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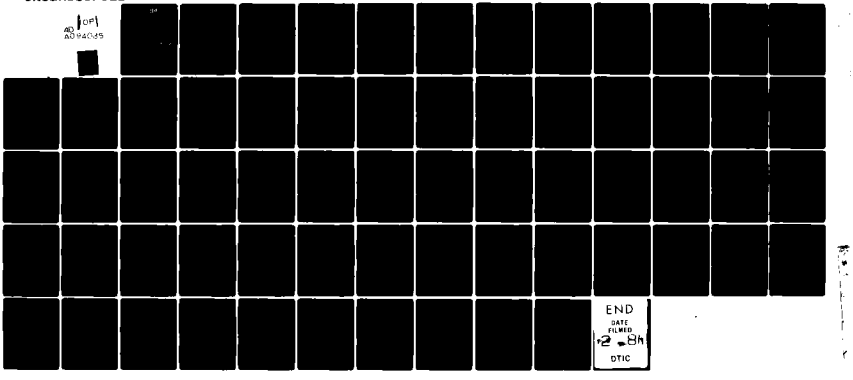

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ASYMPTOTIC TECHNIQUES IN IGNITION THEORY

Final Technical Report

by

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Madrid - 3 (SPAIN)

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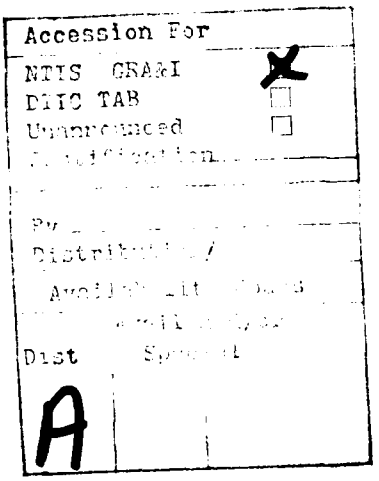

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Final Report for Grant DAERO 79-G-0007

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ABSTRACT

An analysis is given in Part I for the ignition of a reactive material by an electrical wire heated at constant power. The wire temperature history is described for small and large values of the electrical heating rate and for solid and gaseous reactants; in the case of gaseous reactants the effects of convection have also been taken into account. The results of the analysis include closed form expressions for the ignition time in terms of the physicochemical parameters.

An analysis is given in Part II of the process of ignition of reacting particles in a shock tube. An analysis for large activation energy of the ignition reaction shows that for values of the particle diameter smaller than a critical value, ignition does not take place; the ignition time is calculated for diameters larger than the critical.

Analysis of ignition by hot catalytic surfaces and by inert hot spots, and of Lewis number effects on the structure and extinction of diffusion flames due to strain, carried out under this Grant are being published elsewhere.

INTRODUCTION

This report describes the results of applying the technique of large activation energy asymptotics, to several ignition problems of practical interest. This technique, which has been extremely useful in studying many problems in combustion, makes possible the derivation of closed form analytical solutions for the ignition time or other properties of interest, although the original equations describing the problem are complicated non-linear, partial differential equations.

The first part of this report studies the ignition process of a reactive material when an imbedded electric wire is heated by an electric current of constant power. The wire heats up the surrounding reactive material by conduction, until the exothermic reaction leads to a thermal runaway at a finite ignition time. Since the heat conductivity of the wire is often very large compared to that of the reactive medium, the wire temperature is assumed to be uniform. The character of the process is then determined by the ratio of the specific heats per unit volume of the reactive material and of the heating wires; when this ratio is of order unity, the radius of the heated region in the reactive material during the ignition transient is of the order of the wire radius, while if the ratio is small, the radius of the heated region becomes large compared with the wire radius. In the

first case, the chemical heat release is confined to a thin layer adjacent to the wire surface, and it can be calculated in terms of the wire temperature and the heat loss to the surrounding material where the chemical reaction is frozen; the surface temperature and the ignition time are given by the solution of an ordinary differential equation, involving a single parameter. In the second case, which is typical when igniting reacting gases, the characteristic thickness of the reaction region is of the order of the particle radius, and the radius of the heated region is large; therefore forced or free convection effects can be important and have to be retained when analyzing the outer frozen zone of the reactive medium.

The second part of this report studies the process of igniting reacting particles in a shock tube. The particles, which are located in a holder in the shock tube, are dragged, accelerated, and heated by a shock wave. The wave is reflected at the end of the tube, so that when it encounters again the particles, these are decelerated and further heated by the gas behind the reflected shock. In this report we analyze motion and heat transport to the particles as a function of the parameters of the shock wave, and we apply the method of large activation energy asymptotics to study the ignition process. There is a critical value of the diameter of the particles such that for $D<D_{crit}$ ignition does not

occur due to rapid heat transfer from the particle. For $D>D_{crit}$ there is a runaway of surface temperature at a finite ignition time. Figures are provided to compute the ignition time as a function of the parameters of the problem.

An analysis has been carried out under this Grant of the ignition by a hot catalytic surface, taking into account the effect of a nonzero rate for a catalytic consumption of fuel at the plate surface. The analysis leads to an integral equation that describes the evolution of the fuel mass fraction at the surface. A note with the tittle "Ignition by a Hot Catalytic Surface" summarizing this work was written by A. Liñán and Forman A. Williams of the University of California, San Diego. It will appear in SIAM J. of Applied Mathematics.

We have also studied during the term of this contract, the process of ignition of a reactive material by an inert hot spot. The result of this research was presented at the 7th Colloquium on Gasdynamic of Explosions and Reactive Systems in Götingen, August 20-24, and will appear in the Series Progress in Aeronautics and Astronautics.

Finally, an analysis of the "Lewis number effects on the structure and extinction of diffusion flames due to strain" was presented by A. Liñán at the International Conference on the Role of Coherent Structures on Modelling of Turbulence and Mixing, held at the IBM Scientific Center, Universidad Autónoma de Madrid, July 25-27, 1980. The Proceedings will appear in the Lecture Notes on Physics, Springer.

IGNITION OF A REACTIVE MATERIAL BY AN ELECTRICAL WIRE

Part I of the Final Technical Report
for Grant DAERO 79-G-0007

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ABSTRACT

An analysis is given for the ignition of a reactive material by an electrical wire heated at constant power. The wire temperature history is described for small and large values of the electrical heating rate and for solid and gaseous reactants; in the case of gaseous reactants the effects of convection have also been taken into account. The results of the analysis include closed form expression for the ignition time in terms of the physicochemical parameters.

1. INTRODUCTION

Ignition of exothermic materials by hot wires has been extensively studied both theoretically[1,2] and experimentally,[3,4] in order to improve the design of squibs and detonators.

Large activation energy asymptotics have been very useful in describing thermal ignition with different types of heating mechanisms. These methods are used in the present report to analyze hot wire ignition for different values of the parameters of the problem, including the cases of gaseous and condensed exothermic material.

Previous theoretical analyses of ignition of solid materials by wire heating, include the studies of Altman[4] and Grant and Kabik et al.[4], who used the concept of a fixed ignition temperature to correlate their experimental results.

Friedman[1] carried out a theoretical analysis based on the more fundamental kinetic parameters of activation energy and frequency factor, rather than the more ill-defined ignition temperature. The ignition criterion used minimizes the sum of the time to reach temperature T by the inert solution and the explosion time at temperature T. An explicit expression for the ignition time is derived from a simple approximate expression of the inert temperature. This solution is in good agreement with the results of numerical integrations.

Thermal runaway leading to ignition is a strongly dependent function of temperature. Thus, as it will be found later, an analysis such as Friedman's, which correctly describes the inert temperature rise prior to ignition and, approximately, the temperature at which thermal runaway occurs is usually adequate to describe the ignition time to leading order.

The following analysis is an approximate solution of the hot wire ignition problem which is "exact" from an asymptotic point of view. In addition, it includes several ranges of the parameters which are not covered by the approximate analysis of Friedman.

The problem that we consider is that of a cylindrical wire imbedded in a reactive material. A constant rate of thermal energy is supplied to the wire, for instance by an electric current, so that its temperature and the temperature of the surrounding material increase with time.

Initially, the temperatures are low, so that the chemical reaction is frozen. Thus, the temperature profiles are simply given by the solution of the inert problem. However, after a certain time the energy input by the chemical reaction becomes important so that the temperature of the reactive material is further increased. This in turn accelerates the reaction rate, thus leading to ignition, represented by a precipitous rise in temperature, at a finite time. The pur-

pose of this report is to compute this ignition time as a function of the parameters of the problem.

In Section (2) the problem of ignition of reacting-solids by wires is formulated in non-dimensional variables, and the significance of the resulting parameters is discussed. In Section (3) the inert solution is analyzed for several parameter's ranges. These solutions are then used in Section (4) to analyze ignition and in particular to compute the ignition time for several cases. The procedures used in Section (4) may be easily generalized in order to analyze cases in which the values of the parameters are outside the range of validity of the cases presented in Section (4).

In practice, convection is usually important in the case of fluid reactants, so that the ideal solutions presented in Sections (3) and (4) are of little applicability to the case of reacting fluids. In fact, either forced convection is present, or else the temperatures increments associated to the wire are enough to generate a free convective flow which can not be neglected. Thus, in Section (5) the analysis is generalized to include the effect of forced convection, and we indicate how free convective effects can be taken into account. Finally, in Section (6) the results obtained in the report are summarized and discussed.

2. FORMULATION

We consider the case in which a cylindrical wire

imbedded in a reactive material, is heated by circulating an electric current of constant power through it. The thermal conductivity of the wire is considered to be large compared to the conductivity of the reactive material, and therefore its temperature is uniform throughout the process.

A distributed, zero order, exothermic reaction of the Arrhenius type takes place in the reactive material and produces ignition at a finite ignition time.

Assuming constant values for the density ρ, specific heat c, and heat conductivity k, the energy conservation equation in the reactive material becomes

$$A \exp\{-\beta'/(\theta+1)\} + \frac{\partial^2\theta}{\partial r^2} + \frac{1}{r}\frac{\partial\theta}{\partial r} = \frac{\partial\theta}{\partial t} \quad , \qquad (1)$$

where all symbols are defined in the nomenclature. The temperature increment θ is measured with respect to the initial temperature T_o, the space coordinate r with respect to the wire radius R, and the time variable t, with respect to the characteristic heat conduction time (R^2/α_e).

The boundary and initial conditions are

$$\theta(t,\infty) = \theta(0,r) = 0 \qquad (2)$$

$$\delta = \frac{d\theta_s}{dt} - 2\varepsilon \left.\frac{\partial\theta}{\partial r}\right|_s \qquad (3)$$

where the second equation represents a balance between electric energy input, heat absorbed by the wire and heat conducted to the reactive material. The parameter

$$\delta = \frac{B R^2}{K_e T_o} \frac{(\rho c)_e}{(\rho c)_i} \qquad (4)$$

is a ratio between the characteristic heat conduction time and the characteristic heat up time of the wire, and the parameter

$$\varepsilon = \frac{(\rho c)_e}{(\rho c)_i} \qquad (5)$$

is the ratio between the volumetric heat capacity of the reactive material and that of the wire.

Equations (1)-(3) have to be solved in order to obtain the temperature history and the ignition time as a function of the four parameters A, β', δ and ε. The form of the solution depends on the relative magnitude of these parameters. We first analyze in Section 3 the inert solution for different values of δ and ε, and in Section 4, we use this inert solution to analyze ignition in the limit of high activation energy.

Notice that ε is of order unity for ignition of solids and liquids by metal wires, while $\varepsilon << 1$ for ignition of reacting gases. The value of δ depends on the power of the ignition source and the wire radius; δ becomes small for thin wires.

3. INERT SOLUTION

In this section we analyze the process of heating

a material by supplying a constant rate of thermal energy input through a wire of infinite length and large conductivity imbedded in the material.

This process is described by equations (1)-(3) with A=0. The solution[5] can be obtained by means of the Laplace transform technique, for instance, and it may be written as

$$\theta = \frac{2\delta}{\pi} \int_0^\infty \frac{1-\exp(-u^2 t)}{u^2} \frac{\Sigma}{\Delta} \, du \qquad (6)$$

with

$$\Sigma = J_o(ru)\left|uY_o(u)-2\varepsilon Y_1(u)\right| - Y_o(ru)\left|uJ_o(u)-2\varepsilon J_1(u)\right| \qquad (7)$$

$$\Delta = \left|uJ_o(u)-2\varepsilon J_1(u)\right|^2 + \left|uY_o(u)-2\varepsilon Y_1(u)\right|^2 \qquad (8)$$

In particular, the surface temperature is given by

$$\theta_s = \frac{8\delta\varepsilon}{\pi^2} \int^\infty \frac{1-\exp(-u^2 t)}{u^3 \Delta} \, du \qquad (9)$$

which for small and large times results respectively in

$$\theta_s = \delta\left(t - \frac{8\varepsilon}{3\sqrt{\pi}} t^{3/2} + \ldots\right) \qquad (10)$$

$$\theta_s = \frac{\delta}{4\varepsilon}\left(\ln 4t - \gamma + \frac{1}{4t} + \ldots\right) \qquad (11)$$

where γ is Euler constant, $\gamma=0.5772$. Figure (1) shows the value of $(4\varepsilon\theta_s / \delta)$ for different values of ε.

This inert solution is valid up to the time in which the surface temperature increment is such that the reaction term becomes of the order of the heat transport

terms. When that temperature increment is reached, an ignition stage follows in which small temperature increments of order (β'^{-1}) produce changes of order unity in the reaction rate.

The inert surface temperature increment required to start the ignition stage, is generally of the order of the initial temperature, and therefore in this section we analyze the inert solution for times such that θ_s is of order unity. In the following paragraphs we study the inert solution for different asymptotic values of δ and ε, and in each case we obtain the characteristic times necessary to develop temperature increments of the order of the initial temperature.

a) $\delta=O(1)$, $\varepsilon=O(1)$

The surface temperature is given by (9) and the heat up time is of the order of the heat diffusion time, $t=O(1)$. Figure 1 may be used to compute θ_s as a function of δ and ε.

b) $\delta=O(1)$, $\varepsilon<<1$

In this case the surface temperature is given to first order by $\theta_s=t\delta$ and therefore $t=O(1)$. The temperature profile in the reactive material is given by the solution of the heat conduction equation with specified surface temperature.

c) $\delta=O(1)$, $\varepsilon>>1$

In this case $t>>1$, and the solution to equation

(1) is obtained by matching a quasi-steady solution close to the wire, to an unsteady solution far from it. This type of solution exists in all cases in which $t>>1$; that is, when the heat conduction region is large compared to the wire radius.

In the quasi-steady region, in which the time derivative term of equation (1) is negligible, the temperature profile is given by

$$\theta = -\frac{q}{2\pi} \ln r + \theta_s \quad , \tag{12}$$

where q is the rate of heat loss from the wire to the reactive material, $q=-(\partial\theta/\partial r)_s$.

Far from the wire, there is an unsteady region in which the temperature profile is given by

$$\theta = \frac{1}{4\pi} \int_o^t \frac{q}{t-t'} \exp\left(-\frac{r^2}{4(t-t')}\right) dt' \tag{13}$$

Matching this solution for small values of r to the quasi-steady solution, provides a relationship between the heat flux from the wire to the reactive material q, and the surface temperature θ_s, namely

$$\theta_s = \frac{q}{4\pi} (\ln 4t - \gamma) + \frac{1}{4\pi} \int_o^t \frac{q(t-t')-q(t)}{t'} dt' \tag{14}$$

Eq. (14) would be used together with

$$\frac{d\theta_s}{dt} = \delta - \frac{\varepsilon q}{\pi} \tag{14'}$$

and $\theta_s(0)=0$, to calculate the wire temperature history, when-

q is small, so that ignition occurs at large times.

The last term in Eq. (14) is negligible if the relative changes in q are small compared with unity; then $\theta_s(t)$ is given by

$$\frac{d\theta_s}{dt} = \delta - \frac{4\varepsilon\theta_s}{\ln 4t-\gamma} \quad . \tag{15}$$

In the case considered here, $\delta=O(1)$, $\varepsilon>>1$, the power supplied to the wire is mostly used in heating the reactive material; the term $d\theta_s/dt$ is negligible in Eq. (14'), so that q is approximately constant, $q=\pi\delta/\varepsilon$, and

$$\theta_s = \frac{\delta}{4\varepsilon}(\ln 4t-\gamma) \quad . \tag{16}$$

The characteristic heat up time is of the order of $(\exp(\varepsilon))$.

d) $\underline{\delta>>1,\ \varepsilon=O(1)}$

In this case there is a quasi-similarity solution

$$\theta = \tau\theta_1(\eta) + \frac{\tau^{3/2}}{\sqrt{\delta}}\theta_2(\eta) + \ldots \tag{17}$$

where

$$\tau = t\delta \quad , \qquad \eta = \frac{r-1}{2\sqrt{\tau}} \tag{18}$$

$$\theta_1(\eta) = 4\, i^2\, \mathrm{erfc}(\eta) \tag{19}$$

$$\theta_2(\eta) = (3-4\varepsilon)i^3\,\mathrm{erfc}(\eta)+2\left(\eta\,\mathrm{erfc}\,\eta - \frac{\exp(-\eta^2)}{\sqrt{\pi}}\right)$$

Therefore, the surface temperature is given by Eq. (10) and the characteristic heat up time is of order (δ^{-1}).

e) $\delta >> 1$, $\varepsilon = O(\delta)$

Taking the limit $\varepsilon \to \infty$ in Eq. (9), the surface temperature simplifies to

$$\theta_s = \frac{2\delta}{\varepsilon\pi^2} \int_0^\infty \frac{1-\exp(-u^2 t)}{u^3 \left| J_1(u)^2 + Y_1(u)^2 \right|} \, du \qquad (21)$$

The surface temperature in this case may be obtained from figure 1, where the value $4\varepsilon\theta_s/\delta$ is given as a function of ε. The characteristic heat up time is of the order of the heat conduction time, $t = O(1)$.

f) $\delta << 1$, $\varepsilon = O(1)$

This case is analogous to e) and the solution is similarly obtained by matching a quasi-steady region to an unsteady region far from the wire. Since $\delta << 1$ and $\varepsilon = O(1)$ all the energy supplied by the wire is used to heat the reactive material, and therefore $q = \text{const}$. Then Eqs. (14) and (14') simplify again to Eq. (16). The characteristic heat up time is of order $(\exp(\delta^{-1}))$.

g) $\delta << 1$, $\varepsilon = O(\delta \ln \delta) << 1$

As in the previous case, it is necessary to wait times long compared to the characteristic heat conduction time, in order to develop temperature increments of the order of the initial temperature. Therefore, the surface temperature history is obtained by matching a quasi-steady region in which the temperature profile is given by Eq. (12) to an

unsteady region described by Eq. (13); the wire temperature is given by Eqs. (14) and (14'). However, in this case, the values of δ and ε are such that the energy used to heat up the wire is of the same order than the energy used to heat up the reactive material, and therefore all three terms in the surface energy balance given by Eq. (3), or Eq. (14'), have to be retained.

Because we have to wait times of order $1/\delta$ for θ_s to have increments of order unity, it turns out that the last term in Eq. (14) is small, of order $-1/\ln\delta$, relative to the remaining terms, and thus q is given to leading order by

$$q = 4\pi\theta_s/\ln t \quad , \tag{22}$$

and θ_s is given by Eq. (15), which using the definitions

$$\tau = t\,\delta \quad , \qquad \lambda = -\frac{4\varepsilon}{\delta \ln \delta} \tag{23}$$

simplifies to

$$\frac{d\theta_s}{d\tau} + \lambda\,\theta_s = 1 \quad ; \tag{24}$$

therefore

$$\theta_s = \lambda^{-1}(1-\exp(-\lambda\delta t)) \quad . \tag{25}$$

The characteristic heat up time t is of order (δ^{-1}).

h) $\underline{\delta<<1,\ \varepsilon=O(\delta)<<1}$

This case can be considered a particular case of the previous one when $\lambda<<1$ so that in first approximation all the thermal energy supplied through the wire is used to heat

up the wire, and $\theta_s=\delta t$; when two terms are retained in an expansion of Eq. (25) for small λ, the surface temperature is given by

$$\theta_s = \delta t\left(1 + \frac{2\varepsilon t}{\ln\delta}\right) \quad . \qquad (25')$$

The characteristic heat up time is of order δ^{-1}.

4. IGNITION STAGE

In this Section we analyze this ignition stage for $\beta'>>1$, using asymptotic methods in the limit of high activation energy β'.

Initially, there is an inert stage during which the reaction term in Eq. (1) is negligible, and the inert solution analyzed in Section 3 is valid. After this stage there follows an ignition stage in which the temperature of the reactive material is such that the Arrhenius reaction term becomes of the same order than the transport terms. Then small temperature increments of order $1/\beta'$ produce changes of order unity in the reaction term; the energy released by the exothermic reaction further increases the temperature of the material This self-accelerating process leads to ignition, characterized by a precipitous rise in temperature at a finite time.

As discussed in the previous Section there are different cases depending on the relative values of the parameters δ and ε. We will carry out detailed analyses of six of these cases to show the methodology used and to discuss the

different structures of the solutions. Similar procedures may be used to calculate the ignition time in other cases.

a) $\delta=O(1)$, $\varepsilon=O(1)$

In this case the inert temperature of the reactive material increases by a quantity of the order of the initial temperature for times of the order of the characteristic heat conduction time (see Section 3.a). For those times, $t=O(1)$, the size of the heated region in the reactive material is of the order of the wire radius (Figure 2).

From Eq. (1) it is easy to show that the characteristic ignition temperature θ_1, which makes the reaction term of the same order than the transport terms is given by

$$\exp\{\beta'/(\theta_1+1)\}=A/\delta \quad . \tag{26}$$

Introducing this definition and using for convenience as new variables

$$\tau=\delta t \quad , \qquad x=(r-1)\sqrt{\delta} \quad . \tag{27}$$

Equations (1) and (3) become

$$\frac{\partial\theta}{\partial\tau}=\frac{\partial^2\theta}{\partial x^2}+\frac{1}{\sqrt{\delta}+x}\frac{\partial\theta}{\partial x}+\exp\left(\frac{\beta'}{\theta_1+1}\,\frac{\theta-\theta_1}{\theta_1+1}\right) \tag{28}$$

$$1=\frac{d\theta_s}{d\tau}-\frac{2\varepsilon}{\sqrt{\delta}}\left.\frac{\partial\theta}{\partial x}\right|_s \quad . \tag{29}$$

In the limit of high activation energy, the reaction is frozen for $\theta<\theta_1$, while it goes to infinity for $\theta>\theta_1$. To leading order, the ignition time is simply given by τ_1, the time at

which the inert surface temperature reaches θ_1. Equation (9) or Figure (1) may used to compute τ_1 as a function of δ, ε and θ_1, which in turn is related to A and β' through Equation (26).

For times smaller than τ_1 there is an inert stage which is followed by a short transition stage close to τ_1, which leads to a runaway of surface temperature.

During the transition stage there is an inner thin reaction zone close to the wire where the temperature differs from θ_1 by a small quantity of order (β^{-1}), where

$$\beta = \beta'/(\theta_1+1)^2 \ . \qquad (30)$$

This region has to be matched with an outer transient-diffusive zone whose size is of order $(\beta^{-.5})$.

To analyze the reaction zone let's define

$$\psi = \beta(\theta-\theta_I) \qquad (31)$$

and let's expand the inert temperature, θ_I, for times close to τ_1 and for small values of x

$$\theta_I = \theta_1+d_\tau(\tau-\tau_1)+d_x x+.... \qquad (32)$$

where the parameters

$$d_\tau = \left.\frac{\partial\theta_I}{\partial\tau}\right|_{\tau=\tau_1,x=0} \qquad d_x = \left.\frac{\partial\theta_I}{\partial x}\right|_{\tau=\tau_1,x=0} \qquad (33)$$

are obtained from Equations (3) and (9) as a function of δ and ε. Introducing (30)-(32) in the reaction term of Equation

(28) and expanding for large values of β, it is observed that the characteristic variables to analyze the reaction zone during the ignition transient are

$$\sigma-\sigma_1 = d_\tau \beta(\tau-\tau_1) \ , \qquad \xi = - d_x \beta x \tag{34}$$

where

$$\sigma_1 = - \ln(\beta d_x^2/2) \ . \tag{35}$$

With these definitions Equations (28) and (29) result in

$$2 \frac{\partial^2 \psi}{\partial \xi^2} = - \exp(\sigma+\psi-\xi) \tag{36}$$

$$p \frac{d\psi_s}{d\sigma} = \left.\frac{\partial \psi}{\partial \xi}\right|_s \tag{37}$$

where a single parameter appears,

$$p = - \frac{\sqrt{\varepsilon}}{2\varepsilon} \frac{d_\tau}{d\varepsilon} = - \frac{d_\tau(\tau_1)}{d_\tau(\tau_1)-1} \ . \tag{38}$$

Figure (3) shows $d_\tau/4\varepsilon$ as a function of τ for different values of ε.

Equation (36) has to be solved with the boundary condition (37) and with the condition obtained from matching to the outer transient-diffusive zone, namely

$$\lim_{\xi \to \infty} \frac{\partial \psi}{\partial \xi} = 0 \ . \tag{39}$$

Integrating (36) once, and using (39) yields

$$\psi_\xi = 1 - \sqrt{1-\exp(\sigma+\psi-\xi)} \ . \tag{40}$$

Evaluating this expression at the surface and using Equation (37) results in

$$P \frac{d\psi_s}{d\sigma} = 1 - \sqrt{1-\exp(\sigma+\psi_s)} \quad . \qquad (41)$$

Numerical integration of this equation provides the surface temperature history, and in particular the ignition time as a function of P, defined here as the time σ_{ign} when the expression inside the square-root becomes negative. During a short transient, when $\sigma-\sigma_{ign} \sim \beta^{-1}$, the time derivative term which was neglected in Equation (36) should be retained to describe the thermal runaway at the ignition time.

Figure (4) shows the surface temperature history for some values of P, and Figure (5) shows the ignition time as a function of (logP). These results may be approximated by the equation

$$\sigma = - \ln(1+.56/P)$$

which exhibits the correct asymptotic behaviour for small and large P.

Once $\psi_s(\sigma)$ is known from Equation (41), Equation (40) may be integrated to derive the temperature profile and in particular $\psi(\sigma,\infty)$, the apparent increase in wire temperature due to the chemical reaction.

b) $\underline{\delta >> 1,\ \varepsilon = O(1)}$

In this case, the characteristic inert time for a temperature increase of order unity is small, of order (δ^{-1}), compared to the characteristic heat conduction time

(see Section 3.d). Thus, the heated region in the reactive material is small of order ($\beta^{-.5}$), compared to the wire radius (see Figure 6).

The analysis of this case parallels that of the previous Section. As before, there is a transition stage whose structure is described by a thin reactive-diffusive zone close to the wire with size of order ($\beta^{-1}\delta^{-.5}$), which is matched to outer transient-diffusive zone with size of order ($\beta^{-.5}\delta^{-.5}$).

The equations to solve are (28) and (29), which simplify for δ large because then the convective term associated to curvature in Eq. (28), and the energy spent in heating the reactive material in Eq. (29) are negligible, as may be observed in Figure (6).

Introducing the stretched variables (31) and (34) in Eqs. (28) and (29) with

$$\tau_1=1 \ , \qquad d_\tau=1 \ , \qquad d_x=-2\sqrt{\tau_1/\tau} \ , \qquad (42)$$

as obtained from Eqs. (17)-(19), results in Eqs. (36) and

$$\frac{d\psi_s}{d\sigma}=0 \quad \Longrightarrow \quad \psi_s=0 \ . \qquad (43)$$

Integrating (36) once and using conditions (38) and (43) yields

$$\psi_\xi(\sigma,0) = 1 - \sqrt{1-\exp(\sigma)} \qquad (44)$$

and therefore the ignition time may be taken as $\sigma_{ign}=0$, or

$$t_{ign} = \frac{\theta_1}{\delta} + \frac{\ln(2\beta\theta_1/\pi)}{\beta\delta} \quad , \qquad (45)$$

where θ_1 is a function of β, A and δ given by Eq. (26)

c) $\underline{\delta<<1,\ \varepsilon=O(1),\ \beta'=O(\delta^{-1})}$

This condition which corresponds to case (f) of the previous Section is used to introduce the analyses of those cases in which $t_{ign}>>1$, and therefore the structure of the solution consists of a quasi-steady zone close to the wire and an unsteady zone far from it.

Contrary to the previous cases, the reaction zone is not thin compared to the wire radius. However, since the unsteady effects are only important far from the wire, where the temperatures are low and therefore the reaction frozen, the chemical reaction is confined to a thick quasi-steady reaction zone.

To analyze this case, let's introduce

$$\beta = \frac{\beta'}{(\theta_s+1)^2}, \quad \phi = \beta(\theta-\theta_s), \quad K = A\,\beta \exp\left(-\frac{\beta'}{(\theta_s+1)}\right) \qquad (46)$$

where ϕ is of order unity in the reaction zone. Thus, Eq. (1) in the limit of high activation energy simplifies to

$$\frac{\partial^2\phi}{\partial r^2} + \frac{1}{r}\frac{\partial\phi}{\partial r} + K\exp(\phi) = 0 \qquad (47)$$

which replaces in this case the Eq. (36) describing the reaction zone structure in the previous case. To solve this

equation, notice that it is invariant under the group of transformations

$$r \to \alpha r \,, \quad \phi \to \phi + \beta, \quad \alpha^2 \exp(\beta) = 1 \quad . \tag{48}$$

Thus, let's define new variables which are invariant under those transformation

$$\omega = r \frac{\partial \phi}{\partial r} \,, \quad u = K r^2 \exp(\phi) \tag{49}$$

With these variables, Eq. (47) results in

$$(2+\omega) \frac{d\omega}{du} + 1 = 0 \tag{50}$$

which may be integrated once to yield

$$\omega^2 + 4\omega + 2u = \omega_\infty^2 + 4\omega_\infty \tag{51}$$

since in the outer unsteady region for $r \to \infty$, the chemical heat release approaches zero ($u \to 0$). The value of ω_∞, can be written in terms of the heat flux q from the reaction zone towards the thicker unsteady heat conduction region. Because the changes in temperature across the reaction region are small, of order $1/\delta$, the matching procedure, as discussed in Section (3.c), results in a relationship between the surface temperature θ_s and the heat flux, $q = -2\pi\omega_\infty/\beta$, given by Eq. (14).

Because the ignition time t is long, the last term in Eq. (14) can be neglected in first approximation and then we can write

$$\omega_\infty = -\frac{2\beta\theta_s}{\ln 4t - \gamma} \tag{52}$$

although the terms $\ln 4-\gamma$ in the denominator of Eq. (52) are not always consistent with the order of approximation used. The boundary condition at the surface (Eq. (3)) results in

$$\omega_s = \left.\frac{\partial \phi}{\partial r}\right|_s = \frac{\beta}{2\varepsilon}\left(\frac{d\theta_s}{dt} - \delta\right), \qquad u = K \tag{53}$$

From (50) and (53) an equation is derived to describe the surface temperature evolution, namely

$$\omega_s = -2 \pm \sqrt{(2+\omega_\infty)^2 - 2K} \tag{54}$$

or

$$\frac{\beta'}{4\varepsilon(\theta_s+1)^2}\left(\delta - \frac{d\theta_s}{dt}\right) =$$

$$= 1 - \sqrt{\left[1 - \frac{\beta'}{(\theta_s+1)^2}\,\frac{\theta_s}{\ln 4t-\gamma}\right]^2 - \frac{A}{2}\,\frac{\beta'}{(\theta_s+1)^2}\exp\left(-\frac{\beta'}{1+\theta_s}\right)} \tag{55}$$

This equation describes the surface temperature evolution in all cases in which ignition occurs for times large compared to the heat conduction time, and therefore the structure of the heated region in the reactive material is described by a quasi-steady reaction zone near the wire, and an unsteady frozen region far from it.

In the particular case $\delta << 1$, $\varepsilon = O(1)$, $\beta' = O(\delta^{-1})$, the heat absorbed by the wire is negligible compared to the heat supplied to the reactive material, while the chemical heat release is of order δ compared with the external heat input, when the inert reaches a value θ_1 given by

$$\exp\left(\frac{\beta'}{1+\theta_1}\right) = \frac{A}{\delta^2} \quad . \tag{56}$$

From Section (3.f), the characteristic ignition time necessary to reach θ_1 is of order $\left(\exp(\delta^{-1})\right)$, namely

$$t_1 = \frac{1}{4} \exp\left(\gamma + \frac{4\vartheta_1}{\delta}\varepsilon\right) \quad . \tag{57}$$

Introducing in Eq. (55)

$$\tau = t/t_1 \quad , \qquad \theta_s = \theta_{Is} + \phi/\beta \tag{58}$$

with

$$\theta_{Is} = \vartheta_1 + \frac{\delta}{4\varepsilon} \ln \tau \tag{59}$$

and taking the limit of high activation energy β, of order (δ^{-1}), so that the characteristic time in the transition stage is of the same order than the characteristic heat up time, results in

$$\Phi = Q\, \tau^b \exp(\Phi) \quad , \tag{60}$$

where

$$b = \frac{\beta\delta}{4\varepsilon} \quad , \qquad Q = \frac{\theta_1 \beta^2 \delta^2}{4b(b-1)} \quad . \tag{61}$$

This Equation represents a balance between the heat released by the reaction and the increment in heat flux to the outer region as a result of the surface temperature increment Φ. The heat flux from the wire, ω_s, is constant and balanced by the heat flux to the outer region associated to the surface temperature ϑ_1.

The parameter Q is always positive, since b has to be greater than one in order to have the reaction frozen far from the wire. In fact, from (49)

$$\phi = \omega_\infty \ln r + \text{const.} \qquad (62)$$

and since u is assumed to go to zero for $r\to\infty$ in order to write Eq. (51), it is necessary that in the limit $r\to\infty$,

$$u = K r^2 \exp(\phi) \simeq \lim K r^2 r^{\omega_\infty} \to 0 \qquad (63)$$

and therefore $\omega_\infty < -2$. For t large of order t_1, Equations (52) and (57) show that

$$\omega_\infty \simeq -\frac{\beta\delta}{2\varepsilon} \simeq -2b \qquad (64)$$

and therefore $b>1$.

Equation (60) describes the evolution of the surface temperature as a function of τ, and the parameters Q and b. As τ increases, Φ increases, until it reaches a critical value $\Phi=1$ after which the reaction releases heat at a rate too fast to be transmitted to the exterior, and therefore Eq. (60) has no solution. Thus, the ignition time is identified as the time at which $\Phi=1$, so that

$$\tau_{ign} = (Q\,e)^{-1/b} \quad . \qquad (65)$$

Through Eqs. (56) and (61) t_{ign} may be related to the parameters A, β', δ and ε.

d) <u>$\delta<<1$, $\varepsilon=O(1)$</u>

This case is analogous to the preceding one, but

since the characteristic time of the transient ignition stage is short compared to the heat up time, the inert solution may be expanded for times close to t_1, the characteristic time to reach the ignition temperature θ_1 given by Eq. (56). Thus,

$$\theta_s = \theta_1 + \frac{\delta}{4\varepsilon}(\tau-1) + \Phi/\beta \quad , \tag{66}$$

where τ is defined in Eq. (58).

Introducing this relationship in the Arrhenius exponent and expanding for large values of β', it is seen that the characteristic chemical time is of order $(\beta\delta)^{-1}$. Using as time variable

$$\sigma = \frac{\beta\delta}{4\varepsilon}(\tau-1) \tag{67}$$

and taking the limit $\beta\to\infty$ in Eq. (55) results in

$$\Phi = Q\exp(\Phi+\sigma) \tag{68}$$

where

$$Q = 4\theta_1\varepsilon^2 \quad . \tag{69}$$

Thus, the ignition time is simply given by

$$\tau_{ign} = 1 - \frac{4\varepsilon}{\beta\delta}(1+\ln Q) \tag{70}$$

which coincides with Eq. (65) in the limit $b\to\infty$ ($\beta\delta\to\infty$).

e) <u>$\delta<<1$, $\varepsilon=O(\delta\ln\delta)$, $\beta>>\ln\delta$</u>

This case correspond to the inert solution presented in Section)3.g) in which the parameter λ is of order unity. The nondimensional activation energy β' is considered

to be large compared to ($\ln\delta$). Therefore, the reaction zone is small compared to the quasi-steady zone, whose size is of order ($\ln\delta$).

Defining the characteristic ignition temperature θ_1, as

$$\exp\left(\frac{\beta'}{1+\theta_1}\right) = A\,(\ln\delta)^2/2\beta\theta_1^2 \qquad (71)$$

which makes the diffusion and reaction terms of the same order, and introducing (58) in the reaction term, with

$$\theta_{Is} = \theta_1+\delta\exp(-\lambda t_1\delta)\ (t-t_1) \qquad (72)$$

which is obtained by expanding (25) close to t_1, it is found that the characteristic variables in this case are Φ, as defined in (58) and

$$\sigma = \beta\delta\exp(-\lambda\delta t_1)\ (t-t_1)\ . \qquad (73)$$

The characteristic heat up time is of order (δ^{-1}), and therefore the length of the transition stage to ignition is of order (β^{-1}).

Introducing these variables in Eq. (55) and taking the limit $\delta\to 0$, $\beta'=O(\ln\delta)$, $\varepsilon=O(\delta\ln\delta)$, results in

$$\sqrt{1-\exp(\Phi+\sigma)} = 1+\frac{d\Phi}{d\sigma}\left(1-\frac{1}{\lambda\theta_1}\right) \qquad (74)$$

whose solution yields the evolution of the surface temperature with time, as a function of the parameters of the problem. It is observed that this equation coincides with Eq.

(41) if the parameter P is replaced by $(1-1/\lambda\vartheta_1)$, and if the new definitions of σ and Φ are used. Therefore figure (5) may be used to evaluate the ignition time.

f) $\delta \ll 1$, $\varepsilon = O(\delta)$

In this case the inert surface temperature is described by Eq. (25'), and the heat flux to the outer region, ω_∞, is negligible compared to the heat absorbed by the wire. This solution remains valid up to the time in which the heat released by the reaction equals the heat transmitted to the exterior, and therefore the heat flux from the wire, ω_s, becomes zero. This time is considered to be the ignition time, and is obtained by equating to zero the expression inside the square root in Eq. (55). Thus

$$\left(1-\frac{\beta'}{(\delta t_{ign}+1)^2}\frac{\delta t_{ign}}{1+t_{ign}}\right)^2=\frac{A}{2}\frac{\beta'}{(\delta t_{ign}+1)^2}\exp\left(-\frac{\delta'}{1+\delta t_{ign}}\right) \quad (75)$$

The solution of this equation provides t_{ign} as a function of the parameters A, β' and δ. After t_{ign}, the reaction zone ceases to be quasi-steady and Eq. (55) is no longer valid.

5. FORCED CONVECTION

The analyses presented in the preceding sections may be generalized to include the effect of fluid flow normal to the wire, when the Peclet number is small and the parameters δ and ε are such that increments in surface temperature

of the order of the initial temperature occur for times long compared to the characteristic heat conduction time.

Let x be the space coordinate in the direction of flow measured with respect to the wire radius, and let Pe<<1, be the Peclet number associated to the convective velocity U_∞. The Ossen form of energy conservation equation (1)

$$A \exp\{-\beta'/(\theta+1)\}+\nabla^2\theta = \frac{\partial\theta}{\partial t} + Pe\, \vec{v}\cdot\nabla\theta \qquad (76)$$

can be used in first approximation to describe convective effects. This equation has to be solved with conditions (2) and (3).

In Section (3) the inert solution in absence of convection is analyzed, and the characteristic time, t_c, necessary to achieve temperature increments of order of the initial temperature is obtained as a function of β and ε. As discussed in that Section, the structure of the temperature profile for times, t_c, large compared to the heat conduction time ($t_c>>1$), is composed of a quasi-steady region close to the wire and an unsteady region far from it where the temperatures are of order $(1/\ln t_c)$.

From Eq. (76) it is observed that the effect of convection depends on the relative magnitude of Pe and t_c. In fact, the convective terms in the unsteady zone are of order $(Pe/\sqrt{t_c})$, while the transient and diffusive terms are of order (t_c^{-1}). Thus, for $Pe = O(t_c^{-1/2})$ the convective and

unsteady terms have to be retained in the unsteady region for $r=O(t_c^{-1/2})$. For $Pe<<t_c^{-1/2}$, the effect of convection is negligible and the solutions presented in Sections (3) and (4) are valid. For $Pe>>t_c^{-1/2}$ the unsteady term in Eq. (76) is negligible and there is a quasi-steady solution in which the convective terms are balanced by the diffusive terms. For long times, this solution approaches a steady state in which the heat supplied by the wire is convected away by the flow.

To analyze the inert solution for $Pe>>t_c^{-1/2}$, let's introduce as new dependent variable

$$\phi = \vartheta \exp(-Pe\, x/2) \quad , \tag{77}$$

and let's assume that the time derivative term is negligible. Thus, Eq. (76) simplifies to

$$\frac{\partial^2\phi}{\partial r^2} + \frac{1}{r}\frac{\partial\phi}{\partial r} - \frac{Pe^2}{4}\phi = 0 \quad , \tag{78}$$

whose solution with conditions (2) and (3) may be written as

$$\theta = \frac{\left(\delta - \frac{d\theta_s}{dt}\right)}{Pe\ \varepsilon} \frac{K_o(rPe/2)\exp\{Pex(r-1)/2r\}}{K_1(Pe/2) - \frac{x}{r}K_c(Pe/2)} \quad . \tag{79}$$

Thus, the surface temperature history is given by

$$\theta_s = \frac{\delta}{A}\left(1-\exp(-At)\right) \quad , \tag{80}$$

where

$$A = Pe\,\varepsilon\left(\frac{K_1(Pe/2)}{K_o(Pe/2)} - x\right) \quad . \tag{81}$$

In the limit Pe→0,

$$A = 4\varepsilon\{\ln(16/Pe^2)-2\gamma\}^{-1} \qquad (82)$$

so that the characteristic time to reach the steady solution is of order $\{-\ln(Pe)/\varepsilon\}$. If δ/A is small compared to unity, the temperature increment of the steady state is small, and therefore no ignition occurs.

For $Pe=O(t_c^{-1/2})$, there is a quasi-steady region close to the wire in which both the convective and unsteady terms are negligible, and an unsteady zone far from it in which both effects have to be retained.

In the unsteady zone, Eq. (76) has to be solved and matched to the quasi-steady solution. Using, for instance, Laplace transforms the temperature profile in the unsteady region is found to be

$$\theta = \frac{\exp(Pex/2)}{4\pi}\int_0^t \frac{q(\tau)\exp\{-Pe^2(t-\tau)/4\}\exp\{-r^2/4(t-\tau)\}}{t-\tau}\,d\tau \qquad (83)$$

Evaluating this expression for small values of r, provides a matching condition to the quasi-steady region, namely

$$\theta_s = \frac{q(t)}{4\pi}(\ln 4t-\gamma)+\frac{1}{4\pi}\int_0^t \frac{q(\tau)\exp\{-Pe^2(t-\tau)/4\}-q(t)}{t-\tau}\,d\tau \qquad (84)$$

For q(t)=const., when for instance $\delta<<1$, $\varepsilon=O(1)$, so that all the heat supplied by the wire is used to heat up the reactive material, this condition simplifies to

$$\theta_s = \frac{q}{4\pi}\{\ln(16/Pe^2) - E_1(Pe^2t/4) - 2\gamma\} \quad . \qquad (85)$$

Eq. (84) or (85) substitutes Eq. (14) as the matching relationship that together with Eq. (14') is necessary to obtain the surface temperature history. For instance, in the case $\delta<<1$, $\varepsilon=O(1)$,

$$\vartheta_s = \frac{\delta}{4\varepsilon}\left\{\ln(16/Pe^2) - E_1(Pe^2\, t/4) - 2\gamma\right\} \qquad (86)$$

which in the limit $(Pe^2 t)\to 0$, coincides with Eq. (16), and in the limit $(Pe^2 t)\to\infty$ coincides with Eq. (80) with the parameter A given by Eq. (82).

The ignition analyses presented in Sections (3.c) -(3.f) may also be generalized to include the effect of convection. In fact, Eq. (55) is still valid to describe the surface temperature history, if the value ω_∞ given by Eq. (52) is substituted by the new matching relationship

$$\omega_\infty = -\frac{2\,\partial\vartheta_s}{\ln(16/Pe^2)-E_1(Pe^2 t/4)-2\gamma} \quad . \qquad (87)$$

6. CONCLUSIONS

The process of ignition of a reactive material by an imbedded electric wire through which an electrical current of constant power is applied, has been analyzed by means of high activation energy asymptotics.

It is found that the temperature history and therefore the ignition time is a function of four non-dimensional parameters: a Damkhöler Number A, an activation energy β',

an energy input δ and the ratio of volumetric heat capacities ε.

The problem has been analyzed in the limit of large β', and it is found that the solution exhibits different behaviour depending on the relative magnitudes of β', δ and ε. Some of these limiting solutions are derived in Section 4, leading to closed form solutions for the ignition time. The values of δ and ε considered cover both the cases of reactive solids and gases. The same methods presented in that Section may be used to obtain other limiting solutions.

Finally the effect of convection which should be important in the case of reacting gases, is analyzed in Section (5).

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LIST OF SYMBOLS

A , Dimensionless frequency factor, $zR^2Q\rho_e/T_oK_s$.

B , Thermal energy input to the wire per unit volume.

b , Dimensionless parameter defined in Eq. (61).

c , Heat capacity.

d_τ, d_x , Dimensionless parameters defined in Eq. (33).

K , Dimensionless frequency factor defined in Eq. (46).

k , Coefficient of thermal conductivity.

P , Dimensionless parameter defined in Eq. (38).

Pe , Peclet number.

Q , Dimensionless parameter defined in Eqs. (61) or (69).

q , Heat transfer per unit area by conduction.

R , Radius of the wire.

r , Dimensionless radial coordinate r/R.

T , Temperature.

t , Dimensionless time, $t\alpha_e/R^2$.

u , Dimensionless parameter defined in Eq. (15).

x , Dimensionless radial coordinate Eq. (27).

z , Frequency factor.

α , Thermal diffusivity.

β , Dimensionless activation energy defined in Eq. (30).

β' , Dimensionless activation energy, E/RT_o.

δ , Dimensionless parameter defined in Eq. (4).

ϵ , Dimensionless parameter defined in Eq. (5).

η , Similarity variable defined in Eq. (18).

θ , Dimensionless temperature, $(T-T_o)/T_o$.

λ , Dimensionless parameter defined in Eq. (23).

ξ , Stretched radial coordinate defined in Eq. (34).

σ , Stretched time defined in Eqs. (34), (67) or (73).

τ , Stretched time defined in Eq. (18).

Φ , Dimensionless temperature increment defined in Eqs. (60) or (68).

ϕ , Dimensionless temperature increment defined in Eqs. (46) or (77).

ψ , Dimensionless temperature increment defined in Eq. (31).

ω , Transformed variable defined in Eq. (49).

Subscripts

c , Characteristic variable.

e , Exterior conditions.

I , Inert.

i , Interior conditions.

s , Surface.

0 , Initial.

1 , Characteristic conditions at ignition.

∞ , Conditions far from the wire.

LIST OF FIGURES

1. Inert Surface Temperature History for several values of ε.

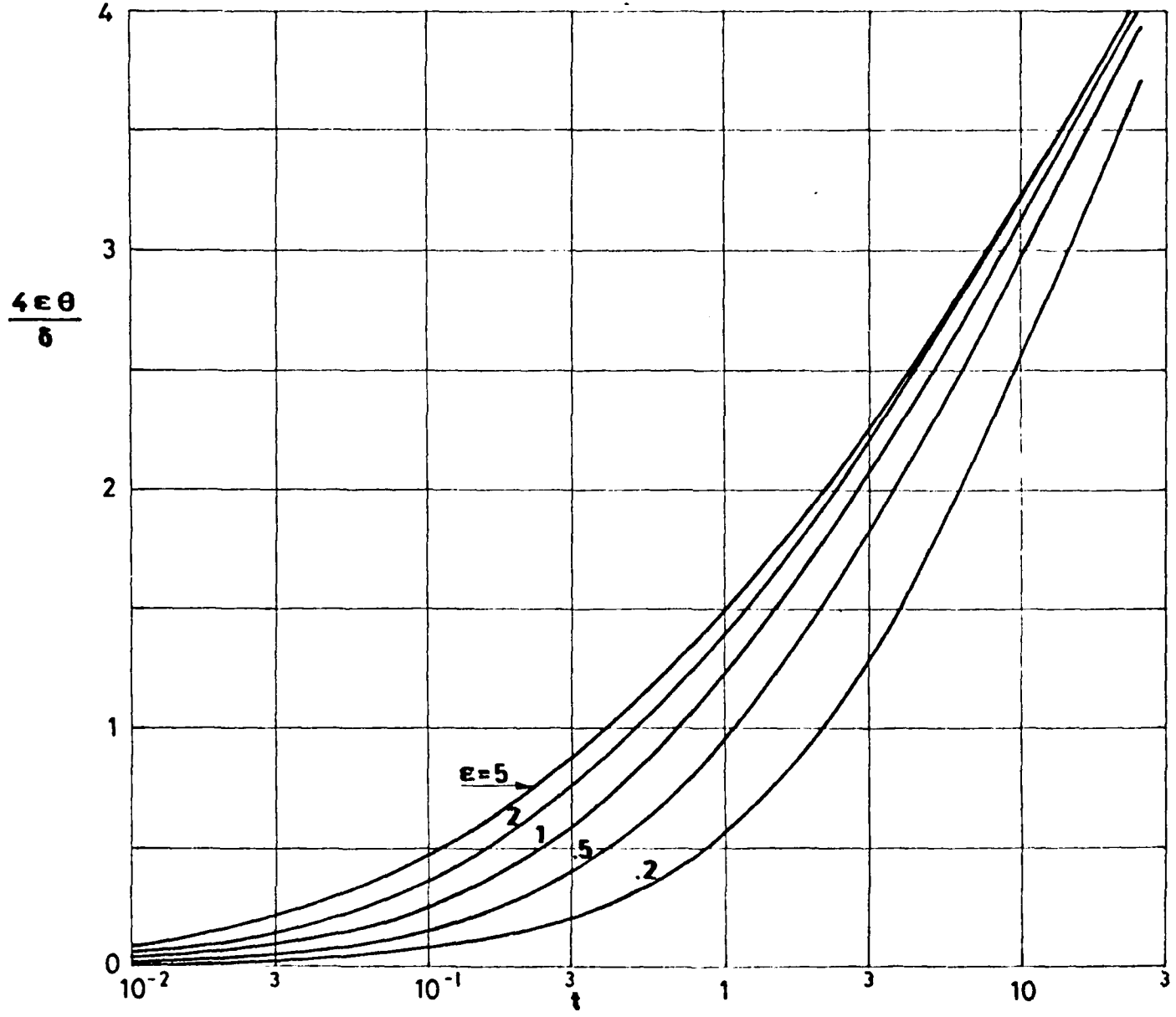
2. Schematic representation of the problem in the case that ε and δ are of order unity.

3. Time derivative at the surface of the inert temperature profile ($d\tau/4\varepsilon$) as a function of t for several values of ε.

4. Surface temperature history ψ_s for different values of the parameter P.

5. Ignition time σ_{ign} as a function of the parameter P.

6. Schematic representation of the problem in the case $\delta>>1$ and ε of order unity.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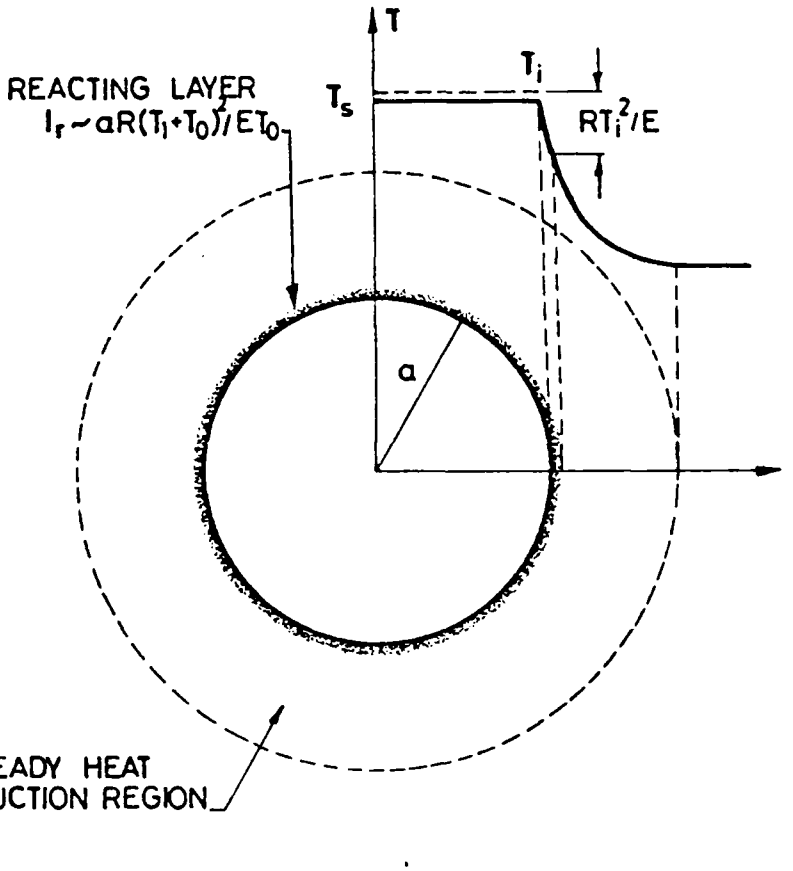
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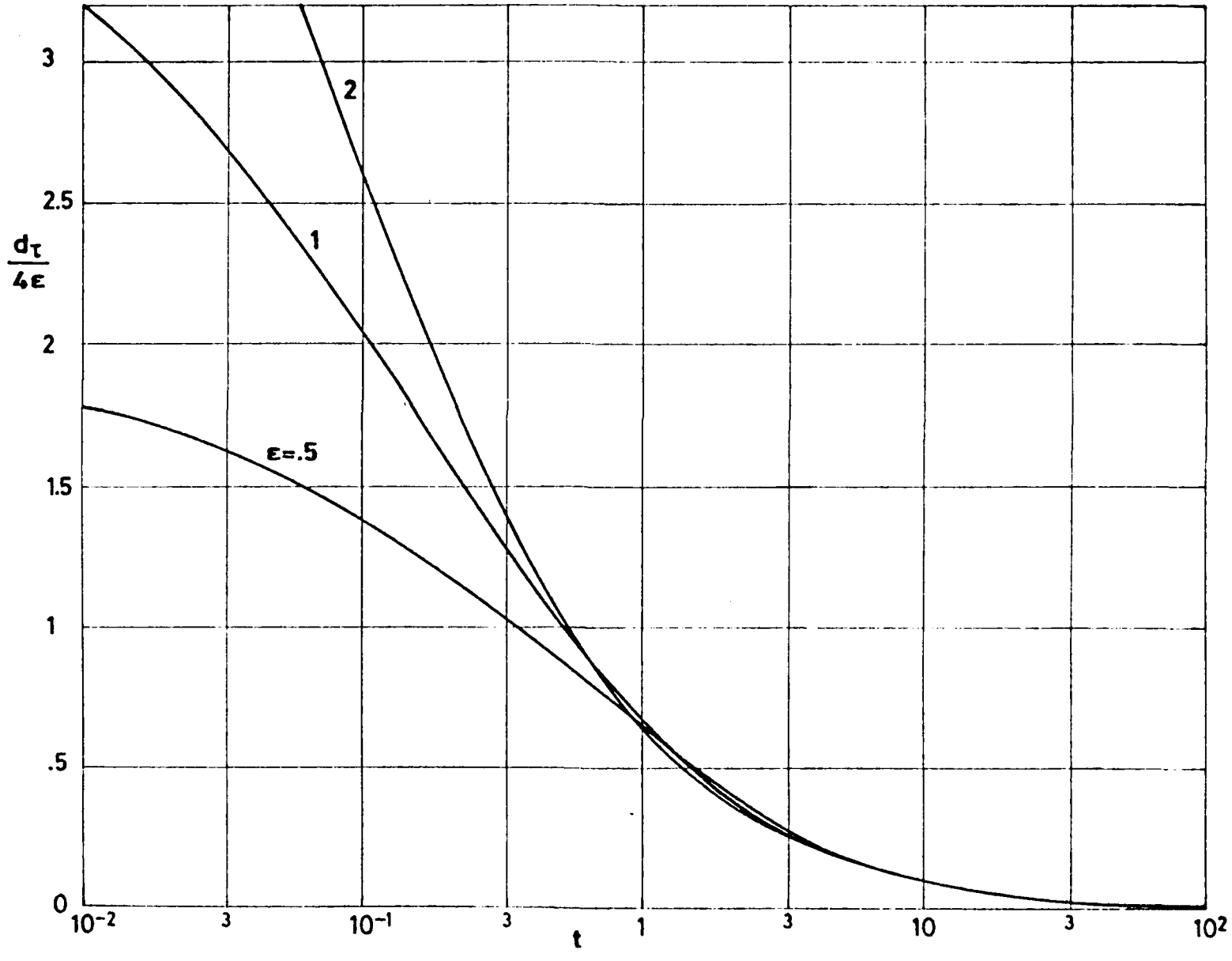
RT_i^2/E
a
UNSTEADY HEAT
CONDUCTION REGION
$l_u - a$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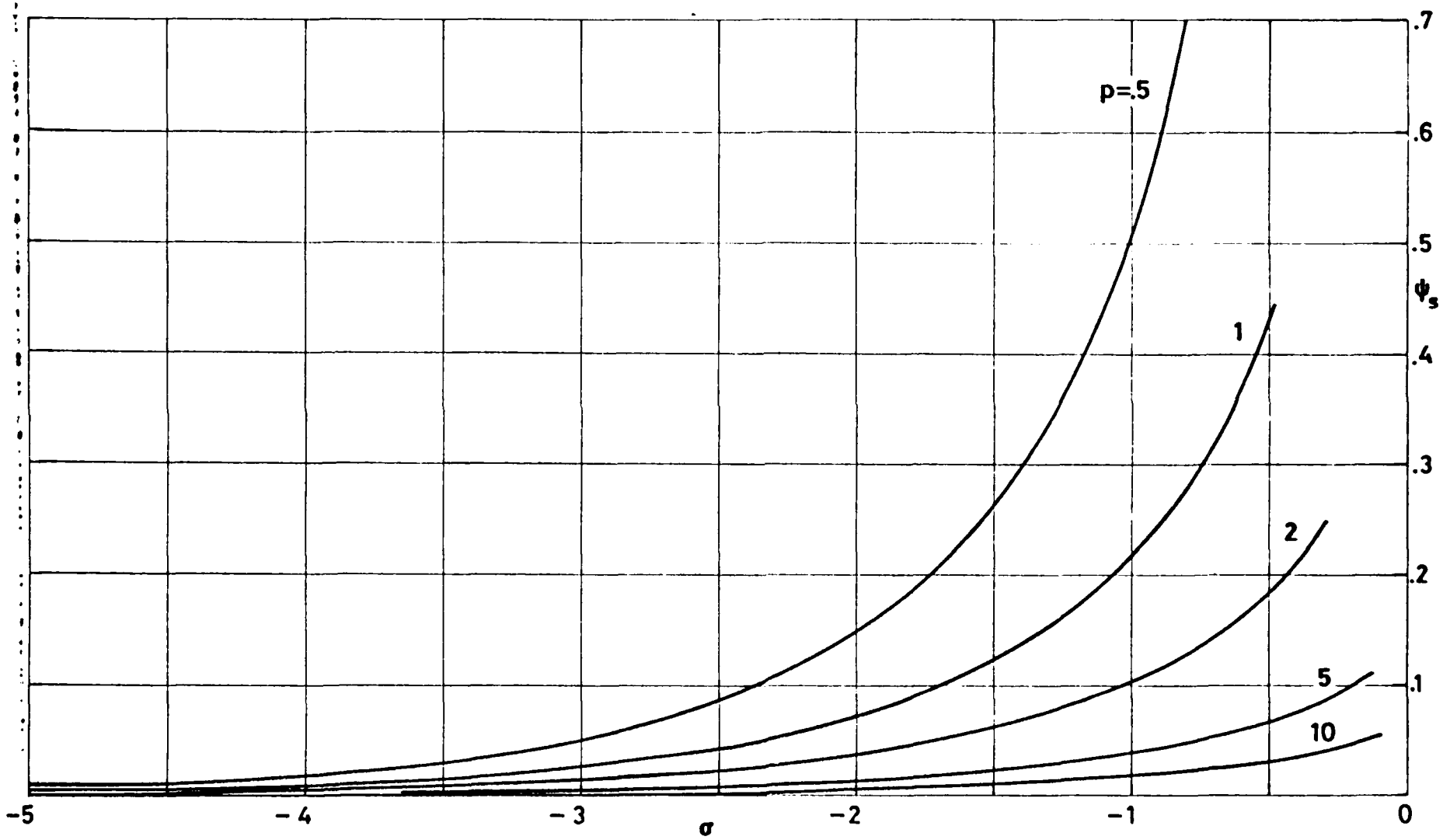
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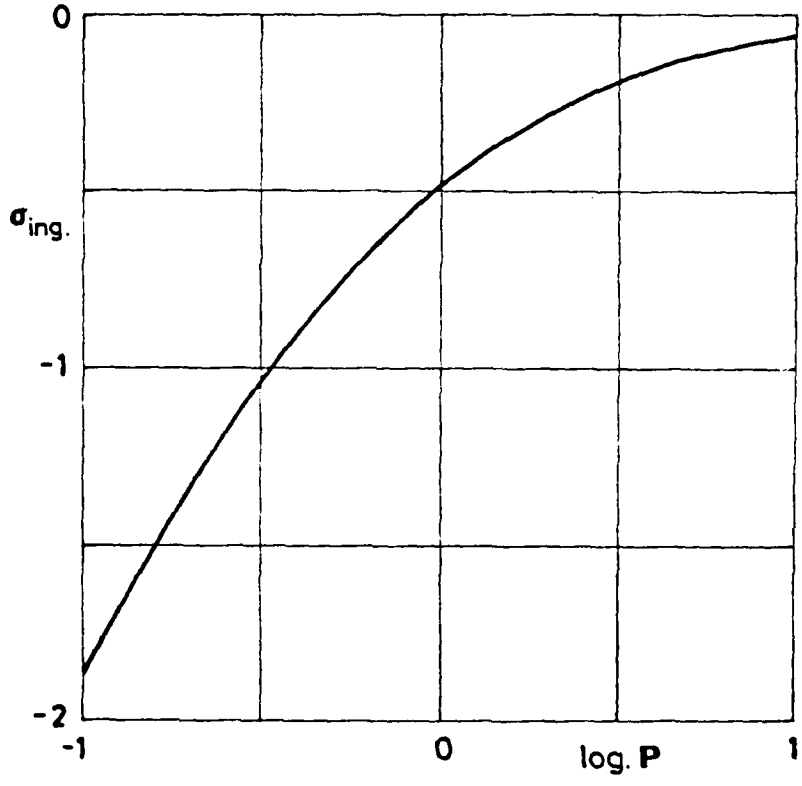
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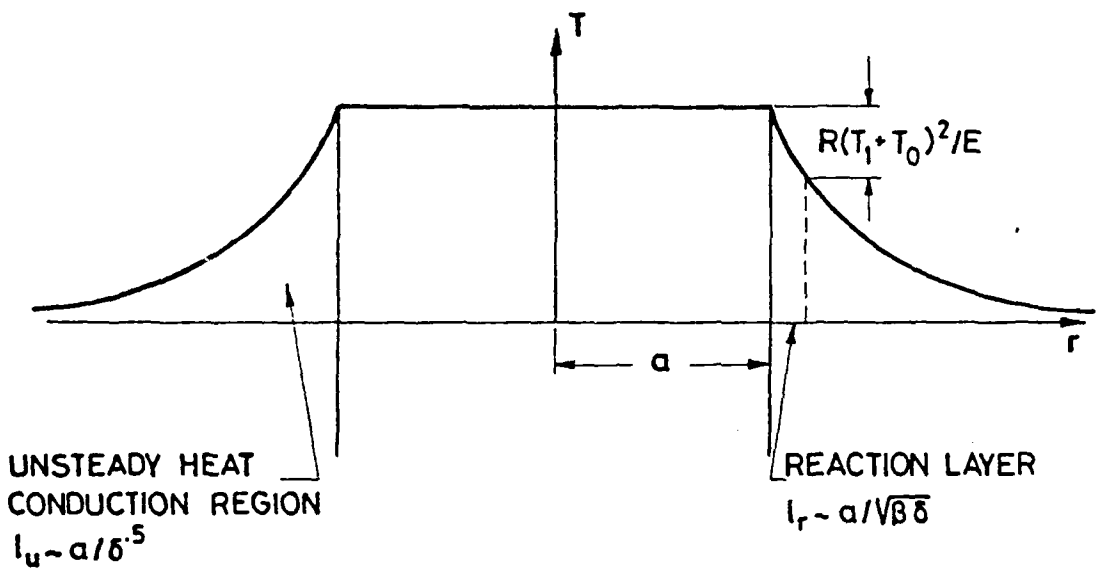

T
$R(T_1+T_0)^2/E$
r
a
UNSTEADY HEAT
CONDUCTION REGION
$l_u \sim a/\delta^{.5}$
REACTION LAYER
$l_r \sim a/\sqrt{\beta\delta}$

ASYMPTOTIC ANALYSIS OF SHOCK TUBE IGNITION OF REACTIVE PARTICLES

1. INTRODUCTION

An interesting method to analyze ignition of reactive particles at high temperature, is the use of a reflected shock technique. In this technique, the reactive particles, which are placed in a holder inside the shock tube, are "dragged", accelerated and heated by the incident shock wave. This wave is reflected at the end wall of the tube and when it encounters the particles, these are decelerated and further heated through a convective-conductive mechanism. These high temperatures accelerate exothermic reactions so that when the rate of chemical heat supply is larger that the heat lost to the gas, ignition occurs at a finite ignition time.

Cohen and Decker[1] used this technique to measure ignition delays of nitrocellulose particles in N_2 at gas temperatures between 600-1200 K and pressures between .1-1.5 MPa. They found that the temperature dependence of these delays changes markedly at 750 K.

The purpose of this Section, is to use high activation energy asymptotics to model ignition delays using the reflected shock technique. The model provides equations for the critical diameter and the ignition time as a function of the parameters of the problem.

No attempt has been made at this stage, to compare

the predictions of the theory, with the experimental results of Cohen and Decker. However, once calculated the gas temperature and velocity behind the incident and reflected shock waves, it is easy to compute the ignition delay time and therefore to estimate the activation energy of the exothermic reaction.

2. FORMULATION

We assume that the particle temperature is uniform and we neglect reactant consumption. Under these conditions the momentum and energy conservation equations, may be written respectively (see Cohen and Decker[1])

$$\frac{dU}{dt} = \frac{3}{4}\frac{C_D}{D}\frac{\bar{\rho}}{\rho}|\bar{U}-U|(\bar{U}-U) \quad , \tag{1}$$

$$\frac{dT}{dt} = \frac{6\bar{h}(\bar{T}-T)}{C\rho D} - \frac{6\varepsilon\sigma T^4}{C\rho D} + \frac{Q A \exp(-E/RT)}{C} \quad , \tag{2}$$

where:

$$C_D = \frac{24}{R_e} + \frac{4}{R_e^{1/3}} \quad ; \qquad R_e = \frac{\bar{\rho} D|\bar{U}-U|}{\bar{\mu}} \tag{3}$$

$$Nu = 2 + .46\, P_r^{.33} R_e^{.55} \quad ; \qquad \bar{h} = \frac{Nu\ \bar{k}}{D} \tag{4}$$

The notation is the same as that used in reference (1), in which a bar is used to distinguish the gas properties from the particle properties. To assume a uniform particle temperature will be correct, as long as the characteristic heat convection time, $\tau_f = \rho C D^2/6\bar{k}Nu$, is large compared to the char-

acteristic heat conduction time, $t_c = \rho C D^2/K$, in the particle.

Eqs. (1) and (2) have to be solved with the appropriate boundary conditions describing the effect of the incident and reflected shock wave to obtain the particle temperature history.

The nondimensional ratio E/RT is usually large. Then, if the reactivity is large enough, the solution exhibits a runaway in the temperature of the particle at a well defined ignition time. However as the reactivity decreases, the ignition temperature increases and approaches the gas-phase temperature $\bar{T}$. There is a critical value of the Damköhler number, such that for values smaller than the critical value, a stationary solution is reached in which the energy released by the chemical reaction is balanced by the energy lost to the gas-phase. For values of the Damköhler number greater than the critical one, there is a well defined ignition time which is derived in this Section as a function of the parameters of the problem.

3. CRITICAL CONDITIONS FOR THERMAL RUNAWAY

To derive the critical Damköhler number, let's consider small increments in the particle temperature above the gas-phase temperature

$$\phi = \frac{E}{R\bar{T}^2} (T-\bar{T}) , \qquad (5)$$

and let's analyze the conditions for the existence of a stationary solution to Eqs. (1)-(2). In the limit of high activation energies, and neglecting radiation, Eq. (2) may be written,

$$\frac{d\phi}{d\tau} = -\phi + \delta e^{\phi} \qquad (6)$$

when the reaction term must be retained because the temperature is close to $\bar{T}$. Here

$$\tau = \frac{12\bar{K}}{\rho C D^2} t \quad , \qquad \delta = \frac{QA\rho D^2 E}{12\bar{K}R\bar{T}^2} \exp\left(-\frac{E}{R\bar{T}}\right) \qquad (7)$$

The Nusselt number has been taken as 2, since critical conditions correspond to long times when the heat transfer by convection is negligible.

The stationary solution of Eq. (6) is given by

$$\delta = \phi \exp(-\phi) \qquad (8)$$

which is shown in Fig. 1 as a function of δ. It is observed that there is a critical value of the Damköhler number ($\delta_c = 1/e$) such that for $\delta > \delta_c$ no stationary solution exists; in this case there is runaway in particle temperature at a finite ignition time. For $\delta < \delta_c$ there are two stationary solutions for each value of δ, of which only the lower one is stable. From the definition of δ, the critical diameter is given by the Semenov result

$$D_c^2 = \frac{4.41\,\bar{K}\,R\bar{T}^2}{\rho EAQ} \exp(E/R\bar{T}) \; . \qquad (9)$$

When the effect of radiation is retained the critical regime occurs with temperatures close to the chemically frozen equilibrium temperature T_f given by

$$\overline{T} = T_f + \frac{\varepsilon\sigma D}{2\overline{K}} T_f^4$$

In this case the critical regime is still given by Eq. (6) and the critical diameter by Eq. (9), is replaced in Eqs. (7) and (9) by $\overline{K}\{1+2\varepsilon\sigma D T_f^3/\overline{K}\}$ and $\overline{T}$ by T_f.

4. IGNITION ANALYSIS

For values of D of the order of D_c the transient temperature history is described by Eq. (6). For values of $D<<D_c$ a thermal runaway will occur when the particle temperature is smaller then $\overline{T}$ (or T_f) by a quantity large compared with $R\overline{T}^2/E$.

To analyze in this case the thermal runaway to ignition, we carry out an asymptotic analysis in the limit of high activation energy of Eqs. (1)-(2), and consider t_c as the time of thermal runaway.

Let's define, $T_c=T_I(t_c)$, as the temperature of the particle at time t_c, with the chemical reaction frozen. In the limit of high activation energy, the chemical heat release is negligible compared to the convection of heat from the hot gases, as long as $T_I<T_c$. Thus, for times smaller than t_c, the temperature history is given by the solution of Eqs.

(1)-(2) without the chemical source term.

For times close to t_c, there follows a short stage of transition to ignition in which the chemical heat release term becomes important and has to be retained. During this stage, the temperature of the particle differs from T_c by a small quantity of order $R T_c^2/E$, but these small changes suffice to produce changes of order unity in the reaction rate.

To study the ignition stage, let's introduce

$$\phi = \frac{E}{R T_c^2} (T-T_I) \quad , \tag{10}$$

and let's expand T, for times close to t_c

$$T = T_c + \left.\frac{dT_I}{dt}\right|_{t_c} (t-t_c) + \frac{R T_c^2}{E} \phi \tag{11}$$

Introducing this expansion in Eq. (2), and defining a stretched time variable

$$\sigma = (t-t_c) \left.\frac{dT_I}{dt}\right|_{t_c} \frac{E}{R T_c^2} \tag{12}$$

results to leading order in

$$\frac{d\phi}{d\sigma} = P \exp(\phi+\sigma) \tag{13}$$

with initial conditions

$$\sigma \to -\infty \qquad \phi = 0 \tag{14}$$

where

$$P = \frac{QA}{c \left.\frac{dT_I}{dt}\right|_{t_c}} \exp(-E/RT_c) \quad . \tag{15}$$

Integrating Eq. (13), results in the following particle temperature history

$$\phi = -\ln(1-P\,e^{\sigma}) \quad . \tag{16}$$

Since t_c is defined as the time at which thermal runaway occurs, Eq. (13) has to satisfy the condition

$$\sigma=0 \quad , \qquad \phi \to \infty \tag{17}$$

and therefore P=1.

Thus, the ignition time is defined by the equation

$$\frac{QA}{C}\exp(-E/RT_I) = \frac{dT_I}{dt} \quad . \tag{18}$$

Therefore, to obtain the ignition time it is only necessary to derive the inert temperature history from Eqs. (1)-(2) without the reaction term, and use that solution in Eq. (18) to deduce the ignition time.

5. INERT SOLUTION

To solve Eq. (1), let's introduce the nondimensional variables

$$x = \frac{Re}{Re_i} \quad , \qquad \tau = \frac{t}{t_c} \quad , \tag{19}$$

where

$$Re_i = \frac{\overline{\rho} D |\overline{U}-U_i|}{\overline{\mu}} \quad , \qquad t_c = \frac{4}{3}\,\frac{\rho D^2}{\overline{\mu}\, Re_i^{2/3}} \quad . \tag{20}$$

With these definitions Eq. (1) results in

$$\frac{dx}{d\tau} = -x^2\left(\frac{4}{x^{1/3}} + \frac{24}{x\,Re_i^{2/3}}\right) \quad , \tag{21}$$

with initial condition

$$x=1 \quad \text{at} \quad \tau=0 \quad , \tag{22}$$

whose solution is shown in Fig. 2 for several values of Re_i. Eq. (21) provides the velocity of the particle behind the incident shock wave, as well as behind the reflected shock wave. In the latter case Re_i is given by Eq. (3), with $\bar{U}=0$ and U the velocity of the particle at the instant the reflected shock wave reaches the particle.

To integrate Eq. (2) we will consider for simplicity the case in which the radiation term is negligible. Introducing

$$z = \frac{\bar{T}-T}{(\bar{T}-T)_i} \quad , \tag{23}$$

and using Eq. (21) results in

$$\frac{d \ln z}{dx} = - \frac{B(2+.46\, Pr^{.33}\, Re_i^{.55}\, x^{.55})}{x^2(4Re_i^{2/3}\, x^{-1/3}+24\, x^{-1})} \quad , \tag{24}$$

with initial condition

$$z=1 \quad \text{at} \quad x=1 \quad , \tag{25}$$

and with

$$B = \frac{8\bar{K}}{\bar{U}C} \quad . \tag{26}$$

This equation has been integrated numerically for Pr=.7, B=8 and different values of Re_i. Fig. 3 shows $Re_i^{.11} \ln z$ as a function of τ for several values of Re_i.

Eq. (18), defining the ignition time, may be written

$$(\bar{T}-T)_i = \frac{QA\rho D^2}{6\bar{K}} \frac{\exp(-E/RT)}{z(2+.46\,Pr^{.33}\,Re_i^{.55}\,x^{.55})} \quad , \qquad (27)$$

which together with Eq. (23) defines x_{ign} and therefore τ_{ign} as a function of the parameters of the problem.

To solve Eq. (27), one may take advantage of the fact that the activation energy is large, and use the following iterative procedure. Let's define

$$f = \frac{QA\rho D^2}{6\bar{K}(\bar{T}-T)_i Pr^{.33}\,Re_i^{.55}} \quad , \quad j = z\left(\frac{2}{Pr^{.33}\,Re_i^{.55}} + .46\,x^{.55}\right) \qquad (28)$$

and let's take the natural log of Eq. (27)

$$-\frac{E}{RT} = -\ln f + \ln j \quad . \qquad (29)$$

However, since j is approximately of order unity, while f is large, one may neglect $\ln j$, and obtain

$$z_{ign} = \frac{\bar{T}}{(\bar{T}-T)_i} - \frac{E}{R(\bar{T}-T)_i}(\ln f)^{-1} \quad . \qquad (30)$$

Figs. 2 and 3 may than be used to derive the zeroth order approximation to x_{ign} and τ_{ign}. Using these values in Eq. (28) a zeroth order approximation to j is calculated, which may then be used in Eq. (29) to derive the first order approximation to z_{ign}. This iterative procedure converges very rapidly to the ignition time.

A further simplification may be introduced by con-

sidering that the initial Reynolds number, Re_i, is large compared with unity, as will usually be the case in practice. Using the solutions to Eqs. (21) and (24) for large values of Re_i in Eq. (30), an explicit zeroth order solution for the ignition time is derived, namely

$$\left(\frac{8\tau_{ign}}{3}+1\right)^{.1666} = 1+\frac{.966\,Re_i^{.111}}{B\,Pr^{.333}}\ln\left(\frac{\bar{T}}{(\bar{T}-T)_i}-\frac{E}{R(\bar{T}-T)_i}(\ln f)^{-1}\right)$$

This equation is valid as long as $T_{ign} << \bar{T}$. When T_{ign} approaches $\bar{T}$, the critical solution discussed in (3) should be used.

It should be pointed out, that in the case in which ignition occurs in the reflected shock region, the ignition is the sum of the time the particle travels until it encounters the reflected shock, and the time from this instant to thermal runaway.

The first component of the ignition time is calculated from the parameters of the incident and reflected shock waves, distance from the holder to the end wall and Fig. 2 giving the particle velocity as a function of the non-dimensional time x. Also, from Fig. 3 the temperature increment in the particle $(\bar{T}-T)$, existing at the instant of encounter with the reflected shock wave, is calculated as a function of the initial temperature increment $(\bar{T}-T)_i$. The resulting temperature increment is used as the initial value in Eqs. (1)-(2) to calculate the time from the instant the particle

enters the reflected shock region until thermal runaway occurs. Analogously, the value Re_i in Eq. (27) is calculated from the particle velocity at the time of entrance in the reflected shock region, which may be obtained from Fig. 2.

REFERENCES

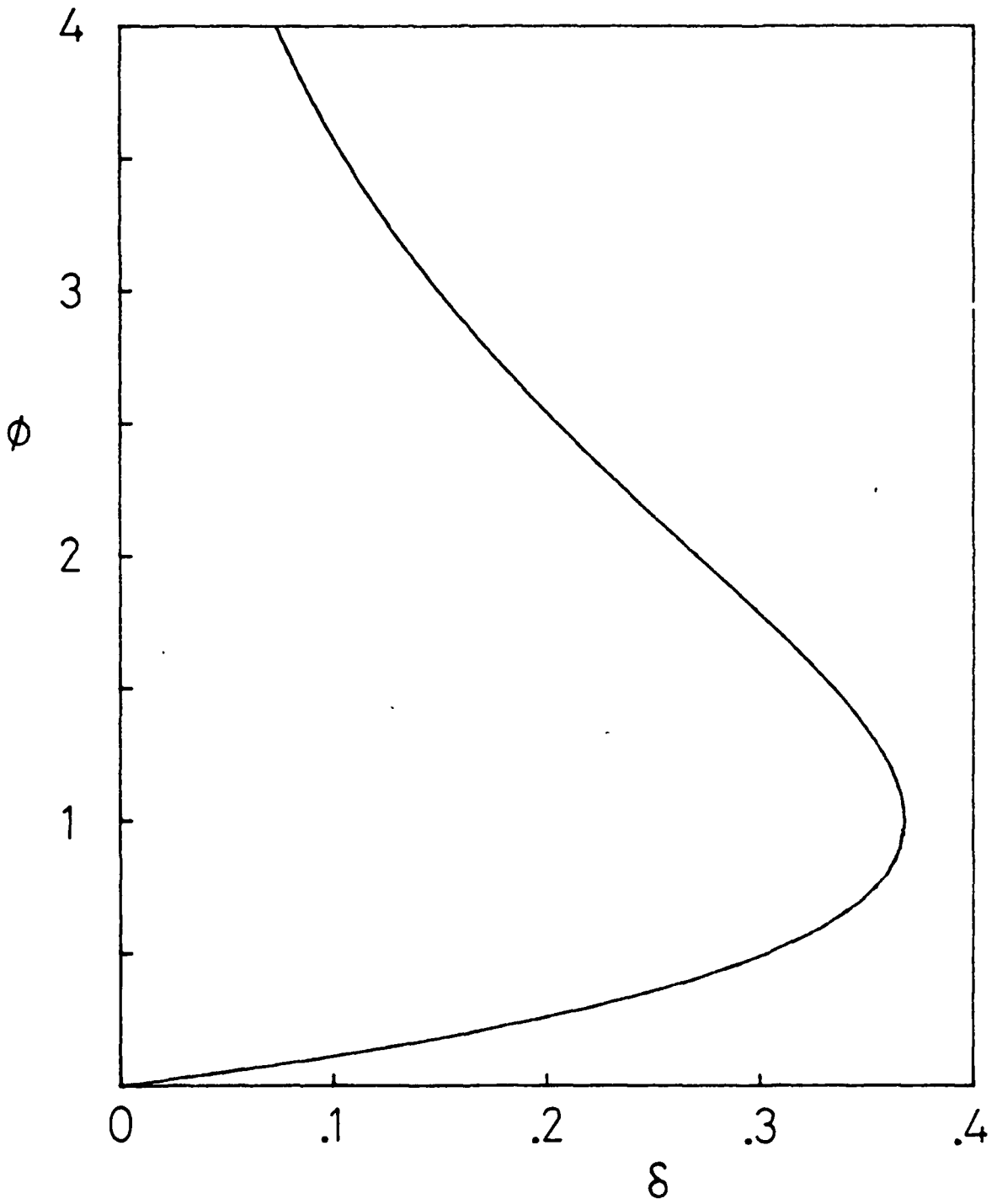
1. Cohen, A. and Decker, L., "Shock Tube Ignition of Nitrocellulose", presented at the Shock Tube Symposium, Jerusalem, Israel, July 1979.

LIST OF FIGURES

1. Stationary temperature increment as a function of $\hat{0}$.

2. Inert non-dimensional Reynolds Number as a function of τ for different values of Re_i.

3. Inert temperature increment $Re_i^{11} \ln z$ as a function of τ for several values of Re_i.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φ
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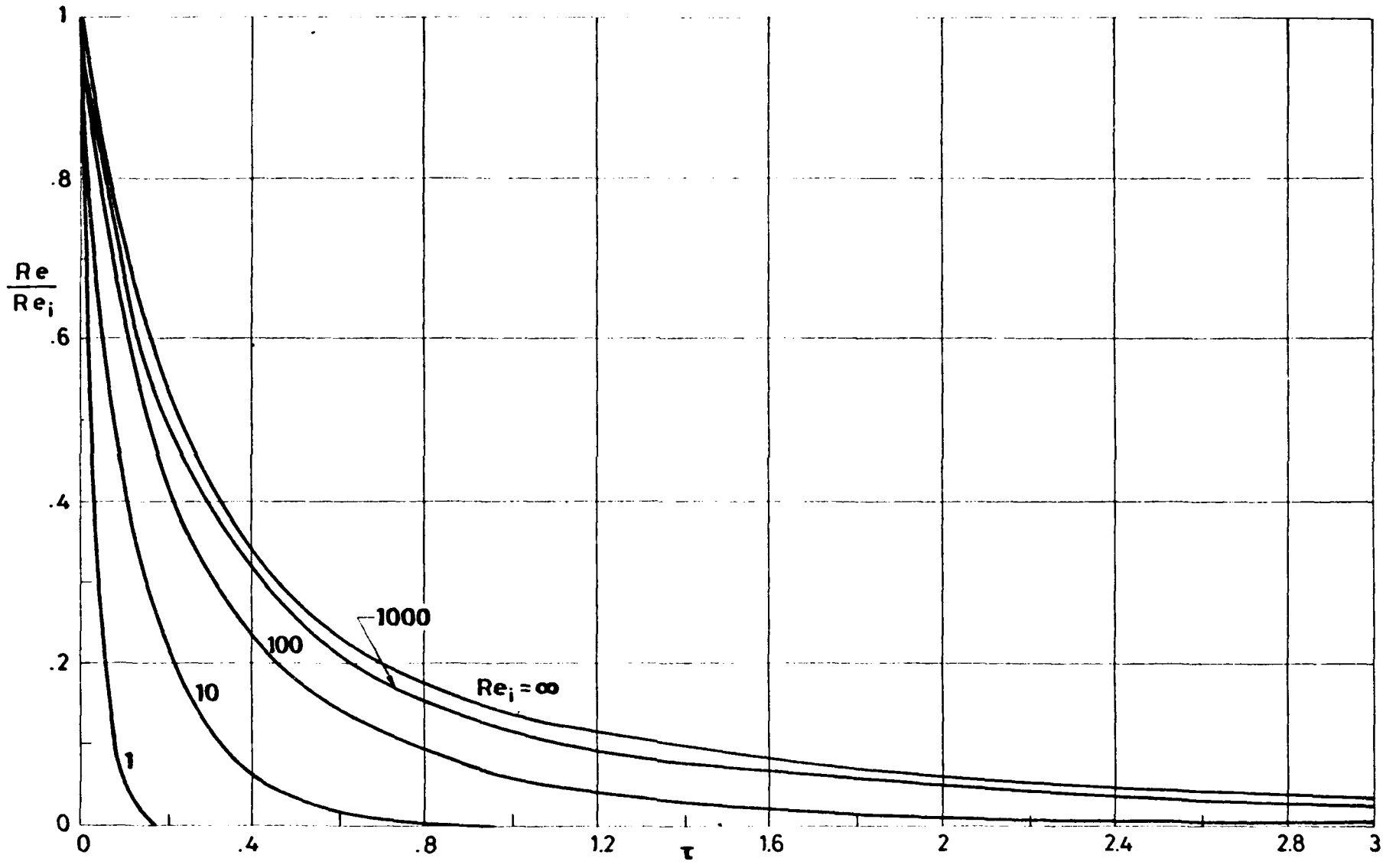
.2
.3
.4
δ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.2
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Re / Re_i
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2.8
3
τ
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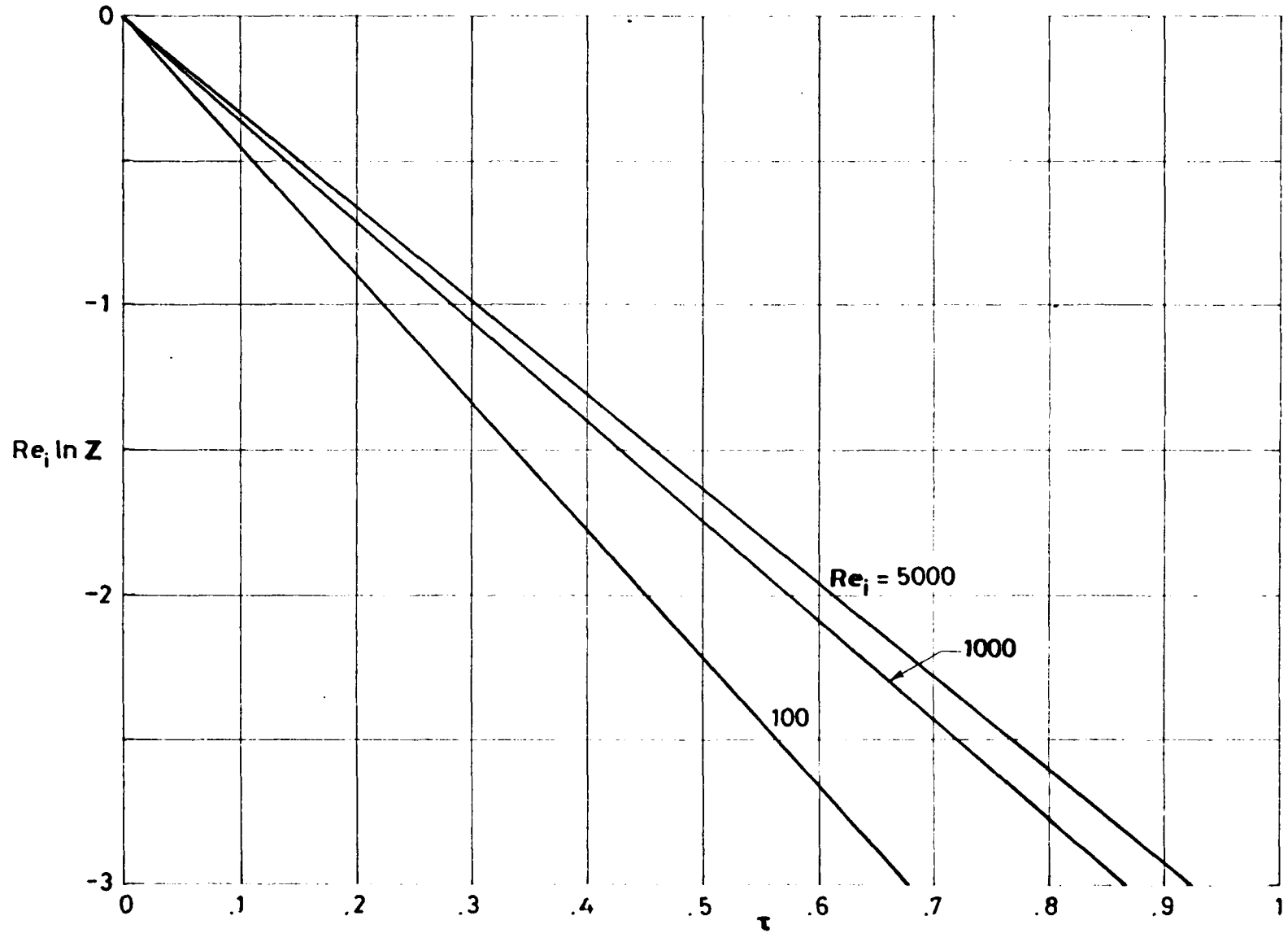
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$Re_i \ln Z$
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.7
.8
.9
1
τ
Re_i = 5000
1000
100


LMED
8