

MWR-MPR-24

RESEARCH AND DEVELOPMENT DEPARTMENT



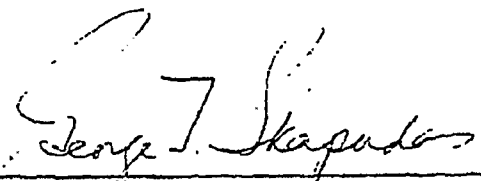
DEVELOPMENT OF KELLOGG COAL GASIFICATION PROCESS

Contract No. 14-01-0001-380

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Progress Report No. 24

APPROVED:

  
Project Manager

  
DIRECTOR

  
MANAGER

RESEARCH & DEVELOPMENT DEPARTMENT

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



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### I. SUMMARY

This progress report is the twenty-fourth 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.

Gasification experiments have been continued in the two-inch-diameter test reactor at three atmospheres pressure to study the effect on gasification rate of feedstock material, ash content of the melt, and hydrogen content of the feed gas.

Gasification data obtained for Renners Cove lignite and a char obtained from FMC (produced from Illinois No. 6 coal) indicated rates about 2.3 and 2.8 times as great as anthracite, respectively. It is felt that the high rate observed for the char is due to its high surface area (80 square meters per gram).

One new run was made to determine the effect of ash level on rate. At 0.5 feet per second superficial gas velocity and with 4 percent ash, a rate was obtained in good agreement with that which was interpolated from previous data.

Four additional runs were made to determine the effect on rate of having hydrogen present in the steam feed. With about 50 percent hydrogen in the steam, there is no detectable change in rate from that obtained with a steam-nitrogen mixture.

Estimated capital investment and operating costs were calculated for a plant capable of producing 250,000,000 SCFD of pipeline gas from anthracite. Estimated capital investment for the plant is about \$169,000,000. Gas selling price is about 90¢/MSCF, based on \$8 per ton anthracite and the OCR standard procedure for calculating gas cost. On the other hand, if large amounts of anthracite could be found in deposits such that it could be provided to the process at \$4 per ton and if process improvements were made to handle the increased ash content of this coal, gas selling price could be reduced to the range of 55 to 60¢/MSCF.

Process calculations were made for the design of a pre-pilot plant reactor which would evaluate the proposed method of melt circulation as well as obtain rate data at high bed depths. This design has been turned over to the Design Engineering Section for its review as well as an estimate of the unit's cost.

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The circulation system for studying flow parameters of a simulated melt material was completed. In addition, the 5 3/4-inch reactor test facility was essentially completed with only a small amount of electrical work remaining to be done.



## II. PROCESS RESEARCH

### A. Accomplishments

Gasification runs using FMC char and Renners Cove lignite completed the study of a diverse range of feedstocks for the Kellogg process. In addition, one run was made to complete the study of the effect of ash on gasification rate and four runs were made to determine if hydrogen had any inhibiting effect on the rate of gasification. These runs are presented in Table I and discussed below.

#### 1. Feedstock Evaluation

The results of all feedstock evaluations have been summarized on Figure 1 which is a standard Arrhenius-type plot of the data. Renners Cove lignite and FMC char derived from Illinois No. 6 coal were added to the figure presented in the last summary.

The data for Renners Cove lignite are in the same region as the South Beulah lignite, another North Dakota lignite. However, the slope of this line is quite different from that of the other feedstocks. Unfortunately, we have run out of this material and cannot check the results. FMC char turned out to be the most reactive feedstock tested. Certainly part of this reactivity appears due to its relatively high surface area compared to the other substances. According to the data received from FMC, this char had a surface area of 80 square meters per gram.

The following tabulation shows the relative average reactivity of the various feedstocks compared with a reactivity of unity for anthracite. Also shown are the apparent activation energies in the temperature range of test, 1640-1740 F:

<u>Feedstock</u>	<u>Relative Gasification Rate</u>	<u>E<sub>a</sub></u>
Anthracite	1	28
Bituminous Coke	1	34
Oxidized Bituminous Coal	1.5	27
So. Beulah Lignite	1.9	19
Elkol Subbituminous Coal	2.4	14
Renners Cove Lignite	2.3	5
FMC Char	2.8	9



Once again it must be mentioned that the gasification rates are conservative since they represent the reactivity of only the fixed carbon in each feedstock. Under the test procedure of feeding the coal into an inert atmosphere on top of the molten salt bed, an appreciable amount of carbon comes off as gas and tar which do not contribute to the above rates of gasification. Under continuous feeding of the fuel into the bottom of the molten salt bed, most, if not all, of this presently ignored carbon will be gasified and will result in an increase of the rates established above.

### 2. Completion of Ash Effect

Run number 34 was made to get one additional point on the 0.5 FPS curve presented in Figure 1 in Progress Report No. 22. Since this point falls on the curve no additional comments are necessary at this time.

### 3. Effect of Hydrogen Pressure

Although early work had established that hydrogen had no effect on the rate of gasification, it was decided to reinvestigate this since the early work was done at one atmosphere total pressure with 30% steam in hydrogen and with bituminous coal as feed. Recent work has shown that this coal could have agglomerated thus possibly clouding the results. In addition, it is now possible to test at super-atmospheric pressure. The last four runs in Table I were made at 5 atmospheres total pressure using 50% hydrogen in steam for two runs and 50% nitrogen in steam for two runs as the basis for comparison.

The first two runs used lightly oxidized bituminous coal. This oxidation of the 12/20 mesh particles was done in an oven at less than 300° F for a period of about 10 days. The gasification rates were 26 and 32 lbs. C gasified/hr./CF for the hydrogen and nitrogen runs, respectively. Although this difference appeared significant, it was confounded by the possibility of non-repeatable agglomeration (still possible with the oxidized coal). In addition the fast rate of reaction made rate interpretation more difficult. Consequently, two runs were now made with non-agglomerating anthracite at a lower temperature to increase the accuracy of determining the rate.

The last two runs, now on a firm basis for establishing the effect of hydrogen, gave 3 lbs. C gasified/hr./CF for the rate of gasification both in hydrogen and in nitrogen. Thus, once again it can be concluded that hydrogen did not affect the rate of gasification under the conditions employed with the molten salt system.

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B. Projections

Evaluation of the effect of steam pressure to 10 atmospheres at 1640°F will be made on the rate of gasification of bituminous coke. This is intended to firm up our pressure effect data. Additional consideration will be given to the inhibiting effect of product gases. Following a cleanup of all our gasification work a re-evaluation of combustion kinetics will be made.

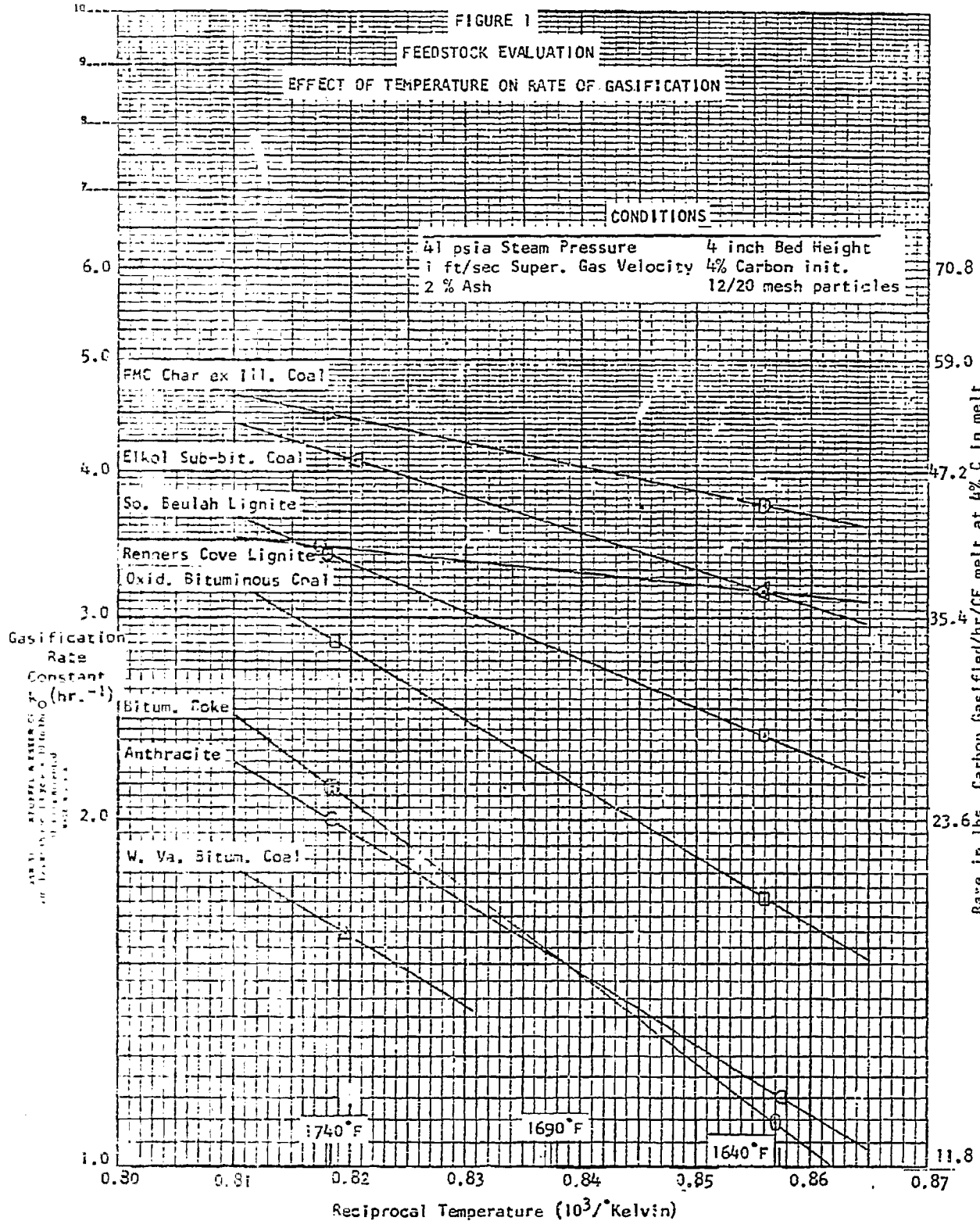
INDEX 1  
GASIFICATION RUNS IN MOLTEN SODIUM CARBONATE (1)

Run No. H- Date 1966	34 6/29	35 7/1	36 7/1	37 7/6	38 7/7	39 7/11	40 7/11	41 7/13	42 7/14
Feed	Bit. Coke VI	Renner's Cove	Lignite	FNC Char ex	Ill. Coal	Oxid. Bit. Coal(4)		Anthracite	
% Fixed Carbon	-	42.5	42.5	76.4	76.4	58.3	58.3	-	-
% Total Carbon	93.2	66.4	66.4	76.8	76.8	81.3	81.3	80.8	80.8
% Vol. Matter	0.6	48.2	48.2	3.5	3.5	37.3	37.3	5.9	5.9
% Ash	6.2	9.3	9.3	20.1	20.1	3.9	3.9	11.7	11.7
gms. charged mesh size	19	40.6	40.6	22.6	22.6	29.6	29.6	21.4	21.4
$\text{Na}_2\text{CO}_3$ - gms	(2)	405.7	(2)	405.7	(2)	405.7	(2)	405.7	(2)
Ash - gms	-	8.3	-	8.3	-	8.3	-	8.3	-
- % in melt	4	2	2.9	2	3	?	2	2	2.6
% C in melt Initial(3)	4	4	4	4	4	4	4	4	4
Bed Height - inches	4	4	4	4	4	4	4	4	4
Conditions									
Temp. - F avg.	1740	1641	1743	1641	1741	1744	1741	1645	1642
Pressure - psia	44.8	45.2	45.0	45.2	44.7	45.2	45.2	45.3	45.3
Steam Pressure-psia	40.4	41.0	41.1	41.5	41.1	24.5	25.9	21.8	24.3
Gas in Steam	←-----N <sub>2</sub> -----→					H <sub>2</sub>	N <sub>2</sub>	H <sub>2</sub>	N <sub>2</sub>
Ft./sec. Steam & Gas	0.51	0.98	1.02	0.97	1.00	0.90	0.90	1.03	0.93
Run Time- min.	40	20	25	25	20	35	25	40	40
cc H <sub>2</sub> O in/hr	589	1194	1193	1196	1195	677	662	667	667
cc Gas in/min	1348	2542	2340	2192	2200	9830	10,420	14,870	11,970
Results									
% C Devolatilized	1.0	8.6	6.8	5.1	6.4	8.1	4.7	12.5	1.7
% C to tar and loss	-	26.6	26.9	1.5	-	20.0	19.3	-	2.4
% C to oxides + CH <sub>4</sub>	102	64.8	66.3	93.4	94.0	71.9	76.0	87.9	95.9
% C to oxides - basis fixed C	105	101	103	98	100	100	106	100	96
Gasif. Rate Const-hr <sup>-1</sup>									
k <sub>1</sub> - Input	1.82	3.21	3.67	3.67	4.50	2.22	3.05	1.12	1.02
k <sub>0</sub> - output	1.73	3.16	3.46	3.75	4.50	2.22	2.73	1.12	1.07
Rate - at 4% C in bed									
lbs C/hr/CF melt	20	37	41	44	53	26	32	13	13
Salt Carryover -gms	14.5	4.5	8.9	3.7	3.5	5.6	12.3	27.1	14.2

- (1) Used 2-inch ID Inconel reactor. Bituminous coke VI made at 950°C.  
 (2) Reused melt from previous run.  
 (3) Based on fixed carbon.  
 (4) Approx. 10 days oxidation in air at 250-300°F.









### III. CHEMICAL ENGINEERING STUDIES AND DEVELOPMENT

#### A. Accomplishments

##### 1. Anthracite Flowsheet

A flowsheet for the production of 250,000,000 SCFD of pipeline gas from anthracite coal has been completed. The processing sequence is the same as that described in Progress Report No. 21 for bituminous coal and so will not be repeated here. The only major difference in the present flowsheet is that there are ten parallel gasification and ash removal trains instead of nine. This increase in number of trains is due to an increase in gas rate in both gasifier and combustor (the number of ash removal trains is increased to equal the number of gasification trains so each gasifier has its own ash removal equipment). The increased gas rate in the gasification section is caused by the liberation of larger amounts of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  (from decomposition of recycle  $\text{NaHCO}_3$ ) than in the case of bituminous coal. Recycle  $\text{NaHCO}_3$ , in turn, is larger because the feed anthracite contains 8.9 percent ash while the bituminous contained only 5.1 percent. The heat to decompose this increased amount of  $\text{NaHCO}_3$  must, of course, be supplied by burning additional coal in the combustor. The increased flow of combustion air (and flue gas) necessitate a bigger combustor, and the net effect is an additional train of equipment.

Estimated capital investment for the case of pipeline gas from anthracite is presented in Table II. As before, shift catalyst and activated carbon have been included in fixed investment because of their long lifetimes. As can be seen, total capital investment is estimated to be about \$169,000,000. This is about twenty percent higher than that for the bituminous plant due to increases in the costs of Sections 200, 600 and 400, Gasification, Ash Removal and Gas Purification. The increased cost of Section 200 is due to the aforementioned increase in number of gasifiers. The cost of ash removal is increased due to the larger amount of ash which must be removed in this section. The gas purification investment increase is the result of the additional  $\text{CO}_2$  released by  $\text{NaHCO}_3$  decomposition which must be removed.

Since essentially all of the investment increase can be traced to the increased ash content of the coal, it is important to consider any schemes which could lessen the impact of this effect. For example, if the ash level in the melt were increased from 8 to 14 percent (corresponding to an ash level in coal increase from 5.1 to 8.9 percent), the amount of  $\text{Na}_2\text{CO}_3$  fed to ash removal would be the same as for the case of bituminous. This would also result in the same amount of  $\text{NaHCO}_3$



being recycled to the gasifier and the increased gasifier heat requirements would be largely eliminated. This, however, would increase melt viscosity quite a bit, and operation at this ash level would have to be checked experimentally.

Another possible means of reducing the gasifier heat load (with its associated investment increases) would be to decompose the recycle  $\text{NaHCO}_3$  before feeding it to the gasifiers. For example, it could be passed through a kiln using relatively low level heat (as opposed to the very expensive high level gasifier heat) to decompose it. The economic potential of this scheme will be evaluated since it could also be used in the bituminous case with possible cost savings.

Estimated annual operating expenses and gas selling price are summarized in Table III. Gas manufacturing cost, with anthracite at \$8 per ton, is calculated to be about 77¢/MSCF, and gas selling price to yield an average 9.4 percent return on equity capital is about 90¢/MSCF. The increases in operating costs over those for bituminous are due to three main causes:

- a. Increase in fixed charges
- b. Increase in coal cost
- c. Increase in sodium carbonate makeup

The reasons for the increase in fixed charges (a reflection of an investment increase) have already been given. The increase in coal cost was due first to the fact that anthracite is a more expensive feed material than bituminous and second because additional coal had to be burned to satisfy the increased process heat demand. The increase in sodium carbonate makeup is due to the increased ash content of anthracite requiring that larger amounts of melt be circulated to the ash removal section with a correspondingly larger loss of  $\text{Na}_2\text{CO}_3$  incurred.

If one of the aforementioned schemes for reducing the gasifier heat requirements could be applied (i.e., increasing ash level or externally decomposing  $\text{NaHCO}_3$ ), it might be possible to decrease fixed charges and  $\text{Na}_2\text{CO}_3$  make-up by as much as 5¢/MSCF. This would result in a gas selling price of about 85¢/MSCF--still economically rather unattractive. It is, therefore, obvious that to make pipeline gas at attractive prices the cost of anthracite must be reduced from the typical \$8 per ton price used in calculating the economics. If, for example, large anthracite deposits could be found capable of yielding coal at a cost of about \$4 per ton, pipeline gas could be produced and sold at a price in the range of 55 to 60¢/MSCF.

## 2. Pre-pilot Gasification Unit

Preliminary process calculations have been completed for a pre-pilot plant reactor which would evaluate the proposed method of melt circulation by density difference. In addition, the unit would be capable of operating with melt depths of



the order of four feet so that the effect of high melt heights on gasification rate could be evaluated. The proposed unit is a 6-inch O. D. vessel which has six 1-1/4-inch O. D. tubes around it. Steam would be fed to the bottom of the reactor causing a density difference between the melt in the vessel and in the tubes thereby inducing melt circulation. The entire vessel would be placed inside a gas-fired furnace where the heat for gasification would be supplied through the tube walls. The preliminary design of this vessel has been turned over to the Design Engineering Section for a review of the mechanical design and for an estimate of its cost.

### 3. Hydrogen Flowsheet

Work on the preparation of a flowsheet for a plant capable of producing hydrogen from bituminous coal was continued. Material balances for the entire process have been essentially completed.

## 8. Projections

### 1. Pipeline Gas

Investment and operating costs will be determined for a plant capable of producing pipeline gas from lignite.

### 2. Hydrogen

Preparation of the "process package" for the hydrogen-from-coal plant will continue. Energy balance calculations will be made and the sizing of process equipment will be begun.



TABLE II

INVESTMENT SUMMARY

PIPELINE GAS FROM ANTHRACITE

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

<u>Title</u>	<u>Bare Cost</u>
100: Coal Storage and Preparation	\$ 4,010,000
200: Gasification	66,950,000
300: Shift Conversion	5,390,000
400: Gas Purification	23,200,000
500: Methanation and Product Gas Compression	10,300,000
600: Ash Removal	6,520,000
1100: Offsite Facilities	<u>17,250,000</u>
Total Bare Cost	\$133,620,000
Interest During Construction and Contractor's Overhead and Profit	<u>23,520,000</u>
TOTAL FIXED INVESTMENT	\$157,140,000
<u>Working Capital</u>	
30 Day Coal Inventory	\$ 4,000,000
30 Day Carbonate Inventory	331,000
30 Day Catalyst Inventory	79,000
Catalyst Charges	649,000
Accounts Receivable	<u>7,141,000</u>
Total Working Capital	\$ 12,200,000
TOTAL CAPITAL INVESTMENT	\$169,340,000



TABLE III

ESTIMATED ANNUAL OPERATING COST

AND GAS SELLING PRICE

PIPELINE GAS FROM ANTHRACITE

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

<u>Item</u>	<u>\$/Year</u>	<u>c/MSCF</u>
Anthracite Coal at \$8/ton	\$43,950,000	53.3
Sodium Carbonate at 1.55¢/lb.	3,643,000	4.4
Miscellaneous Chemicals	258,000	0.3
Sponge Iron Makeup	39,000	0.05
Methanation Catalyst Makeup	685,000	0.8
Direct Labor at \$3.20/man-hour	1,777,000	2.2
Power Credit at 5 mills/kwh	-7,500,000	-9.1
Maintenance at 3% of Bare Cost	4,010,000	4.8
Supplies at 15% of Maintenance	602,000	0.7
Supervision at 10% of Direct Labor	178,000	0.2
Payroll at 10% of Direct Labor + Supervision	196,000	0.2
General Overhead at 50% of Maintenance + Labor + Supervision + Supplies	<u>3,280,000</u>	<u>4.0</u>
Plant Operating Expenses	\$51,118,000	61.8
Depreciation at 5% of Fixed Investment	7,860,000	9.5
Local Taxes and Insurance at 3% of Fixed Investment	<u>4,720,000</u>	<u>5.7</u>
Subtotal	\$63,698,000	77.0
Contingencies at 2%	<u>1,270,000</u>	<u>1.5</u>
<b>TOTAL OPERATING EXPENSE</b>	<b>\$64,968,000</b>	<b>78.5</b>
<b>20 YEAR AVERAGE REVENUE REQUIREMENT</b>	<b>\$74,050,000</b>	
<b>GAS SELLING PRICE</b>		<b>89.9¢/MSCF</b>



#### IV. MECHANICAL DEVELOPMENT

##### A. Accomplishments

###### 1. Environmental Testing of High Temperature Materials

New samples are being prepared and the test facilities made ready to start Corrosion Test #10 using a simulated gasification condition.

###### 2. Mechanical Characteristics Testing

The circulation system for studying flow parameters of a simulated melt material is now complete, and initial testing has just begun. The "air lift" type flow system is designed to allow study of flows over a wide range of operating conditions.

The 5-3/4" reactor test facility is nearly complete, the control wiring and final connection to electrical power is all that remains to be done.

##### B. Projections

###### 1. Environmental Testing of High Temperature Materials

Simulated Gasification Corrosion Test #10 will begin as soon as possible.

###### 2. Mechanical Characteristics Testing

The test facility will be put to use in checking the predicted flow of simulated melt materials under a range of flow conditions.

Installation of the final wiring on the 5-3/4" reactor will continue and testing should begin in the near future.

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#### V. MANPOWER AND COST ESTIMATES

Figure 2 shows the projected breakdown for Phase I for 1966 as well as the actual effort that was made. It can be seen that a 7.4 man-effort was made during July.

Figure 3 shows the expenditures during July. For the month \$16,439 was expended, not including fee and G & A. The total expenditures through July were \$466,863. Including fee and G & A the total expenditures were \$534,012. This is 49% of the encumbered funds.



