

ONLINE MONITORING OF THE BURNING CHARACTERISTICS OF SINGLE PULVERIZED COAL PARTICLE IN O₂/N₂ AND O₂/CO₂ ENVIRONMENTS

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The objective of this study is to compare the ignition and burning characteristics such as burnout times of volatiles and char, and ignition mechanism of single pulverized coal particle burning in air (O₂/N₂) and oxy (O₂/CO₂) environments. An entrained flow reactor with photo detector has been employed for this study. This technique involves online monitoring of radiation emission from an individual coal particle. Individual particles of sub-bituminous or lignite coal particle with size in the ranges of 106 - 125 micron and 180 - 212 micron particles have been injected into an air (O₂/N₂) or oxy (O₂/CO₂) environment inside an entrained flow reactor. The oxygen concentration in the ambient gas is varied between 10% to 50% by volume. The volatile and char burnout times have been obtained from time histories estimated from the radiation emitted by the particle. The results show an obvious increase in the burning rates of volatiles and char with increasing oxygen concentration. Further, when compared to N₂ atmosphere, the burning rate and radiation intensity are found to be lower in CO₂ atmosphere, especially at lower oxygen concentrations. An indication of the possible ignition mechanism has been outlined from the percentages of single and double peaks observed in the radiation emission histories. The complete experimental investigation shows that the burning rates of both volatiles and char are predominantly affected by the oxygen concentration, particle size and gas temperature. The theoretical results from a single particle model has been used to validate the trends and the combustion durations obtained from the present experimental study and is published as a separate manuscript.

Keywords: *Devolatilization, Flame sheet, Oxy-fuel, Ignition, Char, Combustion; char burnout*

1. Introduction

The alarming increase in emission levels and growing concern over global warming necessitate the development of carbon capture and storage (CCS) technologies. The CCS technologies enable substantial reduction in CO₂ emissions, still allowing the use of fossil fuels. Oxy-fuel combustion is one of the CCS technologies, where the energy intensive CO₂ recovery process can be overcome by removing the nitrogen in the air before it enters the furnace. In this mode, combustion takes place in

pure or enriched oxygen leading to the production of a flue gas, which is primarily constituted by CO_2 and H_2O . For a safe and efficient operation, a part of the flue gas leaving the furnace is recycled back into the furnace. In oxy-fuel combustion mode, CO_2 level in the flue gas is enriched up to a higher level, where it can be economically removed. Apart from the capability to produce a highly concentrated CO_2 stream, this technology has also been credited for its flexibility, as it can be adapted in new designs and can also be used as a retrofit option [1-7].

The combustion medium in an oxy-fuel furnace is primarily a mixture of O_2 - CO_2 , as opposed to the normal air used in conventional combustion devices. The presence of CO_2 as a balanced gas alters the flammability and reactivity of the reactants. Therefore, this affects every aspect of combustion right from particle ignition, volatile combustion, flame stability, char reactivity, heat transfer, emission levels and also ash characteristics. Particle ignition and devolatilization under oxy-fuel condition have a substantial effect on near burner heat release rate and combustion stability. It in turn has a significant implication in an oxy-fuel combustor design. This is because of the fact that the burnout and particle residence time depend on the effective heat release rate. The effects of CO_2 can be understood more clearly by comparing the burning of coal particles in normal air and oxy-fuel conditions. This is required to identify the alterations needed for a stable and complete combustion in a O_2 - CO_2 environment.

2. Literature review

2.1. Ignition characteristics in O_2/CO_2 environment

Many studies have been carried out to investigate the combustion characteristics of individual coal particles, coal particle jets and coal dust clouds under oxy-fuel conditions. The experimental studies reported in the literature range from bench scale to the demonstration scale. Few modelling studies have also been reported in the literature. Detailed reviews on oxy-fuel combustion studies have been reported in literature [1-9]. Experimental and modelling studies with respect to pilot plants have reported an altered heat transfer profile in oxy-fuel conditions due to enhancement in the radiation absorption property in the presence of CO_2 when compared to that of N_2 [10, 11]. The combustion temperatures were also found to be lowered due to gas-radiation as well as due to the higher specific heat capacity of CO_2 . Investigations on flame characteristics in oxy-fuel conditions indicated a reduced flame stability, drop in flame temperature and flame propagation velocity [12]. These were attributed to the combined effects of the differences in the properties of the inert gas (N_2 and CO_2) and in the burner configuration having modified aerodynamics. Char reactivity and fuel burnout studies showed mixed trends with increasing or decreasing volatile yields in high-temperature oxidation conditions. Few studies claimed a significant char-gasification reaction [13, 14] and better burnout, while a few other investigators observed contradicting results [15, 16]. In general, coal ignition and devolatilization studies in oxy-fuel conditions indicated a delayed ignition with longer volatile and char burnout times [17, 18]. Studies on gas emissions in oxy-fuel mode reported a significant reduction in NO_x and SO_x emissions as the concentration of CO_2 increased from 16% in air to around 90% in oxy-fuel condition [19, 20]. The lower combustion temperature observed in oxy-fuel conditions had a significant impact on the transformation of mineral matter and vaporization of inorganic materials [21]. The overall conclusion from the previous studies was that there have been some undesirable characteristics when N_2 is replaced by CO_2 in oxy-fuel combustion, and this,

however, can be overcome by enhancing the partial pressure of oxygen in the ambient gas and also with a modified burner design.

Though extensive studies have been reported, there is a definite need for fundamental research that can promote better understanding of burning characteristics of coal in oxygen enhanced mode, in the presence of CO₂. The data obtained from such fundamental studies could serve as inputs for comprehensive models for predicting the dynamics of oxy-fuel combustion closely. Such studies from literature have been reviewed below.

2.2. Single particle studies in O₂/CO₂ environment

An extensive research work has been carried out in the past on single coal particle to draw the effect of various operating conditions on the ignition and combustion behavior in different coal ranks [22-28]. Investigation on a single coal particle eliminates all other effects due to the cloud effect where the volatile cloud predominantly affects the devolatilization and further burning of char. To avoid particle to particle interaction and cloud effect, a single particle study has been carried out. Molina and Shaddix [18] investigated the ignition and devolatilization characteristics of a single pulverized coal particle in an oxy-fuel environment. The experiments were carried out in Sandia's optical entrained flow reactor, where pyrometry and particle imaging techniques were used to measure the particle's temperature and its movement. The influence of CO₂, substituted for N₂, and the effect of enhanced oxygen concentration, were examined. Similar features such as delayed ignition, increased devolatilization duration and reduced particle temperature were observed in CO₂ environment as compared to that in N₂ environment, for the same oxygen levels and ambient gas temperatures. The authors attributed these differences to higher molar heat capacity of CO₂ and lower mass diffusivity in CO₂ compared to N₂ [22].

A detailed investigation on the burning features of a single particle of bituminous coal and that of lignite coal with different size fractions was carried out by Bejarano and Levendis [17]. They conducted their tests in the O₂/N₂ and O₂/CO₂ environment and used an electrically heated drop tube furnace instrumented with pyrometry and obtained similar outcomes as in earlier studies. They also reported that differently ranked coals behaved differently in an oxygen enhanced environment. These differences were observed in studies involving different configurations, such as in coal particle jets and coal dust clouds [19]. The change in the mass diffusivity of oxygen in the presence of CO₂ alters the proportion of mixing and thus, the reactivity of the local fuel-oxidizer mixture. This, in turn, affects the heat release rate, especially during the combustion of volatiles. A higher oxygen mole fraction in the oxidizing stream is expected to enhance the rates of the oxidation reactions, and thereby decrease the ignition delay time [18]. Rathnam *et al.* [14] reported that the differences in reactivity of coal particles burning in O₂/N₂ and O₂/CO₂ environments were almost insignificant.

2.3. Motivation for current study

Pulverised coal combustion has been studied under different ambient media, such as O₂/N₂ and O₂/CO₂. The differences in the thermo-physical properties between different media have been identified as the main reason for the differences in the burning characteristics in those media. Few of the studies have reported that the difference is also due to the coal type and flame aerodynamics (burner configuration). The present study is motivated by seeing the gap in literature with respect to the scarcity of a basic study involving the burning of isolated coal particles in a high heating rate

environment (similar to that of a burner) with different oxidation media. The O_2 levels in the oxidation medium was varied in the range of 10% to 50% by volume. Either N_2 or CO_2 is used as the balance gas. The deliverables from this study will aid in the availability of fundamental data to be used in the modelling of oxy-coal combustion.

3. Experimental

3.1. Entrained Flow Furnace and Methodology

The experiments were conducted in a laminar entrained flow reactor at atmospheric pressure, high temperature simulated air (O_2/N_2) and oxy-fuel (O_2/CO_2) environments. The main advantage of this reactor is that it can attain high heating rates and temperatures comparable to real combustion conditions. The reactor consists of a flat flame burner at the base. The simulated air or oxy-fuel environments with required composition and at the required temperature are obtained by burning a required mixture of $CH_4+H_2+O_2$ with required amounts of N_2 or CO_2 , respectively. The products consisting primarily of CO_2 , H_2O , N_2 and O_2 provide the necessary oxidizing environment for the coal particles to burn. The schematic of the experimental setup, along with the radiation detection system, is shown in Fig.1.

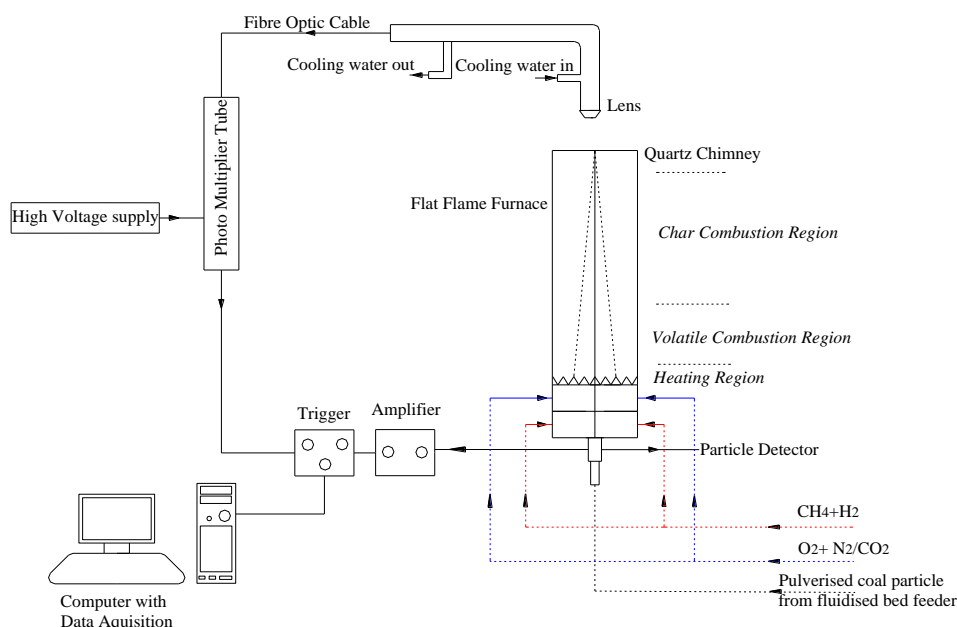


Figure 1. Schematic of the experimental setup with the radiation detection system

The hot products are allowed to steadily flow through a square cross-sectional chamber of a furnace having dimensions of 50 mm × 50 mm. The total gas flow rate through the furnace has been maintained as 40 slpm for all experimental trials. The coal particles are allowed to be entrained by the stream of carrier gas (N_2 or CO_2) into the post flame gas medium by injecting it axially from the bottom using a fluidized bed feeder. The coal feed rate has been maintained low enough (1- 1.5 g/h) to ensure that only a single particle enters the furnace at a given instant. The particle that travels in the axis of the optics alone is considered for sampling. Each particle entering the furnace is detected using a particle detector, which triggers the data collection system. A Photo Multiplier Tube (PMT) collects the radiation emitted by the burning particle at wavelengths within its responding range. The PMT used in this experimental study is Hamamatsu R636, which has a response range of 180-900 nm with

the best quantum efficiency in the wavelength band of 300-800 nm. When a particle enters the furnace and gets ignited, the radiation emitted by the particle has been aligned using a lens (Hamamatsu E717-21) mounted above the furnace onto the optically active cross sectional area of the PMT. The light emission is channelized to the PMT through a water cooled fibre optic cable. The PMT collects emissions at all wavelengths within its responding range but quite insensitive in the infrared range. The radiation intensity collected from the burning particle is used to calculate the burnout times of volatiles and char. The loss of intensity due to the wavelength dependence of the focal distance and particle motion has not been considered as the focus of the present study is primarily to compare the burning characteristics between the medium having N_2 and CO_2 , keeping all other parameters the same. The PMT is housed in a dark enclosure and the experiments were conducted in a darkened room in order to prevent interference from external light. The accuracy check and calibration of the instruments are detailed elsewhere [29]. The intensity is collected every millisecond and the recorded intensities have been stored directly in a computer-based data acquisition system supported by Lab VIEW software.

A separate experiment has been conducted to determine the minimum temperature (brightness) that can be detected by the PMT by using a radiation source of known temperature. The experiments show that the minimum brightness that can be detected by the present PMT is experimentally found to be the intensity more than or equal to that which corresponds to 1300 K.

4. Results and Discussion

The results from single particle experiments are presented in this section. Total wavelength pyrometer (PMT) has been used to indicate the burning patterns and to calculate the burnout times. The volatile (devolatilization time) and char burning times obtained from the PMT radiation history are carried out on two types of coal at two coal particle sizes and gas temperatures with oxygen concentration in the combustion medium varying from 10% to 50% by volume. The present study uses coal particles of sizes larger than the size fraction used in PCI furnaces. However, to eliminate the particle to particle interaction effect

4.1. Ignition Mode from radiation profile

The radiation history obtained from the particles is categorized as single and double peak profiles to identify the mode of ignition. Figure 2 shows the examples of radiation intensity traces obtained from burning particles. The devolatilization and char burnout times in the present study have been calculated by considering the profiles with two peaks alone. The time calculated from the single peak profile can be treated as the total burnout time; however, they are not discussed here.

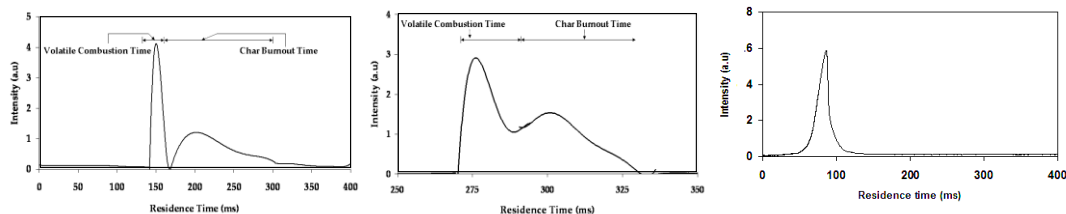


Figure 2. Examples of intensity traces obtained from burning particles separated (left) and overlapping (middle) and single peak (right) traces

In the current study, no clear trend in the mode of ignition is observed with respect to variations in coal type, oxygen concentration, ambient temperature and particle size. Out of the hundreds of profiles collected, the percentage of profiles with double peaks with distinct volatile and char burning stages is seen to be higher for bituminous coal as compared to that of lignite for any given operating condition. The repeatability of the experiment has been verified. The experiments for all the cases were carried out thrice and the statistical analysis carried out showed the results were repeatable with 10% error (the maximum is 10%)

4.2. Burnout times in O_2/N_2 and O_2/CO_2 environments

4.2.1. Effect of oxygen concentration

The volatile burning times obtained from the intensity traces are discussed in the following section. Figures 3 and 4 show the effect of oxygen concentration on devolatilization times of lignite and sub-bituminous coal particles, respectively, having sizes in the range of 180-212 microns at furnace temperature 1800 K. The data points are the mean values of devolatilization times obtained from the intensity profiles recorded from hundreds of particles. The spread in the measured data are represented as one sigma error bars. The oxygen concentration used in the PCI furnace with air lies within 21%. However, in oxy cases, the deviation in the combustion characteristics from air combustion can be matched by increasing the oxygen concentration in the combustion environment beyond 21%. However, the oxygen concentration cannot be increased beyond 50% as the furnace gets overheated.

The devolatilization duration decreases with increasing oxygen concentration. For lignite, it drops from 28 ms at 10% oxygen concentration to 14 ms at 50% oxygen concentration in an N_2 environment. The corresponding values in the CO_2 environment are 38 ms and 16 ms, respectively. In the same range of oxygen concentration, the devolatilization time varies from 25 ms to 14 ms in N_2 and from 33 ms to 17 ms in CO_2 environments in the case of sub-bituminous coal. These results indicate that the volatile burning time is higher in an O_2/CO_2 environment.

In general, the volatile burning time decreases as the oxygen concentration increases. The homogeneous oxidation reaction rates increase as the concentration of oxygen increases. Further, the flame temperature and the rate at which oxygen diffuses to the flame zone increase as the oxygen concentration increases. It is well known that volatile release is a thermal process, when the coal particle is heated to a temperature of around 300 °C to 400°C, volatiles start to evolve. Therefore, the ambient temperature of 1550 K or 1800 K itself is sufficient for volatile release. If the evolved volatiles are burnt at a faster rate, then the rate of mass diffusion of the volatile gases will increase. This is achieved as oxygen concentration is increased. As a result of these effects, volatile burning time decreases.

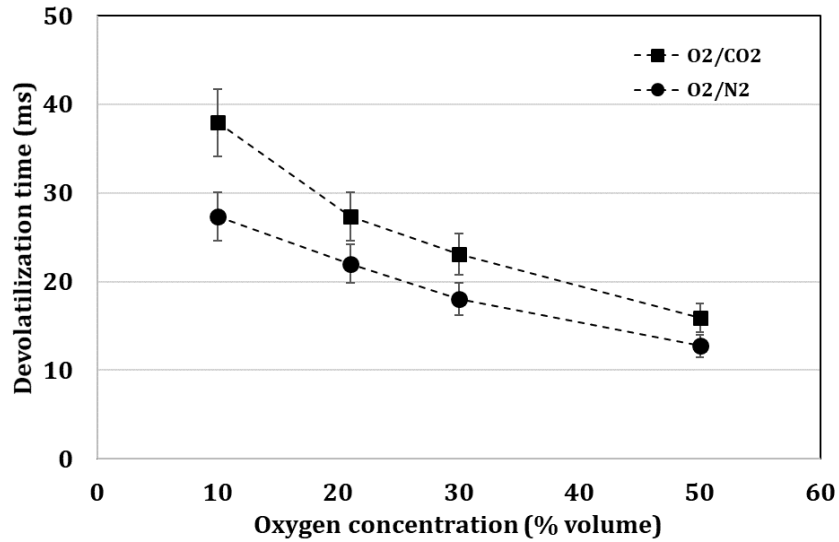


Figure 3. Measured devolatilization time of sub bituminous coal (180- 212 μm) in (O_2/N_2) and (O_2/CO_2) environments burning at 1800 K

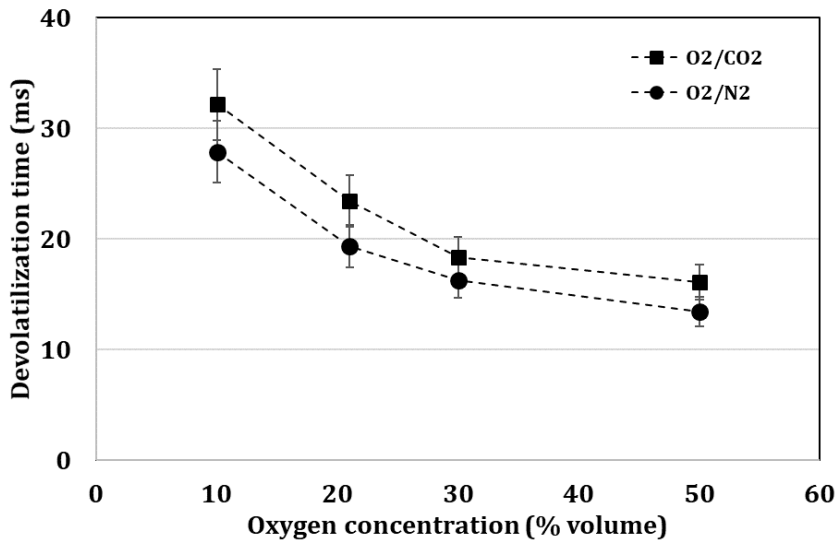


Figure 4. Measured devolatilization time of sub bituminous coal (180- 212 μm) in (O_2/N_2) and (O_2/CO_2) environments burning at 1800 K

In comparison between N_2 and CO_2 environments, the volatile burning time is higher in CO_2 environments. This is due to three-fold effects: (1) higher specific heat of the gas mixture, (2) increased radiation heat loss due to the presence of increased amounts of absorption species such as CO_2 and (3) reduced diffusion rates as a result of the increased molecular mass of the mixture. The first two effects cause a relative decrease in the flame temperature and the third effect causes a relative reduction in the rate of oxygen transport to the flame zone in a CO_2 environment as compared to that in an N_2 environment. Due to this, the volatile burning time is higher in the CO_2 environment.

Figures 5 and 6 show the effect of oxygen concentration on char burnout times. The experimental results show a decreasing trend in the char burn-out time with increasing oxygen concentration. The char burnout time in N_2 and CO_2 increases from around 60 ms to 75 ms at 10% oxygen concentration and from 25 ms to 40 ms, at 50% oxygen concentration for both

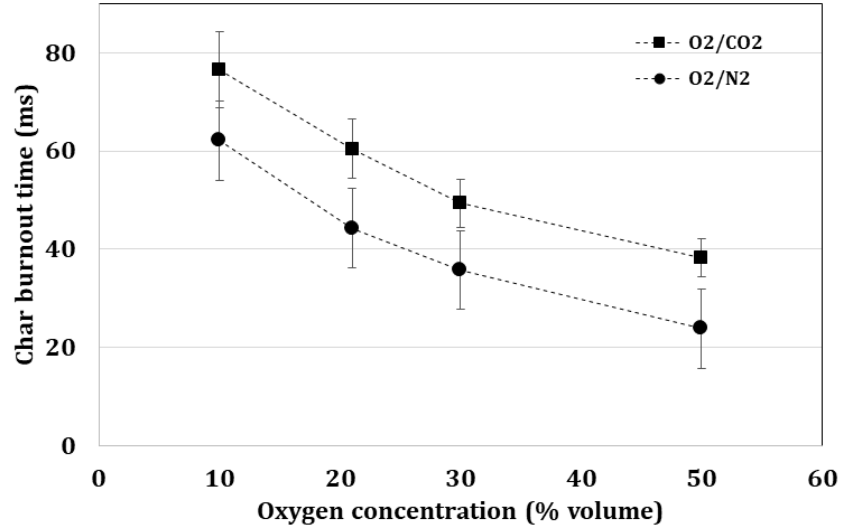


Figure 5. Measured char burnout time of lignite (180- 212 μm) in (O_2/N_2) & (O_2/CO_2) environments burning at 1800 K coals. An Increase in the diffusion of oxygen towards the particle surface forms the primary reason for such variation. The burn-out time in the CO_2 environment is higher. This is because of both reduced oxygen diffusion and flame temperature. The differences in the values between N_2 and CO_2 environments are almost constant for lignite at all oxygen concentrations and the differences decrease with increasing oxygen concentration for sub-bituminous coal. The difference in the fixed carbon content imparts more char burn-out time for sub-bituminous coal.

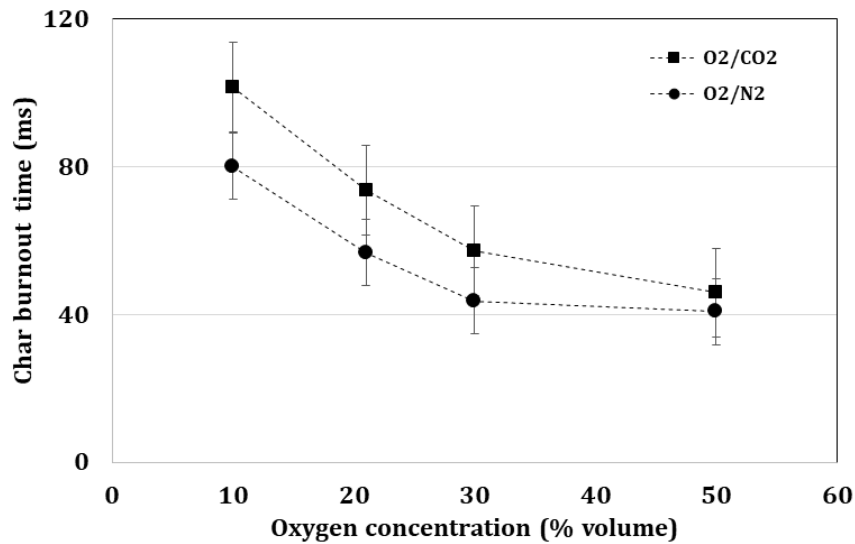


Figure 6. Measured char burnout time of sub bituminous (180- 212 μm) in (O_2/N_2) and (O_2/CO_2) environments burning at 1800 K

It is interesting to observe that in some cases, the magnitudes of char burnout times are comparable to devolatilization times. At higher oxygen concentration, it is attributed to the higher particle heating rate leading to faster char burning. At low oxygen levels, when the radiation intensity levels go below the minimum detectable limit, the intensity drops to a dark intensity. This causes a portion of the otherwise longer profiles to be cut short resulting in reduced combustion durations.

4.2.2. Influence of bulk gas temperature

The devolatilization and char burnout times of the coal samples in air (O_2/N_2) and oxy-fuel (O_2/CO_2) conditions are found to be shorter at 1800 K compared to 1550 K. The temperatures chosen were based on the earlier experimental studies on air combustion and on the limitation of the experimental setup and the range of the measuring devices used. The durations are longer in the CO_2 environment at both the bulk gas temperatures. As the ambient temperature is increased, the reactivity or the reaction rate increases. The effect of furnace temperature on the volatile and char combustion duration are shown in Fig.7 and Fig.8.

The difference in burn out time is attributed to the increase in reactant temperature at the envelope of the volatile cloud and higher oxygen diffusivity at higher temperatures which pushes the flame closer to the particle surface. This impacts both devolatilization and char burning through an improved energy feedback to the particle from the flame sheet. Also, at higher gas temperatures, the heat loss from the particle to the gas will be reduced. Previous studies reported a negligible effect on the combustion behaviour, especially on the devolatilization time [26,28].

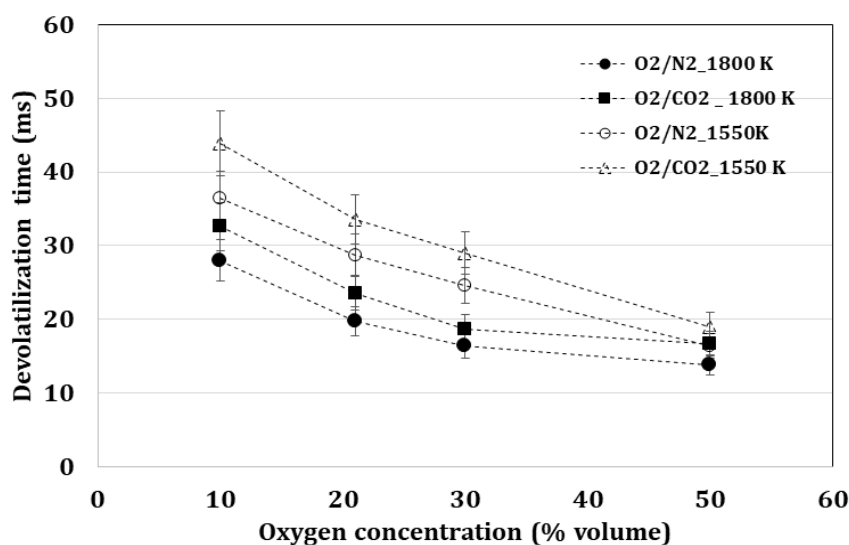


Figure 7. Effect of bulk gas temperature on the devolatilization time of sub bituminous (180- 212 μm) in (O_2/N_2) and (O_2/CO_2) environments

The decrease in char burnout times at higher ambient temperatures can be explained by the improved diffusivity of oxygen at higher gas temperatures. A similar effect has been observed in other oxy-fuel study [21]. The comparison of char burning times shows similar effect like devolatilization time with varying ambient gas temperature. The gap in char burnout time between N_2 and CO_2 widens with decreasing gas temperature. However, char burnout times in both environments follow a decreasing trend in oxygen concentration and the times are shorter at higher ambient temperatures and are attributed to higher heat transfer to the particle and lower heat loss from the particle.

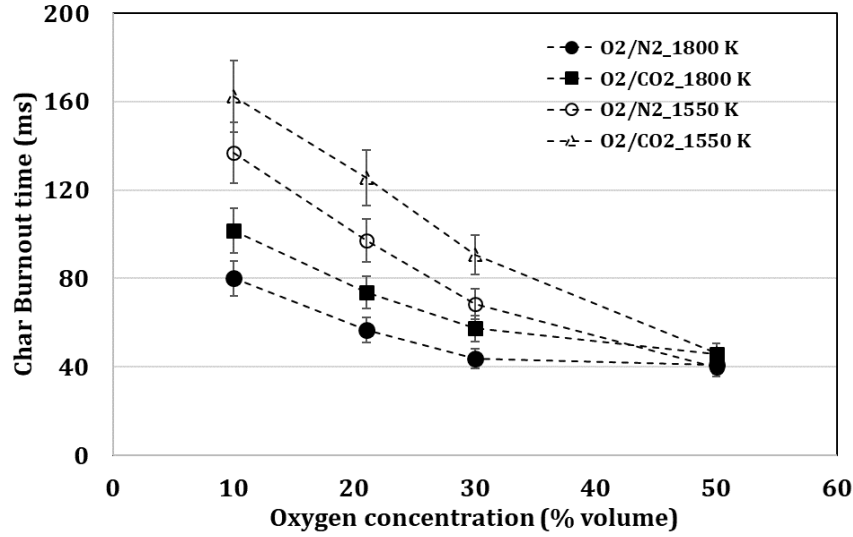


Figure 8. Effect of bulk gas temperature on the char burnout time of sub bituminous (180- 212 μm) in (O_2/N_2) and (O_2/CO_2) environments

4.2.3. Influence of particle size

The experiments were carried out with two particle sizes; 106-125 microns and 180-212 microns. The larger particle size cut has been used to eliminate particle to particle interaction and the cloud effect completely. So, to avoid multiple particles entering the furnace, larger sized cuts were chosen. Figures 9 and 10 show the effect of particle size on volatile and char combustion time of sub bituminous coal particles at 1800 K. As the particle size range is increased, the volatile burning time increases. The difference in the time between the two particle ranges decreases as the oxygen concentration increases. As the particle size is increased, more volatile is evolved from the increased surface area.

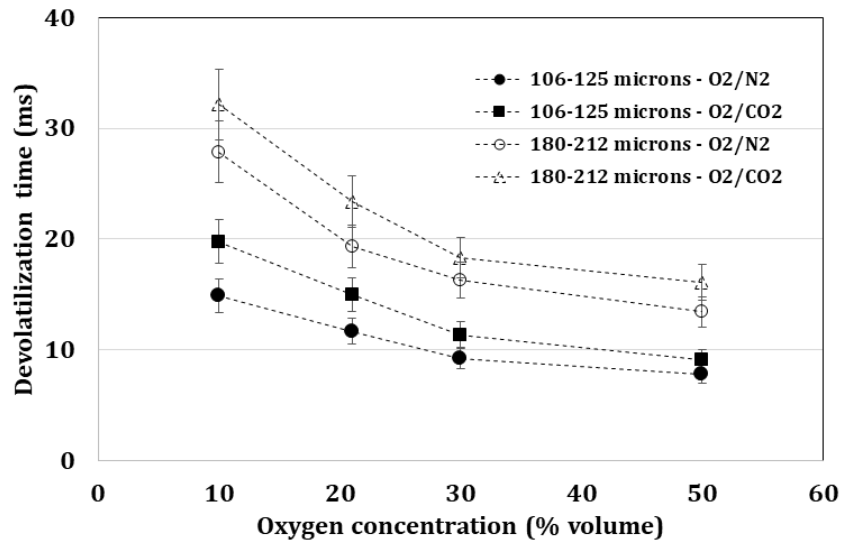


Figure 9. Effect of particle size on the devolatilization time of sub bituminous coal in (O_2/N_2) and (O_2/CO_2) environments at 1800K

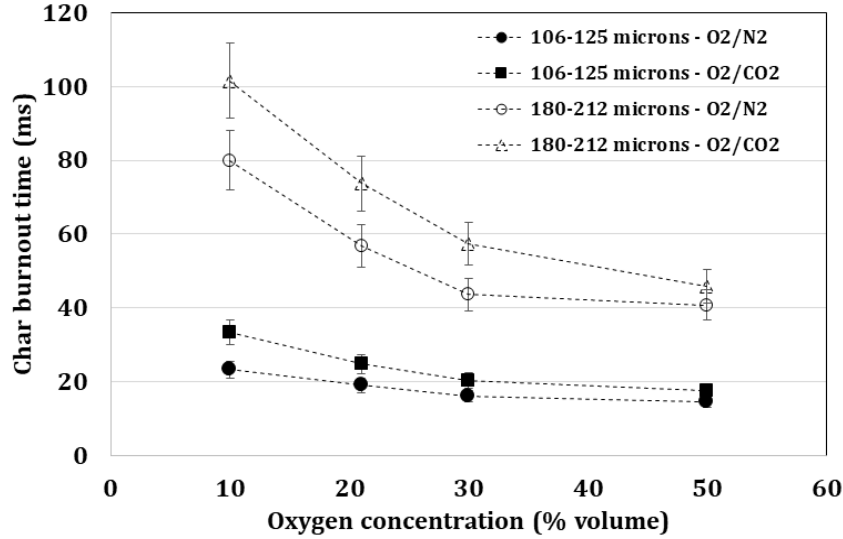


Figure 10. Effect of particle size on the char burnout time of sub bituminous coal in (O₂/N₂) and (O₂/CO₂) environments at 1800K

As the oxygen concentration increases, its diffusion rate towards the particle surface increases, and hence, the volatile burning time decreases [28]. Similarly, as the particle size is increased, the char burnout time is also increased. With an increase in the oxygen concentration, the burnout time decreases. Hence, small particles heat up rapidly and burn more intensely due to higher energy feedback from the flame sheet. Higher particle temperatures in smaller particles due to an increase in surface to volume ratio. This results in an increase in the char burning rate. The trend is similar for lignite coal for the two particle size fractions studied.

5. Conclusion

The combustion behavior of individual bituminous coal particles has been studied in a well-controlled high temperature air (O₂/N₂) and oxy-fuel (O₂/CO₂) environments using an entrained flow reactor using pyrometry. Individual particles of sub-bituminous or lignite coal particle with sizes in the ranges of 106 - 125 microns and 180 - 212 micron particles have been injected into an air (O₂/N₂) or oxy (O₂/CO₂) environment inside an entrained flow reactor. The oxygen concentration in the ambient gas is varied between 10% to 50% by volume. The volatile and char burnout times were measured from the radiation emission history obtained from the burning particle using a photomultiplier tube and other optics. The experiments were performed at gas temperatures of 1550 K and 1800 K with the oxygen concentrations in the bulk gas ranging from 10 to 50% by volume. The results show that the burning rate increases with an increase in oxygen concentration, bulk gas temperature, and smaller particle size. The particle energy traces indicated longer volatile and char combustion times in an oxy-fuel environment than in air, for the same oxygen concentrations. The results indicate that the ignition, combustion, and burning rate of coal particles are significantly influenced by the combustion atmosphere. The measured delay in the volatile and char combustion times under oxy-fuel conditions is attributed to the higher specific heat capacity of CO₂ and lower mass diffusive flux of O₂ in CO₂ mixtures. The experimental results showed that the combustion times in air can be matched by increasing the oxygen concentration to around 30% in the presence of CO₂. The operating range and

the sensitivity of the measuring instruments is found to affect the measured burning times. It has been addressed while validating the theoretical data obtained which is submitted as a separate paper.

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