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# Coupled analytical approach to predict piloted flaming ignition of non-charring polymers



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| <i>Keywords:</i><br>Piloted ignition<br>Thermoplastics<br>Pyrolysis<br>Flammability | Classical thermal theory of piloted ignition is extended by coupling the heat balance at the exposed sample surface<br>and the finite-rate pyrolysis in the material volume. Approximate analytical solutions for the sample temperature<br>are obtained for an arbitrary sample thickness, with the external radiative heating, surface re-radiation, heat of<br>gasification, and the convective heat flux corrected for blowing taken into account. The volatile mass flux is<br>evaluated by integrating the pyrolysis rate throughout the layer, with the assumption of high activation energy<br>limit. Critical mass flux of combustible volatiles is used as the ignition criterion. This enables the ignition tem-<br>perature to be evaluated instead of being pre-assumed as is done in the classical thermal theory. Coupled<br>analytical approach proposed in this work is verified by comparisons to the numerical solution obtained by the<br>Pyropolis model for the same problem setup. This approach has also been validated by comparisons to published<br>experimental data (ignition temperatures and times to ignition) for three non-charring thermoplastics: poly-<br>methylmethacrylate, polyethylene and polypropylene. |  |  |

### 1. Introduction

Classical thermal theory of piloted ignition, for example see Ref. [1], relies on the assumption that flaming ignition occurs as soon as the sample surface attains the specified ignition temperature. The surface temperature is obtained by solving thermal conductivity equation in non-reacting material layer, which is exposed to the external heat flux. With additional assumptions of constant material properties, infinitely thick material layer, negligible heat consumption in pyrolysis, no surface heat losses prior to ignition, no in-depth radiative absorption, the thermal conductivity equation leads to the analytical solution, enabling evaluation of the time to ignition as a function of net heat flux. Suitability of this approach for engineering calculations is justified by the experimental observations indicating weak variation of the surface temperature at piloted ignition observed in a number of works including Refs. [2,3].

There are several approaches to determine the ignition temperature,  $T_{ign}$ . In the first one, surface temperature is measured directly at the onset of either flashing/incipient/transient ignition (flash point) or stable/ sustained burning (fire point) [4–7]. The ignition temperature can also be evaluated from the measured critical heat flux,  $q''_{ext,cr}$ , using the relation,  $\varepsilon q''_{ext,cr} = h(T_{ign} - T_0) + \varepsilon \sigma T^4_{ign}$ , showing that absorbed heat flux is balanced by convective heat losses and surface re-radiation [1]. Finally,

numerical value of the ignition temperature can be calibrated to fit the experimental data for the time to piloted ignition,  $t_{ign}$ , by applying the thermal theory [8,9].

Thus calibrated ignition temperature deviates considerably from the actual measured values of the sample surface at ignition [8]. To obey the measured ignition temperatures within the classical thermal theory, effective material properties are derived in Refs. [6,9] to fit the measured ignition times. It also appears that these properties deviate from those independently measured for the same material. This indicates the deficiency of the classical thermal theory and the need in a predictive approach utilizing as many as possible actual (not effective) material properties.

The above inconsistency also indicates that the surface temperature at ignition is not the fundamental material property. Indeed, in Refs. [6,9,10] the surface temperature at ignition was found to increase with increasing of external heat flux, thereby demonstrating that the ignition temperature is determined by other thermochemical properties and the heat balance at the exposed surface.

An alternative ignition criterion considered in Refs. [3,5], among others, is the critical mass flux of pyrolysis volatiles,  $m''_{ign}$ . This criterion is coupled with volatile concentration at the pilot location corresponding to either lower flammability limit or stoichiometric mixture. Accordingly,

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| Nomenclature   |  | $\Delta h_g$   | heat of gasification, J/kg   |  |
|--|--|--|--|--|
| Latin sym<br>a<br>Bi<br>A<br>c<br>c <sub>P,vol</sub><br>E <sub>a</sub> | nbols<br>thermal diffusivity, m <sup>2</sup> /s<br>Biot number<br>pre-exponential factor, 1/s<br>specific heat of solid material, J/(kg·K)<br>constant pressure specific heat of volatiles, J/(kg·K)<br>activation energy, J/mol | $ \begin{aligned}         \mu_g \\         \omega \\         \sigma \\       $ | volatile mass fraction in pyrolysis<br>adjustable constant<br>Stefan-Boltzmann constant, $5.67 \cdot 10^{-8}$ W/(m <sup>2</sup> ·K <sup>4</sup> )<br>pyrolysis chemical time scale, <i>s</i><br>time period to increase initial surface temperature by one<br>characteristic temperature interval<br>dimensionless surface temperature |  |
| $f(\alpha)$  | conversion function (pyrolysis reaction model)   | Subscript  | 'S   |  |
| Fo   | Fourier number   | cr   | critical   |  |
| h  | heat transfer coefficient, W/(m <sup>2</sup> ·K)   | eff  | effective  |  |
| i  | iteration number   | ext  | external   |  |
| k  | thermal conductivity, W/(m·K)  | ign  | ignition   |  |
| <i>m</i> ″   | mass flux of volatiles, kg/(m <sup>2</sup> ·s)   | net  | net  |  |
| n  | pyrolysis reaction order   | r  | reaction zone  |  |
| $q^{\prime\prime}$   | heat flux, W/m <sup>2</sup>  | rad  | radiative  |  |
| ŕ  | pyrolysis reaction rate, 1/s   | \$   | surface  |  |
| ${\mathcal R}$   | gas constant, 8.314 J/(mol·K)  | Т  | heated layer   |  |
| t  | time, s  | tr   | transition from thermally thick to thermally thin mode   |  |
| Т  | temperature, K   | vol  | volatiles  |  |
| x  | coordinate normal to sample surface, m   | 0  | initial, ambient   |  |
| z  | limensionless mass flux of volatiles   |  |  |  |
| Ze   | Zeldovich number   |  | Abbreviations  |  |
|  |  | PC   | polycarbonate  |  |
| Greek symbols  |  | PE   | polyethylene   |  |
| α  | conversion   | PMMA   | polymethylmethacrylate   |  |
| δ  | sample thickness, m  | POM  | polyoxymethylene   |  |
| δ <sub>r</sub>   | reaction zone thickness, m   | PP   | polypropylene  |  |
| $\delta_T$   | heated layer thickness, m  | PS   | polystyrene  |  |
| $\Delta h_c$   | heat of combustion, J/kg   |  |  |  |

distinct values of the critical mass flux are proposed for flash point and fire point. It was found by Thomson and Drysdale in Ref. [5] that the critical ignition mass flux correlates with the chemical composition of combustible polymers: the hydrocarbon polymers (PE, PP and PS) have the values of around 1  $g/(m^2 \cdot s)$ , while the oxygenated polymers (PMMA) and POM) have the values which are approximately twice as higher. This is explained by the difference in the amount of volatiles needed to reach the lower flammability concentration or to create stoichiometric volatile-air mixture. In Ref. [3], Lyon and Quintiere summarize available measurement data and come to the conclusion that a more general criterion could be the critical heat release rate,  $m''_{ign}\Delta h_c$ , which is about 24 kW/m<sup>2</sup> for the flash point and 66 kW/m<sup>2</sup> for the fire point, with no specific distinction between hydrocarbon and oxygenated polymers. For a wide range of polymers considered in Ref. [3], critical mass fluxes at the flash point vary around 1 g/( $m^2$ ·s) and those at the fire point are 2–3 times higher.

Relevance of the critical mass flux to the limit concentrations in volatile-air mixture makes the value of  $m''_{ign}$  to be a more fundamental ignition criterion than the surface temperature at ignition. In its turn, the volatile mass flux depends on the pyrolysis rate, which is not considered in the classical thermal theory. In this work we aim at developing an analytical approach extending the classical thermal theory by coupling the heat transfer at the exposed sample surface with the finite pyrolysis rate in the material volume. The volatile muss flux is evaluated by integrating the pyrolysis rate throughout the layer, and the approximate analytical solution is obtained in the high activation energy limit. Incorporating this solution in the surface energy balance including external radiative heating, surface re-radiation, heat of gasification, and the convective heat flux corrected for blowing yields the sample surface temperature at ignition. Since the critical mass flux of combustible

volatiles is used as the ignition criterion, the ignition temperature is evaluated instead of being pre-assumed as is done in the classical thermal theory.

To the authors' knowledge, the only previous attempt to allow for the variation of the ignition temperature in analytical simulation of ignition delay was undertaken in Ref. [11], where thermally thick limit is considered, with both surface heat losses and heat of gasification ignored. The internal temperature profile and the temperature dependence of the rate constant are approximated, respectively, by algebraic and power-low analytical expressions, which require fitting constants to be determined. This approach was applied in Ref. [11] to simulate ignition of PMMA, and the predicted times to ignition were shown to be notably underestimated compared to the measured values. In this work, we refine this approach by incorporating a more straightforward technique to evaluate the integral pyrolysis rate (including its extension to the n-th order reactions), couple it to the complete heat balance at the exposed surface, and consider both thermally thick and thermally thin limits. This enables predicting surface temperature and time to ignition, both consistent with the measurement data for three non-charring polymers (PMMA, PE, PP).

#### 2. Model description

The model includes three major components to determine (i) the net heat flux received by the sample, (ii) the integral pyrolysis rate as a function of the sample surface temperature, and (iii) coupling between the sample surface temperature and the net heat flux. The first model component allows for the heat transfer at the gas side of the exposed surface and provides the value of the net heat flux. The second component considers finite-rate pyrolysis in the solid material and provides the Download English Version:

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