

## KINETIC DATA FROM A HIGH TEMPERATURE ENTRAINED FLOW GASIFIER

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### Introduction

An experimental program is currently being carried out at the Eyring Research Institute in which several different coals are being tested in a laboratory scale entrained flow gasifier. The design of the gasifier is essentially a scaled down version of an experimental gasifier operated at the Bureau of Mines Morgantown Coal Research Center (1, 2). This gasifier is being operated under contract with the Energy Research and Development Administration to obtain reaction rate and conversion efficiency data for several different coals and chars.

### Description of Gasifier

A diagram of the gasifier is shown in Figure 1 and a flow diagram for the entire gasification system is presented in Figure 2. The reaction chamber, formed from silicon carbide is 7.6 cm I.D. and 28 cm in length. The coal, pulverized to 70-80 percent minus 200 mesh, is withdrawn with metering augers from a pressurized feed tank and blown into the top of the reaction chamber with recycled product gas. It enters the reactor through a water-cooled 8 mm I.D. nozzle. A preheated mixture of steam and oxygen is injected into the reactor through eight nozzles that closely surround the coal nozzle. These nozzles have rectangular openings, 1.1 x 1.5 mm in cross-section, and the oxygen-steam streams impinge on the coal stream at an angle of 30°.

The reactor effluent stream is cooled by radiation to the water-cooled walls fo the heat recovery unit which is attached to the base of the reactor. The heat recovered in this unit is used to generate the feed steam for the gasifier. Slag droplets and larger solid particles in the effluent stream are separated from the gases and they are then further cooled and cleaned by passage through a convective heat exchanger, a scrubber column, and fabric filters (Figure 2). Gas samples are withdrawn downstream of the filters. The cooled and cleaned gas is then metered and flared.

### Gasifier Operating Conditions

High volatile, low sulfur, bituminous coal from the Deseret Mine in Carbon County, Utah, was used for these tests. This coal has a high heating value of 1430 kcal/kg. It is 70.1 percent carbon, 5.6 percent hydrogen, 11.7 percent oxygen, 1.4 percent nitrogen, 0.6 percent sulfur, and 10.6 percent ash (dry basis). The moisture of the coal as fed to the gasifier was approximately one percent.

To obtain the data reported here the gasifier was operated at a nominal pressure of 10 atmospheres and coal feed rates were 20 kg/hr. The volumetric flow rate of the recycle gas stream was 5 M<sup>3</sup>/hr at STP. The oxygen and steam feed stream was preheated to 400°C for these test. The principal test variable was oxygen feed rate. The steam/coal feed ratio was 0.5 g/g. Product gas samples were not taken until steady-state operation was achieved, 20-30 minutes after start up.

### Test Results

Results from a series of ten test runs in which the oxygen/coal feed rates were systematically varied are presented in Figures 3 and 4. Figure 3 shows the volume of dry gas produced per kg coal fed and the percent of carbon converted to gas. As observed during the Bureau of Mines studies (1,2) the carbon conversion increased markedly with increasing oxygen/coal ratio. Gas volume also increased with increases in this ratio. Figure 4 presents the corresponding gas composition

data. Carbon monoxide increased with increasing oxygen/coal ratio whereas the yield of hydrogen and methane decreased. The carbon dioxide concentration varied only slightly.

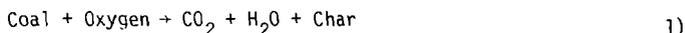
#### Data Analysis Procedure

After careful study of kinetic data available in the literature for the high temperature reaction of coal with oxygen and steam, it was concluded that the rate limiting reactions at the conditions employed in these tests were those of charred coal with steam and with carbon dioxide. The reactions of oxygen with coal volatiles and with char were calculated to be at least an order of magnitude faster. For example, the data of Kimber and Gray (3) indicate the specific reaction rate of both CO<sub>2</sub> and water vapor with 35 micron char particles to be less than 5 grams carbon/gram unreacted carbon per second per atmosphere partial pressure of the CO<sub>2</sub> or water. The data of Khitrin and Golovina (4) suggests rates of 3.3 sec<sup>-1</sup> atm<sup>-1</sup> and 1.9 sec<sup>-1</sup> atm<sup>-1</sup>, respectively, for water and CO<sub>2</sub> with char at 2000°K. On the other hand, the rate equation recommended by Field, et.al., (5) for the reaction of oxygen with char yields a specific rate of 1900 sec<sup>-1</sup> atm<sup>-1</sup> at this temperature.

Reaction rates of coal volatiles with available oxygen are even faster than the oxygen-char rates. Employing the rate equation suggested by Field (5). The specific reaction rate for the carbon monoxide-oxygen reaction is estimated to be of the order of 10<sup>5</sup> sec<sup>-1</sup> for the conditions used in these experiments.

Mixing and diffusion rates were also considered in selecting a procedure for analyzing the data. The gasifier feed nozzle configuration used in these tests was selected after extensive tests of alternate configurations. These tests demonstrated that it provided rapid and efficient mixing of the feed streams. The recirculation rate near the point where the feed streams were injected was estimated to be approximately 150% of the mass flow of the feed streams. Consequently, it seems reasonable that as a first approximation the reactor might be considered as being well-mixed. Diffusion rates of steam and CO<sub>2</sub> to the surface of the char particles were considered by calculating the diffusion limited reaction rates. These were estimated to be of the order of 50-100 sec<sup>-1</sup> at the experimental conditions of this study and were therefore considered to be of secondary importance in limiting the overall reaction rate.

On the basis of the above reasoning the data were analyzed assuming the reactor to be well stirred and that the gasification reactions could be represented by two simple reactions:



with reaction (1), which includes devolatilization of the coal, assumed to be fast relative to reaction (2).

The fraction of carbon gasified by equation (1), Y<sub>1</sub>, may be calculated from the following stoichiometric equation:

$$Y_1 = \Phi - \frac{1}{2}(O/C) (\Phi - \epsilon_O) + \frac{1}{2}(H/C) (\Phi - \epsilon_H) \quad 3)$$

In this equation, Φ is the equivalence ratio of the oxygen and coal feed streams, a value of one corresponding to a feed ratio such that sufficient oxygen is available to convert the carbon to CO<sub>2</sub> and the hydrogen to H<sub>2</sub>O. The ratios (O/C) and (H/C) are the molar ratios of oxygen to carbon and hydrogen to carbon in the coal, and ε<sub>O</sub> and ε<sub>H</sub> are the fractions of oxygen and hydrogen that are gasified by reaction (1). The fraction of carbon gasified by reaction (2), Y<sub>2</sub>, may then be calculated from Y<sub>1</sub> and the total fraction of carbon gasified Y<sub>T</sub>, which is available from the experimental data. Thus,

$$Y_2 = Y_T - Y_1 \quad 4)$$

A further simplification is the assumption that the rate of reaction (2) is first order with respect to the amount of unreacted carbon.

$$\frac{dy}{dt} = k (1 - Y) \quad 5)$$

The equation for the reaction rate constant resulting from integration of equation (5) is

$$k = -(1/t) \ln (1 - Y_T)/(1 - Y_1) \quad 6)$$

This rate constant which is the specific reaction rate, can be utilized as a measure of the reactivity of the coal.

The reaction time was taken to be the ratio of the reactor volume to the volumetric flow rate of the reaction products at the outlet of the reactor. The density at the outlet was computed from the dry gas analysis and an overall oxygen balance and energy balance. The oxygen balance yielded the steam concentration and the overall energy balance yielded the outlet temperature. The temperature calculation was verified by thermocouple measurements that were made in several of the runs.

#### Specific Reaction Rates

The specific reaction rates that were computed from the experimental data are presented in Figure 5 versus the oxygen/coal equivalence ratio. Reactor outlet temperatures are also presented on this figure. It is noted that magnitudes of these rates are somewhat higher than the rates measured by Kimber and Gray (3) and by Khitrin and Golovina (4). A cross-plot of the rate constant and temperature data yielded the following correlating equation:

$$k = 1.4 \times 10^5 \exp (-31,000/RT) \text{ sec}^{-1} \quad 7)$$

The magnitude of the apparent activation energy, 31 kcal, is consistent with the assumption that the char surface reaction with  $\text{CO}_2$  and steam is the rate limiting step in the gasification of this coal.

#### Acknowledgements

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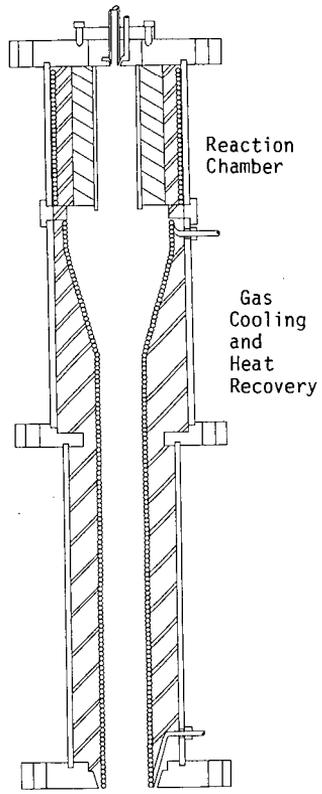


Figure 1. Diagram of laboratory gasifier.



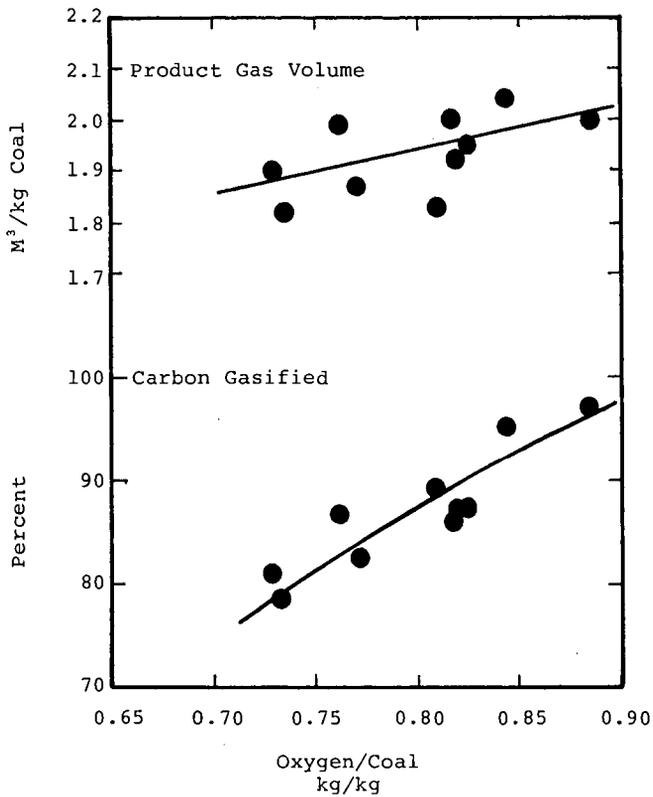


Figure 3. Gas volume and carbon conversion data from tests with Deseret Mine bituminous coal at 20 atm.

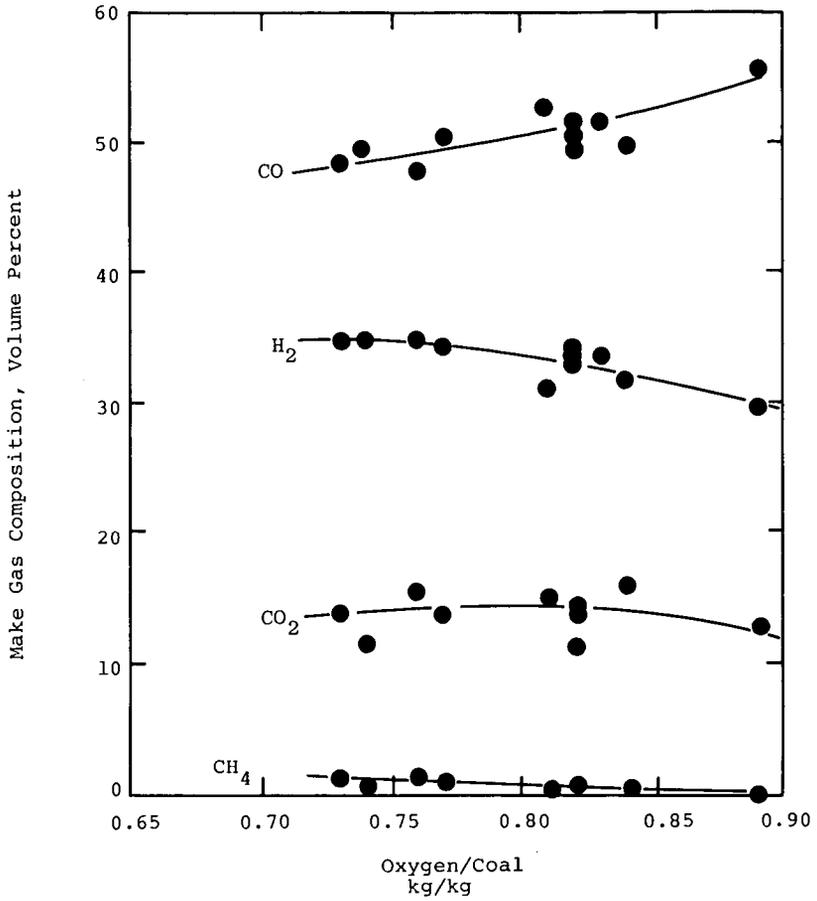


Figure 4. Gas composition data from tests with Deseret Mine bituminous coal at 20 atm.

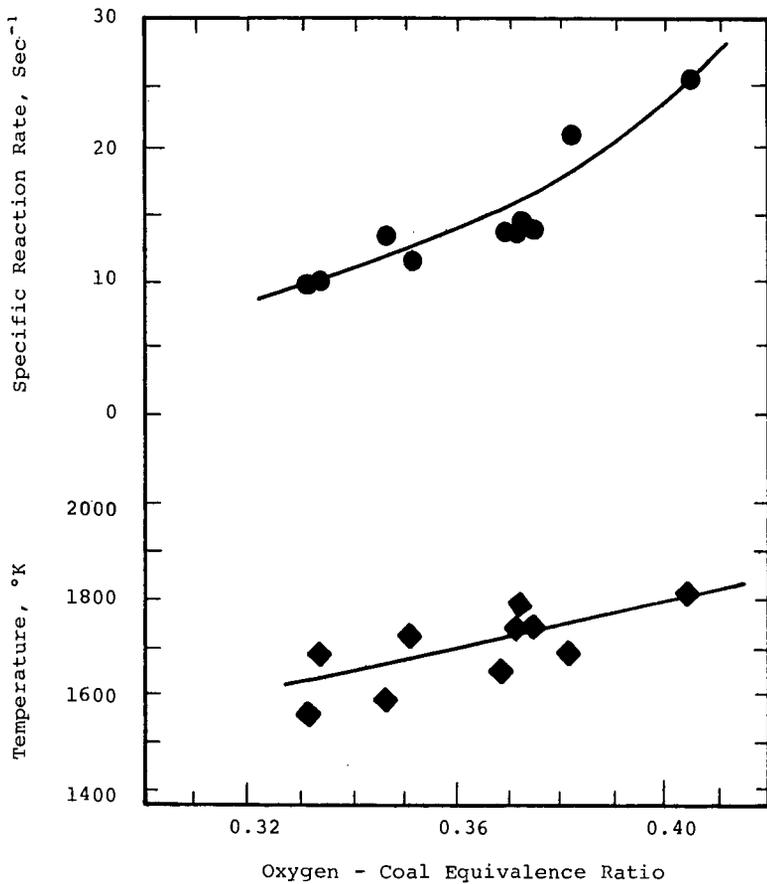


Figure 5. Specific reaction rates and reactor outlet temperatures calculated from Deseret Mine Coal test data.