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Steady and transient pyrolysis of thick clear PMMA slabs

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ABSTRACT

Thermal degradation of solid fuels is an issue of major importance in the prediction of fire ignition and growth. The pyrolysis rate is generally deduced from the energy balance at the surface of the solid material, while all or part of the in-depth loss into the solid material or the net radiation at its surface is disregarded. The aim of the present study is to improve the accuracy and predictive capability of solid fuel pyrolysis models. Both the steady and transient burning of thick clear polymethyl methacrylate (PMMA) slabs are investigated. First, experiments are conducted to evaluate quantitatively the energy flux components at the surfaces of steady-burning vertically oriented slabs. The total heat flux from the flame is measured, showing a decrease from 30.9 to 23.4 kW/m^2 as the sample height increases from 2.5 to 20 cm. A specific procedure for estimating the surface reradiation of burning slabs is conducted, changing from the vertical to the ceiling configuration, where flame self-extinction occurs. At midheight of the slab, the surface reradiation heat flux is as high as 11.5 kW/m^2, which corresponds to the emissive power of an equivalent blackbody at a temperature of 671 K, in accordance with current spectroscopic measurements. Local steady-burning mass loss rates are then deduced from the energy balance at the fuel surface and compared with direct measurements, showing good agreement. Second, the transient burning of thermally thick slabs of clear PMMA is studied experimentally and numerically. Pure pyrolysis, for incident heat fluxes of 14 and 18 kW/m^2, and flaming experiments are conducted. A one-dimensional numerical model is proposed to solve the problem of conjugated heat transfer into the semitransparent material, assuming that the exposed surface is a perfect emitter and a perfect absorber (soot-covered). The Schuster–Schwarzschild approximation is employed to calculate in-depth radiation, using data for the absorption coefficient of clear PMMA with a very fine spectral resolution. Model results exhibit the same trend as that revealed in experiments for the rise in temperature in the sample and the regression rates. Results show that the surface temperature tends asymptotically to a constant value, which increases with the incident heat flux, and that steady burning occurs when the surface temperature saturates. Similar steady-burning regression rates are obtained for the 18 kW/m^2 and flaming configurations. It is found that for the flaming case, the increase in the steady-burning regression rate due to higher incident heat flux and surface temperature is nearly compensated for by the decrease of regression rate caused by higher losses (outward reradiation and heat of gasification)

1. Introduction

For a semitransparent polymer, here clear polymethyl methacrylate (PMMA), the local mass loss rate (MLR) due to pyrolysis, $\dot{m}''$, can be related to heat fluxes through an energy balance equation at the surface of the burning material as (Fig. 1a).

$$\begin{aligned} &\underbrace{q_{fl}^{conv}}_{\text{convection from the flame}} + \underbrace{q_{fl}^{rad}}_{\text{radiation from the flame}} + \underbrace{\dot{m}''h_{ng}}_{\text{heat carried to the surface by non volatile PMMA}} \\ &= \underbrace{q_{rr}}_{\text{outward re-radiation}} + \underbrace{q_{id}^{cond}}_{\text{conduction into the solid}} + \underbrace{q_{id}^{rad}}_{\text{net radiation into the solid}} \\ &+ \underbrace{\dot{m}''h_g}_{\text{heat carried by the PMMA vapors away from the surface}} + \underbrace{q_{fl}^{refl}}_{\text{reflection of flame radiation}} . \end{aligned} \tag{1}$$

The left-hand terms of Eq. (1) represent the heat fluxes from the flame and the heat carried to the surface by the polymer in its nonvolatile state. The right-hand terms represent the outward

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Nomenclature

c_p	heat capacity (J/kg/K)
$E_{b\eta}$	blackbody emissive power (W/m)
g	gravitational acceleration (m/s^2)
h	convective heat transfer coefficient (W/m^2/K)
h_g	heat of gasification (J/kg)
h_{ng}	defined as $h_{ng} = h_{mel} + \int_{T_\infty}^{T_p} c_p(T)dT$ (J/kg)
h_{vap}	heat of vaporization (J/kg)
h_{pyr}	heat of pyrolysis (J/kg)
h_{mel}	heat of melting (J/kg)
L	initial slab thickness (m)
LBL	laminar boundary layer
$\dot{m}''$	pyrolysis rate per unit area (kg/m^2/s)
MLR	mass loss rate
n	refractive index (–)
PMMA	polymethyl methacrylate
q	heat flux (W/m^2)
r	surface reflectivity (–)
RHF	radiative heat flux (W/m^2)
s	regressing surface location (m)
t	time (s)
T	temperature (K)
x	distance from the leading edge (m)
y	space coordinate along the sample thickness direction (m)

Greek letters

ε	emissivity
η	wavenumber (m^{-1})
κ	absorption coefficient (m^{-1})
λ	thermal conductivity (W/m/K)
ρ	density (kg/m^3)
σ	Stefan-Boltzmann constant (=5.67 × 10^{-8} W/m^2/K^4)

Superscripts and subscripts

cond	conduction
conv	convection
fl	flame
id	in-depth
p	pyrolysis
rr	surface reradiation
s	solid
∞	ambient
rad	radiation
η	wavenumber-dependent property

reradiation, the in-depth losses into the solid, the heat carried by the fuel vapors away from the surface, and the part of flame radiation that is reflected by the surface. Since PMMA is a melting polymer, the energy required to raise the temperature of a unit mass of polymer from ambient temperature to the final pyrolysis temperature T_p and to melt it is given by $h_{ng} = h_{mel} + \int_{T_\infty}^{T_p} c_p(T)dT$, where h_{mel} is the heat of melting. The term h_g is the effective heat of gasification [1], assuming that pyrolysis and evaporation of the pyrolysis products only take place at the final pyrolysis temperature (in this case the surface temperature), $h_g = h_{ng} + h_{vap} + h_{pyr}$, where h_{vap} and h_{pyr} are the latent heats of vaporization and pyrolysis.

Eq. (1) slightly differs from that given by Orloff et al. [2], who disregarded melting, assuming that the polymer sublimes at a critical temperature and remains inert at temperatures below this. As stated by Orloff et al. [2,3], for the steady burning of an infinitely thick slab, the heat carried to the surface by the solid exactly equals the conduction plus net radiation into the solid, yielding

$$\dot{m}''h_{ng} = q_{id}^{cond} + q_{id}^{rad}. \tag{2}$$

Eq. (1) thus becomes

$$q_{fl}^{conv} + q_{fl}^{rad} = q_{rr} + \dot{m}''h_g + q_{fl}^{refl} \tag{3}$$

or

$$q_{fl}^{conv} + (1-r)q_{fl}^{rad} = q_{rr} + \dot{m}''h_g, \tag{4}$$

where r is the surface reflectivity.

This indicates that the calculation of the steady burning rate does not require the estimation of heat losses into the solid interior (Fig. 1b).

Most pyrolysis models are based on assumptions regarding the in-depth losses into the solid fuel. Following the pioneering work of Emmons [4], where the flow field was approximated as a two-dimensional reacting-boundary-layer problem for forced flows, some researchers developed the reacting laminar boundary layer (LBL) theory for free flows [5–8]. The LBL theory is based on the assumptions that burning is steady and that convection is the only mode of heat transfer, disregarding the radiative terms in the energy balance equation at the fuel surface (Eq. (4)). Ahmad and Faeth [9] developed an integral model to obtain the pyrolysis mass flow rate, which avoids solving the set of LBL equations numerically. The LBL theory was assessed by comparison with experimental quantities such as the standoff distance [5] or the total burning rate [6]. Furthermore, this theory served as a basis for the determination of the flame height [7,8], as well as other global properties of the flame and of the flow field. These studies showed that the agreement between experimental data and theoretical predictions using the mass transfer number (also called the *B* number) was not satisfactory. The origin of the discrepancies was identified by Torero et al. [10], who demonstrated the need to include nonconvective heat transfers at the surface, especially net radiation and in-depth conduction for thick solid fuels. Another possible approach is to adjust the pyrolysis mass flow rate calculated by the LBL theory using experimental results [11]. An extension of this approach including transient conductive heat losses was presented for inclined burning slabs [12]. Rangwala et al. [13,14] modified the two-dimensional LBL theory to incorporate finite-width (three-dimensional) effects and heat losses, assuming that a fraction of fuel diffuses to the sides.

Experimental studies were also performed to evaluate the contribution of one or more surface energy flux components to the burning rate. Some of the most comprehensive studies are listed in Table 1 and briefly discussed here, focusing on MLR and heat flux measurements. In their pioneering work, Orloff et al. [2] performed a detailed, careful heat transfer analysis using 1.57-m-high, 0.41-m-wide, and 4.5-cm-thick vertical slabs of transparent PMMA. They measured the burning rate and total outward radiation (flame plus surface) at different heights along the slab, attributing the significant increase of the steady burning rate with height to increasing flame radiation. Reradiant heat loss from the fuel surface vs. height was deduced from that measured at midheight by means of a thermopile radiometer, relating the MLR to the surface temperature by a zero-order Arrhenius expression and a unit surface emissivity. Despite the large sample size, the Arrhenius correction induced only small variations of reradiant heat flux, no more than ±7%. The convective heat flux was inferred from the steady surface