

# **AN ANALYSIS OF COAL HYDROGASIFICATION PROCESSES**

**Monthly Technical Progress Report  
for the Period  
1 September - 30 September 1977**

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San Francisco, California 94119  
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## ABSTRACT

This monthly Technical Progress Report covers work performed during the period 1 September 1977 to 30 September 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 September, substantial progress was made on Tasks I, II, and III. Data from nine recent Cities Service hydrogasification tests using Montana Rosebud subbituminous coal were entered into a computerized data base. This base contained data from earlier Cities Service tests using lignite coal and recent Cities Service tests using the subbituminous coal. Semiempirical correlations for predicting carbon conversion efficiency, carbon selectivity to methane, carbon selectivity to ethane, and carbon selectivity to hydrocarbon gas were fitted to the data. Coal type did not appear to have any significant effect on carbon conversion or carbon selectivity. Carbon selectivity to methane increased with increasing gas temperature and residence time; carbon selectivity to ethane (and higher hydrocarbon gases) decreased with increasing gas temperature and residence time. In addition, a thermal efficiency for the Cities Service reactor was defined in terms of the primary desirable gaseous hydrocarbon products, the coal feed, and the hydrogen consumed.

During September, data from 18 tests conducted at the Brookhaven National Laboratory for the rapid gas-phase hydrogenation of coal were entered into the computerized data base. The results from the tests have been tabulated and discussed.

The design of a commercial-scale hydrogasification reactor is continuing. The unit will have a configuration similar to the Rocketdyne reactor assembly, consisting mainly of a preburner, injector element, and an entrained-flow reaction chamber.

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## Section 1

### OBJECTIVES AND SCOPE

This report is the September 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 September 30, 1977. During September, substantial progress was made on Tasks I, II, and III. Actual manhours expended in September were 670; budgeted manhours were 750. 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.

REPORT PERIOD: 1 Feb - 30 September 77

TASK NO.	WORK STATEMENT	SCHEDULE												
		February	March	April	May	June	July	August	September	October	November			
I	DATA COLLECTION	-----			-----	①								
II	DATA ANALYSIS		-----	-----		-----	②							
III	PROCESS MODELING			-----	-----	-----	-----	-----	-----	③				
IV	IDENTIFICATION OF ADDITIONAL DATA AND RECOMMENDED EXPERIMENTAL PROGRAMS							-----	-----	-----	④			
-	FINAL REPORT										-----	-----	⑤	⑥

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

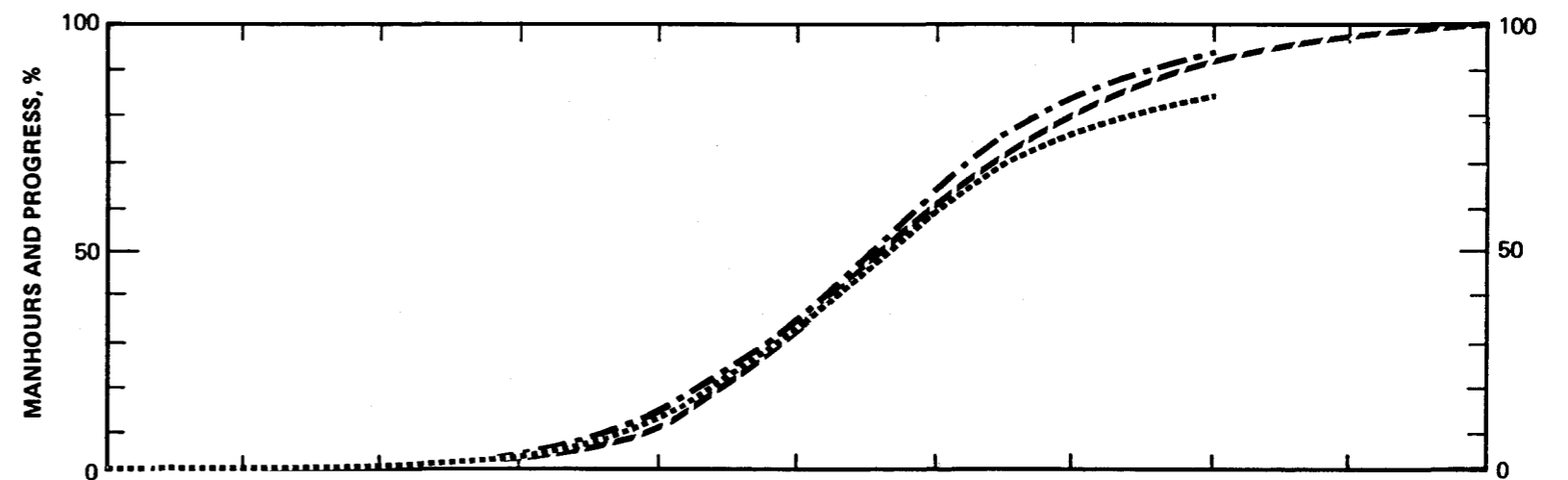


Figure 2-1. Progress and Performance Chart

## Section 3

### TECHNICAL PROGRESS

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

#### 3.1 TASKS I AND II — CITIES SERVICE DATA COLLECTION AND ANALYSIS

During this reporting period, Bechtel received additional data for nine recently completed Cities Service hydrogasification runs using Montana Rosebud subbituminous coal.<sup>1</sup> The data were entered into the computerized data base containing data from 25 lignite runs and five subbituminous runs. A computer listing of all the data is presented in Table 3-1. A detailed discussion of the Cities Service hydrogasification testing program was given in Bechtel's Second Quarterly Progress Report.<sup>2</sup>

The Cities Service subbituminous coal data show a maximum overall carbon conversion of 45 percent for Run MR-15, at a gas temperature of 1,660<sup>o</sup>F (2,160<sup>o</sup>R), a gas residence time\* of 1.18 seconds, and a hydrogen partial pressure of 1,500 psig (see Table 3-1). The highest methane selectivity and yield were also obtained during Run MR-15; carbon selectivity to methane was 54 percent and carbon conversion to methane was 25 percent.

Excellent carbon material balance closures from 96 to 102 percent and ash material balance closures from 85 to 102 percent have been reported<sup>1</sup> for the recent Cities Service subbituminous coal testing.

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\* For the entrained-flow Cities Service reactor, particle residence time is approximately equal to gas residence time.

Table 3-1

## 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.	17.00	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.

### 3.2 TASKS I AND II - BROOKHAVEN DATA COLLECTION AND ANALYSIS

Brookhaven National Laboratory has been conducting an experimental study on rapid gas-phase coal hydrogenation (flash hydropyrolysis). Although the major emphasis in this study was to maximize liquid hydrocarbon yield, appreciable yield of hydrocarbon gases (mainly methane and ethane) were obtained.<sup>3</sup> The bench-scale system incorporates an entrained downflow tubular reactor, 1-inch ID by 8 feet long, with a 3-foot cooling section below. A unique coal-feeding device is used which is capable of delivering coal to the reactor at flow rates as low as 0.1 lb/hr. The reactor has a maximum feed capacity of about 2 lb/hr. Special sets of electrically charged capacitor plates installed at the entrance and exit of the reactor allow accurate measurement of the residence time of coal particles falling through the reactor tube. Preheated hydrogen mixes with 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.<sup>3</sup>

A North Dakota lignite was employed in the initial 18 experiments performed by Brookhaven. The average particle size of the dried lignite feed was less than 150 microns; the analysis of the dried lignite used in these experiments is given in Table 3-2.

The operating conditions and the initial results of the published Brookhaven lignite runs<sup>3</sup> are given in Table 3-3. In these runs, Brookhaven explored reactor temperatures between 890°F (1,350°R) and 1,430°F (1,890°R), reactor pressures of 1,500 and 2,000 psig, and hydrogen-to-coal ratios between 0.9 and 5.8 lb/lb. Particle and gas residence times for each run were not reported, but a range of 12 to 20 seconds was given for the coal particle (and presumably gas) residence times. It was not clear whether the reactor temperature reported by Brookhaven was that of the reactor wall or of the gas-solid mixture flowing through the reactor.

Table 3-2

ANALYSIS OF MOISTURE-FREE NORTH DAKOTA LIGNITE\*  
 USED IN BROOKHAVEN HYDROLYSIS TESTS<sup>3</sup>

Constituent	Composition, wt %
Hydrogen	3.8
Carbon	59.4
Nitrogen	0.8
Oxygen	24.7
Sulfur	1.2
Ash	<u>10.1</u>
Total	100.0

\* From Baunol-Noonan mine with a moisture content as received of approximately 30 wt %

The Brookhaven reactor temperatures are at the lower end of the temperature range investigated by Cities Service (see Table 3-1). The Brookhaven hydrogen partial pressures are at the higher end of the range investigated by Cities Service. The Brookhaven coal particle residence times, however, are significantly higher than the Cities Service particle residence times.

The Brookhaven results from Table 3-3 show overall carbon conversions as high as 89 percent and carbon selectivities to methane as high as 45 percent. The high carbon conversions, compared with Cities Service results, may be due to the larger particle residence times.

Table 3-3

## BROOKHAVEN INITIAL HYDROLYSIS DATA

RUN DESIG- NATION	DATE	COAL TYPE	REACTOR	OVERALL FRACTION CARBON CONVERTED	FRACTION SELEC- TIVITY TO GAS	FRACTION SELEC- TIVITY TO METHANE	FRACTION SELEC- TIVITY TO ETHANE	REACTOR TEMP (DEG R)	HYDROGEN PARTIAL PRESSURE (PSIG)	HYDROGEN TO COAL RATIO (LB/LB)
5	1976	LIGNITE	EF*	.365	.737	.334	.164	1750.	1500.	3.38
7	1976	LIGNITE	EF	.301	.781	.312	.146	1750.	1500.	1.39
8	1976	LIGNITE	EF	.398	.721	.339	.0	1750.	1500.	5.80
9	1976	LIGNITE	EF	.215	.879	.265	.148	1660.	1500.	2.20
10	1976	LIGNITE	EF	.459	.649	.259	.137	1750.	2000.	1.48
11	1976	LIGNITE	EF	.171	.760	.158	.094	1570.	1500.	3.63
12	1976	LIGNITE	EF	.129	.977	.155	.085	1350.	1500.	4.85
13A	1976	LIGNITE	EF	.330	.867	.258	.139	1660.	1500.	5.63
13B	1976	LIGNITE	EF	.234	.855	.299	.167	1660.	1500.	0.90
14	1976	LIGNITE	EF	.566	.716	.387	.143	1890.	1500.	2.33
15	1976	LIGNITE	EF	.586	.759	.449	.089	1960.	1500.	2.80
16A	1976	LIGNITE	EF	.444	.722	.399	.131	1890.	1500.	0.98
16B	1976	LIGNITE	EF	.396	.714	.394	.134	1890.	1500.	1.40
16C	1976	LIGNITE	EF	.580	.705	.409	.133	1890.	1500.	1.53
17	1976	LIGNITE	EF	.692	.711	.397	.133	1870.	1500.	0.95
18A	1976	LIGNITE	EF	.860	.693	.367	.165	1830.	2100.	1.28
18B	1976	LIGNITE	EF	.822	.695	.354	.167	1830.	2100.	0.98
18C	1976	LIGNITE	EF	.888	.703	.359	.164	1830.	2100.	0.95

\* EF refers to a entrained-flow reactor.

During or at the conclusion of each run, actual material balances were calculated from the quantity of coal and hydrogen fed and products measured plus the results of the coal, char, and gas stream analyses. However, no product analyses or material balances were given for the runs presented in Table 3-2.<sup>3</sup>

During the next reporting period, particle and gas residence times for each Brookhaven lignite run will be collected, if possible, and entered into the computerized data base. Subsequently, semiempirical correlations for predicting carbon conversion efficiency and carbon selectivity will be fitted to the data. These correlations can be compared with the correlations fitted to the Cities Service lignite data.

### 3.3 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 products.<sup>2</sup> A computer listing of the correlated variables is given in Table 3-1.

#### 3.3.1 Overall Carbon Conversion

The proposed carbon conversion model was fitted to all the Cities Service data (lignite plus subbituminous), with lignite Run 8 excluded from the analysis. Run 8 had an unusually high reactor pressure of 2,960 psi, which 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 hydrogen partial pressure, particle residence time, gas velocity, coal type, or particle size within the region investigated. The correlation fitted to the Cities Service data is:

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

where,

X = overall carbon conversion, weight fraction

$t_R$  = gas (or particle) residence time, milliseconds

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

$T_G$  = maximum gas temperature, °R

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. This figure shows 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 data from the 14 subbituminous runs shown in Table 3-1 were correlated using the same carbon conversion model. A statistical analysis of the data indicated that carbon conversion for Montana Rosebud subbituminous coal was a function of gas residence time and gas temperature. Other correlated variables, such as coal-to-hydrogen ratio, hydrogen partial pressure, etc., did not significantly affect carbon conversion within the region investigated. The correlation fitted to the Cities Service data is:

$$X = 1 - \exp \left[ -1.59 (t_R)^{0.335} \exp(-7,210/T_G) \right] \quad (2)$$

Equation 2 indicates that carbon conversion for subbituminous coal increases with increase in both gas residence time and temperature in the region investigated. Note that, for the subbituminous data, carbon conversion is not a function of hydrogen-to-coal ratio. This is probably due to the fact that the lignite data showed a much wider variation in hydrogen-to-coal ratio than the subbituminous data (see Table 3-1).

Statistically, Equation 2 accounts for 83 percent of the variation in the fitted data (multiple correlation coefficient of 0.91), with a standard error of estimate of 4 percent in the predicted percent carbon conversion. Measured and predicted carbon conversions are shown in Figure 3-2. Both the statistics and Figure 3-2 indicate the good fit to the subbituminous carbon conversion data using Equation 2.

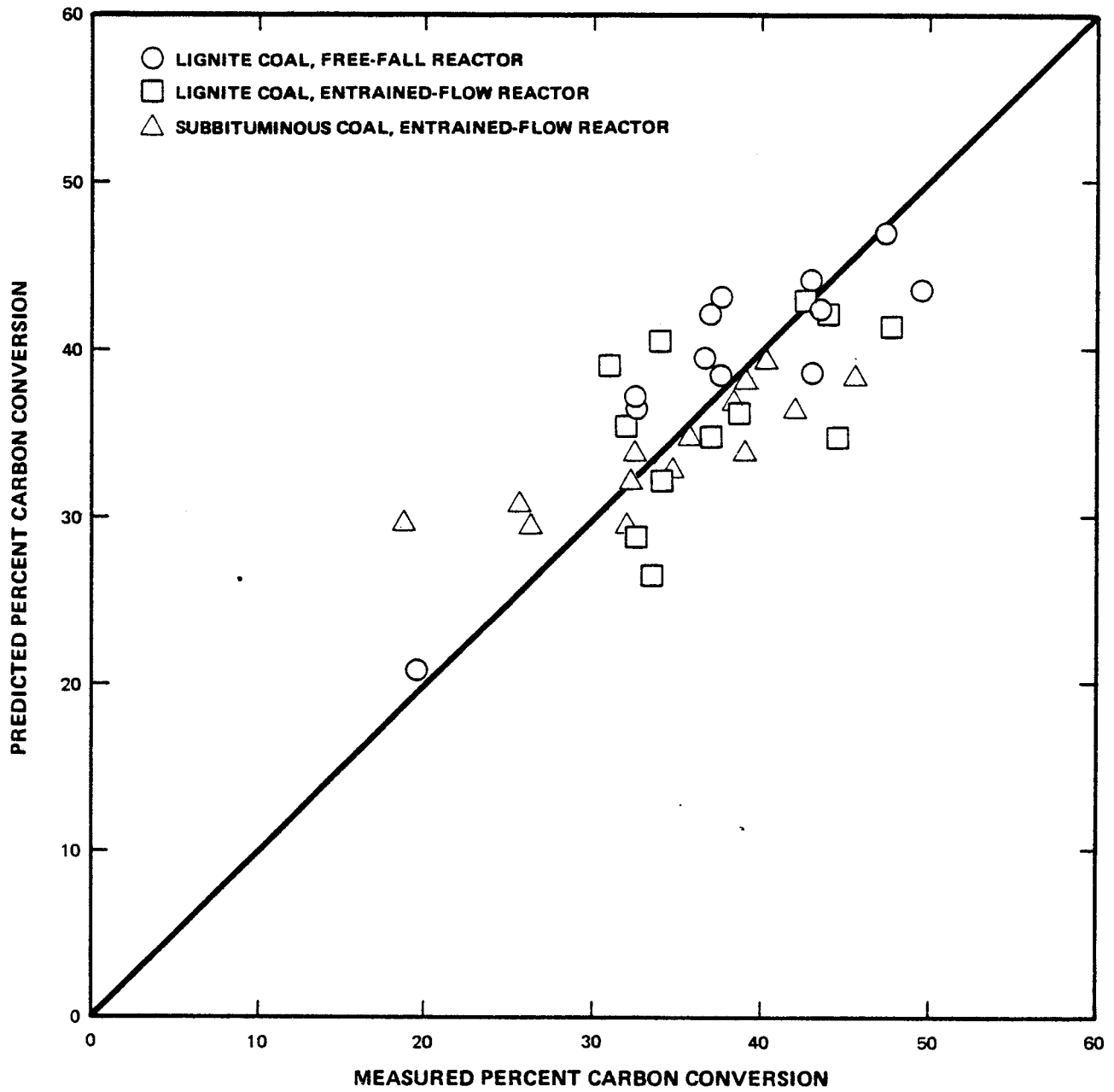


Figure 3-1. Comparison of Measured and Predicted Carbon Conversion for the Cities Service Reactor

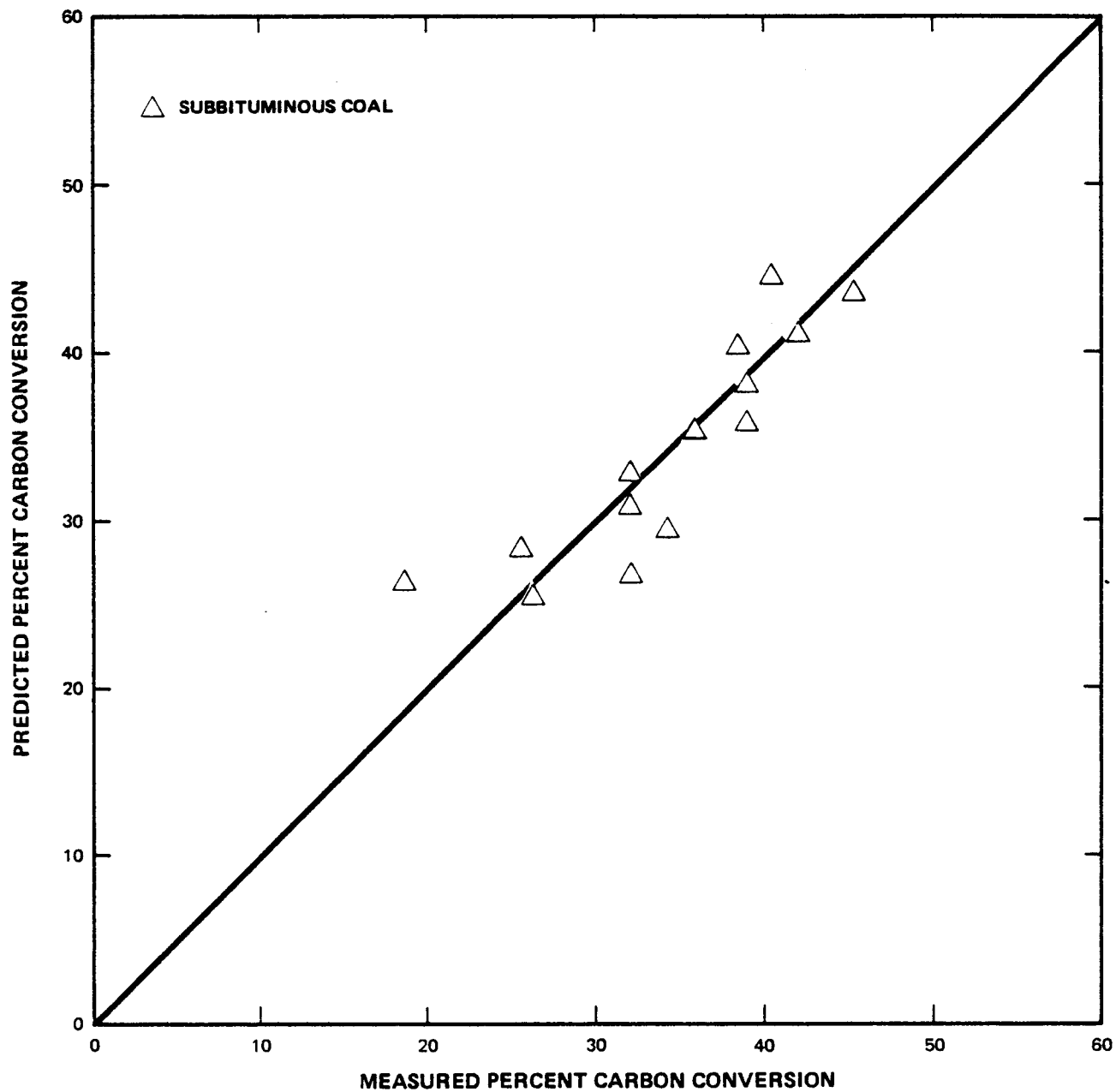


Figure 3-2. Comparison of Measured and Predicted Carbon Conversion for Subbituminous Coal for the Cities Service Reactor

In Figure 3-3, predicted overall carbon conversions from Equation 2 are plotted as a function of maximum gas temperature and gas residence time. This figure shows that a predicted overall carbon conversion of  $50 \pm 4$  percent can be achieved for Montana Rosebud subbituminous coal in the Cities Service bench-scale reactor at a gas temperature of about  $1,800^{\circ}\text{F}$  and at a gas residence time of 1,200 milliseconds. Of course, at higher residence times (e.g., 1.5 to 2 seconds), the same conversion level could be achieved at lower temperatures.

### 3.3.2 Carbon Selectivity to Methane

Fraction carbon selectivity to methane  $\phi_M$  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<sup>2</sup> was fitted to the combined lignite and subbituminous data (excluding Run 8), a statistical analysis showed that methane selectivity was a function of gas temperature and particle residence time (approximately equal to gas residence time for entrained-flow reactors). Methane selectivity was not significantly affected by hydrogen-to-coal ratio, hydrogen partial pressure, particle size, or gas velocity within the region investigated. The correlation fitted to the data is:

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

where,

- $\phi_M$  = carbon selectivity to methane, weight fraction
- $t_{RP}$  = particle residence time, milliseconds
- $T_G$  = maximum gas temperature,  $^{\circ}\text{R}$

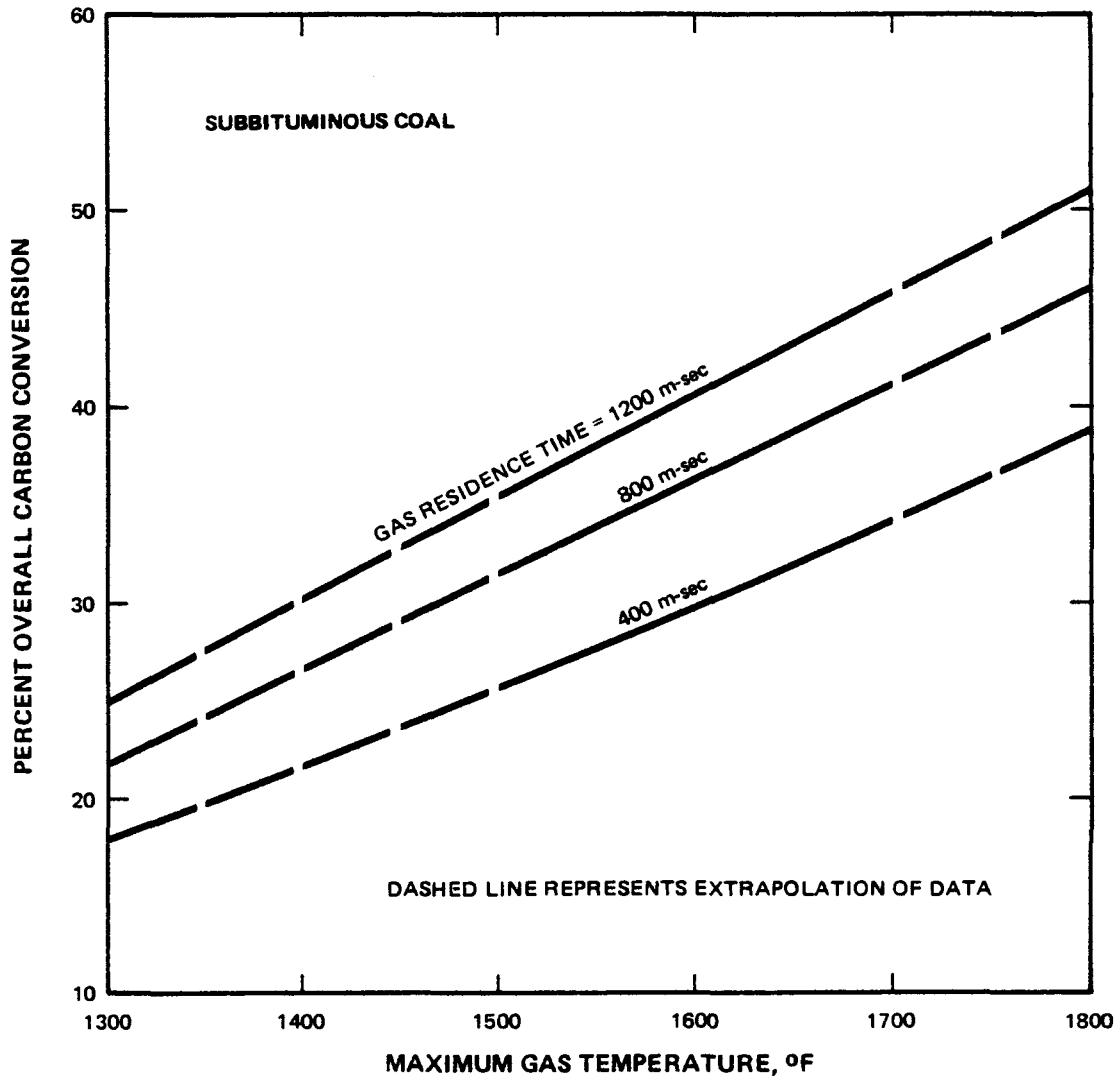


Figure 3-3. Predicted Overall Carbon Conversion for the Cities Service Reactor

As Equation 3 indicates,  $\phi_M$  increases significantly with increase in temperature and increases slightly with increase in 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. This 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.

A statistical analysis of the subbituminous data alone revealed that methane selectivity was a function of hydrogen partial pressure, gas (or particle) residence time, and gas temperature. Methane selectivity was not significantly affected by hydrogen-to-coal ratio, particle size, or gas velocity within the region investigated. The correlation fitted to the subbituminous data is:

$$\phi_M = 1 - \exp \left[ -313(P_{H_2})^{-0.250} (t_R)^{0.415} \exp(-15,300/T_G) \right] \quad (4)$$

where  $P_{H_2}$  is hydrogen partial pressure in psig.

As Equation 4 indicates, methane selectivity increases with increase in residence time and temperature, but decreases somewhat with increased hydrogen partial pressure. This effect of hydrogen partial pressure was not noticeable when the lignite data were included in the fitting, as Equation 3 indicates. Statistically, Equation 4 accounts for 95 percent of the variation in the data (multiple correlation coefficient of 0.98), with a standard error of estimate of 0.02 weight fraction in the predicted fraction selectivity. Measured and predicted methane selectivities are plotted against one another in Figure 3-5. Both the statistics and Figure 3-5 indicate the excellent fit to the methane selectivity data using Equation 4.

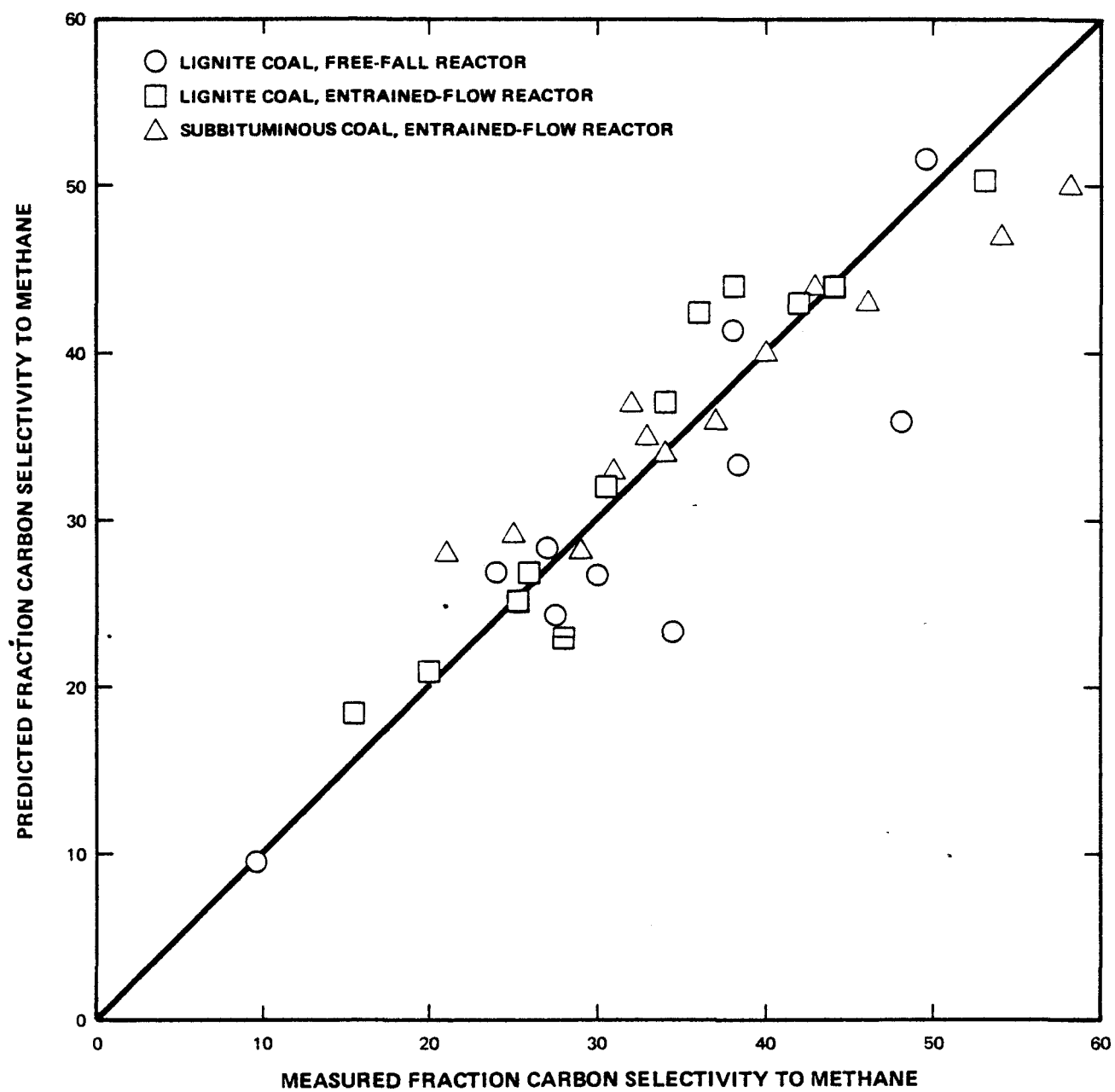


Figure 3-4. Comparison of Measured and Predicted Carbon Selectivity to Methane for the Cities Service Reactor

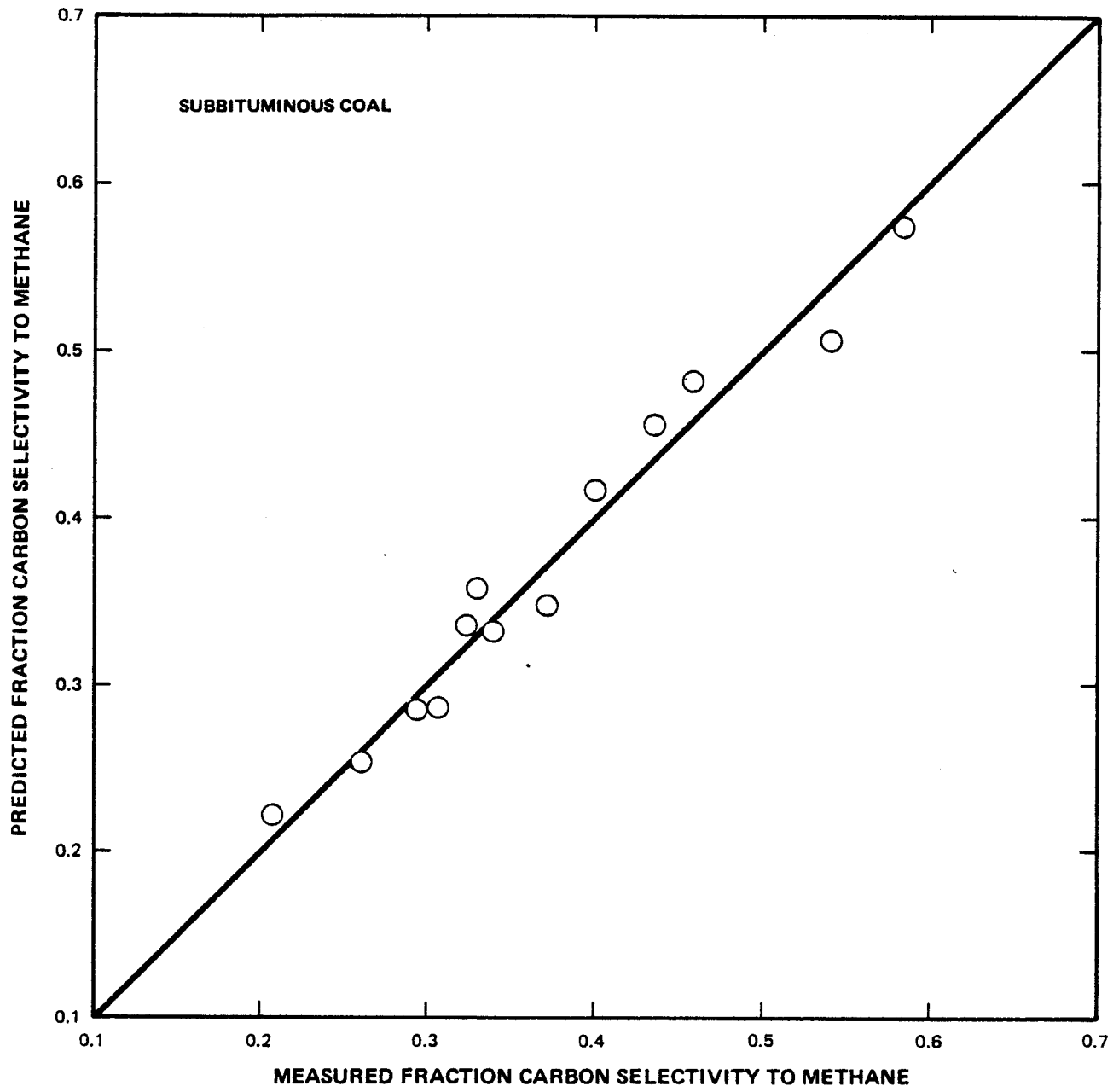


Figure 3-5. Comparison of Measured and Predicted Carbon Selectivity to Methane for Subbituminous Coal for the Cities Service Reactor

In Figures 3-6 and 3-7, methane selectivity predicted from Equation 4 is plotted as a function of maximum gas temperature and gas residence time, respectively. Figure 3-6 shows that selectivities of over  $53 \pm 2$  percent are predicted for the Cities Service reactor for temperatures in excess of  $1,700^{\circ}\text{F}$ , at a residence time of 800 milliseconds and hydrogen partial pressure of 1,000 psig. Figure 3-7 shows that methane selectivities of over  $50 \pm 2$  percent are predicted for the Cities Service reactor for residence times longer than 1,600 milliseconds, at a gas temperature of  $1,600^{\circ}\text{F}$  and hydrogen partial pressure of 1,000 psig.

### 3.3.3 Carbon Selectivity to Ethane

Ethane is another valuable hydrocarbon product formed during hydrogasification of lignite and subbituminous coals in the Cities Service reactor. The fraction carbon selectivity to ethane  $\phi_E$  is defined as the weight of carbon converted to ethane per total weight of carbon converted.

The proposed exponential carbon selectivity model<sup>2</sup> gave a very poor fit to the combined lignite and subbituminous data. 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

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

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

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

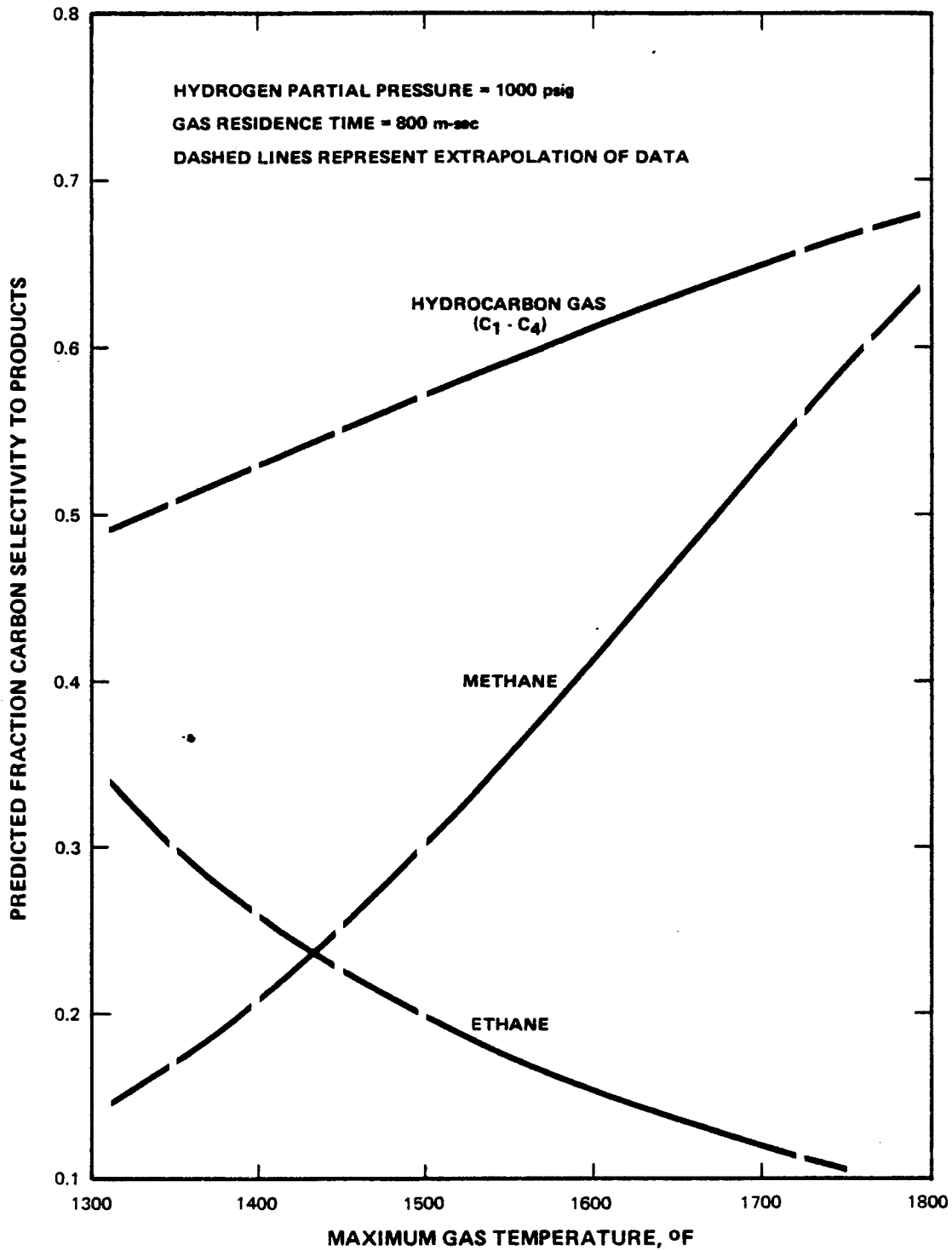


Figure 3-6. Predicted Carbon Selectivity to Hydrocarbon Products as a Function of Temperature for the Cities Service Reactor

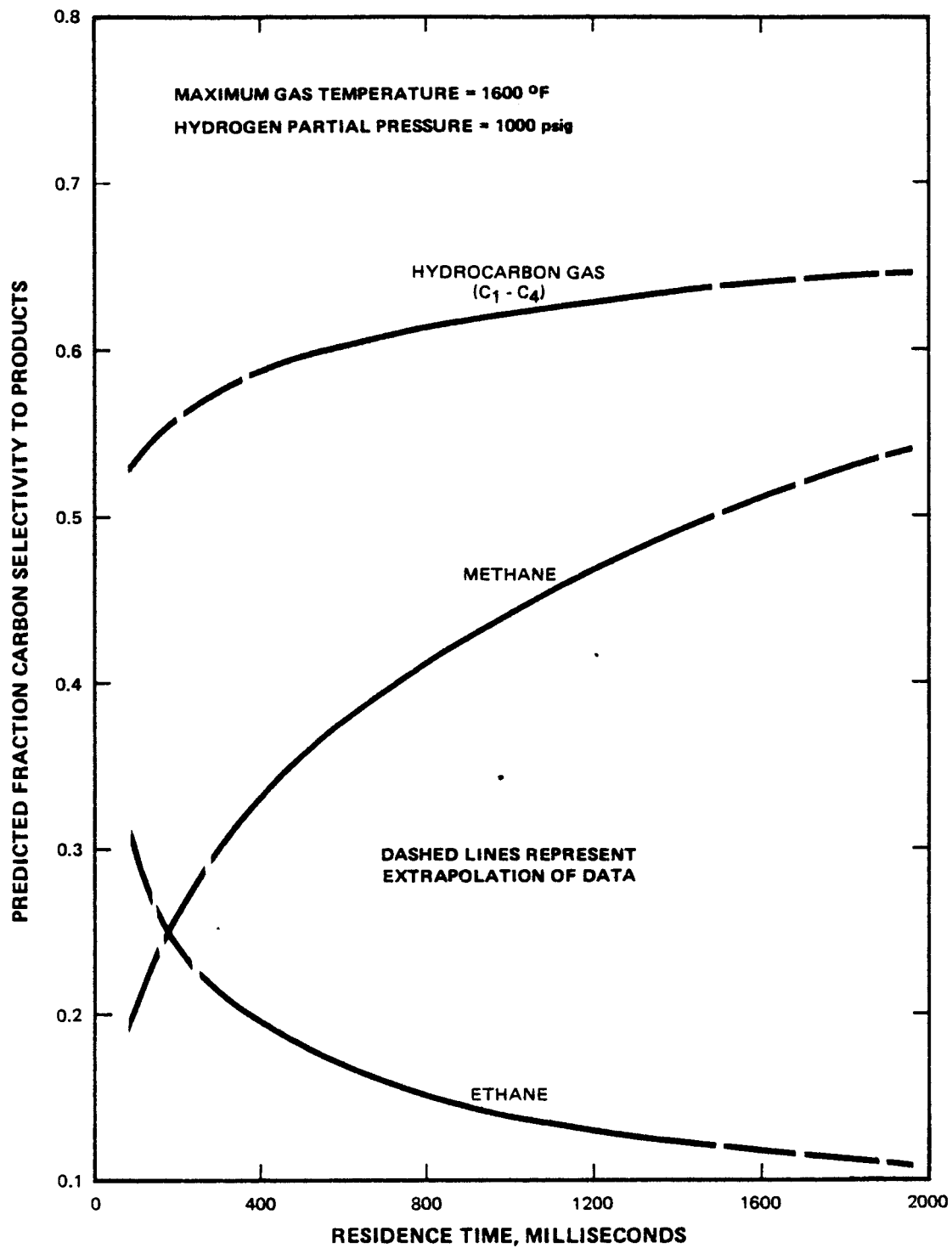


Figure 3-7. Predicted Carbon Selectivity to Hydrocarbon Products as a Function of Residence Time for the Cities Service Reactor

When the exponential selectivity model was fitted to the subbituminous data alone, a much improved fit was obtained. Statistical analysis of these data showed that ethane selectivity was a function of gas temperature and gas residence time. Hydrogen partial pressure, hydrogen-to-coal ratio, particle size, and gas velocity had no significant effect on ethane selectivity. The correlation fitted to the data is:

$$\phi_E = 1 - \exp \left[ -0.00738(t_R)^{-0.374} \exp(11,700/T_G) \right] \quad (5)$$

As Equation 5 indicates, ethane selectivity decreases with increase in residence time and temperature. This result suggests that the ratio of the rate of formation of ethane to the rate of formation of other products decreases at higher temperature and longer residence time. The decrease in this ratio may well be due to the thermal cracking of ethane at higher temperature and increased residence time.

Statistically, Equation 5 accounts for 87 percent of the variation in the data (multiple correlation coefficient of 0.93), with a standard error of estimate of 0.05 weight fraction in the predicted fraction selectivity. Measured and predicted ethane selectivities are plotted in Figure 3-8.

In Figures 3-6 and 3-7, ethane selectivity predicted from Equation 5 is plotted as a function of maximum gas temperature and gas residence time, respectively. Obviously, at temperatures in excess of 1,700°F or at residence times greater than about 2 seconds, the predicted carbon selectivity to ethane is small.

A predicted methane/ethane ratio in the product gas could be estimated at any given temperature and residence time from Equations 4 and 5 (or from Figures 3-6 and 3-7). Since ethane has a high heating value, the selection of a suitable methane/ethane ratio may be important for the economic operation of the reactor to produce pipeline-quality SNG.

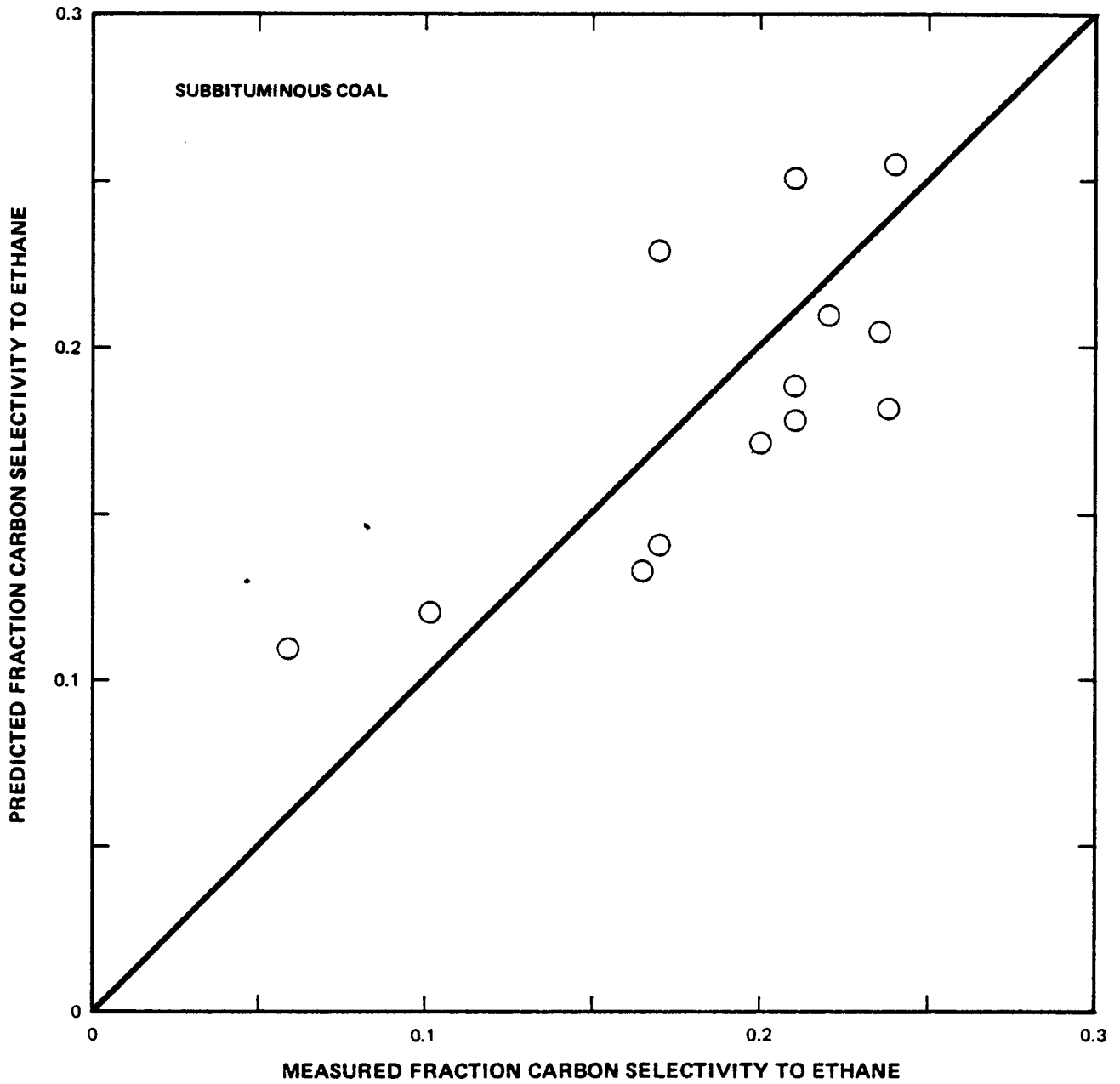


Figure 3-8. Comparison of Measured and Predicted Carbon Selectivity to Ethane for the Cities Service Reactor

### 3.3.4 Carbon Selectivity to Hydrocarbon Gas

In Cities Service subbituminous tests, C<sub>3</sub> and C<sub>4</sub> hydrocarbons were obtained in the product gas. Carbon selectivities  $\phi_{C_3}$  and  $\phi_{C_4}$  were calculated from the data, and each was correlated to the independent variables listed in Table 3-1 using the exponential carbon selectivity model proposed earlier. This model gave a very poor fit to the data for both  $\phi_{C_3}$  and  $\phi_{C_4}$ . Although the model fit was poor, statistical analysis of the fitted data still indicated that both carbon selectivities to C<sub>3</sub> and C<sub>4</sub> were inversely proportional to gas temperature and residence time within the region investigated. This result is not conclusive, but again may indicate that C<sub>3</sub> and C<sub>4</sub> hydrocarbons could be thermally cracking at higher temperatures and longer residence time; these hydrocarbons, whether parafins or olefins, are generally known to be less stable than methane and ethane at the temperature levels studied. A linear carbon selectivity model of the form given previously in Subsection 3.3.3 also gave a very poor fit to the C<sub>3</sub> and C<sub>4</sub> selectivities data.

The fraction carbon selectivity to hydrocarbon gas  $\phi_{C_1-C_4}$  obtained by combining the carbon selectivities to methane, ethane, and C<sub>3</sub> and C<sub>4</sub> hydrocarbons was correlated with the independent variables for the subbituminous data using the exponential carbon selectivity model. Statistical analysis of the fitted data showed that hydrocarbon gas selectivity was a function of hydrogen partial pressure, gas residence time, and maximum gas temperature. Hydrogen-to-coal ratio, particle size, and gas velocity had insignificant effect on hydrocarbon gas selectivity in the region investigated. The correlation fitted to the data is:

$$\phi_{C_1-C_4} = 1 - \exp \left[ -18.7 (P_{H_2})^{-0.227} (t_R)^{0.103} \exp(-4,330/T_G) \right] \quad (6)$$

Equation 6 indicates that hydrocarbon gas selectivity increases slightly with increase in temperature and residence time; but decreases somewhat with increased hydrogen partial pressure. Statistically, Equation 6 accounts for 84 percent of the variation in the data (multiple correlation coefficient of 0.92), with a standard error of estimate of 0.02 weight fraction in the predicted fraction selectivity. Measured and predicted selectivities are plotted against one another in Figure 3-9.

In Figures 3-6 and 3-7, hydrocarbon gas selectivity predicted from Equation 6 is plotted as a function of maximum gas temperature and gas residence time, respectively. Figure 3-6 shows that carbon selectivities to hydrocarbon gas of over  $65 \pm 2$  percent is predicted for the Cities Service reactor for temperatures in excess of about of  $1,700^{\circ}\text{F}$ , at a residence time of 800 milliseconds and hydrogen partial pressure of 1,000 psig. Figure 3-7 shows that hydrocarbon gas selectivities of over  $65 \pm 2$  percent is predicted for residence times greater than about 1.6 seconds, at a temperature of  $1,600^{\circ}\text{F}$  and hydrogen partial pressure of 1,000 psig.

Equation 6 will allow the selection of a suitable combination of temperature and residence time for the reactor operation that will achieve a desirable hydrocarbon selectivity level. The choice of such a level will be dictated, in part, by the desired reactor hydrogasification efficiency, which will be discussed in the following subsection.

In Bechtel's August Monthly Progress Report,<sup>4</sup> a correlation was presented for the fraction carbon selectivity to methane plus ethane  $\phi_{\text{ME}}$  for the combined lignite and subbituminous data. Since methane and ethane represent over 90 percent of the hydrocarbon gas found in both the lignite and subbituminous tests, methane plus ethane selectivity is very close to hydrocarbon selectivity, and the  $\phi_{\text{ME}}$  correlation should approximately represent the hydrocarbon gas selectivity for the lignite plus subbituminous data for the Cities Service reactor.

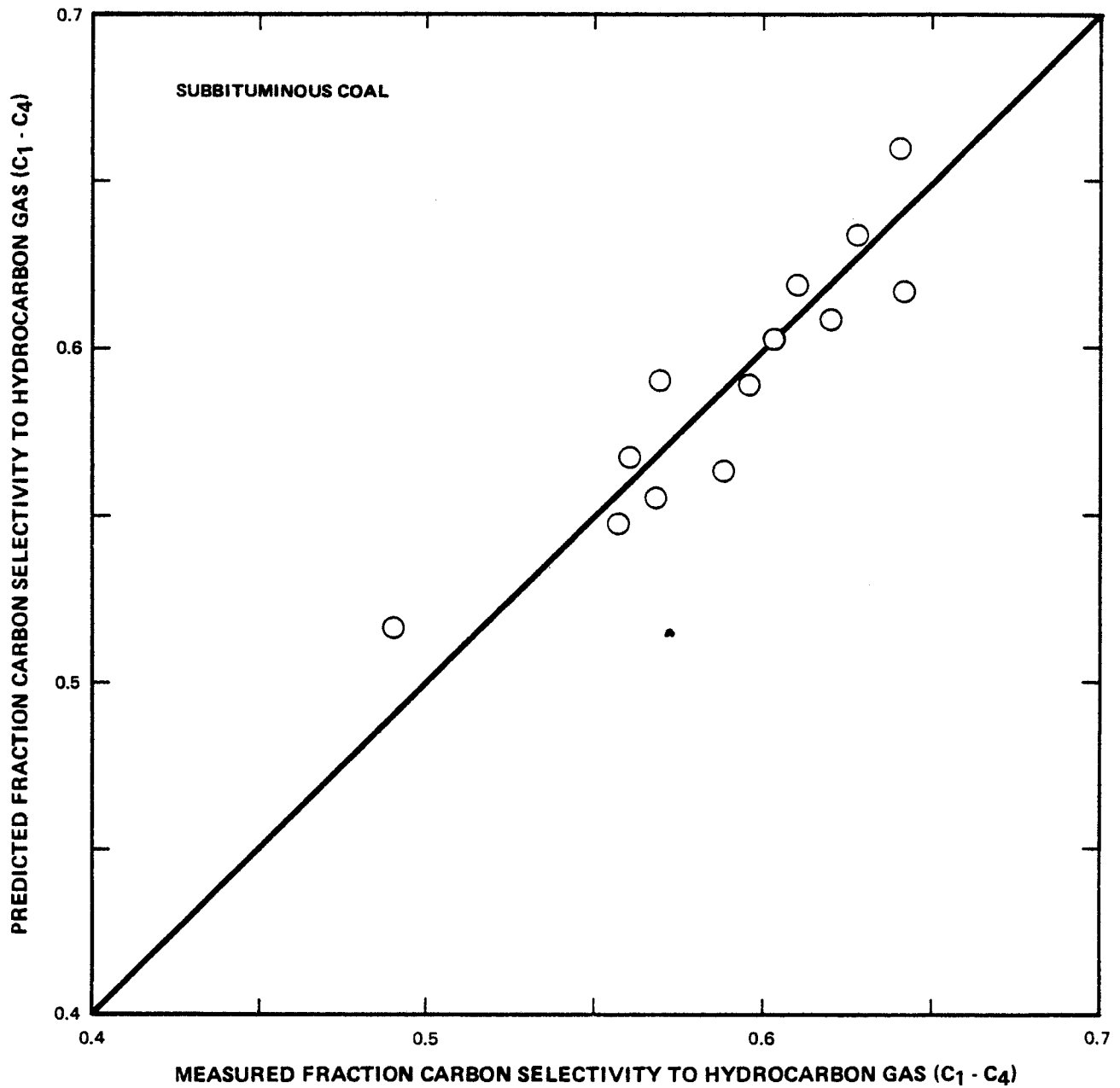


Figure 3-9. Comparison of Measured and Predicted Carbon Selectivity to Hydrocarbon Gas for the Cities Service Reactor

### 3.4 TASK III - CITIES SERVICE REACTOR HYDROGASIFICATION EFFICIENCY

For the Cities Service reactor, a hydrogasification efficiency has been defined as follows:

$$\text{Hydrogasification Efficiency} = \frac{100 \text{ (HV)}_{\text{gas}}}{\text{(HV)}_{\text{coal}} + \text{(HV)}_{\text{hydrogen}}} \quad (7)$$

where,

$\text{(HV)}_{\text{gas}}$  = heat content of hydrocarbon gas product,  
Btu/lb MAF coal

$\text{(HV)}_{\text{coal}}$  = heat content of coal feed, Btu/lb MAF coal

$\text{(HV)}_{\text{hydrogen}}$  = heat content of consumed hydrogen, Btu/lb MAF coal

All heat contents are calculated at 60°F and 1 atmosphere.

The above efficiency represents, in effect, a thermal efficiency for the reactor in terms of the primary desirable products, i.e., methane, ethane, and other hydrocarbon gases. The efficiency by definition is clearly different from a reactor overall efficiency in which the heat content of all products (BTX, light oil, char, and gas) are considered. The heat contents of other undesirable products in the product gas stream were neglected; these products are mainly carbon monoxide. Almost all the carbon monoxide produced in the reactor will be consumed in subsequent shift and/or methanation processing steps before the final SNG product is obtained.

For the Cities Service reactor using subbituminous coal, the heat content of the produced hydrocarbon gases is calculated from the data by summing the individual heat contents of methane, ethane, and C<sub>3</sub> and C<sub>4</sub> hydrocarbons.

The heat content of any of these hydrocarbon species is calculated from the following equation:

$$HV = (\Delta H_c) (MW/C_w) (\phi) (X) (C_F) \quad (8)$$

where,

HV = heat content of hydrocarbon product, Btu/lb MAF coal

$\Delta H_c$  = heat of combustion of hydrocarbon product

MW = molecular weight of hydrocarbon product, lb/lb mole

$C_w$  = weight of carbon in one lb-mole of hydrocarbon product, lb/lb-mole

$\phi$  = carbon selectivity to hydrocarbon product, weight fraction

X = overall fraction carbon conversion

$C_F$  = average carbon content of coal feed, lb/lb MAF coal

The heat content of the hydrocarbon gas product then becomes:

$$(HV)_{\text{gas}} = 0.716(X) \left[ 32,000 \phi_M + 28,000 \phi_E + 26,500 \phi_{C_3} + 25,850 \phi_{C_4} \right] \quad (9)$$

where  $\phi_M$  and  $\phi_E$  are the carbon selectivities to methane and ethane, and  $\phi_{C_3}$  and  $\phi_{C_4}$  are the carbon selectivities to  $C_3$  and  $C_4$  hydrocarbons. The heat of combustions used in Equation 9 were obtained from Janka,<sup>5</sup> assuming  $C_3$  and  $C_4$  to be propane and butane, respectively.

The average heat content of the Montana Rosebud subbituminous coal  $(HV)_{\text{coal}}$  was estimated by using an average composition of the coal<sup>1,6</sup> and the Mott and Spooner formula for calculating heat of combustion (calorific value) of coal.<sup>5</sup> The calculation yielded a value for  $(HV)_{\text{coal}}$  of 12,300 Btu/lb on a moisture- and ash-free basis.

The heat content of consumed hydrogen, (HV)<sub>hydrogen</sub>, was calculated from the reported hydrogen consumptions for the Cities Service runs<sup>1,6</sup> and the heat of combustion of hydrogen.<sup>5</sup>

Table 3-4 lists the calculated heat contents of the hydrocarbon gas product and consumed hydrogen, and the reactor hydrogasification efficiency for the Cities Service subbituminous runs. As can be seen, the reactor hydrogasification efficiencies varied from 15 to 41 percent. During the next reporting period, the reactor hydrogasification efficiency for the subbituminous runs will be correlated with the operating variables listed in Table 3-1 using several semiempirical models.

Table 3-4

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			
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

### 3.5 TASK III -- CONCEPTUAL COMMERCIAL-SCALE REACTOR DESIGN

During this reporting period, work was continued on the design of a commercial-scale hydrogasification reactor system, based upon the reactions envisioned and tested by PERC. Some of the features of the design being considered are discussed below.

The hydrogasification reactor section will be of the multiple mixing nozzle, entrained-flow type being experimentally tested by Rocketdyne. Carbon conversion in the reactor will be roughly 50 percent. The reactor design will be related to the process variable values (e.g., gas temperature, pressure) and conditions specified in Subsection 3.3 of Bechtel's Second Quarterly Progress Report.<sup>2</sup> Additionally, key considerations for commercial-scale process design have been discussed with James Gray of Rocketdyne\* and Marvin Greene of Cities Service.

Hot char from the hydrogasification stage will be transferred to a second reactor stage to react with steam and oxygen to make the hydrogen needed for the hydrogasifier. Two designs for the steam-oxygen-char reactor are being considered: the first will be a fluid-bed type; the second will be a multiple mixing nozzle entrained-flow reactor of the Rocketdyne type.

Heat exchange between the gas product stream from the hydrogasifier and the feed hydrogen will be used to recover process heat and decrease the hydrogen preheat duty as much as possible. A preliminary conceptual design of these heat exchangers will be investigated.

Equipment conceptual design sketches of the reactors, coal and char handling systems, and high-temperature heat exchangers will be presented in future reports.

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\* Presently at PERC.

### 3.6 FUTURE WORK

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

Models developed for correlating the Cities Service carbon conversion data will be updated and improved upon as further test results are obtained. Of special interest will be recent Cities Service results with Montana Rosebud subbituminous coal. Where possible, models will be developed for predicting (1) overall carbon conversion; (2) fraction carbon selectivity to methane and ethane; and (3) reactor hydrogasification efficiency.

Data generated from additional sources, e.g., Brookhaven National Laboratories, will be evaluated.

Design of a reference, commercial-scale hydrogasification reactor will continue. The design will be based on variable levels and conditions specified in Subsection 3.3 of Bechtel's Second Quarterly Progress Report.<sup>2</sup>

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

#### Section 4

#### REFERENCES

1. Combs, L.P. and Greene, M.I. "Hydrogasifier Development for the Hydrane Process," Monthly Progress Report, August 1977, ERDA Contract EX-77-C-01-2518 (September 1977).
2. Bechtel Corporation, "An Analysis of Coal Hydrogasification Processes," Quarterly Technical Progress Report for the Period 1 June - 31 August 1977, ERDA Contract EF-77-A-01-2565 (September 1977).
3. Fallon, P., and Steinberg, M. "Flash Hydrolysis of Coal," presented at the ACS 173d National Meeting, New Orleans (January 1977).
4. Bechtel Corporation, "An Analysis of Coal Hydrogasification Processes," Monthly Technical Progress Report for the Period 1 August - 31 August 1977, ERDA Contract EF-77-A-01-2565 (September 1977).
5. Janka, J. C., and Malharta, R. "Estimation of Coal and Gas Properties for Gasification Design Calculations," NTIS-PB235370 (August 1971).
6. Gray, J. A., and Sprouse, K. M. "Hydrogasifier Development for the Hydrane Process," First Quarterly Report, March-May 1977, ERDA Contract EX-77-C-01-2518 (June 1977).