



A PARAMETRIC INVESTIGATION ON THE TRANSIENT IGNITION OF COAL PARTICLES

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***Abstract.** The transient ignition of single coal particles is studied by modeling. The stability of the flames in large scale boilers is greatly influenced by the fuel ignition characteristics. The ignition model may be homogeneous or heterogeneous depending on particle size and other parameters. In this paper we present a parametric investigation on coal ignition in a variety of particle sizes and two gas temperatures. The results are then compared to some previous work as well as to some experimental results available in the literature of the field. At high heating rates, primary ignition was always heterogeneous for particle size in the range of 10 to 600 microns, for both, lignite and bituminous coal. A transition of the ignition phase was observed only for low heating rates. Models that assume the coal particle temperature as spatially uniform overestimate devolatilization rates, thereby favoring gas phase ignition for large particles. Our predictions have shown that even large particles ignite at the surface if the temperature distribution inside the particle is considered.*

***Key-words:** Coal ignition, Pulverized solid combustion*

1. INTRODUCTION

Stabilization of pulverized solid fuel flames is strongly dependent on particle ignition and extinction. Ignition is an unsteady state combustion phenomenon, which characterizes the transition from slow to fast oxidation followed by a large heat release rate.

Ignition of coal is a great concern since the last century when Faraday and Lyell performed studies in coal-dust flammability (Essenhigh *et al.*, 1989). Since then, the consensus was that ignition takes place in the gas phase. That is, after the coal has released

enough volatile matter to form a flammable mixture around the particle. In the paper of Annamalai and Durbetaki (1977), it is shown theoretically that there is a clear transition of the ignition phase in the burning of coal particles. The motivation for the study seemed to be from the experimental observations of Howard and Essenhigh (1967, 1967a) that small bituminous coal particle ignites most probably at the surface. Annamalai and Durbetaki (1977) showed that a seam bituminous particle which size is below 350 μm might not undergo gas phase ignition.

A comparative study of the ignition behavior of wood, peat and coal was presented by McLean (1982). More recently, Couturier *et al.* (1996) have discussed the ignition and flame stability of pulverized wood flames. Solomon and co-workers (1990) performed experiments in coal combustion. Photographs of several ignited coal particles were carefully observed. They concluded that the injected particles first ignite and appear to burn at the surface of the particle, indicated by the fact that the width of the luminosity is roughly that of the particle diameter. The second ignition phenomenon takes place after the particles have burned for a short time. The calculated dimension of this second luminous zone is about three times the original dimension. This implies a radiant gas flame (CO oxidation) at about two particle radii far from the surface of the particle. More than 80% of Rosebud sub-bituminous coal particles (53-75 μm diameter) surrounded by a dry ambient airflow (850 $^{\circ}\text{C}$, 1 atm) that exhibited the first ignition has also exhibited the second ignition phenomenon.

Fuertes *et al.* (1993) use captive technique for direct measurement of ignition temperatures of coal and char particles. The measured temperatures were in the range from 685 $^{\circ}\text{C}$, for a high-volatile bituminous coal, to 1090 $^{\circ}\text{C}$ for the metallurgical coke. More recently, ignition of coal, char and carbon particles has been also under investigation (Fu and Zhang, 1992, Lee *et al.*, 1995).

Du and Annamalai (1994) presented a transient model for ignition of coal particle. The model was continuous and the kinetics of oxidation of volatiles was that of the oxidation of methane. Coal particle temperatures were time dependent but spatially uniform. For devolatilization, they used the kinetic parameters of two competing reactions given by Ubhayakar *et al.* (1976). They claimed that the transition of ignition phase (TIP), from heterogeneous to homogenous ignition mode occur at particle size of about 400 microns. Particles smaller than the size for TIP ignite heterogeneously at first, while those larger than that experiment volatiles gas phase ignition. Their theoretical investigations concerned a gas at 1500 K containing 23% O_2 (mass fraction).

In this work, we present a transient ignition theory, which resembles that on the paper of Du and Annamalai (1994). Notwithstanding, many of the simplifying assumptions on their work were relaxed here as to give a better insight in such a complex phenomenon.

2. SOLID FUEL IGNITION

In solid fuel combustion two different burning modes exist, gas phase reactions and heterogeneous solid/gas reactions at the particle surface and in its internal porous structure; the first as a consequence of gas phase ignition, of the chain-radical type, while the second due to a solid matrix ignition, of the heterogeneous adsorption/desorption type. Which primary mechanism prevails at a particular case is a function of many interacting factors.

Recent studies have shown that large particles heated slowly ignite homogeneously,

while small particle heated rapidly ignite heterogeneously. Large particles have a smaller outer-surface/volume ratio compared to small ones. Consequently, the heat-loss/production ratio reduces and the heterogeneous ignition temperature decreases.

Direct application of the classical Semenov's thermal theory of ignition to solid fuel particles is not possible without some considerations. Particle ignition would be defined when the heat losses or its rate from the particle equals the heat generation or its rate. In other words ignition occurs when $dT_p/dt = 0$ where T_p is the temperature of the particle and t is time. In a real situation, however, this condition will not be attained if the ambient gas temperature is higher than the minimum ignition temperature but the particle will certainly ignite at some point.

This problem must then be overcome. By manipulating the particle's heat balance equation it is possible to get a more general ignition condition. This general criterion states that $dQ/dT = 0$ and $Q \geq 0$ at the time of ignition, where Q is the net heat supply rate to the particle from all possible sources. We are primarily concerned with ignition of solid particles in real combustors. Thus, we are discharging the condition for which the ambient gas temperature is lower than the critical ignition temperature for the solid fuel particle.

Du and Annamalai (1994) have explored the ignition criterion mentioned above. They applied the inflection condition $dQ/dT_p = 0$ to the heterogeneous ignition for the transient model yielding a critical particle mass loss rate expression. When actual particle mass loss rate is greater than the mass loss rate at the inflection condition, the heterogeneous ignition (HIT) will occur at a specific particle temperature. In pulverized flames, ambient gas temperature is thought to be equal or higher than the minimum temperature for ignition. Therefore, we assume in this work that heterogeneous ignition takes place when $d^2T_s/dt^2 = 0$, where T_s is the temperature of the surface of the particle.

The criterion for homogeneous ignition used by Du and Annamalai (1994) is rather simple. The time any control volume has increased the temperature higher than the neighbors characterizes gas phase ignition (GIT). Since our model is also continuous, we are able, thus, to adopt the same approach. As previously established, heterogeneous ignition (HIT) takes place when d^2T_s/dt^2 equals zero. This is slightly different compared to that employed by Du and Annamalai (1994) because their model has the simplified assumption of uniform particle temperature. Our model is able to predict the temperature distribution inside the particle thereby allowing the transfer of heat to the center of the particle to be included in the surface ignition heat balance where the reactions are, in fact, taking place.

3. NUMERICAL MODEL

In this work, we used the CMVC model ("Continuous Model for Volatiles Combustion") to analyze the transient ignition mode. The model was primarily used to investigate the burning of coal and char particles under pressurized conditions (Gurgel Veras *et al.*, 1999). It was further improved to allow the analysis of NO formation in the devolatilization phase of a burning coal particle (Gurgel Veras *et al.* 1998). More recently, the CMVC model was extended to analyze pulverized biomass combustion. Here, a description of the main characteristics of the CMVC model is provided.

The model was designed to calculate the dynamic chemically reacting flow around an isolated solid fuel particle in one-dimensional geometry. In this continuous model, the

width of reaction zone in the gas phase is not infinitesimal, as assumed in the flame sheet model, but rather established by the local concentration of the reactants and the gas temperature.

Particle temperature distribution is possible along with the transient effects. If heating rates are high, there will be a temperature gradient inside the solid matrix. Devolatilization rates predicted by models that assume coal and biomass particle temperature as spatially uniform far exceed those that take into account the distribution of the temperature in the solid matrix.

Specific heat and thermal conductivity for coal and biomass are function of temperature and the local level of thermal degradation. Char is modeled as carbon with the specific heat calculated with the parameters given by the NASA SP-273 computer code (Gordon and McBride, 1971).

The rates of pyrolysis are calculated by two overall competing reactions (first order). Kinetic constants are taken from the models presented by Kobayashi *et al.* (1976) and Ubhayakar *et al.* (1976). When the ambient gas temperature is low, the constants of the Kobayashi and Gat's models (Gat *et al.* 1983) give lower devolatilization rates compared to the model of Ubhayakar as pointed by Gururajan *et al.* (1988). Gat's parameters predict burnout fairly well but ignition time was unduly long. On the other hand, the constants of Ubhayakar predicted an early ignition time and very short devolatilization period, compared to the experimental results. By virtue of such discrepancies, it is important to investigate the influence of the devolatilization constants to the transient particle ignition mode.

As regard to the composition of volatiles, we assumed it to be a mixture of tar, light hydrocarbons, CO₂, CO, H₂O, H₂, O₂, NH₃ and HCN. For simplicity, tar was assumed to be C₆H₆ and light hydrocarbons CH₄, solely. The composition of the volatiles released from coal was taken from Suuberg *et al.* (1978). At the particle's surface, oxidation of char by oxygen may produce CO or CO₂. The correlation of Monson *et al.* (1995) was used for the ratio CO/CO₂ formed on the surface by the reaction of O₂ with carbon. This correlation is important because of the mass and heat balance to the particle at low temperatures. In addition, the surface mechanisms for the gasification of char by H₂O and CO₂ were included and the rates given by Hobbs *et al.* (1993).

For the gas phase, local specific heat of a mixture, as well as transport properties are dependent of composition, pressure and temperature. The combustion rates of volatiles were calculated with the two step mechanism for hydrocarbon oxidation (Westbrook and Dryer, 1981). Both rates have negative exponents for the concentration of the hydrocarbon. The rates then go to infinity when the concentrations of the hydrocarbons approach zero. We added, in each case, a small reverse rate of reaction. As regarded to dissociation at very high gas temperatures we included a reverse reaction for CO₂, which gives proper heats of reactions for the overall hydrocarbon conversion.

The final gas temperature of rich mixtures is also governed by the concentrations of CO, H₂, CO₂ and H₂O. Therefore, we included the water gas shift reaction along with a reaction for the oxidation of hydrogen. The CO oxidation rate in the boundary layer is dependent on the water vapor concentration (order 0.5). Therefore, CO conversion does not take place if water vapor concentration is zero. We then set the stagnant gas to have 0.1% (mass basis) of water vapor.

4. RESULTS AND DISCUSSION

At first, a high volatile bituminous coal, 30% VM (dry ash free), was chosen for the predictions. Devolatilization rates were modeled by means of single first order and two parallel competing reaction models. Initial particle density was set to 1300 kg/m^3 . Kinetic rate constants for the HVBA (Hobbs *et al.*, 1993) were taken for the oxidation and gasification reactions at the surface of the coal particle. Initially, we set the gas temperature and pressure to be the same as those in the paper of Du and Annamalai (1994), 1500 K, 1 atm, respectively. The oxygen content, however, was reduced from 23% (mass basis) to 16%, for the majority of the cases investigated. The parametric study started by analyzing the effect of particle size on ignition for two different devolatilization models.

Firstly, the kinetic parameters of Ubhayakar for coal devolatilization were used. As pointed by Gururajan *et al.* (1988), their rates of pyrolysis give an early ignition time. Since devolatilization takes place at relatively low temperatures, the mixture around the particle rapidly reaches the flammability limits, thus favoring homogeneous ignition. In the paper of Du and Annamalai (1994) even particles as small as $20 \mu\text{m}$ ignited homogeneously for a certain combination of surface and gas phase kinetics. This is not in accordance with the results presented by Howard and Essenhigh (1967) who concluded that particles less than about 65 microns in size experience simultaneously heterogeneous combustion and gas phase reaction. Particle which are less than 15 microns in size appeared to have significant rate of heterogeneous combustion during rapid volatile evolution. In their investigation, particle size ranged from zero to 200 microns. They concluded that ignition was at the particle surface and soon after significant volatile evolution took place. Therefore, the study of the transient ignition process has to take into account the influence of the kinetic parameters for devolatilization, as well as gas phase reactions.

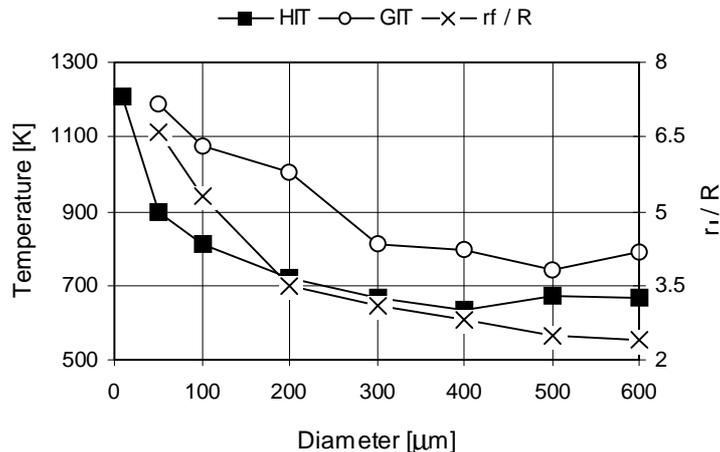


Figure 1: Surface temperature for homogeneous and heterogeneous ignition, and boundary layer location of gas phase ignition with the kinetic parameters of Kobayashi for a bituminous coal for different particle sizes.

Figure 1 presents the particle surface temperature at the instant of both heterogeneous and gas phase ignition. The primary ignition mode is, therefore, that for which the surface

temperature is lower. Figure 1 also shows the boundary layer location of gas phase ignition for different particle sizes (r_f / R). Devolatilization kinetics constants of Kobayashi were used for a gas temperature held at 1500 K with 16% oxygen mass fraction.

Primary heterogeneous ignition prevailed for all the cases analyzed. Secondary homogeneous ignition (higher surface temperature) occurred after surface ignition for particles larger than 50 microns in size. For particles as small as 10 μm , devolatilization rates are too slow to provide a flammable gas mixture in the boundary layer for all the particle's lifetime. In a previous work (Gurgel Veras *et al.* 1999), we have showed that extensive overlap of char combustion and devolatilization occurs for bituminous coal particles smaller than 40 μm (1 bar, 1400 K and 23.3 wt.% O_2). This implies that, in spite of gas temperature or oxygen mass fraction, particle ignition occurs always at the surface, heterogeneously.

. We then extend our analysis using Ubhayakar's pyrolysis constants. Figure 2 shows the same curves as Fig. 1 with the kinetic constants of Ubhayakar with the gas condition unchanged.

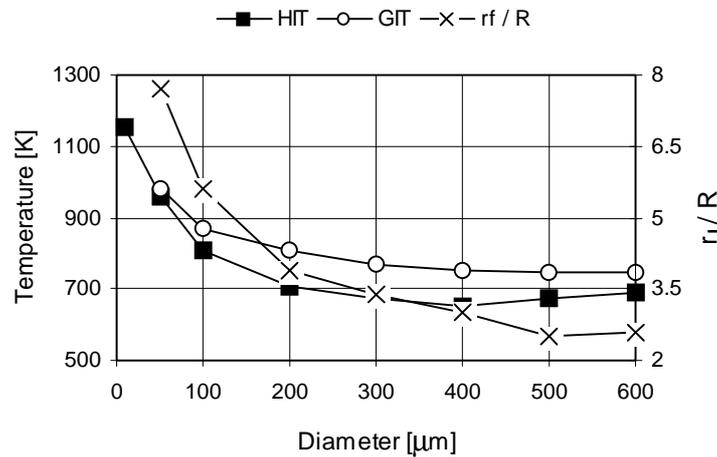


Figure 2: Surface temperature for homogeneous and heterogeneous ignition, and boundary layer location of gas phase ignition with the kinetic parameters of Ubhayakar for a bituminous coal for different particle sizes.

Here, the primary mode of ignition was also heterogeneous for all the cases. Since volatile evolution is much higher than that where Kobayashi's kinetic constants were employed, homogeneous ignition took place shortly after surface ignition. For particle 50 microns in size, ignition was almost simultaneous at the surface and in gas phase. The time delay increases, as particle size is increased until 400 microns, then tending back to simultaneous ignition for particles larger than 500 microns.

As regarded to the transition of ignition phase, our results are neither in accordance with the predictions of the continuous model of Du and Annamalai (1994) nor to that of the flame sheet model of Annamalai and Durbetaki (1977). For bituminous coal, the former found TIP size to be about 500 μm and the latter, 105 μm . However, both models assumed isothermal particles and the global rate expressions for volatiles (CH_4) oxidation was based on one step conversion. In addition, it is not clear what was the heating value of volatiles

they adopted in their predictions. Our model gives preferential release of tar (C_6H_6) at low devolatilization temperatures and the heat value of volatiles were corrected in order to give the overall coal heating value more realistic.

Du and Annamalai (1994) investigated the effect of kinetic parameters on the transient ignition mode. Primary homogeneous ignition was predicted even for small particles when combining fast combustion kinetics for gas with slow surface reactions. On the other hand, ignition was always heterogeneous when using slow volatiles kinetics combined with fast surface reactions. This suggests a strong influence of the kinetic parameters chosen, as well as some of their model assumptions. The CMVC model, however, calculates the temperature distribution inside the particle. The local rate of production of volatiles is a function of the temperature history of that control volume. Therefore, the volumetric rate of volatile evolution from our predictions is much less than where it is assumed uniform coal particle temperature. In addition to this, coal heat capacity is not constant but increases as the local temperature increases, thus retarding the heating of the interior of the particle.

One important characteristic of the transient model of Du and Annamalai (1994) is related to the numerical solution when comparing to the present model. Their domain outside the particle was divided in 10 shells only, while in this work 100 control volumes were employed. The criterion of gas phase ignition was such that the temperature distribution in the boundary layer must show a peak. The gas phase reaction zone, at the instant of ignition is much thinner than the size of their shells; consequently, more volatiles are needed to form a flammable mixture for the onset of fast oxidation.

The global mechanisms for volatile combustion in the CMVC model differs from that employed by Du and Annamalai (1994). The two-step mechanism used here is more realistic for gas phase ignition than one-step models as pointed by Sloane and Ronney (1992). They criticized the use of one-step model for hydrocarbon oxidation in predicting minimum ignition energy. The reason is, predominantly, due to the fact that one-step models were tailored to reproduce the integrated heat release in the flame front, as to enable the model to reproduce burning velocity. The conclusions were supported by calculations, which showed that the homogeneous induction times for one-step model and detailed chemical models are drastically different, although all the models reproduce well steady planar burning velocity. The kinetic constants of the two-step model, used here, were fitted as to give better agreement with detailed mechanism in a broad range of equivalence ratios (Westbrook and Dryer, 1981). Therefore, we may expect that the ambient factors for gas phase ignition, as predicted from our model, are different compared to those from the continuous model presented by Du and Annamalai (1994). Neglecting all of these observations greatly favors homogeneous ignition for larger particles.

In the review of Essenhigh *et al.* (1989), particles smaller than 650 μm are reported to have no volatiles flame but instead ignition was observed to take place only at the surface of the particle. They also mentioned that the ignition was of the char, not of the whole coal. Ignition occurred only after a clear pyrolysis pulse had been released. This experimental observation is very difficult to simulate in the model. The surface reactivity we are employing is for the char derived from the specific coal. Therefore, it is indirectly assumed by the model that the outer surface has released all the volatiles although it might not be the case. However, the small amount of volatiles released before heterogeneous ignition greatly contributes for the surface ignition since the heat released in the gas phase increases the gas temperature near the particle. Figure 3 shows that for particles smaller than 100 microns, heterogeneous ignition is immediately followed by gas phase ignition. These results agree

with the observations discussed in the review of Essenhigh *et al.* (1989) that, at high temperature, the heterogeneous ignition may take place simultaneously with pyrolysis.

Although the mixture around the particle would favor gas phase ignition, the surface of the particle acts as a cold wall, thus retarding homogeneous ignition. Figure 3 shows the boundary layer distribution of temperature and oxygen for two particle sizes prior to gas phase ignition. Dashed line illustrates the profile after possible homogeneous ignition.

For the 50 μm particle, oxygen at the surface is almost the same as in the bulk. This would imply internal reaction to some depth of penetration. Diffusion of heat is faster thus smoothing the temperature decay from about 5 particle radii to the surface. For larger particles (400 μm), it can be seen a steep gradient of temperature near the surface. Oxygen mass fraction presents also an intense decay towards particle's surface. This decay is due to the high volatile evolution from the particle. In spite of the fact that this mixture of combustible gases and oxygen is more favorable to ignite, heat losses to the particle are too high to permit homogeneous ignition. As regarded to temperature, gas phase ignition is facilitated far from the particle, where temperatures are high. On the other hand, concentration ratios (Oxidizer/Fuel), which favor ignition, are located near to the particle's surface, where, conversely, gas temperatures are low. As long as the mixture is flammable, reaction rates are faster where temperatures are higher. Therefore, gas phase ignition will take place at some distance from the particle.

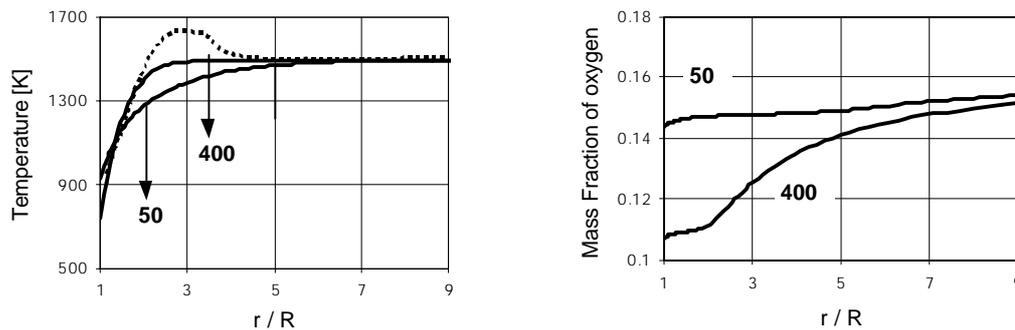


Figure 3: Boundary layer profiles of temperature and oxygen for two particle sizes versus normalized radial distance. Dashed line means the profile after homogeneous ignition

The location of gas phase ignition is shown in Figs. 1 and 2. As particle size increases, homogeneous ignition takes place nearer to the particle. HIT for particles larger than 400 microns increases slightly since the outer parts of such particles are losing more heat to the colder center thus retarding heterogeneous ignition.

Primary ignition was always heterogeneous when making use of Gat's rate constants. Similar to the Kobayashi's model, Gat's give low rates at modest pyrolysis temperatures thus favoring heterogeneous ignition, at least for particles in the range of 10 to 600 μm examined here.

Homogeneous ignition is thought to occur when large particles are heated slowly. We then reduced the gas temperature from 1500 to 1200 K with oxygen mass fraction fixed at 16%.

Primary ignition was homogeneous for particles smaller than 600 microns, exception for the 10 micron particle. For the 400 μm particle, however, ignition was almost simultaneous, prevailing slightly at the gas phase. A further increase in particle size shows a transition of the ignition phase (TIP). The 600 microns particle ignited heterogeneously first. This transition maybe explained by a combination of many factors. At low heating rates, devolatilization starts earlier thus favoring the onset of a flammable mixture near the particle. Therefore, gas phase ignition takes place primarily for particles as large as 400 microns. As the size of the particle increases, heterogeneous ignition temperature decreases, independently of the heating rate. In the case of particle size as large as 600 microns, the flammable mixture is more favorable to ignite very near to the surface but the particle is at lower temperature thereby acting as a cold wall. The heat losses from the gas phase reaction zone to the particle retards homogeneous ignition. The particle then ignites at the surface first. Gas phase ignition follows immediately.

5. CONCLUSIONS

The CMVC model showed results that are not in full agreement with previously published models. Temperature distribution inside the particle was the main reason for the discrepancies. In addition, the usage of a two step global mechanisms for volatiles oxidation and a correction for the heat value of such devolatilization products are deemed more appropriate for this analysis. Most of the predictions indicated that the primary ignition mode was heterogeneous. Only at low heating rates, gas phase ignition was observed to take place firstly.

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