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Madrid, Spain

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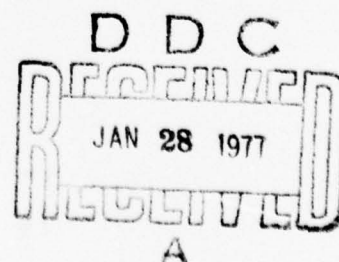
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M. Kindelán and A. Liñán

ABSTRACT

An asymptotic analysis is developed, describing ignition of a reactive solid suddenly exposed to hot gases. It is found that in the limit of high activation energy of the Arrhenius exothermic reaction, the chemical heat release occurs in a thin diffusive-reactive layer near the surface. The solution for this inner layer is then matched to the solution for the outer transient-diffusive region. An explicit formula for the ignition time is obtained.

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1. INTRODUCTION

Several theoretical analyses of the ignition mechanism of a reactive solid have been developed in the past, (see review by Merzhanov and Averson¹). Recently, asymptotic techniques have been successfully applied to describe ignition under different heating mechanisms. Liñan & Williams analyzed ignition of a reactive solid by a constant radiant energy flux absorbed at the surface² or in depth³. Bush & Williams⁴ studied the influence of heat losses by conduction to the ambient gas on the radiant ignition process. Williams and Niiooka⁵ studied ignition by forced convective heating or newtonian heating, and Liñan and Williams⁶ have recently completed the analysis of ignition by a hot plate (surface temperature constant).

In the present study we analyze the ignition process of a reactive solid by conductive heating from a hot ambient gas, so as to complete our understanding of ignition of a reactive solid under all the main types of heating mechanisms. The analysis is also applicable to ignition of a reactive solid by a hot body when the thermal responsivities of both materials are of the same order.

In addition, this study is a further step on our analysis of ignition of a condensed material by hot gases, with different locations of the exothermic reaction. In reference (8) we assumed an heterogeneous reaction occurring at the surface between the condensed fuel and the ambient oxidizer. To analyze ignition by a gas phase reaction, it is necessary to analyze first the gasification process that produces gaseous fuel to react with the ambient oxidizer. We have recently analyzed⁷ this gasification process and at the present time we are using the temperature and mass fraction profiles resulting from this analysis to study gas phase ignition.

These analyses have been developed under similar assumptions, and differ only on the location of the exothermic reaction. It will now be possible to use experimental results in trying to ascertain the location of the reaction producing ignition for a given material under some given conditions.

2. FORMULATION

We assume that no gasification of the solid occurs during the ignition transient. Under assumptions such

as those used in reference (8), the energy conservation equations in the solid and gas-phases, become

$$\frac{\partial \theta}{\partial \tau} = \frac{\partial^2 \theta}{\partial \xi^2} + \exp \frac{\beta \theta}{\Gamma \{1 + \theta (T_{g0}/T_0)\}} \quad (1)$$

$$\frac{\partial \theta_g}{\partial \tau} = \frac{\partial^2 \theta_g}{\partial \xi^2} \quad (2)$$

Where the second term in the right hand side of equation (1) represents the effect of an exothermic distributed reaction of the Arrhenius type.

The boundary and initial conditions are

$$\left. \frac{\partial \theta}{\partial \xi} \right|_s + \Gamma \left. \frac{\partial \theta_g}{\partial \xi} \right|_s = 0 \quad (3)$$

$$\theta(\tau, 0) = \theta_g(\tau, 0) \quad , \quad \theta(0, \xi) = \theta(\tau, \infty) = -\Gamma$$

$$\theta_g(0, \xi) = \theta_g(\tau, \infty) = 1 \quad (4)$$

where all symbols are defined in the nomenclature.

For small values of τ the effects of the heat release can be neglected so that the temperature profiles take their inert values:

$$\theta_{Ig} = -\theta_I/\Gamma = \text{erf} (\xi/2\sqrt{\tau}) \quad (5)$$

The analysis that follows is based on the assumption that the nondimensional activation energy β/Γ is a large parameter. We consider Γ to be of order unity, although the results are also valid when Γ is small. Since β/Γ is large, small increments of order Γ/β in the nondimensional temperature θ , above its inert value θ_I , produce changes in the reaction rate by a factor e , and these small increments are sufficient to produce a thermal runaway in surface temperature at a finite ignition time τ_{ign} . We anticipate that the time τ required to produce these increments is of order unity and therefore τ_{ign} is of order unity.

From equation (5) it is observed that the inert temperature in the condensed phase decreases towards the interior, so that the exothermic reaction only occurs in a small layer near the surface where $\beta\xi/\Gamma\sqrt{\tau}$ is of order unity. Thus, two different regions exist in the

condensed phase: an inner layer with characteristic space dimension $z=\beta\xi$, in which the reaction takes place, and an outer region with characteristic space dimension ξ . To analyze these regions, we use the following expansions

Inner:

$$\theta = -\frac{\Gamma z}{\beta\sqrt{\pi\tau}} + \left(\frac{\Gamma}{\beta}\right)\chi_1 + \left(\frac{\Gamma}{\beta}\right)^2\chi_2 + \frac{\Gamma z^3}{12\beta^3\tau\sqrt{\pi\tau}} + \dots \quad (6)$$

Outer:

$$\theta = -\Gamma \operatorname{erf}\left(\frac{\xi}{2\sqrt{\tau}}\right) + \frac{\Gamma}{\beta} X_1 + \dots \quad (7)$$

Gas phase:

$$\theta_g = \operatorname{erf}\left(\frac{\xi}{2\sqrt{\tau}}\right) + \frac{\Gamma}{\beta} \chi_{g1} + \dots \quad (8)$$

The variables χ_1, χ_2 are functions of z and τ and the variables X_1 and χ_{g1} are functions of ξ and τ .

Introducing these expansions in equations (1) and (2), expanding and collecting like powers of β , the following equations are obtained

$$\frac{\partial^2 \chi_1}{\partial z^2} = 0 \quad (9)$$

$$\Gamma^2 \frac{\partial^2 \chi_2}{\partial z^2} = -\exp \{ \chi_1 - z/\sqrt{\pi\tau} \} \quad (10)$$

$$\frac{\partial X_1}{\partial \tau} = \frac{\partial^2 X_1}{\partial \xi^2} \quad (11)$$

$$\frac{\partial \chi_{g1}}{\partial \tau} = \frac{\partial^2 \chi_{g1}}{\partial \xi^2} \quad (12)$$

From the boundary and initial conditions (3) and (4) the following conditions are obtained

$$\left. \frac{\partial \chi_1}{\partial z} \right|_s = 0 \quad \left. \frac{\partial \chi_2}{\partial z} \right|_s = - \left. \frac{\partial \chi_{g1}}{\partial \xi} \right|_s \quad (13)$$

$$\chi_1(\tau, 0) = \chi_{g1}(\tau, 0) \quad , \quad \chi_1(\tau, \infty) = \chi_{g1}(\tau, \infty) = 0 \quad (14)$$

$$\chi_1(0, z) = \chi_2(0, z) = \chi_1(0, \xi) = \chi_{g1}(0, \xi) = 0 \quad (15)$$

Equations (9)-(12), can be integrated with conditions (13)-(15) to give

$$\chi_1(\tau, z) = \chi_{1s}(\tau) \quad (16)$$

$$\frac{d\chi_2}{dz} = \frac{d\chi_2}{dz} \Big|_s + \frac{\sqrt{\pi\tau}}{\Gamma^2} \exp(\chi_{1s} - z/\sqrt{\pi\tau}) - \frac{\sqrt{\pi\tau}}{\Gamma^2} \exp(\chi_{1s}) \quad (17)$$

$$\chi_1 = - \frac{1}{\sqrt{\pi}} \int_0^\tau \frac{\frac{\partial \chi_1}{\partial \xi} \Big|_s \exp \{-\xi^2/4(\tau-\tau')\}}{\sqrt{\tau-\tau'}} d\tau' \quad (18)$$

$$\chi_{g1} = - \frac{1}{\sqrt{\pi}} \int_0^\tau \frac{\frac{\partial \chi_{g1}}{\partial \xi} \Big|_s \exp \{-\xi^2/4(\tau-\tau')\}}{\sqrt{\tau-\tau'}} d\tau' \quad (19)$$

From equations (16) and (17) the following matching conditions with the solution for the outer zone are derived

$$\chi_1(\tau, 0) = \chi_{1s} \quad (20)$$

$$\frac{\partial \chi_1}{\partial \xi} \Big|_s = \Gamma \frac{\partial \chi_2}{\partial z} \Big|_\infty = \Gamma \frac{\partial \chi_2}{\partial z} \Big|_s - \frac{\sqrt{\pi\tau}}{\Gamma} \exp(\chi_{1s}) \quad (21)$$

From equations (18), (19) and using equations (13), (14), (21), the value of the surface temperature deviation from its inert value is found to satisfy the integral equations

.....

$$x_{1s} = - \frac{1}{\sqrt{\pi}} \int_0^\tau \frac{\Gamma \frac{\partial x_2'}{\partial z} \Big|_s - \frac{\sqrt{\pi\tau'}}{\Gamma} \exp(x_{1s}')}{\sqrt{\tau-\tau'}} d\tau' \quad (22)$$

$$x_{1s} = \frac{1}{\sqrt{\pi}} \int_0^\tau \frac{\frac{\partial x_2'}{\partial z} \Big|_s}{\sqrt{\tau-\tau'}} d\tau' \quad (23)$$

Combining these equations to eliminate $\partial x_2'/\partial z \Big|_s$ and defining a new variable $\sigma = \tau / \{\Gamma(1+\Gamma)\}$ the following integral equation for x_{1s} is derived

$$x_{1s} = \int_0^\sigma \frac{\sqrt{\sigma'}}{\sqrt{\sigma-\sigma'}} \exp(x_{1s}') d\sigma' \quad (24)$$

Equation (24) has been numerically integrated by a procedure paralleling the one used in reference (8). The results of this integration are shown in figure (1). Runaway in surface temperature is found to occur at a finite ignition time $\sigma_{ign} = 0.393$. In dimensional variables the ignition time is given by

$$t_{ign} = 0.393 \frac{c(T_{go} - T_j) \Gamma(1+\Gamma)}{AQ} \exp(E/RT_j) \quad (25)$$

where T_j is the jump temperature, given by

$$T_j = T_o + (T_{go} - T_o) \frac{\Gamma}{1+\Gamma} \quad (26)$$

when the thermal responsivities are constant. Since the ignition time is strongly dependent on T_j the value of T_j should be correctly computed taking into account the temperature dependence of the thermal responsivities as was done in reference (8).

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NOMENCLATURE

A Pre-exponential factor

c Specific heat at constant pressure

E Activation Energy

Q Heat released by the reaction per unit mass of reactant

R Universal gas constant

T Temperature

t Time

x Space coordinate in the condensed phase

z Stretched space coordinate $z = \beta \xi$

β Dimensionless activation energy $\beta = E(T_j - T_o) / RT_j^2$

Γ Ratio of thermal responsivities $\Gamma = \sqrt{(\rho \lambda c)_g / (\rho \lambda c)}$

θ Dimensionless temperature $\theta = (T - T_j) / (T_{go} - T_j)$

λ Thermal conductivity

ξ Dimensionless space coordinate:

$$\text{Condensed phase } \xi = -x \sqrt{\rho A Q \exp(-E/RT_j) / \{\lambda (T_{go} - T_j)\}}$$

$$\text{Gaseous phase } \xi = \psi \sqrt{Q A c_g \exp(-E/RT_j) / \{\rho_g \lambda_g c (T_{go} - T_j)\}}$$

ρ Density

σ Reduced time $\sigma = \tau / \{\Gamma(1 + \Gamma)\}$

τ Dimensionless time $\tau = t A Q \exp(-E/RT_j) / \{c (T_{go} - T_j)\}$

X Reduced temperature defined in Eq(7)

χ Reduced temperature defined in Eqs(6) and (8)

Ψ Mass coordinate $\Psi = \int_0^x \rho_g dx$

SUBSCRIPTS

g Gas

I Inert

j Jump value

s Surface

o Initial

1 First order perturbation

2 Second order perturbation

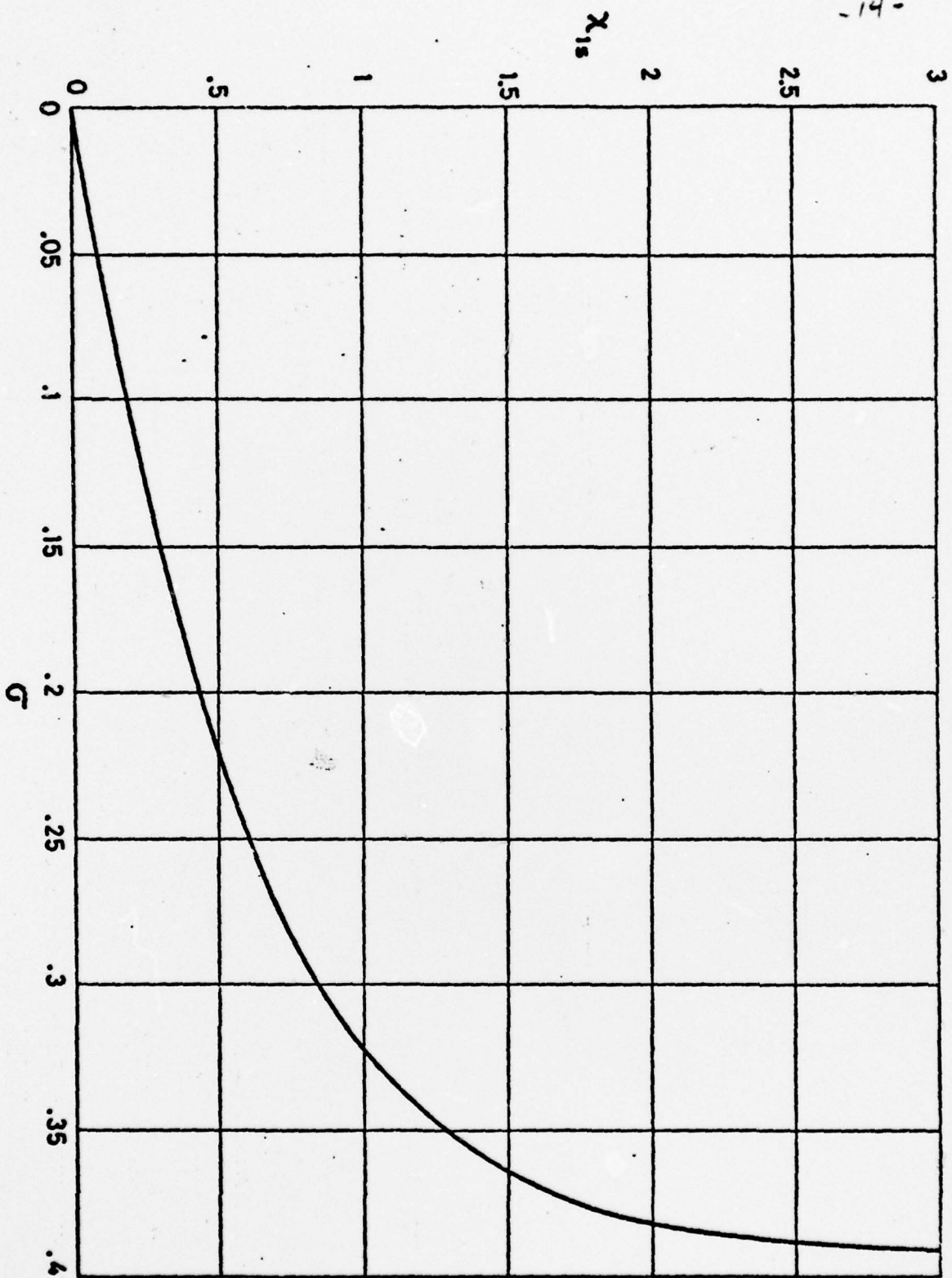


Fig.1.- Surface temperature $X_{1s} = (T_s - T_1)E/RT_1^2$ as a function of the nondimensional time σ .