MATHEMATICAL SIMULATION OF THE INFLUENCE OF VOLATILE MATTER CLOUD ON HETEROGENEOUS IGNITION OF SINGLE COAL PARTICLES

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A simple model has been developed to predict the heterogeneous ignition temperature for single coal particles under conditions of negligible, natural and forced convections. These conditions cause different sizes of volatile matter cloud that surround the coal particle. The model is based on the ignition of a single coal particle heated by irradiation from spot heaters in a cold (ambient) surrounding. Energy from the particle surface oxidation, volatile matter combustion and physical heating from the spot heaters are all included in the particle energy balance. General ignition criterion is used to determine the ignition point. The agreement between the experimental and predicted results is fairly good as far as the effects of particle size and volatile matter are concerned. The gas temperature emerged as the most effective model parameter influencing the ignition temperature. In the absence of volatiles in the particle vicinity, as for the forced convection case, the gas temperature remained almost constant and so did the ignition temperature. For the cases where volatiles surround the particle, high gas temperatures as well as ignition temperatures were obtained. This is mainly attributed to the combustion of the volatiles contributing to the gas temperature rise and possibly raising the particle temperature.

Keywords: *Coal ignition; volatile cloud; ignition mechanism; convection*

INTRODUCTION

Mathematical modeling of heterogeneous ignition (i.e. ignition by direct attack of oxygen on the surface of the particle) has been undertaken quite extensively by a number of researchers. Results from most of these models have shown a decreasing ignition temperature with increasing particle size. On the other hand, the effect of volatile matter on the heterogeneous ignition temperature and ignition mechanism has received less attention. In their transient model, Du and Annamalai (1994), presented the effect of a number of parameters on ignition temperature of single coal particles and showed a weak dependence of the heterogeneous ignition temperature on volatile matter. Zhang and Wall (1993) reported that the effect of volatile matter on the heterogeneous ignition temperature is reduced when the oxygen concentration is increased. At low oxygen concentrations, the contribution of volatile matter combustion to ignition is significant and the ignition seems to be in the gas phase (homogeneous). They did not, however, specify the limit of oxygen concentration above which the contribution of volatile matter ceases to be significant.

Recent work on the effect of particle size on ignition temperature found the trend to be opposite of what have been generally reported in literature (Katalambula et al., 1996). However, the findings were mainly due to the fact that the heating rate was maintained almost constant, something which seemed to be missing in most of the other cases. In addition, not a single mathematical model has been made to describe these new finding, hence prompting the need to develop one for the purpose.

Since ignition is due to the interaction between chemical and physical processes occurring in a system, the onset of ignition must be defined to include not only the chemical processes but the physical ones as well (Phuoc et al., 1993). A number of models have considered only the heat generated by the char surface oxidation and did not consider the effect of volatiles combustion on the heterogeneous ignition. Generally, during the volatile (gas phase) combustion, heat from the volatile flame is transferred to the particle by conduction, thus contributing to heating up the particle (Gururajan et al., 1990).

As far as the model assumptions are concerned, many authors have assumed a symmetrical distribution of volatiles around the particle and no relative motion between the particle and the gas (Du and Annamalai, 1994; Phuoc et al., 1993; Yang and Wang, 1990; Gururajan et al., 1990). Some have neglected the effect of radiation and natural convection (Zhang and Wall, 1993; Phuoc et al., 1993). While it is a common practice to draw some assumptions in order to simplify the development of the model, it is sometimes better to simplify the process/phenomenon itself so that the assumed condition is achieved. For example, for an experiment done under normal gravity conditions, no symmetry of the released volatiles can be achieved since natural convection sweeps away some of the volatiles. In so doing, the amount of volatiles taking part in the combustion reaction is affected and consequently affecting the final results. If the experiment is performed under microgravity condition where natural convection is negligible, a good symmetry of released volatiles will be achieved and a better agreement between experimental and theoretical results can be realized.

As for the determination of the ignition point, some models have applied the char weight loss as an indicator (Du and Annamalai, 1994; Yang and Wang, 1990), while some have employed Semenov's thermal explosion theory (Zhang and Wall, 1993; Wall and Gururajan, 1986; Chen, 1996; Chen et al., 1994; Krishna and Berlad, 1980). Others defined the ignition point as a transition from a low temperature or kinetically controlled combustion regime to a high temperature and diffusion controlled combustion regime (Gururajan et al., 1990). Phuoc et al., (1993) used the ratio between chemical energy and laser energy as an indicator of the ignition. Since the experiments on which this model is based (Katalambula et al., 1996, 1997a, 1997b) determined the ignition temperature by a sudden jump on the particle temperature history, the theoretical part will also be based on the same principle employing the general ignition criterion.

Given the above few shortcomings, this work aims at developing a model which will address the effect of volatile matter as well as particle size on the ignition mechanism and ignition temperature. The model will also demonstrate how convection plays part in determining the ignition mechanism.

MODEL FORMULATION

Considered is a problem of a single coal particle ignition in cold (ambient) surroundings. It is initially heated by infrared radiation from spot heaters which raises the particle temperature. Heat generated by volatile's combustion heats up the particle. The surrounding gas is then heated through the particle-gas heat transfer. The particle can also directly exchange heat with the surrounding gas by radiation. To change the size of the volatile matter cloud, three conditions are employed, namely negligible, natural and forced convections (Katalambula et al., 1997b). Under negligible convection, all the volatiles released stay in the particle vicinity thus giving the maximum volatile cloud size, while under natural convection, only a portion of the released volatiles remain in the particle vicinity, the rest are swept away. Under forced convection, all the released volatiles are swept away from the particle as soon as they are released, therefore there are no volatiles left in the particle vicinity.

Assumptions

The assumptions put forward in the development of the present model are:

1) Heterogeneous ignition takes place soon after volatile combustion is completed.
2) The particle is dry, ash-free and spherical in shape.

3) Negligible temperature gradient exists within the particle.
4) The char oxidation is expressed as a first order irreversible reaction with respect to oxygen concentration and evaluated at the external surface of the particle. The product of oxidation is CO_2.
5) The heat of devolatilization is neglected as it is small compared to the heat of the surface oxidation and volatile matter combustion.
6) Boundary layer diffusion is neglected because the layer is not established until an appreciable reaction is in progress, i.e. until after the ignition

Governing equations

Heat balance for a coal particle subjected to an external heat supply is given by

$$m_p c_p \left[\frac{dT_p}{dt}\right] = Q = Q_{gain} - Q_{loss} \tag{1}$$

The heat gained by a particle, Q_{gain}, is made up of three sources, namely:

a) Heat due to physical heating, Q_p, which could be from hot gases in a hot furnace, laser beam, electrical heating coil or spot heaters as in the present case.

b) Heat due to volatile combustion, Q_v, originating from the combustion of volatiles taking place in the vicinity of the particle, also referred to as heat feedback to the particle (Masami and Okazaki, 1988) and

c) Heat due to char oxidation, Q_c.

So the total heat gained can be written as

$$Q_{gain} = Q_p + Q_v + Q_c \tag{2}$$

The physical heat from the spot heaters, Q_p, is calculated based on the light concentration principle. The spot heaters' light concentrator has a truncated cone shape shown in Figure 1 from which its area can be calculated. Knowing the focal area (manufacturer specified) gives the energy concentration factor.

The spectral absorption efficiency, a_s, is estimated from the Mie theory and in this case is the same as that used by Zhang et al., (1994). Electrical to light to heat energy conversion factor, e_{cf}, is taken as 30% while n_s is the number of spot heaters and p_{pa} is the power delivered to the particle per unit particle surface area. So, the physical heat can be given by the following equation:

$$Q_p = e_{cf} a_s n_s p_{pa} \tag{3}$$

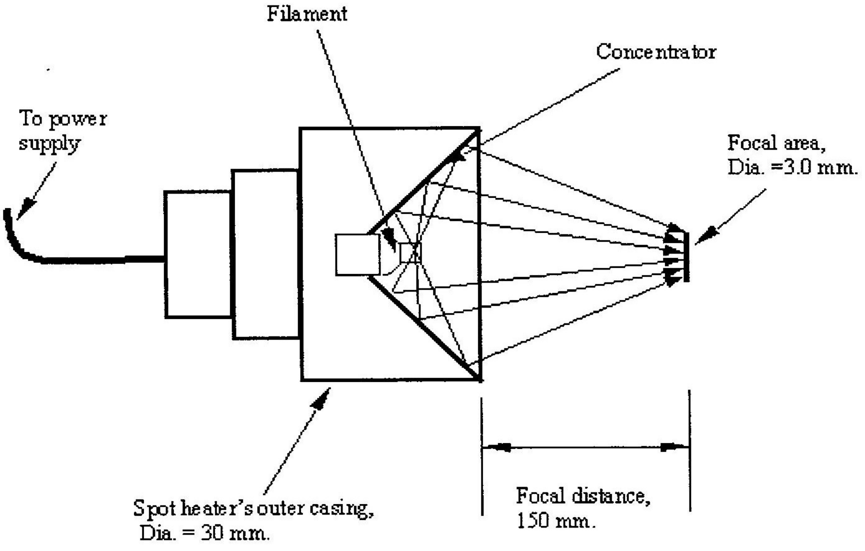

Figure 1: Cross-section of spot heater showing shape of the light concentrator

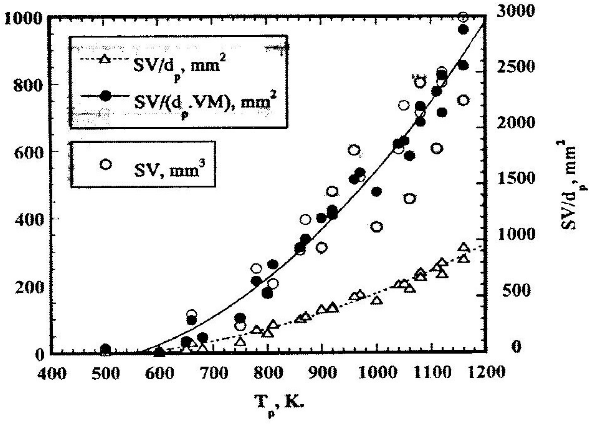

Figure 2: Cross-section of spot heater showing shape of the light concentrator

The heat due to volatile combustion, Q_V, can be calculated by

$$Q_v = H_v v_{cf} \frac{d(m_{vr})}{dt} \tag{4}$$

where v_{cf} is the volatile concentration factor and the rate of volatile release is given by

$$\frac{d(m_{vr})}{dt} = A_v\left(m_{vt} - m_v(T_p)\right)\exp\left(\frac{-E_v}{RT_p}\right) \tag{5}$$

m_{vt} is the total mass of volatiles contained in a coal particle and is obtained by the product of particle weight and volatile matter content. $m_V(T_p)$ is the amount of volatiles released until the particle reaches a temperature T_p and is estimated by equation 6. The parameter $m_v(T_p)$ is important in estimating the time dependent changes in the mass of char or volatiles which were not measured experimentally. This is achieved by making use of the time dependent changes in the size of the volatile cloud by assuming it to behave as a one component gas made of CH_4.

$$m_v(T_p) = v(T_p) d_p w_{VM} \rho_v \tag{6}$$

$v(T_p)$ is the correlation for volatile volume as a function of T_p obtained from the experimental results reported previously (Katalambula et al., 1997a). The observed volatile volumes are first converted to standard temperature and pressure (STP) to give the standard volume, SV. The standard volume is then divided by the particle size, SV/d_p and then by volatile matter content, SV/(d_p.w_{VM}). The obtained results are shown in Figure 2. This correlation is given in Equation7, representing the volume of volatiles released from a particle of unit size and unit volatile matter content:

$$v(T_p) = 576 - 3.65T_p + 0.0047{T_p}^2 \tag{7}$$

Heat due to char oxidation, Q_C, is given by

$$Q_C = S_p H_c {Y_{O_2}}^N \left.\frac{dC}{dt}\right|_{T_p} \tag{8}$$

where

$$\left.\frac{dC}{dt}\right|_{T_p} = A_c\left(\frac{m_{ct} - m_c(T_p)}{m_{ct}}\right)\exp\left(\frac{-E_c}{RT_p}\right) \tag{9}$$

and m_{ct} is the total mass of char given by

$$m_{ct} = m_{coal} - m_v\left(T_p\right) \tag{10}$$

The heat loss sources include convection and radiation which are written as

$$Q_{loss} = \frac{Nu\lambda}{d_p}\left(T_p - T_g\right) + \varepsilon\sigma\left({T_p}^4 - {T_g}^4\right) \tag{11 a}$$

where Nu is the Nusselt number given by:

$$Nu = 2.0 + 0.6\left(\frac{d_p u_g \rho_g}{\mu_g}\right)^{1/2}\left(\frac{c\mu}{\lambda}\right)_g^{1/3} \tag{11 b}$$

Assuming a linear increase of the particle temperature with time, the particle temperature can be expressed as a function of time as shown below:

$$T_p = T_{p0} + \frac{dT_p}{dt}t \tag{12}$$

In order to estimate gas temperature, T_g, two heat sources are considered, namely heat due to combustion of volatiles taking place around the particle, Q_V, and that due to the radiation from the particle to the volatiles Q_{lr}. Substitution of T_p in the respective equation gives Q_V as a function of time, t, and Q_{lr} as a function of both t and T_g. Thus, the heat balance for the volatiles yields.

Table 1: Model Parameters and Values

Parameter	Description	Value	Units
A_c	Pre-exponential factor for char oxidation	$5.41\cdot10^3$	kg/m^{2}s
A_v	Pre-exponential factor for devolatilization	$6.74\cdot10^7$	s^{-1}
E_c	Activation energy for char oxidation	150.0	MJ/Kmol
E_v	Activation energy of devolatilization	113.0	MJ/Kmol
Hc	Heat of reaction for char oxidation	32790	kJ/kg
H_v	Heat of reaction for volatiles (CH_4)	22000	kJ/kg
N	Order of reaction	1.0	-
R	Universal gas constant	$8.314\cdot10^{-3}$	kJ/mol.K
Y_{O2}	O_2 mole fraction at particle surface	0.23	-
□	Emmisivity of coal	0.8	-
□	Stefan-Boltzmann constant	$5.67\cdot10^{-8}$	W/m^{2}K^4
□coal	density of coal particle	$1.2\cdot10^3$	kg/m^3
v_{cf}	volatiles concentration factor	1.0 for microgravity	-
	0.5 for normal gravity	-	
	0.0 for forced convection	-	

$$m_v(t)c_{pv}\frac{dT_g}{dt} = Q_v + Q_{lr} \qquad (13)$$

A numerical solution for the above equation by the Runge Kutta-Gill method gives T_g as a function of t and hence as a function of T_p for a given particle heating rate.

Based on the general ignition criterion (Wong et al,. 1995., Essenhigh et al., 1989), ignition occurs when:

$$Q_{gain} \geq Q_{loss} \qquad (14)$$

Equation1 is then solved for T_p which gives the particle temperature at ignition.

Kinetic parameters

Kinetic parameters were determined stagewise employing different conditions. First stage involved determining the kinetic parameters under forced convection. Under this condition, T_g remained constant since all volatiles were blown away as soon as they were released, hence they did not ignite in the particle's vicinity. The volatile concentration factor, v_{cf}, in this case is zero and the pre-exponential factor A_c was determined while H_c and E_c are obtained from literature (Du and Annamalai, 1994). The determination of the pre-exponential factor was done by varying the value of A_c until a reasonable fit of the curve to the experimental data was obtained. The second stage was under negligible convection. In this case all the volatiles accumulated around the particle, hence $v_{cf} = 1.0$. For volatiles combustion, A_v was then determined and H_v and E_v were obtained from literature (Du and Annamalai, 1994; Phuoc et al., 1993). The third stage involved fitting theoretical to experimental results employing parameters established above. The v_{cf} was adjusted so as to obtain the best fit. The value of

Table 2:Sensitivity Check: Percentage change in ignition temperature due to +/- 10% change in a given parameter.

	Forced Convection		Natural Convection		Negligible Convection	
Parameter	+10%	-10%	+10%	-10%	+10%	-10%
H_v, A_v	0.0	0.0	0.0	-0.1	0.1	0.0
E_v	0.0	0.0	-0.3	0.8	-0.3	0.9
H_c, A_c	0.0	0.0	0.0	0.0	0.0	0.0
E_c	0.0	0.2	-0.1	0.4	-0.2	1.2
Q_p	0.8	-0.7	0.6	-0.7	0.7	-0.5

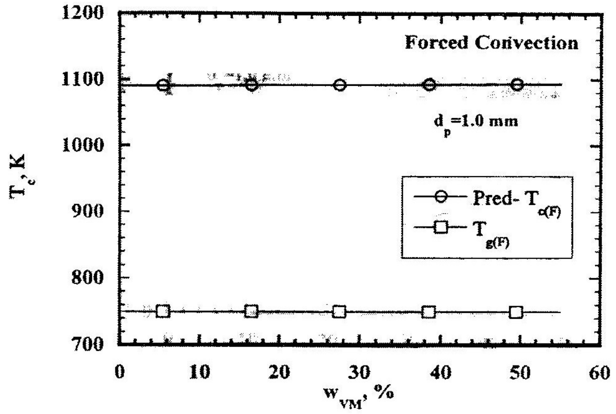

Figure 3: Variation of gas temperature and ignition temperature with volatile matter content under forced convection

v_{cf} indicates the proportion of the released volatiles taking part in the combustion reaction. Other model parameters and their values are listed in Table 1 and a sensitivity check for these parameters is presented in Table 2. It can be seen that with a change of +/- 10% of the value of a given parameter (when all other parameters are held constant), the resulting change in the heterogeneous ignition temperature is negligibly small, implying that small variations in these parameters will not affect the model results.

RESULTS AND DISCUSSION

The experimental results used for validation of this model are previously reported (Katalambula et al., 1996, 1997a, 1997b) and all the ignition temperatures refer to heterogeneous ignition temperatures.

Effect of Volatile Matter Content on Ignition

Under the forced convection, the ignition temperature, $T_{c(F)}$, as well as the gas temperature, T_g, remained unchanged with a change in the volatile matter content, w_{VM}, (coal type) as depicted in Figure 3. This shows that under forced convection, the volatiles have no effect at all since they are all swept away as soon as they are released, and hence they do not have time to burn and cause any temperature change on either the surrounding gases or the particle itself. The gas temperature, T_g, in this case remained constant and about the same as that of the hot air blown to the particle for forced convection. This implies that T_g depends to a large extent on the volatiles combustion.

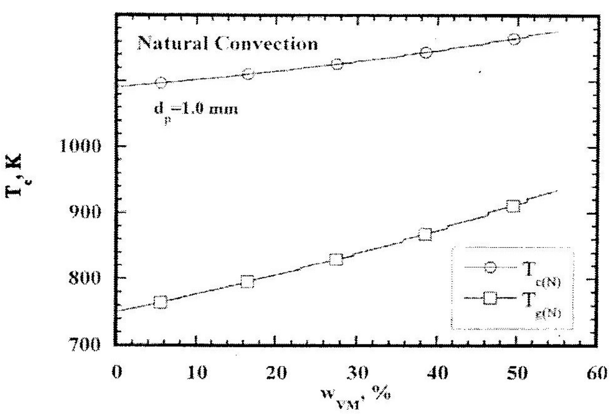

Figure 4: Variation of gas temperature and ignition temperature with volatile matter content under natural convection

Figure 4 shows the variation of T_g and T_c with w_{VM} under the natural convection. In this case, as opposed to the forced convection, it is seen that both T_g and T_c increases with w_{VM}, but T^g increases much faster than T_c. The increase of T_g is attributed to the presence of volatile matter in the particles vicinity, the combustion of these volatiles in the gas phase causes T_g to rise. Similar explanation can be given for the case of the negligible convection shown in Figure 5. Both T_g and T_c increases with increasing w_{VM}. In this case, all the volatiles released stay in the particle vicinity and upon ignition and combustion, they contribute significantly to the

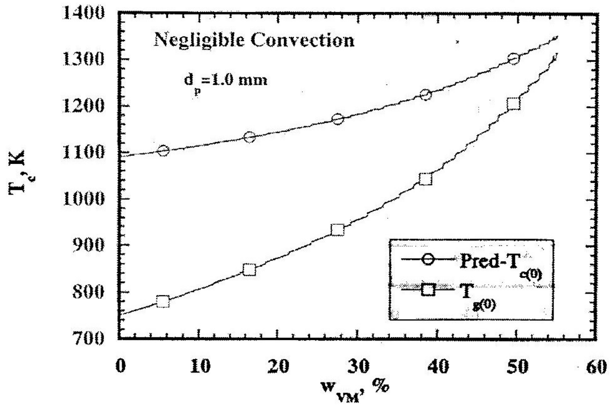

Figure 5: Variation of gas temperature and ignition temperature with volatile matter content under negligible convection

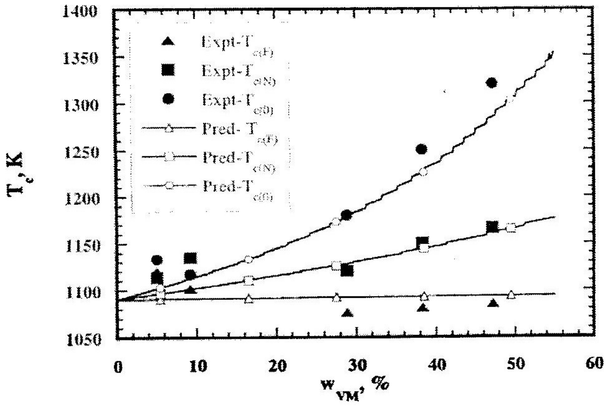

Figure 6: Comparison of experimental and predicted results on the variation of ignition temperature with volatile matter content under negligible, natural and forced convection

rise in T_g. At higher values of w_{VM}, Tg exceeds the particle temperature. At this point, the gas heats up the particle. Masami and Okazaki (1988) have reported that when the evoluting flux of volatile matter from the particle surface in the stage of volatile matter combustion is larger than the equivalent oxygen flux to the particle from the surrounding air, the reaction/flame zone is pushed away from the particle surface. The feedback rate of the heat released in this flame zone to the particle depends on how far the flame zone is away from the particle. Hence, it can generally be said that, as the volatile evolution decreases, the flame zone comes closer and closer and finally touches the particle, hence giving a maximum heat feedback.

Figure 6 puts together the above cases and shows the variation of the heterogeneous ignition temperature, T_c, with volatile matter content, w_{VM}, under the different experimental conditions. Both the experimental and calculated results are presented for comparison. For the natural convection case, the calculated results were fitted to the experimental ones by fixing the value of v_{cf} at 0.4 to 0.5. This indicates that less than a half of the released volatiles are combusted, while the rest are swept away by natural convection. The volatile matter content values used are the ones which were obtained experimentally using the current experimental setup. These values are used because volatile yield increases with heating rates. The standard w_{VM} values and the experimentally obtained ones are presented in Table 3. It can be seen that the model can predict quite well the variation of T_C with w_{VM} under different conditions as exhibited by the good agreement between experimental and model results. The difference in ignition temperatures between different conditions decreases with decreasing w_{VM} of the coal. As far as ignition mechanism is concerned, the model shows that for lower volatile matter contents, the effect of convection becomes insignificant, suggesting a transfer of ignition phase (TIP) at volatile matter contents below 10%.

Table 3: Standard and experimental volatile matter content values

Coal name	Standard VM values, %	Experimental VM values, %
Pennslyvania	4.4	5.1
Mt. Klappan	8.0	9.2
Datong	26.0	28.9
Coal Valley	32.5	38.3
Taiheiyo	40.2	47.2

Effect of particle size

For the case of forced convection, the variation of T_g and $T_{c(F)}$ with particle size, d_p, is presented in Figure 7. As in the case of variation with the volatile matter, T_g remains unchanged when d_p is varied. Generally, when d_p is varied, the absolute

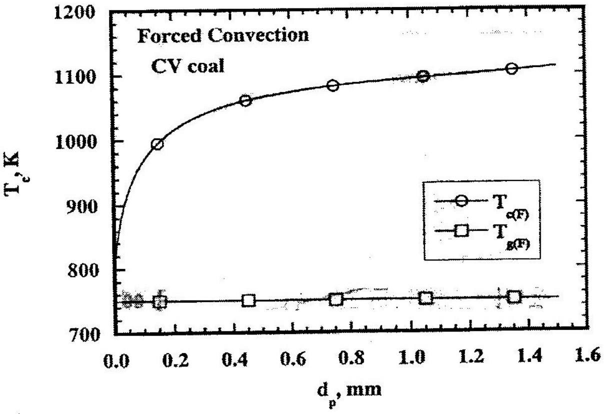

Figure 7: Variation of gas temperature and ignition temperature with particle size under forced convection

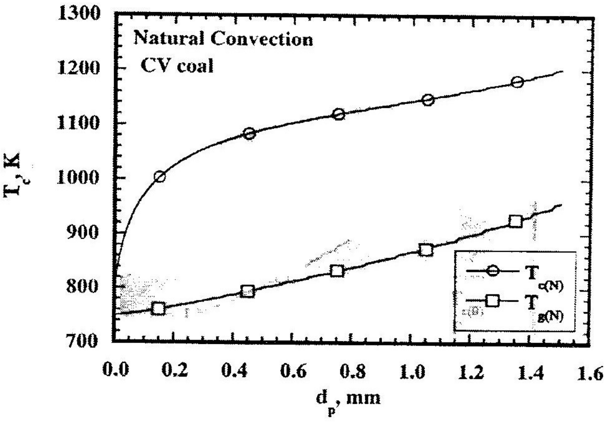

Figure 8: Dependence of gas temperature and ignition temperature on particle size under natural convection

amount of volatiles in the particle vicinity also changes, but this change does not cause any gas temperature increase because all the volatiles are swept away before they ignite. T_c, however, rises quite sharply at first, but it then gradually slows down with increasing d_p. From the exhibited trend, it seems that for d_p above 400μm, an increase in d_p do not have a significant effect on $T_{c(F)}$, although it is not negligible. Figure 8 also shows the variation of T_g and $T_{c(N)}$ with dp. Here, Tg as well as T_c are seen to increase with dp. The increase of Tg could be attributed to the availability of volatiles in the particle vicinity which upon combustion contributes to an increase in T_g. Figure 9 presents the negligible convection case where Tg increases until it exceeds $T_{c(O)}$ at d_p of about 1.4 mm. This is due to the fact that the absolute amount of volatiles increases with particle size. Accumulation of these volatiles around the particle under the negligible convection condition leads to higher T_g after the volatiles ignition and combustion.

The experimental and predicted results for the effect of particle size, d_p, on T_c are shown in Figure 10. The model shows an increasing Tc with increasing d_p under all the three conditions as previously reported (Katalambula et al., 1997a). The agreement between the predicted and experimental results is quite reasonable. On the other hand, the model shows that convection has no effect on small particles (below 200μm), implying a transition of the ignition phase from homogeneous to heterogeneous ignition. Whereas $T_{c(F)}$ and $T_{c(N)}$ seems to increase almost linearly after the initial sharp rise, $T_{c(O)}$ forms a sigmoid shaped curve exhibiting a second sharp rise as dp increases.

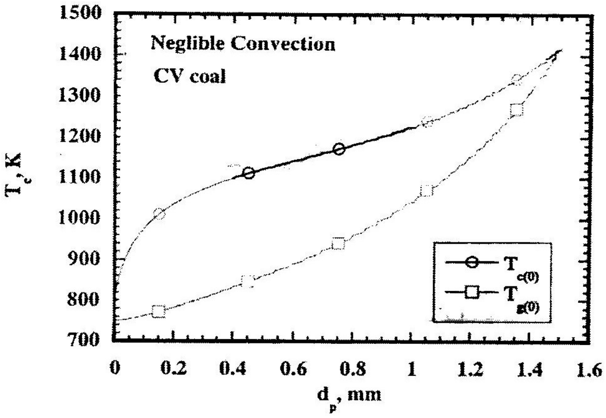

Figure 9: Variation of gas temperature and ignition temperature with particle size under negligible convection

GENERAL OBSERVATION

In the present model, the gas temperature, T_g, has emerged as a very strong parameter in influencing the ignition temperature. In the absence of the volatiles in the particle vicinity, as for the forced convection case, T_g has remained almost constant and so has $T_{c(F)}$. For the cases where the volatiles surrounded the particle, T_g as well as T_c increased

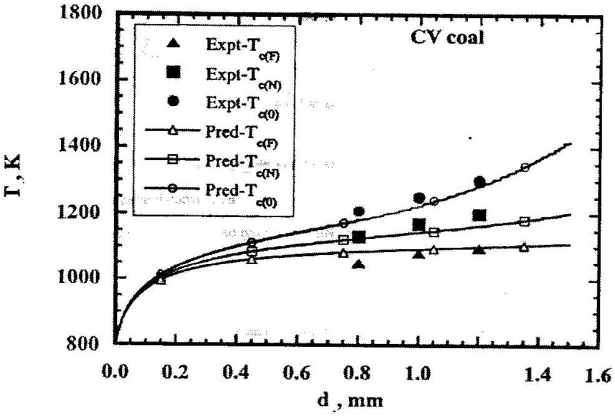

Figure 10: Effect of particle size on ignition temperature under negligible, natural and forced convection: Comparison of experimental and predicted results

simultaneously, mainly being due to the combustion of the volatiles which contributed to the gas temperature rise and possibly raising the particle temperature.

CONCLUSION

A model for predicting the effect of convection on heterogeneous ignition of single coal particles has been developed. The predicted results on the effects of volatile matter, particle size and heating rate on the heterogeneous ignition temperature have shown a good agreement with the experimental ones. Within the limits of the current model, the followings are concluded:

1) The amount of volatiles surrounding a coal particle influences significantly the ignition mechanism and ignition temperature.
2) The gas temperature is a very strong parameter in influencing the ignition temperature in any condition. At the same time, the gas temperature strongly depends on the amount of the volatiles in the particle vicinity.
3) Under forced convection, ignition characteristics are independent of coal type.
4) Under normal gravity, only 40-50% of the released volatiles is combusted, the rest is swept away by natural convection.
5) Both the volatile matter content and the particle size have an appreciable influence on the ignition mechanism.

NOMENCLATURE

A_c	pre-exponential factor for char oxidation [kg·m^{-2}s]
A_v	pre-exponential factor for volatile release [s^{-1}]
A_s	spectral absorption efficiency [-]
c_p	specific heat [J.kg^{-1}K]
d_p	particle diameter [m]
E_c	activation energy for char oxidation [MJ·Kmol^{-1}]
E_v	activation energy of devolatilization MJ·Kmol^{-1}]
e_{cf}	energy conversion factor *[-]*
H_c	heat of reaction for char oxidation [kJ·kg^{-1}]
H_v	heat of reaction for volatiles [kJ·kg^{-1}]
m	mass [kg]
N	order of reaction [-]
n_s	number of spot heaters [-]
P_{pa}	power delivered to the particle [kJ s^{-1}]
Q	net heat supply rate [kJ·s^{-1}]
Q_c	heat due to char oxidation [kJ·s^{-1}]
Q_g	rate of heat generation/gain [kJ·s^{-1}]
Q_l	rate of heat loss [kJ·s^{-1}]
Q_p	heat due to physical heating [kJ·s^{-1}]
Q_v	heat due to volatile combustion [kJ·s^{-1}]
R	universal gas constant [kJ·mol^{-1}K^{-1}]
S_p	particle surface area [m^2]
t	time [s]
t_C	time taken for char ignition [s]
T	temperature [K]
T_c	particle temperature at char ignition [K]
U	gas velocity [m.s^{-1}]
v	volume [m^3]
v_{cf}	volatiles concentration factor [-]
w_{VM}	volatile matter content of coal[wt%]
YO_2	oxygen mole fraction [-]

Greek symbols

ε	emmisivity of coal [-]
λ	thermal conductivity of gas [W·m^{-1}K^{-1}]
ρ	density [kg·m^{-3}]
σ	Stefan-Boltzmann constant [W·m^{-2}K^{-4}]
μ	viscosity [kg.m^{-1}s^{-1}]

Subscripts

c	char
g	gas
p	particle

v volatiles

Others

(O) under microgravity with negligible convection

(N) under normal gravity with natural onvection

(F) under normal gravity with forced convection

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