

STUDIES ON IGNITION OF COAL: THE EFFECTS OF RANK, TEMPERATURE, VOLATILE CONTENT, AND LITHOTYPE

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ABSTRACT

The rates of CO and CO₂ production from single particles of bituminous coal and lignite introduced into a hot, flowing oxidant gas stream were measured as a function of residence time. The time between injection of the particles into the combustion furnace and initial CO₂ production was defined as the ignition delay. These delays, as well as the maximum combustion rates and CO₂:CO product ratios are reported. Two gas temperatures were employed, 923 K and 1273 K. For the bituminous coal studies, temperature was the only parameter varied. For the lignite studies, the effects of volatile content and lithotype were examined in addition to temperature. The effect of volatile matter content was ascertained by comparing a coal to its char. Ignition delays were most affected by the gas temperature, decreasing substantially with increased gas temperature. Activation energies were estimated from the ignition delays. Variations of ignition delay are discussed in terms of the ignition mechanism.

INTRODUCTION

An important consideration in the design of coal combustion systems is ignition delay. In qualitative terms, the ignition delay can be described as the time from the combination of a fuel with an oxidant at high temperature to the beginning of the exotherm. Ignition characteristics of a given coal are important in the prevention of spontaneous ignition, in the production of stable flames, and in the establishment of optimum temperatures in fluidized bed combustion.

Several reviews of pulverized fuel combustion behavior are available (1,2,3). Parameters which affect the ignition behavior of coal and chars include rank and gas temperature. Both ignition delay and gas ignition temperature have been shown to increase with increasing coal rank (4,5). Lignite chars have been shown to be more reactive than other chars (6) and mineral matter has been found to increase the rate of combustion (7). Volatile matter has been shown to affect the mode of ignition of subbituminous and bituminous coals (8,9). Surface area has also been shown to influence ignition behavior (10). The overall objective of this study is to define the effects of moisture content, lithotype, volatile matter concentration, and mineral matter distribution on the ignition delay and combustion rate of single lignite, and subbituminous and bituminous coal particles. To date, data has been obtained from one lignite and one bituminous coal.

The most commonly monitored events used to determine ignition points are visible light production, mass loss, heat effects, and detection of products. Wall, et al. (11) have examined the correlation of light emission with combustion product formation and concluded the latter to be a better

indicator of the onset of stable combustion. Evolution of CO and CO₂ was chosen as the primary means of determining ignition because it is relatively unresponsive to volatile production and provides input for a mass balance.

Apparent Arrhenius activation energies and preexponential factors can be determined from gas ignition temperatures and ignition delays (4,12,13). The analysis assumes heterogeneous ignition and is accomplished through a heat balance on the particle. The unsteady-state balances have been integrated in this work to obtain Arrhenius parameters.

UNSTEADY STATE PARTICLE TEMPERATURES

The ignition process is modeled by assuming that a spherical particle is contacted by hot gas (14,15). The particle is heated by the environment and begins to react with the oxygen present in the atmosphere. The reaction increases the particle temperature which can be substantially higher than the gas temperature. The particle temperature, as a function of time, is determined from unsteady-state energy and mass balances:

$$dm/dt = A_s \rho \quad 1)$$

$$mC_p (dT_p/dt) = A_s \rho \Delta H - hA_s (T_p - T_g) - \epsilon \sigma A_s (T_p^4 - T_w^4). \quad 2)$$

Here m is the mass of the particle, while A_s is the surface area. The energy balance equation accounts for the heating of the particle by reaction, where ρ is the overall rate of reaction and ΔH is the enthalpy of reaction, and the loss of heat by conduction to the ambient gas and by radiation to surrounding walls. The temperatures of the particle, wall, and gas are T_p , T_w , and T_g , respectively. The heat transfer coefficient, h , is determined from the Nusselt number, Nu , which is given by

$$Nu = hd/\lambda \quad 3)$$

The limiting value of the Nu is two and Nu depends on d , the particle diameter and λ , the thermal conductivity of the gas, which is a function of temperature. The emissivity, ϵ , is taken to be unity, and σ is the Stefan-Boltzmann constant.

Mass and area are related to the diameter through

$$m = \pi \rho_p d^3/6 \text{ and} \quad 4)$$

$$A_s = \pi d^2 \quad 5)$$

The overall rate of combustion is determined from the rate of diffusion of oxygen to the surface, ρ_{diff} , and the rate of chemical reaction, ρ_{chem} , where these terms are given by the equations:

$$\rho_{diff} = k_{diff} (P - P_s). \quad 6)$$

where k_{diff} is given by

$$k_{diff} = 48D_o/dRT_m \quad 7)$$

and

$$\rho_{chem} = k_{chem} (P_s)^n \quad 8)$$

Here the chemical rate constant is assumed to have Arrhenius form:

$$k_{chem} = A \exp(-E/RT) \quad 9)$$

Since the surface pressure (P_s) is unknown, an approximation employed here is to use the maximum rate of both diffusion and reaction and to calculate the overall rate from a resistances in series formula (16)

$$1/\rho = 1/\rho_{diff} + 1/\rho_{chem} \quad 10)$$

where n has been assumed to be unity. The diffusion coefficient, D_o , depends on temperature and is evaluated at the mean temperature of the particle and the gas.

The enthalpy of reaction and the heat capacity are functions of temperature and in these calculations the char particle is assumed to be composed entirely of carbon. Combustion is assumed to proceed to CO as the product, because, for short times, the temperature versus time curve is insensitive to product mix. The unsteady-state balances, Equations 1 and 2, are solved by numerical techniques to yield particle temperature versus time curves. The initial particle temperature is assumed to be 300 K. This calculation is repeated for several values of the activation energy and preexponential factor until the calculated and experimental ignition delays agree. Several temperature versus time curves are shown in Figure 1 for gas temperatures of 1273 K and 923 K.

EQUIPMENT AND EXPERIMENTAL PROCEDURE

The ignition apparatus was similar to that employed by Gomez and Vastola (8). A detailed side view of the ignition apparatus combustion zone is shown in Figure 2. The system includes a tube furnace with hot zone, a particle feeder, and a gas mixing system. The combustion products are measured with CO and CO₂ analyzers which are connected to an A/D converter and a minicomputer to collect concentration versus time data. Thermocouples monitor wall and gas temperatures. The particle feeder is a stainless steel tube and is grounded to prevent electrostatic charge accumulation. A quartz window provides optical access for the trigger system.

Coal samples were obtained from the Pennsylvania State University Sample Bank and included a Beulah lignite (PSOC-1507) and Pittsburgh #8 (PSOC-1451) bituminous coal. The lithotypes vitrain, attritus, and fusain were physically separated from the coal samples. Particles were sized under a microscope and were observed to be irregular in shape. Hence, an aspect ratio was calculated from the formula $2a/(b+c)$, where the axes are ordered $a > b > c$.

The coal or char particles are first picked up by a suction bulb and

sized under a microscope. The experiment is started by placing the particle in a syringe needle and pushing it into the drop tube with a thin wire. Data collection from the CO/CO₂ analyzers is started when the particle travels past the trigger. The gas flow rate through the apparatus was 200 ml/min and the gas composition was 80% N₂ and 20% O₂. The linear velocity was 26 cm/sec.

The apparatus was calibrated using malonic acid to produce CO₂ at 413 K which acted as a marker to measure the time for flow between the source and the detector. Further details are available (17).

Char of the untreated lignite was produced in an air natural gas flame at approximately 3% excess oxygen. The chars were collected with a nitrogen-quenched extraction probe at 200 mm above the injection point, corresponding to a residence time of 150 ms. Gas temperature varied from approximately 1500 K at the particle injection point to 1050 K at the collection point. After collection, the chars were sieved, and size ranges were selected for combustion tests. The coals and chars were stored over concentrated H₂SO₄ or water at 30°C for two weeks to establish relative moisture contents of 0 and 100%, respectively (18).

RESULTS AND DISCUSSION

The CO₂ concentration versus time curve yields the ignition delay once the system calibration constant is known. The total carbon production is determined from the summation of the CO and CO₂ concentrations. The data is then differentiated to determine the gas production rate as a function of time. Examples of the rate versus time curves are given in Figures 3 and 4. The peaks in these curves give the maximum CO and CO₂ production rates. Arrhenius parameters were estimated by varying the E and A constants in Equation 9 to match the ignition delay time.

Water had no discernable effect on the ignition delay, combustion rate, or CO₂/CO product ratio of Beulah lignite vitrain, as shown in Table 1. Lithotype had an effect on the ignition delay of Beulah lignite with fusain igniting the fastest. Karz et al. (13) have reported that fusains have shorter ignition delays than vitrains obtained from an anthracite and a bituminous coal.

The effect of volatiles on the combustion behavior of Beulah lignite can be seen by comparing the results obtained from char to those obtained from burning the parent lignite (see Table 1). The char reacted as rapidly as the most reactive lithotype and produced a similar, yet somewhat lower, maximum combustion rate. The overall CO and CO₂ production profiles obtained from the char did not differ significantly from those observed for the lignite shown in Figure 4. Heterogeneous ignition of the lignite is indicated since its combustion behavior is similar to that of its char, which can only ignite in a heterogeneous manner.

The effect of material rank on ignition delay and combustion rate can be seen from comparing the Beulah lignite results to those obtained from Pittsburgh #8 bituminous coal. These two materials behaved similarly at 1273 K, however, at 923 K a substantial difference was noted. At 923 K, the bituminous coal was less reactive than the lignite, exhibiting a longer ignition delay and a lower maximum combustion rate. A comparison of the

overall profiles, Figures 3 and 4, shows that the bituminous coal burns in two distinct stages as opposed to the single stage of the lignite. The two stages represent initial volatiles combustion followed by char burnout. The single stage combustion of Beulah lignite indicates that its char is highly reactive, a fact supported by the char data.

Increasing the gas temperature dramatically decreased the ignition delay time for both the Pittsburgh #8 vitrain and the Beulah vitrain. The Arrhenius activation energy is estimated to be about 20 kcal/mole for a preexponential factor of 200 g/(cm²s) when determined from the delay times. The particle temperature versus time curves for two gas temperatures calculated using Equations 1-10 are presented in Figure 1. These compare favorably with the 29.6 and 20 kcal/mole used by Bandyopadhyay and Baduri (4) for bituminous coal and lignite, respectively. In addition, Karcz et al. (13) obtained 17.6 kcal/mole from experiments on bituminous vitrain. Wall and Gururajan (12) cite a value of 17 kcal/mole for a lignite, and Patel et al. (18) used TGA to measure an activation energy of 18 kcal/mole for a char burned for 100 ms at 1030 K in a flame burner.

CONCLUSIONS

The production of CO and CO₂ was measured as a function of time for several low-rank coals and chars. Detection is possible for individual particles of about 200 μm. Activation energies obtained from ignition delay times agree well with measurements from TGA experiments as well as steady-state Semenov analyses. Ignition delay depends strongly on gas temperature and, to a minor extent, on lithotype. The Beulah lignite char was demonstrated to be a highly reactive material and comparison of its ignition behavior to that of the lignite indicated that the lignite ignited heterogeneously.

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TABLE 1
IGNITION RESULTS FOR SEVERAL COALS AND CHARs

Material	Number of particles	I (K)	Particle Volumex, 10^{-6} (cm ³)	Aspect Ratio	Maximum Rate $\times 10^{-6}$ (g/sec)	CO ₂ /CO Product Ratios	Delay (ms)
Beulah Vitrain, wet	7	923	8.3 ± 1.5	1.6	3.9 ± 1.1	0.99 ± 0.21	376 ± 62
Beulah Vitrain, dry	11	923	5.4 ± 1.3	1.7	4.5 ± 1.1	0.84 ± 0.16	419 ± 51
Beulah Fusain	7	923	6.7 ± 1.9	1.8	6.3 ± 1.4	0.88 ± 0.33	302 ± 51
Beulah Attritus	9	923	9.8 ± 5.1	2.1	7.2 ± 2.7	0.78 ± 0.25	371 ± 36
Beulah Vitrain	8	1273	6.9 ± 2.0	1.7	5.3 ± 1.2	1.19 ± 0.36	137 ± 38
Pitts #8 Vitrain	7	1273	6.2 ± 1.8	1.4	4.5 ± 1.8	2.01 ± 0.53	120 ± 79
Pitts #8 Vitrain	8	923	6.5 ± 2.9	1.7	0.99 ± 0.25	--- ^a	480 ± 34
Beulah Char	7	923	7.4 ± 3.1	2.5	4.2 ± 2.4	1.06 ± 0.30	309 ± 63

a. Not available

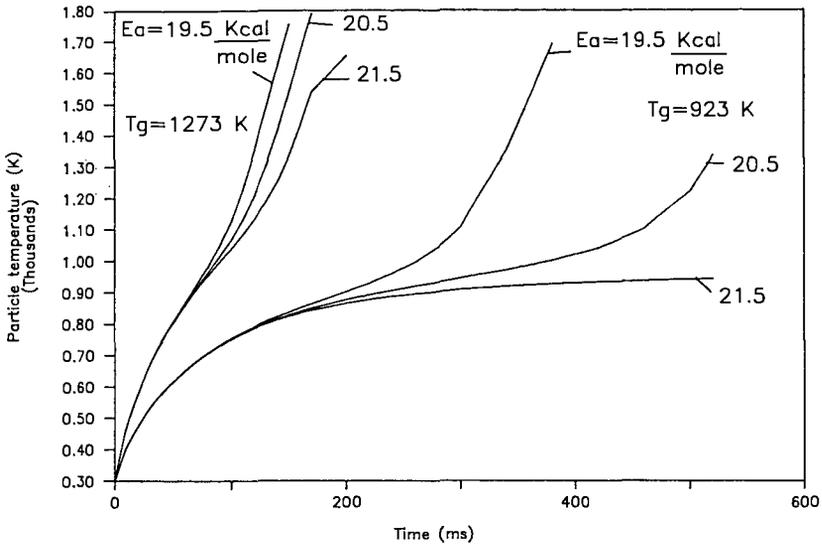


Figure 1. Calculated Temperature Profiles for 200 μm Particles

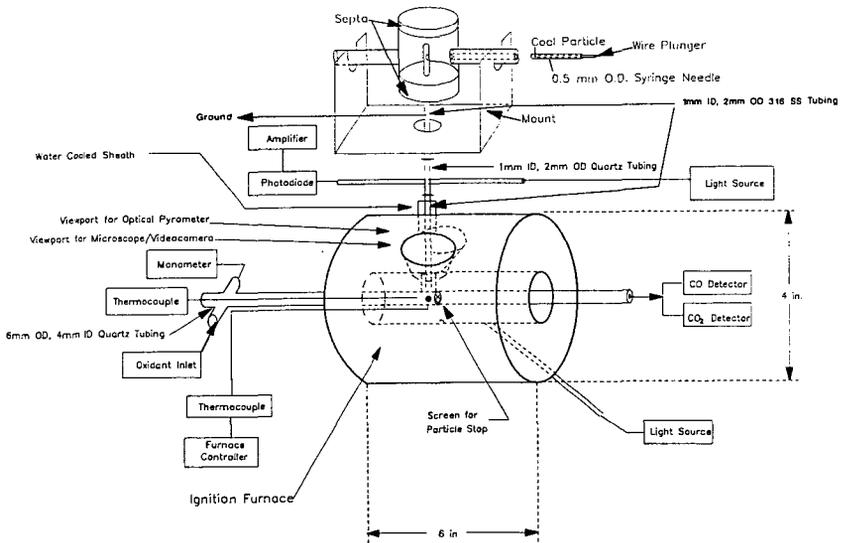


Figure 2. Detailed Side View of the Ignition Apparatus

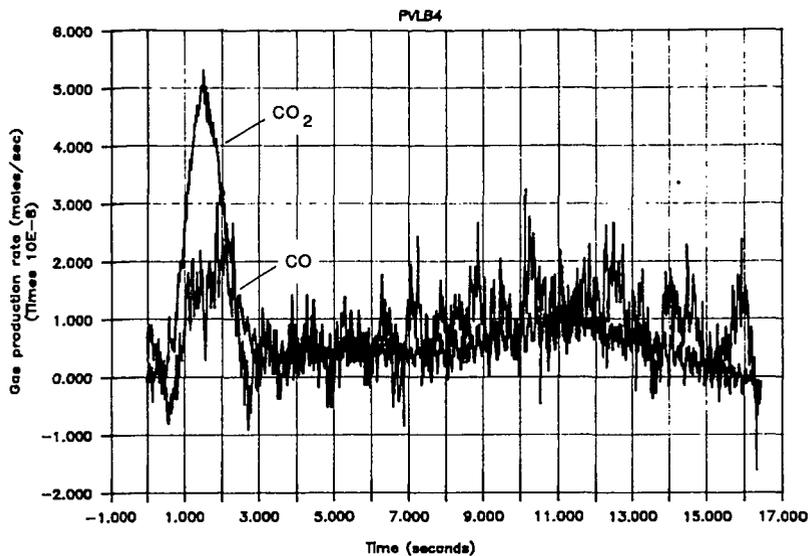


Figure 3. CO and CO₂ Production Rate Profiles of Pittsburgh #8 Coal at 923 K

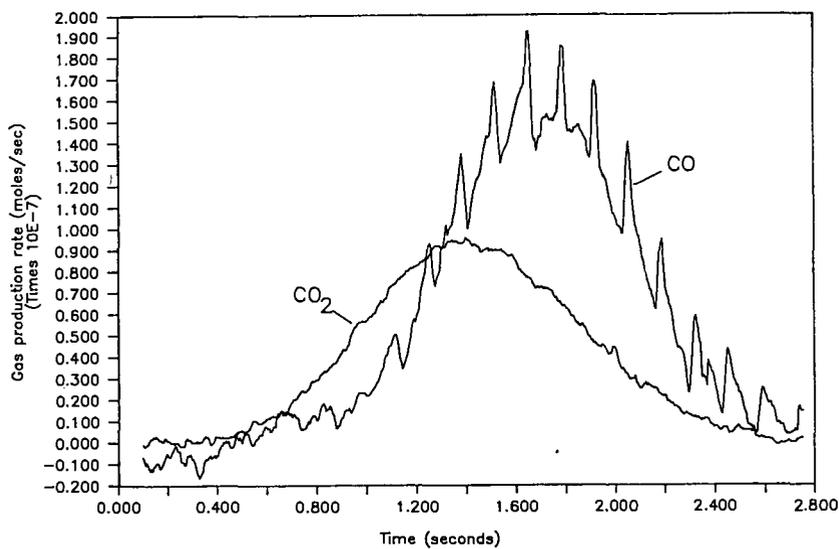


Figure 4. CO and CO₂ Production Rate Profiles of Beulah Lignite at 923 K