

AN INTEGRAL MODEL FOR THE PYROLYSIS OF NON-CHARRING MATERIALS

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ABSTRACT

An integral model is presented for the heat-up and pyrolysis phases of the combustion of non-charring thermoplastic materials. The heat transfer within the material is calculated by a one-dimensional transient heat conduction model. For each phase, the original partial differential equations are transformed into a set of two ordinary differential equations by assuming a known temperature profile throughout the solid and using a moment technique. The model is different from previous integral models, since instead of a linear or exponential temperature profile, a quadratic temperature profile is assumed within the solid. The model provides for a specified external radiant heat flux and heat losses, including re-radiation and convection. The accuracy of the integral model is evaluated by comparison of the results with analytical solutions, numerical simulations and experiments. Since the model is simple, and computationally cheap, it can be incorporated into computer fire codes where the available memory and CPU time are always serious limitations. Calculations presented here show that it is ideal for this task.

NOMENCLATURE

c_p	Specific heat of solid [kJ/kg/K]
$E(t)$	Total absorbed energy function [mK]
H	Enthalpy [kJ/kg]
H_v	Latent heat of pyrolysis [kJ/kg]
h	Convective heat transfer coefficient [W/m ² /K]
k	Thermal conductivity [W/m/K]
L	Thickness [m]
\dot{q}_{net}''	Net heat flux [kW/m ²]
t	Time [s]
T	Temperature [K]
T_p	Pyrolysis temperature [K]
T_s	Surface temperature [K]
T_0	Initial temperature [K]
T_∞	Ambient temperature [K]
x	Distance from the front surface [m]
α	Thermal diffusivity, $(k/\rho c_p)$, [m ² /s]
δ	Thermal penetration depth [m]
δ_p	Depth of the pyrolysed material [m]

ε	Emissivity
θ	Temperature rise, $(T - T_0)$, [K]
ρ	Density [kg/m ³]
σ	Stefan-Boltzmann constant [W/m ² /K ⁴]

INTRODUCTION

Non-charring thermoplastic materials like poly-methyl-methacrylate (PMMA) constitute a substantial fraction of the fuel load in many building fires. Thus, an understanding of their behaviour under high temperature burning conditions is very important. In this regard, the pyrolysis process is of particular interest, especially from the fire dynamics point of view, because pyrolysis plays a key role in the ignition, flame spread and burning processes during the early stages of fire growth.

A substantial amount of experimental and theoretical work on the heat-up and pyrolysis of non-charring thermoplastic materials has already been done. The experimental work was extensively reviewed by Khalturinskii [1] and will not be addressed here. The previous theoretical models can generally be classified into three main groups: (a) simple algebraic models, (b) analytical models and (c) numerical models.

In the algebraic models [2] most of the physical and chemical processes are ignored except for the gasification rate which is assumed to be proportional to the net absorbed heat flux once some ignition criterion has been met. On the other hand, analytical models, such as that reported in [3] are more general but their range of applicability is limited to restricted cases, since an analytical solution is, in general, not available.

Numerical models tend to fall into two categories; detailed partial differential equation (PDE) models that attempt to provide a comprehensive description of the heat-up and pyrolysis processes and integral models. The problem with the detailed numerical models is that they are neither simple to use in computer fire codes nor comprehensive enough to include all of the relevant physics and chemistry. Integral models are an attempt to develop simple and practical numerical models by transforming the original partial differential equations to a system of ordinary differential equations (ODE). This is done by assuming a known temperature profile through the solid and integrating the governing equations across the solid thickness. Different temperature profiles have been considered for the integral model approximation, such as a linear temperature profile [4] and an exponential temperature profile [5,6]. In spite of the reasonable accuracy of the previous integral models, they are not mathematically consistent, because the slopes of their assumed temperature profiles do not match both of the heat flux boundary conditions. However, this deficiency can be overcome if a quadratic temperature profile is employed. Therefore, the purpose of this study is to utilise a quadratic temperature profile for developing an integral model of the pyrolysis process of non-charring thermoplastic materials, to include this model in a CFD fire code and to validate the integral model.

PHYSICAL DESCRIPTION OF THE MODEL

The sequence of events occurring in a slab of a non-charring solid object when it is exposed to a fire environment can be divided into: (a) a preheat phase, and (b) a pyrolysis phase. The initial heating phase, which is also called the 'inert heating' stage, is characterised by the constant density heat-up of the material. In this phase a fraction of the heat flux received by the surface is transferred into the interior of the solid by conduction while the rest is re-radiated or convected back to the surrounding. As a result, the surface and interior

temperatures gradually rise with time. This effect is confined to a thin layer near the surface, which is called the 'thermal penetration depth (δ)'. The solid at a depth greater than the thickness of this thermal layer can be considered to be at the initial temperature. Depending on the ratio of (δ/L) the material may behave as a thermally thick solid, a thermally thin solid or a thermally intermediate solid. In this study only the thermally thick ($\delta/L \ll 1$) and the thermally intermediate ($\delta/L \approx 1$) cases are considered.

As heating continues, the surface temperature increases to a high value which can initiate the pyrolysis process. The influence of temperature on the rate of pyrolysis can be represented satisfactorily using a first order Arrhenius type rate equation. However, for many thermoplastic materials the activation energy is so high that the solid may be assumed to pyrolyse abruptly when it reaches a so-called pyrolysis temperature. This assumption has been shown to be a good representation of the pyrolysis process in thermoplastic materials [7]. Once pyrolysis starts the material transforms solely to gases, the front face regresses and the surface temperature remains at the pyrolysis temperature.

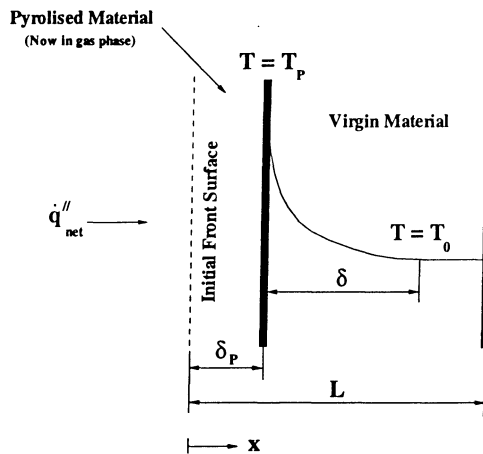


Figure 1: The physical configuration.

Figure 1 illustrates the physical configuration of the present model. An external radiant heat flux is applied at the front surface and the back surface is assumed to be adiabatic. The initial depth of material has been divided into two regions, the region of totally pyrolysed material (burnt material) and the region of unpyrolysed (virgin) material. If an exponential temperature profile ($\theta = \theta_s e^{-x/\delta}$) is assumed, then the adiabatic boundary condition will not be satisfied when the material behaves as a thermally intermediate solid. However, if a quadratic temperature profile is assumed within the thermal layer and the adiabatic boundary condition is applied at $x=\delta$, then even for a thermally intermediate solid there is no mathematical inconsistency. The assumed temperature profile in this study is defined in eq. (1):

$$\frac{T - T_0}{T_s - T_0} = \left[\left(\frac{x}{\delta} \right) - 1 \right]^2 \qquad \frac{T - T_0}{T_p - T_0} = \left[\left(\frac{x - \delta_p}{\delta} \right) - 1 \right]^2$$

(a) Heat-up Phase

(b) Pyrolysis Phase

(1)

THE MATHEMATICAL MODEL

In this section the governing equations and numerical solution procedures for the preheat stage and the pyrolysis phase are presented.

THE INTEGRAL MODEL FOR HEAT-UP

The preheat phase requires a solution to the one dimensional heat conduction equation. If the thermal properties k , ρ and c_p are assumed to be constant (*i.e.* independent of temperature) the heat conduction equation takes the following form:

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} \quad (2)$$

$$\text{subject to the initial condition: } T(x,0) = T_0 \quad (3)$$

and boundary conditions:

$$-k \left. \frac{\partial T}{\partial x} \right|_{x=0} = \dot{q}_{net}'' \quad , \quad -k \left. \frac{\partial T}{\partial x} \right|_{x=\delta} = 0 \quad (4)$$

where an adiabatic boundary condition is assumed at $x = \delta$. The net heat flux absorbed at the front surface can be expressed in terms of the external radiant heat flux, convective and radiative heat losses, *i.e.*

$$\dot{q}_{net}'' = \dot{q}_{ext}'' - \epsilon \sigma T_s^4 - h(T_s - T_\infty) \quad (5)$$

To develop the integral model for the heat-up phase the first two moments of the conduction equation (2) are utilised, as well as the temperature profile specified in equation (1a). If equation (2) is rearranged based on the temperature rise θ , then the first two moments of the heat conduction equation are:

$$\int_0^\delta \frac{\partial \theta}{\partial t} dx = \alpha \int_0^\delta \frac{\partial^2 \theta}{\partial x^2} dx \quad (6)$$

$$\int_0^\delta \theta \frac{\partial \theta}{\partial t} dx = \alpha \int_0^\delta \theta \frac{\partial^2 \theta}{\partial x^2} dx \quad (7)$$

The solution of equations (6) and (7) provides the following equations for the heat-up phase:

$$\frac{d}{dt} (\theta_s \delta) = \frac{3 \dot{q}_{net}''}{\rho c_p} \quad (8)$$

$$\frac{1}{10} \frac{d}{dt} (\theta_s^2 \delta) = \frac{\theta_s \dot{q}_{net}''}{\rho c_p} - \frac{4 \alpha \theta_s^2}{3 \delta} \quad (9)$$

where the quadratic temperature profile in eq. (1a) was utilised to transform the original PDE equations to the above system of ordinary differential equations (ODE) for the surface temperature rise (θ_s) and thermal penetration depth (δ) as a function of time.

THE INTEGRAL MODEL FOR PYROLYSIS

As illustrated in Fig. 1, after pyrolysis starts the front surface recedes at a rate given by $(d\delta_p/dt)$, where δ_p is the depth of pyrolysed material. Also, a rapid rise in the volatile mass flux is observed and re-radiation and convective heat losses become important. In this phase the heat transfer process to the solid is still expressed by eq. (2), but the boundary conditions take a new form:

$$-k \frac{\partial T}{\partial x} \Big|_{x=\delta_p} = \dot{q}_{net}'' - H_v \dot{m}'' \quad , \quad -k \frac{\partial T}{\partial x} \Big|_{x=\delta_p+\delta} = 0 \quad (10)$$

where the mass flux of pyrolysed gases at the front surface is calculated from:

$$\dot{m}'' = \rho \frac{d\delta_p}{dt} \quad (11)$$

Similarly, the integration limits of the first two moments of the heat conduction equation are modified from $(0, \delta)$ to $(\delta_p, \delta_p+\delta)$. If the quadratic temperature profile of eq. (1b) is used and the first two moments of the conduction equation with the modified integration limits are evaluated across the thermal layer, then the resulting equations for the pyrolysis phase are:

$$\frac{d\delta}{dt} + 3 \left(1 + \frac{H_v}{c_p \theta_p} \right) \frac{d\delta_p}{dt} = \frac{3 \dot{q}_{net}''}{\rho c_p \theta_p} \quad (12)$$

$$\frac{d\delta}{dt} + 5 \left(1 + \frac{2H_v}{c_p \theta_p} \right) \frac{d\delta_p}{dt} = \frac{10 \dot{q}_{net}''}{\rho c_p \theta_p} - \frac{40\alpha}{3\delta} \quad (13)$$

Equations (12) and (13) constitute a system of ordinary differential equations for δ and δ_p as a function of time with initial conditions:

$$\delta(t=t_p) = \delta_{heat-up} \quad , \quad \delta_p(t=t_p) = 0 \quad (14)$$

NUMERICAL SOLUTION METHODS

To solve the system of equations for the pyrolysis phase (eqs. (12)-(13)) a standard fourth-order Runge-Kutta method has been used without any difficulty. However, for the heat-up phase this method is not suitable due to the singularity in eq. (9) at $t=0$, where the thermal depth (δ) is zero. To avoid this singularity the method reported in [5] was employed. In this method, new variables δ^2 and $g=(\theta_p/3\delta)$ are introduced and then after some algebra the heat-up equations (8) and (9) become:

$$\delta^2 \frac{dg}{dt} + g \frac{d\delta^2}{dt} = \frac{\dot{q}_{net}''}{\rho c_p} \quad (15)$$

$$\delta^2 \frac{dg}{dt} + \left(\frac{3}{4} \right) g \frac{d\delta^2}{dt} = \left(\frac{5}{3} \right) \frac{\dot{q}_{net}''}{\rho c_p} - \frac{20\alpha g}{3} \quad (16)$$

Next the energy function $E(t)$ is introduced by integrating eq. (8):

$$E(t) = 3 \int_0^t \frac{\dot{q}_{net}''}{\rho c_p} d\tau = \theta_s \delta. \quad (17)$$

Therefore g in eqs. (15) and (16) can be replaced by $E(t)$. Solution of these equations for $(d\delta^2/dt)$ provides the following equation for δ^2 :

$$\frac{d\delta^2}{dt} = \frac{80\alpha}{3} - \left(\frac{\dot{q}_{net}''}{\rho c_p} \right) \frac{8\delta^2}{E(t)}. \quad (18)$$

Equation (18) can be solved easily using a standard integration scheme, such as the implicit trapezoidal method of Adams-Moulton. Once the thermal penetration depth (δ) is obtained, the surface temperature rise (θ_s) can be calculated from eq. (17).

RESULTS AND DISCUSSION

In this section the integral model for the heat-up and pyrolysis is validated against several analytical and numerical solutions, as well as experimental data. The performance of the model when it is included in a CFD computer fire code is also considered.

VALIDATION OF THE HEAT-UP MODEL

In order to validate the integral model for the heat-up phase two series of calculations were carried out for PMMA. The first set of calculations was performed for the case of a constant net heat flux, in which re-radiation and convection heat losses were ignored. The external radiant heat flux was set to 10 kW/m² and the sample thickness was assumed to be 0.015 m. The material properties used in this calculation were: $\rho = 1190$ kg/m³, $c_p = 2.2$ kJ/kg/K and $k=0.23$ W/m/K.

For the constant heat flux case and thermally thick solids, it can be shown [8] that there is an exact analytical solution to the one-dimensional heat conduction equation:

$$\theta_{s,exact} = \sqrt{\frac{4t}{\pi\alpha} \frac{\dot{q}_{net}''}{\rho c_p}} \quad (19)$$

where $\theta_{s,exact}$ is the exact surface temperature rise of the solid from its initial temperature, T_0 . Fig. 2(a) shows the front surface temperature as a function of time, calculated by this analytical method and the integral model, as well as a numerical solution to the heat conduction equation using a control volume approach. As this figure indicates, the integral model produces results that are in good agreement with those obtained from the analytical and numerical solutions. The integral model slightly under-predicts the front surface temperature. This can be explained if the heat-up equations (8) and (9) are solved analytically for the constant net heat flux case. The resulting expression for the surface temperature rise in this case is:

$$\theta_s = \sqrt{\frac{99t}{80\alpha} \left(\frac{\dot{q}_{net}''}{\rho c_p} \right)}. \quad (20)$$

If this equation is divided by eq. (19), then the ratio of the calculated surface temperature rise to the exact surface temperature rise is:

$$\frac{\theta_s}{\theta_{s, exact}} = \sqrt{\frac{99\pi}{320}} = 0.986 \quad (21)$$

Since this ratio is always less than one, the integral model under-predicts the surface temperature. However, equation (21) shows only a 1.4% error in the solution of the surface temperature rise, which is quite acceptable.

The second set of calculations deals with the situation where heat losses were taken into account. In this case an emissivity equal to unity was assumed for the front surface and the convective heat transfer coefficient was set to 13.5 W/m²/K. The material properties used were the same as before.

Fig. 2(b) illustrates the results of the second set of calculations which simulate the analytical results of Steckler [9] and experiments conducted by Kashiwagi [10] on vertical samples of PMMA in an inert atmosphere (no combustion). As this figure shows the general agreement is satisfactory and the integral model follows the same trend as the experimental data and the analytical results. The predicted surface temperature in Fig. 2(b) at a radiant flux of 17 kW/m² is higher than the experimental data mainly due to the fact, that the integral model ignores the in-depth absorption of the radiant energy, whereas in reality some of the energy is actually transmitted and absorbed within the solid. Since higher heat fluxes are associated with shorter time-scales, conduction cannot smooth out the effects of in-depth absorption. Therefore a greater discrepancy between experiment and the integral model prediction is expected for higher heat fluxes. Fig. 2(b) also shows a difference between the analytical results and the integral model predictions. The main reason for this difference is that the analytical model utilises variable thermal properties, whereas the present integral model assumes that all properties are constant. However, as Fig. 2(b) indicates this difference is not significant.

Apart from these two sets of calculations, a number of tests were also carried out to evaluate the performance of the integral model under extreme conditions. In particular the heat-up model was tested for the following cases: a sudden step in the external heat flux, a variable external heat flux and to examine the effect of cooling after a primary heating period. The heat up model performed well in all tests except for the cooling one. However, this is not surprising because under cooling conditions there is no longer a monotonic variation of temperature with depth and the chosen temperature profile in the integral model is not valid [6].

VALIDATION OF THE PYROLYSIS MODEL

The accuracy of the integral model for the pyrolysis phase is investigated in this section by comparing the integral model predictions with the available numerical solutions and experimental data.

Fig. 3 illustrates a comparison between the mass flux of pyrolysed gases calculated using the integral model and a numerical solution to the full partial differential equations for thermal pyrolysis [5]. The radiant heat flux was set to a fixed value of 20 kW/m² and all heat losses were ignored. The properties used in the simulation were: $\rho=1000$ kg/m³, $c_p=1.5$ kJ/kg/K, $k=0.15$ W/m/K, $H_v = 1700$ kJ/kg, $T_p=367^\circ\text{C}$ and $L=0.015$ m.

Despite the neglect of detailed chemical kinetic (Arrhenius) pyrolysis expressions, the numerical and integral model results shown in Fig. 3 are in a very good agreement, with an

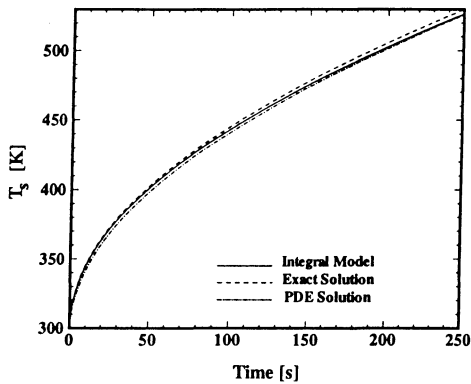


Figure 2(a): The front surface temperature calculated by the integral model, exact solution and PDE solution.

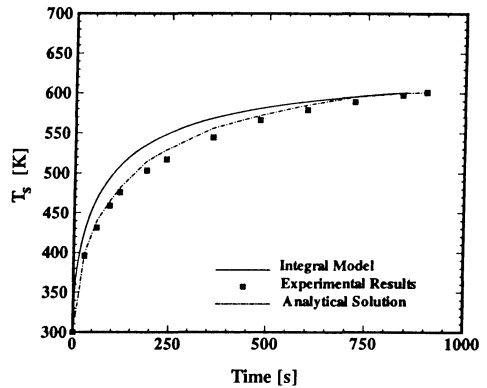


Figure 2(b): A comparison of the calculated and experimental front surface temperature.

error of less than 4%. This suggests that the assumption of a characteristic pyrolysis temperature is reasonable for non-charring materials. The only major difference between the integral model calculations and the numerical results is that the integral model predicts a small positive jump in the mass flux at the beginning of pyrolysis, while the numerical results do not show such a trend. However, this behaviour is limited to a short period of time when t is close to the heat-up time and has no effect on the overall solution. Besides, this initial jump facilitates the computation since small time steps are not needed to describe the steep rise in the mass flux during the early stages of pyrolysis.

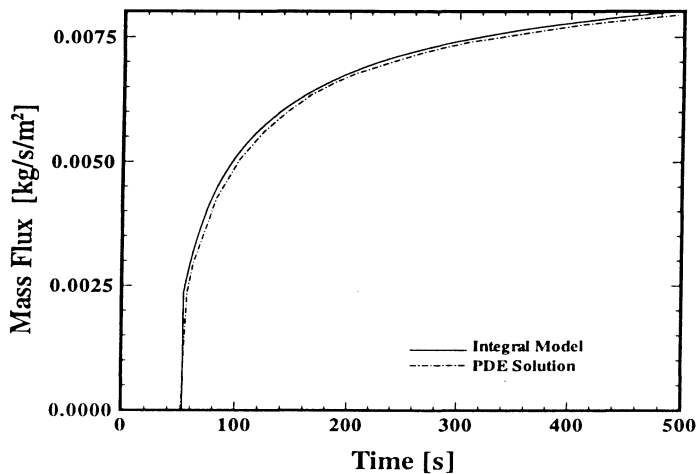


Figure 3: The mass flux history calculated by the integral model and a numerical solution.

A second set of numerical experiments was carried out to make a comparison with experimental data obtained for PMMA in an inert atmosphere [11]. The calculations were performed for two radiant heat fluxes: 14 kW/m² and 17 kW/m². The physical and thermal properties used in the simulation were: $L = 0.015$ m, $\rho = 1200$ kg/m³, $c_p = 2.2$ kJ/kg/K, $k = 0.2825$ W/m/K, $H_v = 1654$ kJ/kg [11]. The pyrolysis temperature was set to a value of 300°C for the 14 kW/m² heat flux and a value of 310°C for the 17 kW/m² heat flux [10]. In these calculations the effect of heat losses was also included.

Figure 4 shows a comparison of the calculated and the experimental mass loss rate for this case. For both heat fluxes, the worst discrepancy is observed to occur during the heat-up phase when in contrast to the experiment, the integral model does not predict any mass loss. However, the resulting error is not significant since at the early stages of the process the mass loss is generally low. Apart from that, the agreement between the theoretical and experimental results for the 14 kW/m² incident heat flux is remarkably good, especially for times greater than 1000 s. The mass loss rate results for the 17 kW/m² exposure are also good. However, the integral model and experiment follow different curves in the later stages of gasification. This is due to the fact that for the 17 kW/m² exposure the material becomes thermally thin and the heat loss from the back surface becomes significant after about 2100 s from the start, while the integral model assumes an adiabatic boundary condition at the back surface. The in-depth absorption of the radiant energy accounts for the discrepancy between the calculated and the experimental mass loss rates in the early stages of pyrolysis (300 s < t < 1000 s).

A number of other numerical experiments were also carried out to verify the accuracy of the present integral model, including comparison with the numerical and experimental results of Chen [6], Kashiwagi [10] and Delichatsios [5,12]. The results were acceptable in all cases.

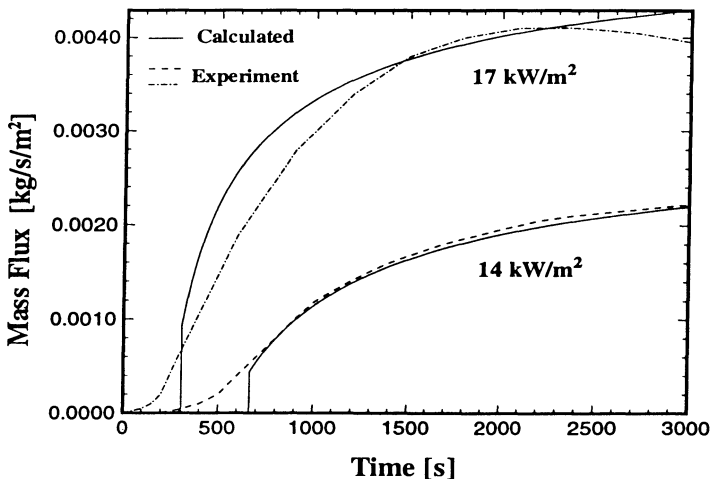


Figure 4: A comparison of the calculated and experimental mass loss rate for PMMA in an inert atmosphere.

APPLICATION OF THE INTEGRAL MODEL IN A CFD CODE

A set of calculations for the pyrolysis of PMMA in air was carried out to evaluate the

performance of the integral model when it is incorporated in a CFD fire code. This allowed us to consider the coupling between the solid and gaseous phases. In this case, heat losses from the burning surface do not have prescribed values but depend on the solution obtained for the gas phase.

To follow the experimental setup of Vovelle [11] we modelled PMMA degradation in a 0.4m x 0.4m x 0.5m chamber. A modified version of the FURNACE code was used to perform the calculations. This code has been used widely for the study of coal-fired boilers [13] and tunnel fires [14]. The code allows us to consider solid phase pyrolysis and provides an accurate coupling between the solid and gas phases. Combustion of volatiles is modelled using an Eddy-Breakup model [15]. Three different values for the external radiant heat flux (11 kW/m², 13 kW/m² and 16 kW/m²) were considered to make a comparison with the experimental results. The pyrolysis temperature was set to 300°C, 310°C and 320°C, respectively. The other parameters were the same as for the inert atmosphere case.

The results are shown in Fig. 5. As for the inert situation, the worst discrepancy was observed to occur during the initial stage of the process. In particular, the slope of the calculated mass loss rate curve is infinite at the moment of ignition whereas the experimental curve has finite slope. This is due to the fact that the integral model assumes that pyrolysis starts once the front surface temperature reaches a prescribed value. Apart from that initial stage, the prediction is quite accurate especially for low heat fluxes (11 kW/m² and 13 kW/m²). Again the discrepancy at the later stages of the process (the decrease in the mass loss rate) is explained in terms of the heat loss from the back surface, which is not accounted for in the integral model. This discrepancy becomes significant after about 1500 s for the 16 kW/m² exposure, 1800 s for the 13 kW/m² exposure and 2500 s for the 11 kW/m² exposure.

From a numerical point of view, the integral model provides a significant saving of CPU time. For this particular case, each iteration took 0.453 (s) when the integral model was employed, while the required CPU time for the PDE approach was 0.902 (s) per iteration.

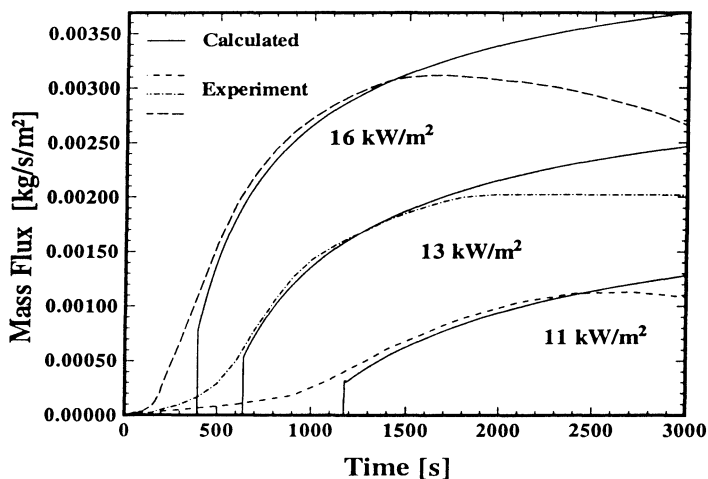


Figure 5: A comparison of the calculated and experimental mass loss rate for PMMA in an oxidising atmosphere.

CONCLUSIONS

An integral model based on a quadratic temperature profile has been developed and applied to the transient pyrolysis of non-charring materials. The model has been validated against several analytical and numerical solutions as well as experimental results. It has been shown that in all cases the integral model is able to simulate accurately the pyrolysis process. The present integral model is computationally cheap and has been incorporated successfully into a CFD fire code for the numerical investigation of the fire safety issues. Future work will consider the removal of some of the restrictions inherent in the present approach, e.g. consideration of thermally thin solids and allowing for in-depth absorption of radiation.

ACKNOWLEDGMENTS

This work was funded by an Australian Research Council Award in collaboration with the WorkCover Authority of NSW and Wormalds.

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