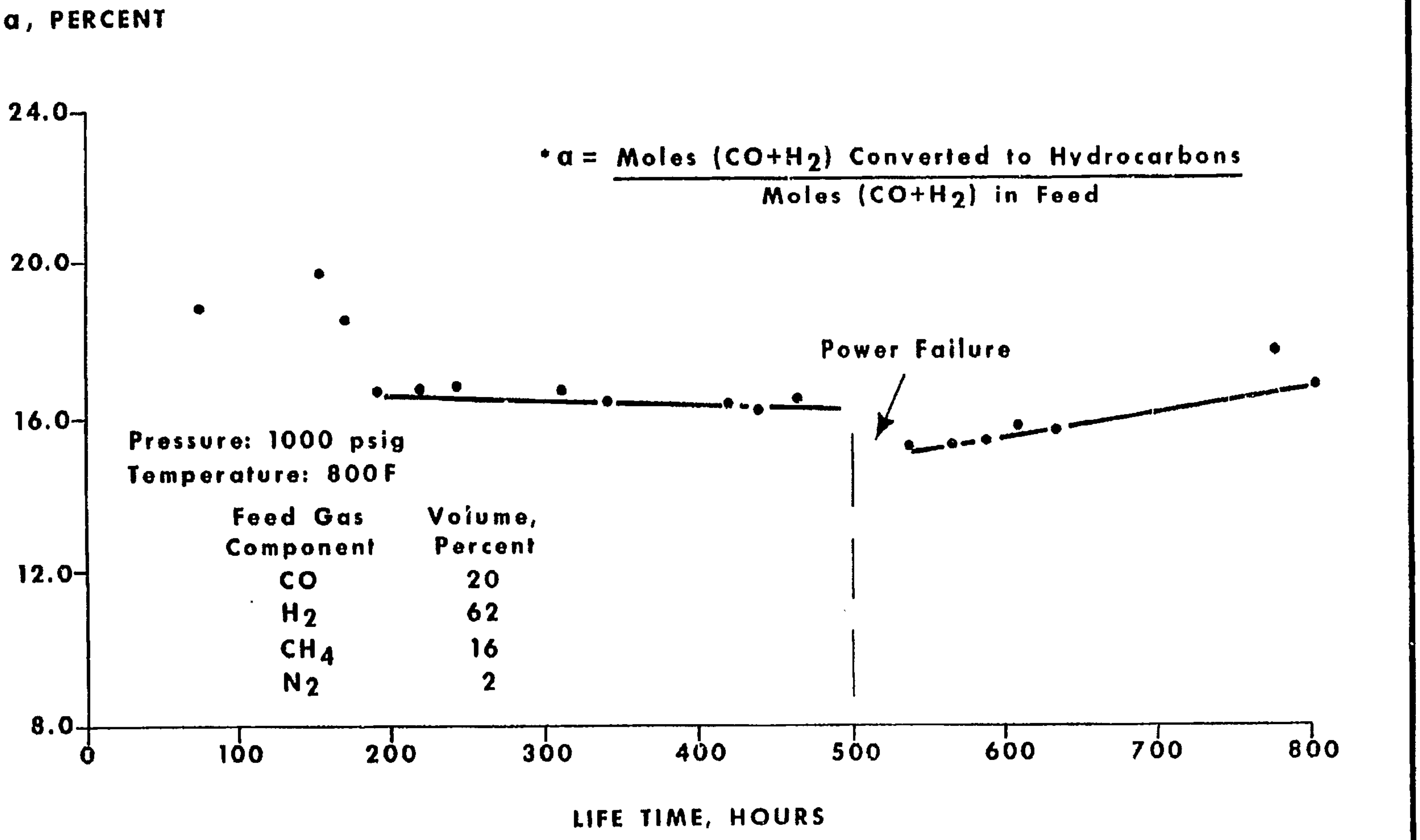
[1] Standard volumes/volume catalyst/hr at inlet conditions

[2] α, useful conversion, 100 x (moles ($CO + H_2$) converted to hydrocarbons)/(total moles ($CO + H_2$) fed)

[3] β, selectivity, moles ethane produced/total moles hydrocarbons produced

[4] K_s, shift constant, $(P_{CO_2})(P_{H_2})/(P_{CO})(P_{H_2O})$ at outlet

[5] τ, residence time assuming 30% bed expansion and $U = (U_{in} + U_{out})/2$



Bituminous Coal Research, Inc. 8040G49

Figure 99. Extended Activity Data for Life Test 2904 Using Chromic Oxide on Alumina Catalyst (BCR Lot 2904)

oil was decomposed again, exposing the entire surface for reaction. The lack of effect of temperature on catalyst stability is not unexpected since chromic oxide is used as a high temperature reforming catalyst. Even at 2700 F, rapid crystalline growth and sintering are not problems. Chromic oxide is also very stable in the presence of hydrogen. Equilibrium calculations indicate the chromic oxide will not be chemically reduced at 800 F.

Product distribution over chromic oxide shows methane, ethane, and carbon dioxide as main products. Since the test is not being operated at high conversions, actual product distributions are not indicative of what might be obtained for higher alpha (α) values. The catalyst does promote the shift reaction readily as do all the other hydrogenation catalysts studied to date.

New Life Test Unit: Parts have been ordered to permit the construction of a new life tester containing four reactors. These will be constructed of 0.08-inch ID stainless steel tubing and the overall design will be the same as the current unit. All four reactors, however, will operate from the same feed system and a common heater-controller system. A sketch of the unit is presented in Figure 100. Mass transfer calculations indicate that the system will be kinetically controlled as is the current reactor system. The unit should be completed in about a month.

b. PEDU Studies: Significant progress was achieved on the methanation PEDU during the month of February.

(1) Design Engineering: The basic design of the PEDU system has been completed for some time. Recently, the system concept has been modified to handle feed gases containing not only the main synthesis gas components, carbon monoxide, hydrogen, and methane, but also the addition of carbon dioxide and hydrogen sulfide so as to permit operation under process schemes shown as Cases A, B, and C of Figure 97. The pertinent material balance data have been supplied by Koppers in the form of drawings 2415-2A27 and 28. Since the issuing of the P & I drawings for the PEDU, several alterations have been made in the system. These include addition of hydrogen sulfide and carbon dioxide (4-ton) systems, more detailed alarm and annunciator systems, addition of another CO-alarm safety system for the cubical area, and addition of nitrogen purge system. Koppers is making pencil changes in these drawings and will not reissue them until they are so notified.

The location of the hydrogen sulfide and carbon dioxide storage tanks has been finalized. These will be located in the northeast corner of Building 3 across the road from the nitrogen and oxygen systems. The tanks are to be completely fenced in, and any controls for the hydrogen sulfide system will be located in a locked compartment.

Koppers is designing a hydrogen sulfide removal system for the PEDU to be included in the system ahead of the thermal oxidizer.

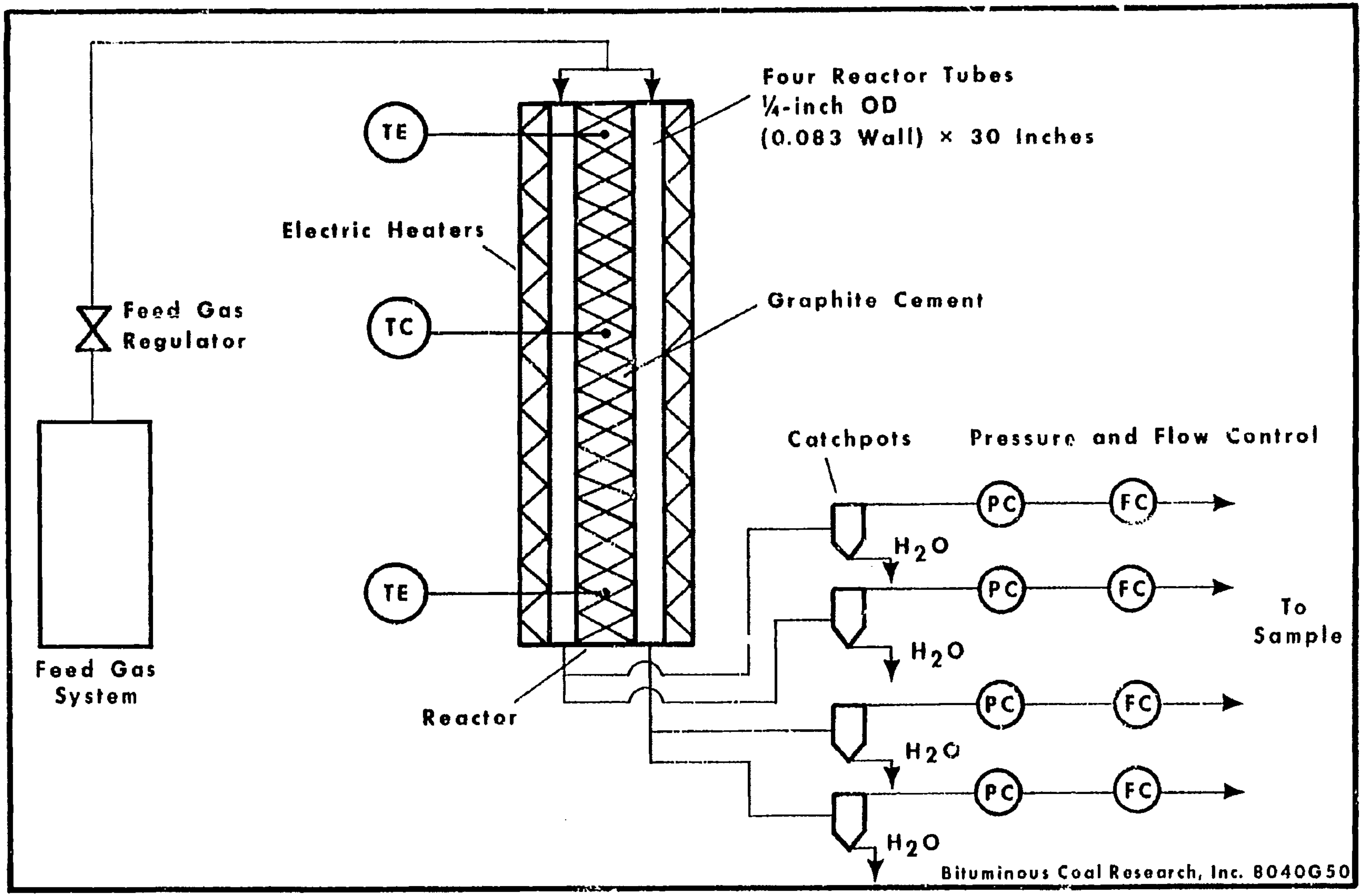


Figure 100. Revised Life Test System (2-72)

(2) Detail Engineering: To permit the solicitating of quotes on PEDU items, Koppers has had to perform some detail engineering on vessels and equipment. Currently, they are on interim funding until final approval of the detail engineering proposal and schedule is given.

In the area of vessel and equipment detailing, Koppers has provided services in reviewing specifications for bidding purposes and supplying necessary sketches to be included with quotes. The following drawings have been received for quotation purposes:

2415-9A701 and 702	Panel board design and specifications
2415-2A700 and 2A714	Methanator detail drawings. (On this vessel, BCR altered the design temperature to 1050 F to provide safety margin.)
2415-2A722	Demister detail drawing
2415-2A721	Cooling water hold tank detail drawing
2415-2A724	Water metering tank detail drawing
2415-2A723	Filter blowback heater detail drawing (Holding for further review)

Other drawings are being prepared by Koppers.

During the past several weeks, Koppers has been developing a detail engineering program for the methanation PEDU. A preliminary schedule and cost estimate was provided to BCR on February 11, 1972. This estimate is being reviewed and Koppers is currently reducing the manday requirements for the job based on BCR's recommendations.

(3) Status of Quotations: Bids on a number of items have been requested. The remainder of the items will be handled as soon as drawings and specifications become available from Koppers.

Quotations on the following items have been received and are being reviewed:

MX 720	Steam Boiler
ME 700	Water Cooler
MP 710	Cooling Water Pump
MX 750	Demineralizer
MX 500	Therminol System
ME 405 A & B	Feed Gas Heaters

(4) Other Work

(a) Building Permits: Required forms are being prepared.

(b) Air Pollution Permit: Preliminary contacts have been made, but more cannot be done until a final design and quotation for the thermal oxidizer system are available.

c. Cold Model Studies: Data were reported last month for several 6-inch cold model tests using a sand-freon system. While data from these studies have been encouraging, it has been apparent that the fluidization properties of the sand mix used are such that extreme channeling occurs in the bed. Therefore, cold model tests have been temporarily postponed until a batch of Harshaw (BCR Lot 2903) molybdenum oxide catalyst can be obtained. Upon receiving the catalyst, cold model tests will be reinitiated.

d. Other Work: BCR Suggestions 193, 194, (M. S. Graboski), and 196 (E. E. Donath), have been reviewed by the BCR attorney. He has recommended that these be combined and a single invention disclosure be prepared covering all three. On this basis, a report will be submitted to OCR in the near future.

e. Future Work: The following work will be conducted according to the gas processing schedule during the coming month:

At the bench-scale level, fluidization tests of Lot 2904 chromic oxide catalyst will be conducted under Case A and Case B conditions. Life testing of chromic oxide will continue, and the new life test will be constructed.

PEDU work will continue. Bids on equipment will be solicited and evaluated; approval to purchase items will be requested from CCR. Koppers will continue detail engineering; a final cost and scope of work will be determined. Continued emphasis will be placed on acquisition of the necessary permits for construction.

Model studies will be completed pending the early arrival of the Lot 2903 catalyst.

4. Analytical Services (J. E. Noll): During the past month, 45 samples were analyzed by gas chromatography. The types of analyses requested were as follows:

Type of Analysis Requested	No. of Samples Analyzed
Gas Chromatography	
Methanation Unit Gas Samples	45
TOTAL	45

5. *Gas Chromatographic Procedures (J. E. Noll)*: No work was required in this area during the past month.

Future Work: Performing sample analyses is the only work planned.

B. Stage 2 Process and Equipment Development Unit--100 lb/hr (R. J. Grace, E. E. Donath, and R. L. Zahradnik)

During the month, time was distributed between attendance at the Subcommittee Work Group for the AGA Materials Evaluation program, answering requests from bidders for clarification of various items in the MPRF bid package, discussing possible application of mathematical modeling to our cold model experiments as suggested by representatives of United Aircraft Research Laboratories and Dynamic Sciences Inc., completion of supervision of dismantling operations of the Stage 2 100 lb/hr PEDU, and preparation of the final summary report, Phase II, Process and Equipment Development.

1. Dismantling of the 100 lb/hr PEDU: Dismantling of the 100 lb/hr PEDU equipment was essentially completed during the month of February. However, disposition or relocation of the four major items located in the safety cubicle, namely the reactor, scrubber, scrubber cooler, and steam superheater, requires an immediate decision in order to clear the area for the new PEDUs.

2. Inspection of Stage 1 Cooling Coils and Refractories: As discussed in Section B, Item 1, page 276 of Progress Report No. 5, portions of the refractories and a section of the Stage 1 cooling coil were delivered to the Koppers Company for inspection and analysis. Kopper Company letter report of February 7, 1972 on the cooling coil is summarized as follows:

> A uniform rusting and scale, visible on the outer surface of the carbon-steel cooling coil, was probably the corrosion that occurred when the wet refractory cement was applied during fabrication. No water-side corrosion or scale was observed.
>
> Metallographic specimens were prepared representing (1) the metal nearest the outer flange, (2) the midwall area, and (3) the point nearest the reactor center line. The structures were that of annealed, low-carbon steel. Macroetching indicated seamless pipe.
>
> No evidence of carburization oxidation or excessive temperature excursions was observed. Hardness values of 53/58 Rockwell B were measured which are well below the maximum RB 77 permitted by ASTM A 192 for carbon-steel boiler tubes.
>
> Based on these observations it was concluded that the cooling coil was fabricated from seamless, low-carbon steel boiler tube and that under the conditions of service in the PEDU the tubes were satisfactory.
>
> The refractory samples were submitted to Mr. D. M. Mitsak for examination by others. Results are not available at this time.

3. Status of Phase II Summary Report: Typing of the first rough draft of the Phase II Summary Report has been completed. Editing and assembly of graphs and figures is progressing. The final draft should be completed early in March.

4. Future Work Future work will include:

a. Report of Koppers Company findings relative to inspection of Stage 1 refractories.

b. Allocation of surplus items from 100 lb/hr PEDU.

c. Completion of final draft of Phase II Summary Report.

C. Cold Flow Model Experiments--5 ton/hr Two-stage Gasifier (R. J. Grace, J. E. Noll, R. D. Harris, R. L. Zahradnik, and E. E. Donath)

Work during the month consisted of modifying the cold flow model to allow operation of the Stage 1 section of the model at higher air rates, which simulate the expected pilot plant characteristics near the reactor walls.

The model studies to be carried out in the following months are expected to indicate both the location and size of nozzles and the shape of the reactor which will avoid excessive localizing of temperatures near the reactor walls. Stages 1 and 2 of the gasifier are to be studied independently at first, followed by tests of the two stages together.

The Stage 1 studies have been divided into three phases, now underway: (I) single-burner tests, (II) multiple-burner tests, and (III) multiple-burner tests plus simulated slag. The experimental requirements for the third phase were considered to be more difficult than those for the other phases. Provisional Phase III work was therefore being undertaken to solve some of the experimental problems so that Phase III data can be obtained immediately following Phase II tests.

The Phase II and III studies for Stage 1 of the two-stage gasifier were proceeding as planned and satisfactorily when it appeared expedient to review some of the assumptions on steam and char flow rates.

The model constructed to date consists of the shell for Stage 1 and the lower part of Stage 2 and a set of burners for Stage 1. The burners are socket mounted so that they can be aimed horizontally and at the center line and can be pointed downwards and to the right of the center line.

1. Initial Model Tests: The Stage 1 model was first operated at air rates corresponding to the feed rates (cu ft/sec) of the steam and oxygen to the pilot plant. These tests were conducted principally to develop the techniques required for Phase III. For these tests, a single 200 scfm blower was used. The test set-up was shown in Figure 62, page 199, of Progress Report No. 3. Tests were conducted using 100 to 200 scfm air with fine plastic tracers, coarse plastic flow indicators, and glycerol spray from a compressed air atomizer. The tests indicated that rotation of the air in the 4 ft high-2 ft diameter chamber could be induced by aiming the burners horizontally and 1-1/2 inches from the center line. The air stream from the three burners appeared to mix readily at the center and heavier particulates readily migrated to the periphery. Tests with a glycerol spray from a nozzle selected to produce 25 micron droplets indicated that the droplets were quickly collected as a film on the interior surface.

This film readily drained into the simulated slag collecting vessel and most of 1/2 gallon of spray material was recovered.

These provisional tests indicated:

(1) The burners should be aimed horizontally.

(2) The streams from the three burners blended and produced rotation as desired.

(3) Fine droplets were readily deentrained and made to coalesce on the surfaces.

(It should be pointed out that the effects of a horizontal burner alignment in the model would be obtained in the pilot plant with a burner aimed down to compensate for the buoyancy of the hotter reaction products.)

The tests described above were principally for the development of experimental techniques. More detailed investigation was interrupted by the investigation of the properties of the FEDU chars. During the course of these investigations, the model test program was reviewed and it was concluded that two important aspects of the operation of Stage 1 needed to be investigated.

(1) The proper mixing of particles to enable adequate reactions.

(2) The possibility of severe slag build-ups near the entrance to Stage 2.

It was decided to defer consideration of particulate mixing since the size of the char to be fed to Stage 1 was uncertain. It was also decided that the tangential forces that satisfactorily deentrained the glycerol droplets in the preliminary tests were lower than those expected in the pilot plant. It was therefore concluded that the slag droplet produced in the pilot plant would be readily deentrained. However, the lateral and upward forces on the simulated slag layer were also lower than those expected in the pilot plant. It was, therefore, concluded that it would not be realistic to assume that the slag layer in the pilot plant could be drained effectively without such problems as a film being induced upwards along the crown and thereby being reentrained in the outlet gas stream or freezing at the cooler throat.

2. Further Model Tests

a. Modifications to the Equipment: Calculations indicated that the most severe conditions for entraining slag droplets and inhibiting slag drainage could be simulated by operating at 1,600 scfm to simulate the momentum at the burner exits and at 1,150 scfm to simulate particulate lifting velocity. These rates could not be supplied by available blowers so plans were made to operate at 575 scfm to simulate the Stage 1 lifting velocity at 50 percent of rating.

These air rates were supplied by using a larger available blower to feed two burners and the initial blower to feed a single one. Larger bore burners with mushroom shaped heads were also fabricated to provide the appropriate velocities at the burner heads. The revised installation is shown in Figure 101.

b. Test of Higher Capacity Unit: To date the unit has been tested once to determine the requirement for photographing particulate and droplet injections. The unit was operated with air and with the simultaneous injection of water from the spray nozzle and small increments of Elkol char. Figure 102 shows the effects of a water spray from the atomizing nozzles. Small amounts of Elkol subbituminous coal char were also being injected but the particles were apparently dispersed too rapidly to be seen on any of several sequences of similar pictures.

3. Future Work: Greater blower capacity than presently available will eventually be necessary to complete the presently planned model program. In order to accommodate a likely need for an expanded model program, a search is now under way for a suitable blower that could be immediately installed.

If such a blower is soon found, the tests of simulated slag drainage will be conducted at higher air rates than the 600 scfm presently planned.

D. Data Processing (R. K. Young and D. R. Hauck)

1. Automated Data Acquisition: The PDP8/E computer and peripherals were delivered on February 16, 1972. Installation and initial checkout of the equipment is in progress.

Several computer programs are being written to demonstrate the system's capabilities. These programs will be ready for use during March, 1972. The software system (computer programs) necessary for interfacing the bench-scale methanation unit with the computer is being written and should be completed during the next two months.

2. Future Work: Plans for the next report period include:

a. Continue writing the software system for interfacing the bench-scale methanation unit with the computer.

b. Generate simulation runs with subroutine GASIFY as requested and authorized.

E. Engineering Design and Evaluation

1. BI-GAS Process: There were no requests for gasifier simulation runs during the period.

No further work was requested regarding the determination, by equation form, of oxygen requirements for gasification of selected coals. Information pertinent to West Kentucky No. 11 seam coal, Pittsburgh seam coal, lignite, and Elkol coal was presented in Progress Report No. 5.

2. OCR/BCR Gasification--Power Generation: At the request of Mr. Neal Cochran, a meeting has been scheduled for March 3, 1972, with representatives of

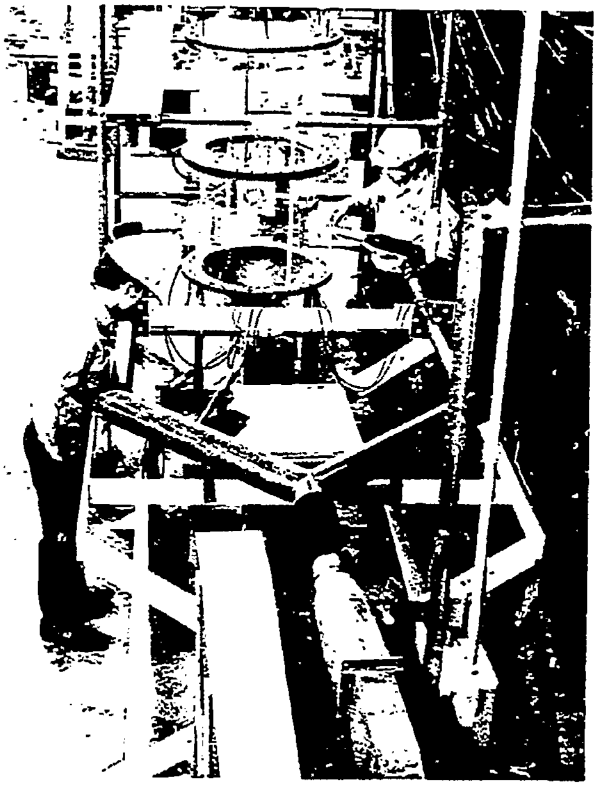


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Figure 101. Stage 1 Model Showing Added Larger Capacity Blower

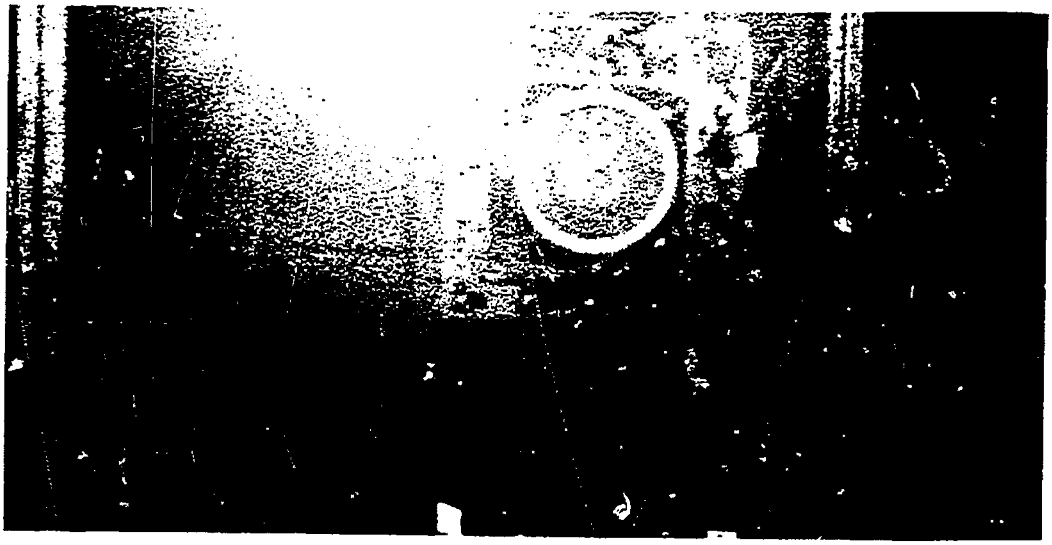

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Figure 102. Stage 1 Model Showing Water Spray from Simulated Burner

OCR, BCR, Westinghouse, Consolidation Coal Company, and Bechtel Corporation, to discuss the two-stage air-blown gasifier.

F. Multipurpose Research Pilot Plant Facility (MPPF)

A transcript of the proceedings of the bidders conference held January 19, 1972, was mailed to all bidders on February 4 together with photographs of the process area model and the plot model.

During the conference, bidders were advised that questions submitted in writing would be distributed, together with BCR's answers, to all bidders. We have received seven questions to date, and by letters dated February 10, February 18, and February 25, 1972, question and answer-type letters have been distributed to all bidders with copies to OCR. In response to an inquiry regarding water quality in Two Lick Creek at the pilot plant site, an analysis of stream samples was also sent to bidders.

February 16 and February 23 were the dates selected for plant site visitations by interested bidders. Four companies visited the site on the first date and two companies visited the site on the last date.

By letter dated February 7, 1972, BCR transmitted to OCR a copy of Koppers Memorandum, "Review of Capital Cost Estimates." This review was prepared for the purpose of comparing preliminary estimates of capital costs as presented in Volume VI of the Engineering Bid Package dated January 14, 1972, and in the base proposal contained in Koppers Evaluation Report for a Multipurpose Gasification Research Facility dated January 19, 1970. The review points out the major difference in basis, design, and scope pertinent to the estimates.

An updated bidders list was sent to OCR on February 17, 1972. This list also indicated those bidders who have advised BCR that they are unable to bid on construction of the pilot plant. When notified of an organization's decision not to bid, BCR has requested that copies of the Engineering Bid Package be returned.

1. Materials Evaluation Program: A meeting of the Task Group on material data for coal gasification equipment was held at 10 a.m. January 26, 1972, at the Pittsburgh Airport Hotel to review the report submitted by the ad hoc working group. The objective of this meeting was to formulate the program for materials evaluation of coal gasification equipment program.

Phase I on high-pressure and high-temperature evaluation of materials of construction for coal gasification has been drafted and approved by the Task Force. BCR has been requested and has reissued additional copies of the report "Report of Ad Hoc Working Group, Task Group on Materials Design Data for Coal Gasification Equipment, Subcommittee 8 on Corrosion, Metals Properties Council, January 4, 1972" to the Task Group members, 10 copies to AGA, and 10 copies to the chairman of the Task Group.

Detailed minutes of this meeting will be sent to all Task Force members.

2. Model Status: The process pilot plant model (1/4 inch = 1 foot) and the overall site model (1 inch = 20 feet) were completed and delivered to BCR on January 17, 1972.

3. Desk Type Model: During this report period, Visual Industrial Products, Inc., of Indianola, Pennsylvania, was authorized to construct a desk-top scale model (1/32 inch = 1 foot). Approval to purchase this model was given by Mr. Neal Cochran per telephone conversation of February 7, 1972; a letter confirming the conversation was sent to OCR on the same date.

The desk top model is based on the following design: A 250 MM scf/day SNG plant based on the process design as given in the Air Products report entitled, "Engineering Study and Technical Evaluation of the Bituminous Coal Research, Inc., Two-stage Superpressure Gasification Process," (OCR R & D Report No. 60).

The model is scheduled for completion on February 26 and will be delivered to OCR in Washington, D. C., on Monday, February 28, 1972.

4. Pollution Control Permits - Homer City, Pa.: On Monday, February 14, 1972, initial contact was made with the Pennsylvania Department of Environmental Resources in preparation for an industrial waste permit for the Homer City pilot plant.

G. Literature Search (V. E. Gleason)

Annotated literature references completed during the month are listed in Appendix B.

H. Other

1. Prime Contract Matters: Additional surplus equipment lists are being compiled and will be submitted in March. Early action is requested for the list submitted in September, as well as the lists to be submitted, because of limited storage facilities and the need for space for the new PEDUs.

By letter dated January 12, 1972, Mr. Howard E. Thunberg requested BCR to submit details regarding estimated costs for fiscal years 1972 and 1973 under Contract 14-32-0001-1207 for his use in submitting a statistical report to the National Science Foundation. This information was transmitted to OCR on January 24, 1972.

2. Outside Engineering and Services: Koppers continues to provide engineering assistance as required and as reported in their Progress Report No. 31 in Appendix C.

Action by OCR on the request for approval of proposed Amendment No. 7 to Subcontract No. 2, signed by Koppers and submitted to OCR on October 20, 1971, has not been received.

3. Brigham Young University: The project entitled "Study of High Rate, High Temperature Pyrolysis of Coal" with joint funding by Brigham Young University

and BCR is now in its eleventh month. Figure 103, Monthly Progress Chart, Expenditures, shows the current budget status. The letter report of progress by BYU is as follows:

During February a series of test runs were made with the 3/4-inch reactor. Serious plugging of the reactor was encountered under the feed conditions selected, namely 3.3 pounds per hour of coal and 1.0 pounds per hour oxygen-hydrogen combustion gas, but some data were obtained on the percent coal gasified and the gas composition for short duration runs. Representative data are listed in Table 80. Comparison of these data with prior data obtained with near the same feed rate in the 1-1/2-inch reactor shows that the percent coal gasified and the yield of hydrocarbon gases from these runs are lower than obtained previously. These tests are continuing, varying the feed rates of both coal and combustion gas.

The porous ceramic filter elements for use in the effluent sample collection bottle have proved satisfactory in the test runs.

A problem with the electrical heating elements shorting out has been encountered. The elements were replaced and better insulated to prevent this problem. Problems with air contamination of the gas samples have also been encountered. Better procedures for collecting the samples are being developed.

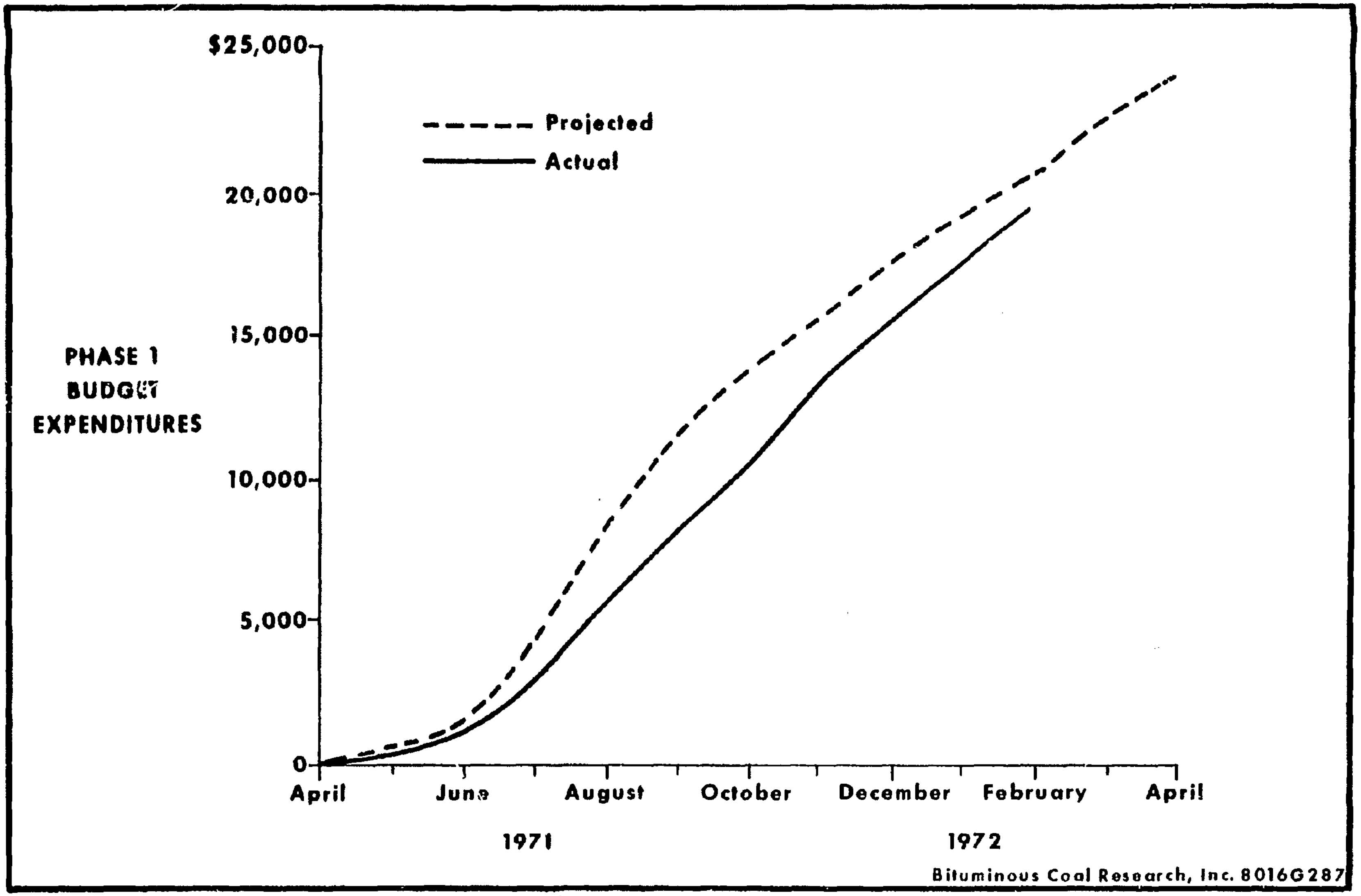
4. FPC National Gas Survey - Economics of Manufacturing SNG from Coal: BCR has completed a report, "Economics of Manufacturing SNG by BCR/OCR BI-GAS Coal Gasification Process," to produce 250 MM scf/day of pipeline gas. Copies of this report have been distributed to all Task Force members for their review in preparation for the next synthetic gas-coal Task Force meeting set for February 17, 1972.

5. Reports and Papers: A talk on the status of the BCR/OCR BI-GAS Process will be presented by J. W. Tieman on March 23, 1972, at a regional Meeting of the Air Pollution Control Association in Buffalo, New York.

6. Patent Matters: Worthwhile ideas continue to be written up as invention disclosures for submission to OCR for consideration.

a. OCR-866 and OCR-1078: A U.S. patent application based on the new process concept (E. E. Donath, December 11, 1970) has been filed and given Serial Number 182,652. The application, entitled "Gasification of Carbonaceous Solids," contains nine claims. The appropriate document assigning rights to the U.S. Government was filed with the patent application on September 22, 1971. BCR has applied for and received a license for foreign filing. On February 23, 1972, patent applications were forwarded for filing in Australia, India, South Africa, and Great Britain. Applications are also being prepared for filing in Canada, France, Japan, and West Germany.

Confirmatory license to the government was executed by BCR and returned to Mr. Ernest Cohen, Assistant Solicitor, Branch of Patents, under date of January 12, 1972.



Bituminous Coal Research, Inc. 8016G287

Figure 103. Monthly Progress Chart, Expenditures, Brigham Young University

TABLE 80. AVERAGES OF DATA FROM THREE RUNS
WITH 3/4-INCH DIAMETER BYU REACTOR

Feed Rates, lb/hr	Scf/hour	lb/hour
Combustion Gas		
Oxygen	10	0.9
Hydrogen	20	0.1
Entrained Coal		
Hydrogen	20	0.1
Coal	--	3.3
Nitrogen Purge	5	0.4
Reactor Temperature		1700°F
Gas Analysis, Mole Percent (dry)		
Hydrogen		61.4
Carbon Monoxide		13.5
Carbon Dioxide		2.8
Methane		2.9
Ethane		----
Ethylene		----
Acetylene		0.8
Oxygen		2.5
Nitrogen		15.6
Percent Coal Gasified		30

Note: Temperatures measured and samples withdrawn at a location 2 inches below point of coal injection.

b. New Invention Disclosures: Formal Invention Disclosures (Form DI-1217) for six individual BCR suggestions were submitted to OCR on May 7, 1971. These were listed in Progress Report No. 1.

Inasmuch as 90 days have elapsed since the submission of these disclosures, in accordance with the patent clause under Contract 14-01-0001-324, BCR is proceeding, as reported last month, to develop patent applications for filing in the U.S., first obtaining the approval of the Solicitor's Office. Action taken on these disclosures is as follows:

(1) OCR-1860 and OCR-1861: As previously reported, Disclosures 1860 and 1861 have been combined into a single patent application entitled "Two-stage Gasification of Pretreated Coal." A copy of this application was mailed to Mr. Howard Silverstein, Branch of Patents, on December 6, 1971, for his review and authorization for filing by BCR. The appropriate document assigning rights to the government has been prepared.

By letter dated December 28, 1971, Mr. George Fumich authorized BCR to file a foreign patent application on the subject invention. This letter was signed by John W. Igoe and returned to Mr. Fumich under date of January 11, 1972, agreeing that BCR will file and prosecute applications; the government will pay the patent filing and issue fees. Other expenses incurred by BCR, including preparation and prosecution of applications, are non-reimbursable.

During the month this patent application was reviewed again by the inventor and certain revisions have been made. Copy of the revised application will be forwarded to Mr. Silverstein.

Confirmatory license will be prepared after application is filed in U.S. Patent Office and serial number is received.

(2) OCR-1862: As previously reported, a U.S. patent application has been prepared on Disclosure 1862 entitled "Three Stage Gasification of Coal." A copy of this application was sent to Mr. Howard Silverstein, Branch of Patents, on December 13, 1971, for his review and authorization for filing by BCR. The appropriate document assigning rights to the U.S. Government has been prepared.

By letter dated December 28, 1971, Mr. George Fumich authorized BCR to file a foreign patent application on the subject invention. As stated above, this letter was signed by John W. Igoe and returned to Mr. Fumich on January 11, 1972.

This patent application was reviewed again by the inventor and revisions have been made. Copy of the revised application will be forwarded to Mr. Silverstein.

Confirmatory license will be prepared after the application is filed in the U.S. Patent Office and serial number is received.

(3) OCR-1863: As previously reported, a U.S. patent application has also been prepared on Disclosure OCR-1863 entitled "Two-stage Downflow Gasification of Coal." A copy of this application was forwarded to

Mr. Howard Silverstein, Branch of Patents, on December 13, 1971, for his review and authorization for filing by BCR. The appropriate document assigning rights to the U.S. Government has been prepared.

By letter dated December 28, 1971, Mr. George Fumich authorized BCR to file a foreign patent application on the subject invention. As stated above, this letter was signed by John W. Igoe and returned to Mr. Fumich on January 11, 1972.

This patent application was reviewed again by the inventor and certain revisions have been made. Copy of the revised application will be forwarded to Mr. Silverstein.

Confirmatory license will be prepared after application is filed in U.S. Patent Office and serial number is received.

(4) OCR-1864: As previously reported, a U.S. patent application was prepared on Disclosure OCR-1864 entitled "Two-stage Gasification of Coal with Forced Reactant Mixing and Steam Treatment of Recycled Char." A copy of this application was mailed to Mr. Howard Silverstein, Branch of Patents, on December 8, 1971, for his review and approval for filing by BCR. The appropriate document assigning rights to the U.S. Government has been prepared.

By letter dated December 28, 1971, Mr. George Fumich authorized BCR to file a foreign patent application on the subject invention. As stated above, this letter was signed by John W. Igoe and returned to Mr. Fumich on January 11, 1972.

This patent application has also been reviewed again by the inventor and revisions made to the original application. Copy of the revised application will be forwarded to Mr. Silverstein.

Confirmatory license will be prepared after application is filed in U.S. Patent Office and serial number is received.

c. Invention Disclosure--Brigham Young University: During the course of work under Subcontract No. 3, Professor R. L. Coates, Brigham Young University, developed a new concept of pyrolyzing coal which may be patentable. An Invention Disclosure (Form DI-1217) entitled "Process for High Temperature Pyrolysis of Coal," was prepared and submitted to Mr. George Fumich on January 6, 1972, for his consideration.

I. Visitors During February, 1972

February 1, 1972

Mr. R. Hinderliter
Pittsburgh & Midway Coal Mining Co.
9009 W. 67th Street
Merriam, Kansas 66202

Mr. D. M. Mitsak
Mr. A. E. Miller
Koppers Company, Inc.
Koppers Building
Pittsburgh, Pennsylvania 15219

Mr. Nelson Gardner
Mr. E. Samuels
Mr. Karl Wilks
Mr. R. Cwiklinski
Case Western Reserve University
Cleveland, Ohio 44106

February 7, 1972

Mr. James W. Clark
United Aircraft Corporation
East Hartford, Connecticut 06108

Mr. S. M. Tymiak
Mr. F. L. Brown
Koppers Company, Inc.
Koppers Building
Pittsburgh, Pennsylvania 15219

February 11, 1972

Mr. D. M. Mitsak
Mr. S. M. Tymiak
Koppers Company, Inc.
Koppers Building
Pittsburgh, Pennsylvania 15219

February 23, 1972

Mr. W. J. Kerr
Koppers Company, Inc.
Koppers Building
Pittsburgh, Pennsylvania 15219

February 24, 1972

Mr. Uzi Halevy
Project Manager
The Litwin Corporation
P.O. Box 282
Wichita, Kansas 67201

Mr. D. M. Mitsak
Koppers Company, Inc.
Koppers Building
Pittsburgh, Pennsylvania 15219

Mr. H. E. Evans
Koppers Company, Inc.
Koppers Building
Pittsburgh, Pennsylvania 15219

J. Trips, Visits, and Meetings during February, 1972

February 16, 1972	Office of Coal Research Washington, D. C.	E. K. Diehl J. T. Stewart

K. Requests for Information

Mrs. Anita Pancook
Combustion Engineering, Inc.
Windsor, Connecticut 06095

Mr. James Rooney
USS Agri-Chemicals
10 E. Temple
Salt Lake City, Utah 84111

Mr. Joseph Ilaqua
Assistant Professor
Department of Economics
Bryant College
Smithfield, Rhode Island 02917

Continental Oil Company
Research and Development Dept.
Drawer 1267
Ponca City, Oklahoma 74601

III. WORK PLANNED FOR MARCH, 1972

The work planned for March will basically be a continuation of the on-going program which has been underway for the past few months.

The final summary report on the coal composition and beneficiation studies will be mailed during the first full week in March.

As a result of the February 16, 1972, meeting with OCR concerning the fluidized-bed PEDU gasifier, and submission of a preliminary design on February 25, 1972, based on these discussions, no work is planned during the coming month until a decision by OCR is reached. Tests in the laboratory-scale batch reactor will, however, continue.

Tests in the bench-scale methanator will continue in order to evaluate suitable catalysts. Tests are planned in the life-test unit and construction of the new unit will proceed. Bids on equipment for the methanation PEDU will be solicited and, after evaluation, approval to purchase will be requested. Work on the model studies is planned providing the catalyst is received.

The first draft of the final summary report on the Stage 2 PEDU (100 lb/hr) will undergo final editing. An early mailing date is planned.

Installation of a large blower is planned for the model studies of the 5 ton/hr two-stage gasifier. Higher air rates than 600 cfm will be required for tests of simulated slag drainage in the first stage.

Installation and checkout of the PDP8/E computer and peripherals will continue. Several programs will be written to demonstrate the system's capabilities. Data from simulation runs will be evaluated as needed.

With respect to the Homer City pilot plant, meetings are planned with organizations interested in providing project management services to BCR.

A. Trips and Meetings Planned

April 10-14, 1972	163rd National Meeting of American Chemical Society Division of Fuel Chemistry Boston, Massachusetts	J. W. Tieman and/or E. K. Diehl

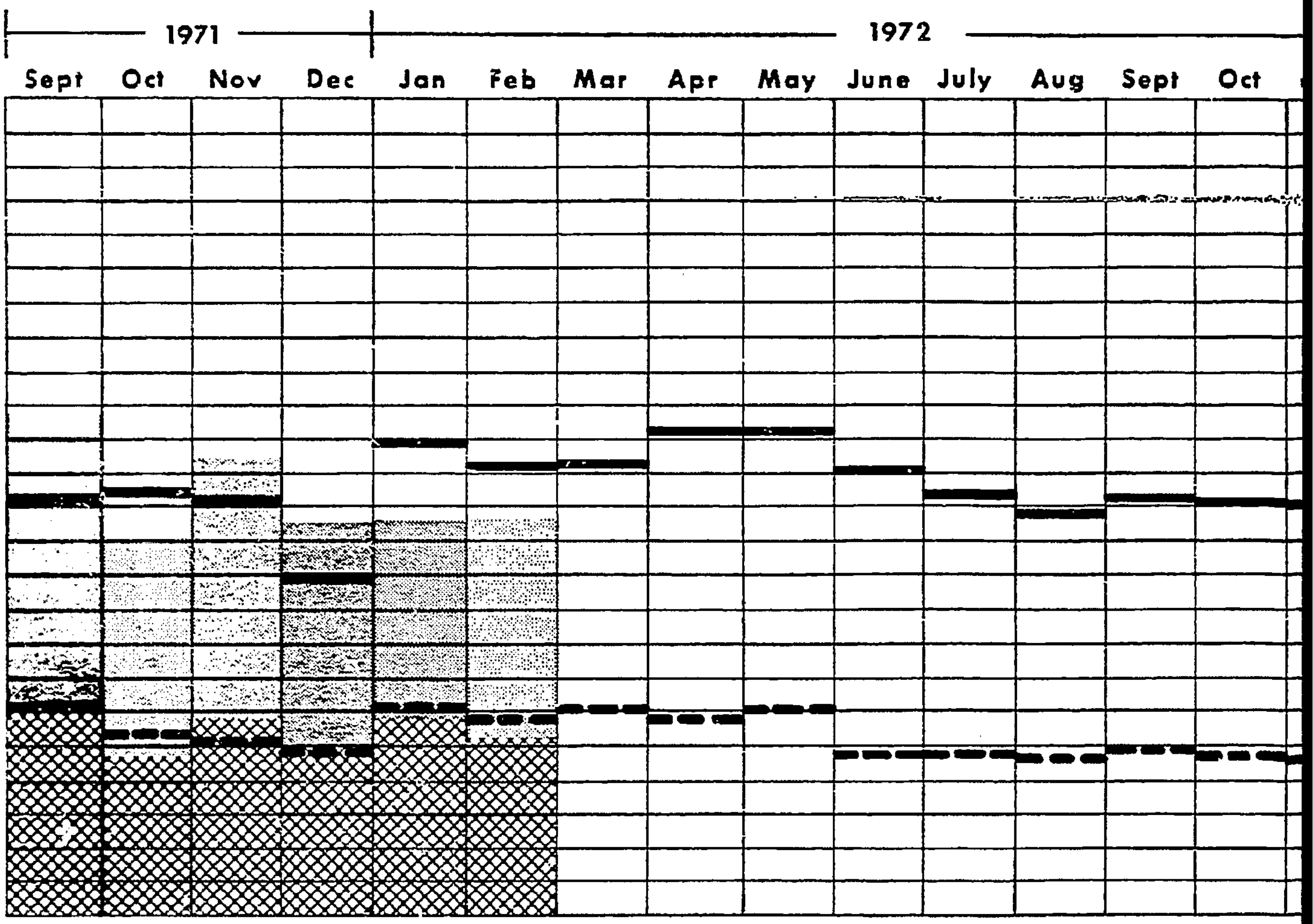
B. Papers to be Presented

None

C. Visitors Expected

None

MANHOURS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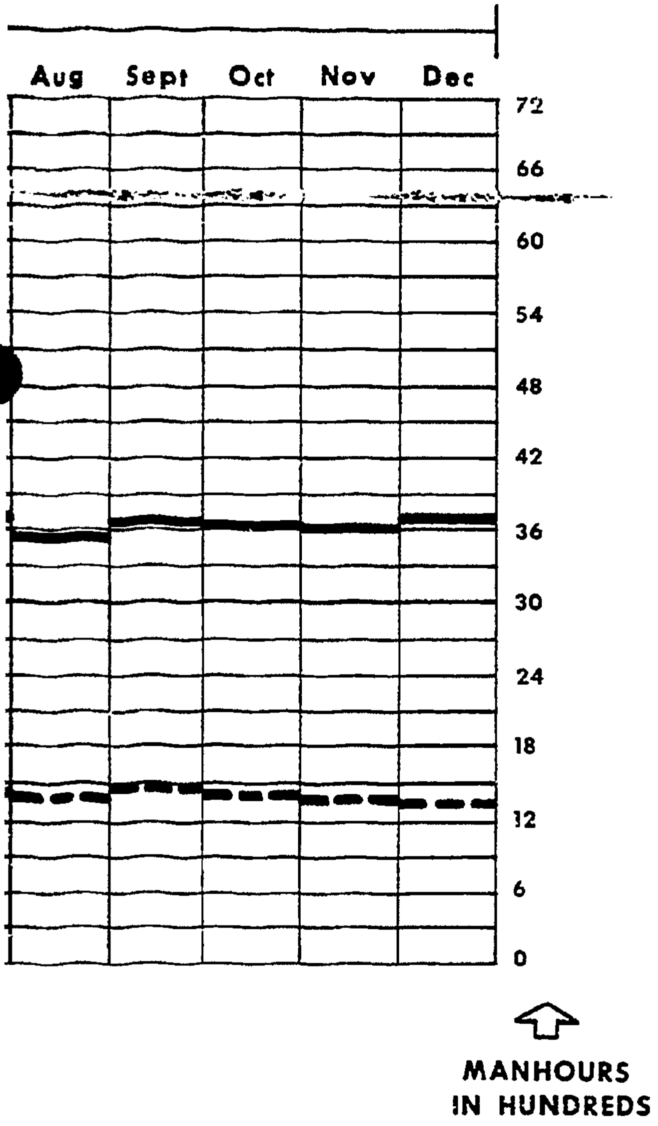

MONTHLY PROGRESS CHART
PART 1 MANHOURS

Bituminous Coal Research, Inc.
350 Hochberg Road Monroeville, Pa.

OFFICE OF COAL RESEARCH
DEPARTMENT OF THE INTERIOR

CONTRACT NO. 14-32-0001-1207

- ▬ Predicted Professional and Non-professional
- ▬ ▬ Predicted Professional
- ☐ Actual Non-professional
- ▨ Actual Professional

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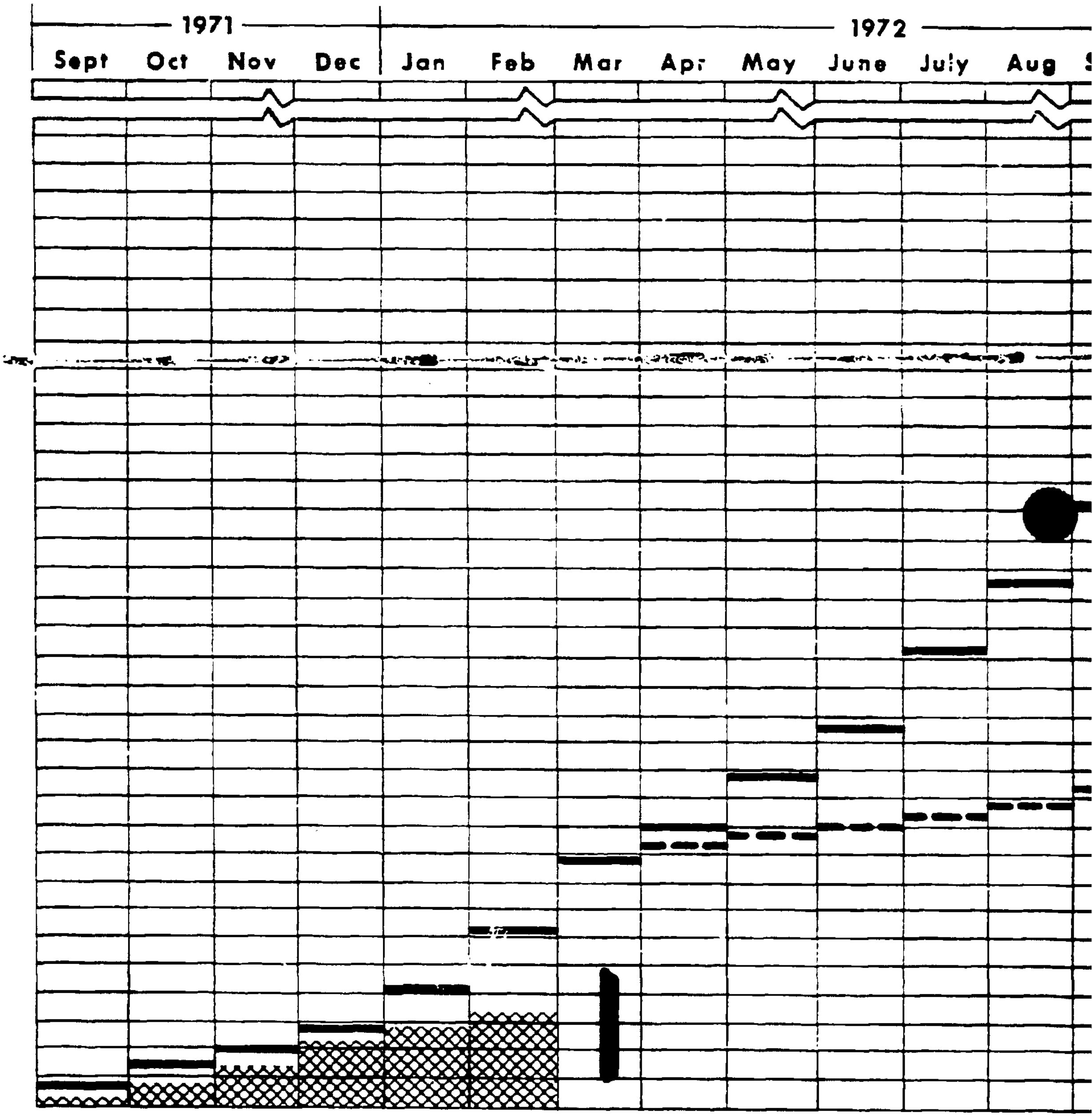


MONTHLY EXPENDITURES (All Costs, in D

		Sept	Oct	Nov	Dec	Jan	Feb	Mar	Apr	
Monroeville	**Predicted**	129,991	129,991	129,991	129,991	323,486	382,228	558,454	105,058	8
	Actual	63,610	121,696	146,834	144,590*	103,147	86,250*			
Homer City	**Predicted**								154,000	2
	Actual									
Total	**Predicted**	129,991	129,991	129,991	129,991	323,486	382,228	558,454	259,058	3
	Actual	63,610	121,696	146,834	144,590*	103,147	86,250*			

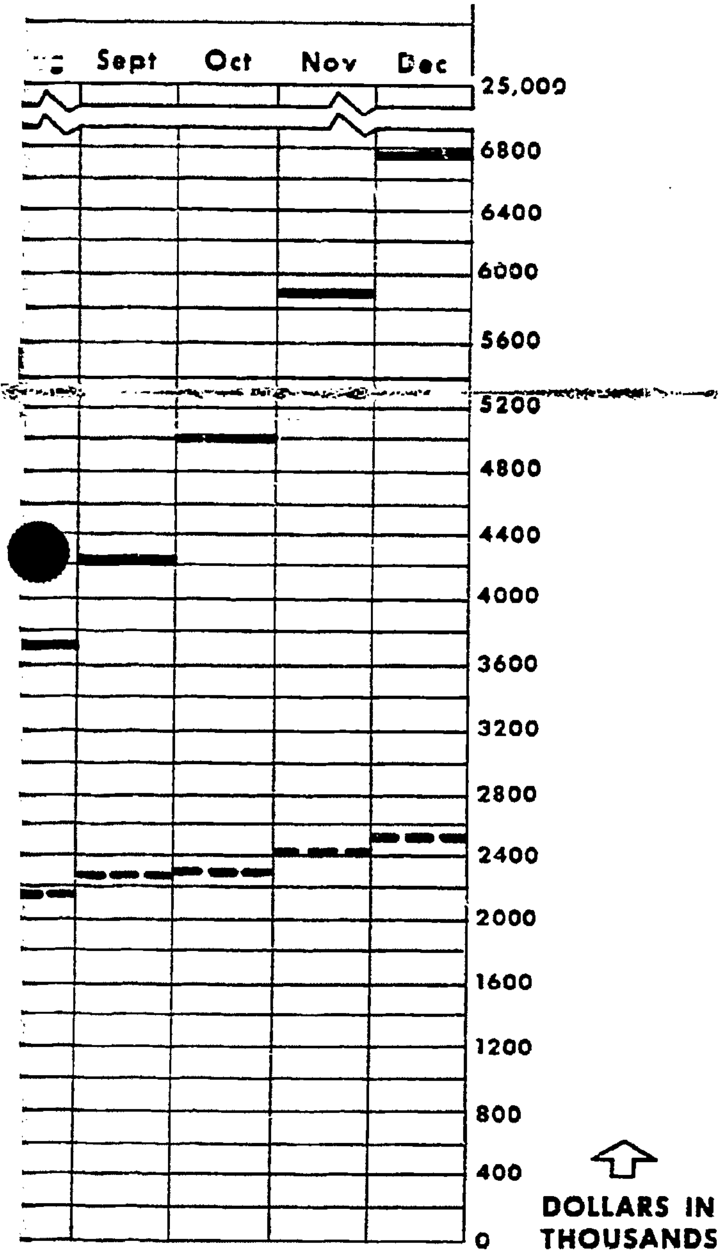
CUMULATIVE EXPENDITURES

MONTHLY PROGRESS CHART
PART 2 EXPENDITURES

Bituminous Coal Research, Inc.
350 Hochberg Road Monroeville, Pa.

OFFICE OF COAL RESEARCH
DEPARTMENT OF THE INTERIOR

CONTRACT NO. 14-32-0001-1207



Expenditures, Cumulative

▨ Actual, Monroeville
☐ Actual, Homer City

Predicted Expenditures, Cumulative

— Total, Monroeville and Homer City
--- Monroeville

2

, in Dollars)

	May	June	July	Aug	Sept	Oct	Nov	Dec
8	86,238	86,240	65,813	65,813	74,746	62,273	62,273	62,275
	215,600	280,400	444,300	444,300	444,400	760,600	760,600	760,800
	301,838	366,640	510,113	510,113	519,146	822,873	822,873	823,075

* Estimated

APPENDIX B

ADDITIONS TO ABSTRACT FILE, FEBRUARY 1972

"Coal gasification technology at center stage," Chem. Eng. News 50 (2), 36, 38 (Jan. 10, 1972). 540.000 72-1

> Four domestic gasification processes are considered to to the most likely to be able to compete with foreign technology, particularly the commercially available Lurgi process. The four, BCR's BI-GAS, Bureau of Mines' Synthane, IGT's HYGAS, and Kellogg's molten salt processes, are reviewed and their latest developments summarized.

PATENT

Tornquist, E. L. (to Northern Illinois Gas Co.), "Gasification of carbonaceous solid fuels," U.S. Pat. 3,004,839 (Oct. 17, 1961). 12 pp. 540.000 61-2

> Methane is produced by hydrogenation of coal or other solid fuel at about 40 atmospheres pressure and at temperatures in the range of 1500-1700 F. The gasification apparatus has two reaction zones in direct heat transfer relation so that the exothermic heat of the reaction can be used in producing heat for the process. Additional heat that may be necessary is supplied by burning some of the fuel with oxygen. Alternative steps are proposed to adjust the concentration of the reactants, to separate and remove the reaction products, and to adapt the system to use by caking coals.

C-362.

APPENDIX C

PROGRESS REPORT #31

Bituminous Coal Research, Inc.
Coal Gasification

February 1972

Koppers Contract 2415

I. STATUS OF CONTRACT

A. Pilot Plant Engineering Bid Packages

Step No. 1: Pilot Plant for oxygen-blown, two stage coal gasification system, including general facilities: design and models.

(Work Completed)

Step No. 2: Fluidized bed system.

(Work Deferred)

B. Engineering Assistance and Recommendations for PEDU Program

1. Informal review of PEDU Program was made at BCR on February 11, 1972 and reported in Koppers letter C-313.

2. Koppers continued to provide engineering assistance for Fluid Bed Methanation PEDU. Following items were completed:

   a. Panel Board arrangement was completed, approved by BCR and released for bids.

   b. Drawings of vessels for fabrication were completed, approved by BCR and released for bids.

   c. Revised design and materials of construction of methanation unit to permit addition of H_2S and CO_2 to feed gas and to increase design temperature from 900°F. to 1050°F. was approved by BCR and released for bids.

   d. BCR and Koppers reviewed sites for storage of CO_2 and H_2S. A site was selected, subject to addition of a fence and other safety measures.

   e. Koppers reviewed H_2S removal system and proposed inclusion of two (2) 3 ft. 6 in. by 15 ft. high iron oxide towers to be installed before thermal oxidizer. Additional costs submitted to BCR for their approval.

f. Reviewed Engineering Manday Schedule and reduced the scope of work to approximately 900 Mandays. Awaiting BCR's approval of the revised scope of detail engineering.

3. Revised scope of work to provide for three-stage Coal-Char Fluid Bed Gasification PEDU (including estimated capital costs) was submitted to BCR in Koppers letter C-321 of February 22, 1972. Much of the flexibility and control included in the original design was eliminated to reduce costs. However, increasing the number of vessels required from one (1) to three (3) resulted in a cost about equal to that of the original design.

4. No further work is being performed on the Char Fluid Bed Gasification PEDU pending receipt of further direction from BCR.

II. CONTRACT EVALUATION

Four (4) copies of Amendment No. 7 to Amended Subcontract No. 2, including Appendices I through VIII, signed by Mr. J. D. Rice, Vice President, Engineering and Construction Division, Koppers Company, Inc., were transmitted to BCR in our letter C-183 dated October 18, 1971. Receipt of these copies was acknowledged by BCR in their letter dated October 18, 1971.

J. F. Farnsworth
Project Manager