

MWR-MPR-26

RESEARCH AND DEVELOPMENT DEPARTMENT



DEVELOPMENT OF KELLOGG COAL GASIFICATION PROCESS

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THE M. W. KELLOGG COMPANY
A DIVISION OF PULLMAN INCORPORATED



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RESEARCH & DEVELOPMENT DEPARTMENT

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1. SUMMARY

This progress report is the twenty-sixth since the awarding of the contract. It is concerned with the first phase of the contract and summarizes the progress that has been made in the three principal areas now being studied: process research, chemical engineering studies and mechanical development.

Nine new combustion runs have been made at three atmospheres total pressure using anthracite, a South Beulah lignite, an oxidized bituminous coal and a sub-bituminous coal with initial temperatures ranging from 1650 to 1930°F. During each of these runs an increase in melt temperature was noted, although this phenomenon was most pronounced with the reactive feedstocks, namely, lignite and subbituminous. The combustion rates for these two materials were so rapid that all of the oxygen fed to the system was consumed during the first few minutes of each run.

The combustion rates of anthracite and an oxidized bituminous coal were not nearly as high as for the other two fuels. Also, the combustion rates observed were lower than the gasification rates obtained at comparable conditions. However, the rates obtained are quite consistent with those assumed for the conceptual process designs which have been made.

Two additional combustion runs were made--one each with anthracite and oxidized bituminous coal. In each of these runs the fuels were burned in a melt containing about 4 percent lignite ash. For these runs, combustion rates were increased by a factor of 2.5 or higher. Work is continuing to better elucidate these results.

Economics have been calculated for the manufacture of 250,000,000 SCFD of pipeline gas from lignite and an FMC char. Two processing schemes were considered for the lignite case--one essentially the same as that used for bituminous, sub-bituminous and anthracite and the other involving decomposition of the recycle sodium bicarbonate outside of the gasifiers. With such a scheme plant investment was reduced by about 20 percent to approximately \$162,000,000 and gas selling price was lowered from 63¢/MSCF to about 58¢/MSCF. Total investment for the char processing plant, with outside bicarbonate decomposition, is estimated to be about \$192,000,000 and gas selling price, with char taken at 10¢/MM BTU, is calculated to be about 72¢/MSCF. The chief factor in the high costs obtained in these cases is the high ash content of the fuels considered.

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Work has continued on the preparation of the process flowsheet for the hydrogen-from-coal-plant.

Corrosion Test #10, designed to study the resistance to a reducing atmosphere of Monofrax A blocks under compressive stress, has been begun.

Simulation studies of the air-lift circulation system have been continued. A plastic model has been constructed consisting of two vessels with two air-lift pumps capable of providing cross circulation between the vessels. This equipment will enable the degree of cross contamination by the two lift gases to be measured.



11. PROCESS RESEARCH

A. Accomplishments

Process research has attempted to evaluate the combustion rates of the same feedstocks used in the gasification studies. The standard conditions employed consisted of: 3 atmospheres absolute pressure, 1 ft./sec. superficial air velocity, 4 inch bed height, 2% ash in melt, 4% carbon initially present based on the fixed carbon in the feedstock, and a 5 minute devolatilization period in N₂ after charging the feedstock and before introducing air. The combustion runs are summarized in Table I.

1. Combustion of Char, Subbituminous Coal and Lignite

Three of the feedstocks, namely, FMC char, Elkol subbituminous coal and South Beulah lignite reacted at maximum combustion rate, 70 lbs carbon burned per hour per cubic foot of melt. This represents complete utilization of the oxygen. The temperatures of the tests are shown in the following table:

<u>Feed</u>	<u>Temperatures - °F</u>		
	<u>Initial</u>	<u>Average for Rate</u>	<u>Maximum</u>
FMC char	1640 1740	1762 1870	1847 1940
Elkol subbit. coal	1740	1831	1925
S. Beulah lignite	1640 1600	1753 1690	1874 1821

The maximum combustion rates are well reflected by the immediate transfer of heat to the salt and the Inconel reactor. Even the last run with S. Beulah lignite at the lowest possible initial test temperature of 1600°F (melting point of Na₂CO₃ is 1560°F) was sufficient to have all the oxygen consumed in the first few minutes.

The combustion rates for the char, subbituminous coal and lignite are faster than the gasification rates, thus rate of combustion will not be a limiting factor. Control of temperature in a large unit with these feedstocks may have to be regulated by control of the amount of air introduced for combustion.



2. Combustion of Anthracite, Bituminous Coal (oxidized) and Coke

Combustion runs have been made on anthracite and oxidized bituminous coal (1 week at 150°C in air) at three temperatures. These runs, given in Table 1, together with previous runs on coke derived from the same bituminous coal have been used to give the following table:

Feed	Temperatures-°F			Rate of Combustion lbs/hr/CF
	Average	Initial	Maximum	
Anthracite	1745	1749	1766	9.0
	1842	1840	1870	14.9
	1957	1930	1980	31.7
Bitum. Coke	1743	1740	1754	11.7
	1846	1840	1863	21.5
	1973	1940	1985	40.5
Ox. Bitum. Coal	1659	1650	1690	14.4
	1760	1740	1788	19.5
	1892	1840	1910	33.7

The average temperature is that during which the rate is obtained, that is, during 50% consumption of the feed. Even in these runs where the rates of combustion are not high, the transfer of combustion heat to the molten salt is reflected in the average and maximum temperatures. Arrhenius plots of the data show curvature at high temperature for anthracite and oxidized bituminous coal. Interpretation of this effect is not known as yet.

In the following tabulation, gasification rates are compared to the combustion rates at 1740°F for these three feedstocks. Combustion rates are 37 to 54% of the gasification rates.

	Rates at 1740°F		%
	Combustion	Gasification	
Anthracite	8.8	23.6	37
Bitum. Coke	11.8	25.3	47
Oxid. Bit. Coal	18.3	33.8	54

This phenomenon, contrary to literature data for the rates of combustion and steam gasification of coal, must await further experimentation for explanation. However, based upon the effects of variables in gasification, it is hypothesized that differences in carbon distribution between the steam-melt and air-melt systems may be the underlying cause.

It should be pointed out here, however, that although these rates seem low, they are still quite consistent with those used in the conceptual process designs. For example, for bituminous coal a combustion rate of about 12 pounds carbon/hr/CF was assumed.



3. Combustion of Anthracite and Bituminous Coal in a Melt Containing Lignite Ash

After the last run with South Beulah lignite (H-87), the melt was reused to test anthracite in run H-88 and oxidized bituminous coal in run H-89. The lignite ash content in the melt had built up to 4.4%. In run H-89, the ash from the anthracite feed in run H-88 represented less than 0.6% basis the melt. The following table compares the results obtained with expected values for combustion in melts containing 2% ash basis the coal burned and expected values for gasification. Combustion rates were improved by a factor of 2.5 or better and, in addition, they were higher than the gasification rates.

<u>Feed</u>	<u>Avg. Temp. F</u>	<u>Combustion Rates in Melts of</u>		<u>Gasification Rate</u>
		<u>4.4% Lignite Ash</u>	<u>2% Feed Ash</u>	
Anthracite	1933	>70	26	60.2
Oxid. Bit. Coal	1731	44.3	17.8	31.8

Exact explanation of this effect must await further testing. Whereas anthracite and bituminous coal ashes are high in silica and alumina, lignite ash is high in calcia, magnesia, iron oxide and sulfur. Lignite ash tends to give much lower viscosity melts compared to bituminous and anthracite melts. This effect could be attributed to a catalytic action or to a physical action which improved air-carbon contact.

B. Projections

Evaluation of the effect of air pressure on the rate of combustion will be determined. Effect of the other combustion variables will also be ascertained.

TABLE I

SUMMARY OF COMBUSTION RUNS IN MOLTEN SODIUM CARBONATE (1)

Run No. H- Date - 1966	80 9/2 Elkol	81 9/6	82 9/7	83 9/9	84 9/19	85 9/20	86 9/22	87 9/23	88 9/26	89 9/27	90 9/29
Feed	Subbit. Coal	Anthracite		Oxidized Bituminous Coal			S. Beulah Lignite	Anthracite	Ox. Bit. Coal	Anthracite	
% Fixed Carbon	48.5	-	-	58.3	58.3	58.3	42.88	42.88	-	58.3	-
% Total Carbon	73.5	80.8	80.8	81.3	81.3	81.3	66.35	66.35	80.8	31.3	80.8
% Vol. Matter	48.3	5.9	5.9	37.3	37.3	37.3	44.0	44.0	5.9	37.3	5.9
% Ash	3.2	11.7	11.7	3.9	3.9	3.9	13.1	13.1	11.7	3.9	11.7
gms. charge	35.37	21.35	21.35	29.60	29.60	29.60	40.24	40.24	21.35	29.60	21.35
mesh size	12/20	12/20	12/20	12/20	12/20	12/20	12/20	12/20	12/20	12/20	12/20
% C in melt-initial	-----										
Na ₂ CO ₃ - gms.	405.7	405.7	(3)	405.7	(3)	(3)	405.7	(3)	(4)	(4)	405.7
Ash - gms.	8.3	8.3	-	8.3	-	-	8.3	-	-	-	8.3
% Ash in bed	2	?	2	2	2	2	2	3.2	4.4	4.4	2
Bed Height - Inches	-----										
Conditions											
Temp. °F - Initial	1740	1749	1930	1740	1840	1650	1641	1600	1845	1650	1840
" - average (2)	1831	1745	1957	1760	1892	1659	1753	1690	1933+	1731	1842
" - maximum	1925	1766	1980	1788	1910	1690	1874	1821	2000+	1802	1870
Pressure - psia	44.9	45.5	46.2	46.2	44.6	45.6	45.2	44.9	45.8	44.5	45.0
Sup. Gas Vel. - ft/sec	0.93	0.99	1.03	0.94	1.00	0.93	0.91	0.90	0.97	0.89	0.96
Run Time - min.	10	65	25	30	15	45	10	7.5	8.3	10	40
Air Rate - liters/min	24.5	26.1	25.0	24.8	25.2	25.8	25.2	25.1	24.6	24.8	24.4
Results - Prod. Gas											
% CO ₂ - 5 min.	19.5	3.0	7.5	6.4	12.0	6.0	18.0	19.0	16.5	16.0	4.2
- 35 min.	-	1.7	-	-	-	1.75	-	-	-	-	1.8
- end	0.5	1.0	1.6	2.2	4.2	1.3	1.7	1.1	1.0	6.8	1.3
% O ₂ - 5 min.	1.0	18.0	13.0	15.5	10.0	15.5	2.0	2.0	2.8	7.0	17.0
- 35 min.	-	20.5	-	-	-	19.0	-	-	-	-	18.5
- end	20.0	20.0	20.0	18.2	17.5	19.0	20.3	19.0	17.8	15.5	18.3
% CO - 5 min.	0.3	0	0	0	0	0	1.5	1.1	0	0.1	0
Combustion Rate Constant	>6	0.76	2.69	1.65	2.86	1.22	>6	>6	76	3.75	1.26
Total % Fixed Carbon Consumed	109	99.4	100.6	97.5	102.2	95.4	112	100	100	96.0	99.7
Rate - lbsC/hr/GF	>70	9	31.7	19.5	33.7	1.44	>70	>70	>70	44.3	14.9
% of Total Carbon - Devolatilized	17.7	12.5	12.2	14.0	10.6	15.7	14.1	16.0	14.6	13.9	14.7
% of Total Carbon - Combusted	71.9	87.0	83.3	63.9	73.3	68.4	72.4	64.6	85.4	68.8	85.0
% of Total Carbon - Tar + Loss	10.4	0.5	-	16.1	16.1	15.9	13.5	19.4	-	17.3	0.3

(1) Used 2" ID Inconel reactor. Feed charged then 5 min. devol. period in 0.1 ft/sec N₂ before air in.

(2) Average temperature during 50% carbon consumption.

(3) Reused melt from previous run.

(4) Reused previous run melt which contained lignite ash.



III. CHEMICAL ENGINEERING STUDIES AND DEVELOPMENT

A. Accomplishments

1. Pipeline Gas

Flowsheets have been completed for the production of pipeline gas from lignite and an FMC char. The Case I lignite flowsheet is substantially the same as the bituminous, subbituminous, and anthracite flowsheets previously reported. The Case II lignite and the char flowsheets differ inasmuch as the recycle sodium bicarbonate from the ash removal section is calcined to sodium carbonate before being returned to the gasifiers. The purpose in doing this is to lessen the heat load on the gasifiers brought about by the large heat of decomposition of the bicarbonate as well as reducing the CO₂ removal requirements of the gas purification system. The investment for each of these cases is presented in Table 2 and the resulting gas manufacturing costs are given in Table 3.

In comparing the investments for Cases I and II for lignite it is apparent that the plant cost is much lower with the return of sodium carbonate instead of sodium bicarbonate to the gasifiers. By adding a kiln to each ash removal train, a substantial reduction is made in the cost of the gasification and gas purification sections which more than offsets the increased ash removal cost.

The removal of water and carbon dioxide from sodium bicarbonate reduces the gasifier effluent and therefore the equipment sizes and costs. However, the reduced amount of gasifier effluent also means there is less heat to be removed thereby reducing steam production and power credit. The reduction in the power credit is more than offset by the reduction in fixed charges.

The investment and operating costs for char are noticeably higher than for lignite. The reason for this is the high ash level (17.4%) of the char. The investment increase appears in the gasification and ash removal sections. The increase in equipment in the ash removal section is due to the large amount of sodium carbonate to be processed. The increase in the gasification section is the result of an increase in the heat of reaction due to the small amount of methane formation and the high melt heat effects associated with the large melt recycle rate. The operating costs also show this effect of ash level. The major cost



increases over Lignite II are due to higher fixed charges and ash losses. The power credit is also reduced. This is caused by the higher preheat demands due to increased combustion air rate, and lower flue gas heat content, as no moisture is associated with the char. Thus less heat is available for steam production.

The cost of char production is dependent on the cost of char. In this case the FMC figure of 10¢/MM BTU was used. However, even if the char were free the selling price would be 50.6¢/MSCF.

2. Hydrogen

Work has continued on the preparation of a flowsheet of a plant capable of producing hydrogen from bituminous coal.

B. Projections

1. Hydrogen

Calculations will continue on the hydrogen-from-coal plant.

2. Secondary Product Recovery

A study will be conducted to determine the economic incentives in recovering various by-products from the coal and coal ash. This study will include investigating:

- (a) Estimated markets in 1980
- (b) Products which can be recovered
- (c) Cost of recovery
- (d) Effect on pipeline gas economics of recovery.

3. Pre-Pilot Plant Unit

A coal combustion calorimeter will be designed in an effort to provide a better means of evaluating the efficiency of combustion in the melt. Since it is extremely important to the process economics that the heat of combustion of coal be transferred as efficiently as possible to the melt, it is felt that such a combustion system should be experimentally demonstrated before building the one-ton-per-hour pilot plant.



TABLE 2

INVESTMENT SUMMARY

PIPELINE GAS FROM LIGNITE AND CHAR

Basis: 250,000,000 SCFD of Pipeline Gas

	<u>Lignite I</u>	<u>Lignite II</u>	<u>Char</u>
Section 100:Coal Handling	\$ 8,550,000	\$ 8,270,000	\$ 5,830,000
Section 200:Gasification	76,860,000	49,560,000	63,100,000
Section 300:Shift Conversion	4,800,000	3,455,000	3,030,000
Section 400:Gas Purification	26,800,000	18,220,000	18,670,000
Section 500:Methanation	10,310,000	10,310,000	11,080,000
Section 600:Ash Removal	11,780,000	23,800,000	37,550,000
Section 1100:Offsites	<u>21,940,000</u>	<u>18,170,000</u>	<u>16,620,000</u>
 TOTAL BARE COST	 \$161,040,000	 \$131,785,000	 \$155,380,000
Contractor's Overhead and Interest	<u>28,260,000</u>	<u>23,115,000</u>	<u>27,420,000</u>
 TOTAL FIXED INVESTMENT	 \$189,300,000	 \$154,900,000	 \$183,300,000
 <u>Working Capital</u>			
30 Day Coal Supply	\$ 1,739,000	\$ 1,630,000	\$ 1,540,000
30 Day Carbonate Supply	569,000	512,000	908,000
30 Day Catalyst Makeup	101,000	79,000	84,000
Shift Catalyst Charge	608,000	391,000	344,000
Accounts Receivable	<u>4,590,000</u>	<u>4,380,000</u>	<u>5,460,000</u>
 WORKING CAPITAL	 <u>\$ 7,707,000</u>	 <u>\$ 6,992,000</u>	 <u>\$ 8,336,000</u>
 TOTAL CAPITAL INVESTMENT	 \$197,007,000	 \$161,892,000	 \$191,636,000



TABLE 3

GAS COSTS AND SELLING PRICES

PIPELINE GAS FROM LIGNITE AND CHAR

Basis: 250,000,000 SCFD of Pipeline Gas
90% Stream Efficiency

	<u>Lignite I</u>	<u>Lignite II</u>	<u>Char</u>
(Lignite \$1.50/ton			
Coal (Char 10¢/MM BTU	23.2	21.3	20.5
Sodium Carbonate	8.9	6.8	12.1
Miscellaneous Chemicals	0.5	0.2	0.2
Sponge Iron Makeup	0.1	0.1	0.1
Methanation Catalyst Makeup	0.8	0.8	0.9
Direct Labor	2.0	2.0	2.0
Power Credit	-15.8	-8.7	-5.9
Maintenance	5.9	4.8	5.7
Supplies	0.9	0.7	0.8
Supervision	0.2	0.2	0.2
Payroll Overhead	0.2	0.2	0.2
General Overhead	<u>4.5</u>	<u>3.9</u>	<u>4.4</u>
PLANT OPERATING COSTS	31.4	32.3	41.2
Depreciation	11.5	9.4	11.1
Local Taxes and Insurance	<u>6.9</u>	<u>5.6</u>	<u>6.7</u>
SUBTOTAL	49.8	47.3	59.0
Contingencies	<u>1.0</u>	<u>0.9</u>	<u>1.2</u>
TOTAL OPERATING COSTS	50.8	48.2	60.2
GAS SELLING PRICE	62.8	58.3	72.1



IV. MECHANICAL DEVELOPMENT

A. Accomplishments

1. Environmental Testing of High Temperature Materials

The gas cylinders for Corrosion Test #10 have been received and the test is in progress. The test is similar to Corrosion Test #9 except that the specimen support rack has been modified to minimize chances of the Monofrax A blocks being damaged by support failure.

2. Mechanical Characteristics Testing

The 5-3/4" reactor test facility has been completed. Equipment for the first scheduled tests on bed expansion has been installed and tests will proceed immediately. A series of tests on bed expansion of water, ethylene glycol and glycerine-water mixture (100 C.P. viscosity) have been completed in a 5-1/2" I.D. plastic tower. The data from these tests are being studied and will be compared and presented with the melt expansion data.

3. Melt Circulation

Studies of the "air-lift" type flow system continue. In addition to the air-water and air-glycerine water (100 C.P. viscosity) systems, an air-ethylene glycol system has been explored. Although results of this system are not as good as results obtained for the air-water system, a prediction of flow within -50 to +38 percent was made over a wide range of air rates and submergence. The predictions and experimental data were obtained for a 1-3/8" I.D. air lift tube 9 feet long.

Before any more substantial amount of work can be done in the circulation area it is obvious that additional physical property data are required for the melt material; i.e., viscosity, bed expansion vs. superficial velocity, and entrainment. The first two items will be covered under Part 2, Mechanical Characteristics Testing. The latter item, entrainment, will be covered in this part.

A plastic circulation model has been constructed to allow both circulation and entrainment tests. The system contains two vessels with two air-lift circulation pumps to provide cross circulation between the two vessels. Entrainment will be measured at various flow rates and bed expansion using two different aeration gases. Cross contamination will be measured by means of exit gas analysis.



B. Projections

1. Environmental Testing of High Temperature Materials

Corrosion Test #10 will continue to completion.

2. Mechanical Characteristics Testing

Bed expansion tests will be carried out with bed heights up to three feet and superficial velocities up to two feet per second, or higher where possible.

3. Melt Circulation

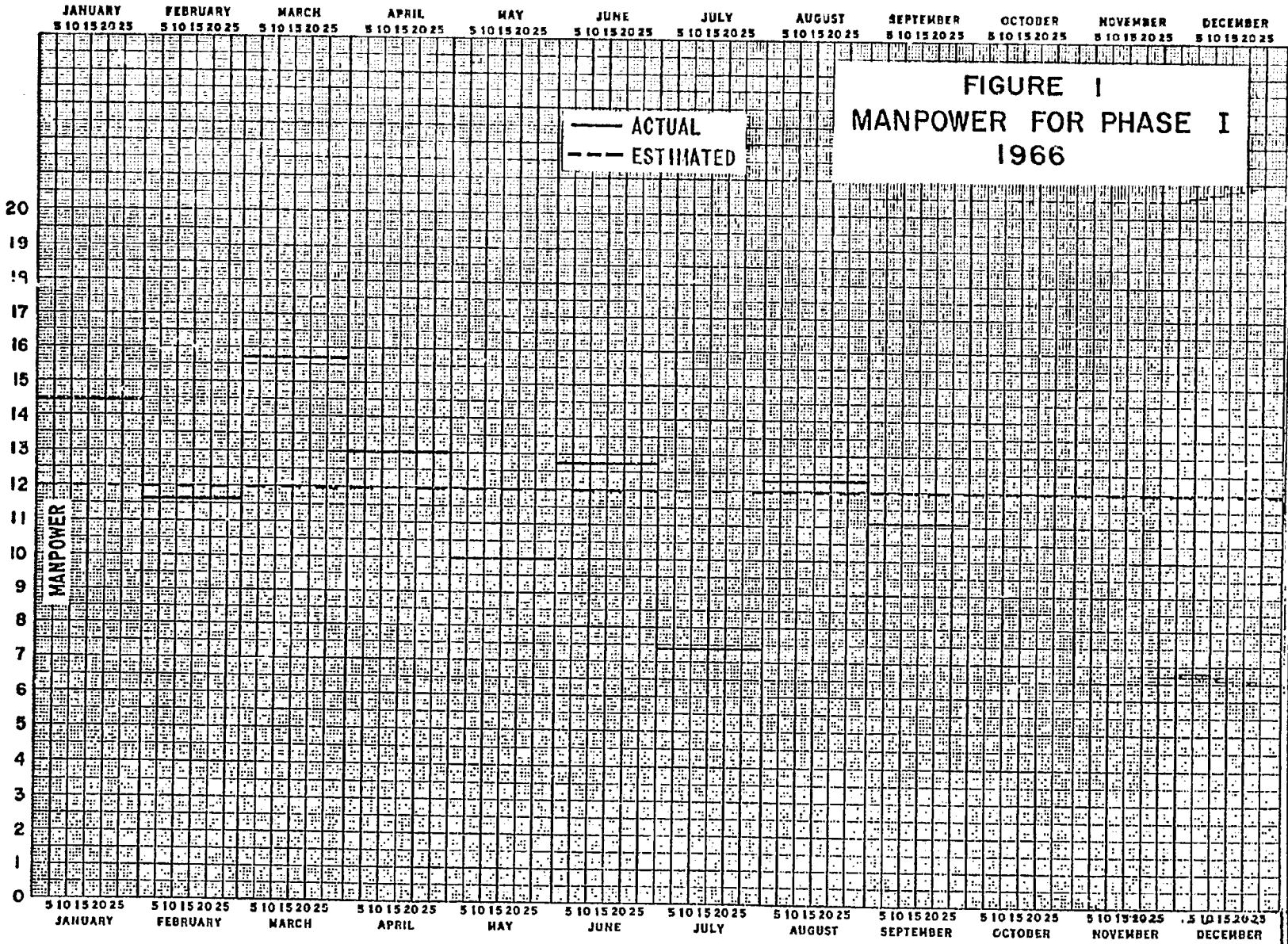
Work will continue on circulation and entrainment using the crossflow model.



V. MANPOWER AND COST ESTIMATES

Figure 1 shows the projected breakdown for Phase I for 1966 as well as the actual effort that was made. It can be seen that an 11.1 man-effort was made during September.

Figure 2 shows the expenditures during September. For the month, \$19,201 was expended, not including fee and G & A. The total expenditures through September were \$507,273. Including fee and G & A the total expenditures were \$579,830. This is 53% of the encumbered funds.



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