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AN ANALYSIS OF COAL HYDROGASIFICATION PROCESSES

Monthly Technical Progress Report for the Period 1 October - 31 October 1977

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ABSTRACT

This monthly Technical Progress Report covers work performed during the period 1 October 1977 to 31 October 1977 for a program entitled "An Analysis of Coal Hydrogasification Processes." This program is being performed in four sequential tasks: Task I — Data Collection; Task II — Data Analysis; Task III — Process Modeling; and Task IV — Identification of Additional Data and Recommended Experimental Programs.

During October, substantial progress was made on Tasks I, II, and III. Data from three recent Rocketdyne tests using subbituminous coal and a recent Rocketdyne test using bituminous coal were entered into the computerized data base. Also, data from five recent Cities Service tests using subbituminous coal were entered into the data base. The correlation previously developed for predicting carbon conversion, based on Cities Service subbituminous tests, gave results that were in reasonable agreement with the measured conversions for the recently completed Rocketdyne subbituminous tests. This indicates that the Cities Service and Rocketdyne reactors behave similarly for the same coal. The measured value of carbon conversion for the Rocketdyne test using bituminous coal was much higher than the value predicted from the correlation based on subbituminous coal. This was expected, since previous Rocketdyne bituminous coal data have shown higher levels of carbon conversion than recent data with subbituminous coal under similar operating conditions. A correlation for predicting reactor hydrogasification efficiency was fitted to the Cities Service subbituminous data.

Operating variable levels and size constraints were chosen for the design of a conceptual full-scale hydrogasification reactor. These levels and constraints were based on data gathered in the Cities Service and Rocketdyne reactors using subbituminous coal, together with

predictive reactor performance models fitted to the data. A conceptual design was presented for a full-scale reactor facility which consists primarily of a hydrogasification stage to produce methane-rich product gas from the coal, and a steam/oxygen stage to produce hydrogen-rich product gas from the unreacted char.

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Section 1

OBJECTIVES AND SCOPE

This report is the October Monthly Technical Progress Report for a program entitled, "An Analysis of Coal Hydrogasification Processes." The program is being performed for ERDA by Bechtel Corporation under ERDA Contract No. EF-77-A-01-2565. Work on this program was initiated on February 1, 1977.

The major objective of the program is "to conduct an analytical study which will investigate the operability potential and scaleup feasibility of the Cities Service, Rocketdyne, and ERDA Pittsburgh Energy Research Center (PERC) coal hydrogasification processes, relative to ERDA plans for a Hydrane process development unit (PDU)." To accomplish the objective, four sequential program tasks have been established.

The primary objective of Task I is to conduct a survey of information in the public domain relative to the above three processes. This survey is to be supplemented with visits to the process contractors for discussion, expansion, and updating.

The primary objective of Task II is to perform a detailed analysis of the data, as required to evaluate the information for a pilot plant application. Consideration will be given to reactor heat and mass balances, reaction kinetics, actual or predicted data on the product gas yield and composition, and all other relevant factors. In addition, conceptual designs, where available, will be analyzed for potential operational problems and scaling.

Task III has two primary objectives: (1) to perform reactor model studies, where available data permit, for each of the three processes; and (2) to generate a conceptual, full-scale, optimum reactor design in consultation with ERDA. The reactor model study will attempt to predict, where possible, overall carbon conversion, carbon selectivity to gas, and carbon selectivity to methane and ethane for the three processes. In conjunction with the modeling study, a sensitivity analysis will be performed that will determine the influence of the degree of uncertainty of the basic information used in the prediction of reactor performance.

The primary objectives of Task IV are to: (1) identify critical data gaps and point out specific data that are missing and are required for reliable pilot plant design; (2) recommend experiments to acquire the necessary data, and estimate the number of experiments and man-hours to obtain these data; and (3) assess the impact on the Hydrane process design phase, in case the necessary data cannot be experimentally determined.

Section 2

PROGRESS SUMMARY AND OPEN ITEMS

2.1 PROGRESS SUMMARY

Figure 2-1 summarizes the program progress between February 1, 1977 (the program start date) and October 31, 1977. During October, substantial progress was made on Tasks I, II, and III. Actual manhours expended in September were 610; budgeted manhours were 700. As can be seen in Figure 2-1, actual manhours expended are less than planned, while program progress is on schedule.

2.2 OPEN ITEMS

As presently scheduled, the completed results from the Cities Service and Rocketdyne ERDA hydrogasification test programs will not be available for analysis until about the end of January 1978. Accordingly, Bechtel will not be able to incorporate into its program the wide range of data needed to effectively perform Tasks III and IV within the present program schedule (see Figure 2-1). Bechtel recommends, therefore, that the period of performance of the program be extended to reflect the delay in the acquisition of Cities Service and Rocketdyne hydrogasification data.

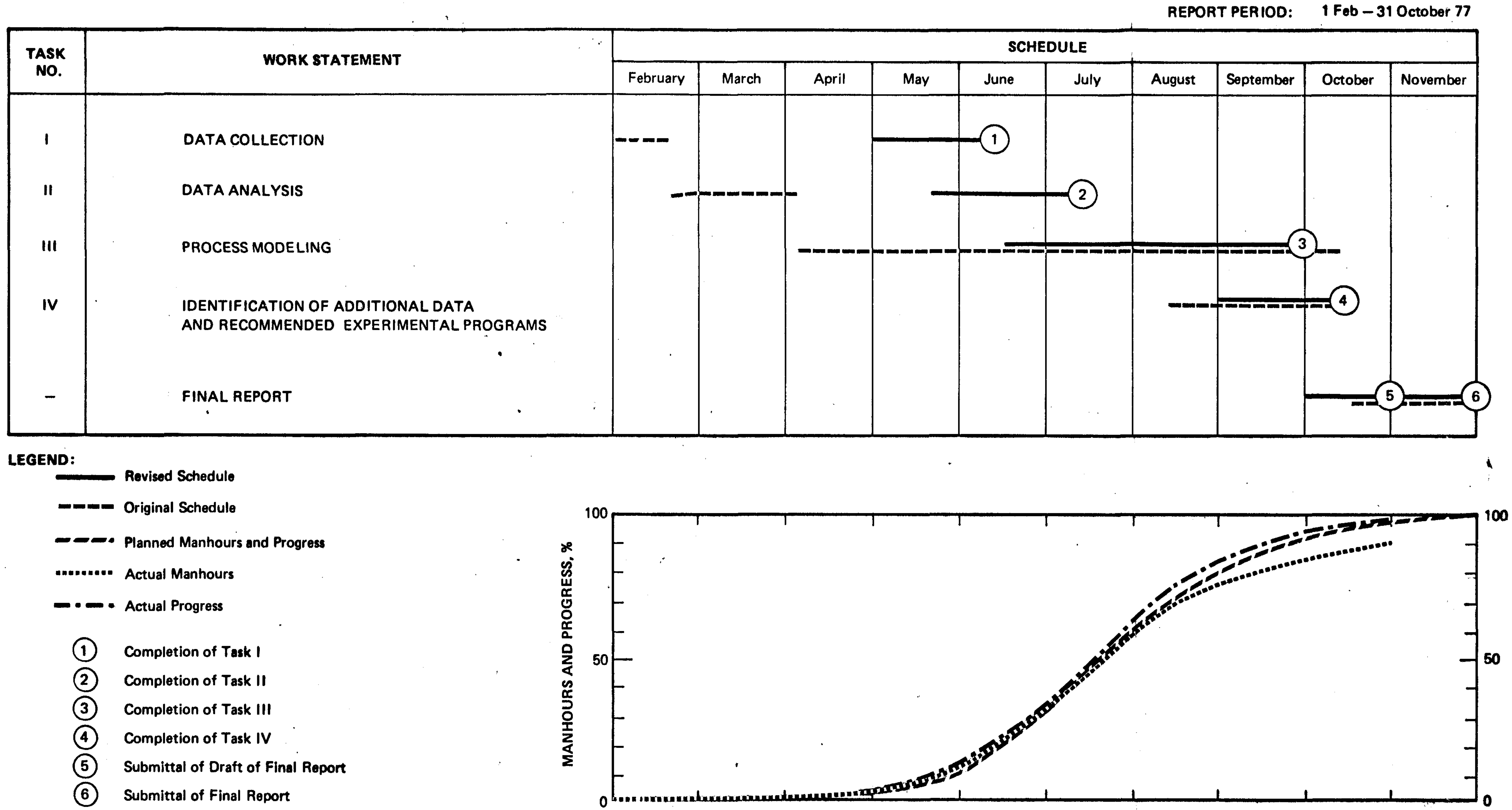

Figure 2-1. Progress and Performance Chart

Section 3

TECHNICAL PROGRESS

This section describes the technical progress for Tasks I, II, and III during the reporting period.

3.1 TASKS I AND II — ROCKETDYNE DATA COLLECTION AND ANALYSIS

During this reporting period, Bechtel reviewed data from Rocketdyne[1] for five recently completed hydrogasification tests conducted in the Rocketdyne 1/4-ton/hr reactor test facility. Three of the tests used Montana Rosebud subbituminous coal and two of the tests used Western Kentucky bituminous (HvAb) coal.

The data were entered into the computerized data base containing data from 11 previous partial liquefaction tests generated in the Rocketdyne 1-ton/hr reactor facility using the Kentucky HvAb coal. A computer listing of all the data is presented in Table 3-1.

The recent Rocketdyne hydrogasification data were generated in an entrained-downflow tubular reactor, 1.88 inches in diameter and 15 feet in length. All five tests (see Table 3-1) were made at reactor pressures of approximately 1,000 psig, gas (or particle) residence times between 520 and 550 milliseconds, and gas outlet temperatures ranging from 1,475°F to 1,900°F (1,935°R to 2,360°R). Overall carbon conversions for the subbituminous tests were between 32 and 44 percent, and carbon selectivities to gaseous products were between 60 and 87 percent. The overall carbon conversions for subbituminous coal were in substantial agreement with the Cities Service bench-scale test results with the same subbituminous coal at comparable operating conditions (see Subsection 3.4 of this report).

Table 3-1

ROCKETDYNE HYDROGASIFICATION DATA

RUN DESIG-NATION	DATE	COAL TYPE	REACTOR	OVERALL FRACTION CARBON CONVERTED	FRACTION SELEC-TIVITY TO GAS	OUTLET GAS TEMP (DEG R)	HYDROGEN PARTIAL PRESSURE (PSIG)	RESI-DENCE TIME (MILLISEC)	HYDROGEN TO COAL RATIO (LB/LB)
5	1/31/77	HVAB	1/4 TPH	.382		1800.	1000.	155.	.250
6	2/ 3/77	HVAB	1/4 TPH	.542	0.397	2160.	1000.	130.	.478
7	2/ 7/77	HVAB	1/4 TPH	.615	0.483	2370.	1000.	120.	.775
8	2/17/77	HVAB	1/4 TPH	.596	0.485	2160.	1000.	270.	.365
9	2/22/77	HVAB	1/4 TPH	.645	0.760	2260.	1500.	410.	.365
10	3/ 1/77	HVAB	1/4 TPH	.609	0.782	2050.	1500.	490.	.314
11	3/ 4/77	HVAB	1/4 TPH	.627	1.000	2060.	1500.	630.	.344
12	3/ 9/77	HVAB	1/4 TPH	.576	0.672	2060.	1000.	430.	.333
13	3/23/77	HVAB	1/4 TPH	.538	0.348	2160.	1000.	60.	.292
14	3/25/77	HVAB	1/4 TPH	.570	0.507	2070.	1500.	100.	.397
15	3/29/77	HVAB	1/4 TPH	.526	0.382	2160.	700.	45.	.403
011- 7	9/21/77	HVAB	1 TPH	.520		2130.	1003.	550.	.386
011- 8	9/29/77	HVAB	1 TPH			2270.	1007.	550.	.391
011- 2	8/30/77	SUBBTM	1 TPH	.319	0.596	1930.	1021.	540.	.617
011- 4	9/ 9/77	SUBBTM	1 TPH	.435	0.874	2360.	987.	520.	.691
011- 5	9/15/77	SUBBTM	1 TPH	.365	0.822	2190.	995.	550.	.411

The tests using Kentucky HvAb coal (Runs 011-7 and 011-8) are being conducted in order to determine whether results in the 1/4 ton/hr reactor assembly can duplicate earlier test results obtained with the HvAb coal in the 1-ton/hr reactor assembly. There are not, however, sufficient data to permit any conclusions at this time. Nonetheless, it should be noted that the carbon conversion of 52 percent reported for HvAb Run 011-7 is much higher than the carbon conversion of 37 percent reported for subbituminous Run 011-5, which was conducted under similar operating conditions.

Rocketdyne has not yet reported the product gas analyses and material balances for the recent tests. These data will be presented and discussed in future Bechtel reports as they are received.

3.2 TASKS I AND II — CITIES SERVICE DATA COLLECTION AND ANALYSIS

During this reporting period, Bechtel received additional data for five recently completed Cities Service hydrogasification tests using Montana Rosebud subbituminous coal.[1] The data were entered into the computerized data base containing data from 14 earlier completed subbituminous tests. A computer listing of all the subbituminous data is presented in Table 3-2.

The recently acquired Cities Service data (Runs MR-2, 3, 16, 17, and 18) showed overall carbon conversions ranging from 33 to 43 percent at gas temperatures from 1,520°F to 1,710°F (1,980°R to 2,170°R), hydrogen partial pressures from 500 to 1,500 psig, and gas residence times from 312 to 656 milliseconds (see Table 3-2). The highest methane selectivity and yield were obtained in Run MR-18; carbon selectivity to methane was 39 percent and carbon conversion to methane was 17 percent.

Actual carbon mass balance closures ranging from 97 to 110 percent and ash balance closures ranging from 85 to 92 percent were reported for the recent five Cities Service runs.[1]

Table 3-2

CITIES SERVICE
HYDROGASIFICATION DATA

RUN DESIGN-NATION	DATE	COAL TYPE	REAC-TOR	OVERALL FRACTION CARBON CONVERTED	CARBON SELEC-TIVITY TO METHANE	CARBON SELEC-TIVITY TO ETHANE	CARBON SELEC-TIVITY TO C1-C5 GAS	MAXIMUM GAS TEMP (DEG R)	HYDROGEN PARTIAL PRESSURE (PSIG)	GAS VELOCITY (FT/SEC)	GAS RESI-DENCE TIME (MSEC)	PARTICLE RESI-DENCE TIME (MSEC)	HYDROGEN TO COAL RATIO (LB/LB)	MEAN PARTICLE SIZE (MICRONS)
MR- 4	6/13/77	SUBBTM	EF	.390				1970.	500.	20.90	1521.	1521.	1.40	45.
MR- 1	6/16/77	SUBBTM	EF	.319	.295	.238	.621	1960.	500.	9.60	416.	416.	0.76	45.
MR-10	6/22/77	SUBBTM	EF	.186	.210	.172	.489	1960.	1500.	9.60	417.	417.	0.83	45.
MR-13	6/27/77	SUBBTM	EF	.390	.372	.213	.587	1990.	1500.	16.70	1086.	1086.	0.80	45.
MR-14	6/29/77	SUBBTM	EF	.421	.435	.166	.603	2090.	1500.	17.00	1060.	1060.	0.74	45.
MR-28	7/ 6/77	SUBBTM	EF	.262	.260	.214	.569	2010.	1000.	13.30	295.	295.	0.79	45.
MR-29	7/ 8/77	SUBBTM	EF	.344	.340	.235	.596	2100.	1000.	13.30	297.	297.	0.99	45.
MR-30	7/12/77	SUBBTM	EF	.324	.401	.204	.611	2180.	1000.	12.80	307.	307.	0.85	45.
MR-11	7/15/77	SUBBTM	EF	.255	.306	.224	.557	2070.	1500.	13.20	299.	299.	0.78	56.
MR-12	7/19/77	SUBBTM	EF	.321	.321	.212	.561	2130.	1500.	13.00	304.	304.	0.75	56.
MR-25	7/21/77	SUBBTM	EF	.359	.331	.234	.568	1980.	1000.	16.70	1081.	1081.	0.98	56.
MR-26	7/25/77	SUBBTM	EF	.382	.458	.170	.628	2080.	1000.	16.70	1078.	1078.	0.88	56.
MR-27	7/27/77	SUBBTM	EF	.402	.585	.057	.642	2160.	1000.	16.60	1085.	1085.	0.93	56.
MR-15	7/29/77	SUBBTM	EF	.453	.541	.102	.642	2120.	1500.	15.30	1175.	1175.	0.87	56.
MR- 2	8/ 3/77	SUBBTM	EF	.339	.327	.212	.546	2070.	500.	29.80	313.	313.	0.89	56.
MR- 3	8/ 5/77	SUBBTM	EF	.330	.352	.109	.461	2170.	500.	29.90	312.	312.	0.97	56.
MR-16	8/ 8/77	SUBBTM	EF	.379	.256	.172	.433	1980.	1500.	14.30	654.	654.	0.91	56.
MR-17	8/10/77	SUBBTM	EF	.430	.319	.153	.472	2070.	1500.	14.30	651.	651.	1.24	56.
MR-18	8/12/77	SUBBTM	EF	.430	.388	.158	.547	2110.	1500.	14.20	656.	656.	0.93	56.

3.3 TASK III — CITIES SERVICE REACTOR MODELING

Bechtel has developed semiempirical models to correlate the previously acquired Cities Service subbituminous data (Runs MR-4 through MR-15 in Table 3-2). Correlations have been presented in Bechtel's September Progress Report[2] for predicting overall carbon conversion and carbon selectivities to methane, ethane, and hydrocarbon gas. In this subsection, these correlations will be used to compare predicted and measured values of conversions and selectivities for the recently acquired data (Runs MR-2, 3, 16, 17, and 18 in Table 3-2). When additional subbituminous data become available during the next reporting periods, the models proposed by Bechtel will be refitted to all of the subbituminous data. In addition, a correlation for predicting reactor hydrogasification efficiency, which has been fitted to the Cities Service subbituminous data, is presented.

3.3.1 Overall Carbon Conversion

Equation 2 of Bechtel's September Progress Report[2] has been used to predict carbon conversions for the five new subbituminous runs. The predicted and measured conversions for the five new runs are shown in Figure 3-1, together with the measured and predicted conversions for the previously fitted subbituminous runs. As can be seen, the model proposed for carbon conversion in Bechtel's September Progress Report[2] gives results that are in reasonable agreement with the measured conversions for the newly received data; i.e., the errors in predicted conversion fall within the estimated error range of the proposed model.

3.3.2 Carbon Selectivity to Methane

Equation 4 of Bechtel's September Progress Report[2] has been used to predict carbon selectivity to methane for the five new subbituminous runs. The predicted and measured selectivities for the five new runs are shown in Figure 3-2, together with the measured and predicted

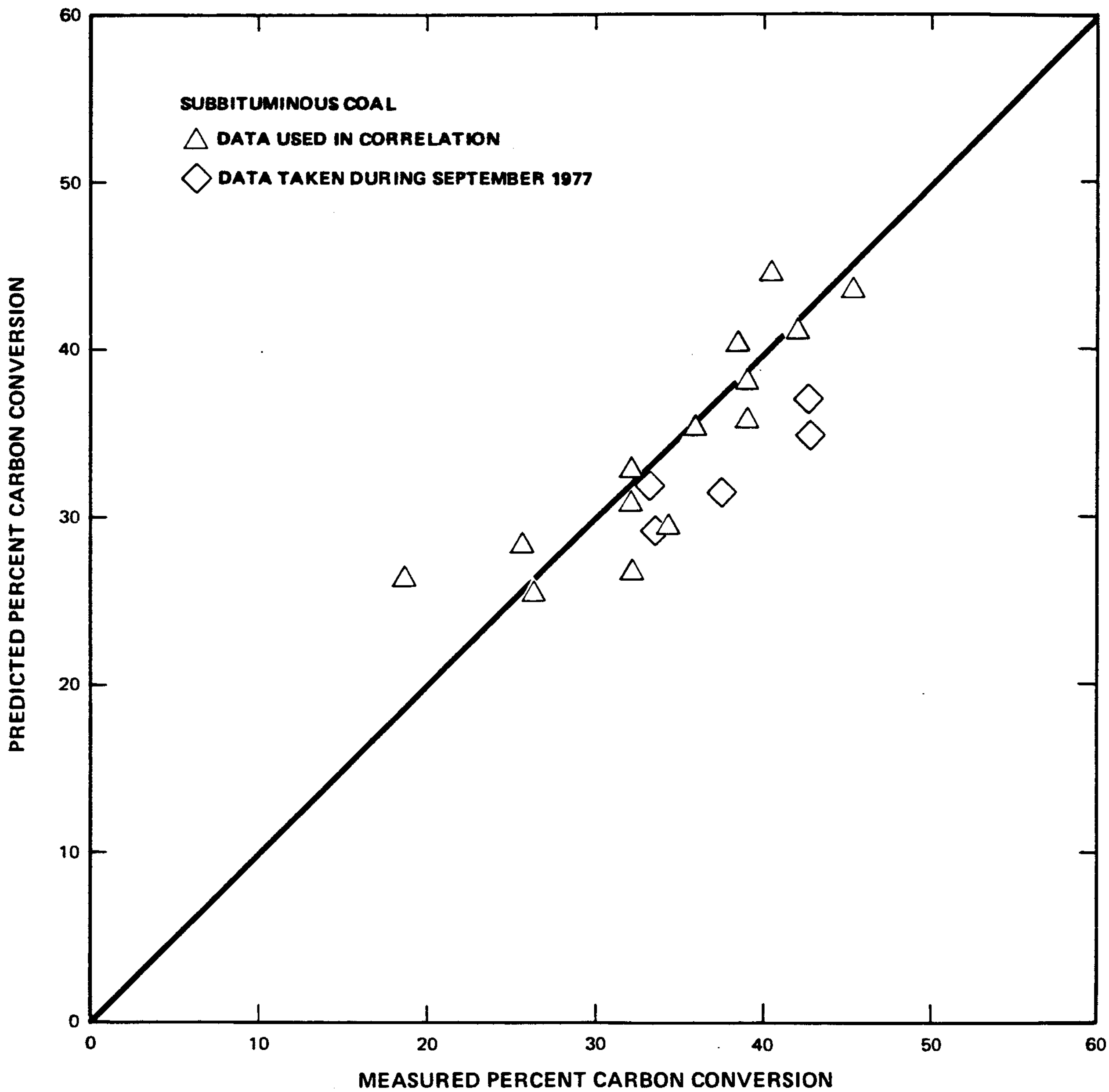

Figure 3-1. Comparison of Measured and Predicted Carbon Conversion for Subbituminous Coal for the Cities Service Reactor

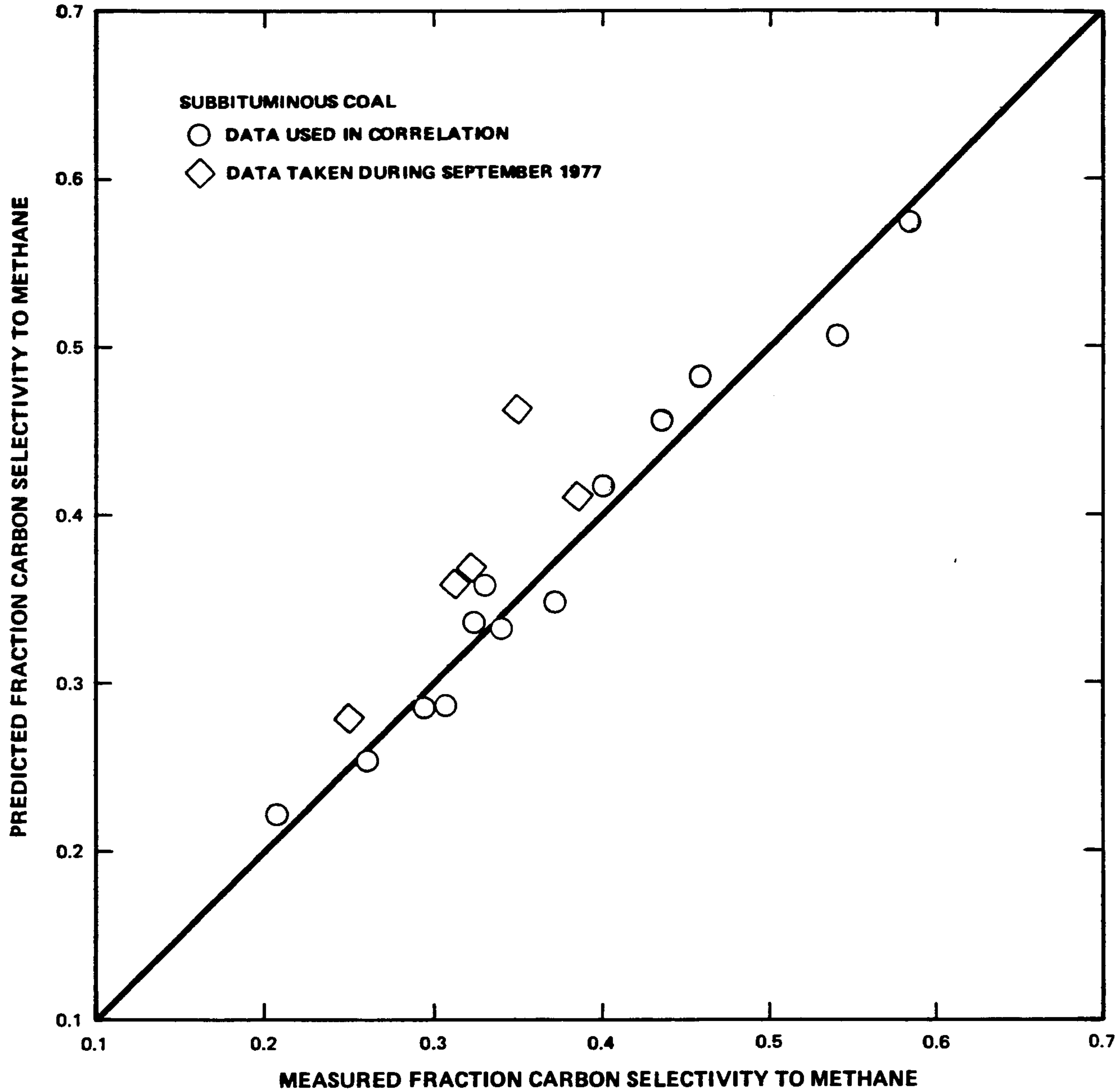

Figure 3-2. Comparison of Measured and Predicted Carbon Selectivity to Methane for Subbituminous Coal for the Cities Service Reactor

selectivities for the previously fitted runs. As can be seen, the model proposed for methane selectivity gives results that are in agreement with the measured selectivities for the newly received data, with the exception of Run MR-3, which has a measured selectivity of 35.2 percent. The excellence of the fit for the rest of the data suggests the possibility that the measurement of product gas methane content for Run MR-3 is in error.

3.3.3 Carbon Selectivity to Ethane

Equation 5 of Bechtel's September Progress Report[2] has been used to predict carbon selectivity to ethane for the five new subbituminous runs. The predicted and measured selectivities for the five new runs are shown in Figure 3-3, together with the measured and predicted selectivities for the previously fitted runs. As can be seen, the model proposed for ethane selectivity gives results that are in reasonable agreement with the measured selectivities for the newly received data.

3.3.4 Reactor Hydrogasification Efficiency

In Bechtel's September Monthly Report,[2] a reactor hydrogasification efficiency was defined and discussed in detail. The efficiency takes into account the heat contents of the product hydrocarbon gases, the feed coal, and the hydrogen consumed.

Reactor hydrogasification efficiencies have been calculated for the Cities Service subbituminous runs using Equations 7 through 9 of Bechtel's September Progress Report.[2] These calculated efficiencies are listed in Table 3-3, along with the calculated heat contents of hydrocarbon product gas and consumed hydrogen.

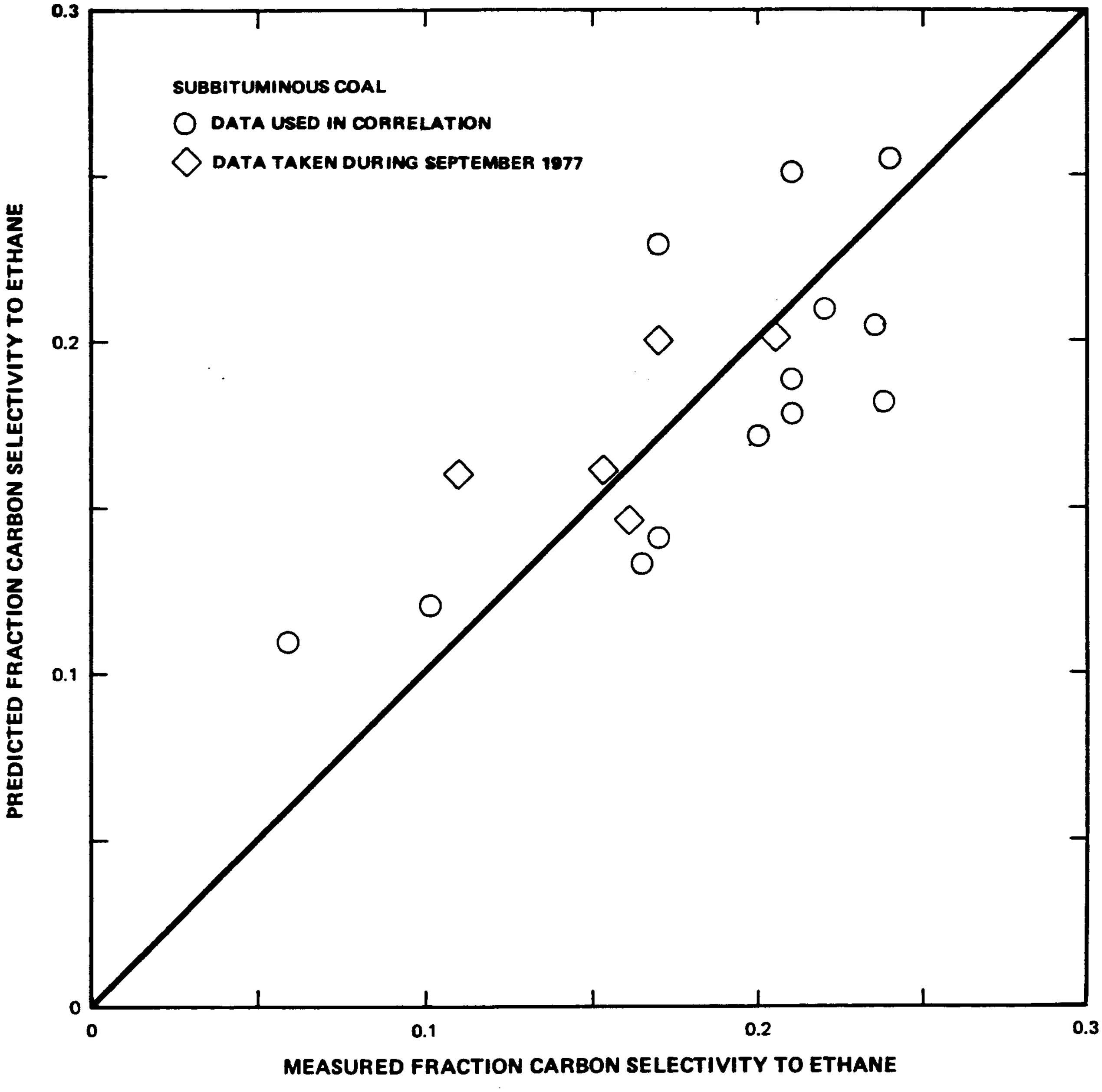

Figure 3-3. Comparison of Measured and Predicted Carbon Selectivity to Ethane for the Cities Service Reactor

Table 3-3

CITIES SERVICE REACTOR HYDROGASIFICATION EFFICIENCY*

RUN NO.	HEAT CONTENT OF HYDROCARBON GASES (BTU/LB MAF COAL)	HEAT CONTENT OF CONSUMED HYDROGEN (BTU/LB MAF COAL)	REACTOR HYDROGASIFICATION EFFICIENCY
MR- 4	1000.	1000.	
MR- 1	4201.	1472.	30.4
MR-10	1911.	591.	14.8
MR-13	4999.	2571.	33.5
MR-14	5608.	2707.	37.3
MR-28	3149.	1154.	23.4
MR-29	4431.	1839.	31.3
MR-30	4334.	2035.	30.2
MR-11	3058.	1355.	22.3
MR-12	3888.	1570.	28.0
MR-25	4424.	2038.	30.8
MR-26	5306.	2642.	35.4
MR-27	5837.	2785.	38.6
MR-15	6526.	3309.	41.7
MR- 2	4019.	1482.	29.1
MR- 3	3375.	806.	25.7
MR-16	3559.	1180.	26.3
MR-17	4456.	1632.	31.9
MR-18	5183.	2408.	35.2

*Based on Montana Rosebud subbituminous coal with an average heat content of 12,330 Btu per pound of MAF coal.

The reactor hydrogasification efficiencies listed in Table 3-3 were correlated with the operating variables listed in Table 3-2 using the following model:

$$\eta = 1 - \exp\left[-\alpha_1 \ (t_R)^{\alpha_2} \ (u_G)^{\alpha_3} \ (P_{H_2})^{\alpha_4} \ (H_2/\text{coal})^{\alpha_5} \ (d_P)^{\alpha_6} \ \exp\ (-\alpha_7/T)\right] \quad (1)$$

where,

η = fraction hydrogasification efficiency

$\alpha_1, \alpha_2 \ldots \alpha_7$ = fitted coefficients

t_R = gas (or particle) residence time

u_G = superficial gas velocity

P_{H_2} = hydrogen partial pressure

H_2/coal = hydrogen-to-coal ratio

d_P = mean particle diameter

T = reactor temperature

A statistical analysis of the data revealed that the reactor efficiency was a function of gas residence time and gas temperature. Reactor efficiency was not significantly affected by hydrogen partial pressure, hydrogen-to-coal ratio, particle size, or gas velocity within the region investigated. The correlation fitted to the Cities Service subbituminous data is:

$$\eta = 1 - \exp\left[-2.28 \ (t_R)^{0.338} \ \exp\ (-8{,}255/T_G)\right] \quad (2)$$

where,

t_R = gas residence time, milliseconds

T_G = maximum gas temperature, $^{\circ}R$

Equation 2 indicates that reactor efficiency increases with increase in residence time and temperature within the region investigated. Statistically, Equation 2 accounts for 79 percent of the variation in the data (multiple correlation coefficient of 0.89), with a standard error of estimate of 4 percent in the predicted percent hydrogasification efficiency. Measured and predicted efficiencies for the Cities Service reactor are illustrated in Figure 3-4. Both the statistics and Figure 3-4 indicate the good fit to the defined Cities Service hydrogasification efficiency using Equation 2. In Figure 3-5, the predicted efficiencies are plotted as a function of maximum gas temperature and gas residence time.

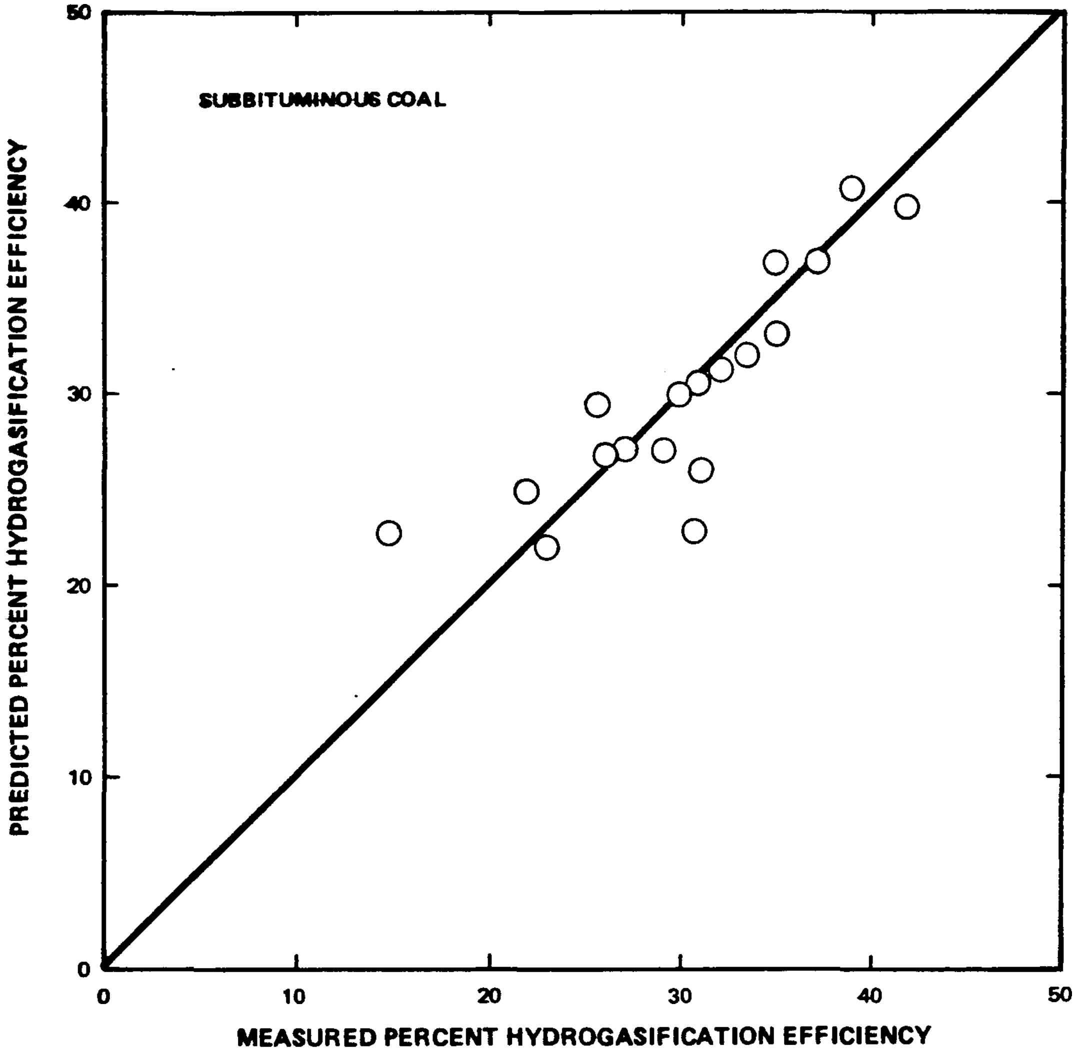

Figure 3-4. Comparison of Measured and Predicted Hydrogasification Efficiency for Subbituminous Coal for the Cities Service Reactor

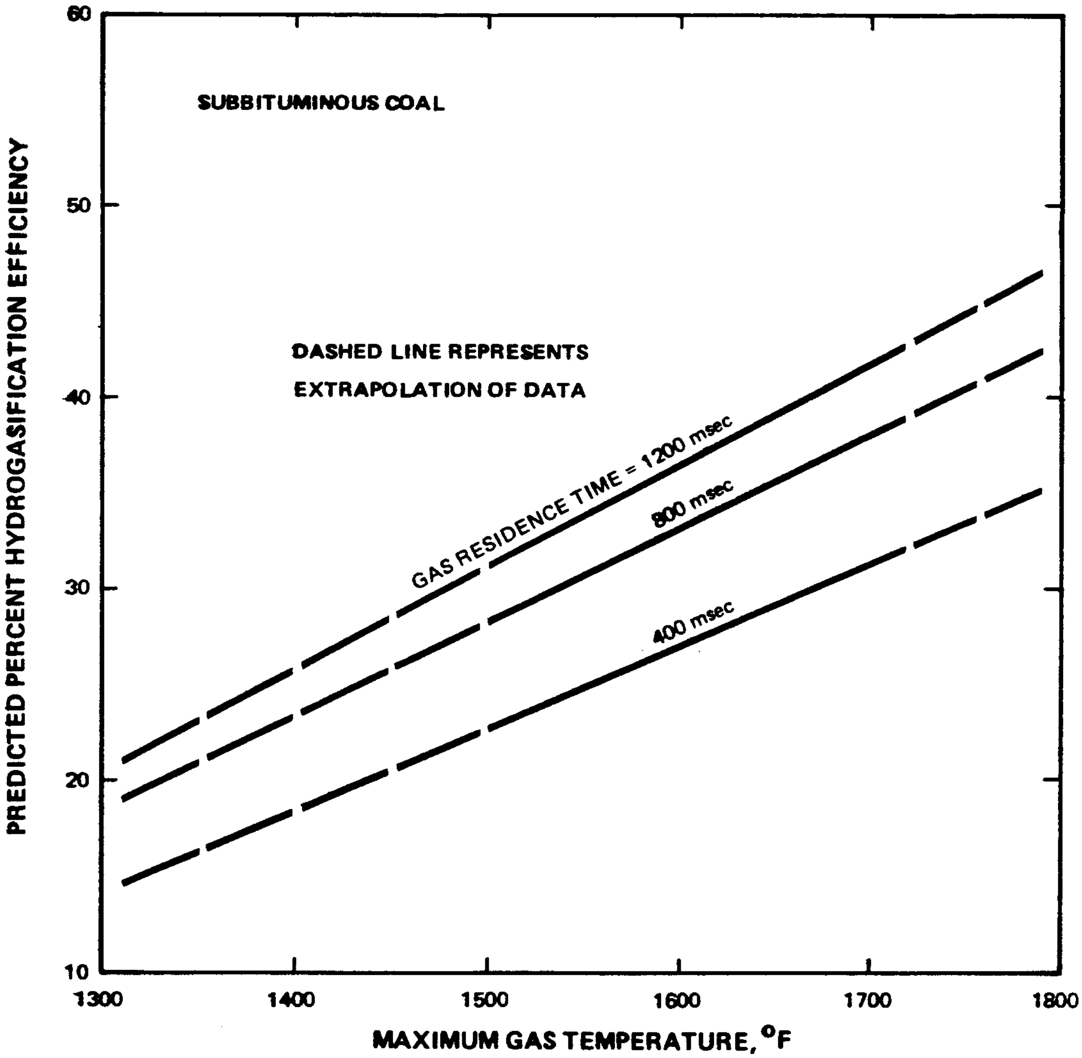

Figure 3-5. Predicted Hydrogasification Efficiency for Subbituminous Coal for the Cities Service Reactor

3.4 TASK III — ROCKETDYNE REACTOR MODELING

The semiempirical model developed by Bechtel to predict overall carbon conversion for the Cities Service subbituminous tests (Equation 2 in Bechtel's September Progress Report[2]) has been used to predict the overall carbon conversion for the three Rocketdyne hydrogasification tests conducted in the 1/4-ton/hr reactor using Montana Rosebud subbituminous coal. (A computer listing of the data from the three Rocketdyne subbituminous tests is shown in Table 3-1.)

The predicted and measured carbon conversions for the Rocketdyne subbituminous tests are shown in Figure 3-6, along with the predicted and measured conversions from the Cities Service subbituminous tests. As can be seen from this figure, the model developed for the Cities Service reactor gives results that are in reasonable agreement with the measured conversions for the Rocketdyne reactor, i.e., the errors in the predicted Rocketdyne conversions are well within the estimated error range of the correlation. This is an indication that the Cities Service bench-scale reactor and the Rocketdyne 1/4-ton/hr reactor behave similarly for the same coal within the region investigated. It is also an indication that the model developed by Bechtel for predicting carbon conversion for Montana Rosebud subbituminous coal under rapid-rate hydrogasification conditions (flash hydropyrolysis) is sound within the region of operating variables investigated to date by Cities Service and Rocketdyne. Of course, as more subbituminous data are generated by Rocketdyne and Cities Service, the proposed correlation will be further refined, and the comparative behavior of the two reactors verified.

The correlation developed for the Cities Service subbituminous coal has also been used to predict overall carbon conversion for Rocketdyne bituminous coal Run 011-7 (see Table 3-1). The predicted and measured carbon conversion for Run 011-7 is also shown in Figure 3-6. As can be

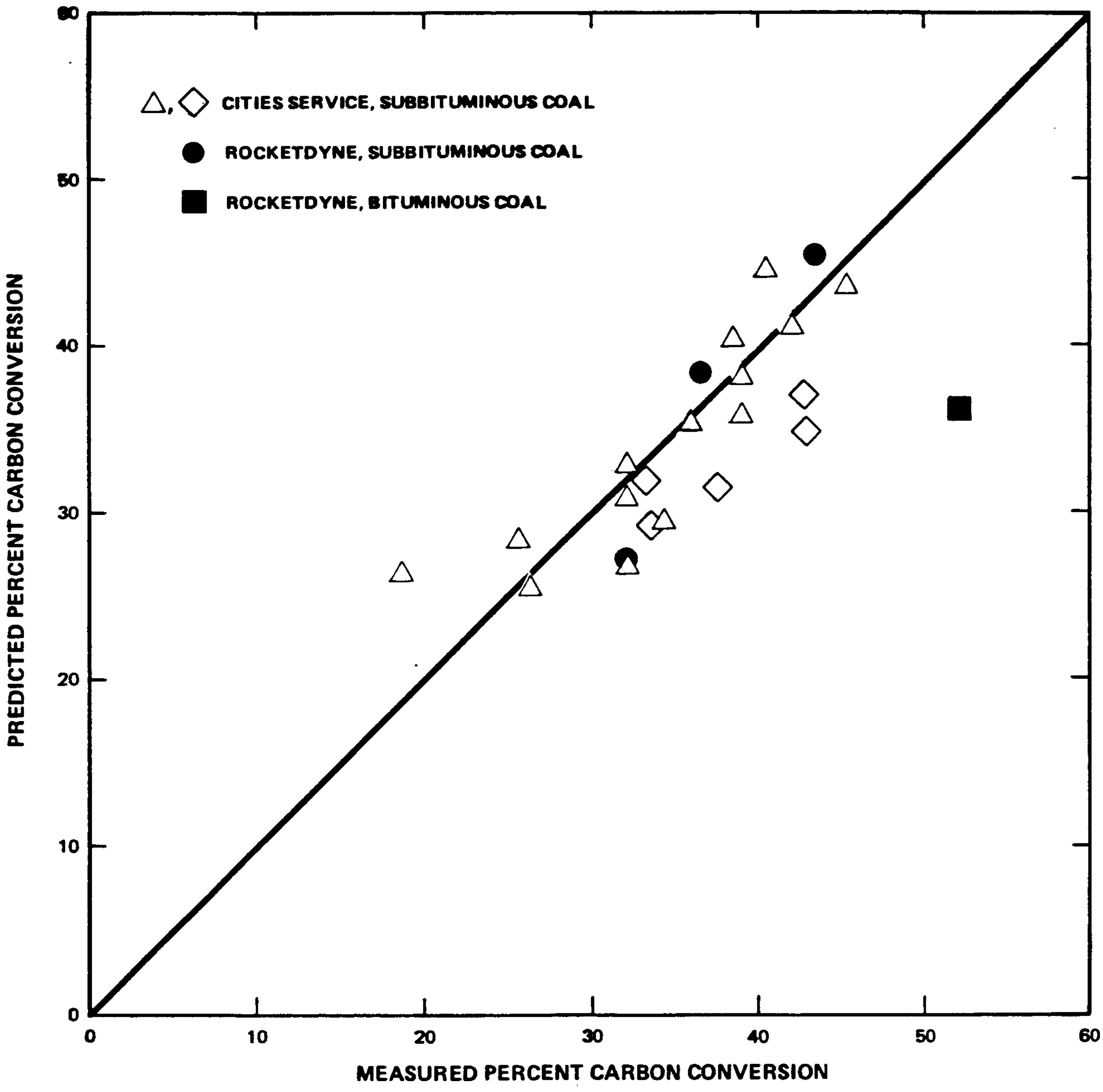

Figure 3-6. Comparison of Measured and Predicted Carbon Conversion for the Cities Service and Rocketdyne Reactors

seen in this figure, the predicted carbon conversion is significantly lower than the measured conversion, which indicates that there is an apparent effect of coal type that is not accounted for in the sub-butiminous coal model. This result was expected, since previous Rocketdyne data with bituminous coal[3] have shown higher levels of carbon conversion than recent data with subbituminous coal under similar operating conditions.

The semiempirical model developed by Bechtel to predict overall carbon conversion for the Rocketdyne partial liquefaction bituminous coal tests in the 1-ton/hr reactor was used to predict carbon conversion for Rocketdyne bituminous Run 011-7. Carbon conversion predicted with this model (Equation 5 in Bechtel's June-August 1977 Quarterly Progress Report[4]) was 61 percent, compared with a measured conversion of 52 percent. As further bituminous coal data are generated in the Rocketdyne 1/4-ton/hr reactor, the general exponential model proposed by Bechtel for correlating carbon conversion[4] will be fitted to all of the bituminous data in the 1-ton and 1/4-ton/hr reactors.

3.5 TASK III — CONCEPTUAL DESIGN BASIS FOR FULL-SCALE REACTOR

This subsection describes the basis for the selection of operating variable levels and size constraints for the hydrogasification stage of a proposed integrated full-scale reactor facility for converting coal to pipeline-quality gas. The integrated reactor facility consists of a hydrogasification stage to produce methane-rich product gas from the coal, and a hydrogen production stage to produce hydrogen-rich product gas from the unreacted char. Thus far, the development effort has been devoted primarily to the hydrogasification stage. However, future development of the char-to-hydrogen stage also is required to have an economic process. A possible reactor scheme for the unreacted char is shown to illustrate the concept of an integrated reactor and is not intended to be a recommendation. Further work will be devoted to development of designs for the hydrogen production stage. A sketch and a detailed description of the conceptual reactor facility are presented in Subsection 3.6.

The conceptual full-scale hydrogasification stage will have a configuration similar to the Rocketdyne reactor assembly, which consists mainly of a preburner, injector nozzles, and a tubular entrained-downflow reactor chamber. Details of the Rocketdyne reactor assembly have been given elsewhere.[5]

Bechtel has already developed a reference design basis for a conceptual full-scale hydrogasification reactor stage.[4] This design basis was developed employing data gathered in the Rocketdyne 1-ton/hr reactor using Kentucky HvAb coal,[3] together with predictive reactor performance models fitted to the data by Bechtel.[4] For this design basis, a maximum reactor temperature of 1,400°F was required to achieve an overall carbon conversion of 50 percent. Recent data from Cities Service and Rocketdyne, however, have shown that higher temperatures (about 1,800°F) may be required to attain 50 percent carbon conversion for Montana Rosebud subbituminous coal.

In view of the above considerations, Bechtel has decided to revise the previous design basis in order to select a set of operating parameter levels consistent with the use of the less reactive subbituminous coal. A reactor design based on the higher required reaction temperature will obviously allow for the handling of a wider range of coals. The revised operating levels will be based on subbituminous coal data generated at Cities Service and Rocketdyne, together with the predictive reactor performance models fitted to the data by Bechtel.[2] As shown earlier in this report, the models fitted to the Cities Service subbituminous data appear to correlate well with the recent Rocketdyne subbituminous data.

The revised design basis for the conceptual full-scale hydrogasification reactor stage is given below. This design basis should be considered preliminary, since it will be updated as more subbituminous coal data are generated by Cities Service and Rocketdyne.

Selected Operating Parameters:

Coal type	Montana Rosebud subbituminous
Coal mean particle size	40 to 50 microns
Coal feed rate	108 tons/hr
Nominal reactor pressure	1,000 psig
Hydrogen preheat temperature	3,000^{o}F
Coal-hydrogen mix temperature	1,700^{o}F
Maximum reactor temperature	1,800^{o}F
Overall carbon conversion	50 percent

Calculated Operating Parameters:

Hydrogen-to-coal ratio	0.20 lb/lb
Nominal gas (or particle) residence time	1,120 milliseconds
Carbon selectivity to hydrocarbon gas	70 percent

The coal type and size are those used in the recent Cities Service and Rocketdyne testing; the average coal composition has been given elsewhere.[6] The reactor pressure selected is within the middle of the range (500 to 1,500 psig) covered in the Cities Service testing (see Table 3-2). Note that a statistical analysis of the Cities Service subbituminous coal data presented in Bechtel's September Progress Report[2] showed that carbon conversion was relatively unaffected by reactor pressure within the region investigated.

The selected coal feed rate of 108 tons/hr is based on a recommendation by Gray[7] for a maximum coal capacity for a single injector element of 3 tons/hr and a maximum number of 36 injector elements per head. Gray has also recommended a hydrogen preheat temperature of 3,000°F, which can be easily achieved by combustion with a relatively small amount of oxygen in a preburner placed ahead of the reactor injection head. To date, Rocketdyne and Cities Service have used hydrogen preheat temperatures of approximately 2,000°F and 1,600°F, respectively.[3,6]

The coal-hydrogen mix temperature (initial reaction temperature) of 1,700°F was selected since it has been demonstrated that this temperature is easily attainable with the Rocketydne injection nozzle.[5] The maximum reaction temperature of 1,800°F was estimated by conducting a heat balance around the reactor, assuming adiabatic operation and including reaction heat effects of initial devolatilization (endothermic) and the coal-hydrogen reactions (exothermic).

An overall carbon conversion of about 50 percent was chosen because previous studies[6] have shown that this is approximately the desired conversion level required for an overall balanced process. A balanced process is a process where the char by-product from hydrogasification is further gasified (probably with steam and oxygen) to make the required process hydrogen. Although the maximum carbon conversion achieved to date in the Cities Service and Rocketdyne subbituminous tests is about 45 percent (see Run 011-4 in Table 3-1 and Run MR-15 in Table 3-2), planned future subbituminous tests at extended residence times are expected to yield higher conversions.

The hydrogen to coal ratio of 0.20 lb/lb was calculated from a simple heat balance around the coal-hydrogen mixing injector nozzle, assuming coal is fed at 60°F, hydrogen is fed at 3,000°F, and the final mix temperature is 1,700°F. This hydrogen-to-coal ratio is lower than the levels used by Rocketdyne and Cities Service in their testing to date. It should be noted that a statistical analysis of the Cities Service subbituminous coal data presented in Bechtel's September Progress Report[2] showed that carbon conversion was relatively unaffected by hydrogen-to-coal ratio within the region investigated.

The nominal gas (or particle) residence time for the entrained-flow reaction chamber was calculated from the correlation developed by Bechtel for predicting carbon conversion for the Cities Service reactor with subbituminous coal (Equation 2 in Bechtel's September Progress Report[2]). The residence time t_R was obtained by substituting the selected carbon conversion and maximum reaction temperature into the correlation:

$$0.50 = 1 - \exp\left\{-1.59\ (t_R)^{0.335}\ \exp\left[-7{,}210/(1{,}800 + 460)\right]\right\}$$

$$t_R = 1{,}120 \text{ milliseconds}$$

The carbon selectivity to hydrocarbon gas,* $\Phi_{C_1-C_4}$, was calculated from the correlation developed by Bechtel for the Cities Service

*Hydrocarbon gas consists of methane and ethane, plus other paraffins and olefins.

subbituminous data (Equation 6 in Bechtel's September Progress Report[2]), at the conditions of pressure, temperature, and residence time previously defined:

$$\Phi_{C_1-C_4} = 1 - \exp\left\{-18.7\ (1{,}000)^{-0.227}\ (1{,}120)^{0.103} \exp\left[-4{,}330/(1{,}800 + 460)\right]\right\}$$

The hydrogen mass feed rate is easily calculated from the given hydrogen-to-coal ratio and the coal feed rate. At the specified average reactor temperature, pressure, and hydrogen feed rate, the average volumetric flow rate of the gas through the reactor can be estimated from the ideal gas law, assuming negligible change in the total number of moles of gas flowing through the reactor. This assumption appears reasonable, since calculations based on the results from Cities Service Run MR-15, in which a carbon conversion of 45 percent was achieved, showed a total change of only about 5 percent in the total number of moles of gas inside the reactor. For these assumptions, the average volumetric flow rate of gas V_G is approximately 510,000 ft^3/hr (142 ft^3/sec).

The reactor dimensions are related to the nominal superficial gas velocity as follows:

$$S = V_G/u_G = 142/u_G \qquad (3)$$

and

$$L = t_R u_G = 1.12\ u_G \qquad (4)$$

where,

S = reactor cross-sectional area, ft^2

L = reactor length, feet

u_G = superficial gas velocity, ft/sec

For any specified gas velocity, the reactor cross-sectional area and length can be calculated using the above equations. A superficial gas velocity range of from 10 to 25 ft/sec has been selected for the reactor design, based on recommendations by Gray[7,8] and the conditions tested at Cities Service and Rocketdyne. At 10 ft/sec gas velocity, the required reactor cross-sectional area from Equation 3 is 14 ft^2, and the required reactor length from Equation 4 is 11 feet. At 25 ft/sec gas velocity, the required cross-sectional area is 6 ft^2, and the required length is 28 feet.

3.6 TASK III — CONCEPTUAL DESIGN OF FULL-SCALE REACTOR

In this section, a detailed description is given of a design concept proposed by Bechtel for a full-scale reactor facility for converting coal to pipeline-quality gas. The proposed reactor design concept is preliminary and is intended to provide a basis for further modifications and studies. Further study should result in simplification and improve-

Other design concepts have been considered and should be investigated. One plan, based on a Rocketdyne-type reactor for hydrogen generation from unreacted char, is of great interest but does present real developmental problems. These should be explored further.

A detailed sketch of the reactor assembly is given in Figure 3-7. Basically, the reactor vessel consists of three sections. The uppermost part of the vessel contains a shell and tube heat exchanger; the middle part includes a coal hydrogasification reactor and a cyclone separator; and the lower part includes a steam-oxygen-char gasification reactor. As discussed in the previous subsection, the hydrogasification reactor would have a length roughly between 10 and 30 feet, depending on the gas velocity.

In the hydrogasification section, hot hydrogen at 3,000^{o}F is contacted with coal feed in a total of 36 mixing-injection nozzles; each nozzle handles a maximum of 3 tons of coal per hour, as has been discussed previously in Subsection 3.5. The nozzle design is similar to that developed and used by Rocketdyne in its 1-ton/hr and 1/4-ton/hr hydrogasification reactor facilities. The mixing nozzles are arranged in single rank in a circle. Coal enters each through a central tube, and hot hydrogen enters through annular nozzles around the coal tubes.

Char and product gas flow downward in an entrained-flow manner through the annuli formed by the inner wall of reactor vessel shell and the outer shell of a central pipe (or duct) through which the product gas leaves the hydrogasifier. The coal char solids and the gas stream are

Figure 3-7. Conceptual Design of a Full-Scale Reactor

separated in a cyclone which sends the product gas stream back up through the central pipe or duct and sends the char downward through a cyclone dipleg. The char next collects in a surge volume section and is held there as a feed material for the second reaction stage. The cyclone is constructed so that it can be moved vertically and hence could be used to control the residence time of char and gas inside the reactor. A water or gas quench system is also installed near the bottom of the central pipe to provide an extra or standby facility for quickly controlling the reaction, if necessary.

Product gas from the hydrogasifier cyclone flows upwards through the tube side of a shell and tube heat exchanger where it is cooled from 1,800^{o}F to about 1,000^{o}F by heat exchange with cold feed hydrogen flowing downward through the exchanger shell side. This hydrogen stream is assumed to enter at 100^{o}F and is heated to about 1,100^{o}F.

The hydrogen effluent from the exchanger is further heated to about 3,000^{o}F by combustion with oxygen, which is injected into the hydrogen stream near the exchanger outlet, as shown in Figure 3-7. This hydrogen preburner section should be relatively short since combustion and heating are rapid, but if experience shows otherwise, the preheater section could be easily made longer than indicated in Figure 3-7.

Char from the hydrogasification reactor, containing about 50 percent of the feed carbon, is then reacted with steam and oxygen in a second stage to produce most of the process hydrogen required for hydrogasification. Because only limited data are presently available on the reactivity of char from the Rocketdyne-type hydrogasifier used here, it is assumed that several minutes of holding time will be required to gasify the char to produce acceptable yields of hydrogen. This suggests that a dense-phase fluid bed should be used for the steam-oxygen-char reactor, as is shown in Figure 3-7. The fluid-bed nominal reaction temperature is assumed to be about 1,800^{o}F.

The char solids are fed to the fluid-bed reactor via a fluidized standpipe and throttle valve combination; this combination is simple but considered eminently suitable for this severe service. Oxygen and steam are fed to the fluid bed via a gas distributor manifold near the bottom of the reactor. The product gas from this reactor leaves at the top through a cyclone separator, and the entrained fines are collected and returned by the cyclone dipleg and the oxygen carrier gas stream, as is illustrated in Figure 3-7. The spent char (mostly ash in composition) leaves the fluid-bed reactor at the bottom and goes down to a quench pot where it is sprayed with sufficient water to make up an ash-water slurry for transfer to pressure letdown and eventual disposal.

A small direct-fired heater may be used to start up the fluid-bed reactor, as is shown in Figure 3-7. Hot gases from this heater may also be sent to the hydrogasification section during startup. This startup heater is shown here primarily as a reminder that a practical startup procedure must be developed for this full-scale reactor facility.

The reactor vessel shell shown in Figure 3-7 has internal refractory insulation and a bare metal shell free of external insulation. Although this "hot-wall" design is typical of catalytic cracking practice, the higher temperature (1,800°F) and pressure (1,000 psig) within the shell demand careful attention in the interest of operating reliability and overall safety. One approach would be to provide infrared scanning and hot-spot alarm instrumentation for the outer shell wall, whose surface temperature would be kept between 250°F and 400°F. A screen of louvers would shield the bare metal shell from rain and weather-induced thermal stresses. This vessel shell design will certainly require alloy lining.

Although the hydrogasification reactor section may be only 10 to 30 feet in length (see Subsection 3.5), Figure 3-7 (drawn roughly to scale) suggests that the overall reactor system, including the two reaction sections and related equipment, could be roughly 150 to 200 feet high. This reactor length is not excessive; it is needed to assure smooth solids transfer between the different sections of the reactor. The method proposed here for removing the hot char from the hydrogasification section and transferring it without cooling to the char gasification section has to be regarded as probably the simplest and most direct method that can be devised. Some of the seemingly excessive height is also due to the assumed size of the steam-oxygen-char reactor, which was based on a char holding time on the order of minutes, as has been discussed previously. Of course, as more is learned about the operation of the hydrogasifier and fluid-bed reactor, and about the properties of the char from the hydrogasification section, it is likely that some of the safety factors incorporated in prototype designs as the one given here can be reduced and the height of a commercial-size unit decreased.

The control methods and systems needed for the reliable and safe operation of the reactor, with special attention to the control of solids flow through the reactor, are being studied and will be presented in future reports. In addition, other approaches to reactor vessel shell design are being considered; one approach will be to use a system pressure water jacket inside the vessel strength shell to keep the metal temperature as low as 550°F. Alternative designs for the steam-oxygen-char reactor are also being considered. These designs are aimed at reducing the reaction time (holding time) and may include reactor operation at much higher temperatures (2,600°F to 2,800°F), the Rocketdyne design principle of rapid heating in mixing-injection nozzles, and the use of entrained flow.

3.7 FUTURE WORK

During the next reporting period, work will be conducted in the areas discussed below.

Models developed for correlating the Rocketdyne and Cities Service carbon conversion and carbon selectivity data will be updated and improved upon as further tests results are obtained with Montana Rosebud subbituminous coal and with Western Kentucky bituminous coal.

Models will be developed, where possible, for correlating the carbon conversion and carbon selectivity data received to date from Brookhaven National Laboratories.

Conceptual design of a reference, full-size hydrogasification reactor will be continued.

Additional data that may be required for reliable pilot plant design will be identified, and experimental programs necessary for the generation of the additional data will be recommended.

Section 4

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7. Personal communication between M. Epstein (Bechtel) and J. A. Gray (Rocketdyne)* on August 18, 1977.

8. Personal communications between L. S. Daniels (Bechtel) and J. A. Gray (Rocketdyne) on September 14, 1977.

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