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Energy and Environment Division

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THE COMBUSTION OF SOLVENT REFINED COAL
IN AN OPPOSED FLOW DIFFUSION FLAME*

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ABSTRACT

Interest in the combustion characteristics of coal and coal derived fuels relates primarily to their adaptation in existing and planned combustion devices and their propensity to produce increased pollutant emissions in comparison with petroleum fuels and natural gas. This work on solvent refined coal (SRC) is part of an experimental study of the mechanism of the burning of coal and coal derived fuels employing an opposed flow diffusion burner. SRC is a reconstituted tar-like material which has been solvent extracted from coal. This refined product is low in both sulfur and ash in comparison with the parent coal.

Cylindrical rods were cast for burning, some containing fine imbedded thermocouples. The burning rate of the SRC was measured for a range of oxidizer (oxygen and nitrogen) blowing velocities and oxidizer oxygen mole fractions and the dependency on these parameters determined. The temperature profile in the condensed phase fuel was determined from a time to distance transformation made possible by the steady state nature of the burning. The similarity of the burning characteristics of SRC and pressed pulverized coal suggests that similar mechanisms control the burning of these two fuels in our experiments.

INTRODUCTION

The continuing depletion of cleaner fuels such as natural gas and petroleum has made coal once again a primary energy source. Due to the increasing importance of pollutant emissions requirements, the direct combustion of coal necessarily encounters difficulties. Through solvent refining, the sulfur and ash levels of coal may be reduced. Solvent refined coal (SRC) is a reconstituted tar-like material which has been dissolved and chemically reacted with some cleaning agent, filtered, and separated from its solvent. Because this is a newly developed fuel, its combustion characteristics have not been extensively evaluated. Early combustion tests of SRC were conducted by Combustion Engineering, Babcock and Wilcox, and U.S. Bureau of Mines (1964-1965) and recently by McCann et al. (1976) in a multiburner. Difficulties in pulverization, transport, and burner plugging were reported by most investigators due to low fusion temperature (438°K). Successful burner modifications have increased the operation time; however, the clogging problem remains unresolved.

We have been studying the mechanism of the burning of coal and coal derived fuels in an opposed flow diffusion flame burner. An earlier experimental study on the steady state combustion of graphite and of pressed pulverized coal with significant ash production in the opposed flow of oxygen/nitrogen mixtures has been reported, Chin and Sawyer (1977). This previously developed apparatus, Holve and Sawyer (1974), was employed to observe the steady state diffusion flame combustion of solvent refined coal. The test fuel came from the Fort Lewis, Washington pilot plant owned by the Pittsburg & Midway Coal Mining Company and sponsored by the Department of Energy.
The feed coal for the SRC was a mixture of Kentucky 9 and 11. The properties of the SRC are summarized in Table 1.

Table 1. SRC properties, Pittsburg & Midway Coal Mining Co. (1976)

<table>
<thead>
<tr>
<th>Ultimate, mass %:</th>
<th>Carbon 86.00</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon</td>
<td>86.00</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>5.89</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>2.19</td>
</tr>
<tr>
<td>Sulfur</td>
<td>0.65</td>
</tr>
<tr>
<td>Oxygen</td>
<td>5.13</td>
</tr>
<tr>
<td>Ash</td>
<td>0.14</td>
</tr>
<tr>
<td>Higher Heating Value:</td>
<td>36.7 MJ/kg</td>
</tr>
<tr>
<td>Fusion Point:</td>
<td>438° K</td>
</tr>
<tr>
<td>Average density:</td>
<td>1.22 gm/cc</td>
</tr>
</tbody>
</table>

EXPERIMENTAL METHODS

The utility of the opposed flow diffusion flame (OFDF) apparatus for the study of the combustion of gaseous, liquid, and polymer fuels has been demonstrated by a number of researchers: Blazowski, et al. (1973), Evans and Emmons (1977), Holve and Sawyer (1974), Richard, et al. (1974), Sashadri and Weinberg (1965), Tien, et al. (1976) and Pendya and Weinberg (1965). We have employed this configuration, Figure 1, to observe the steady state combustion of solvent refined coal in a mixture of nitrogen and oxygen.

The advantages of this geometry lie primarily in the ability to observe the combustion of coal and coal derived fuels in a well controlled, steady state environment. It provides a simple geometry for analysis and also serves as a means of comparing the combustion
Figure 1. Combustion of solvent refined coal in an opposed flow diffusion flame.
of different types of coal and coal related fuels such as coal, pulverized coal, char, coke, solvent refined coal and coal liquids under similar burning conditions. The limitations of this technique lie in its failure to simulate the rapid heating rates and radiation environment which are likely to be characteristic of SRC combustion in existing combustion equipment. Additionally, observation of the combustion of particles or droplets is not possible.

A schematic of the opposed flow diffusion flame apparatus as used for study of SRC is shown in Figure 2. The fuel holder and oxidizer nozzle are contained in a 10 cm diameter pyrex cross to eliminate external disturbances to the flame and to aid in the collection of combustion product gases. One side of the cross serves as a viewing port and the other provides access for temperature and gas sampling probes. The oxidizer enters through the top flange and the fuel feed mechanism through the lower flange.

The oxidizer flow, consisting of nitrogen and oxygen, metered through calibrated rotometers, passes through a gas mixer and then enters through a converging nozzle filled with 3.0 mm steel balls followed by a collimated holes structure with holes of 0.01 mm in diameter and a wire screen to provide a flow of uniform velocity and composition. A portable torch is used to ignite the fuel sample. Once ignition takes place, the torch is removed and steady state burning is established within several minutes.

The sample feed system has been designed to provide for the accurate placement and determination of the location (within ± 10 μm) of the fuel surface. A laser beam and photo diode detector are used to sense the burning fuel surface, providing a signal to a stepping motor which
Figure 2. Opposed flow diffusion flame apparatus.
automatically positions the fuel rod. A 100 nm narrow band optical filter was positioned in front of the photo diode to eliminate the background noise from the burning surface. The linear regression rate is found by an electronic counter which reports the number of pulses to the stepping motor over a selected time interval. Data can be recorded and presented either as accumulated counts (total linear regression) as a function of time or the average regression rate for a selected time interval as a function of time. The steady state of the combustion can be established either by the linearity of the former or constancy of the latter. Also a digital to analog converter is used to provide direct measurement of the displacement of the sample surface as a function of time and is recorded by means of a strip chart recorder. The presently reported burning rates are determined from a best linear fit to the position-time data.

Both the fuel and the oxidizer were at a laboratory ambient temperature, approximately 20° C, and all experiments reported were conducted at atmospheric pressure. The measurements of the burning rate of the SRC were made by varying the primary experimental parameters--oxidizer flow velocity and oxygen fraction.

RADIATION HEAT LOSS

The geometry of the experiment is such that there is little radiant heat transfer from high temperature product gases in the combustion environment to the burning fuel. Rather, there is substantial heat loss from the burning fuel surface to the surroundings. The rate at which a solid body emits radiation is given by Stefan Boltzman law:

$$q_R = \varepsilon \sigma T_s^4$$  \hspace{1cm} (1)
where $q_R$ is the radiant heat flux emitted by the surface, $\bar{\varepsilon}$ is the total hemispherical emissivity of the surface, $\sigma$ is the Boltzmann's constant $5.6693 \times 10^{-12}$ W/cm$^2$°K$^4$. The calculation of radiant heat exchange between bodies and surfaces of irregular geometry is a fairly complex task because the procedure has to involve the heat balances in which account must be taken of the reflected and absorbed radiation. In our small scale experiment, although gases like water vapor and carbon dioxide are both absorbing and emitting gases, the optical length is small. So for the purpose of simple analysis of radiant heat exchange with the surroundings, we can assume the product gases are transparent and also the surface is isothermal. Furthermore, if we assume that the burning surface is a char, its properties may be approximated by those of graphite. The total hemispherical emissivity for graphite at $1777°$ K, which is the surface temperature measured with an oxidizer flow velocity of 131 cm/sec and an oxygen mole fraction of 0.8, is 0.7, Gubareff et al. (1976). For radiant energy exchange between two bodies, the expression is given by

$$ q_{1z2} = \bar{\varepsilon} \sigma F_{1-2} (T_1^4 - T_2^4) $$

where $F_{1-2}$, the shape factor, for energy exchange between a surface and its surroundings is 1. From (2) $q_{1z2}$, the radiant heat loss from the surface, is found to be 39.54 W/cm$^2$ or 100.2 W if we assume an emitting surface is a hemisphere of radius 0.635 cm. This is about 30% of the rate of energy release from combustion. Because of this heat loss, it is not possible to burn most coal or coal derived fuel samples in air, but oxygen enrichment is required to sustain combustion. The limiting oxygen index for the burning of coal in this geometry
is greater than .21 with the determination of the precise level difficult because of poorly defined extinction and ignition conditions.

PREPARATION OF FUEL SAMPLE

Because of the low melting temperature of SRC, it was possible to cast cylindrical samples of 12.77 mm in diameter and about 70 mm in length in a pyrex mold. The samples were then machined to have flat surfaces at both ends. Thermocouples made of platinum and platinum 13% Rhodium of 0.25 mm diameter were used to sense the temperature distribution along the fuel in some of the samples. The thermocouple was imbedded into the sample at the time of casting the sample. The thermocouple leads come out radially on opposite sides of the rod to reduce the importance of heat loss by conduction through the wire. The sample's density was measured by a standard displacement method.

CHAR PROBLEM

During the combustion of SRC, the surface geometry was quite irregular. This irregular geometry on occasion would block the laser beam that was supposed to signal the feedback mechanism to raise the surface. To solve this problem we have constructed a mechanical ash removing device consisting of a fine tungsten wire which was driven in a translating motion above the burning surface by a variable speed motor. Samples removed from the surface by the mechanical device were collected and analysed by combustion analysis and the results are compared with the SRC sample in Table 2.
Table 2. Analysis of fuel and removed char/ash residue.

<table>
<thead>
<tr>
<th></th>
<th>SRC fuel</th>
<th>sample</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weight %:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon</td>
<td>86</td>
<td>81.36</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>5.89</td>
<td>0.93</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>2.19</td>
<td>1.46</td>
</tr>
<tr>
<td>Sulfur</td>
<td>0.65</td>
<td>0.4</td>
</tr>
<tr>
<td>Oxygen</td>
<td>5.13</td>
<td>10.55*</td>
</tr>
<tr>
<td>Ash</td>
<td>0.14</td>
<td>5.3</td>
</tr>
</tbody>
</table>

* by difference

From the analysis we know that the samples collected are largely char, that is, they contain large amounts of carbon. The effect of the mechanical device was tested by three separate experimental observations at different oxygen concentrations and blowing velocities, with and without the mechanical device to remove the irregular char surface which blocks the positioning beam. The results showed (somewhat surprisingly) that the regression rates obtained were all within ± 20% with no consistent pattern which would suggest that mechanical ash/char removal accelerates burning. This is consistent with an estimate that less than about 10% of the fuel is mechanically removed and fails to burn.

**BURNING RATES**

Burning rates were measured and recorded as linear rates for SRC for a range of oxidizer blowing velocities from 131 cm/sec to 381 cm/sec and oxygen mole fractions from 0.45 to 1. A typical record of the regression rate history of an SRC sample burned at 131 cm/sec and oxygen mole fraction of 0.8 is shown in Figure 3. The irregularities are caused
Figure 3. Experimental regression curve for solvent refined coal burning in mixture of \(0.8 \text{O}_2 + 0.2 \text{N}_2\); oxidizer blowing velocity, 131 cm/sec; sample density, 1.22 gm/cc.
by the build up of the ash and char at the surface and their irregular removal. As the ash and char begins to develop on the burning surface, the apparent burning rate falls as the feedback control mechanism cannot distinguish between the ash and unburned fuel. When the ash is removed, the sample is then advanced to bring the fuel surface up to the set point for the fuel location, producing a stepped output in the regression record.

The formation of the rounded structure of the char at the surface (because the burning surface was set above the cooling collar) and destruction of the structure by the blowing effect and combustion are all taking place during the time interval at which no fuel advancement occurs. Averaged over these unsteady ash build-up and removal processes, however, the burning rate is nearly constant with time.

The reproducibility of the sample preparation and burning rate measurements is shown in Figure 4. Four different samples, all with a density of $1.22 \text{ gm/cm}^3$, oxidizer composition (0.50 $\text{O}_2 + 0.50 \text{N}_2$), and oxidizer flow velocity of 381 cm/sec are represented. The regression rates found by best squares fit are within $\pm 20\%$ of the mean of the regression rates. The reproducibility of burning rates measurements for multiple samples was also noted at other oxidizer flow velocities and oxygen mole fractions. It was found that the deviations in the regression rates were constant. The deviation could possibly be due to errors made in the initial positioning of the sample surfaces as this affects area exposed to combustion, and therefore, the regression rate.

The effect of oxidizer concentration was investigated by burning samples at several different oxidizer blowing velocities, Figure 5.
Figure 4. Experimental regression curve for solvent refined coal samples in a mixture of .5 O₂ + .5 N₂; oxidizer flow velocity, 380 cm/sec; sample density, 1.22 gm/cm³.
Figure 5. Experimental regression rate dependence on oxygen concentration. Blowing velocity: \( \bigcirc \), \( \square \), \( \Diamond \) are 381 cm/sec, 263 cm/sec and 131 cm/sec respectively; sample density, 1.22 gm/cm\(^3\).
The dependence of regression rate on oxygen mole fraction at different oxidizer blowing velocities is seen to increase with oxygen concentration in a fashion suggesting a power law dependence. These same data, relative to the lowest burning rate data point, are shown on a log-log plot in Figure 6. Assuming a linear fit, the empirical regression rate dependence on oxygen mole fraction at a fixed blowing velocity has the following form

\[ (r - r_{ref}) = A(X_{O_2} - X_{ref, O_2})^B \]  

where the values of B are .76, .54 and .45 corresponding to the blowing velocities of 381 cm/sec, 263 cm/sec, and 131 cm/sec respectively. The coefficient A is influenced by the blowing velocity.

The dependence of regression rate data on oxidizer blowing velocity at different oxygen concentrations is shown in Figure 7. At each oxygen concentration, the linear regression rates are found to be linearly dependent on oxidizer blowing velocity with slopes varying from \(3.0 \times 10^{-5}\) to \(3.5 \times 10^{-5}\). Holve and Sawyer (1974), found a 0.5 power dependence of regression rate on the oxidizer blowing velocity for both charring and non-charring polymers. Our value of 1.0 is higher than both their prediction and measurement. This could be due to the changing of the surface geometry with flow velocity. At lower flow velocity the surface area available for combustion is less than the case of higher flow velocity (the surface becoming more rounded as flow velocity increases). Radiation heat loss is likely to be more significant at lower flow velocities and may also contribute to the observed burning rate dependence on flow velocity.
Figure 6. Power law dependence of regression rate of solvent refined coal on oxygen concentration. Oxidizer blowing velocity: O, □, ◊ are 381 cm/sec, 263 cm/sec and 131 cm/sec respectively; sample density, 1.22 gm/cm³.
Figure 7. Dependence of experimental regression rate on oxidizer blowing velocity. Oxygen mole fractions: □, ○, △, ▽ are .5, .6, .7, .8 and 1 respectively; sample density, 1.22 gm/cm³.
COMPARISON WITH THE COMBUSTION OF PRESSED PULVERIZED COAL

In a previous study of the burning of pressed pulverized coal (Chin and Sawyer, 1977), a 0.74 power dependency on oxygen mole fraction was found at a blowing velocity of 381 cm/sec. This is surprisingly close to the .76 power dependency found for a blowing velocity of 381 cm/sec for SRC. Linear regression and mass burning rates for solvent refined coal are approximately two times those of pressed pulverized coal under similar conditions of oxidizer velocity and oxidizer oxygen mole fraction. This difference is consistent with the dependence of the burning rate of condensed fuels on the mass transfer number and that the mass transfer number of SRC will be greater than that of the pressed pulverized coal (although the values for the fuels used are not known). The similarity of the burning characteristics of these two fuels suggests that the rate limiting process for both may be established by the char oxidation.

TEMPERATURE PROFILES

The condensed phase temperature distribution in the SRC sample was measured by means of an imbedded thermocouple, Figure 8, for a blowing velocity of 131 cm/sec and an oxygen mole fraction of 0.8. The change of phase (solid to liquid) takes place at about 4 mm from the char surface. The maximum temperature measured was about 1777° K.

CONCLUSIONS

The experimental program has demonstrated the suitability of the opposed flow diffusion flame technique for studying the combustion of SRC, graphite, and pressed pulverized coal. Samples can be burned under steady state conditions for as long as 15 minutes and burning
Figure 8. Condensed phase temperature distribution for solvent refined coal at a blowing velocity of 131 cm/sec; oxygen mole fraction of 0.8, and sample density of 1.22 gm/cm³.
rate data obtained. The technique is particularly useful in the comparison of combustion of different fuels under similar conditions. The interpretation of fundamental processes controlling the combustion is complicated by the formation of ash deposits on the surface of the sample, by the rounding of the burning surface, and by a large radiation heat loss from the burning surface.

Under the conditions of these experiments, the burning rate of SRC was found to have a first power dependency on oxidizer blowing velocity and a .46 to .76 power dependency on oxidizer oxygen mole fraction. Temperature profile measurements establish that the melt interface lies about 4 mm below the burning surface and that the surface temperature is 1777° K (uncorrected).
REFERENCES


Seshadri, K. and Williams, F.A. Structure and extinction of counterflow diffusion flames above condensed fuels.

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