# A Finite Element Analysis on the Modeling of Heat Release Rate, as Assessed by a Cone Calorimeter, of Char Forming Polycarbonate

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**Abstract:** During the pyrolysis and combustion of polymers, heat is released and is typically measured with a cone calorimeter to better assess the polymer's flammability. Key data from a cone calorimeter include the rate of heat release, peak heat release rate, and time to ignition. Modeling heat release rate, as assessed by cone calorimetry, has not been extensively studied for char-forming polymers, such as, polycarbonate. Here we determine the heat release rate with the help of a one-dimensional transient finite-element model using commercially available COMSOL Multiphysics software. This model considers the heat and mass transport phenomena taking place throughout the thickness of the polymer and the growing char that is forming during the pyrolysis and combustion reaction. This model accurately predicts the heat release rate curve for char forming polycarbonate. This model should also be applicable to other systems that contain flame retardants where only the pyrolysis kinetics are different.

**Keywords:** Modeling Heat Release Rate, Char Forming Polycarbonate.

## 1. Introduction

The cone calorimeter is a small-scale testing apparatus that can effectively predict real world fire behavior. It can determine fire characteristics, such as, ignition time, weight loss, heat and smoke release rates, heat of combustion, and the average specific extinction area. A sample placed on a load cell is heated with a conical heater, sparked with an igniter and combusted in atmospheric air. The exhaust gasses are analyzed to determine heat and smoke release rates. The heat release rate is found by knowing the percentage of oxygen consumed during combustion. 1g of oxygen consumed in burning of most organic materials leads to the release of about 13kJ of heat.[1]

## 2. Description of Heat and Mass Transfer Processes

The modeling of heat and mass transfer during the heating, pyrolysis, and char formation of a polymeric material to predict the heat release rate, as assessed by the Cone Calorimeter, has not been examined. One paper by Fredlund[3] looked at the modeling of wood structures under fire but is quite complex and does not apply the results to heat release rates.

To describe the combustion process in detail, a step by step analysis follows. First, a flat plate of material (3mm x 100mm x 100mm) is placed in a holding device that is held up by a thick layer of insulating material. The cone calorimeter uses a circular radiant cone heater to deliver heat to the sample. This cone heater is preheated and equilibrated before any experiment is performed. Once the cone is at equilibrium, the polymer sample (at ambient temperature) is placed under the cone heater. The sample is first heated by radiation from the cone heater. As the sample heats, the polymer begins to break down into smaller gas molecules through a process called pyrolysis. The pyrolysis gases then diffuse out of the sample and into the atmosphere just above the sample where air is present. These gases then react with oxygen in the air, and thus, combustion takes place. A small amount of the heat of combustion is then fed back to the polymer providing more heat to the sample, however, most of the heat is lost to the atmosphere in heating up the surrounding air. During the process, some polymers form a char layer (intumescent, carbon, ceramic like), which has a lower thermal conductivity and diffusivity than the host polymer. These char properties help in lowering the peak heat release rate and spread the heat release rate over a much broader time frame. Ultimately, the polymer will be consumed during the reaction, residual char will be left behind, and combustion will cease.

Polycarbonate is a char forming polymer. Typical results from cone calorimeter data for polycarbonate are shown in Figure 1. The data were taken at a frequency of 1 Hz and the radiant heat flux was 50 kW/m$^2$. This heat flux corresponds to a cone temperature of 975 K [4]. Examining the heat release rate curve, for char forming polymers, after the peak heat release rate, a plateau region is observed before returning to zero. This is due to char formation. During the combustion, char formation occurs during pyrolysis. As the char grows thicker, the physics of the problem balance and give rise to a plateau and second peak. Thus, heat is released at a lower rate and over a broad time range, which is a major advantage for char forming polymers.
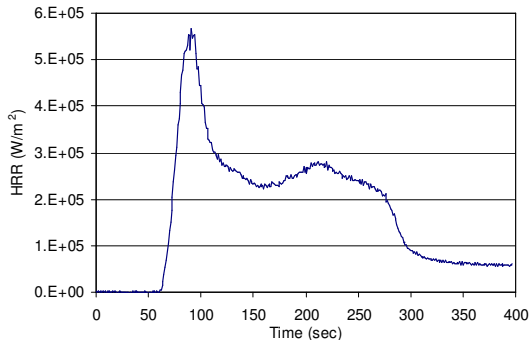


**Figure 1.** Heat Release Rate data from Cone Calorimetry for Polycarbonate. 50kW/m$^2$ Radiant Heat Flux.

To model the heat release rate observed in a cone calorimeter, partial differential equations, of mass and heat transfer that accurately depict the pyrolysis reaction and the temperature profile, must be solved. Due to the nature of this problem, the partial differential mass and heat transfer equations are coupled together, and an analytical solution does not exist. Therefore, a numerical solution must be sought. However, this three dimensional transient problem can be simplified to a one dimensional transient problem, lowering the computational power required to solve this system of equations. Figure 2 shows the physical situation involved, where $x_0$-$x_1$ (3mm) is the polymer zone, and $x_1$-$x_2$ (1nm initially, grows to ~ 2 cm) is the char formation zone, where a very thin char layer is introduced and grows at a velocity corresponding to the rate of pyrolysis.

The numerical technique used is a finite element model. The commercial software package COMSOL Multiphyics® was purchased and used for this modeling. COMSOL Multiphysics® is a user friendly, efficient, finite element method program, where many equations can be coupled together at one time and solved.
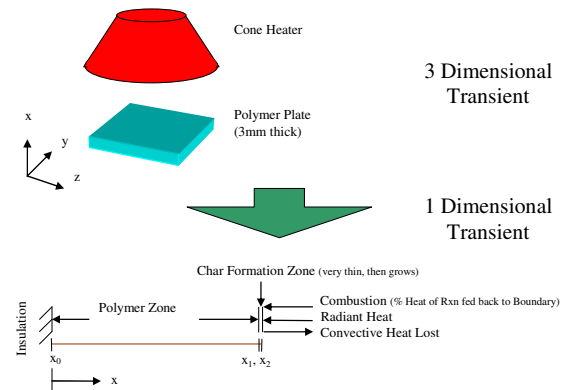


**Figure 2.** Schematic and geometry of physical model.

The partial differential equations that describe this system are discussed next.

## 3. Mass Transfer

Upon heating, the polymer pyrolyzes into gas and char. To model the problem, two distinct regions have been selected to account for the physics involved. The polymer zone accounts for the pyrolysis reaction; polymer is consumed and gas is produced. The char formation zone accounts for the char growth and acts only as a barrier to heat and mass transfer.

### 3.1 Pyrolysis Reaction inside the Polymer Zone [$x_0$-$x_1$]

The polymer must first react in the polymer zone. Reaction mechanisms have been greatly simplified to bring out the essential physics: polymer goes to gas plus char. The reaction scheme is shown:

$$Polymer \xrightarrow{\;k_0\;} Gas + Char$$
$$or$$
$$P \xrightarrow{\;k_0\;} \alpha \cdot G + (1-\alpha) \cdot C$$
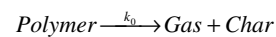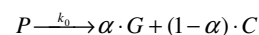
During this reaction, mass of polymer is consumed and produces a fraction, $\alpha$, of gas and

the remaining char. The first order reaction rate for polymer mass consumption is:

$$\frac{\partial m_p}{\partial t} = -k_0 \cdot m_p$$

where $m_p$ = Mass of polymer [kg], $t$ =Time [sec], $k_0$ =Rate constant for pyrolysis reaction [1/sec].

Further, the rate constant for the pyrolysis reaction, $k_0$, is a function of temperature and is better described by the Arrhenius relationship:

$$k_0 = A_0 \cdot \exp\left[\frac{-E_{A0}}{R \cdot T}\right]$$

where $A_0$ =Pre-exponential factor of pyrolysis reaction [1/sec], $E_{A0}$ =Activation energy of pyrolysis reaction [kJ/mol], $R$ =Gas constant [J/mol/K], $T$ =Temperature [K].

The pre-exponential factor and activation energy can be found by thermogravimetric analysis.

Considering the physics of the problem, in reality, the boundary, $x_1$, would move to the left as polymer is consumed. During the numerical modeling, it is assumed that polymer zone, $x_0$-$x_1$, is constant, thus, the volume is constant, and instead concentration (or density) changes with time. By dividing each side by a constant volume gives a concentration equation, which is easier to work with in COMSOL Multiphysics®. Re-writing the equation:

$$r_P = \frac{\partial c_p}{\partial t} = -k_0 \cdot c_p$$

where $r_P$ = Rate of polymer consumption during pyrolysis [kg/m$^3$/sec], $c_P$ =Concentration of polymer [kg/m$^3$].

When the polymer is consumed during the reaction, gas is produced. The production of gas in the polymer zone [$x_0$-$x_1$] must also diffuse through this zone to reach the char formation zone [$x_1$-$x_2$]. Writing the mass balance on the gas species that is produced:

$$r_{G\circ P} = \frac{\partial c_G}{\partial t} - D_{polymer} \cdot \frac{\partial^2 c_G}{\partial x^2} = \alpha \cdot k_0 \cdot c_p$$

where $r_{G\circ P}$ = Rate of gas evolution during pyrolysis [kg/m$^3$/sec], $c_G$ =Concentration of pyrolysis gases [kg/m$^3$], $D_{polymer}$ =Diffusion coefficient of pyrolysis gases through polymer [m$^2$/sec], $x$ =Length in the x-direction [m], $\alpha$ =Mass fraction of gas that is produced.

## 3.2 Mass Transfer in Char Formation Zone [$x_1$-$x_2$]

When the gas species that is produced during pyrolysis in the polymer zone [$x_0$-$x_1$] reaches the char formation zone [$x_1$-$x_2$], it must diffuse through to reach the outside boundary that is exposed to atmospheric oxygen. Writing the mass balance in the char formation zone:

$$\frac{\partial c_G}{\partial t} - D_{char} \cdot \frac{\partial^2 c_G}{\partial x^2} = 0$$

where $D_{char}$ =Diffusion coefficient of pyrolysis gases through char [m$^2$/sec].

## 4. Heat Transfer

Before the cone calorimeter experiment takes place, the thin (3mm) plaque of polymer is placed in a sample holder and suspended by a thick layer of insulating material. The polymer starts off at ambient conditions. When the polymer is placed under the cone heater, the outside surface is heated by radiation. Heat is transferred throughout the polymer by conduction. The pyrolysis reaction may also give off or consume heat. If there are other additives or fillers in the polymer, they too may react in an exo- or endothermic fashion. When the gas is produced and it diffuses out of the char layer, it reacts with oxygen to produce more heat. Part of the heat of combustion of this reaction is fed back to the surface, whereas the rest leaves to the atmosphere.

### 4.1 Heat Transfer in Polymer Zone [$x_0$-$x_1$]

In the polymer zone [$x_0$-$x_1$], heat is transferred by conduction. Heat of volatilization from pyrolysis and heat from other chemical processes (for example, endothermic reaction of aluminum hydroxide) are also shown:

$$\rho_{polymer} \cdot Cp_{polymer} \cdot \frac{\partial T}{\partial t} - k_{polymer} \cdot \frac{\partial^2 T}{\partial x^2} = -\Delta H_0 \cdot k_0 \cdot c_P + \Delta H_e$$

where $\rho_{polymer}$=Density of polymer [kg/m$^3$], $Cp_{polymer}$=Heat capacity of polymer [J/kg/K], $k_{polymer}$=Thermal conductivity of polymer [J/m/K/sec], $\Delta H_0$=Heat of volatilization of polymer [J/kg], $\Delta H_e$=Heat released or absorbed in other chemical processes (e.g. decomposition of additives) in polymer, *negligible* [J/kg].

## 4.2 Heat Transfer in Char Formation Zone [x$_1$-x$_2$]

In the char formation zone [x$_1$-x$_2$], heat is transferred through the char by conduction. The energy balance leads to:

$$\rho_{char} \cdot Cp_{char} \cdot \frac{\partial T}{\partial t} - k_{char} \cdot \frac{\partial^2 T}{\partial x^2} = 0$$

where $\rho_{char}$=Density of char [kg/m$^3$], $Cp_{char}$=Heat capacity of char [J/kg/K], $k_{char}$=Thermal conductivity of char [J/m/K/sec].

## 4.3 Combustion on the Surface Boundary

At the surface of the char, the mechanism of heat transfer is very complex. Heat is being introduced to the surface by radiation. Some of this heat may be reflected back. Combustion is taking place on the surface and a fraction of this heat of reaction is fed back to the boundary. Heat is also leaving the surface by convection to the outside air that is passing the sample and being swept away through the exhaust tube. At this boundary, the heat flux entering the surface is described as:

$$\dot{Q}_{Surface} = \phi \cdot \Delta H_1 \cdot (-\frac{\partial c_G}{\partial x})\Big|_{Surface} \cdot D_{Char} - h \cdot (T - T_{atm}) + \varepsilon \cdot \sigma \cdot \left(T_{cone}^4 - T^4\right)$$

where $\dot{Q}_{Surface}$=Heat flux at surface [J/m$^2$/sec], $\phi$=Percent heat transferred by heat of combustion, $\Delta H_1$=Heat of combustion [J/mol], $h$=Heat transfer coefficient of gas at surface [J/m$^2$/K/sec], $T_{atm}$=Temperature of atmosphere [K], $\varepsilon$=Emissivity, $\sigma$=Stefan-Boltzmann Constant [J/m$^2$/K$^4$/sec], $T_{Cone}$=Temperature of cone heater [K].

For modeling convenience, the convection heat loss term can be combined into the heat of combustion term, therefore, eliminating one parameter that is not exactly known. Now, it is easy to adjust one parameter, $\phi$, which sets the percentage of heat that is transferred back to the surface. The heat of combustion term, $\Delta H_1$, can be found from the cone calorimeter experimental data by integrating the heat release rate versus time curve or by simple means of disassociation of bond energies.

## 4.4 Heat Release Rate

The heat release rate versus time is measured by heat of combustion multiplied by the flux of gas diffusing out of the surface:

$$HRR = \Delta H_1 \cdot (-\frac{\partial c_G}{\partial x})\Big|_{Surface} \cdot D_{Char}$$

where *HRR* =Heat Release Rate [W/m$^2$].

## 5. Moving Mesh during Char Formation

For the model geometry, the char formation zone [x$_1$-x$_2$] is initially set to 1nm, as to try to minimize the influence of this barrier at the beginning of the computations. As time proceeds, the polymer heats, and as the temperature rises, gas is produced, but ignition does not start until some minimum level of gas surface concentration is reached (or minimum level of polymer loss). When this minimum level of gas surface concentration is reached, ignition begins along with moving mesh to simulate char formation.

Inside the model, each subdomain zone is set to 'free displacement,' meaning that, the mesh is not fixed in each subdomain and can move. For x$_0$ and x$_1$ the mesh displacement was set to zero, meaning that it was fixed in space. For x$_2$, the mesh growth velocity must be set so that the char grows at the correct rate. To arrive at the mesh growth velocity term, it is important to recognize the governing phenomena that control this complex system.

First, rate of mass growth of char is equal to the same polymer pyrolysis kinetics which can be equated as:

$$\frac{\partial m_C}{\partial t} = -(1-\alpha) \cdot \frac{\partial m_P}{\partial t} = (1-\alpha) \cdot k_0 m_P$$

where $m_C$ = mass of char [kg].

Dividing by $\rho_{char}$ the left hand side becomes

$$\frac{1}{\rho_{Char}} \frac{\partial m_C}{\partial t} = \frac{\partial \left( \frac{m_C}{\rho_{Char}} \right)}{\partial t} = \frac{\partial V}{\partial t} = \frac{\partial Ax}{\partial t} = A \frac{\partial x}{\partial t}$$

where $V$ is the volume and $A$ is the area and $x$ is the displacement of the char. The complete balance is

$$A \frac{\partial x}{\partial t} = -\frac{(1-\alpha)}{\rho_{Char}} \frac{\partial m_P}{\partial t} = \frac{(1-\alpha) \cdot k_0}{\rho_{Char}} m_P$$

Now by dividing both sides by $A$ and multiplying the right hand side top and bottom by a constant thickness, $\chi$, which is the distance between $x_0$ and $x_1$, 3mm.

$$\frac{\partial x}{\partial t} = -\frac{(1-\alpha) \cdot \chi}{\rho_{Char} \cdot A \cdot \chi} \frac{\partial m_P}{\partial t} = \frac{(1-\alpha) \cdot k_0 \cdot \chi}{\rho_{Char}} \frac{m_P}{A \cdot \chi}$$

By recognizing that $(A \cdot \chi)$ is a constant volume, $m_P/(A \cdot \chi) = c_P$ and $m_G/(A \cdot \chi) = c_G$. Therefore, the mesh velocity, *mvel*, can be defined as:

$$mvel = \frac{\partial x}{\partial t} = -\frac{(1-\alpha) \cdot \chi}{\rho_{Char}} \frac{\partial c_P}{\partial t} = \frac{(1-\alpha) \cdot k_0 \cdot \chi}{\rho_{Char}} c_P$$

This relationship directly shows how the physical phenomena are related to one another during char growth.

For modeling purposes, the $c_P$ time derivative is used and must be specified at a particular boundary, because $c_P$ is 0 at boundary $x_2$ where char growth is taking place. Therefore, the char growth mesh velocity becomes:

$$mvel = \frac{(1-\alpha) \cdot \chi}{\rho_{Char}} \cdot \left. \left( -\frac{\partial c_P}{\partial t} \right) \right|_{x_0}$$

where *mvel* =Char/mesh formation velocity [m/s], $\chi$ =Thickness constant, $x_1$-$x_0$ [m].

This function should only become active when $c_G$ reaches the minimum level of gas surface concentration for ignition and char growth to take place (or a minimum level of polymer loss). If it were active at all times, then char growth would be taking place in the very beginning. Even though this value might be quite small, it would affect the heat transfer of the problem enough to have drastic effects on the end results. In transient numerical modeling, a basic step function (on/off) can not be used due to convergence issues. Rather, a smoothed step function is used that lets the transition take place over some given range. The step function used in COMSOL Multiphysics® is called *flc1hs*. Now *mvel* can be written as:

$$mvel = flc1hs(\rho_{polymer} - c_P|_{x_0} - \min, scale) \cdot \frac{(1-\alpha) \cdot \chi}{\rho_{Char}} \cdot \left. \left( -\frac{\partial c_P}{\partial t} \right) \right|_{x_0}$$

where *flc1hs* =Smoothing step function in COMSOL Multiphysics®, min =Minimum level of polymer concentration reduction for ignition and char growth to begin [kg/m³], *scale* =Range over which transition takes place [kg/m³].

For the solver to use $\left. \left( -\frac{\partial c_P}{\partial t} \right) \right|_{x_0}$ and $c_P|_{x_0}$, they must be defined in COMSOL Multiphysics® in the *Extrusion Coupling Variables / Boundary Variables* section. Here, names are created, cPt3 and cP3, that will be valid on surface $x_2$ and will equal the negative of the $c_P$ time derivative evaluated at $x_0$ and the polymer concentration evaluated at $x_0$, respectively.

## 6. Initial and Boundary Conditions

Initial conditions for each zone of the problem are shown in Table 1.

**Table 1.** Initial Conditions, t=0.

| Initial Condition (x,0) | Zone | |
|---|---|---|
| | Polymer [$x_0$-$x_1$] | Char Formation [$x_1$-$x_2$] |
| $c_P$ | $c_P = \rho_{polymer}$ | $c_P = 0$ |
| $c_G$ | $c_G = 0$ | $c_G = 0$ |
| T | $T = T_{initial}$ | $T = T_{initial}$ |

where $T_{initial}$ =Initial Temperature [K].

Boundary conditions were set at the $x_0$ and $x_2$ boundaries. At $x_1$ all boundary conditions are assumed to be constant interfacial boundary conditions where temperature and concentration are held constant at the interface. Boundary conditions are shown in Table 2.

**Table 2.** Boundary conditions at $x_0$ and $x_2$.

| Boundary Condition $(x_0;x_3,t)$ | Boundary | |
|---|---|---|
| | $x_0$ | $x_3$ |
| $c_P$ | Flux=0 | Flux=0 |
| $c_G$ | Flux=0 | $c_G=0$ |
| T | Flux=0 | Flux = $\phi \cdot \Delta H_1 \cdot (-\frac{\partial c_G}{\partial x}) \cdot D_{Char} + \varepsilon \cdot \sigma \cdot \left(T_{cone}^{4} - T^4\right)$ |

# 7. Constants & Variables; COMSOL Conversion Chart

The names of the variables and constants described above must be changed to use in COMSOL Multiphysics®. Conversion charts for the variables, constants, and parameters are shown in Table 3-5. Also included in the tables are the values for the constants and parameters used to best fit the experimental data. All parameters in Table 5 is referenced at the bottom of the table. Some of the parameters come from TGA data and came from the physical properties of polycarbonate. The parameters that had to be adjusted to fit the data are $D_{char}$, $\rho_{char}$, $Cp_{char}$, $k_{char}$, $\phi$, $\varepsilon$, $scale$, and $c_{G\min}$. These parameters are within a realistic range.

**Table 3.** Variable conversion chart for COMSOL.

| Variables | COMSOL Name | Unit |
|---|---|---|
| $c_P$ | c_P | mol/m³ |
| $c_G$ | c_G | mol/m³ |
| $T$ | T | K |
| $t$ | t | sec |
| $x$ | x | m |

**Table 4.** Constants conversion chart for COMSOL.

| Constants | COMSOL Name | Value | Unit |
|---|---|---|---|
| $R$ | Rconst | 8.314 | J/(mol·K) |
| $T_{Cone}$ | Theater | 975 | K |
| $T_{initial}$ | Tinitial | 300 | K |
| $\sigma$ | sigma | 5.6703e-8 | J/(m²·K⁴·sec) |
| $\chi$ | mconst | 0.003 | m |

**Table 5.** Parameters conversion chart for COMSOL.

| Parameter | COMSOL Name | Value | Unit |
|---|---|---|---|
| $A_0$ | kconst0 | 45420 [a] | 1/s |
| $E_{A0}$ | Eact0 | 66.59 [a] | kJ/mol |
| $k_0$ | kP | kconst0*exp(-Eact0/Rconst/T) | 1/s |
| $D_{polymer}$ | Dpolymer | 10 [b] | m²/s |
| $\rho_{polymer}$ | rpolymer | 1200 [c] | kg/m³ |
| $Cp_{polymer}$ | Cppolymer | 1200 [c] | J/(kg·K) |
| $k_{polymer}$ | kpolymer | 10 [b] | J/(m·K·s) |
| $D_{char}$ | Dchar | 1.8*10⁻⁶ [d] | m²/sec |
| $\rho_{char}$ | rchar | 35 [d] | kg/m³ |
| $Cp_{char}$ | Cpchar | 100 [d] | J/(kg·K) |
| $k_{char}$ | kchar | 0.05 [d] | J/(m·K·s) |
| $\Delta H_1$ | DH1 | 23655000 [e] | J/kg |
| $\alpha$ | alpha | 0.8 [a] | |
| $\phi$ | HRfrac | 0 [d] | |
| $\varepsilon$ | Emissivity | 0.23 [d] | |
| $scale$ | scale | 100 [d] | kg/m³ |
| $c_{G\min}$ | min | 150 [d] | kg/m³ |
| $mvel$ | mvel | Flc1hs(rpolymer-cP3-min,scale)*(cPt3)/rchar*mconst*(1-alpha) | m/s |

a ~ TGA experimentally determined data.
b ~ Polymer zone assumed to diffuse and heat equally.
c ~ Typical polycarbonate data.
d ~ Reasonably adjusted parameters to fit experimental data.
e ~ $\Delta H_1$ found by integrating HRR curve of PC (see Figure 2) and dividing by $\rho_{polymer} \chi$.

With everything mentioned, the model is complete. Adjustments to the parameters can then be made to fine tune the system. Results of the computations are shown next.

# 8. Numerical Modeling Results

## 8.1 Polycarbonate with No Flame Retardant

Fine tuning of the model parameters must be made initially to fit the time of ignition data. This is done by setting the char velocity to zero and adjusting the emissivity, $\varepsilon$, value. One other piece of data to check is the Total Heat Released, $\Delta H_1$, which is the area under the curve of the heat release rate versus time plot.

Now that the emissivity term has been set and the total area under the curves are the same, char growth can now be implemented. Turning the char formation velocity term on, the values *min* and *scale* can be set. These values come from looking at a plot of $c_P$ versus time and observing when char formation should occur. The value that works best is a 1/24 loss of polymer concentration for char formation to begin. Therefore, *min* = 150 and *scale* = 100, so that char formation begins at (150-100)/1200 = 1/24 loss of polymer concentration.

Figure 3 shows the modeled COMSOL data compared to the cone data. Parameters used are those listed in Table 5.
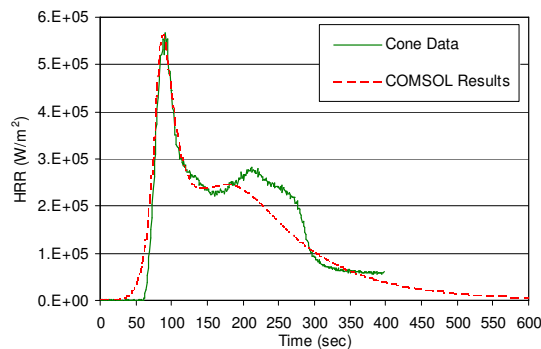


**Figure 3.** Heat Release Rate versus Time of PC No FR; COMSOL Model Results with Char Growth compared to Cone Data.

It is instructive to explain in more detail the physical phenomena taking place. These explainations can be found in Statler's disseration[5] and will be presented at the COMSOL 2008 Boston Conference.

### 8.2 The Influence of Flame Retardant Additive

The use of two different flame retardants have be exaimed and numerical data fit well with the experimental data. These data can be found in Statler's disseration[5] and will be presented at the COMSOL 2008 Boston Conference.

### 9. Conclusions

A mathematical model has been developed to predict accurately the heat release rate of a char forming polymeric material during combustion described under the cone calorimeter. From the model, time to ignition and peak heat release rate can be predicted for char forming materials using computational methods. The HRR after the peak (plateau region) can also be modeled with minor error. For this model to predict the heat release rate curves of a polymeric material some physical properties must be known, such as, polymer: thermal conductivity, density, heat capacity, and char: thermal conductivity, density, heat capacity, and diffusivity. These are quantities that can be measured by other instruments. Kinetic rate constants and amount of char formed can be found by thermogravimetric analysis. Heat of combustion of a polymer can be calculated by simple bond energies. From these data, heat release rate curves can be predicted and the use of a cone calorimeter is not necessary. However, a cone calorimeter will prevail until confidence has been built in the use of the model for different polymeric materials.

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### 11. Acknowledgements