

COAL GASIFICATION IN A LOW PRESSURE,
LOW RESIDENCE TIME, ENTRAINED FLOW REACTOR

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INTRODUCTION

Prior studies in which finely-ground coal was heated very rapidly have shown that the fraction of the coal that can be volatilized increases with both the rate of heating and the final temperature to which the coal is heated. For example, Eddinger, et al ¹, have presented data from an entrained flow reactor which show that volatile products amounting to 49.9 percent of the coal fed may be produced from a finely-ground coal having an ASTM volatility of only 35.5 percent, even though maximum reactor temperature was less than the 950°C reached in the standard volatility test. Kimber and Gray ² reported coal pyrolysis data in an entrained flow reactor operated at temperatures up to 2200°K. They observed volatiles as much as 87% greater than that from the standard test and concluded both higher heating rates and higher final temperatures increase the amount of volatile products.

Another characteristic of high-rate, high-temperature pyrolysis of coal that is not found in normal carbonization is the production of significant quantities of acetylene and ethylene in the pyrolysis gas. These products are commonly observed during coal pyrolysis in a plasma or by flash heating.³

The present study was undertaken to investigate the potential for increased volatility and also the production of unsaturated hydrocarbons as a result of rapid pyrolysis using partial combustion as the source of pyrolysis energy. This paper represents a progress report on the experimental work accomplished to date.

EXPERIMENTAL PROCEDURES

An entrained flow reactor was designed in which the finely-ground coal could be rapidly mixed with oxidizing combustion gases. The combustion gases were produced from a pre-mixed flame of pure oxygen with hydrogen. The reactor volume was designed for short residence times, and the products were quenched by water spray immediately downstream of the reactor.

A diagram of the reactor is shown in Figure 1. The reaction tube was made of alumina. This tube was placed inside an annular electrical heating element for preheating and to reduce heat loss during the run. The reaction tube and heating elements were insulated with a fibrous alumina insulation and encased with a water-cooled section of 6-inch aluminum pipe. Reaction tubes 4 5/8 inches in length with inside diameters of 3/4, 1 1/2, and 2 inches were tested. The use of smaller diameter tubes permitted testing at reduced residence times. The water-cooled injector head was made of aluminum. The coal was injected through two copper injectors located 180° apart and at an angle of 30° with the centerline of the reaction tube. The impingement point for these injectors was a distance of 3 inches below the orifice through which premixed combustion gases were fed to the reactor. A platinum/13% platinum-rhodium thermocouple was inserted near the base of the reactor to record the reactor temperature.

The coal tested was a high volatile B Utah coal from the Orangeville, Carbon County, area. Typical proximate and ultimate analyses of coal from this area are listed in Table 1. The coal was dried, ball-milled, and screened to minus 200 mesh for these tests. The moisture as used in the tests was less than one percent.

The coal was entrained into a stream of carrier gas, either hydrogen or nitrogen, with an auger-driven feeder. A variable-speed auger drive was employed to obtain feed rates ranging from 0.5 to 5.0 pounds of coal per hour. Entraining gas flows of from 13 to 15 SCFH were used in the 1/4-inch diameter feed line.

The product gas was separated from the quench water, passed through a filter and then through a gas meter. Samples of filtered gas were withdrawn for analysis with a gas chromatograph. The char was filtered from the quench water, dried and analyzed for ash content to verify material balance calculations.

The operating parameters varied were the feed rates of the coal and combustion gases and the stoichiometry of the combustion gases. Run times following preheating of the reactor ranged from 2 to 22 minutes. The range of feed rate variables tested and the range of reactor operating conditions that resulted are listed in Table 2.

RESULTS

A total of thirty-two test runs were made with the 2-inch diameter reaction tube, twenty with hydrogen as the carrier gas and twelve with nitrogen. Twelve tests were made with the 1 1/4-inch diameter reaction tube, and seven tests were made with the 3/4-inch diameter tube. Typical data obtained from these tests are presented in Table 3.

Effect of Hydrogen Concentration. It was observed that conversions to the hydrocarbon gases were generally higher the greater the concentration of hydrogen in the reactor. Data illustrating this effect are presented in Figure 6. In this figure the conversions to methane, ethylene, and acetylene at temperatures ranging from 1200 to 1400°K are plotted versus the hydrogen partial pressure at the reactor outlet. The conversion to methane is shown to be the most sensitive to this operating variable.

Although the observed effect of hydrogen concentration on the methane yield is in the direction expected from the hydrogenation reaction, i.e., $C + 2H_2 = CH_4$, the equilibrium constant, K_p , for this reaction is much lower than the observed ratio of $P_{CH_4}/P_{H_2}^2$. The observed ratios are compared with the curve representing hydrogenation equilibrium in Figure 7. It seems clear from this comparison that the hydrocarbon gases are nonequilibrium species resulting from pyrolysis reactions.

Steam Carbon Reaction. The composition and volumes of the product gas indicated that a significant fraction of the steam produced by the combustion gases reacted with the coal to form hydrogen and carbon monoxide. The calculated steam decomposition is plotted versus the oxygen/coal ratio in Figure 8. This plot also shows the effect of the two carrier gases, hydrogen and nitrogen. The higher hydrogen concentrations resulting from the use of hydrogen carrier gas is shown to suppress the steam decomposition. The approach of the reaction $C + H_2O = CO + H_2$ toward equilibrium is indicated by the data presented in Figure 9. It is apparent from this comparison that the steam-carbon reaction is far from equilibrium for all of the run conditions that were tested.

Shift Reaction. The conversion of carbon to carbon dioxide was observed to be rather low relative to the conversion to carbon monoxide, as mentioned above. In all cases the equilibrium constant, K_p , for the shift reaction, $CO + H_2O = CO_2 + H_2$, was observed to exceed the observed ratio of $P_{CO_2} P_{H_2}/P_{CO} P_{H_2O}$. This is illustrated by the data shown in Figure 10. The observed ratios are seen to correspond to equilibrium K_p 's at substantially higher temperatures than those measured at the reactor outlet.

Volume and Heating Value. The volume of dry gas produced, less the volume of coal carrier gas fed to the reactor, is shown in Figure 11 as a function of the oxygen/coal ratio. The volume produced is seen to increase uniformly with this ratio. The corresponding heating value of the dry, carrier-free gas is shown in Figure 12.

Combustion Equivalence Ratio. The effect of varying the equivalence ratio of combustion hydrogen to combustion oxygen was tested by operating the reactor with coal feed rate, oxygen to coal ratio, and carrier gas rate constant, and varying the combustion hydrogen feed rate. Combustion hydrogen was varied to give an equivalence ratio, i.e., the moles H_2 per mole O_2 , from 0.4 to 1.1. As illustrated in Figure 13, the weight of carbon in hydrocarbon gases per 100 pounds of coal increased and the molar ratio of carbon dioxide to carbon monoxide decreased with the equivalence ratio.

Reactor Temperature. Analysis of the data showed that the primary variable governing the composition of the reactor products was the temperature. The temperature as indicated by the thermocouple measurements was observed to increase with the amount of combustion gas fed to the reactor per pound of coal. Although the reactor tube was electrically heated, the feed rates and heat transfer area were such that the heating elements exerted a rather small effect on the reaction temperature, serving primarily to reduce heat losses. Figure 2 presents the measured temperatures as a function of the ratio of combustion oxygen per pound of coal. The effects of reaction tube diameter and coal feed rate are also indicated on this figure.

Effect of Temperature. Figure 3 presents data showing the conversion of the carbon in the coal to the hydrocarbon gases methane, acetylene, and ethylene, and to carbon monoxide and carbon dioxide. The conversion data are plotted versus the measured temperature without regard for variations in the other operating variables. These conversions were computed from the measured volume and composition of the gas produced, after condensation of the water vapor, and the feed rate of the coal. It is of interest to note first that in contrast to gasification products of conventional low-pressure coal gasifiers, significant conversion to the three hydrocarbon gases, methane, acetylene, and ethylene, were observed. Secondly, the trend of conversion with reactor temperature is clearly evident. The methane conversion rises to a maximum and then decreases with increasing temperature, the conversion to acetylene increases with temperature, and the conversion to ethylene decreases with temperature.

The effect of replacing hydrogen as the coal carrier with nitrogen on the conversion to carbon oxides is also indicated on this figure. The conversion to carbon monoxide appears to be principally dependent on temperature; however, the carbon dioxide yield is significantly greater with the lower hydrogen concentrations resulting from the use of nitrogen carrier gas. It is of interest to note that the amount of carbon dioxide produced relative to the amount of carbon monoxide is low as compared to the products from conventional gasifiers.

Effect of Residence Time. The effect of average residence time in the reactor on the conversion to the three hydrocarbon gases is indicated by the data shown in Figures 4 and 5. Figure 4 shows conversion data from three different sizes of reactor tubes each operated at a coal feed rate of 1.2 pounds of coal per hour. These data show only a slight effect of reactor size on the product yields. In Figure 5 the conversions for reactor temperatures in the range 1200-1400°K are plotted versus the average residence time. Although the data are scattered, there is a trend to lower conversions with increasing residence time. This trend is most evident in the conversion to acetylene. The data corresponding to Figure 4 and 5 show that reactor residence times of less than fifty milliseconds are sufficient for gasification to occur in this temperature range and that these products tend to disappear as the residence time increases.

SUMMARY

The observations and conclusions drawn from the experimental runs may be summarized as follows:

1. Reactor temperature was found to be the most important parameter affecting the overall gasification of coal. The temperature was controlled principally by the combustion oxygen fed per pound of coal. As much as 57 percent of the mass of coal fed was gasified, and the space time conversion corresponding to this yield was 408 lb carbon gasified/ft³ hr.
2. Significant yields of methane, ethylene and acetylene were produced. Up to 14 percent of the coal carbon was converted to these gases. The yield of ethylene was observed to decrease with increasing reactor temperature while the yield of acetylene increased. The conversion to methane was observed to pass through a maximum at a reactor temperature of about 1200°K. Maximum yields of methane, ethylene and acetylene were 6, 4, and 6 percent of the coal carbon respectively.
3. The yield of hydrocarbon gases was observed to increase with increasing hydrogen concentration and to decrease with increasing residence time. Residence times shorter than 50 milliseconds are indicated for optimum yield of hydrocarbon gases. Comparison of the methane yield data with hydrogenation equilibrium indicates that the hydrocarbons in the product result not from hydrogenation reactions but from non-equilibrium pyrolysis reactions.
4. Although significant steam decomposition was observed, conversion of carbon to carbon monoxide was substantially less than predicted for steam, carbon equilibrium. The conversion of carbon to carbon dioxide was observed to be much lower than predicted by the shift reaction equilibrium.
5. The yield of hydrocarbon gases and the yield of carbon dioxide relative to carbon monoxide depends strongly on the stoichiometry of the combustion gas. Combustion gases lean in hydrogen fuel cause the hydrocarbon gas yield to be reduced and the ratio of CO₂ to CO to be increased from the yields with stoichiometric combustion gases.
6. The volume of gas produced per pound of coal increases uniformly with the oxygen/coal ratio. At the ratio corresponding to maximum hydrocarbon gas yield the volume produced is 22 SCF per pound of coal.
7. The carrier-free heating value of the product gas decreases uniformly with increasing oxygen/coal ratio. At the ratio corresponding to maximum hydrocarbon gas yield, the heating value is in the range of 370-420 BTU/SCF.

ACKNOWLEDGEMENT

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REFERENCES

1. Eddinger, R. T., Friedman, L. D., and Rau, E., "Devolatilization of Coal in a Transport Reactor," Fuel 45, 245-252 (1966).
2. Kimber, G. M., and Gray, M. D., "Rapid Devolatilization of Small Coal Particles," Combustion and Flame 11, 360-362 (1967).
3. Bond, R. L., Ladner, W. R., and McConnell, G. I., "Reactions of Coal in a Plasma Jet," Fuel 45, 381-395 (1966).

TABLE 1.- Coal analysis - weight percent as received

PROXIMATE		ULTIMATE	
Moisture	5.65	Carbon	70.05
Ash	6.20	Hydrogen	5.76
Volatile Matter	34.35	Nitrogen	1.30
Fixed Carbon	53.80	Sulphur	0.64
	<u>100.00</u>	Oxygen	10.40
		Moisture	5.65
		Ash	6.20
			<u>100.00</u>

Coal size (-200 MESH)

TABLE 2.- Range of Feed Rate Variables

Coal Feed Rate, lbs/hr	0.7-4.1
Oxygen/Coal Ratio	0.3-1.6
Combustion Gas Equivalence Ratio	0.4-1.1
Coal Carrier Gas (13-15 SCFH)	Nitrogen or Hydrogen

Range of reactor operating conditions

Average Temperature	1200-2500°F
Average Residence Time	0.012-0.343 sec.
Space Time Conversion	13-408 lbs C gasified/ft ³ -hr
Steam Partial Pressure (Reactor exit)	0.125-0.255 atm
Hydrogen Partial Pressure (Reactor exit)	0.194-0.553 atm

TABLE 3.- Typical data obtained from gasification tests

Run No.	5-8-4	5-8-2	5-8-1	6-9-4	7-31-1	8-23-2
Reactor Diameter, inches	2.000	2.000	2.000	2.000	1.250	0.750
Feed Rates, lbs/hr						
Coal	1.590	1.590	1.590	2.043	1.670	1.180
Hydrogen Carrier	0.082	0.082	0.082	--	0.082	0.082
Hydrogen Combustion	0.069	0.102	0.140	0.167	0.167	0.113
Oxygen	0.540	0.800	1.050	1.260	1.340	0.900
Nitrogen Carrier	--	--	--	1.020	--	--
Oxygen/Coal Ratio	0.339	0.503	0.660	0.616	0.802	0.762
Combustion Equivalence Ratio	1.022	1.025	1.066	1.060	0.997	1.008
Reactor Temperature, °F	1445	1750	1955	1966	2157	1913
Volume Gas Produced (Dry)						
Total, SCFH	30.9	37.6	45.9	56.5	60.3	39.7
Carrier Free Basis, SCF/lb Coal	10.2	14.4	19.6	21.2	27.3	21.1
Gas Analysis (Dry, Volume %)						
Hydrogen	74.12	69.88	69.75	26.38	64.23	67.14
Oxygen	0.20	0.42	0.15	8.30	1.65	1.39
Nitrogen	0.81	1.33	0.45	46.89	4.12	4.66
Methane	5.33	5.02	3.85	1.81	2.34	3.22
Carbon Monoxide	15.35	18.41	20.85	12.96	23.48	18.83
Ethane	0.14	0.04	0.01	0.00	0.00	0.01
Ethylene	1.62	1.26	0.64	0.12	0.19	0.52
Carbon Dioxide	1.55	2.06	2.51	2.68	2.39	2.57
Acetylene	0.88	1.58	1.79	0.86	1.60	1.66
Carrier Free Heating Value, BTU/CF	449.4	430.2	395.0	358.9	365.7	392.1
Steam Decomposed, Percent	18.28	28.79	38.81	48.67	46.86	36.84
Ash in Char, Weight percent	9.3	14.9	15.8	16.5	15.68	13.94

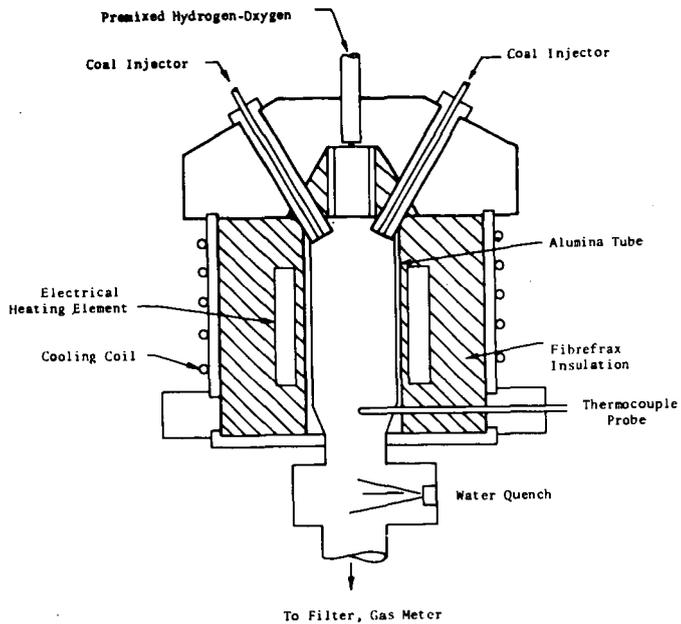


Figure 1. Schematic Diagram of Reactor.

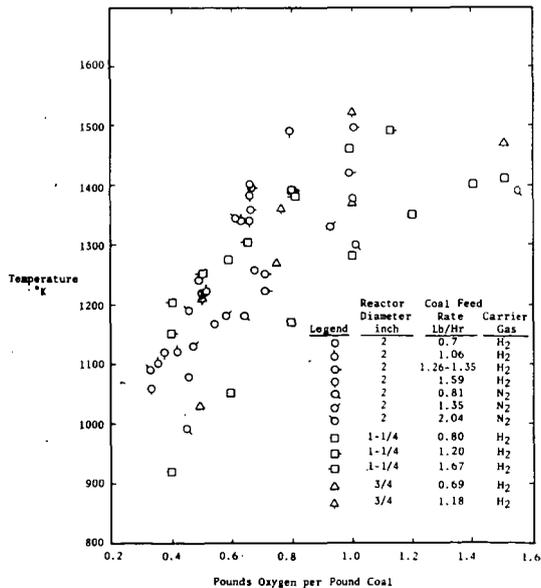


Figure 2. Measured reactor outlet temperature versus the oxygen fed per pound of coal.

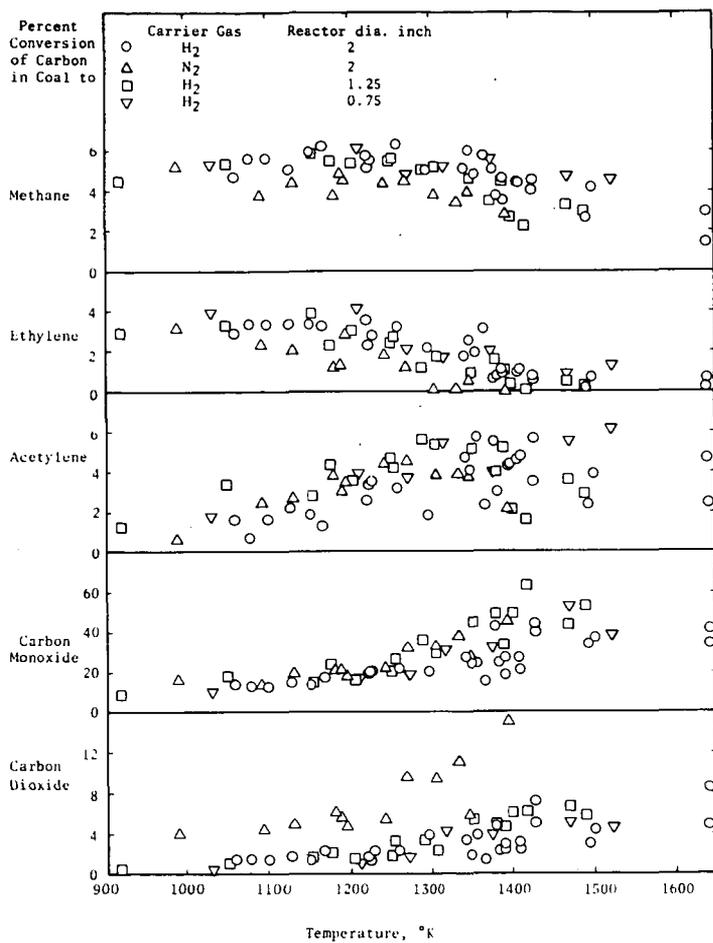


Figure 3. Conversion of Carbon in Coal to Hydrocarbon Gases, carbon monoxide and carbon dioxide versus reactor temperature.

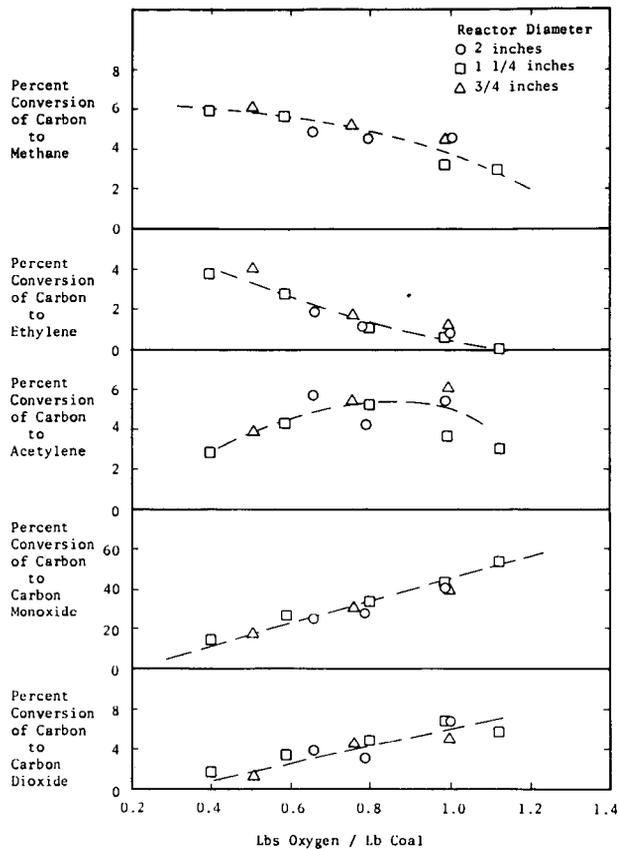


Figure 4. Conversion data showing small effect of varying the reactor size. Data shown are for a coal feed rate of 1.2 lbs/hr.

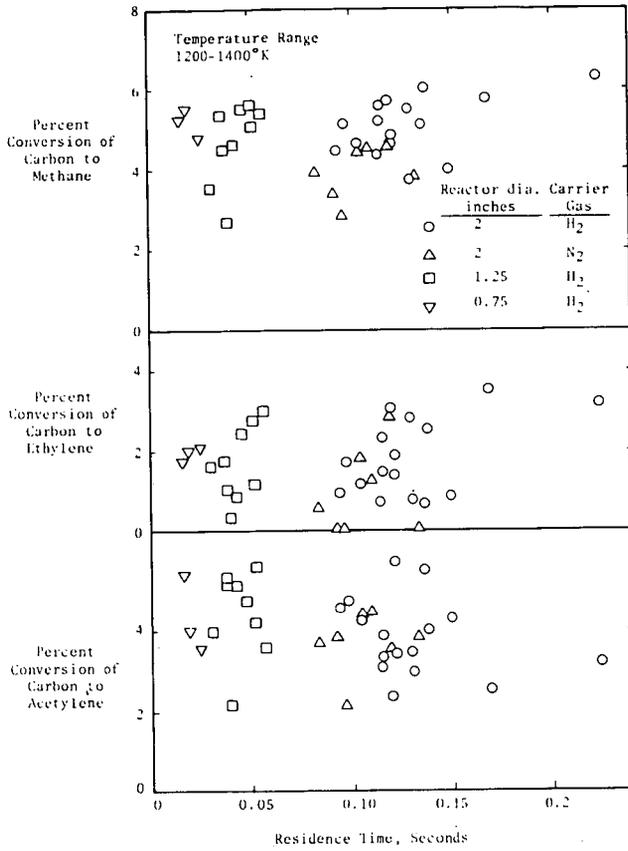


Figure 5. Conversion data showing small effect of average reactor residence time. All data shown are for a coal feed rate of 1.2 lb/hr.

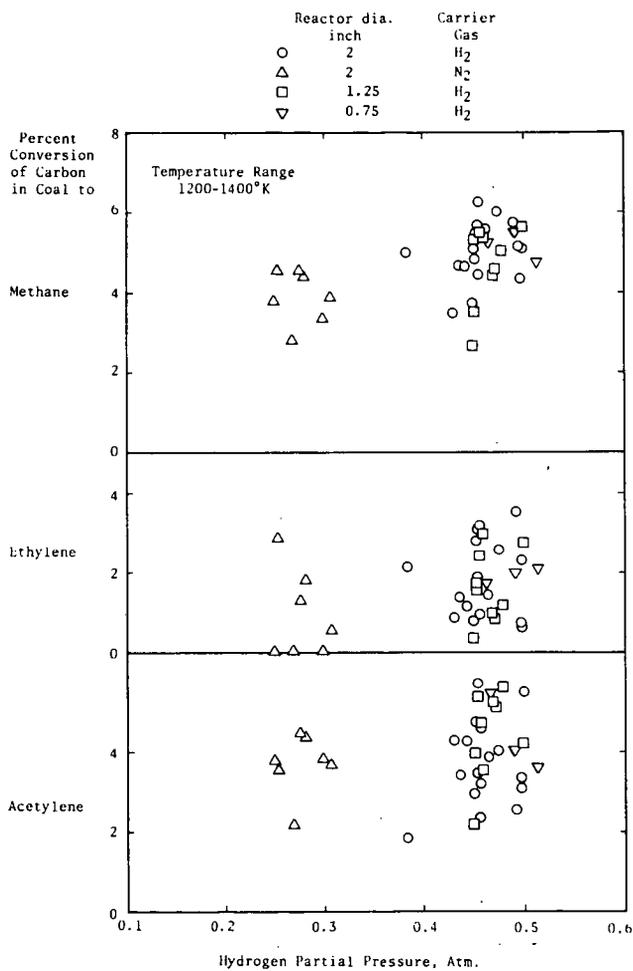


Figure 6. Data showing the effect of hydrogen partial pressure on hydrocarbon yields.

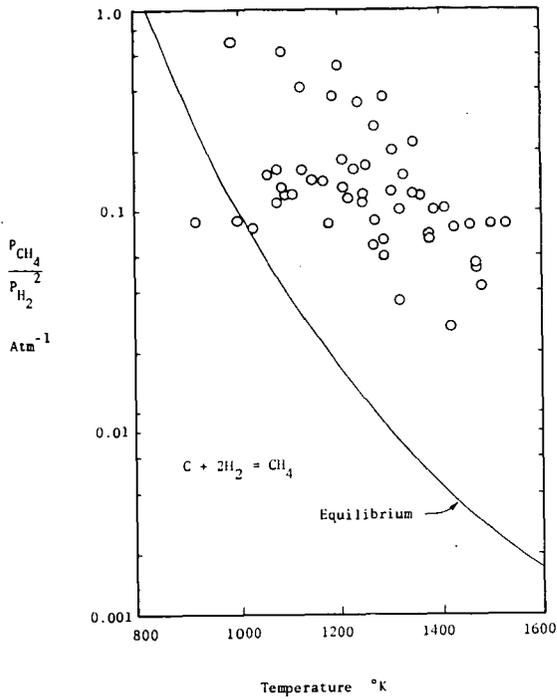


Figure 7. Comparison of equilibrium pressure ratios for the hydrogenation reaction with measured ratios.

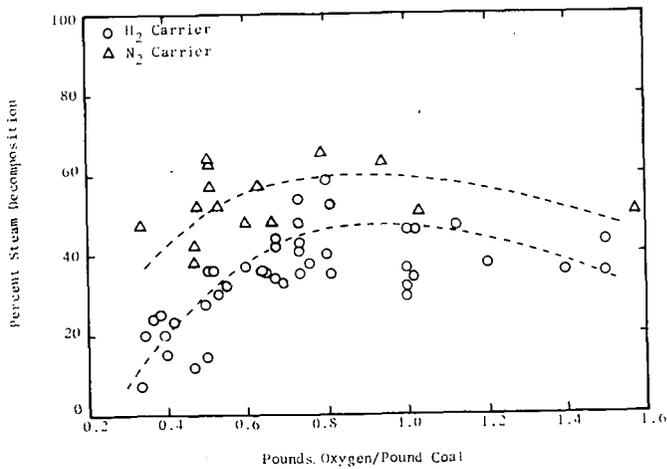


Figure 8. Calculated steam decomposition versus the oxygen fed per pound of coal.

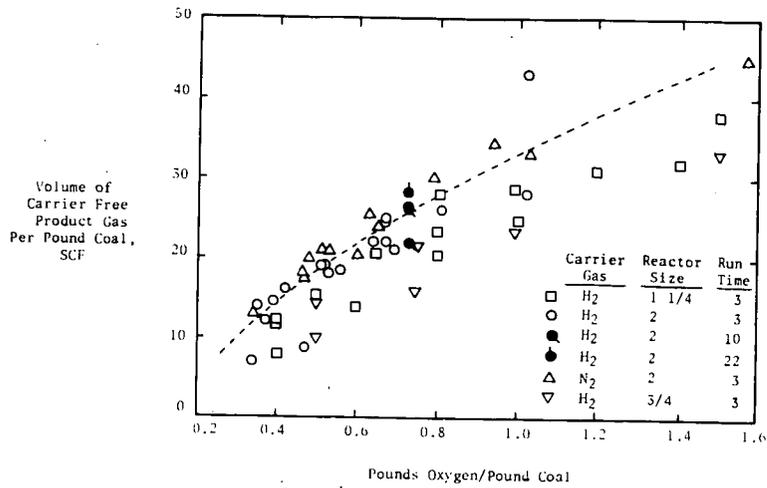


Figure 11. Net volume of product gas per pound of coal fed as a function of oxygen to coal ratio.

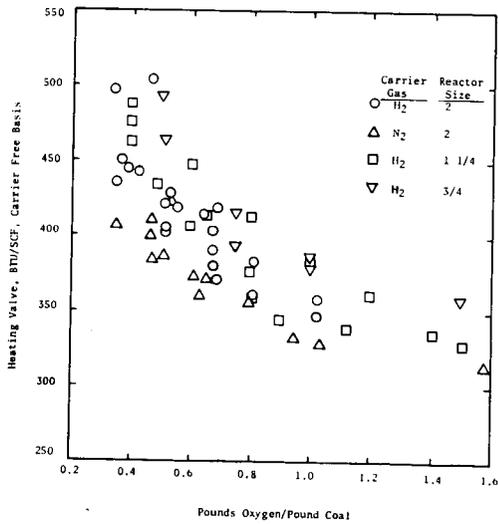


Figure 12. Data showing the variation of the heating value of the dry, carrier-free product gas with the oxygen-coal ratio.

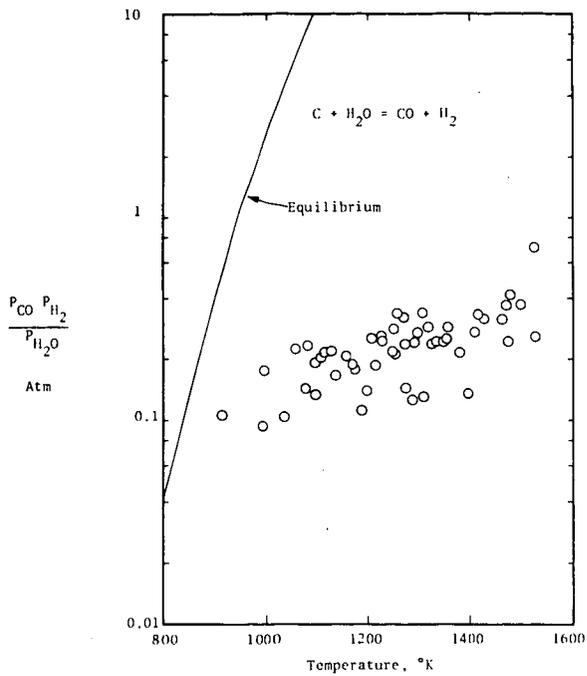


Figure 9. Comparison of equilibrium pressure ratios for the steam-carbon reaction with measured ratios.

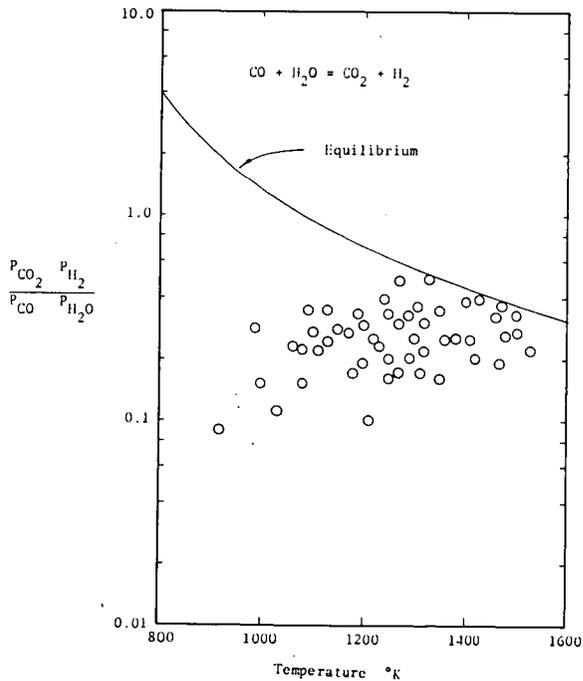


Figure 10. Comparison of equilibrium pressure ratios for the shift reaction with measured values.

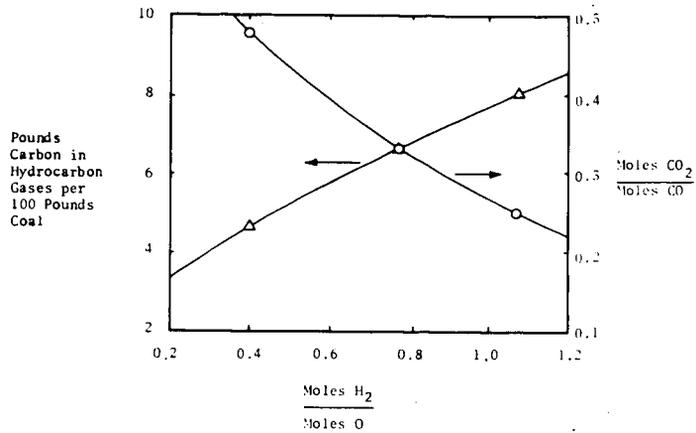


Figure 13. Data showing the effect of varying combustion gas equivalence rates. The coal feed rate was 2 lbs/hr and the O₂/coal ratio was 0.51.