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AN ANALYSIS OF COAL HYDROGASIFICATION PROCESSES

Quarterly Technical Progress Report for the Period 1 September - 30 November 1977

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ABSTRACT

This Quarterly Technical Progress Report covers work performed during the period 1 September 1977 to 30 November 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 the reporting period, substantial progress was made on Tasks I, II, III, and IV. Data from six recent Rocketdyne tests using subbituminous coal and four recent Rocketdyne tests using bituminous coal were entered into the computerized data base. Also, data from 16 recent Cities Service tests using subbituminous coal were entered into the data base. The base contained data from earlier Rocketdyne tests with bituminous coal, earlier Cities Service tests with lignite coal, and recent Cities Service tests with subbituminous coal.

Semiempirical correlations for predicting carbon conversion efficiency and carbon selectivity to methane and ethane were fitted to the Cities Service lignite and subbituminous data. Coal type did not appear to have any significant effect on carbon conversion or carbon selectivity. The correlation developed for predicting carbon conversion for the Cities Service subbituminous tests gave results that were in good 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. Substantially higher carbon conversions were obtained in the Rocketdyne tests with bituminous coal than in the Cities Service and Rocketdyne tests

with subbituminous coal at comparable operating conditions. The measured carbon conversions for the recent Rocketdyne bituminous tests (in the 1/4-ton/hr reactor) were from 10 to 15 percent lower than the measures conversions for the earlier Rocketdyne bituminous tests (in the 1-ton/hr reactor). Results from the recent 1/4-ton/hr reactor testing are in doubt because of the relatively short duration of the runs. Bechtel has recommended that replicate runs be added to the Rocketdyne and Cities Service experimental designs, and that Cities Service conduct some additional tests at 750 and 500 psig hydrogen partial pressure.

During the reporting period, data from 48 tests conducted at the Brookhaven National Laboratory using lignite coal were entered into the computerized data base. The results from the tests are tabulated and discussed.

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 the full-scale hydrogasification reactor. Also, hydrogen production using unreacted char in a steam/oxygen gasification stage was discussed.

During November, visits were made to the City College of the City University of the New York and Brookhaven National Laboratory. The purpose of the visits were to: (1) discuss the ongoing technical programs, (2) obtain additional hydropyrolysis data, and (3) observe the testing facilities.

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Section 1

OBJECTIVES AND SCOPE

This report is the third Quarterly Technical Progress Report for a program entitled, "An Analysis of Coal Hydrogasification Processes." The program is being performed for DOE by Bechtel Corporation under DOE 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 Pittsburgh Energy Research Center (PERC) coal hydrogasification processes, relative to DOE plans for a hydrogasification 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 DOE. 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 needed 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

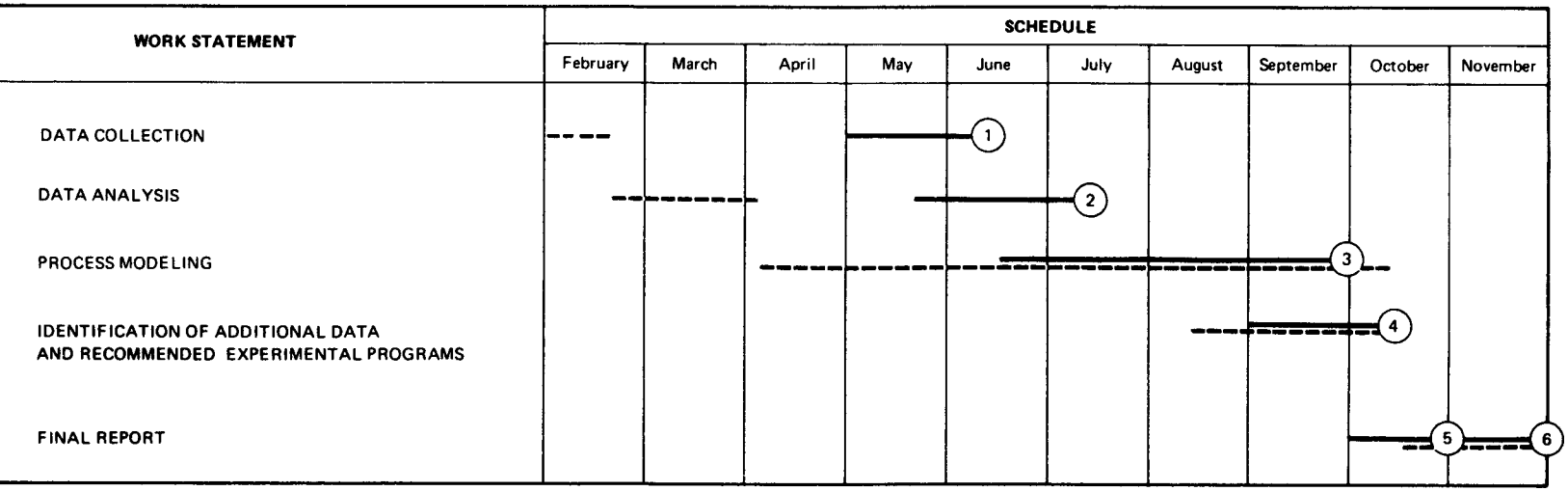
Figure 2-1 summarizes the program progress between Febraury 1, 1977 (the program start date) and November 30, 1977. During the reporting period, substantial progress was made on Tasks I, II, III, and IV. The technical progress for each subject task is presented in Section 3. As can be seen in Figure 2-1, actual manhours expended are less than planned, while program progress is on schedule.

2.2 OPEN ITEMS

As presently schedule, the completed results from the Cities Service and Rocketdyne DOE 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.

REPORT PERIOD: 1 Feb – 30 November 77

TASK NO.	WORK STATEMENT	SCHEDULE
I	DATA COLLECTION	
II	DATA ANALYSIS	
III	PROCESS MODELING	
IV	IDENTIFICATION OF ADDITIONAL DATA AND RECOMMENDED EXPERIMENTAL PROGRAMS	
–	FINAL REPORT	



LEGEND:

- Revised Schedule
- Original Schedule
- Planned Manhours and Progress
- Actual Manhours
- Actual Progress

(1) Completion of Task I
(2) Completion of Task II
(3) Completion of Task III
(4) Completion of Task IV
(5) Submittal of Draft of Final Report
(6) Submittal of Final Report

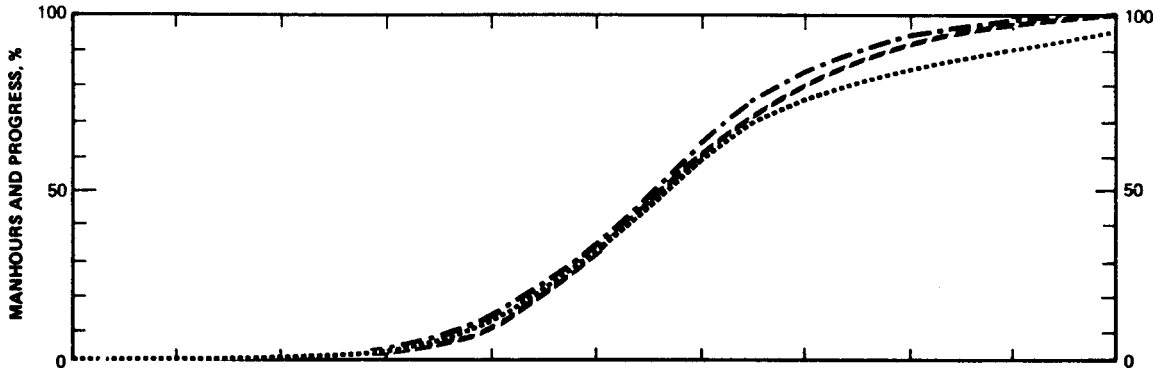


Figure 2-1. Progress and Performance Chart

Section 3

TECHNICAL PROGRESS

This section describes the technical progress for Tasks I, II, III, and IV during the reporting period. Visits made to the City College of New York and Brookhaven National Laboratory are discussed in the Appendix.

3.1 TASKS I AND II — ROCKETDYNE DATA COLLECTION AND ANALYSIS

During this reporting period, Bechtel received data from Rocketdyne[1] for 10 recently completed hydrogasification tests conducted in the Rocketdyne 1/4-ton/hr reactor test facility. Six of the tests used Montana Rosebud subbituminous coal and four used Western Kentucky bituminous (HvAb) coal. Complete analyses of both coals have been given elsewhere.[1] Five of the tests (Runs 011-2, 4, 5, 7, and 8) contain data that were presented previously[2] and that have been revised by Rocketdyne.

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

All the recent hydrogasification data were generated in an entrained-downflow tubular reactor, 1.88 inches in diameter and 15 feet in length. The subbituminous tests (Runs 011-2, 4, 5, 11, 12, and 13) were conducted at reactor pressures of 1,000 to 1,500 psig, outlet gas temperatures of 1,470°F to 1,900°F (1,930°R to 2,360°R), and gas (or particle) residence times of 570 to 870 milliseconds. Preliminary analytical

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 TPH	.382		1800.	1000.	155.	.250
6	2/ 3/77	HVAB	1 TPH	.542	0.397	2160.	1000.	130.	.478
7	2/ 7/77	HVAB	1 TPH	.615	0.483	2370.	1000.	120.	.775
8	2/17/77	HVAB	1 TPH	.596	0.485	2160.	1000.	270.	.365
9	2/22/77	HVAB	1 TPH	.645	0.760	2260.	1500.	410.	.365
10	3/ 1/77	HVAB	1 TPH	.609	0.782	2050.	1500.	490.	.314
11	3/ 4/77	HVAB	1 TPH	.627	1.000	2060.	1500.	630.	.344
12	3/ 9/77	HVAB	1 TPH	.576	0.672	2060.	1000.	430.	.333
13	3/23/77	HVAB	1 TPH	.538	0.348	2160.	1000.	60.	.292
14	3/25/77	HVAB	1 TPH	.570	0.507	2070.	1500.	100.	.397
15	3/29/77	HVAB	1 TPH	.526	0.382	2160.	700.	45.	.403
011- 7	9/21/77	HVAB	1/4 TPH	.473	0.416	2130.	1000.	651.	.356
011- 8	9/29/77	HVAB	1/4 TPH	.532	0.586	2270.	1010.	509.	.421
011- 9	10/ 4/77	HVAB	1/4 TPH	.588	0.724	2420.	1500.	757.	.499
011-10	10/ 7/77	HVAB	1/4 TPH	.562	0.740	2370.	1490.	756.	.506
011- 2	8/30/77	SUBBTM	1/4 TPH	.289	0.495	1930.	1020.	619.	.592
011- 4	9/ 9/77	SUBBTM	1/4 TPH	.332	0.910	2360.	990.	568.	.512
011- 5	9/15/77	SUBBTM	1/4 TPH	.365	0.627	2190.	1000.	608.	.401
011-11	10/14/77	SUBBTM	1/4 TPH	.435	0.885	2320.	1500.	818.	.543
011-12	10/18/77	SUBBTM	1/4 TPH	.362		2050.	1500.	867.	.559
011-13	10/21/77	SUBBTM	1/4 TPH	.290		1930.	1500.	810.	.541

results indicate overall carbon conversions of 29 to 44 percent and carbon selectivities to gaseous products of 50 to 90 percent (carbon selectivity to gases was not reported for Runs 011-12 and 13).

The bituminous tests (Runs 011-7, 8, 9, and 10) were conducted at reactor pressures of 1,000 to 1,500 psig, outlet gas temperatures of 1,670°F to 1,960°F (2,130°R to 2,420°R), and gas residence times of 510 to 760 milliseconds. Preliminary analytical results show overall carbon conversions ranging from 47 to 59 percent and carbon selectivities to gases ranging from approximately 40 to 75 percent.

The overall carbon conversions for the Rocketdyne subbituminous tests appear to be in substantial agreement with those of the Cities Service bench-scale subbituminous tests at comparable operating conditions (see Subsection 3.6 of this report). Also, substantially higher conversions were obtained with the Kentucky bituminous coal than with the Montana subbituminous coal at comparable operating conditions. However, the overall carbon conversions reported for the 1/4-ton/hr reactor bituminous tests (Runs 011-7, 8, 9, and 10) appear to be consistently lower by about 10 percent from those obtained earlier with the same coal during the 1-ton/hr reactor bituminous testing (Runs 5 through 15).

Probable uncertainties in the values for carbon conversion for subbituminous and bituminous Runs 011-2, 4, 5, 7, 8, 9, and 10 were mentioned by Rocketdyne;[1] these uncertainties stem mainly from the relatively short durations (approximately 3 minutes) of the tests, which, owing to operational problems, had all been terminated prematurely. For these tests, marked variations occurred among the product gas samples taken at various time intervals, and an average of these samples was used to calculate the conversions. In addition, poor carbon balance closures were reported by Rocketdyne for bituminous Runs 011-5, 7, 8, 9, and 10 in the 1/4-ton/hr reactor.

Rocketdyne has not yet reported product gas analyses and completed material balances for a majority of its recent tests. These results will be incorporated into the Bechtel data base as they become available.

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

During this reporting period, Bechtel received additional data from 16 recently completed Cities Service bench-scale hydrogasification tests using Montana Rosebud subbituminous coal.[1,2,3] These data were entered into the computerized data base containing data from Cities Service for 9 earlier subbituminous tests and 25 earlier lignite tests. A computer listing of all the data is presented in Table 3-2.

The 16 recent subbituminous tests were made in a bench-scale helical entrained-flow reactor having a nominal diameter of 0.21 inch and heated length of 60 feet. These tests were conducted at reactor pressures of 500 to 1,600 psig, maximum gas temperatures of 1,520°F to 1,730°F (1,980°R to 2,190°F), and gas (or particle) residence times of 304 to 3,480 milliseconds. Overall carbon conversion ranged from 32 to 51 percent. The highest carbon conversion of 51 percent was achieved in Run MR-21, at 1,600 psig hydrogen partial pressure, 1,590°F maximum gas temperature, and 3,480 milliseconds residence time.

Run MR-27 gave the highest methane yield and selectivity, while Run MR-20 gave the highest ethane yield and selectivity. Run MR-39 had a carbon conversion to ethane of 0.4 percent and a carbon selectivity to ethane of 0.9 percent. These ethane values are questionably low when compared with results from other runs under similar operating conditions.

Because of operational problems, Run MR-5 was ended prematurely. Since no gas samples were taken in that run, no product analysis was reported.

Excellent carbon mass balance closures ranging from 96 to 108 percent and ash balance closures ranging from 86 to 91 percent were reported for the recent subbituminous tests.

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)
1	1975-6	LIGNITE	FF	.472				2040.	1500.	0.49	6300.	1180.	1.40	175.
2	1975-6	LIGNITE	FF	.434				1960.	1500.	0.46	6600.	880.	1.30	250.
3	1975-6	LIGNITE	FF	.366	.243	.197	.451	1940.	1500.	0.50	3000.	430.	1.30	200.
4	1975-6	LIGNITE	FF	.377	.276	.196	.507	1890.	1500.	0.45	6800.	460.	1.60	470.
5	1975-6	LIGNITE	FF	.323	.300	.183	.489	1960.	750.	0.90	1700.	350.	1.20	200.
6	1975-6	LIGNITE	FF	.435	.345	.214	.563	1970.	1500.	0.40	7700.	155.	0.90	190.
7	1975-6	LIGNITE	FF	.369	.382	.157	.540	2080.	580.	1.70	1800.	880.	1.40	190.
8	1975-6	LIGNITE	FF	.816	.635	.089	.725	1940.	2960.	0.20	14700.	2470.	1.00	190.
9	1975-6	LIGNITE	EF	.429	.361	.226	.588	1990.	1000.	7.70	2400.	2400.	2.00	190.
10	1975-6	LIGNITE	FF	.374	.382	.160	.544	1920.	1500.	0.30	10400.	1520.	0.48	190.
11	1975-6	LIGNITE	FF	.430	.498	.109	.608	2000.	1500.	0.12	24700.	6290.	0.18	56.
12	1975-6	LIGNITE	FF	.492	.482	.110	.592	1950.	2000.	0.28	10800.	1650.	0.90	190.
13	1975-6	LIGNITE	FF	.326	.273	.156	.445	1970.	1000.	0.74	1300.	410.	1.20	190.
14	1975-6	LIGNITE	EF	.383	.337	.154	.496	2030.	1000.	77.50	800.	800.	1.20	190.
15	1975-6	LIGNITE	EF	.479	.532	.109	.643	2080.	1500.	24.00	2500.	2500.	1.00	190.
16	1975-6	LIGNITE	EF	.310	.423	.123	.552	2080.	500.	58.30	1000.	1000.	1.30	150.
17	1975-6	LIGNITE	EF	.442	.380	.156	.537	1990.	1000.	6.70	3000.	3000.	1.50	150.
18	1975-6	LIGNITE	EF	.443	.255	.153	.463	1780.	1500.	23.50	2500.	2500.	1.60	150.
19	1975-6	LIGNITE	EF	.327	.156	.128	.391	1940.	1000.	46.60	90.	90.	2.30	109.
20	1975-6	LIGNITE	FF	.197	.096	.076	.294	1460.	1000.	0.17	24900.	7500.	0.17	109.
21	1975-6	LIGNITE	EF	.331	.202	.142	.405	2010.	1000.	48.90	70.	70.	1.20	109.
22	1975-6	LIGNITE	EF	.343	.449	.082	.531	2080.	300.	44.60	1300.	1300.	1.50	109.
23	1975-6	LIGNITE	EF	.341	.264	.152	.443	2120.	1000.	58.00	70.	70.	2.40	161.
24	1975-6	LIGNITE	EF	.321	.305	.171	.495	2060.	1000.	13.80	290.	290.	1.90	161.
25	1975-6	LIGNITE	EF	.369	.279	.157	.496	2050.	1000.	57.30	70.	70.	5.10	63.
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.	17.60	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.

*FF refers to a free-fall reactor. EF refers to an entrained-flow reactor.

Table 3-2 (Cont'd)

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- 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.
MR-37	8/16/77	SUBBTM	EF	.334	.338	.168	.506	2000.	750.	25.30	2397.	2397.	1.08	56.
MR-38	8/18/77	SUBBTM	EF	.414	.488	.065	.553	2100.	765.	20.60	2956.	2956.	0.97	56.
MR-39	8/22/77	SUBBTM	EF	.455	.497	.009	.505	2190.	750.	21.10	2868.	2868.	0.98	56.
MR- 5	8/24/77	SUBBTM	EF	.418				2090.	750.	63.50	956.	956.	1.23	56.
MR-20	9/15/77	SUBBTM	EF	.460	.365	.239	.604	1980.	1600.	18.00	3458.	3458.	0.91	56.
MR-21	9/20/77	SUBBTM	EF	.507	.438	.134	.572	2050.	1600.	17.90	3482.	3482.	0.94	56.

3.3 TASKS I AND II — BROOKHAVEN DATA COLLECTION AND ANALYSIS

Brookhaven National Laboratory has been performing an experimental study on rapid gas-phase hydrogenation (flash hydropyrolysis) of a lignite coal. Although the major emphasis in this study has been to maximize liquid hydrocarbon yield, an appreciable yield of hydrocarbon gases (mainly methane and ethane) has been obtained. The bench-scale system incorporates an entrained downflow tubular reactor, 1 inch inside diameter by 8 feet long, with a 3-foot cooling section below.

The unit is designed to feed coal at up to 2 lb/hr at design temperatures to 1,500°F and pressures to 4,000 psi. The coal used to date is a North Dakota lignite with an average particle size less than 150 microns. Preheated hydrogen mixes with the coal, and the mixture then falls through the reactor tube, which is electrically heated through the walls. A more detailed description of the reactor system has been given by Fallon.[4]

During the reporting period, all of the published[4,5,6] Brookhaven lignite data were entered into the computerized data base. A computer listing of the data is presented in Table 3-3.

The Brookhaven tests were conducted at reactor pressures of 1,000 to 3,000 psig, reactor wall temperatures of 890°F to 1,500°F (1,350°R to 1,960°R), particle residence times of approximately 2 to 12 seconds, gas residence times of approximately 11 to 56 seconds, and hydrogen-to-coal ratios of approximately 0.5 to 6 lb/lb. Gas residence time was calculated by Bechtel using the inlet reactor conditions and the reactor length. Particle residence times for the earlier 18 tests (Runs 5 through 18C) were not available.

The Brookhaven results given in Table 3-3 show overall carbon conversions rainging from 13 to 89 percent and carbon selectivity to methane

Table 3-3

BROOKHAVEN HYDROPYROLYSIS DATA

RUN DESIG-NATION	DATE	COAL TYPE	OVERALL FRACTION CARBON CONVERTED	CARBON SELEC-TIVITY TO GAS	CARBON SELEC-TIVITY TO METHANE	CARBON SELEC-TIVITY TO ETHANE	REACTOR WALL TEMP (DEG R)	HYDROGEN PARTIAL PRESSURE (PSIG)	HYDROGEN TO COAL RATIO (LB/LB)	GAS VELOCITY (FT/SEC)	GAS RESIDENCE TIME (SEC)	PARTICLE RESIDENCE TIME (SEC)
5	1976	LIG	.365	.737	.334	.164	1750.	1500.	3.38	.226	35.3	
7	1976	LIG	.301	.781	.312	.146	1750.	1500.	1.39	.239	33.4	
8	1976	LIG	.398	.721	.339	.0	1750.	1500.	5.80	.462	17.3	
9	1976	LIG	.215	.879	.265	.148	1660.	1500.	2.20	.439	18.2	
10	1976	LIG	.459	.649	.259	.137	1750.	2000.	1.48	.177	45.2	
11	1976	LIG	.171	.760	.158	.094	1570.	1500.	3.62	.415	19.3	
12	1976	LIG	.129	.977	.155	.085	1350.	1500.	4.85	.309	25.9	
13A	1976	LIG	.330	.867	.258	.139	1660.	1500.	5.63	.408	19.6	
13B	1976	LIG	.234	.855	.299	.167	1660.	1500.	0.90	.378	21.2	
14	1976	LIG	.566	.716	.387	.143	1890.	1500.	2.33	.481	16.6	
15	1976	LIG	.586	.759	.449	.089	1960.	1500.	2.80	.500	16.0	
16A	1976	LIG	.444	.722	.399	.131	1890.	1500.	0.98	.447	17.9	
16B	1976	LIG	.396	.714	.394	.134	1890.	1500.	1.40	.447	17.9	
16C	1976	LIG	.580	.705	.409	.133	1890.	1500.	1.53	.447	17.9	
17	1976	LIG	.692	.711	.397	.133	1870.	1500.	0.95	.426	18.8	
18A	1976	LIG	.860	.693	.367	.165	1830.	2100.	1.28	.286	28.0	
18B	1976	LIG	.822	.695	.354	.167	1830.	2100.	0.98	.286	28.0	
18C	1976	LIG	.888	.703	.359	.164	1830.	2100.	0.94	.286	28.0	
21	11/ 5/76	LIG	.428	.717	.348	.178	1800.	2000.	1.24	.213	37.5	8.6
22	1/13/77	LIG	.475	.680	.356	.168	1840.	2000.	1.32	.272	29.5	11.4
23	1/25/77	LIG	.448	.596	.368	.109	1910.	2000.	1.46	.240	33.4	12.2
24	1/27/77	LIG	.595	.655	.469	.094	1940.	2000.	3.62	.278	28.7	11.5
25	1/28/77	LIG	.381	.714	.336	.171	1800.	2000.	2.24	.270	29.6	11.1
26	1/31/77	LIG	.360	.647	.275	.150	1750.	2000.	2.20	.263	30.4	11.3
27	2/ 2/77	LIG	.388	.696	.317	.165	1820.	2000.	1.86	.273	29.3	11.2
28	2/ 3/77	LIG	.438	.710	.388	.148	1880.	2000.	2.29	.282	28.3	11.2
29	2/ 3/77	LIG	.358	.771	.377	.156	1880.	1500.	1.92	.342	23.4	10.5
46	4/26/77	LIG	.511	.818	.538	.115	1890.	2000.	0.42	.284	28.2	9.9
47	4/27/77	LIG	.467	.722	.358	.212	1910.	2000.	1.13	.273	29.3	8.3
48	5/ 6/77	LIG	.325	.800	.422	.178	1890.	1500.	0.66	.396	20.2	6.5

Table 3-3 (Cont'd)

RUN DESIG-NATION	DATE	COAL TYPE	OVERALL FRACTION CARBON CONVERTED	CARBON SELEC-TIVITY TO GAS	CARBON SELEC-TIVITY TO METHANE	CARBON SELEC-TIVITY TO ETHANE	REACTOR WALL TEMP (DEG R)	HYDROGEN PARTIAL PRESSURE (PSIG)	HYDROGEN TO COAL RATIO (LB/LB)	GAS VELOCITY (FT/SEC)	GAS RESIDENCE TIME (SEC)	PARTICLE RESIDENCE TIME (SEC)
49	5/ 9/77	LIG	.637	.804	.557	.104	1900.	1500.	0.97	.345	23.2	6.8
50A	5/12/77	LIG	.407	.779	.474	.135	1930.	1500.	0.91	.380	21.1	6.8
50B	5/12/77	LIG	.591	.934	.766	.076	1930.	2500.	1.04	.224	35.8	8.8
51A	5/13/77	LIG	.503	.847	.630	.093	1930.	2000.	1.08	.264	30.3	8.1
51B	5/13/77	LIG	.634	.964	.801	.091	1930.	3000.	1.26	.171	46.9	9.5
52	5/16/77	LIG	.587	.818	.555	.164	1840.	3000.	0.89	.181	44.2	9.5
53	5/17/77	LIG	.482	.869	.643	.180	1890.	3000.	1.32	.176	45.5	9.5
55	6/ 7/77	LIG	.611	.975	.881	.074	1930.	3000.	0.51	.160	50.0	9.5
56	6/15/77	LIG	.384	.792	.477	.190	1840.	3000.	0.89	.143	56.1	10.0
57	6/16/77	LIG	.492	.758	.429	.207	1830.	3000.	1.23	.150	53.5	9.9
58	6/20/77	LIG	.497	.831	.551	.111	1840.	2000.	0.53	.201	39.8	8.7
59	6/21/77	LIG	.478	.799	.502	.142	1840.	1500.	0.61	.295	27.1	7.4
60A	6/23/77	LIG	.627	.986	.871	.030	1930.	2500.	0.63	.179	44.6	9.2
60B	6/23/77	LIG	.601	.938	.837	.035	1930.	2500.	0.63	.179	11.1	2.3
61A	6/27/77	LIG	.518	.809	.519	.158	1840.	2500.	0.62	.165	48.5	9.6
61B	6/27/77	LIG	.454	.722	.445	.156	1840.	2500.	0.62	.165	12.1	2.4
62	6/28/77	LIG	.663	.807	.572	.139	1840.	3000.	0.58	.134	59.6	2.5
63	6/29/77	LIG	.353	.824	.405	.167	1840.	1000.	0.60	.438	18.3	6.4

ranging from 16 to 88 percent. The highest carbon conversion was achieved in Run 18C at 2,100 psig and 1,370°F; the highest methane selectivity and yield were obtained in Run 55.

Several tests conducted under comparable conditions of temperature, pressure, residence time, and hydrogen-to-coal ratio have resulted in some discrepancies. For example, two product samples drawn in Run 16 (16B and 16C) gave substantially different carbon conversions: one was 40 percent; the other was 58 percent. Also, comparable Runs 48 and 49 and comparable Runs 52 and 56 gave significantly different conversions for approximately the same operating conditions (see Table 3-3). Some of the discrepancies may be due to analytical errors, reactor transient behavior that results in large differences between measures wall temperature and gas/particle temperature, or fluctuations in coal feed.

Actual mass balances for carbon, hydrogen, oxygen, nitrogen, and sulfur have been presented by Brookhaven[5,6] for all the lignite tests listed in Table 3-3. Almost all tests show excellent balance closures.

3.4 TASK III — CITIES SERVICE REACTOR MODELING

The Cities Service lignite and subbituminous data received to date have been fitted to semiempirical models proposed by Bechtel for predicting carbon conversion and carbon selectivity to gaseous products.[9] A computer listing of the correlated variables is given in Table 3-2.

3.4.1 Overall Carbon Conversion

The proposed carbon conversion model was fitted to lignite data (with Run 8 excluded) and to subbituminous Runs MR-4 through MR-15.* Lignite Run 8 was excluded since its unusually high reactor pressure (2,960 psi) was well outside the region of interest of the current hydrogasification program.

A statistical analysis of the data revealed that overall carbon conversion was a function of gas temperature, gas residence time,** and hydrogen-to-coal ratio. Carbon conversion was not significantly affected by coal type, hydrogen partial pressure, particle residence time, or particle size within the region investigated. The correlation fitted to the Cities Service data is:

$$X = 1 - \exp\left[-1.14(t_{RG})^{0.146} (H_2/Coal)^{0.201} \exp(-3,960/T_G)\right] \qquad (1)$$

where,

X = overall carbon conversion, weight fraction

t_{RG} = gas residence time, milliseconds

$H_2/Coal$ = hydrogen-to-coal ratio, lb/lb

T_G = maximum gas temperature, ^{o}R

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* Only Runs MR-4 through MR-15 were available in September 1977, when the proposed model was fitted to the lignite and subbituminous data.

** For entrained-flow tests, the gas and particle residence times are nearly identical. For free-fall tests, the gas residence times are greater than the particle residence times (see Table 3-2).

Equation 1 indicates that carbon conversion increases with increase in residence time, hydrogen-to-coal ratio, and temperature within the region investigated. Statistically, Equation 1 accounts for 61 percent of the variation in the data (multiple correlation coefficient of 0.78), with a standard error of estimate of 5 percent in the predicted percent carbon conversion. Measured and predicted carbon conversions are illustrated in Figure 3-1. Note that there is no apparent effect of coal type or reactor flow regime (entrained-flow versus free-fall) on coal conversion, within the precision of the measured and predicted data.

Next, the carbon conversion for the 25 subbituminous tests shown in Table 3-2 were correlated with the reactor operating variables. A statistical analysis of the fitted data indicated that carbon conversion for the Montana Rosebud coal was a significant function of gas (or particle) residence time* and maximum gas temperature. Carbon conversion was not significantly affected by hydrogen partial pressure, hydrogen-to-coal ratio, or particle size within the region investigated. The correlation fitted to the Cities Service subbituminous carbon conversion data is:

$$X = 1 - \exp\left[-1.40\ (t_R)^{0.232}\ \exp\ (-5{,}520/T_G)\right] \qquad (2)$$

where,

X = overall carbon conversion, weight fraction

t_R = gas (or particle) residence time, milliseconds

T_G = maximum gas temperature, ^{o}R

It can be seen from Equation 2 that carbon conversion for the subbituminous coal increases with increasing gas residence time and maximum

* All of the subbituminous tests are entrained-flow tests. Hence, gas and particle residence times are nearly identical.

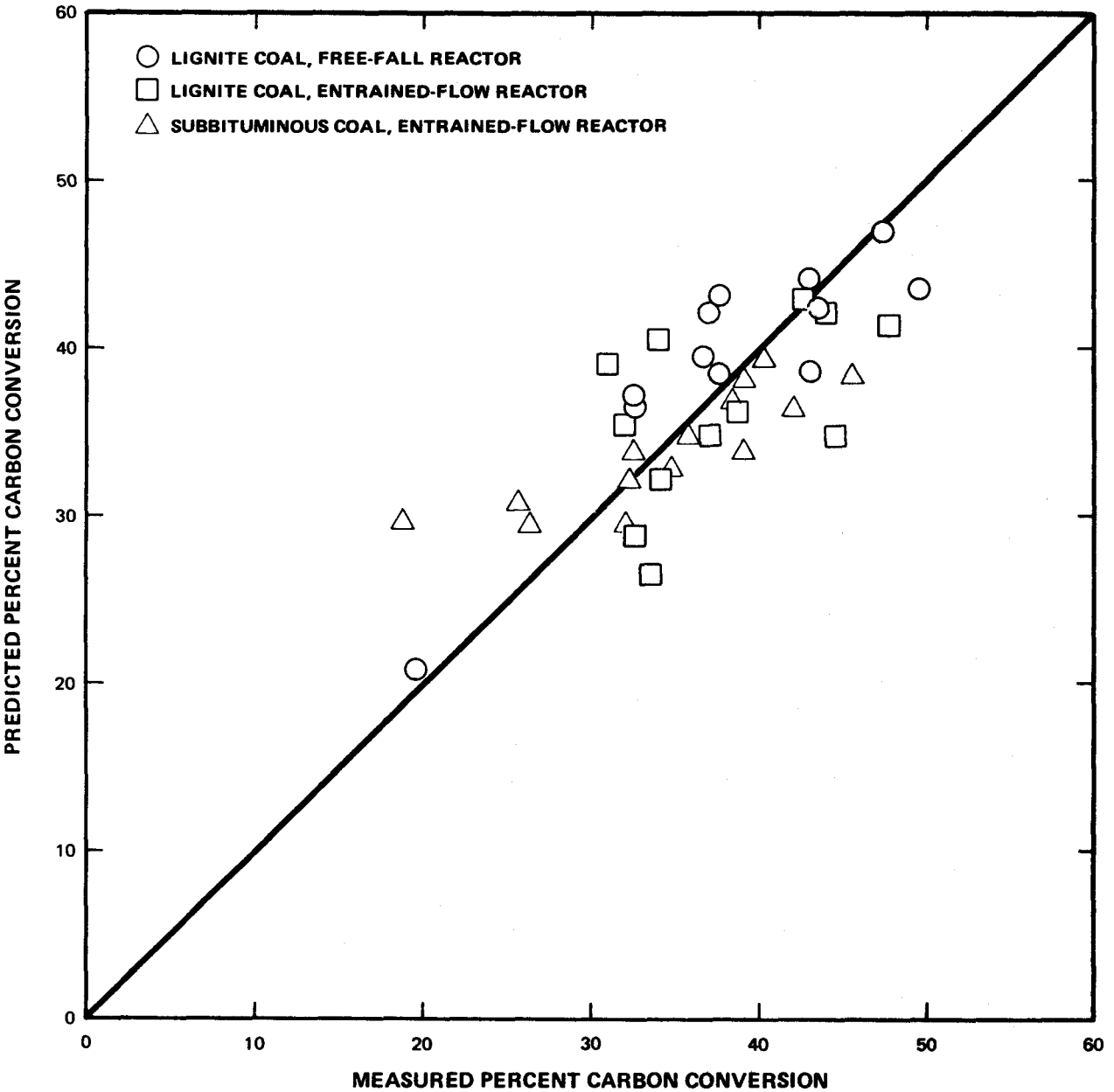

Figure 3-1. Comparison of Measured and Predicted Carbon Conversion for the Cities Service Reactor

gas temperature. Statistically, Equation 2 accounts for 71 percent of the variation in the fitted data (multiple correlation of 0.84), with a standard error of estimate of 5 percent in the predicted percent carbon conversion. The measured and predicted carbon conversions are shown in Figure 3-2. Both the statistics and Figure 3-2 indicate a somewhat poorer overall fit to the subbituminous data using Equation 2 than that obtained previously[10] when fewer subbituminous data were fitted to the same semiempirical model.

In Figure 3-3, the predicted carbon conversions from Equation 1 are plotted against the maximum gas temperature for different gas residence times. Note that a predicted carbon conversion of 50 ± 5 percent can be obtained for the Montana Rosebud coal at a maximum gas temperature of about 1,750°F and gas residence time of 2,500 milliseconds. Longer residence times (2,500 to 3,500 milliseconds) will result in the same carbon conversion at lower temperatures.

3.4.2 Carbon Selectivity to Methane

Fraction carbon selectivity to methane ϕ_M is defined as the weight of carbon converted to methane per total weight of carbon converted. When the carbon selectivity model proposed earlier by Bechtel[9] was fitted to the lignite data (excluding Run 8) and subbituminous Runs MR-4 through MR-15, a statistical analysis showed that methane selectivity was a function of gas temperature and particle residence time. Methane selectivity was not significantly affected by coal type, hydrogen-to-coal ratio, hydrogen partial pressure, gas residence time, or particle size within the region investigated. The correlation fitted to the data is:

$$\phi_M = 1 - \exp\left[-10.8(t_{RP})^{0.250} \exp(-10{,}700/T_G)\right] \qquad (3)$$

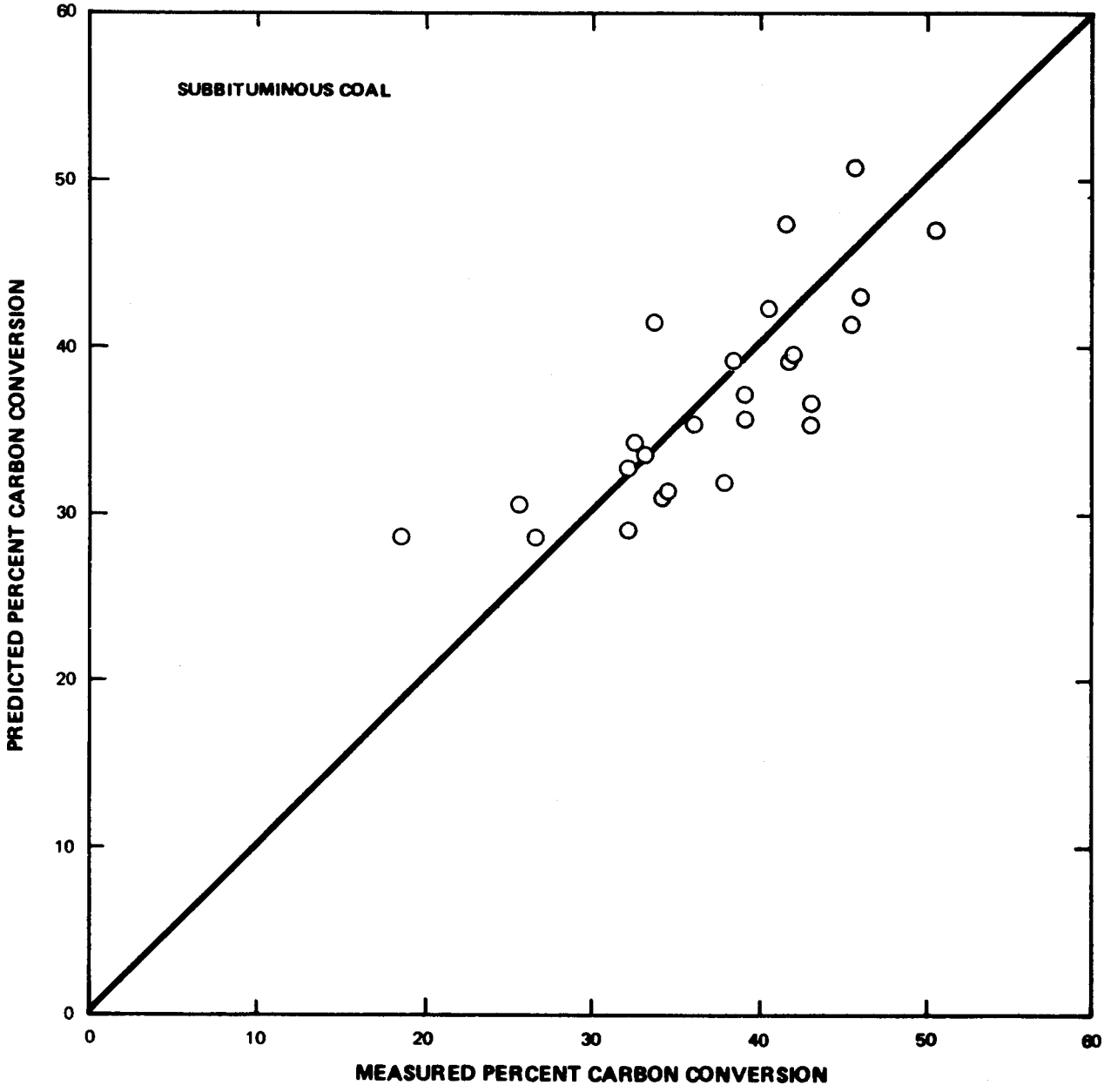

Figure 3-2. Comparison of Measured and Predicted Carbon Conversion for the Cities Service Reactor

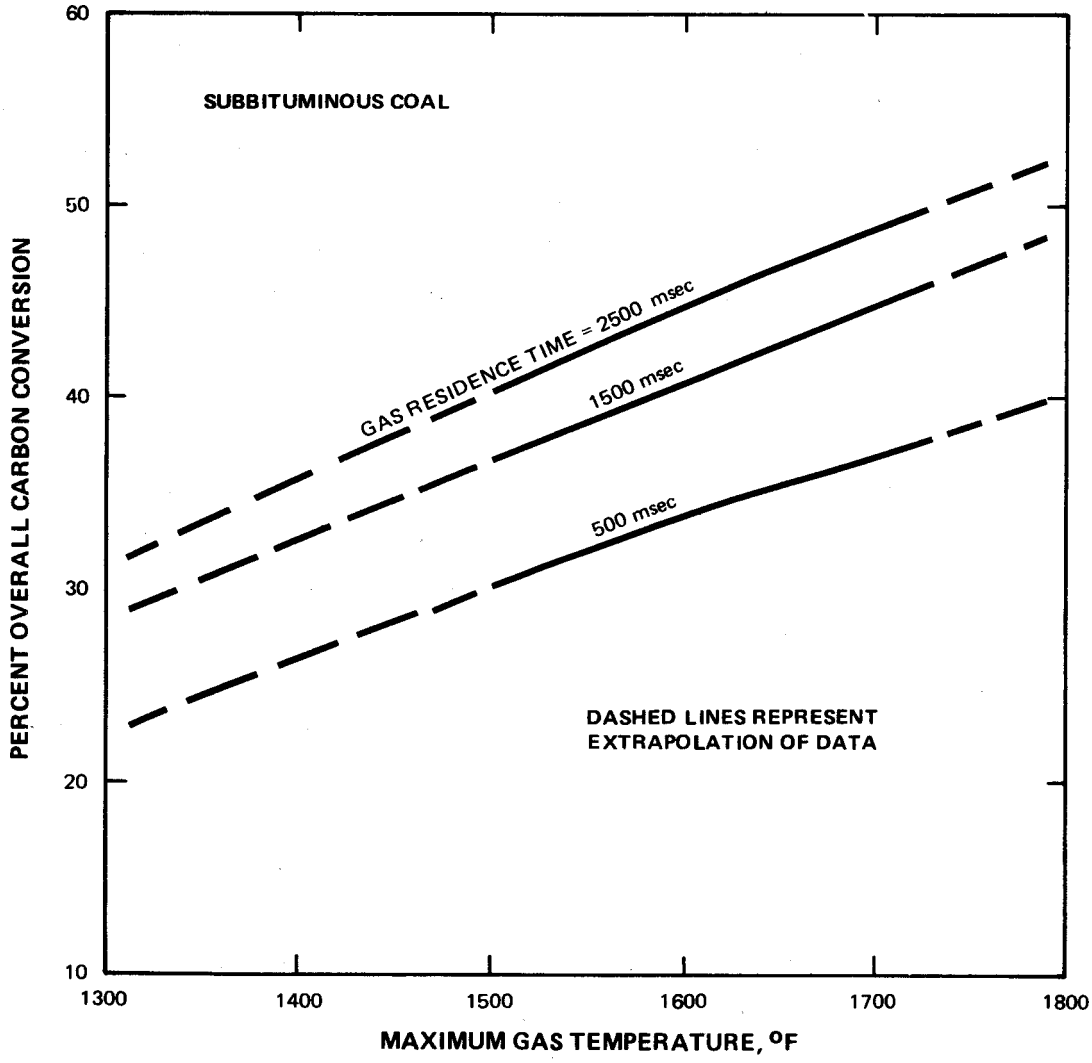

Figure 3-3. Predicted Overall Carbon Conversion for the Cities Service Reactor

where,

ϕ_M = carbon selectivity to methane, weight fraction

t_{RP} = particle residence time, milliseconds

T_G = maximum gas temperature, ^oR

As Equation 3 indicates, ϕ_M increases with increasing temperature and coal particle residence time. Statistically, Equation 3 accounts for 82 percent of the variation in the data (multiple correlation coefficient of 0.90), with a standard error of estimate of 0.05 weight fraction in the predicted fraction selectivity. Measured and predicted selectivities are illustrated in Figure 3-4. The figure shows that there is no apparent effect of coal type or reactor flow regime on methane selectivity, within the precision of the measured and predicted data.

When the carbon selectivity model proposed earlier by Bechtel[9] was fitted to the methane data from all of the 25 subbituminous tests given in Table 3-2, a statistical analysis showed that methane selectivity was a function of maximum gas temperature and gas (or particle) residence time. Methane selectivity was not significantly affected by hydrogen-to-coal ratio, hydrogen partial pressure, or particle size in the region investigated. The correlation fitted to the subbituminous data is:

$$\phi_M = 1 - \exp\left[-52.7\ (t_R)^{0.225}\ \exp\ (-12{,}900/T_G)\right] \qquad (4)$$

As can be seen from Equation 4, methane selectivity increases with increasing gas residence time and temperature. Statistically, Equation 4 accounts for 78 percent of the variation in the data (multiple correlation coefficient of 0.89), with a standard error of estimate of 0.05 weight fraction in the predicted fraction carbon selectivity to

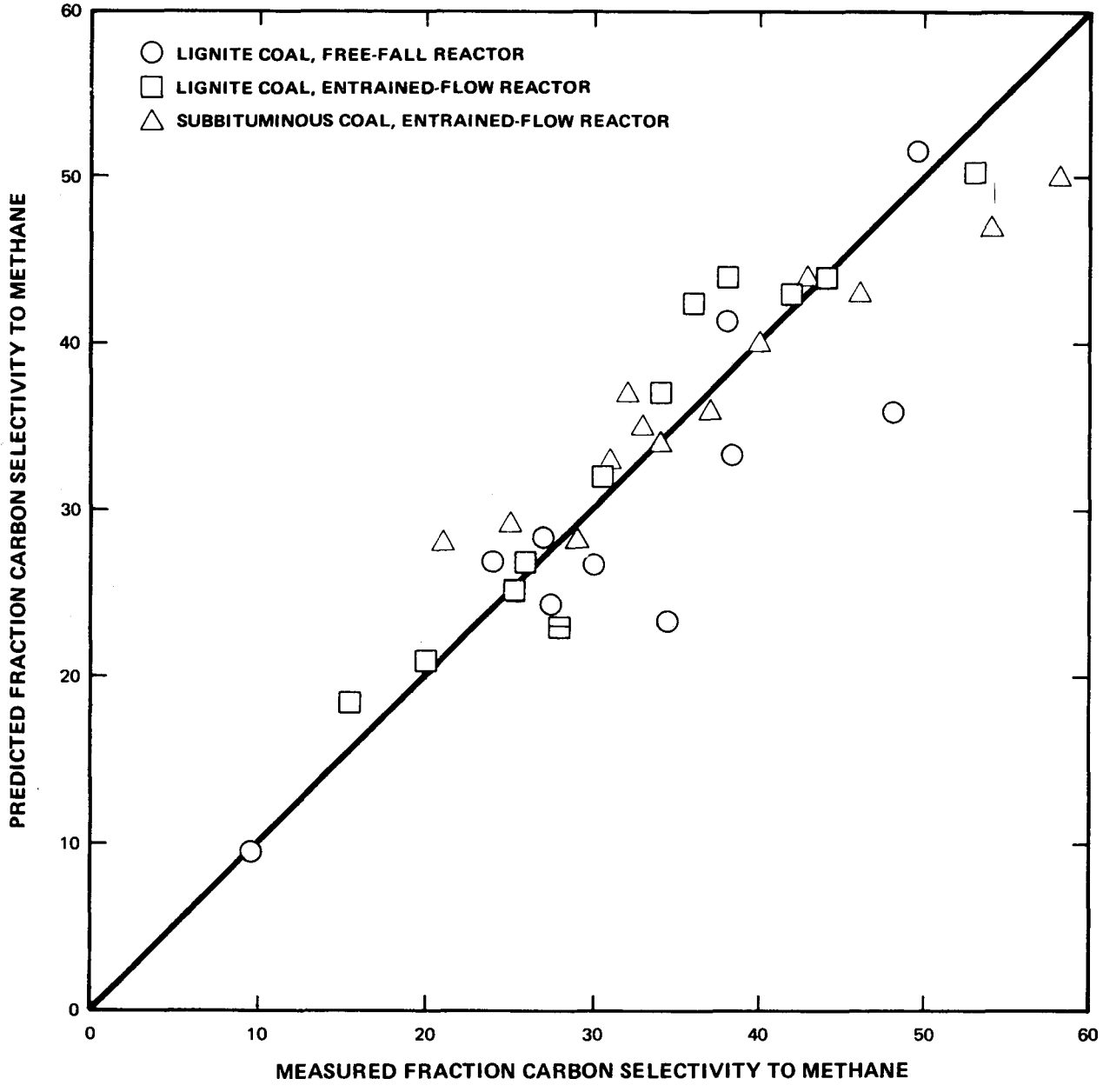

Figure 3-4. Comparison of Measured and Predicted Carbon Selectivity to Methane for the Cities Service Reactor

methane. Measured and predicted methane selectivities are plotted against one another in Figure 3-5. Runs MR-15, 37, and 39, which gave the highest measured methane selectivities of all the fitted data, appear to give the largest error in predicted methane selectivity, as shown in Figure 3-5.

In Figures 3-6 and 3-7, methane selectivity predicted from Equation 4 is plotted against the maximum gas temperature and gas residence time, respectively. Figure 3-6 shows that methane selectivities of over 50 ± 5 percent are predicted for the Cities Service reactor for temperatures in excess of 1,700°F, at a residence time of 1,500 milliseconds. Figure 3-7 shows that methane selectivities of over 55 ± 5 percent are predicted for the Cities Service reactor for residence times longer than 2,500 milliseconds, at a maximum gas temperature of 1,700°F.

3.4.3 Carbon Selectivity to Ethane

The fraction carbon selectivity to ethane ϕ_E is defined as the weight of carbon converted to ethane per total weight of carbon converted. The proposed exponential carbon selectivity model[9] gave a very poor fit to the combined lignite and subbituminous data. A statistical analysis of the fitted data showed that ethane selectivity did not appear to be significantly affected by any of the independent variables included in the fitting, i.e., temperature, hydrogen partial pressure, residence time, hydrogen-to-coal ratio, particle size, gas velocity, and coal type.

A linear selectivity model of the form

$$\phi_E = a + b_1V_1 + b_2V_2 + \ldots,^*$$

*V_1, V_2, etc., refer to correlated independent variables.

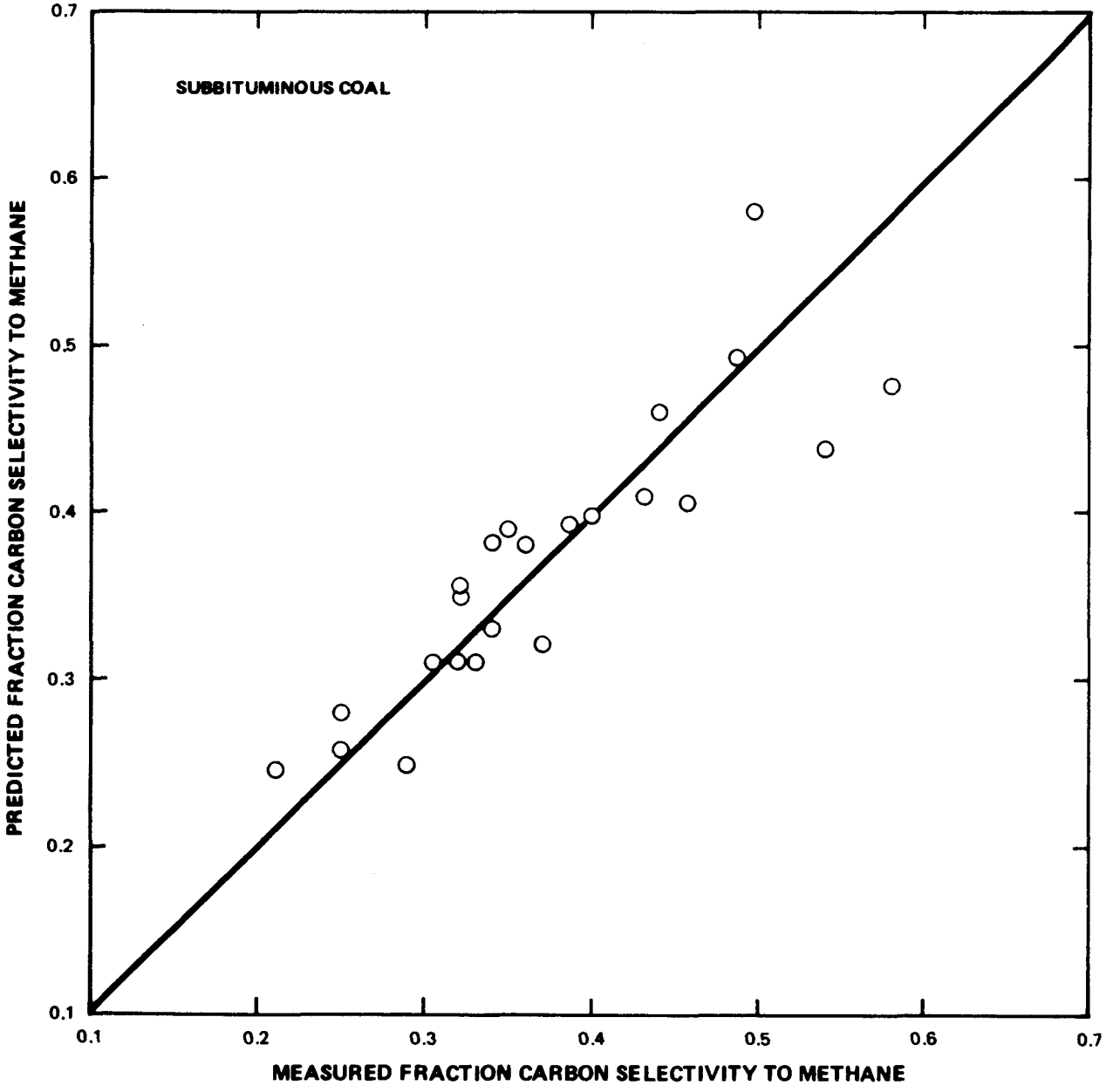

Figure 3-5. Comparison of Measured and Predicted Carbon Selectivity to Methane for the Cities Service Reactor

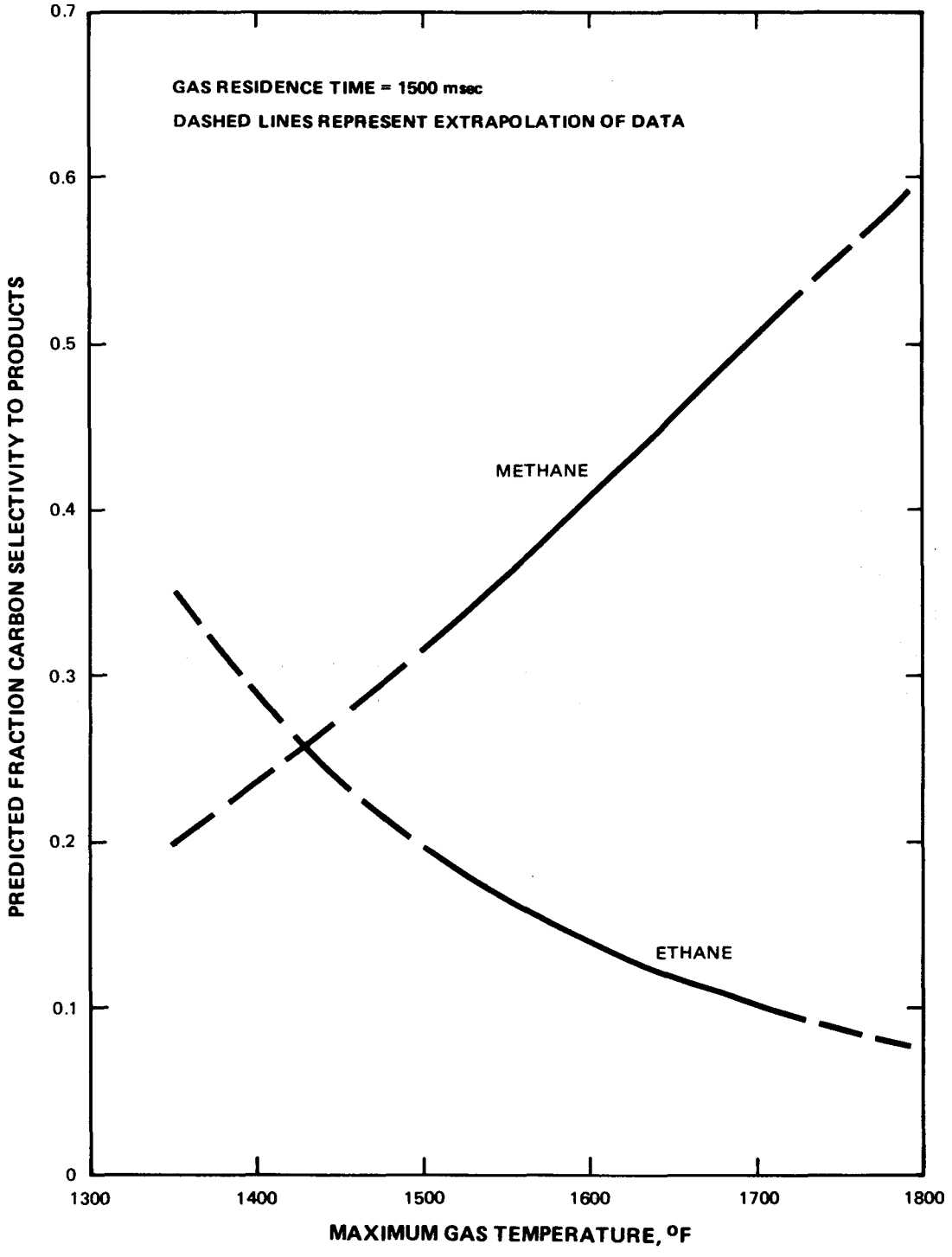

Figure 3-6. Predicted Carbon Selectivity to Methane and Ethane as a Function of Temperature for the Cities Service Reactor

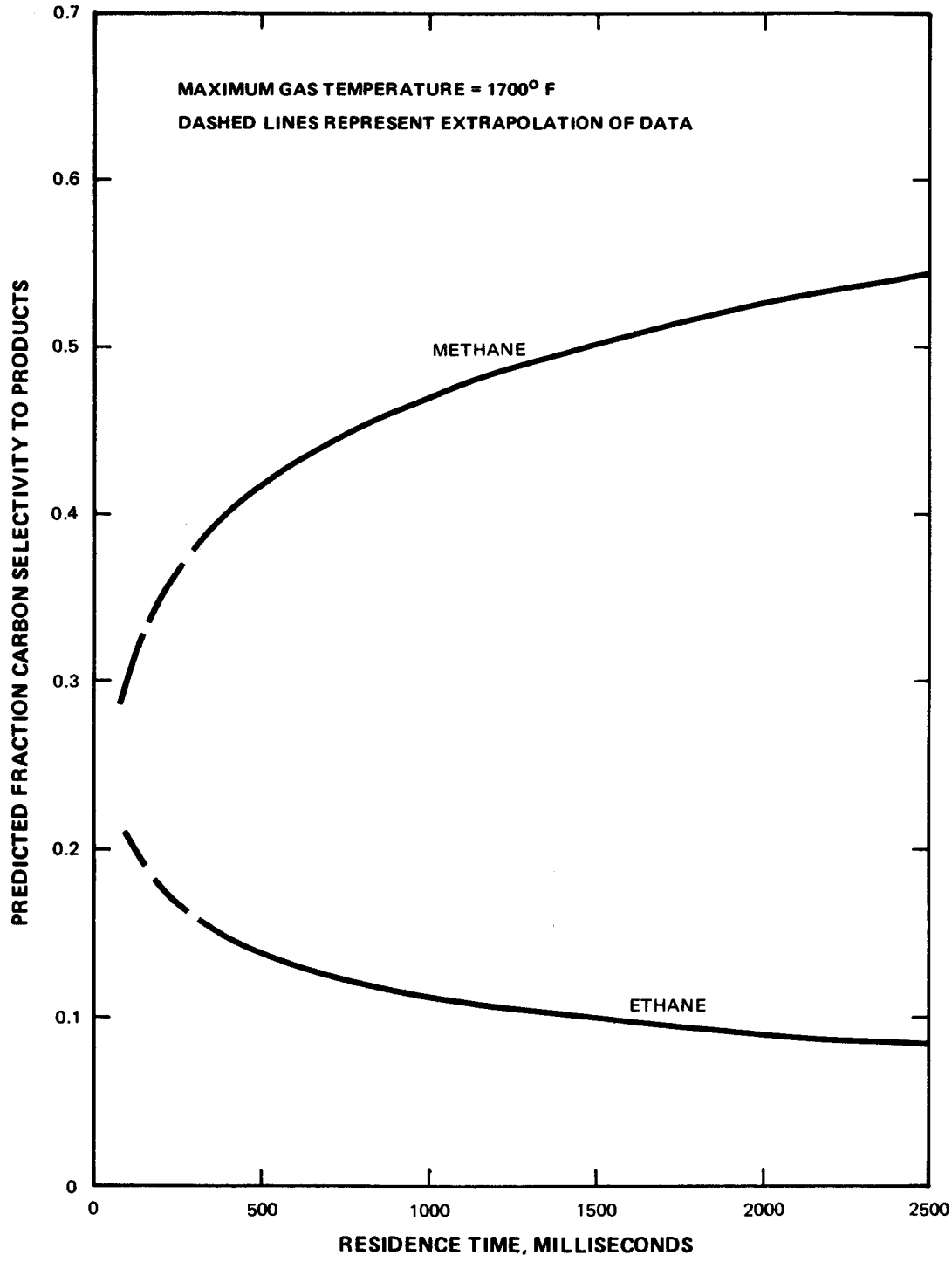

Figure 3-7. Predicted Carbon Selectivity to Methane and Ethane as a Function of Residence Time for the Cities Service Reactor

which did not fit boundary constraints, also gave a poor fit to the data. This poor fit to the data by the two models may be attributable to inaccuracies in the lignite ethane data and/or to inadequacy of the models employed.

The proposed exponential carbon selectivity model also gave a poor fit to the ethane selectivity data from the 25 subbituminous tests listed in Table 3-2. However, a much improved fit to the data resulted when the questionably low ethane selectivity of Run MR-39 (see Table 3-2) was excluded from the fitting. A statistical analysis of the fitted data revealed that ethane selectivity was a function of maximum gas temperature and gas residence time. Hydrogen partial pressure, hydrogen-to-coal ratio, and particle size did not have a significant effect on ethane selectivity within the region investigated. The correlation fitted to the subbituminous data is:

$$\phi_E = 1 - \exp\left[-0.000505\ (t_R)^{-0.261}\ \exp(15{,}700/T_G)\right] \qquad (5)$$

As can be seen from Equation 5, ethane selectivity for the Montana Rosebud coal decreases with increasing residence time and temperature, thus suggesting possible secondary cracking of ethane at higher temperatures and extended residence times. Statistically, Equation 5 accounts for 77 percent of the variation in the data (multiple correlation coefficient of 0.88), with a standard error of estimate of 0.05 weight fraction in the predicted fraction selectivity to ethane. Measured and predicted ethane selectivities for the Cities Service reactor are plotted in Figure 3-8.

In Figures 3-6 and 3-7, the ethane selectivity predicted from Equation 5 is plotted as a function of the maximum gas temperature and gas residence time, respectively. The two plots indicate that at temperatures in excess of 1,700°F or at residence times longer than about 2 seconds, the predicted carbon selectivity to ethane is less than 10 percent.

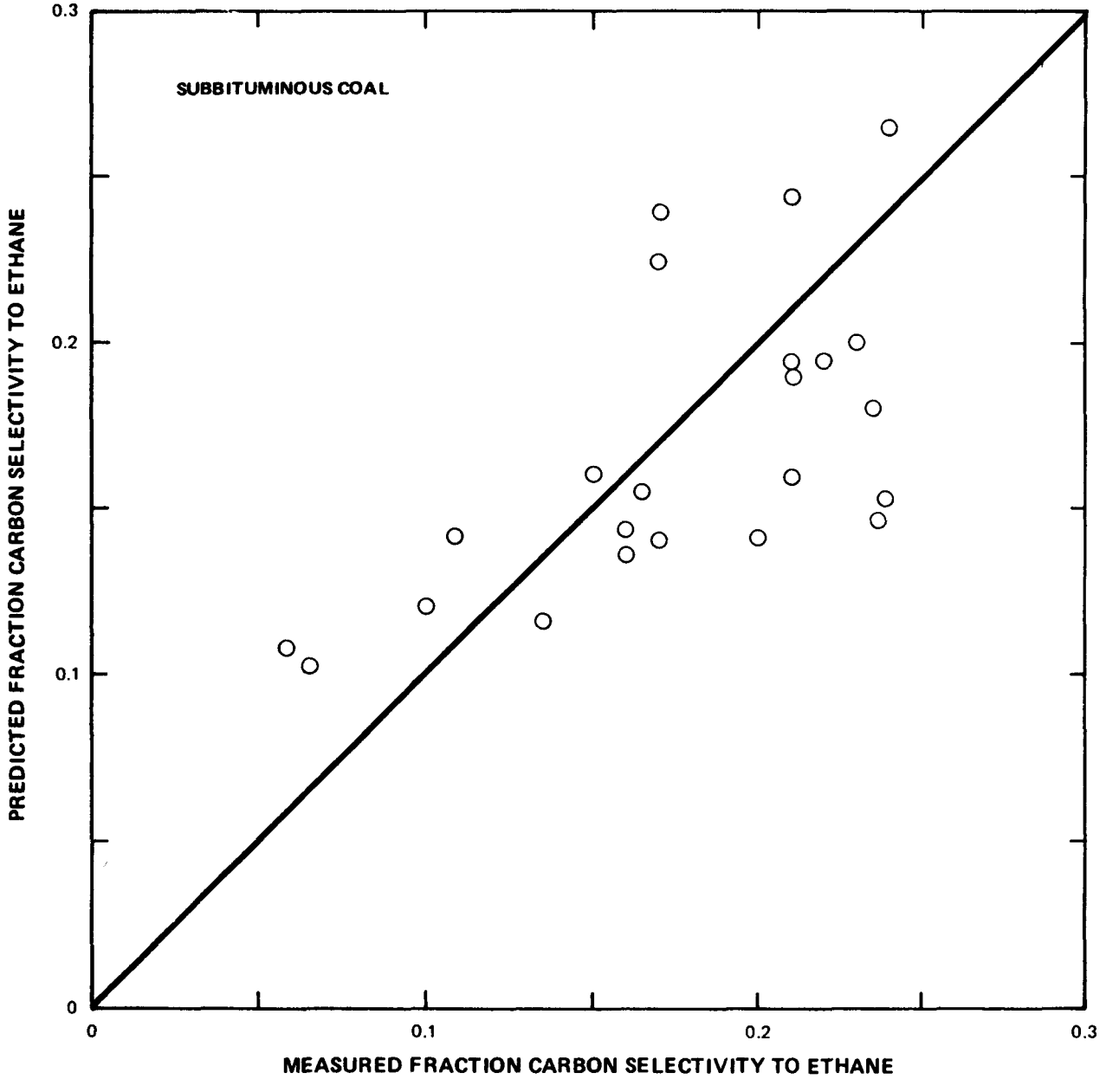

Figure 3-8. Comparison of Measured and Predicted Carbon Selectivity to Ethane for the Cities Service Reactor

3.5 TASK III — ROCKETDYNE REACTOR MODELING

The correlation developed to predict the overall carbon conversion for the Cities Service subbituminous tests (Equation 2 in this report) was used to predict the overall carbon conversion for the six Rocketdyne subbituminous tests conducted in the 1/4-ton/hr reactor (see Table 3-1).

The predicted and measured carbon conversions for the Rocketdyne subbituminous tests are shown in Figure 3-9, along with the predicted and measured conversions for the Cities Service subbituminous tests. Aside from the result for Run 011-4 at a measured carbon conversion of 33 percent (see Table 3-1), the predicted and measured conversions for the Rocketdyne subbituminous tests are in excellent agreement. The substantial disagreement between the measured and predicted conversion for Run 011-4 may be attributed to the short duration (3 minutes) of the test, as discussed in Subsection 3.1.

Figure 3-9 clearly suggests that the Cities Service bench-scale reactor and the Rocketdyne 1/4-ton/hr reactor achieve similar carbon conversions with Montana Rosebud coal under comparable operating conditions within the region investigated, i.e., the predicted results for the Rocketdyne tests are well within the standard error of estimate of the Cities Service correlation. Of course, as more subbituminous data are generated by Cities Service and Rocketdyne, the comparative behavior of the two reactors will be verified further.

The correlation developed earlier by Bechtel to predict the overall carbon conversion for the Rocketdyne bituminous tests in the 1-ton/hr reactor (Equation 5 in Bechtel's June-August 1977 Quarterly Progress Report[9]) was used to predict the carbon conversion for the four recent bituminous tests made in the 1/4-ton/hr reactor (see Table 3-1). The conversions predicted from this correlation were, however, consistently higher (about 12 percent) than the measured conversions reported for the four recent bituminous tests. Rocketdyne[1] has attributed this

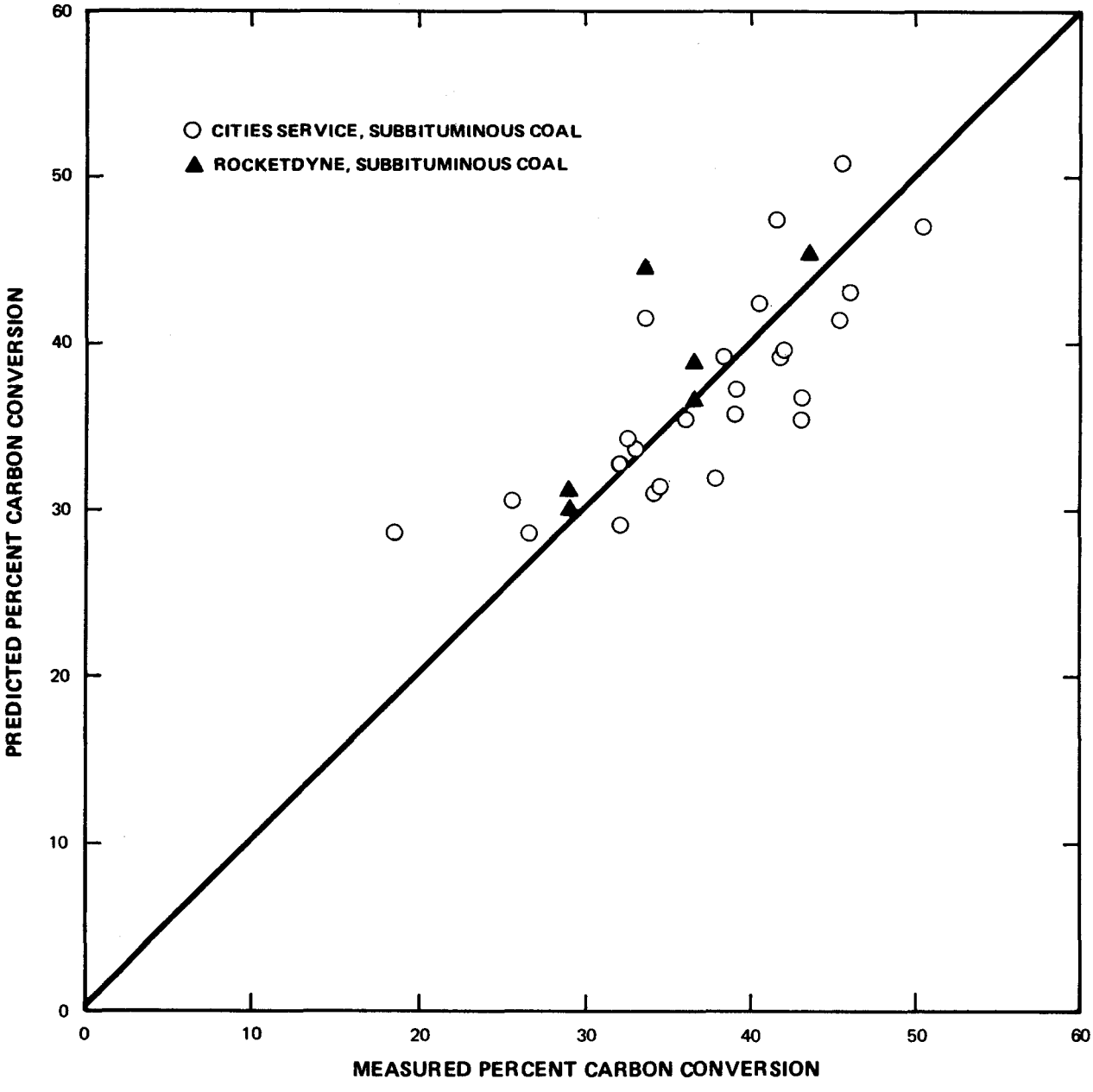

Figure 3-9. Comparison of Measured and Predicted Carbon Conversion for the Cities Service and Rocketdyne Reactors

discrepancy to probable uncertainties in the results of the recent 1/4-ton/hr bituminous coal tests. These uncertainties may be due to the relatively short duration (3 minutes) of the tests.

When all the bituminous data from the two reactors (see Table 3-1) were fitted to the semiempirical model proposed earlier for carbon conversion,[9] a poor fit resulted, as was expected. When more data from longer duration bituminous tests in the 1/4-ton/hr reactor are available, the bituminous conversion results from the two Rocketdyne reactors will be compared further.

3.6 TASK III — BROOKHAVEN REACTOR MODELING

The carbon conversion data from the 48 lignite hydropyrolysis tests given in Table 3-3 were fitted to the semiempirical carbon conversion model proposed earlier by Bechtel.[9] A statistical analysis of the fitted data showed that the variation in carbon conversion was mainly due to the variations in reactor wall temperature and hydrogen partial pressure within the region investigated. However, the resulting fit was poor, as indicated by a multiple correlation coefficient of 0.66 and a standard error of estimate of 12 percent in the predicted percent carbon conversion.

The poor fit for carbon conversion may be attributable to the apparent inconsistencies in results from several tests conducted under comparable conditions of temperature, pressure, residence time, and hydrogen-to-coal ratio. These inconsistences have been discussed in Subsection 3.3.

As more information and data are obtained from Brookhaven National Laboratory, correlating the Brookhaven lignite data will be continued.

3.7 TASK IV — ROCKETDYNE AND CITIES SERVICE EXPERIMENTAL DESIGNS

The goodness of fit of the models proposed for carbon conversion and carbon selectivity depends primarily on three factors: (1) the accuracy of the functional form of the proposed models within the range of the data generated, (2) the experimental error associated with the generated data, and (3) the design of the experiments.

The experimental error and the variability associated with generated data are often estimated from results of replicate runs. (Replicate runs have identical levels of the independent variables.) Unfortunately, replicate runs have not been included in the proposed experimental designs[11,12] for the current Rocketdyne and Cities Service hydrogasification programs.* It is recommended that replicate runs be added to the Rocketdyne and Cities Service test matrices for each coal studied. If possible, the replicate runs should be conducted at the mid-range of the independent variables investigated.

A statistical analysis of the Cities Service subbituminous data has indicated that carbon conversion and carbon selectivity to products are not significant functions of hydrogen partial pressure over the pressure range 750 to 1,600 psig (see Equations 2, 4, and 5 in Subsection 3.5). Since it may be advantageous to operate a commercial-scale hydrogasifier at reduced pressure while maintaining 50 percent carbon conversion, it is important to verify the results of the statistical analysis for the lower pressures. Therefore, it is recommended that additional tests be conducted by Cities Service at hydrogen partial pressures of 750 and 500 psig, with a nominal gas temperature and residence time of 1,700°F and 3,500 milliseconds, respectively.

*The fact that good carbon material balance closures have been obtained for the Cities Service subbituminous tests strongly suggests, however, that the experimental error in measured carbon conversion and selectivity is small for each run at Cities Service.

3.8 TASK III — CONCEPTUAL DESIGN BASIS FOR A FULL-SCALE HYDROGASIFICATION REACTOR

This subsection describes the basis for the selection of operating variable levels and size constraints for the hydrogasification stage of a proposed full-scale reactor facility for converting coal to pipeline-quality gas. As presently envisioned, the reactor facility will consist 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.

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.[13] A sketch and a detailed description of the conceptual hydrogasification reactor are presented in Subsection 3.10.

Bechtel had previously developed a reference design basis for a conceptual full-scale hydrogasification reactor stage.[9] This design basis was developed employing data gathered in the Rocketdyne 1-ton/hr reactor using Kentucky HvAb coal,[14] together with predictive reactor performance models fitted to the data by Bechtel.[9] 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 parameters 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 models fitted to the data by Bechtel. As shown earlier in this report, the models fitted to the Cities Service subbituminous data 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
Maximum reactor temperature	1,800°F
Overall carbon conversion	50 percent

Calculated Operating Parameters:

Hydrogen injection temperature	1,900°F
Coal-hydrogen mix temperature	1,300°F
Hydrogen-to-coal ratio	0.40 lb/lb
Nominal gas (or particle) residence time	1,700 milliseconds
Carbon selectivity to methane	61 percent
Carbon selectivity to ethane	7 percent

The coal type and size selected are those used in the recent Cities Service and Rocketdyne testing; the average coal composition has been

given elsewhere.[11] 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 given earlier in this report showed that carbon conversion and carbon selectivity to products were 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[15] for a maximum coal capacity for a single injector element of 3 tons/hr and a maximum number of 36 injector elements per head.

The selected hydrogen-to-coal ratio of 0.4 lb/lb is within the lower range investigated by Rocketdyne. Note that a statistical analysis of the Cities Service coal data presented earlier in this report showed that carbon conversion and carbon selectivity to products were relatively unaffected by hydrogen-to-coal ratio within the region investigated.

The selected maximum reactor temperature (exit gas temperature) of 1,800°F is at the upper range of temperature studied by Rocketdyne in the 1-ton/hr and 1/4-ton/hr reactors (see Table 3-2). This temperature is required to achieve 50 percent carbon conversion for subbituminous coal at gas residence times less than 2 seconds.

An overall carbon conversion of about 50 percent was chosen because previous studies[13,16] had shown that this value approximates the desired conversion level for an overall balanced process. A balanced process is a process in which the unreacted char from the hydrogasification stage is further gasified (probably with steam and oxygen) to make the required process hydrogen. The maximum carbon conversion achieved so far in the Cities Service subbituminous testing is 51 percent (see Run MR-21 in Table 3-2).

The coal-hydrogen mix temperature (reactor gas inlet temperature) of 1,300°F was calculated from a heat balance around the reactor for an exit gas temperature of 1,800°F. The heat balance included the heat released during the coal-hydrogen exothermic reaction, and assumed adiabatic reactor operation.

The hydrogen injection temperature of 1,900°F was calculated from a heat balance around the coal-hydrogen mixing injector nozzle, assuming that coal is fed at 77°F, and using the coal-to-hydrogen ratio and coal-hydrogen mix temperature given above. As shown by Rocketdyne,[13,14] this injection temperature can be readily achieved by combusting preheated hydrogen with a relatively small amount of oxygen in a preburner placed ahead of the reactor injection head.

The nominal gas (or particle) residence time for the entrained-flow reactor was computed using the correlation given earlier in this report (Equation 2 in Subsection 3.5) for predicting carbon conversion for the subbituminous coal Cities Service tests. (Note that the Cities Service and Rocketdyne subbituminous tests are in substantial agreement at comparable operating conditions.) The residence time t_R was obtained by substituting the selected carbon conversion and maximum reaction temperature into the correlation:

$$0.50 = 1 - \exp\left\{-1.40\ (t_R)^{0.232}\ \exp\left[-5{,}520/(1{,}800 + 460)\right]\right\}$$

$$t_R = 1{,}700 \text{ milliseconds}$$

Carbon selectivity to methane and ethane were calculated using the correlations fitted to the Cities Service subbituminous data presented earlier in this report (Equations 4 and 5 in Subsection 3.5). From these correlations, carbon selectivity to methane and ethane were found to be 61 and 7 percent, respectively.

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 932,000 ft^3/hr (258 ft^3/sec).

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

$$S = V_G/u_G = 258/u_G \tag{6}$$

and

$$L = t_R u_G = 1.70\ u_G \tag{7}$$

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[15,17] and the conditions tested at Cities Service and Rocketdyne. At 10 ft/sec gas velocity, the required reactor cross-sectional area from Equation 6 is 26 ft^2, and the required reactor length from Equation 7 is 17 feet. At 25 ft/sec gas velocity, the required cross-sectional area is 11 ft^2, and the required length is 43 feet.

3.9 TASK III — CONCEPTUAL DESIGN OF A FULL-SCALE HYDROGASIFICATION REACTOR

As discussed in the previous subsection, a full-scale reactor facility will consist 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. In this section, a conceptual design of a full-scale hydrogasification section is presented in detail, followed by a discussion of the char gasification stage.

3.9.1 Hydrogasification Stage

A detailed sketch of the conceptual full-scale hydrogasification reactor stage is shown in Figure 3-10. The hydrogasification reactor vessel consists of two sections. The upper section of the vessel contains a shell and tube heat exchanger, and the lower section includes a hydrogasifier and a cyclone separator. As discussed in the previous subsection, the hydrogasification reactor would have a length roughly between 15 and 45 feet, depending on the gas velocity.

In the hydrogasification section, hot hydrogen at 1,900^{o}F is contacted with coal feed at 77^{o}F in a total of 36 mixing-injection nozzles; each nozzle handles a maximum of 3 tons of coal per hour, as has been discussed in Subsection 3.9. 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 the 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 separated in a cyclone which sends the product gas stream back up through

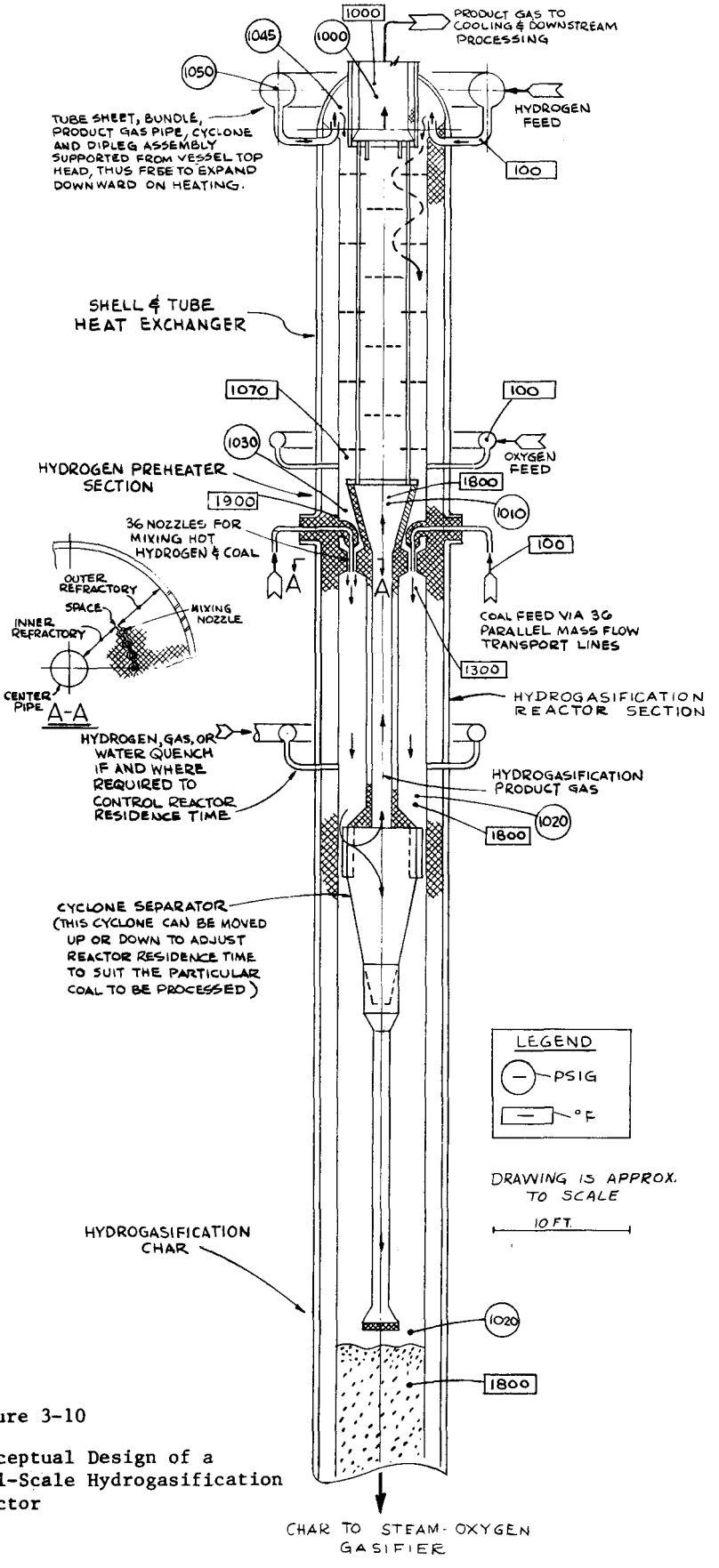

Figure 3-10

Conceptual Design of a Full-Scale Hydrogasification Reactor

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 hydrogen production. 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°F to about 1,000°F by heat exchange with cold feed hydrogen flowing downward through the exchanger shell side. This hydrogen stream is assumed to enter at 100°F and is heated to about 1,100°F.

The hydrogen effluent from the exchanger is further heated to about 1,900°F by combustion with oxygen, which is injected into the hydrogen stream near the exchanger outlet, as shown in Figure 3-10. 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 3-10.

The reactor vessel shell shown in Figure 3-10 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.

Other approaches to reactor vessel shell design are being considered. One approach will be to use a pressurized water jacket inside the vessel strength shell to keep the metal temperature as low as 550°F.

3.9.2 Char Gasification Stage

Efficient hydrogen generation from the hydrogasifier char product will have a major impact on process thermal efficiency and economics. Gasification of the unreacted char with steam and oxygen at temperatures of 1,800°F to 2,700°F will generally produce a gas consisting mostly of carbon monoxide and hydrogen. Further processing of this raw gas (shift and purification) will yield the process hydrogen required for the hydrogasification stage.

The following versions of the char/oxygen/steam reactor design should be investigated:

- An entrained-downflow reactor incorporating Rocketdyne-type injectors for rapid mixing of the reactants
- A dense-phase, fluid-bed reactor similar in design to the IGT-HYGAS char gasifier
- A Texaco high-pressure, entrained-flow gasifier
- A Koppers-Totzek entrained-flow gasifier

As an example, if a dense-phase, fluid-bed reactor scheme is considered, the char solids from the hydrogasifier would normally be transferred at the hydrogasifier pressure (about 1,000 psig) to the char gasifier via a standpipe. If the fluid-bed nominal temperature equals that of the incoming char (about 1,800°F), several minutes[18] of holding time will probably be required to produce acceptable conversion of char.

Oxygen and steam would enter the char fluid bed via a gas distributor manifold near the bottom of the reactor, and the product gas would leave the reactor at the top through a cyclone separator. The entrained fines

would be collected and returned to the reactor by the cyclone dipleg and the oxygen carrier stream. The spent char (mostly ash) would leave the fluid-bed reactor at the bottom and go to a quench pot, where it would be sprayed with sufficient water to make up a slurry suitable for transfer to pressure letdown and eventual disposal.

The above considerations suggest that the fluid-bed char gasifier operating at 1,800°F could be roughly 100 to 150 feet high. Higher reactor operating temperatures (2,500°F to 2,700°F) could reduce reaction time (holding time) and reactor size, but the reactor would be operating in the slagging region.

3.10 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 test 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

CONCLUSIONS

Semiempircal correlations, based on presently available data from Rocketdyne, Cities Service, and PERC, can be developed to predict carbon conversion efficiency and carbon selectivity to products for the reactor systems. The fitted models show that the Cities Service bench-scale reactor and the Rocketdyne 1/4-ton/hr reactor achieve similar carbon conversions with Montana Rosebud subbituminous coal under comparable operating conditions.

Substantially higher carbon conversions were obtained in Rocketdyne tests with bituminous coal than in Rocketdyne and Cities Service tests with subbituminous coal. In Cities Service tests, lignite and subbituminous coals achieved similar carbon conversions at comparable operating conditions.

An overall carbon conversion of about 50 percent is required for a balanced system, where char by-product is gasified to make the process hydrogen. The data indicate that a balanced process can be achieved with the Rocketdyne reactor using bituminous and subbituminous coals, with the Cities Service reactor using lignite and subbituminous coals, and with the PERC reactor using lignite coal.

Preliminary operating variable levels and size constraints have been selected for the design of a conceptual full-scale, entrained-downflow hydrogasifier. These levels and constraints are based on data gathered in the Cities Service and Rocketdyne reactors using subbituminous coal,

together with the reactor performance models fitted to the data. For 50 percent carbon conversion at a reactor exit gas temperature of 1,800°F, the required gas residence time would be 1.7 seconds. For a coal rate of 108 tons/hr and a superficial gas velocity of 15 ft/sec, the required reactor cross-sectional area would be 17 ft^2 and the required reactor length 25 feet.

A conceptual design has been presented for a full-scale hydrogasification reactor using subbituminous coal, based on the selected operating variables and size constraints. At the reactor inlet, hot hydrogen at 1,900°F is contacted with coal in a total of 36 mixing-injection nozzles. The nozzle design is similar to that developed and used by Rocketdyne. Char and product gas flow downward in an entrained-flow manner through the annuli formed by the inner wall of the reactor vessel shell and the outer shell of a central pipe (or duct) through which the product gas leaves the gasifier. The exiting coal-char solids and gas stream are separated in a cyclone, which can be moved vertically to control the residence time of char and gas inside the reactor. Product gas from the cyclone flows upward through the tube side of an integrated shell and tube heat exchanger, where it is cooled by heat exchange with hydrogen flowing downward through the exchanger shell side.

Bechtel has recommended that replicate runs be added to the Rocketdyne and Cities Service experimental design, to directely determine experimental error. Since results to date have indicated that carbon conversion and carbon selectivity are not significant functions of hydrogen partial pressure, Bechtel has recommended that additional tests be conducted at lower levels of hydrogen partial pressure (i.e., 500 and 750 psig). Although there appears to be sufficient data to scale up the Rocketdyne and Cities Service reactors for subbituminous and lignite coals, additional data are required to determine the effect of bituminous coals on reactor performance.

Section 5

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Appendix

VISITS TO THE CITY COLLEGE OF NEW YORK AND BROOKHAVEN NATIONAL LABORATORY

During November 1977, Michael Epstein of Bechtel visited with personnel from the City College of the City University of New York (CUNY) and Brookhaven National Laboratory. The objectives of the visits were to: (1) discuss the ongoing technical programs, (2) obtain additional data, and (3) observe the test facilities. The information gathered during these visits is summarized below.

A.1 THE CITY COLLEGE OF NEW YORK

On November 15, 1977, Michael Epstein of Bechtel met with Professor Robert A. Graff of the Chemical Engineering Department of the City College of New York. The City College of New York has developed a laboratory reactor system for the determination of products obtained from the flash heating of coal in flowing hydrogen at pressures up to 100 atmospheres and temperatures up to 1,000^{o}C.

The reactor system is designed for independent control of three important time parameters: (1) the time period over which the coal is heated from room temperature to a selected final temperature (heating rate), (2) the time period over which the vapor products evolved from the coal are maintained at temperature (vapor residence time), and (3) the time allowed for the reaction of coal and derived species not vaporized (solids contact time).

In a conventional experiment, about 10 milligrams of powdered coal are deposited as a thin ring at a selected position in a reactor tube of 316 stainless steel, 6.35 millimeters in outside diameter, 5.1 milli-

meters in inside diameter, and approximately 300 millimeters in length. Hydrogen at pressure continuously flows through the tube to downstream analytical equipment. A high current is passed through the tube wall to flash-heat the coal and hydrogen to reaction temperature. After the temperature is reached, a controller switches to a lower current to maintain the reactor temperature for the duration of the run.

The college has published[7] results of various tests that show the effect of the three time parameters on overall carbon conversion, and fraction carbon conversion to gas and liquid products. Additionally, a comparative study of a suite of coals broadly representative of the U.S. spectrum has been carried out. Results of this testing have also been published.[8]

During the visit, Professor Graff made available to Bechtel recent data obtained at the college that showed the effect of the time parameters and coal type on carbon conversion and product distribution. A tour was made of the test facility, and a run was observed in the course of which liquid products were condensed in a liquid nitrogen trap downstream of the reactor tube.

A.2 BROOKHAVEN NATIONAL LABORATORY

On November 16, 1977, Michael Epstein of Bechtel met with Meyer Steinberg, Peter Fallon, and Barrat Baht of the Brookhaven National Laboratory. Brookhaven has developed a coal flash hydropyrolysis unit to obtain process chemistry data for a noncatalytic, gas-phase, coal liquefaction process. The unit is described in Subsection 3.3 of this report. Also presented in Subsection 3.3 are results of testing at the laboratory during 1976 and 1977.

During the visit, recently generated data obtained at the facility were presented to Bechtel. Meyer Steinberg informed Bechtel that particle residence times for Runs 5 through 18C (see Table 3-3) would be calcu-

lated and available in the near future. Bechtel was told that gas temperatures are now being measured within the reactor. (In earlier tests, only wall temperature was measured.) There is not much difference between gas and wall temperatures once steady state has been reached. Significant differences between wall and gas temperatures, however, have been observed during line-out in certain of the runs with high methane yield.

The Syntron vertical vibratory coal feeder has been replaced by a screw-type feeder. In recent tests, product samples have been obtained at various locations along the length of the reactor. This permits the direct determination of the effect of gas and particle residence time on carbon conversion and carbon selectivity for a specific set of operating conditions. Brookhaven has defined an "effective reactor efficiency," which takes into account the heat content of the gaseous and liquid products, and the heat content of the coal and consumed hydrogen. (For the calculation, all of the BTX liquid product is assumed to be benzene.)

The discrepancies in measured values for carbon conversion in some of the runs having similar operating conditions were discussed at the meeting. (The problem is described in detail in Subsection 3.3 of the report.) Some of the discrepancies may be due to analytical errors, reactor transient behavior that results in large differences between measured wall temperature and gas/particle temperature, or fluctuations in coal feed rate.

At the conclusion of the visit, a tour was made of the reactor facility.

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