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COMBUSTION RATES OF COAL-WATER SLURRY DROPLETS.

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Abstract

This paper reports on experimental findings and theoretical calculations on the combustion rates of coal-water fuels. Combustion of coal-water fuels (CWF) takes place upon evaporation of the water and heat-up of the remaining solid agglomerate. Combustion takes place in two distinct phases: (i) burning of the volatiles in an envelope flame, and (b) burning of the char heterogeneously. Volatile combustion takes place at high temperature (2500-2800 K in air at gas temperatures of 1050-1450 K). The Chars ignite at temperatures in the vicinity of 1400-1500 K and burn a little above 2000 K in air. Swelling of the bituminous coals was observed during the devolatilization stage. Fragmentation at the end of the char combustion was occasionally observed. All experiments were conducted in a bench scale, high temperature furnace electrically heated to gas temperatures up to 1600 K. Single CWF droplets (100-600μm, in diameter) were generated inside the furnace, and upon ignition their combustion behavior was monitored with a specially developed three wavelength (640, 810 and 998 nm) pyrometer. This instrument has been calibrated against sources of known temperature, such as an NIST tungsten lamp and the melting point of pure metal wires. Pyrometric observations on the behavior of burning CWF particles were conducted from the top of the furnace so that the complete combustion behavior of the particles, from ignition to extinction, could be monitored. Experiments took place in a variety of furnace temperatures and oxidizing atmospheres. Char surface or volatile flame temperature data collected as a function of time in the course of these experiments have been used, in conjunction with particle physical properties, to derive combustion rates for CWFs.

INTRODUCTION

Coal-water fuels (CWF) also called coal-water slurry fuels can be used as a substitute for oil in oil-fired furnaces[1,2,3] with a small, in principle, loss in energy[4]. Thus, depending on their price competitiveness with petroleum in the international markets CWF can provide coal-rich countries with an energy alternative. They can even be more attractive than dry pulverized coal because they can be transported easily and safely in pipelines with centralized crashing units. They are also optimum for micronization and beneficiation and for mixing with sulfur removal agents since dewatering is not required. Problem areas include those associated with ignition and flame stability because of the large water content, with the selection of materials end equipment to handle the abrasiveness of CWF, with slurry settling and solidification, etc.

Coal water fuels typically contain 65-70% by weight coal[5] the balance being water and a small amount (<1%) of surfactants and emulsifiers. Regular grind slurries, for utility boiler combustion applications, have mass median diameters (MMD) of $\approx 50\mu$m; micronized slurries, for special applications such as diesel engines and turbines, have

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MMD of >10μm. The latter, even if more costly, are superior because they enable a higher degree of beneficiation, i.e. removal of inorganic matter.

The water evaporation and combustion characteristics of coal-water fuels has been investigated in a fair amount of studies many of which have been described in the review papers of McHale[5], Papachristodoulou and Trass[6] and Miller[7]. Such studies varied from single drop combustion in bench scale equipment[4,8,9] to large scale spray-flames in pilot plant furnaces[1-3,10,11]. Moreover, similar studies involving fuels of coal-oil mixtures (COM), tar-coal mixtures, coal-alcohol mixtures, etc. have been investigated[12-14]. The economics of using coal-water fuels, however, are more favorable.

The fundamental, small scale studies seem to concentrate into understanding the differences of the combustion of CWF and COM drops/particles from those of more traditional fuels like liquid fuel droplets or solid coal particles. Upon evaporation of the water content, the combustion of CWF certainly resembles that of pulverized parent coal, however, typical sizes are larger, the porosity and pore structure are different and as result the mode of combustion may be different. The size of the agglomerates may be large enough to promote oxygen consumption at a high rate on the surface, thus, preventing combustion inside the pore network and resulting in longer burnout times. Because of the sensitivity of the combustion behavior on particle size, and particle structural parameters[15,16], as well as combustion environments[17], it is important to obtain combustion rates at conditions pertinent to those encountered in actual boilers.

Combustion of single CWF drops has been experimentally observed with a variety of techniques and equipment ranging from suspending droplets on thermocouple wires and quartz fibers over flat flame burners, to free falling droplets in similar furnaces, and levitating drops in electrodynamic balances. Four different stages have been identified: (i) evaporation of water, (ii) heat-up and pyrolysis of coal particle agglomerates, (iii) evolution and gas-phase combustion of volatiles, and (iv) heterogeneous combustion of the remaining char. The last step was observed to be the slowest requiring up to 90-95% of the droplet life-time[12]. Thus, provisions to provide adequate residence time in a boiler should be incorporated for complete burnout of the char.

The evaporation of water from the CWF droplet has been studied by Zghoul and Essenhigh[26] who observed that heat-up, evaporation and ignition times for CWS were proportional to the initial droplet diameter. Moreover, Maloney and co-workers observed water evaporation on levitated drops in an electrodynamic balance[27,28] and reported on secondary atomization and microexplosions under intense heating rates. On the other hand the combustion of the devolatilized char residue (fourth stage) has been modeled in the manner of a burning carbon sphere[5] since it was determined that the coal burns as a whole agglomerate particle rather than individual coal particles.

The present study is fundamental in nature, and aims in understanding the different stages of CWF combustion. For this purpose isolated CWF drops were monitored in a non-intrusive manner throughout their lifetimes in high temperature environments. The purpose is to help clarify uncertainties on the elevated temperature combustion of small size drops/particles, investigate the combustion characteristics, obtain accurate surface or flame temperatures and investigate combustion rate trends. Such information will facilitate calculation of radiative fluxes and residence times in furnaces.

In the present study experiments were conducted in bench scale drop-tube furnace where a special technique was used to obtain the time for evaporation of water from the CWF droplet. Ratio optical pyrometry[19-21] was employed to measure particle/flame temperatures and burnout times while SEM microscopy was used to infer structural data. A simplified model[21] was used to obtain apparent reaction rates.

EXPERIMENTAL

The slurries employed in this investigation are made of micronized and beneficiated coal supplied by *Otisca*. The coal is bituminous with 37.50% volatiles, 61.56% fixed carbon and 0.94% ash; the particle size varies from 0.5 to 7.5 μm (MMD $\approx 3.5\mu$m). The solid loading is typically 50%, the rest was water. Water was occasionally replaced by acetone to facilitate evaporation at low temperature experiments.

Production of Single Drops.

Techniques to produce single droplets of coal-water slurries have been described previously[22]. The injector used herein utilized an electrically actuated plunger to push a small quantity of slurry out of a thin needle. The size of the drops was controlled by various diameters of needle-plunger combinations. Particle sizes in the range of 100-600μm were generated with this technique. A photograph of the single droplet generator is shown in Fig. 1. At the upper end of the solenoid actuator a light source and a photodetector were mounted to sense the motion of the solenoid plunger. Thus, whenever the plunger moved downwards to expel a droplet from the injector the photocell was activated and a signal was recorded by the high speed data aquisition system. In this manner the onset of particle injection was monitored.

High Temperature Furnace and Optical Pyrometer.

The furnace is a drop tube, laminar flow device the shell of which is manufactured by *ATS*, utilizing *Kanthal Super 33* molybdenum disilicide heating elements, to heat a 25 cm long radiation cavity to temperatures up to 2000 K. A 60 cm long 7 cm i.d. high purity alumina tube, manufactured by *Coors*, has been installed along the centerline of the furnace, Fig. 2. Observations of the combustion events in the furnace were conducted from the top and the bottom.

A movable, stainless steel, water cooled injector, 57 cm long and 1.25 cm i.d., has been inserted at the top of the drop tube furnace. The injector was designed to to perform a dual function: (a) enable injection of particles or drops at various heights in the furnace radiation cavity, and (b) enable pyrometric observations of burning particles from the top of the furnace. Thus, the width and length of the injector were selected to maximize collection of light emitted from burning particles ($I_{rad} \propto (distance)^{-2}$), while simultaneously avoiding observation of *direct* radiation from the furnace walls.

Light Collection Optics. Optical observations were conducted from the top of the injector, along the centerline of the furnace. To monitor the complete burnout history of the solid particle, from ignition to extinction, the light collection optics were situated at the top of the furnace injector. Furthermore, at the top of the injector both the particle and drop generation systems were also located. To avoid obstruction of the optics the particle/droplet injectors had to be as small as possible, preferably less than 1 mm in diameter.

At the top of the furnace a collimating lens was placed to focus the light from burning particles on to one end of an optical fiber and transmit it to the optical pyrometer which is situated at the other end. The optical fiber serves as the coupling medium between the furnace and the pyrometer. The single optical fiber, *General Fiber Optic*, used in this design has 1 mm diameter and is 1.6 m long.

An inverted collimator lens (*Oriel*, 11 mm diameter, 19 mm focal length lens) is used to focus light on the optical fiber. To avoid *reflected* furnace radiation from climbing inside the injector (after multiple reflections on the inner walls) and reaching the optical fiber the injector was black passivated and a pinhole was used between the lens and the fiber to define an optical field width.

Optical Pyrometer. A three-color electronic pyrometer has been designed and constructed [23]. This instrument uses high transmission efficiency dichroic beam splitters to separate the radiation spectrum to the three detectors Three medium bandwidth (FWHM = 70 nm) interference filters (*Oriel*) were used to define the working wavelengths of 0.640, 0.810 and 0.998 μm. In conjunction with these interference filters, three 19-mm- focal-length aspheric lenses were used to focus the light beams onto the photo-detectors. Solid state silicon photo-detectors *Oriel* were used to transform the incident light into voltage signals. Photo-currents, generated at the detectors, are converted to voltages and are amplified 10^9 times using linear amplifiers (*Analog Devices, model #LF347*). The signals are then conducted to linear amplifiers (*Analog Devices*, model *#AD524*). All devices were powered by a solid state ±15 V power supply.

Data Acquisition. To automatically record the signals generated by the photodetectors to an IBM-AT microcomputer a *Data Translation DT2828* A/D converter with a *DT707* screw terminal board were employed. The DT2828 is a high speed simultaneous sample and hold converter, and has a maximum throughput of 100 kHz over 4 channels with single-ended input. Three channels are currently used, each at 33 kHz. When the fourth channel was used to record the motion of the droplet generator plunger, as mentioned before, the aquisition rate dropped to 25 kHz per channel. The A/D board has a 12 bit resolution over the 10 volt range, producing a voltage resolution of 2.44 millivolts.

The pyrometer was calibrated with a tungsten filament lamp purchased from NIST and with the melting point of pure metal wires[23].

EVAPORATION AND HEAT-UP PERIODS.

The time for evaporation of the liquid portion of the droplet had been determined earlier by theoretical calculations [24] for different coal loadings and different solvents such as water, acetone and ethanol. To experimentally determine this time a photo diode was connected across the solenoid of the droplet generator, as described earlier, so as to create a spike (of a certain voltage, in this case of 4V) whenever the solenoid was actuated. The wall temperature of the furnace for these experiments was maintained at 1500K. The flow of air through the injector was 0.1 lpm and an additional flow through the furnce was 2.0 lpm. The injected droplet covered the cool environment in the injector tube, where some evaporation took place, before entering the hot radiation zone. Upon entering the radiation zone massive evaporation (boiling) took place at ≈100°C. Upon completion of evaporation the particle (agglomerate) heated to the temperature of the gas and ignited (ignition of volatiles). This onset of luminous combustion was recorded by the pyrometer. Signals obtained from three different CWF droplets; two of them having diameters ≈ 250μm and the third ≈ 500μm are shown in Fig. 3. The recorded time for evaporation and subsequent particle heat-up was around 600 msec (time is counted between the middle of the photocell spike and the start of the luminous combustion). Only one of the intensity signals, the one obtained at 0.640μm, is shown on the plot. These signals were recorded with logarithmic main amplifiers, which are inverting in nature, so any increase in temperature and/or light intensity will be presented by a decrease in voltage signal. From the signal trace we can see an initial peak followed by a secondary hump. The initial peak is related to the release and combustion of the volatiles; the hump represents the combustion of the char residue. More on the combustion behavior of slurries will be given in the next section. This experimentally obtained water evaporation times can be contrasted with those calculated numerically by Cumper *et al.*[24] for 250μm size CWS drops. It was calculated that 250μm size droplets need ≈680 ms for complete evaporation if injected from the same height and the same environment as in the experiments. This is similar as the experimentally observed time. Of course there is some considerable uncertainty in the initial size of the drops generated by this device, and the actual mechanism of evaporation of water at the presence of the coal slurry, (i.e. formation of a shell with water trapped inside, swelling, further water evaporation, etc.) is different than the "water shell and carbon core" model that the calculation assumed. Furthermore, it is possible that leaching organics from the coal tars to the water alter the properties of the latter to a degree. Nevertheless, the agreement was very good indicating that the evaporation behavior of CWF droplets can be approximated by a simple model.

PYROMETRIC TIME-TEMPERATURE PROFILES OF BURNING CWF PARTICLES.

These studies were conducted with pre-dried CWF particles. The reason for this is given in the following. Since the CWF droplets obtained from the droplet generator were not of accurately controlled and repeatable size, the initial diameter of the dry CWF agglomerated particle was not known. This impaired the accuracy of subsequent combustion studies. Hence, the droplet generator was used in a separate reactor[25] to produce dried agglomerates. The temperature in that reactor was kept at the minimum required for complete water evaporation (600°C) and the atmosphere was nitrogen. The sizes of agglomerate particles were first measured under a microscope and subsequently they were dropped into the high temperature furnace, through the port for dry injection, by means of a bevel-shaped needle.

Upon entering the radiation cavity of the furnace the agglomerate ignited and burned in a fraction of a second. The pyrometer recorded the total burnout time, from ignition to extinction, of single agglomerate particles along with surface temperature. Temperature was derived using a Planckian analysis to the radiation intensities of three channels of the pyrometer[23].

Two typical profiles of CWF particles burning in air and oxygen are shown in Figs. 4 and 5, respectively. The top plot in each figure shows the radiation intensity traces at the three wavelengths of the pyrometer. The bottom plot depicts the flame/surface temperatures derived at the three wavelength ratio combinations. The gas temperature was 1350 K. Both initial dry particle diameters were $\approx 435 \mu m$. Given that these particles (agglomerates) have been observed to swell during devolatilization, with mean swelling factors of 1.12 the actual size was probably ca. $487 \mu m$. Strong signals, associated with volatile combustion in envelope flames are evident in both cases. Volatile combustion duration for these particles was in the order of 25% of the total. Total burnout times were 375 and 175 ms in air and O_2, respectively. Volatile flame temperatures were $\approx 500°$ higher than particle surface temperatures in the subsequent char combustion. The char burned at 2000 K in air and at 3000 K in oxygen. Combustion traces, such as those presented above, were recorded for particles of different sizes at gas temperatures in the range of 1250-1450 K.

REACTION RATES

This section reports on reaction rates for the char residue, i.e. after devolatilization is complete. The burning time for a spherical carbonaceous particle of radius, a, under diffusion control can be estimated using the analysis of Levendis *et al.*[21], assuming first order kinetics. Including the convective flow away from the char surface (Stefan flow), the oxygen conservation equation can be written:

$$\dot{m}_{O_2} = -\sigma_g D_b \frac{dY_{O_2}}{dr} + Y_{O_2} \dot{m}_{tot} \ , \tag{1}$$

where $\dot{m}_{O_2}$ is the flux of oxygen (g/cm^{2}s), $\dot{m}_{tot}$ is the net mass flux (g/cm^2 s), D_b is the bulk diffusion coefficient, σ_g is the average gas density in the film, r is the distance from the center of the particle (cm) and Y_{O_2} is the oxygen mass fraction. Assuming the product of combustion to be only CO, the following mass balance holds:

$$\dot{m}_{O_2} = -16/12 \dot{m}_{tot} \ , \tag{2}$$

Integrating between the particle surface, $r = a$ and infinity, $r = \infty$ and making use of the mass balance at the particle surface:

$$\sigma_a \left(\frac{dr}{dt} \right)_a = -\dot{m}_{tot,s} \ . \tag{3}$$

Using the diffusion equation to calculate the oxygen concentration at the particle surface, thereby correcting for the external diffusion resistance, the observed combustion rate can be related to the apparent reaction rate constant. Combining Eq. (2) and (3):

$$\dot{m}_{tot,s} = \sigma_g \frac{D_b}{a} \ln\left(\frac{1+3/4Y_{O_2\infty}}{1+3/4Y_{O_2s}}\right) = R_c Y_{O_2s}\sigma_g \qquad \mathrm{g/m^2s}, \tag{4}$$

where R_c is the chemical rate coefficient and the reaction order has been assumed to be equal to unity. Y_{O_2s} can be eliminated for the above equation by substituting: $Y_{O_2s} = Y_{O_2\infty}(1-\psi)$ and approximating the logarithmic term as

$$\ln\left(\frac{1+3/4Y_{O_2\infty}}{1+3/4Y_{O_2s}}\right) \approx 3/4\ \psi \frac{Y_{O_2\infty}}{1+3/4Y_{O_2\infty}}. \tag{5}$$

Substituting this expression in Equation (4), expressions for ψ and $\dot{m}_{tot,s}$ as a function of R_c can be obtained. The expression for $\dot{m}_{tot,s}$ is then used to integrate Eq. (3) and obtain an expression for R_c. Then, the apparent reaction rate constant is given as $\hat{R}_a = 32R_c/(RT)^n$, where 32 is the molecular weight of oxygen, R is the ideal gas constant (82.06 atm cm^3 mole^{-1} K^{-1}). An Arrhenius plot of the apparent rate constant vs temperature is shown in Fig. 6;[1] plotted on the same figure are the rates of various carbonaceous spheres, described in Ref. [21]. As expected the reaction rate increases with temperature. It can be seen that fair agreement is obtained between the beneficiated slurries of the present study and the high purity chars of Ref. [21]. The apparent rates of the CWF are a little higher, as may be expected by their enhanced porosity and cenospheric nature. The rate at temperatures above 3000 K drops, as observed earlier[21].

SUMMARY

Fundamental studies on the combustion of single coal-water fuel drops have been undertaken. Most studies were conducted with pre-dried CWF agglomerates to facilitate precise measurement of the initial particle size. The combustion behavior, the particle (agglomerate) temperature and the particle burntime were monitored with ratio optical pyrometry. It was found that CWS particles of a few hundred micrometers in diameter (100-600μm) burn with two distinct phases of volatile flame combustion and char oxidation. In air, at furnace temperatures in the range of 1250-1450 K the volatile combustion of these particles accounted for ca. 20% of the burnout time. The char burns at ca. 500 K above the gas temperature and the volatiles a few hundred degrees higher than the char. Apparent reaction rates for these chars are a few percent higher than those of pure carbonaceous spheres of various porosities.

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[1] Figure 6 appears at the end of the text before Figs 4 and 5 to comply with the page limit of the manuscript.

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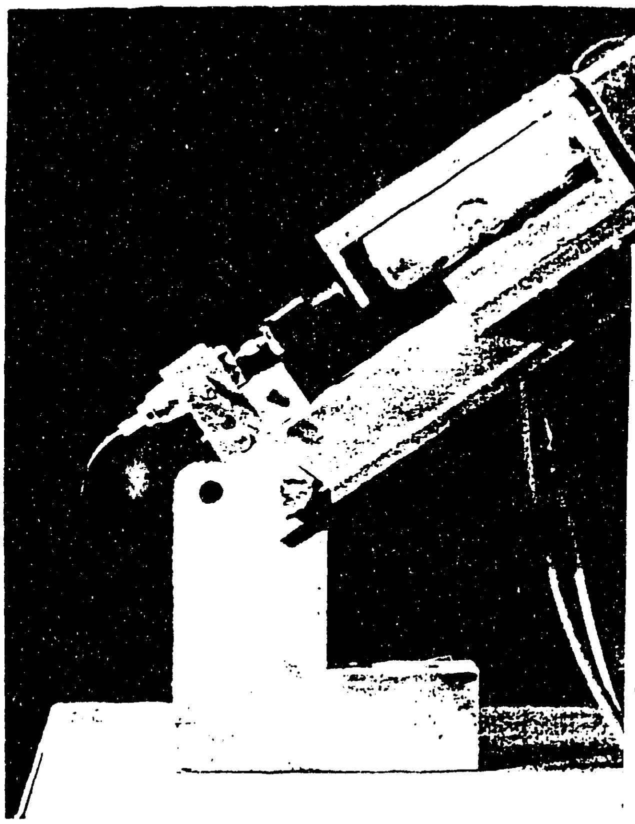
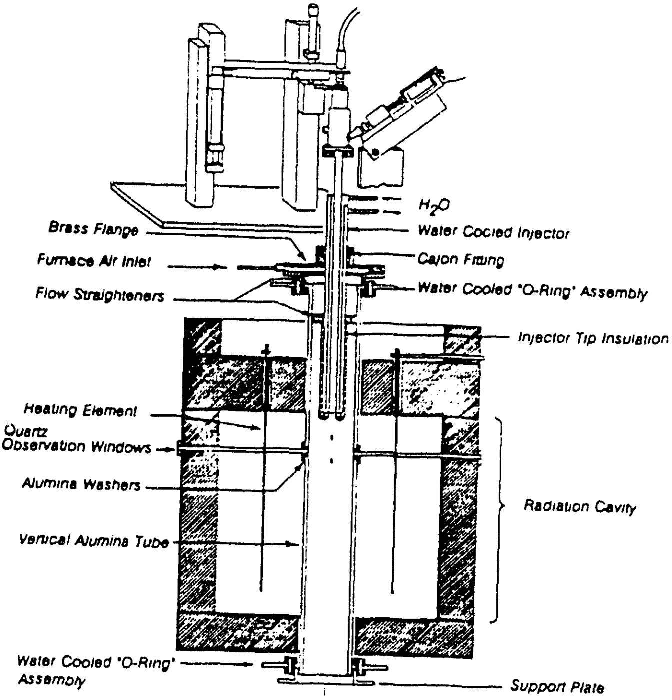
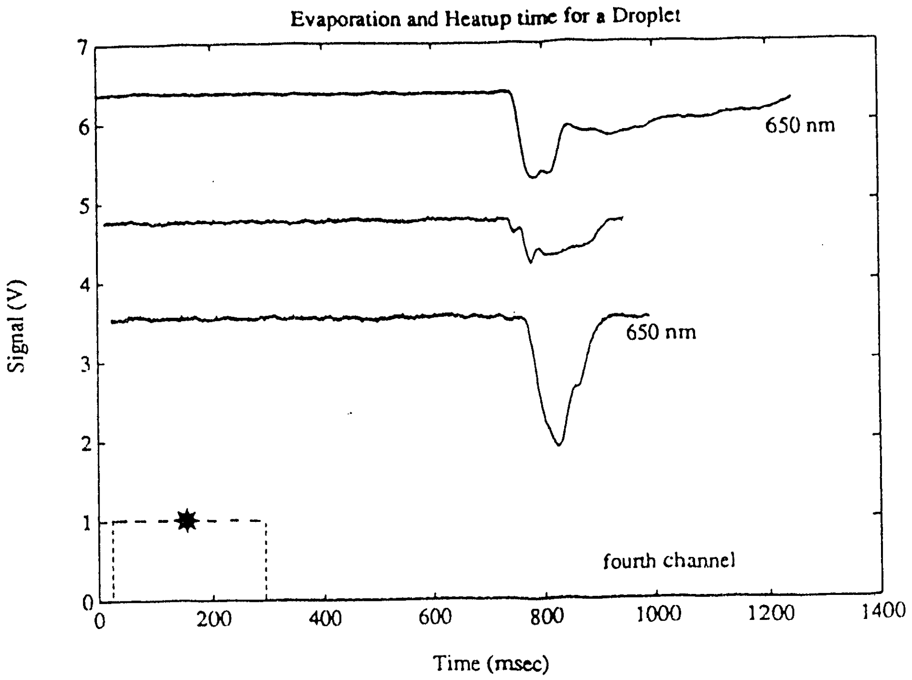


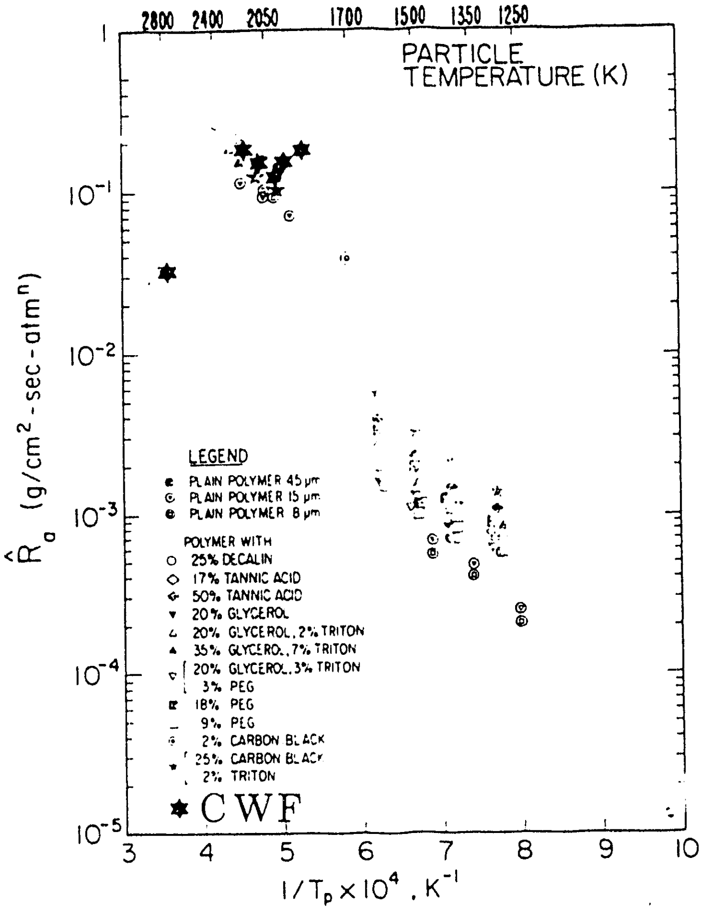


1. *FIGURE 1. (a) Mechanical plunger-needle drop generator. A photocell is incorporated in the housing at the top of the generator.*

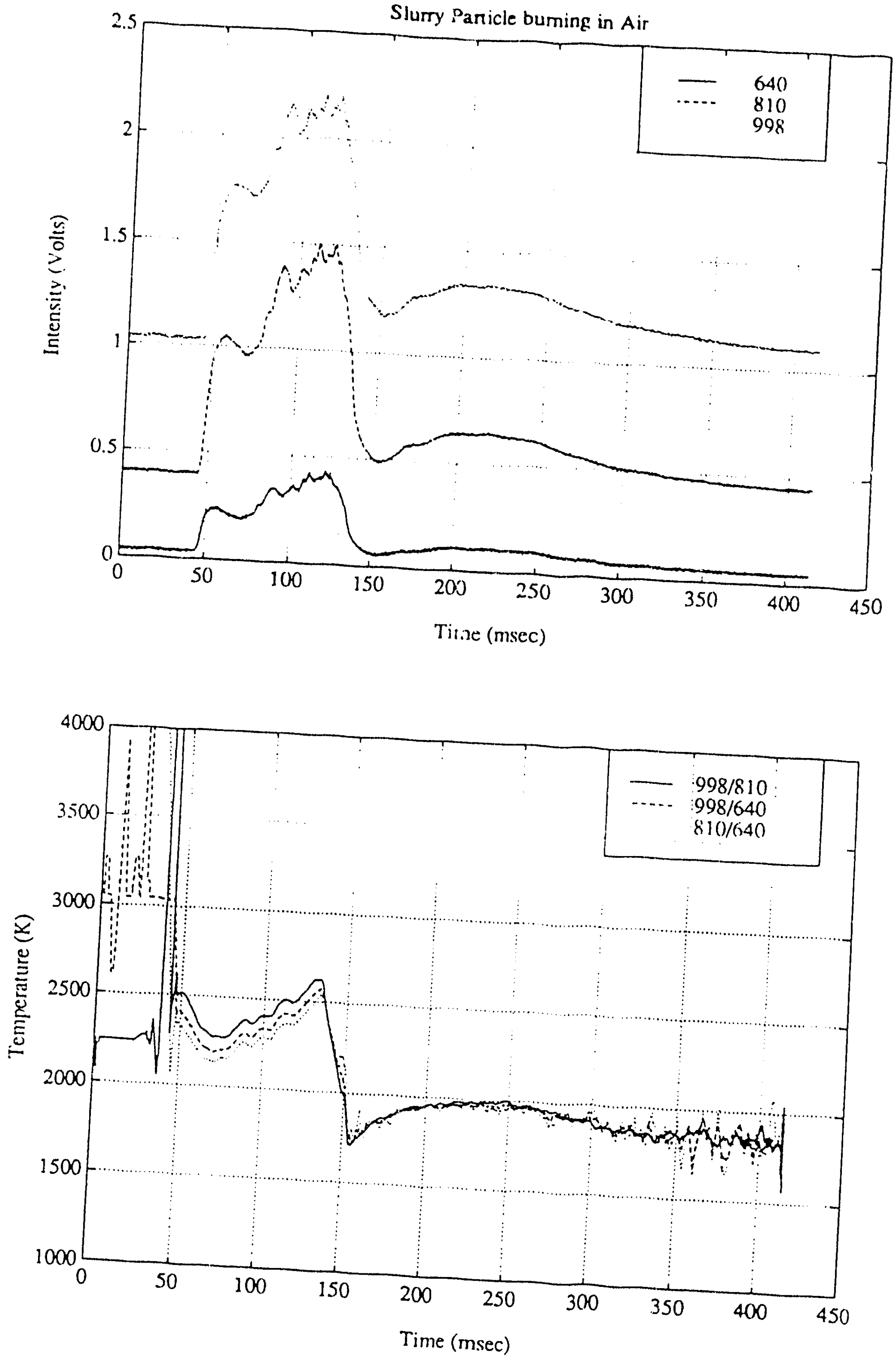
2. *FIGURE 2. Schematic of the laminar, drop-tube furnace, the injector assembly, and the light collection optics.*



3. *FIGURE 3 Water evaporation and combustion of three CWF drops. Signals were monitored by the 650 nm channel of the pyrometer. The square pulse was generated by the photocell. The three particle sizes are (a) and (b) $\approx 250\mu m$ and (c) $\approx 500\mu m$.*



6. *FIGURE 6. Apparent combustion rate constant vs $1/T$ for CWS agglomerates at T_g = 1450 K. Data on pure carbon chars from Ref. [21] is also shown.*



4. *FIGURE 4. (a) Intensity-time profiles, recorded by the ratio optical pyrometer. (b) Temperature-time profiles. Dry CWS-agglomerate, 435μm diameter, burning in air at T_g = 1350 K.*

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