SOME   IGNITION   PROBLEMS

WITH   SPHERICAL   SYMMETRY

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ABSTRACT

Several problems of thermal ignition from a
spherical hot 'pocket' in a cool infinite medium have
been investigated theoretically. It is assumed that
at some initial time a hot 'pocket' is introduced
into a cool medium, where either the inner region, or
the medium, or both are chemically reactive. Energy
conservation equations have been examined with a view
to finding the minimum initial radius of the hot 'pocket'
for which ignition will just take place, and its de-
pendence on transport properties and chemical kinetic
parameters.

Firstly the case of a hot spherical gas bubble of
reaction products in a stagnant atmosphere of fuel
and oxygen has been considered. By assuming the reaction
rate to be of simple step-function form, solutions valid
for small values of the time have been found. On the
basis of these, critical bubble sizes have been de-
termined as a function of ignition temperature.

The same problem was then examined with a polynomial
form of reaction rate function and thermal properties
varying with temperature. An adaptation of 'profile'
or 'integral' methods due to Spalding has been used and

the effect on critical radius of changes in the temper-
ature dependence of the transport coefficients, and
variations in the activation energy parameter have been
evaluated numerically.

A corresponding problem in thermal explosion theory
of hot spots in solids was then examined, using both
linear and power law assumptions for chemical reaction
rate. In the former case a first approximation to the
formal solution has been used to determine a critical
condition for ignition, while in the latter case the
profile method has been employed.

Finally the behaviour of a hot inert gas bubble in
an infinite sea of cool reactive liquid has been investi-
gated. The profile method has again been used and an
" inner solution" valid for small values of time has been
found by singular perturbation methods. It has been
shown that in this time region, inertia and chemical
reaction effects are negligible. An " outer solution"
has been found numerically using the properties of the
" inner solution" .

The results we have obtained are in good agreement
with exact numerical solutions, when they exist, and
in reasonable agreement with experimental work.

CONTENTS

CHAPTER I

INTRODUCTION

The various problems considered in this work
exhibit several common features and hence have been
treated by related methods. Basically, we have that
at some initial time, a hot 'pocket' is introduced
into a cool infinite medium; either this spherical
pocket, or the surroundings, or both, are chemically
reactive. Consequently the initial temperature profile
is of rectangular 'step' form. A balance must exist
between conduction of heat away from the pocket, the
production of heat by reaction within the reactive part
of the medium, and the transport of energy by inertia
effects, if any. Hence our primary concern is with
energy conservation equations. The dominance of chemical
reaction rate terms leads to ignition in the pocket or
the medium, whilst dominance of heat conduction leads to
final extinction. If these effects were comparable
throughout the process, an oscillatory state could be
achieved. We therefore seek to determine a critical
condition for ignition of the pocket or medium, separa-
ting the two regimes of ignition and extinction. A

parameter which describes such a critical condition is the critical initial radius of the hot pocket for which ignition will just take place.

The initial infinite temperature gradient at the pocket interface leads to very rapid changes in the temperature in that region; at the outset conduction is the controlling factor. Ignition processes take place within times of the order of several milliseconds, and are, as a result, difficult to observe accurately; for this reason relatively little experimental work has been attempted. The rapidity of the process also enables us to make several simplifying assumptions concerning, for example, thermal properties etc.

Strong non-linearity is introduced into the energy conservation equation by the chemical reaction rate term; for this reason transient solutions have not been obtained in analytical form. This term is assumed to be temperature explicit, but as it involves an Arrhenius rate factor, it has been necessary to use various approximations to it. Formal solutions can only be obtained for the simpler of such approximations. For the less crude models where non-linearity is not eliminated an approximate technique must be used.

Consequently, extensive use has been made of an

adaptation of the profile methods of boundary layer
theory. The method was originally proposed by
Pohlhausen (1921) who applied it to von Karman's
momentum equation (1921).

The exact boundary-layer equations are frequently
not amenable to solution; von Karman's equation was
derived by integrating the boundary layer equations
across the boundary-layer, and so reducing the partial
differential equation to an ordinary non-linear diff-
erential equation; this enables approximate solutions
of the exact equation to be found. This procedure
was extended to all boundary layer problems (heat
transfer, compressible flow, etc.).

The procedure adopted by Pohlhausen involved the
approximation to a dimensionless velocity by a known
simple function of a dimensionless distance variable.
Ensuring that this function satisfies the appropriate
boundary conditions, it may be substituted into von
Karman's momentum equation, reducing it to an ordinary
non-linear differential equation which may be solved
for the boundary layer thickness. Pohlhausen used a
fourth degree polynomial, in order to satisfy the four
boundary conditions. The success of this approximate
method, even when a very simple expression is assumed

for the velocity in the boundary layer shows that such a reduction is of fundamental importance.

The basic principles of this method have been adapted to ignition problems by Spalding (1958). The energy conservation equation is integrated over the space variable, the integrals being evaluated by assumptions as to the form of the temperature distribution or 'profile'. Two or more time-dependent parameters are introduced to evaluate the integrals, the number being determined by the number of equations and boundary conditions to be satisfied. Thus a set of ordinary differential equations is obtained for these parameters. In this investigation only the simple cases of linear and parabolic profiles have been considered. Spalding (1958) has shown that profile methods give an accuracy to within 20 per cent. Choice of profile assumption does not appear to affect the results greatly, provided that the profile exhibits the correct physical characteristics.

The primary disadvantage of this method is the somewhat arbitrary choice of time-dependent parameters. We have used here quantities such as the temperature at the centre, or other convenient position of the pocket, and the pocket 'size', defined as the region

wherein the temperature is above the initial ambient value. The latter can only be specified in terms of simple linear, parabolic or other profiles having a discontinuity in temperature gradient at this point; only such profiles have been used by Spalding. A 'continuous' profile given by, say, a negative exponential term, does not uniquely define a second time-dependent parameter, and hence, although physically more realistic, appears to have little application except for qualitative work.

This degree of arbitrariness in the specification of the size of the pocket is exactly analogous to the definition of a boundary layer, as that part of the flow wherein the velocity increases from zero to <u>some</u> fixed proportion of the main stream velocity. This inadequacy is not felt in hydrodynamics since quantitative values for the boundary layer thickness are not often required.

A further disadvantage of the method is the difficulty of simulating the original rectangular step distribution of temperature by the assumed profile. The problem actually undertaken thus represents the solution of the appropriate equations with slightly altered initial conditions.

Despite these shortcomings profile methods are shown to give good agreement with exact numerical results.

The first problem examined has been that of flame ignition from a spherical hot gas pocket. Although much study has been made of steady laminar flame propagation, transient solutions have not been obtained. We assume that the chemical reaction rate may be made temperature explicit. Spalding (1957) has shown that this assumption is valid for the cases of i) single step reactions ii) chain reactions obeying the steady state approximation iii) simple chain branching reactions. It is necessary that the diffusion coefficients of all components should be 'normal' i.e. equal to each other and to the thermal diffusivity, and that the flames should be adiabatic. Case (i) is assumed to be satisfied here.

In Chapter 2 is considered the case of step-function reaction rate, using the concept of an ignition temperature; a formal solution is obtained. Chapter 3 deals with a polynomial approximation to reaction rate function applied to the same problem; the approximation is due to Spalding (1957) and Rosen (1958); the profile method discussed above is used. The assumption of constant

thermal properties used in the preceeding chapter is
lifted, and numerical results are obtained.

The classical theory of the thermal explosion of
a combustible mixture is due to Semenov (c.f. Frank-
Kamenetskii (1955)). It considers a closed vessel
filled with a combustible mixture, and is most easily
understood graphically. Figure 1 represents curves
of heat liberation (I) and heat loss (II) as a function
of temperature. The quantity of heat liberated per
unit time in a stationary process must evidently equal
the quantity of heat given off in unit time by the
vessel to the ambient medium; graphically this corres-
ponds to the points of intersection of curves I and II.
It is possible that two such stationary points exist
(points A and B), or only one (point C), or there may
be no possible stationary states. The region of the
point C is taken to be the critical ignition region and
gives an ignition temperature. It is not difficult to
show that the stationary point A is stable, while B
is unstable. It follows from the Semenov theory that
ignition conditions, in particular, ignition temperature,
are determined by all the conditions for the growth of
the process in a system, rather than by single properties
of the mixture. Thus the concept of an ignition temp-

erature,as used in Chapter 2,is more of a mathematical
idealization than representative of physical reality;
nevertheless it does provide a simple means of defining
the 'flame radius'.

Laminar flame theory regards the flame as a zone
of heat conduction, diffusion, reaction and viscous
effects; the latter are usually neglected. Flow
velocities are assumed small and hence the assumption
of constant pressure is not unreasonable. The chemical
nature of the gas changes across the flame zone.

Much attention has been devoted to the determina-
tion of flame speed. This is defined as the linear
velocity of displacement of the flame front with res-
pect to fresh gas entering the combustion zone in a
direction normal to the flame front, and is often used
to test the correctness of a theory. The dimensionless
Peclet number, based on laminar flame speed has been
used to illustrate the results obtained with polynomial
form of reaction rate function.

Another useful concept is that of Lewis number
defined as the ratio of diffusion coefficient to
thermal diffusivity; hence it represents the ratio of
energy transferred by diffusion to that transported by
conduction. It is most often taken to be constant,

and generally assumed equal to unity. This is in accordance with the kinetic theory of gases. Consequently the combined heat and mass transfer equations take a simple form, and are reduced to an enthalpy conservation equation. The case of combustion in premixed gases considered here does not involve mass transfer considerations explicitly and is equivalent to a unit Lewis number assumption.

Chapter 4 considers a similar problem in the thermal theory of hot spot explosion. Such 'pockets' arise in reactive solids, and clearly safety criteria for handling such solids are of some importance industrially – the critical initial radius for ignition is just one such criterion. The formation of hot spots is often due to the adiabatic heating of entrapped bubbles by compression. These hot spots, though small in diameter, are often large compared with molecular dimensions, and they may perhaps be of order of 10^{-5} to 10^{-3} cm. in diameter. In this problem, unlike that of the two previous chapters where the adiabatic flame temperature was the maximum attainable, there is no upper limit to the temperature rise of the hot spot.

Two methods of solution have been attempted. The first assumes a linear reaction rate term and a formal

solution of the energy equation has been found by a
method based on a continuous distribution of heat
sources. Using a first approximation to the solution
for the temperature at the centre of the hot spot, a
critical condition was obtained. The problem was then
considered using a power law approximation to the reaction
rate function; the previous profile method was used.
Both theories give good agreement with exact numerical
solution.

The development of detonation in solids from an
initiating source passes through the following phases:
a) Initiation of reaction. b)The growth of this re-
action from the decomposition region to a burning region.
c) Acceleration of the burning and a sharp transition
to low velocity detonation. d) Propagation of low
velocity detonation. e) Propagation of high velocity
detonation. Of these a) is the best understood and it
is this phase that we consider. It is generally accepted
that the initiation mechanism is thermal in nature,
although hot spots may be formed in several ways.

Since it is essential to have instruments capable
of measuring times much less than a microsecond and
apparatus which can analyse reactions taking place in
a region perhaps only one-tenth of a millimetre thick,

it is not surprising that little progress has been made in the experimental field.

The original theory of thermal explosion of Frank-Kamenetskii (1955) and others considers the stationary state condition where the temperature distribution over space only is taken into account and not its variation with time. The conditions under which a stationary solution is just not possible will then give the critical condition for explosion; this is obtained in the form of critical values of one or more dimensionless parameters.

Since then, several theoretical formulations have been proposed using the transient energy equation. These consider various approximations to the reaction rate function. Merzhanov et al. (1963) have obtained numerical solutions using the exponential approximation. The various theories are compared in a paper by Thomas (1965); a graphical comparison is made which we, too, have utilised.

The final chapter contains a study of the behaviour of a hot inert gas bubble in an infinite sea of cool reactive liquid; this is probably the most interesting problem considered here and correspondingly the most complex. It has possible applications in liquid fuel

rockets and chemical reactors.

The interest of the problem lies in the interplay of thermal, inertial, and chemical reaction effects. It is assumed that the bubble has uniform pressure, temperature and density at any instant in time; this is justified by its small size; surface tension,too,is neglected. In addition to the non-linearity introduced by the reaction rate which is approximated by simple power law expression, a further complication is introduced in the form of a small parameter. Neglect of this parameter would lower the order of the equations; consequently the problem is ideally suited to the methods of singular perturbation theory.

Once again the profile method has been used; it has been necessary to introduce an additional time-dependent parameter in order to satisfy the temperature gradient condition at the bubble interface. Thermal and transport properties of both media are assumed constant.

This investigation is thus an extension of classical hot spot theory to the case of liquid explosives. With liquids, however, in addition to conduction, convection plays an important part in the transfer of heat; the effect of this is to increase the value of the critical

initial radius above which explosion can occur.

Despite its obvious interest, theoretical work of other authors on such problems appears limited to dynamics of phase growth. Scriven (1959) has formulated the equations governing spherically symmetric phase growth in an infinite medium. These are then simplified to describe growth controlled by the transport of heat and matter. Similarity solutions are obtained for conditions typical of bubble growth in the nucleate boiling of a) pure materials and b) binary mixtures. There appear to be no formulations of the problem considered here, even in a simple form.

CHAPTER 2

FLAME IGNITION FROM A SPHERICAL
HOT GAS POCKET

1. Introduction

The theory of steady laminar flame propagation has been studied extensively and is well understood, but transient solutions of the energy balance equation have not been obtained in analytical form due to the non-linearity introduced by the reaction rate term.

The case of transient propagation from a one dimensional slab has been solved graphically by Spalding (1955), using a finite difference method. It is possible to use a similar method for flame propagation from a spherical hot gas pocket but analytical solutions are preferable since they give some insight into the ignition process.

The problem of flame propagation from a hot gas slab has also been considered by Spalding (1958) using a profile method. This approach will be discussed in greater detail in the following chapter. Profile methods are found to give an accuracy to within twenty per cent.

Steady laminar flame propagation with spherical

symmetry has been studied by Jain (1961), the flame
being supported by an outwards streaming source of
premixed gas. He was able to obtain analytical
solutions for step-function reaction rates and for
Adams type reaction rate functions; Adams used the
assumption of a constant temperature gradient in the
reaction zone. Although the step-function reaction
rate is not very realistic in shape, it is nevertheless
more representative of reality than the spiked Adams'
type of reaction rate curves.

The propagation of flames in combustible gases
has been studied for many years from the experimental
point of view, partly because of its importance in
coalmines and in petrol engines. Experimental obser-
vations on ignition and initial flame growth in hydro-
carbon-air mixtures have been made by Arnold and
Sherburne (1953). Premixed fuel was introduced into
the burner and sparked. Simultaneously a series of
light flashes was triggered to produce Schlieren
images of the flame growth on photographic film;
pictures were taken at time intervals of about 400
microseconds after the sparking. They were able to
obtain the growth rates of hot gas bubbles for varying
energy inputs. Critical bubble sizes below which

extinction always occurred were also deduced; a critical radius versus time curve was plotted and this was extrapolated back to the time origin to yield critical initial bubble radii.

The present chapter is concerned with the theory of flame ignition from a spherical hot gas pocket. It is assumed that at some initial time a mass of hot reaction products is introduced into a stagnant infinite medium of cold combustible gas. Attention has been concentrated on finding the minimum size of hot gas bubble for which flame propagation will take place and its dependence on chemical kinetic parameters. In order to facilitate the solution the following simplifying assumptions have been made: (i) The system is adiabatic, (ii) the chemical reaction rate may be made temperature-explicit, (iii) the gases have constant properties. Assumptions (i) and (ii) are often made in steady flame theory and (iii) may be justified on the grounds that attention is restricted to small values of the time where gas expansion effects are not likely to be of importance.

With the assumptions made, the mathematical problem is that of heat generation and conduction with a moving boundary. Thus at any time sufficiently

small, there is a variable inner region within which
heat is produced at a constant rate, the surface being
kept at the ignition temperature. For an initial
stepped temperature distribution it is required to
find the step size for which the maximum central
temperature will just fall to the ignition temperature.
This determines the critical state, separating final
ignition from extinction.

2.1. Formulation of the problem

With spherical symmetry, the energy conservation
equation may be written

$$c\rho \, \frac{\partial T}{\partial t} = \rho \, \frac{\partial}{\partial \psi} \, (K\rho r^4 \, \frac{\partial T}{\partial \psi}) + Q \qquad (1)$$

where the independent variable ψ, which is effectively
a stream function, is defined by

$$\psi = \int_0^r \rho r^2 dr \qquad (2)$$

Here ρ is an assumed function of T, and

$$r = \left\{ 3 \int_0^\psi \frac{d\psi}{\rho(T)} \right\}^{1/3} \qquad (3)$$

The variables above have the following significance:

r   Radial distance.

t   Time.

ρ   Gas density.

c   Specific heat.

K   Thermal conductivity.

T   Absolute temperature.

Q   Volumetric heat release rate due to chemical
     reaction, in general a function of temperature
     and fuel concentration. As shown by Spalding
     (1957), under adiabatic conditions and for
     'normal' diffusion, it can be made a function
     of temperature alone.

To deal with equation (1) we shall assume the gas
properties $c, \rho,$ and k to be independent of T.
Although this is not realistic in general, it is prob-
ably permissible for small values of time.

With the above assumptions, it is convenient to
introduce the following dimensionless variables.

Distance
$$d\xi = \left\{ \frac{\overline{Q}}{k(T_b - T_u)} \right\}^{\frac{1}{2}} dr \qquad (4)$$

Time
$$d\theta = \frac{\overline{Q}}{c\rho(T_b - T_u)} dt \qquad (5)$$

$$\text{Temperature} \quad \tau = \frac{T-T_u}{T_b-T_u} \tag{6}$$

$$\text{Reaction rate} \quad \phi(\tau) = Q/\bar{Q} \tag{7}$$

where $\bar{Q} = \displaystyle\int_0^1 Q\,d\tau$.

The suffices u,b refer to the unburnt and burnt states of the gas respectively.

On introducing the dimensionless variables into the energy equation this becomes

$$\frac{\partial \tau}{\partial \theta} = \frac{1}{\xi^2}\frac{\partial}{\partial \xi}\left(\xi^2\frac{\partial \tau}{\partial \xi}\right) + \phi(\tau) \tag{8}$$

The initial and boundary conditions applicable to equation (8) are

$$\theta = 0 : \quad \begin{aligned} 0 \le \xi \le \xi_0 \, , & \quad \tau = 1 \, , \\ \xi > \xi_0 \, , & \quad \tau = 0 \, . \end{aligned} \tag{9}$$

$$\theta > 0 \quad \begin{aligned} \xi = 0 \, , & \quad \partial \tau/\partial \xi = 0 \, , \\ \xi = \infty \, , & \quad \tau = 0 \, . \end{aligned} \tag{10}$$

In (9), ξ_0 denotes the initial dimensionless radius of the hot gas pocket.

Definition (7) shows that the area under $\phi(\tau)$ between $\tau = 0$ and $\tau = 1$ is unity. A typical reaction rate function is sketched in Fig.2. and we approximate it by a simple step-function

$$\bar{\phi}(\tau) = \begin{cases} 0, & 0 \leq \tau < \tau_i , \\ 1/(1-\tau_i), & \tau_i < \tau \leq 1 , \end{cases} \qquad (11)$$

where τ_i is the dimensionless ignition temperature.
We can now define $\xi_i(\theta)$ to be the radius of the
flame front at time θ, by

$$0 \leq \xi \leq \xi_i(\theta) : \tau \geq \tau_i ; \qquad \xi \geq \xi_i(\theta): \tau \leq \tau_i \quad (12)$$

Subsequently we shall refer to that part of the medium,
where the temperature is greater than or equal to the
ignition temperature as the <u>flame</u>.

On writing $\omega = \xi\tau$, equation (8) becomes

$$\frac{\partial\omega}{\partial\theta} = \frac{\partial^2\omega}{\partial\xi^2} + \phi(\xi) \qquad (13)$$

where

$$\phi(\xi) = \begin{cases} \xi/(1-\tau_i), & 0 \leq \xi < \xi_i , \\ 0, & \xi > \xi_i . \end{cases} \qquad (14)$$

2.2. Formal solution of the problem

Since ω is an odd function of ξ (τ is an even
function due to (10)) it is convenient to solve (13)
by application of Fourier sine-transforms. We put

$$\bar{\omega} = \sqrt{\frac{2}{\pi}} \int_0^\infty \omega \sin(\xi\eta) d\xi \qquad (15)$$

On applying the transformation to equation (13), this becomes

$$\frac{d\bar{\bar{\omega}}}{d\theta} + \eta^2 \bar{\bar{\omega}} = - \frac{1}{1-\tau_i} \sqrt{\left(\frac{2}{\pi}\right)} \ \frac{\partial}{\partial \eta} \left(\frac{\sin \xi_i \eta}{\eta} \right) \ . \ (16)$$

Equation (16) implies that

$$\lim_{\xi \to \infty} \omega = \lim_{\xi \to \infty} \partial \omega / \partial \xi = 0 \ .$$

Initial conditions (9) give

$$\theta = 0 : \bar{\bar{\omega}}_0 = - \sqrt{\left(\frac{2}{\pi}\right)} \ \frac{\partial}{\partial \eta} \left(\frac{\sin \xi_0 \eta}{\eta} \right) \quad (17)$$

On solving equation (16) subject to (17), one finds

$$\bar{\bar{\omega}} = - \sqrt{\left(\frac{2}{\pi}\right)} \left(\frac{\sin \xi_0 \eta}{\eta} \right) \ \exp(-\eta^2 \theta) -$$

$$- \frac{1}{1-\tau_i} \sqrt{\left(\frac{2}{\pi}\right)} \int_0^\theta \frac{\partial}{\partial \eta} \left(\frac{\sin \xi_i' \eta}{\eta} \right) \exp\left\{ -\eta^2 (\theta-\theta') \right\} d\theta',$$
$$(18)$$

where $\quad \xi_i' = \xi_i(\theta') \ .$

Expression (18) can now be formally inverted to give the required temperature distribution. Thus

$$\xi\tau = - \frac{2}{\pi} \int_0^\infty \frac{\partial}{\partial \eta} \left(\frac{\sin \xi_0 \eta}{\eta} \right) \exp(-\eta^2 \theta) \sin(\xi\eta) d\eta -$$

$$- \frac{2}{\pi} \frac{1}{1-\tau_i} \int_0^\infty \int_0^\theta \frac{\partial}{\partial \eta} \left(\frac{\sin \xi_i' \eta}{\eta} \right) \exp\left\{ -\eta^2 (\theta-\theta') \right\}$$
$$\sin(\xi\eta) d\theta' d\eta \ . \quad (19)$$

By using Parseval's theorem for Fourier transforms
(c.f. Sneddon (1951), it is possible to change (19) into
a more recognizable form.   One finds

$$\xi\tau = \sqrt{\left(\frac{\theta}{\pi}\right)}\,[\exp\{-(\xi_0+\xi)^2/4\theta\}-\exp\{-(\xi_0-\xi)^2/4\theta\}] +$$

$$+ \frac{\xi}{2}[\mathrm{erf}\{(\xi_0+\xi)/2\sqrt\theta\} +\mathrm{erf}\{(\xi_0-\xi)/2\sqrt\theta\}] +$$

$$+ \frac{1}{1-\tau_i}\frac{1}{\sqrt\pi}\int_0^\theta \sqrt{\theta'}[\exp\{-(\tilde\xi_i+\xi)^2/4\theta'\}-\exp\{-(\tilde\xi_i-\xi)^2/4\theta'\}]d\theta' +$$

$$+ \frac{1}{1-\tau_i}\frac{\xi}{2}\int_0^\theta [\mathrm{erf}\{(\tilde\xi_i+\xi)/2\sqrt{\theta'}\} + \mathrm{erf}\{(\tilde\xi_i-\xi)/2\sqrt{\theta'}\}]d\theta' \quad (20)$$

with $\tilde\xi_i = \xi_i(\theta-\theta')$.   In (20) the first two terms are
the usual ones which arise in heat conduction theory
for the given initial conditions (7), whereas the inte-
grals are the contribution to the temperature from the
heat source term.

By definition, when $\xi = \xi_i$, $\tau = \tau_i$, giving an
integral equation for the flame front radius at any
time θ.   Thus

$$\xi_i\tau_i = \sqrt{\left(\frac{\theta}{\pi}\right)}\,[\exp\{-(\xi_0+\xi_i)^2/4\theta\}-\exp\{-(\xi_0-\xi_i)^2/4\theta\}] +$$

$$+ \frac{\xi_i}{2}[\mathrm{erf}\{(\xi_0+\xi_i)/2\sqrt\theta\} +\mathrm{erf}\{(\xi_0-\xi_i)/2\sqrt\theta\}] +$$

$$+ \frac{1}{1-\tau_i}\frac{1}{\sqrt\pi}\int_0^\theta \sqrt{\theta'}[\exp\{-(\tilde\xi_i+\xi_i)^2/4\theta'\}-\exp\{-(\tilde\xi_i-\xi_i)^2/4\theta'\}]d\theta'$$

$$+ \frac{1}{1-\tau_i}\frac{\xi_i}{2}\int_0^\theta [\mathrm{erf}\{(\tilde\xi_i+\xi_i)/2\sqrt{\theta'}\} + \mathrm{erf}\{(\tilde\xi_i-\xi_i)/2\sqrt{\theta'}\}]d\theta'.$$

$$(21)$$

From (19) the temperature at the centre of the hot gas pocket is obtained as

$$\tau(\theta) = -\frac{2}{\pi}\int_0^\infty \eta\frac{\partial}{\partial\eta}\left(\frac{\sin\xi_0\eta}{\eta}\right)\exp(-\eta^2\theta)d\eta -$$

$$-\frac{2}{\pi}\frac{1}{1-\tau_i}\int_0^\infty\int_0^\theta \eta\frac{\partial}{\partial\eta}\left(\frac{\sin\xi_i'\eta}{\eta}\right)\exp\left[-\eta^2(\theta-\theta')\right]d\theta'd\eta \tag{22}$$

We shall show from (22) that extinction will occur if the temperature at the centre of the bubble falls to τ_i. Suppose that at some time $\theta = \theta_1$, $\xi_i = 0$. Then at time $\theta_1 + \delta\theta$, where $\delta\theta$ is a small time increment,

$$\tau(\theta_1+\delta\theta) \simeq -\frac{2}{\pi}\int_0^\infty \eta\frac{\partial}{\partial\eta}\left(\frac{\sin\xi_0\eta}{\eta}\right)\exp\left[-\eta^2(\theta_1+\delta\theta)\right]d\eta -$$

$$-\frac{2}{\pi}\frac{1}{1-\tau_i}\int_0^\infty\int_0^{\theta_1'} \eta\frac{\partial}{\partial\eta}\left(\frac{\sin\xi_i'\eta}{\eta}\right)\exp\left[-\eta^2(\theta_1+\delta\theta-\theta')\right]d\theta'd\eta$$

$$< \tau(\theta_1) = \tau_i .$$

Hence, if the temperature at the centre falls to τ_i, it will continue to fall below this value. Since τ is now everywhere below the ignition temperature, there is no heat addition and the process becomes one of pure conduction. It is easily shown (e.g. Carslaw and Jaeger (1959) that under these conditions the temperature $\tau(\theta)$ decreases to zero.

2.3. Series approximation for small values of θ

Although representing a complete analytical solution, the complexity of the integral equation for $\xi_i(\theta)$ does not permit solution in closed form. Since the life history of the hot gas bubble is likely to be determined by its behaviour for small time, we derive a solution for $\xi_i(\theta)$ in the form of a polynomial in $\theta^{\frac{1}{2}}$. Let

$$\xi_i(\theta) = \xi_0 - a_1\theta^{\frac{1}{2}} - a_2\theta - a_3\theta^{3/2} \qquad (24)$$

where the a_i are constants depending on the two parameters ξ_0 and τ_i. For very small θ, $\xi_0 - a_1\theta^{\frac{1}{2}}$ will be dominant in expression (24), and as it is expected that the flame initially contracts due to contact with the adjacent cold gas, we should have $a_1 > 0$. For larger time the term $-a_3\theta^{3/2}$ dominates, and so, in order that the flame continues to propagate, it is necessary that $a_3 < 0$.

We make the reasonable assumption that for small θ and θ'

$$\exp\left\{-(\widetilde{\xi}_i+\xi_i)^2/4\theta'\right\} \to 0 \ , \quad \mathrm{erf}\left\{(\widetilde{\xi}_i+\xi_i)/2\sqrt{\theta'}\right\} \to 1 \ .$$

The evaluation of the coefficients is facilitated by writing (24) as $\xi_i = \xi_0 + \theta^{\frac{1}{2}}S(\theta)$, which on substitution

in (21) gives an integral equation for $S(\theta)$. On expanding $S(\theta)$ and equating coefficients of powers of $\theta^{\frac{1}{2}}$ one obtains

$$\operatorname{erf}(\tfrac{1}{2}a_1) = 2\tau_i - 1 , \tag{25}$$

$$a_2 = 2/\xi_0 . \tag{26}$$

In order to obtain a_3 it is necessary to consider the coefficient of θ in (21). The term of lowest order in the first integral of (21) is proportional to $\theta^{3/2}$. Hence, of the source terms, only the second integral need be considered. For small values of θ and θ' we may take

$$\operatorname{erf}\left\{(\tilde{\xi}_i - \tilde{\xi}_i)/2\sqrt{\theta'}\right\} \simeq \operatorname{erf}\left\{a_1(\sqrt{\theta}-\sqrt{\theta-\theta'})/2\sqrt{\theta'}\right\} .$$

It is necessary to evaluate the integral

$$I(a_1) = \int_0^\theta \operatorname{erf}\left\{a_1(\sqrt{\theta}-\sqrt{\theta-\theta'})/2\sqrt{\theta'}\right\} d\theta' .$$

Use of the substitution $y = (\sqrt{\theta}-\sqrt{\theta-\theta'})/\sqrt{\theta'}$ yields

$$I(a_1) = \theta\left\{\operatorname{erf}(\tfrac{a_1}{2}) + a_1^2 \operatorname{erf}(\tfrac{a_1}{2}) - \frac{2a_1}{\sqrt{\pi}}\left[-\tfrac{1}{2}e^{-\tfrac{1}{4}a_1^2} + \right.\right.$$

$$\left.\left. + (1 + \tfrac{1}{2}a_1^2) \int_0^1 \frac{e^{-\tfrac{1}{4}a_1^2 y^2}}{(1+y^2)} \, dy \right. \right. .$$

The integral in the above expression may be evaluated

by use of the technique of differentiation under the
integral sign with respect to a_1; this gives a first
order linear ordinary differential equation which
may be solved to give the required integral. Finally,
comparison of coefficients of θ in (21), and use
of the previous expressions (25) and (26) for a_1, a_2,
gives

$$a_3 = \frac{a_1}{\xi_0^2} - \frac{1}{1-\tau_i} \left[a_1 + \sqrt{\pi}\exp(\tfrac{1}{4}a_1^2)\left\{ 1 + (1+a_1^2)\operatorname{erf}\tfrac{1}{2}a_1 \right\} - \right.$$

$$\left. - \frac{\pi}{2}a_1(1+\tfrac{1}{2}a_1^2)\exp(\tfrac{1}{2}a_1^2)\left\{ 1 - (\operatorname{erf}\tfrac{1}{2}a_1)^2 \right\} \right]. \quad (27)$$

For specified τ_i and ξ_0 all coefficients a_i are
determined. Realistic values of τ_i are greater than
$\tfrac{1}{2}$ and (25) shows that a_1 is then positive. It may
be shown that the second term in (27) is always positive
so that, for a given τ_i, a_3 can be either positive or
negative, depending on the magnitude of ξ_0 .

2.4. Critical conditions

Using the expression for the radius of the flame
front valid for small time, we now determine the
critical conditions for flame propagation. This pro-
cedure is justified since the initial behaviour of the
central gas core determines whether the process will

lead to ignition or extinction. Consider the expression (24) for a given value of τ_i . Initially $\xi_i = \xi_0$ and its value then decreases with increasing θ. If ξ_i falls to zero the process becomes one of pure heat conduction and the incipient flame is eventually extinguished. For ξ_0 sufficiently large, ξ_i is always positive, has a minimum at some $\theta = \theta_c$ and finally increases again with increasing θ. The two possible modes of behaviour of ξ_i are separated by a critical curve which just touches the θ axis. Figure 3.shows qualitatively how ξ_i varies with ξ_0 for a given τ_i .

The critical condition is expressed mathematically by

$$\theta = \theta_c \; : \; \xi_i(\theta_c) = 0 \; , \quad d\xi_i/d\theta = 0 \; . \tag{28}$$

With (24) these conditions become

$$(\xi_0)_c - a_1\theta_c^{\frac{1}{2}} - a_2\theta_c - a_3\theta_c^{3/2} = 0 \; , \tag{29}$$

$$a_1 + 2a_2\theta_c^{\frac{1}{2}} + 3a_3\theta_c = 0 \; . \tag{30}$$

Eliminating a_3 between (29) and (30) and using (26) the following quadratic in $\theta_c^{\frac{1}{2}}$ is obtained:

$$2\theta_c + 2a_1(\xi_0)_c\theta_c^{\frac{1}{2}} - 3(\xi_0^2)_c = 0 \; . \tag{31}$$

The appropriate root of (31) is

$$\theta_c^{\frac{1}{2}} = \tfrac{1}{2}(\xi_o)_c \left\{ (a_1^2 + b)^{\frac{1}{2}} - a_1 \right\}. \qquad (32)$$

Equation (32) shows that the time interval $0 \leq \theta \leq \theta_c$ is small for either $(\xi_o)_c$ small or $\tau_i \to 1$. The critical radius $(\xi_o)_c$, is obtained on substituting for $\theta_c^{\frac{1}{2}}$ in (30). The resulting expression is

$$(\xi_o^2)_c =$$

$$= \frac{\tfrac{1}{3}(1-\tau_i)[4(a_1^2+b)^{\frac{1}{2}} - 2a_1 + 3a_1\{a_1^2 - a_1(a_1^2+b)^{\frac{1}{2}}+3\}]/\{a_1^2 - a_1(a_1^2+b)^{\frac{1}{2}}+3\}}{a_1 + \sqrt{\pi}\{1 + (1+a_1^2)\,\mathrm{erf}\tfrac{1}{2}a_1\}\exp\tfrac{1}{4}a_1^2 - \tfrac{1}{2}\pi a_1(1+\tfrac{1}{2}a_1^2)\{1-(\mathrm{erf}\tfrac{1}{2}a_1)^2\}\exp\tfrac{1}{2}a_1^2}.$$

Values of $(\xi_o)_c$ have been calculated from (25) and (33) for τ_i in the range $\tfrac{1}{2} \leq \tau_i \leq 1$ and are plotted in Fig.4. The curve so obtained has a maximum at $\tau_i = 0.61$ and then gradually decreases, becoming zero at $\tau_i = 1$.

From first considerations one might expect the values of $(\xi_o)_c$ to increase steadily with increasing τ_i, since the unburnt gas would begin to react at a higher temperature, which it could not attain so easily.

This trend is counteracted, however, by the increase in reaction rate with increasing ignition temperature. The appearance of the $(\xi_o)_c$ versus τ_i curve results from the balancing of these two effects. From the arguments above it is seen that initial values in the

region above the curve always lead to ignition whereas those below the curve lead to extinction.

3.   Discussion

Although some of the data is not given, a rough comparison with a result of Arnold and Sherburne is possible by assigning reasonable values to the relevant physical parameters. In their experiments, critical sizes are determined by allowing premixed fuel and air to pass through a nozzle burner, and observing the behaviour of the hot gas bubble formed on sparking the mixture. For propane-air in stoichiometric proportions, they find a critical radius of about 0.3 cm.

From definition (4) it follows that

$$(\zeta_o)_c = \left\{ \frac{\overline{Q}}{k(T_b - T_u)} \right\}^{\frac{1}{2}} r_c \, , \qquad (34)$$

where r_c is the measured critical radius. Taking $\overline{Q} = 10^3$ cal/cm^{3}sec, $k = 2 \times 10$ cal/cm^{o}Ksec, $T_b = 2000^o$K, $T_u = 300^o$K, $r_c = 0.3$cm, one finds that $(\zeta_o)_c = 16.3$.

Comparison shows that this critical radius is about 25 times greater than the maximum value found by us above. Apart from uncertainties in the data, contributing factors to the discrepancy are the following.

i) The reaction rate is assumed of step-function form. With a more conventional rate function a larger reacting volume is required to have the same rate of heat addition at any time.

ii) The gas properties are assumed constant. Expansion effects would tend to increase the critical bubble size.

iii) The initial conditions are not identical with our assumptions. For the experimental bubbles the temperature decreases from the centre outwards.

iv) Schlieren photographs would tend to give an upper limit to the bubble size. Weinberg (1955) has shown that the effective Schlieren radius is at about one and a half times the unburnt gas temperature.

v) Convective effects probably increase the rate of heat transfer from the bubble surface. Heat may also not be conducted away through the electrodes.

vi) The results of Arnold and Sherburne were not obtained at the time origin. In order to find the critical initial bubble radius, the critical bubble size versus time curves were

extrapolated back, thus introducing a
a possible error.

Comparison of our method of approach with that
of Spalding (1958) shows that the two are complementary.
Instead of starting with a temperature profile, whose
dependence on time for a given reaction rate is to be
determined, we choose a profile for the reaction rate
and determine the temperature distribution to match it.
One could, of course, take reaction rate functions
which approximate more closely to conventional ones.
If these are made to consist of a series of straight
lines, the energy conservation equation is still linear
and an analytical solution can be found.

An independent check of this result has been
provided by using the temperature profile method to
evaluate the problem of this chapter. The advantage
of Spalding's method is that one can obtain approximate
solutions of equation (1) for more general reaction
rate functions and non-constant gas properties. Such
a calculation has been undertaken and is presented in
the following chapter, in which also, the two sets of
results are compared.

CHAPTER 3

THE DEPENDENCE OF FLAME IGNITION ON TRANSPORT PROPERTIES AND CHEMICAL KINETIC PARAMETERS

1. Introduction

Although much theoretical work has been done on steady laminar flame propagation, relatively little attention has been paid to transient flames. This is mainly a result of the mathematical difficulties, the relevant partial differential equations being strongly non-linear due to the dependence of chemical reaction rate on temperature and concentrations. Transient flame problems may be divided into two broad classes, (i) ignition problems; in which energy is supplied to a system capable of reacting exothermally and establishing a flame front, (ii) stability studies; in which a steadily propagating flame is subjected to some form of disturbance. In (i) the interest often centres on determining the minimum energy necessary for a flame to establish itself and in (ii) on finding conditions under which the system will once again reach a steadily propagating state.

The present study is concerned with flame ignition
from a spherical hot gas pocket. It is assumed that
at some initial time a mass of hot reaction products
is introduced into a stagnant infinite medium of
premixed fuel and air. Attention has been concentrated
on finding the minimum size of hot gas bubble for which
flame propagation will take place and its dependence on
chemical kinetic parameters and gas transport properties.
In order to facilitate solution, some of the simplifying
assumptions often introduced into steady laminar flame
theory have also been made. Thus it is assumed that
the system is adiabatic and that the chemical reaction
rate may be made temperature explicit.

In the previous chapter, the reaction rate was
taken to be of simple step-function form and the gas
properties assumed independent of temperature. Under
these conditions analytical solution by integral
transform methods was possible, leading to an integral
equation for the effective flame front radius. For more
conventional rate functions analytical solutions cannot
be obtained.

An approximate method for dealing with transient
flame problems, based on a profile technique of boundary

layer theory, has been given by Spalding (1958), who
considered the behaviour of some 'slab' flames. The
transient heat balance equation was studied using the
assumption that the product of the thermal conductivity
and the density of the gas is constant; our assumption
will be less stringent. The heat equation was then
integrated formally over two ranges of the space variable;
it was assumed that the temperature may be expressed as
a simple function containing time-dependent parameters
which were chosen suitably. Thus two ordinary differential
equations for these parameters were obtained. For the
purpose of simplification attention was restricted to
symmetrical profiles and the reaction rate in cooler
half of the flame was neglected.

Within this general framework four problems were
discussed by Spalding, the first being that of the sudden
contact of semi-infinite burned and unburned gas masses;
variation of flame thickness and position of the centre
point of the flame ~~and position of the centre point of
the flame~~ with time were investigated. Clearly in this
problem the maximum temperature remains at its initial
value. Interest was centred on determining the time
for development of a steadily propagating flame, the

velocity of steady propagation, and the stability of the flame. Next was considered the contact of a large mass of cold unburned gas with an adiabatic catalyst; the catalyst has the property of acting as a sink for the reactants so that the concentration of at least one of these is reduced to zero at the catalyst wall. It was found that initially the flame was 'attached' to the catalyst wall where the temperature gradient was initially steeply negative, gradually becoming less negative and finally tending to zero when the second phase commenced; the flame then detached itself from the wall and the equations describing the first problem determined the subsequent behaviour of the flame. Thirdly, Spalding considered the immersion of a finite slab of unburned gas in a large mass of burned; attention was focussed on finding how long it would take for the cold slab to be consumed. Initially the temperature distribution was of rectangular 'well' form. Subsequently flames propagated into the pocket from both sides; finally a composite flame was formed.

Finally Spalding investigated the immersion of a finite slab of burned gas in a large mass of unburned; our interest lies mainly in this study due to its

similarity to our problem. It was found that for
sufficiently large slab widths the temperature at the
centre of the slab remained at its initial value while
the flame spread outwards; this was described by the
equations for the sudden contact of semi-infinite burned
and unburned gas masses. For smaller initial slab widths
the process falls into three parts; initially the
maximum temperature remains at its original value
while the temperature distribution is 'smoothed' out by
conduction. Next the central temperature starts to
fall below its initial value; if this temperature
continues to decrease to the ambient value, the flame
is extinguished. If, on the other hand, this temperature
passes through a minimum and thereafter rises above its
original value, the process is once again described by
the equations of the initial phase and ignition occurs.
A critical initial slab width separating these two
regimes was obtained by Spalding.

Such profile methods, as described above, are
ideally suited for ignition problems and the procedure
has been used by us below.

2.1 Energy conservation equation

Without heat losses and with spherical symmetry, the equation of energy conservation may be written

$$C_p\rho \frac{\partial T}{\partial t} = \rho \frac{\partial}{\partial \psi} (K\rho r^4 \frac{\partial T}{\partial \psi}) + \dot{q}''' \tag{1}$$

where the independent variable ψ, which is effectively a stream function, is defined by

$$\psi = \int_0^r \rho r^2 \, dr \tag{2}$$

Here ρ is assumed a function of T and

$$r = \left\{ 3\int_0^\psi \frac{d\psi}{\rho(T)} \right\}^{1/3} . \tag{3}$$

In the equations above, the variables have the following significance:

r   Radial distance

t   Time

ρ Local gas density

C_p Average specific heat at constant pressure

K   Local thermal conductivity

T   Absolute temperature

$\dot{q}'''$ Volumetric heat release rate due to chemical reaction, in general a function of temperature and fuel concentration. Spalding (1957) has shown that for normal diffusion and certain reactions it may be made a function of temperature only.

To write equation (1) in dimensionless form we introduce the following dimensionless variables:

Stream function

$$d\zeta = \frac{1}{\rho_u} \left\{ \frac{\overline{\dot{q}'''}}{K_u(T_b-T_u)} \right\}^{3/2} d\psi \quad (4)$$

Time

$$d\Theta = \frac{\overline{\dot{q}'''}}{C_p \, \rho_u(T_b-T_u)} \, dt \quad (5)$$

Temperature

$$\tau = \frac{T-T_u}{T_b-T_u} \quad (6)$$

Reaction rate

$$\phi(\tau) = \left(\frac{\rho}{\rho_u}\right)\left(\frac{\dot{q}'''}{\overline{\dot{q}'''}}\right) \quad (7)$$

where

$$\overline{\dot{q}'''} = \int_0^1 \left(\frac{\rho}{\rho_u}\right)\dot{q}''' \, d\tau .$$

The suffices u, b refer to the unburnt and burnt states of the gas respectively.

Introducing the function

$$R = \left(\frac{k\rho}{k_u\rho_u}\right)^{1/4} \left\{ 3 \int_0^\zeta \left(\frac{\rho_u}{\rho}\right)d\zeta \right\}^{1/3} \quad (8)$$

which is proportional to the dimensionless radial distance, the energy conservation equation becomes

$$\frac{\partial \tau}{\partial \Theta} = \frac{\partial}{\partial \zeta} \ (R^4 \ \frac{\partial \tau}{\partial \zeta}) + \emptyset(\tau) \tag{9}$$

2.2 Temperature dependence of transport coefficients

The above formulation assumes that diffusion is normal, namely, that the diffusion coefficients of the reactants are equal to each other and equal to the thermal diffusivity.

In deriving a transient, dimensionless energy equation, Spalding (1958) assumed that $k\rho$ = constant. For the usual constant pressure assumption and ideal gas law this means that k is directly proportional to T. A more convenient form of these relationships is

$$\frac{K}{K_u} = \frac{\rho_u}{\rho} = 1 + a\tau \tag{10}$$

where a is a parameter. For $a = 0$ the gas properties are constant and with increasing a equation (10) approximates to the ideal gas law. By definition

$$a = \frac{k_b}{k_u} - 1 = \frac{\rho_u}{\rho_b} - 1 . \tag{11}$$

With (10), the diffusion coefficients may be written

$$D = (\frac{K_u}{C_p \rho_u}) (1 + a\tau)^2 . \tag{12}$$

The function defined by (8) can now be reduced to

$$R = \left\{ 3 \int_0^{\varsigma} (1 + a\tau) d\varsigma \right\}^{1/3} \tag{13}$$

2.3 Reaction-kinetic assumptions

It is assumed that the flame propagates by means
of a first-order, single-step reaction and that the
conditions under which the reaction rate may be made
temperature-explicit, are satisfied (c.f. Spalding (1957)).
We can then take

$$\emptyset(\tau) = (n+1)(n+2)(1-\tau)\tau^n \tag{14}$$

where   n   is a constant, which in general depends on
the activation energy and on parameter 'a'. For
large activation energy, it may be shown to be nearly

independent of 'a'. In equation (14), τ^n approximates the usual Arrhenius term and $(n+1)(n+2)$ ensures that the area under $\phi(\tau)$, between $\tau = 0$ and $\tau = 1$ is unity.

2.4 Solution by profile method

Integrating the energy equation from $\zeta = 0$ to $\zeta = \infty$, we obtain

$$\frac{d}{d\Theta} \int_0^\infty \tau \, d\zeta = \int_0^\infty \phi(\tau) \, d\zeta \qquad (15)$$

since when $\zeta = \infty$: $\tau = 0$. Similarly an integration from $\zeta = \zeta_i$ to $\zeta = \infty$, where ζ_i is a function of Θ only gives

$$\frac{d}{d\Theta} \int_{\zeta_i}^\infty \tau \, d\zeta + \tau_i \frac{d\zeta_i}{d\Theta} = - R_i^4 \left(\frac{\partial \tau}{\partial \zeta}\right)_{\zeta_i} + \int_{\zeta_i}^\infty \phi(\tau) \, d\zeta \quad (16)$$

where, when $\zeta = \zeta_i$: $\tau = \tau_i$, $R = R_i$. To evaluate the integrals in (15) and (16) we make the profile assumption

$$\tau = \tau_m f\left(\frac{\zeta}{\zeta_i}\right) \tag{17}$$

in which $f(x)$ is a monotonically decreasing function of x, and $f(0) = 1$, $f(\infty) = 0$. τ_m is the temperature at the centre of the gas pocket and is a function of Θ.

It is convenient to introduce the following notation:

$$K_1 = f'(1), \qquad K_2 = f(1),$$

$$J_1 = \int_0^1 f(x)\,dx, \qquad J_2 = \int_1^\infty f(x)\,dx,$$

$$I_1(\tau_m) = \int_0^1 \emptyset\left\{\tau_m f(x)\right\}\,dx, \tag{18}$$

$$I_2(\tau_m) = \int_1^\infty \emptyset\left\{\tau_m f(x)\right\}\,dx.$$

Substituting (17) into (15) and (16), we obtain the two equations

$$(J_1 + J_2)\frac{d}{d\Theta}(\zeta_i \tau_m) = \zeta_i\left\{I_1(\tau_m) + I_2(\tau_m)\right\} \tag{19}$$

$$J_2 \frac{d}{d\theta}(\mathfrak{I}_i \tau_m) + K_2 \tau_m \frac{d\mathfrak{I}_i}{d\theta} = - K_1 [3 \mathfrak{I}_i (1+J_1 a \tau_m)]^{4/3} \frac{\tau_m}{\mathfrak{I}_i} +$$

$$+ \mathfrak{I}_i I_2(\tau_m) \qquad (20)$$

from which the required functions τ_m and $\mathfrak{I}_i$ can be found. K_j , J_j are constants, depending on the choice of profile, and $I_j(\tau_m)$ are determined by $\emptyset(\tau)$ and $f(x)$.

Since θ does not occur explicitly, (19) and (20) can be combined to give a single equation. Solving for the derivatives and dividing one equation by the other we find

$$\frac{d\tau_m}{d\mathfrak{F}_i} = \frac{3\tau_m}{2\mathfrak{I}_i} \left\{ \frac{\xi_i + \psi_1(\tau_m)}{\xi_i + \psi_2(\tau_m)} \right\} \qquad (21)$$

where

$$\mathfrak{I}_i = \frac{1}{3} \xi_i^{-3/2} \qquad (22)$$

and

$$\psi_1(\tau_m) = \frac{1}{9} \frac{(K_2+J_2)I_1(\tau_m) + (K_2-J_1)I_2(\tau_m)}{K_1(J_1+J_2)(1+J_1 a\,\tau_m)^{4/3}\tau_m} \quad ,$$

$$(23)$$

$$\psi_2(\tau_m) = \frac{1}{9} \frac{J_2 I_1(\tau_m) - J_1 I_2(\tau_m)}{K_1(J_1+J_2)(1+J_1 a\,\tau_m)^{4/3}\tau_m} \quad .$$

Equation (21) must be solved subject to the boundary condition

$$\xi_i = (\xi_i)_o \; : \; \tau_m = 1 \qquad\qquad (24)$$

2.5 Approximate analytical solution

In order to obtain an approximate solution of equation (21) we suppose that the reaction rate in the cooler part of the medium may be neglected and hence that $I_2(\tau_m) = 0$; this is equivalent to the assumption made by Spalding (1958). The functions $\psi_1(\tau_m)$, $\psi_2(\tau_m)$ then simplify to

$$\psi_1(\tau_m) = \frac{1}{9} \ \frac{(K_2+J_2)I_1(\tau_m)}{K_1(J_1+J_2)(1+J_1 a\, \tau_m)^{4/3}\tau_m}$$

$$\tag{25}$$

$$\psi_2(\tau_m) = \frac{1}{9} \ \frac{J_2 I_1(\tau_m)}{K_1(J_1+J_2)(1+J_1 a\, \tau_m)^{4/3}\tau_m}$$

Since the K_i, J_i are constants, it is easily seen that $\psi_1(\tau_m)$ and $\psi_2(\tau_m)$ are simply multiples of one another. We now write

$$\psi_1(\tau_m) = -L_1 \ \tau_m^k$$

$$\tag{26}$$

$$\psi_2(\tau_m) = -L_2 \ \tau_m^k$$

where L_1, L_2, k are positive constants. By definition

$$\frac{L_1}{L_2} = 1 + \frac{K_2}{J_2} > 1 \qquad \therefore \ L_1 > L_2$$

If this assumption (26) is written in the form

$$I_1(\tau_m) = \int_0^1 \phi\left\{\tau_m f(x)\right\} \, dx$$

$$= A_1(1+J_1 a\tau_m)^{4/3}\tau_m^{k+1}$$

where $A_1 = -9L_1 K_1 \dfrac{(J_1+J_2)}{K_1+J_2}$, it is readily seen that,

given $f(x)$, this is an integral equation for $\phi(\tau)$. Conversely, given $\phi(\tau)$ as an appropriate power series in τ , the function $f(x)$ is specified.

Under these assumptions equation (21) may be written

$$\frac{d\tau_m}{d\xi_i} = \frac{3\tau_m}{2\xi_i}\left\{\frac{\xi_i - L_1\tau_m^k}{\xi_i - L_2\tau_m^k}\right\} \tag{27}$$

This must be solved subject to the initial condition $\tau_m = 1 : \xi_i = \xi_0$. Integral curves of this equation have a zero gradient along $\xi_i = L_1\tau_m^k$ and infinite gradient along $\xi_i = L_2\tau_m^k$. In the region between these two isoclinals, the integral curves have negative gradient while elsewhere in the positive quadrant the slope is positive. The integral curves satisfying equation (27) will either have a minimum on $\xi_i = L_1\tau_m^k$ or pass through the origin which is a singular point of the equation. The initial value $\xi_i = (\xi_0)_c : \tau_m = 1$

separating these two regimes defines a critical condition for ignition.

On substituting $y = \tau_m^k / \xi_i$ a separable equation is obtained which is readily solved to yield

$$\xi_i^{\frac{3k}{2}-1} = By\left\{(\tfrac{3k}{2}L_1 - L_2)y - (\tfrac{3k}{2} - 1)\right\}^{\sigma} \qquad (28)$$

where $\sigma = \dfrac{-\frac{3k}{2}(L_1-L_2)}{\frac{3k}{2}L_1-L_2}$ and B is a constant.

Use of the initial condition yields

$$(\xi_i)_o^{\frac{3k}{2}-1} = \frac{B}{(\xi_i)_o}\left\{\frac{1}{(\xi_i)_o}(\tfrac{3k}{2}L_1-L_2) - (\tfrac{3k}{2}-1)\right\}^{\sigma}$$

In order that an integral curve should pass through a minimum it must cross the isoclinal of zero slope where $y = \dfrac{1}{L_1}$; hence at the intersection

$$\xi_i^{\frac{3k}{2}-1} = \frac{B}{L_1}\left(1-\frac{L_2}{L_1}\right)^{\sigma} \,.$$

On elimination of B it follows that

$$\left\{\frac{(\xi_i)_o}{\xi_i}\right\}^{\frac{3k}{2}-1} = \frac{L_1}{(\xi_i)_o}\left\{\frac{\frac{1}{(\xi_i)_o}(\tfrac{3k}{2}L_1-L_2)-(\tfrac{3k}{2}-1)}{(\xi_i)_\sigma \left(1 - \frac{L_2}{L_1}\right)}\right\}^{\sigma} \,.$$

To ensure positive real $\bar{\xi}_i$, the expression in brackets on the right hand side of this equation must be positive; the vanishing of this expression yields a critical condition for ignition. Hence

$$(\bar{\xi}_i)_c = L_1 + \frac{(L_1 - L_2)}{\frac{3k}{2} - 1} . \tag{29}$$

For values of $\bar{\xi}_o$ greater than this, extinction will always occur as the integral curves pass through the origin.

The integral curves of the simplified equation (27) exhibit similar characteristics to those of the exact equation and predict an extinction condition.

2.6 Linear profile assumption

As a simple illustration, we shall consider the linear profile

$$\begin{aligned} 0 \leq x \leq 2 : f(x) &= 1 - \tfrac{1}{2}x \\ x \geq 2 : &= 0 \end{aligned} \tag{30}$$

Substituting in (18) we find

$$K_1 = -\frac{1}{2} , \quad K_2 = \frac{1}{2} , \quad J_1 = \frac{3}{4} , \quad J_2 = \frac{1}{4} .$$

For reaction rate function (14) and n large, $I_2(\tau_m)$ is small compared to $I_1(\tau_m)$ and can be neglected (c.f. Spalding (1958)). We then obtain

$$\psi_1(\tau_m) = 3\,\psi_2(\tau_m) = \frac{-\left\{n+2 - (n+1)\tau_m\right\}}{3(1 + \frac{3}{4}a\,\tau_m)^{4/3}}\,\tau_m^{n-1} \quad (31)$$

2.7 Numerical results

We have obtained numerical solutions of equation (21) for the linear profile (30). Typical integral curves for $n = 11$, $a = 1$, are shown in Fig.5. Examination shows that the integral curves have a zero gradient where the numerator of (21) vanishes and infinite gradient when the denominator is zero. These two curves are marked by S_0 and S_∞ in the diagram. For initial conditions (24), step-by-step backward integration results in either integral curves with a minimum on S_0 , (a) and (b), or curves which pass through the singular point $\xi_i = 0$, $\tau_m = 0$, (c) and (d). The former correspond to flame ignition, the central temperature first falling to a minimum and

then rising again to $\mathcal{T}_m = 1$, whereas the latter relate
to flame extinction. The two possible types are
separated by an integral curve which just passes through
the origin and represents the critical state. We have
determined initial conditions $\xi_i = (\xi_i)_c$, $\mathcal{T}_m = 1$,
corresponding to the critical state.

For the linear profile the temperature just
vanishes where $\zeta = 2\xi_i$. It is thus convenient to
put $\delta = 2\xi_i$, where δ is the 'radius' of the hot
gas pocket at any time. Critical values δ_c have been
calculated from (22) for $n = 11, 20$ and values of a ,
the resulting curves being shown in Fig. 6.

2.8 Physical interpretation of results.

Substituting the differential form of (2) in (4)
and integrating

$$\int_0^{\zeta} (1+a\mathcal{T})d\zeta = \tfrac{1}{3}r^3 \left\{ \frac{\overline{\dot{q}'''}}{K_u(T_b-T_u)} \right\}^{3/2} . \qquad (32)$$

For the critical profile $\theta = 0 :$ $\mathcal{T}_c = 1 - \zeta/\delta_c$,
(32) gives

$$(1+\tfrac{1}{2}a)\delta_c = \tfrac{1}{3}r_c^3 \left\{ \overline{\dot{q}'''}/k_u(T_b-T_u) \right\}^{3/2} \qquad (33)$$

where r_c is the measured critical radius.

For large activation energy

$$\frac{\bar{\dot{q}'''}}{(\dot{q}''')^{\ast}} = 1 + a \, \frac{\int_0^1 \tau \, \dot{q}''' \, d\tau}{\int_0^1 \dot{q}''' \, d\tau} \simeq 1 + a \qquad (34)$$

where $(\ )^{\ast}$ is the appropriate variable with $a = 0$. From (33) we obtain

$$\frac{r_c}{r_c^{\ast}} = \frac{(1 + \frac{1}{2}a)^{\frac{1}{3}}}{(1 + a)^{1/2}} \left(\frac{\delta_c}{\delta_c^{\ast}}\right)^{\frac{1}{3}} . \qquad (35)$$

Expression (35) has been evaluated as a function of a using the previous results and is shown in Fig. 7.

The critical radius for constant gas properties can be found from (33). Thus

$$r_c^{\ast} = (3\delta_c^{\ast})^{1/3} \left\{ K_u(T_b - T_u)/(\bar{\dot{q}'''})^{\ast} \right\}^{1/2} . \qquad (36)$$

Taking $(\bar{\dot{q}'''})^{\ast} = 10^3$ cal/cm^{3}sec , $k_u = 2 \times 10^{-4}$ cal/cm$^{\circ}$K sec, $T_b = 2000^{\circ}$K , $T_u = 300^{\circ}$K we find

$$n = 11: \quad r_c^{\neq} = 3.56 \times 10^{-2} \text{ cm} ,$$

$$n = 20: \quad r_c^{\neq} = 3.74 \times 10^{-2} \text{ cm} .$$

For the values chosen, n is nearly independent of a, hence Fig. 7 shows how r_c varies with changing gas properties and approximately constant activation energy parameter. By use of the results of Rosen (1958), n can be related to the usual activation energy.

3. Discussion

One can make general predictions about the general behaviour of the integral curves of equation (21). For large activation energy, $I_2(\tau_m)$ can be neglected compared to $I_1(\tau_m)$, since $\phi(\tau)$ is very peaked, and hence $\psi_1(\tau_m)$ and $\psi_2(\tau_m)$ differ only by a constant multiplier. In general J_1, J_2, $K_2 > 0$ and $K_1 < 0$, so that in the (ξ_i, τ_m) plane there exist limiting curves similar to S_0 and S_∞ where the integral curves have zero and infinite gradients, respectively. The integral curves which satisfy (24) will either have a minimum on S_0 or pass through the singular point $(0,0)$. The starting value $\xi_i = (\xi_i)_c : \tau_m = 1$, separating the two behaviours, determines the critical

state where ignition will just take place.

It is possible to obtain energy criteria based on simple energy balance requirements and these may be expressed in various alternative forms. A useful concept, introduced by Putnam and Jensen (1949), is the dimensionless Peclet group based on laminar flame speed. The critical Peclet number may be written

$$(Pe)_c = 2Su \ r_c/D^x \tag{37}$$

where, for large activation energy, the flame speed is

$$S_u \simeq \frac{1}{C_p \ \rho_u} \left\{ \frac{2k_u \overline{\dot{q}^{nt}}}{(T_b - T_u)} \right\}^{1/2} . \tag{38}$$

Substituting (12), (33) and (38) into (37), this becomes

$$(Pe)_c = 2\sqrt{2} \left\{ 3(1 + \tfrac{1}{2}a)\delta_c \right\}^{1/3} . \tag{39}$$

Function (39) has been plotted in Fig. 8. for various values of n and a . Spalding (1955) has pointed out that although $(Pe)_c$ must vary with different gas conditions its value should be about twelve.

Experimental observations on ignition and initial flame growth in hydrocarbon-air mixtures have been made by Arnold and Sherburne (1953) and by Olsen, Gayhart and Edmondson (1953). Critical sizes were determined by allowing premixed fuel and air to pass through a nozzle burner and observing the behaviour of the hot gas bubbles formed on sparking the mixture. For gas mixtures involving either ethane, methane, butane or propane and air, Arnold and Sherburne found critical radii in the range 0.2 to 0.3 cm. The values calculated by us above agree within an order of magnitude with these measurements.

We now compare the results of this study with those of the previous chapter. Further inspection of the variables used shows that

$$\delta = \frac{2}{3} \, \tilde{\xi}_i^{\,3} \qquad\qquad (40)$$

where $\tilde{\xi}_i$ is the flame radius of Chapter 2. It is also possible to relate the ignition temperature τ_i to the parameter n ; if Υ_i' is taken to be the centroid temperature of the reaction rate function we have

$$\tau_i = \frac{n+1}{n+3} \tag{41}$$

In the previous chapter, thermal and transport properties were assumed constant; this is equivalent to putting the parameter a equal to zero.

With these assumptions, the critical Peclet number becomes

$$(Pe)_c = 2\sqrt{2}.2^{1/3} (\xi_o)_{cr}. \tag{42}$$

Hence for n = 11 , when $\tau_i \simeq .857$, $(Pe)_c \simeq 1.02$, while for n = 20 , when $\tau_i \simeq .913$, $(Pe)_c \simeq 1.53$. These points are shown on the plot of critical Peclet number versus the parameter a (Fig.8); it is shown that the profile method investigation gives better agreement both with Spalding's estimate of twelve and with experimental observations.

The investigations of the two previous chapters have been published as papers (c.f. Zaturska and Adler (1966) and Adler and Zaturska (1966)).

CHAPTER 4

THERMAL EXPLOSION OF A HOT SPOT

1. Introduction

The question of hot spot explosion has been discussed extensively in the literature and several theoretical formulations have been proposed. Such localised regions of high temperature may be produced under impact conditions or by irradiation with light, neutrons, and fission fragments or by the arrival of s shock wave at an impedance discontinuity. They have also been attributed to tribochemical effects i.e. that in some way the combined application of high pressures and the fast shearing of adjacent molecular layers either causes a direct rupture of the molecule or produces sufficient deformation to bring about rapid chemical reaction. Nevertheless there is strong evidence that in the majority of cases the initiation mechanism is predominantly thermal.

Thermal explosion results when the rate of heat production due to chemical reaction exceeds the rate of heat loss to cold surroundings. Under these conditions

the reaction accelerates to a great speed and explosion
may occur.

The physical and chemical changes which take place
in the initiation of such an explosion are completed in
milliseconds, thus imposing limitations on the experi-
mental approach, since elaborate electronic or photographic
equipment is needed to record this rapid sequence of
events.

The various theoretical formulations of this problem
have been compared in survey papers by Thomas (1965) and
Merzhanov (1966). All the authors concerned have neglected
reactant consumption, variation of thermal properties
with temperature, and differences in these between the
interior and exterior of the hot spot. Chemical reaction
has been assumed to be of single-step form, and a
function of temperature alone, but various approximations
to it have been used. In all but one model a spherical
hot spot has been considered, but the method used in
the case of the slab hot spot has been extended to cover
the spherical case.

Briefly the methods may be summarised as follows:
Merzhanov et al. (1963) obtained numerical solutions of
the energy conservation equation using the exponential

approximation to the reaction rate term. Friedman (1963)
considered the case of the slab hot spot but his method
was extended by Thomas (1965) to cover the cylindrical
and spherical cases. It was assumed that chemical
reaction within the hot spot takes place at a constant
rate, while the environment remains inert. A critical
condition was obtained, without actually solving the
energy equation, for which the hot spot temperature fell
after first rising. Boddington (1963) assumed the
exponential approximation for reaction rate and integrated
the energy equation over the hot spot volume, introducing
mean values with respect to volume. He then found the
critical value of the dimensionless parameter for which
the hot spot temperature rose after first falling. Zinn
(1962) considered first the cooling of an inert hot
spot and obtained the time at which cooling by conduction
becomes important and equated this to the adiabatic
induction period of thermal explosion at the initial
hot spot temperature. Thomas proposed a method which
was essentially a combination of the Zinn and Boddington
theories. He also postulated a simple model, equating
the heat required to maintain the initial temperature
indefinitely at the centre of the bubble, with the heat

produced in the hot spot at that temperature; the generation of heat outside the hot spot was disregarded. Each author sought to determine a critical condition separating the regimes of extinction and explosion. The theories were compared on a graph which will be utilised to demonstrate our results.

Hot spots may arise in both liquids and solids; this chapter considers the solid case, while that of the liquid medium will be investigated in Chapter 5.

2. Formulation of the problem

Due to insufficient information on the exact nature of a hot spot, its initial temperature profile is assumed to be rectangular, with the temperature within the hot spot equal to T_1 and that of the cool sur-roundings T_0. Let us denote the radius of the hot spot by a . We make the additional simplifying assumption that reactant consumption may be ignored.

Under these conditions the energy conservation equation with spherical symmetry, constant specific heat and thermal conductivity, is

$$c\rho\frac{\partial T}{\partial t} = \frac{k}{r^2} \frac{\partial}{\partial r} (r^2 \frac{\partial T}{\partial r}) + Qfe^{-E/RT_1} \tag{1}$$

where the notation is identical with that in Chapters 2 and 3. The density, ρ, is plainly constant for solids. Clearly the assumptions of constant thermal properties are not very realistic; they have however been used in all theoretical formulations for the sake of simplicity.

The initial conditions are

$$t = 0 \; : \; T = T_1 \qquad r < a \; ,$$
$$= T_0 \qquad r > a \; , \qquad (2)$$

Mathematically speaking, the difference between this problem and that dealt with previously is that, whereas in the flame ignition problem the temperature was bounded above by the adiabatic flame temperature, that of the hot spot may continue to rise indefinitely, and this situation is to be interpreted as initiation of explosion.

3.1 Reaction rate a linear function of temperature

In view of the chemical reaction rate being of greater significance in the higher temperature range we may write

$$e^{-E/RT} \simeq e^{-E/RT_1} + \frac{(T-T_1)}{RT_1^2} e^{-E/RT_1}$$

$$= e^{-E/RT_1} \tau_\iota$$

where $\quad \tau_\iota = 1 + \frac{E}{RT_1^2}(T-T_1),\quad$ a dimensionless quantity.

We now take

$$e^{-E/RT} = e^{-E/RT_1} \tau_\iota \qquad\qquad \tau_\iota \geq 0$$

$$\qquad\qquad = 0 \qquad\qquad\qquad \tau_\iota \leq 0 \qquad\qquad (3)$$

i.e. chemical reaction rate is taken to be a linear function of a dimensionless temperature.

This enables us to define a convenient set of dimensionless variables, namely:

temperature $\qquad:\qquad \tau_\iota = 1 + \frac{E}{RT_1^2}(T-T_1)$

distance $\qquad\quad:\qquad \xi_\iota = \frac{r}{a}$

time $\qquad\qquad\;\;:\; \theta_\iota = \frac{Qfe^{-E/RT_1} \cdot E}{c\rho\, RT_1^2} \cdot t$.

It is appropriate to introduce the dimensionless parameter

$$\delta = \frac{Qf\ Ea^2 e^{-E/RT_1}}{KRT_1^{\ 2}}$$

We further define

$$-\tau_1 = 1 + \frac{E}{RT_1^{\ 2}}\ (T_0 - T_1)$$

In terms of these new variables the energy equation becomes

$$\frac{\partial \tau_l}{\partial \Theta_l} = \frac{1}{\delta}\ \frac{1}{\xi_l^2}\ \frac{\partial}{\partial \xi_l}\ \left(\ \xi_l^2\ \frac{\partial \tau_l}{\partial \xi_l}\ \right) + \tau_l \quad \text{for } \tau_l \geq 0,$$

$$\tag{4}$$

$$\frac{\partial \tau_l}{\partial \Theta_l} = \frac{1}{\delta}\ \frac{1}{\xi_l^2}\ \frac{\partial}{\partial \xi_l}\ \left(\ \xi_l^2\ \frac{\partial \tau_l}{\partial \xi_l}\ \right) \quad \text{for } \tau_l \leq 0$$

At $\Theta_l = 0$: $\quad \tau_l = 1 \qquad 0 \leq \xi_l \leq 1$,

$$\qquad\qquad\qquad\qquad = -\tau_1 \qquad \xi_l > 1 \ . \tag{5}$$

Substitution of $\omega = \xi_l \tau_l$ yields

$$\frac{\partial \omega}{\partial \Theta_l} = \frac{1}{\delta}\ \frac{\partial^2 \omega}{\partial \xi_l^2} + \emptyset(\omega) \tag{6}$$

where $\phi(\omega) = \omega \qquad \omega \geq 0$

$\qquad\qquad = 0 \qquad \omega \leq 0$.

·The initial and boundary conditions are

$\theta_l = 0 \qquad \omega = \xi_l , \qquad 0 \leq \xi_l \leq 1 ,$

$\qquad\qquad = -\tau_1 \xi_l , \qquad \xi_l > 1 .$

$\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad (7)$

$\theta_l > 0 \qquad \omega = 0 , \qquad \xi_l = 0 ,$

$\qquad\qquad \omega = 0 , \qquad \xi_l = \xi_{l_i} (\theta_l) .$

$\dfrac{\partial \omega}{\partial \xi_l}$ continuous at $\xi_l = \xi_{l_i} (\theta_l)$. i.e. at

$\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \omega = 0.$

This defines the radius of the hot spot $\xi_{l_i}(\theta_l)$ as a function of time; the hot spot is taken to be that part of the medium in which the dimensionless temperature τ_l is greater than or equal to zero. Hence it is assumed that no chemical reaction takes place outside the hot spot. The problem of linear reaction rate in a finite slab, with no flow of heat across one boundary, zero temperature at the other, and zero initial temperature throughout, is considered by Carslaw and Jaeger (1959a) using the Laplace transform method.

3.2 Formal solution of the problem

The temperature distribution due to an instantaneous heat source at ξ_L' and time θ_L' is given by

$$\frac{1}{2\sqrt{\pi}}\frac{\delta\omega(\xi_L',\theta_L')}{(\theta_L-\theta_L')^{1/2}}\left[e^{\frac{-(\xi_L-\xi_L')^2}{4(\theta_L-\theta_L')}}-e^{\frac{-(\xi_L+\xi_L')^2}{4(\theta_L-\theta_L')}}\right]$$

(c.f. Carslaw and Jaeger (1959)).

This is a solution of the heat equation (6) with zero reaction rate, and represents the instantaneous generation of a quantity of heat. We may regard our problem as a continuous distribution of such sources from distance 0 to $\xi_{L_i}(\theta_L)$, and over time 0 to θ_L . Further, we have to incorporate the initial 'step' condition.

Thus

$$\omega(\xi_L,\theta_L)=\frac{1}{2\sqrt{\pi}}\int_0^{\theta_L}\int_0^{\xi_{L_i}(\theta_L)}\frac{\delta\omega(\xi_L',\theta_L')}{(\theta_L-\theta_L')^{1/2}}\left\{e^{\frac{-(\xi_L-\xi_L')^2}{4(\theta_L-\theta_L')}}-\right.$$

$$\left.e^{\frac{-(\xi_L+\xi_L')^2}{4(\theta_L-\theta_L')}}\right\}\,d\xi_L'\,d\theta_L'$$

$$+\frac{1}{2\sqrt{\pi}}\int_0^{\infty}\frac{\xi_L'\tau_k'}{\theta_L^{1/2}}\left\{e^{\frac{-(\xi_L-\xi_L')}{4\theta_L}}-e^{\frac{-(\xi_L+\xi_L')^2}{4\theta_L}}\right\}\,d\xi_L' \quad (8)$$

where $\tau_k = 1$ $\quad 0 \leq \xi_L < 1$

$$= -\tau_1 \qquad \xi_L > 1 \tag{9}$$

The second integral in this expression (8) represents the temperature distribution due to the initial 'step' temperature profile.

Denote the temperature at the centre of the hot spot $\tau_L(0, \theta_L)$ by $\tau_L(\theta_L)$.

Then $\quad \tau_L(\theta_L) = \lim\limits_{\xi_L \to 0} \dfrac{\omega(\xi_L, \theta_L)}{\xi_L}$.

Consequently

$$\tau_L(\theta_L) = \frac{d}{2\sqrt{\pi}} \int_0^{\theta_L} \int_0^{\xi_{Li}(\theta_L)} \frac{\xi_L \, \omega(\xi_L, \theta_L')}{(\theta_L - \theta_L')^{3/2}} e^{\xi_L^2/4(\theta_L - \theta_L')} \, d\xi_L \, d\theta_L'$$

$$+ \frac{1}{2\sqrt{\pi}} \int_0^{\infty} \frac{\xi_L^2 \tau_k}{\theta_L^{3/2}} e^{-\xi_L^2/4\theta_L} \, d\xi_L \tag{10}$$

Our concern is primarily with the ignition process, i.e. with small values of time. It is reasonable to expect that, for such a situation, ξ_{Li} remains approximately equal to unity, while the temperature profile is not altered appreciably, and hence

$$\omega \left(\xi_\iota , \theta_\iota' \right) \simeq \xi_\iota \tag{10a}$$

Under these assumptions we have

$$\tau_\iota(\theta_\iota) = \frac{\delta}{2\sqrt{\pi}} \int_0^{\theta_\iota} \int_0^1 \frac{\xi_\iota^2 \, e^{-\xi_\iota^2/4(\theta_\iota - \theta_\iota')}}{(\theta_\iota - \theta_\iota')^{3/2}} \, d\xi_\iota \, d\theta_\iota'$$

$$+ \frac{1}{2\sqrt{\pi}} \int_0^1 \frac{\xi_\iota^2}{\theta_\iota^{3/2}} \, e^{-\xi_\iota^2/4\theta_\iota} \, d\xi_\iota$$

$$- \frac{\tau_1}{2\sqrt{\pi}} \int_1^\infty \frac{\xi_\iota^2}{\theta_\iota^{3/2}} \, e^{-\xi_\iota^2/4\theta_\iota} \, d\xi_\iota \tag{11}$$

which, on simplication, may be written

$$\tau_\iota(\theta_\iota) = \frac{4}{\sqrt{\pi}} \int_0^\sigma z^2 e^{-z^2} dz - \frac{4\tau_1}{\sqrt{\pi}} \int_\sigma^\infty z^2 e^{-z^2} dz$$

$$+ \frac{\delta}{\sigma^2} \int_0^\sigma z.\text{erfc}\,z.dz \tag{12}$$

where $\sigma = \dfrac{1}{2\sqrt{\theta_\iota}}$.

Differentiation with respect to θ_ι yields

$$\frac{d\tau_\iota}{d\theta_\iota} = -2\left[\frac{4}{\sqrt{\pi}}(1 + \tau_1)\sigma\, e^{-\sigma^2} - \frac{2\delta}{\sqrt{\pi}}\left\{-\frac{\sigma e^{-\sigma^2}}{2} + \frac{\sqrt{\pi}}{4}\,\text{erf}\,\sigma\right\}\right]. \tag{13}$$

Initially $\theta_\iota = 0$, i.e. $\sigma \longrightarrow \infty$. Thus, initially,

$\dfrac{d\tau_t}{d\Theta_t} = \delta$ which is always positive by definition. We deduce that the temperature at the centre of the hot spot always rises initially.

3.3 Critical Condition for ignition

Consider the expression for $\tau_t(\Theta_t)$, the central temperature of the hot spot. Initially this is an increasing function of time. If this should continue to rise indefinitely, ignition and finally explosion, would result. The case of this function rising and subsequently decreasing would describe a subcritical state.

At the outset, when σ is large, the δ term dominates in (13); this is strictly positive. We may rewrite (13) in the form

$$\frac{d\tau_t}{d\Theta_t} = \sigma^5 e^{-\sigma^2}[-\frac{8}{\sqrt{\pi}}(1+\tau_1) + \frac{\delta}{A(\sigma)}] \tag{14}$$

where $A(\sigma)$ is given by

$$A(\sigma) = \frac{\sigma^5 e^{-\sigma^2}}{\text{erf}\,\sigma - \frac{2\sigma}{\sqrt{\pi}}\,e^{-\sigma^2}} \geq 0. \tag{15}$$

The minimum value of this positive term in δ, i.e. the maximum value of $A(\sigma)$, gives the position where the sign of $\dfrac{d\tau_\iota}{d\Theta_\iota}$ is most likely to become negative. If $\dfrac{d\tau_\iota}{d\Theta_\iota}$ is exactly equal to zero at this point, a critical condition is obtained.

Calculation has shown that the maximum of A is attained for $\sigma = 1.365$ where $A = 1.0396$ (to 4 decimal places). Figure (9) shows a plot of $A(\sigma)$ against σ. The critical condition, where $\dfrac{d\tau_\iota}{d\Theta_\iota} = 0$, is hence given by the expression

$$\delta_{cr} = 4.689\ (1 + \tau_1)\ . \tag{16}$$

3.4 Discussion

The various critical conditions obtained by the several formulations are compared in the paper of Thomas (1965); a plot of δ_{cr} versus $(1 + \tau_1)$ is given. Figure (10) reproduces this graph incorporating the results obtained in this section and in the next. It may be seen that the relation obtained in (16) is almost identical with that obtained by the method of Friedman, which assumed constant reaction rate within the hot spot regardless of temperature, while outside

the hot spot the reaction rate was assumed to be zero
i.e. a step-function reaction rate. It has been shown
by Merzhanov (1966) that this approximation gives good
results for low values of $(1+\tau_1)$. Low values of
$(1+\tau_1)$ correspond to low values of the activation energy
E , and hence are suitable for a linear reaction rate
assumption. Consequently we may expect to find good
agreement in this range. For higher activation energies
a polynomial reaction rate is more applicable and this
will be considered in the following section.

Merzhanov's exact numerical computation also shows
that for intermediate and higher values of δ , the
temperature at the centre of the bubble rises initially;
this behaviour is shown in our model.

The remarkable similarity of our result and that
of Friedman deserves further comment. Although our
reaction rate is a linear function of temperature and
not a step-function; the additional assumptions (10a),
valid for small values of time, are in fact equivalent
to a step function assumption. However, the expression
(10) is in fact a complete analytical expression for the
central temperature with a linear reaction rate function.

4.1 Polynomial form of reaction rate function

The reaction rate was assumed to be zeroth order and of single-step form. It has been shown by Rosen (1958) that the Arrhenius term may be approximated in the following way

$$e^{-E/RT} \simeq e^{-E/RT_1} \left(\frac{T-T_o}{T_1-T_o} \right)^n \tag{17}$$

where $n = \dfrac{E(T_1-T_o)}{RT_1^2}$. This is obtained by equating the Arrhenius reaction rate function and its gradient to the corresponding approximations, at the upper temperature T_1 , where reaction rate is more significant.

4.2 Dimensionless form of the equations

The approximation (17) enables us to introduce the following dimensionless variables

Temperature: $\quad \mathcal{T} = \dfrac{T-T_o}{T_1-T_o}$

Time $\quad : \quad \Theta = \dfrac{Qfe^{-E/RT_1}}{\rho C(T_1-T_o)} \cdot t$.

Distance $\quad : \quad \xi = \left(\dfrac{Qfe^{-E/RT_1}}{K(T_1-T_o)} \right)^{1/2} r$

Written in terms of these new variables, the energy equation is

$$\frac{\partial \tau}{\partial \Theta} = \frac{1}{\xi^2} \frac{\partial}{\partial \xi} \left(\xi^2 \frac{\partial \tau}{\partial \xi} \right) + \tau^n \tag{18}$$

with initial conditions

$$\Theta = 0 : \quad \tau = 1 \quad , \quad 0 \le \xi < \xi_o \, ,$$
$$= 0 \quad , \quad \xi > \xi_o \, , \tag{19}$$

where ξ_o denotes the initial dimensionless radius of the hot spot.

4.3 Solution by profile method

For convenience, we rewrite the energy conservation equation in terms of $\omega = \xi \tau$.

We have

$$\frac{\partial \omega}{\partial \Theta} = \frac{\partial^2 \omega}{\partial \xi^2} + \frac{\omega^n}{\xi^{n-1}} \tag{20}$$

with $\Theta = 0 : \quad \omega = \xi \, , \quad 0 \le \xi > \xi_o \, ,$
$$= 0 \, , \quad \xi > \xi_o \, ,$$

72.

We denote by ξ_i the distance from the centre of the hot spot at which some fixed value of the temperature is attained. This value is somewhat arbitrary and in this sense ξ_i is analogous to boundary layer thickness; its physical significance not immediately apparent. Consequently we make use of the profile method again.

We make the profile assumption

$$\tau = \tau_m f\left(\xi^2 / \xi_i^2 \right) \tag{21}$$

where f satisfies the following:

$f(0) = 1$; $f(\infty) = 0$; $f'(\infty) = 0$; and $f'(x) \leq 0$ for all $x \geq 0$.

τ_m denotes the maximum or central temperature of the hot spot, and is a function of time, θ, alone, as also is ξ_i . An even function of ξ is used to ensure zero temperature gradient at the centre of the hot spot.

Integration of the energy equation (20) in the ξ - direction from $\xi = \lambda \xi_i$ to $\xi = \infty$, where λ is an arbitrary constant and $0 \leq \lambda < 1$, and use of the

profile assumption, gives

$$\frac{d}{d\Theta}\left\{\frac{\tau_m \xi_i^2}{2}\right\} \int_{\lambda^2}^{\infty} f(\eta)\,d\eta + \frac{\lambda^2 f(\lambda^2)}{2} \tau_m \frac{d}{d\Theta}(\xi_i^2)$$

$$= -\tau_m\left\{f(\lambda^2) + 2\lambda^2 f'(\lambda^2)\right\} + \frac{\tau_m^n \xi_i^2}{2}\int_{\lambda^2}^{\infty} f^n(\eta)\,d\eta \quad (22)$$

The significant part of the ignition process takes place close to the centre of the hot spot; for this reason our interest lies in small values of λ. Consequently we expand the equation (22) in ascending powers of λ^2 and compare coefficients of terms in λ^0 and λ^2, since we have only two time-dependent parameters and hence require only two equations.

It follows that

$$A_1 \frac{d}{d\Theta}\left(\frac{\tau_m \xi_i^2}{2}\right) = -\tau_m + \frac{\tau_m^n \xi_i^2}{2} A_n \quad (23)$$

and

$$\frac{d\tau_m}{d\Theta} = \frac{2}{\xi_i^2}\left\{3\tau_m f'(0) + \frac{\tau_m^n \xi_i^2}{2}\right\} \quad (24)$$

where
$$A_1 = \int_0^\infty f(\eta)\,d\eta$$

$$A_n = \int_0^\infty f^n(\eta)\,d\eta$$

In obtaining these, it is assumed that $f(x)$ may be expanded in a Taylor series about the origin.

Since Θ does not occur explicitly, equations (23) and (24) may be combined to give a single equation. Solving for the derivatives and dividing one equation by the other, we have

$$\frac{d\,\tau_m}{d\,\xi_i^2} = \frac{A_1\,\tau_m}{\xi_i^2}\left\{\frac{6f'(0) + \tau_m^{n-1}\,\xi_i^2}{-2(1+3f'(0)A_1) + (A_n-A_1)\,\tau_m^{n-1}\,\xi_i^2}\right\}. \tag{25.}$$

This equation must be solved subject to the boundary condition

$$\xi_i^2 = \xi_o^2 \;:\; \tau_m = 1. \tag{26}$$

The integral curves of this equation have zero gradient where the numerator of (25) is zero, and infinite gradient where the denominator vanishes. These curves, S_o and S_∞, are shown qualitatively in Figure 11.,

which also indicates the behaviour of integral curves of the equation.

On substituting $y = \tau_m^{n-1}$, $\zeta = \dfrac{1}{\xi_i^2}$ equation (26) becomes

$$\frac{dv}{d\zeta} = \frac{-(n-1)A_1}{\zeta}\left\{ \frac{6f'(0)\zeta + y}{-2(1+3\,f'(0)A_1)\zeta + (A_n-A_1)y} \right\} \quad (27)$$

which is a first order ordinary differential equation of the homogeneous type. The solution of (25) is thus

$$\left(\tau_m^{n-1}\frac{\xi_i^2}{\xi_0^2}\right)^{A}\frac{\xi_i^2}{\xi_0^2}\left\{[-2+6f'(0)A_1(m-2)] + \tau_m^{n-1}\,\xi_i^2[A_1(n-2)\right.$$

$$\left. + A_n]\right\}^{\dfrac{B}{A_1(n-2)+A_n}}$$

$$= \left\{[-2+6f'(0)A_1(n-2)] + \xi_0^2[A_1(n-2)+A_n]\right\}^{\dfrac{B}{A_1(n-2)+A_n}} \quad (28)$$

where A and B are given by

$$A = \frac{1+3f'(0)A_1}{-1+3f'(0)A_1(n-2)}$$

$$(29)$$

$$B = \frac{-A_1\left\{(n-1)+6f'(0)A_n\right\}}{-1 + 3f'(0)A_1(n-2)} \; .$$

4.4 Critical conditions section

Equation (28) may be rewritten in the form

$$\frac{-C + \tau_m^{n-1} \xi_i^2 D}{-C + \xi_o^2 D} = \left(\frac{\xi_o}{\xi_i}\right)^{2(F+1)} \left(\frac{1}{\tau_m^{n-1}}\right)^F = P(\xi_i, \tau_m)$$

where C, D and F are positive and dependent only on n and the assumed profile; since we are concerned with real, positive ξ_i, τ_m, $P(\xi_i, \tau_m)$ is therefore essentially positive with $P(\xi_o, 1) = 1$. Consequently the above expression may be rearranged to give

$$\tau_m^{n-1} = \frac{C + (-C + \xi_o^2 D) P(\xi_i, \tau_m)}{D \xi_i^2} .$$

Now, $\xi_o^2 D > C$ always gives a positive value for τ_m, hence describing a supercritical case; $C > \xi_o^2 D$ makes it possible for τ_m to become negative, so becoming equal to zero at some stage — extinction. The case of $\xi_o^2 D = C$ may hence be regarded as the critical condition separating these two regimes.

This condition may be written

$$(\xi_o^2)_{crit} = \frac{2-6A_1 f'(0)(n-2)}{A_1(n-2) + A_n} \cdot \tag{30}$$

The corresponding critical integral curve is

$$\tau_m^{n-1} \xi_i^2 = \frac{2-6f'(0)A_1(n-2)}{A_1(n-2) + A_n}$$

$$= -6f'(0) + \frac{6f'(0)A_n+2}{A_n+A_1(n-2)}$$

$$> - 6f'(0) \qquad \text{since} \quad A_n \searrow 0 \quad \text{as} \quad n \to \infty \text{ and}$$

the expression is valid for all m.

Hence it lies to the right of S_o as expected.

In order to compare this result with those of other authors, we consider the parameter $\delta_{cr} = (\xi_o^2)_{crit} \, n$. The parameter n is identical with the Θ_o of Thomas and with the $(1+\tau_1)$ of the previous investigation.

Hence

$$\delta_{cr} = n \cdot \frac{2-6A_1 f'(0)(n-2)}{A_1(n-2) + A_n} \cdot \tag{31}$$

For large values of n it may be seen that

$$\delta_{cr} \longrightarrow -6f'(0) \cdot n. \tag{32}$$

since A_n decreases to zero with increasing n . Thus δ_{cr} depends only on the value of $f'(0)$ and not on the exact nature of the profile used, for large n . But the profile assumption may be written

$$\tau = \tau_m \left\{ 1 + \frac{\xi^2}{\xi_i^2} f'(0) + \ldots \right\} \tag{33}$$

since it is assumed possible to expand f in a Taylor series about the origin. For this reason, a simple parabolic profile would appear most natural as it does not involve any unnecessary information.

We therefore write

$$\tau = \tau_m \left(1 - \frac{\xi^2}{\xi_i^2} \right) \quad , \quad 0 \leq \xi \leq \xi_i \quad ,$$

$$= 0 \quad , \quad \xi \geq \xi_i .$$

The critical condition for this profile becomes

$$\delta_{cr} = \frac{2(n+1)(3n-4)}{(n-1)} . \tag{34}$$

For large n , $\delta_{cr} \rightarrow 6n$.

4.5 Discussion

Our interest lies mainly in the region $10 < n < 20$, for which it may be seen that in the parabolic profile case δ_{cr} is very close to $6n$. This is somewhat high and one might hope to improve on it by a more realistic form of profile assumption. Use of an exponential profile, $f(x) = e^{-\alpha x}$, would appear to be more representative of reality, but since there is no precise way of defining $f(1)$ a degree of arbitrariness is introduced in the form of the parameter α. Such a profile would yield a critical condition of

$$\delta_{cr} = 2\alpha \left(\frac{n}{n-1}\right)^2 [1+3(n-2)]$$

The parameter α is indeterminate inasmuch as there is no unique way of defining the size of the hot spot at any time, other than as that part of the medium wherein the temperature is above a given fraction of the central hot spot temperature. This would be analogous to the concept of boundary layer thickness, but as our interest in ignition is not only of a qualitative, but also of a quantitative nature, such profiles are not suitable for our purposes. Spalding (1958) too,

in his adaptation of profile methods to ignition problems has used only the simpler, well-defined profiles (e.g. linear profile) which have a discontinuity in the gradient at the hot pocket interface.

The result of the parabolic profile case is plotted on the same graph as our previous investigation (see Figure 10.). It compares favourably with the criteria obtained by other authors, although somewhat higher. Since the accuracy of profile methods is to within the order of 20% this shows good agreement.

Further study of the integral curves of Figure 11. indicates that except for very small hot spots, the central temperature always rises before subsequently falling, or just continues to increase with time. This is in agreement with the exact numerical results of Merzhanov et al (1963).

It is worth noting that although the choice of profile is restricted practically by the form of the reaction rate function, and hence the computation of A_n , this has no effect on the result for large values of n , and for such n the qualitative behaviour of δ_{cr} is identical for all profiles.

5. Conclusion

Comparison of the results obtained in the investigations of sections 3 and 4 of this chapter shows that in both cases the temperature of the hot spot centre rises first before falling. This is fully in agreement with the exact work of Merzhanov et al. although our results are higher. The first study is valid for lower values of the activation energy E and is a good approximation for such values.

Profile methods have been found to give reasonable agreement quantitatively although the use of crude profiles appears preferable. The use of 'smooth' profiles has been found inadequate except for quali- tative studies. The limitation of profile methods has been shown in their arbitrary nature.

CHAPTER 5

THERMAL STABILITY OF A HOT INERT GAS BUBBLE
IN A COOL REACTIVE LIQUID

1. Introduction

The problem of the behaviour of a hot gas bubble in a cool reactive liquid medium has not yet attracted attention, in spite of its obvious application in liquid fuel rockets and chemical reactors. Such hot spots may also arise in explosives from (i) the adiabatic compression and heating of enclosed gas spaces, (ii) the reinforcement or the local distortion of the explosive into fine particles and, (iii), the formation of micro Munroe jets. For gentler shocks it is necessary for some discontinuity, cavity or bubble to be present where the energy can be concentrated and a local hot spot formed.

Case (i) could be achieved practically by subjecting the fuel or explosive to some form of impact e.g. by accidentally dropping it. The importance of micro Munroe jets in initiation and growth of explosion has been studied experimentally by Bowden and McOnie (1967). In their experiments nitroglycerine was placed on a flat anvil and subjected

to impact by a flat hammer.   It was shown that micro Munroe jets can be formed at the surface of a compressed gas bubble;   these jets are projected at high speed into the gas as the compression continues.   It is believed that these jets facilitate initiation of explosion by dispersing the liquid within a bubble of hot compressed gas and by increasing the impact velocity of the liquid.   Such jets are also formed between two drops of explosive coalescing on impact.

Initiation of explosion in liquids, whether by impact or shock, is generally held to be thermal in origin.   As such it is an obvious extension of conventional hot spot theory.

Work on problems of a similar nature appears to be limited to considerations of the dynamics of phase growth. Scriven (1959) has formulated the general case of equations governing a spherical vapour bubble growing in a quiescent, superheated liquid of infinite extent.   These are then simplified to describe growth controlled by the transport of heat and matter.   Similarity solutions are then obtained for conditions typical of bubble growth in the nucleate boiling of a) pure materials and b) binary mixtures.   The effect of radial convection resulting from

unequal phase densities is then established, the position of the phase boundary being asymptotically proportional to the square root of time. Although the chemical reaction rate term is included in the initial formulation of the problem, this is subsequently ignored.

Much consideration has also been given to the theory of underwater explosions (c.f. Cole (1948)). This regards the explosion as a chemical reaction in a substance which converts the original material into a gas at very high temperature and pressure, the process occurring with extreme rapidity and evolving a great deal of heat. Reactions of this kind can be initiated if sufficient energy is provided at some point of the explosive. A shock wave is set up by the arrival of the pressure wave at the water-explosive interface. Subsequently motion of the gas sphere is considered and it is shown, both theoretically and experimentally, that the bubble radius oscillates in time. Only hydrodynamical effects are considered; the bubble sizes considered in this kind of theory, although dependent on the charge used, are nevertheless large e.g. of the order of one foot in radius.

Our problem deals with minute bubbles, of order 10^{-3} cms. or so, where temperature gradients have much

greater significance. Thus our approach studies thermal as well as hydrodynamical effects; nevertheless a useful energy conservation equation derived from the first time integral of the equation of motion appears in both theories.

2.1. Formulation of the problem

The formulation naturally falls into two parts:

Gas bubble:-

For simplicity it is assumed that the gas of the bubble obeys the ideal gas law. Since the bubble is small, it is convenient to take pressure, temperature and density within the bubble to be uniform at any instant.

Hence the equation of state may be written

$$\left(\frac{p_s}{p_1}\right)\left(\frac{\mathbb{R}}{\mathbb{R}_1}\right)^3 = \frac{T_s}{T_1} \tag{1}$$

where p_s, T_s, $\mathbb{R}$ denote the pressure, temperature and radius of the bubble, while the suffix 1 refers to initial values of these variables.

We introduce an extra parameter associated with the equation of state, namely the radius $\mathbb{R}_o$ to which the bubble radius would tend as it approached a state of thermal equilibrium; this is defined by

$$\left(\frac{R_o}{R_1}\right)^3 = \frac{T_o}{T_1} \qquad (2)$$

where T_o denotes the initial ambient temperature and hence $T_o < T_1$.

<u>Liquid</u>:-

The liquid is taken to be inviscid, irrotational, incompressible, and under the action of no body forces. Further, the system is assumed to be spherically symmetrical. Under these conditions the equation of continuity is

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 v_r) = 0 \qquad (3)$$

where v_r is the radial fluid velocity. Since there is no flow across the bubble interface

$$(v_r)_{r=R} = \dot{R}(t) \qquad (4)$$

which simplifies to

$$v_r = \left(\frac{R}{r}\right)^2 \dot{R}(t) . \qquad (5)$$

The independent variables r and t denote radial distance and time respectively. The radial momentum equation is

$$\frac{\partial v_r}{\partial t} + v_r \frac{\partial v_r}{\partial r} = -\frac{1}{\rho_L} \frac{\partial p}{\partial r} \qquad (6)$$

where p, ρ_L are the pressure and density of the fluid, the latter assumed constant. This, together with (5), on integrating over the space variable, gives at $r = \mathcal{R}$

$$\frac{1}{\rho_L}(p_s - p_l) = \frac{3}{2}\dot{\mathcal{R}}^2 + \mathcal{R}\ddot{\mathcal{R}} \qquad (7)$$

which may be rewritten in the form

$$(p_s - p_l)\frac{d}{dt}(\frac{4}{3}\pi\mathcal{R}^3) = \frac{\rho_L}{2}\frac{d}{dt}(4\pi\mathcal{R}^3\dot{\mathcal{R}}^2) \qquad (8)$$

thus expressing conservation of inertial energy; the expression on the right hand side is a rate of change of kinetic energy. This is exactly equivalent to the energy conservation energy given by Cole (1948).

The heat conservation equation for the fluid may be written

$$c_p\rho_L(\frac{\partial T}{\partial t} + v_r\frac{\partial T}{\partial r}) = \frac{k}{r^2}\frac{\partial}{\partial r}(r^2\frac{\partial T}{\partial r}) + \dot{q}''' \qquad (9)$$

where T denotes temperature within the liquid, c_p, k are the specific heat at constant pressure and the thermal conductivity of the liquid and are both assumed constant, while $\dot{q}'''$ is the volumetric heat release rate due to chemical reaction.

A zeroth order reaction with no reactant consumption is assumed, and the reaction rate term is taken to be of standard Arrhenius form.

The boundary condition at the bubble interface, which expresses the balance of the rate of inflow of heat into the bubble, and the rate of increase of heat within it, making use of the combined first and second laws of thermodynamics, is

$$4\pi R^2 k \left(\frac{\partial T}{\partial r}\right)_{r=R} = C_v \left(\frac{\partial T}{\partial t}\right)_{r=R} + p_s \cdot 4\pi R^2 \dot{R} \qquad (10)$$

where C_v is the constant thermal capacity of the gas bubble.

2.2. Dimensionless form of the equations

To write the above equations in dimensionless form, we introduce the following dimensionless variables:

distance
$$\zeta = \sqrt{\frac{Q f e^{-E/RT_1}}{K(T_1 - T_0)}} \cdot r$$

time
$$\theta = \frac{Q f e^{-E/RT_1}}{\rho_L C_p (T_1 - T_0)} \cdot t$$

temperature
$$\tau = \frac{T - T_0}{T_1 - T_0}$$

pressure
$$\pi_s = \frac{p_s}{p_1}$$

bubble radius $\qquad \eta = \sqrt{\dfrac{Qfe^{-E/RT_1}}{K(T_1 - T_o)} \cdot \mathcal{R}}$

where the reaction rate term is approximated by

$$\dot{q}''' = Qfe^{-E/RT}$$

$$\simeq Qfe^{-E/RT_1} \cdot \left(\frac{T - T_o}{T_1 - T_o}\right)^n$$

and n is given by

$$n = \frac{E}{RT_1^2}(T_1 - T_o)$$

as in previous chapters.   This gives a very good degree of accuracy for n of order 10. T_o denotes the initial temperature of the cool liquid.

Written in terms of these new variables the equations are:

Equation of state

$$\tau_s = \frac{\pi_s \left(\frac{\eta}{\eta_o}\right)^3 - 1}{\left(\frac{\eta_1}{\eta_o}\right)^3 - 1} \qquad (11)$$

Momentum

$$3\dot{\eta}^2 + 2\eta\ddot{\eta} = \alpha_3(\pi_s - 1) \qquad (12)$$

Energy conservation

$$\frac{\partial \tau}{\partial \theta} + \frac{\eta^2}{\xi^2} \dot{\eta} \frac{\partial \tau}{\partial \xi} = \frac{1}{\xi^2} \frac{\partial}{\partial \xi} \left(\xi^2 \frac{\partial \tau}{\partial \xi} \right) + \tau^n \qquad (13)$$

while initial conditions are:

$$\theta = 0 \quad : \quad \tau_s = 1$$

$$\eta = \eta_1$$

$$\tau = 0 \quad \text{for} \quad \xi > \eta_1$$

$$\pi_s = 1$$

$$\dot{\eta} = \ddot{\eta} = 0 \ ,$$

and for $\theta > 0$ we have

$$\left(\frac{\partial \tau}{\partial \xi} \right)_{\xi = \eta} = \frac{\alpha_1}{\eta^2} \left(\frac{\partial \tau}{\partial \theta} \right)_{\xi = \eta} + \alpha_2 \pi_s \dot{\eta} \ . \qquad (15)$$

The parameters α_1, α_2, α_3 are defined by

$$\alpha_1 = \frac{1}{3} \frac{C_v}{\frac{4}{3} \pi R_1^3 \rho_L C_p} \cdot \eta_1^3 = \Gamma^1 \eta_1^3$$

$$\alpha_2 = \frac{p_1}{\rho_L C_p (T_1 - T_o)} \qquad (16)$$

$$\alpha_3 = \frac{2 p_1}{\rho_L} \frac{K(T_1 - T_o)}{Q f e^{-E/RT_1}} \cdot \frac{1}{\left(\frac{K}{C_p \rho_L} \right)^2}$$

and together with η_1, η_o, and n, a six-parameter system is defined.

2.3. Physical discussion of parameters

In order to reduce the system to one of only four parameters we make the following simplifying assumptions

$$\eta_o = 1$$

$$\alpha_3 = 1 \quad .$$

(17)

It may be seen from (16) that Γ represents the ratio of thermal capacity of the gas bubble to that of an equal volume of liquid; due to low density of gases this is a very small quantity unless a highly compressed gas bubble is considered. In fact, generally $\alpha_1 \sim 3 \times 10^{-4}$. Nevertheless it cannot be neglected as the term $\frac{\alpha_1}{\eta^2}(\frac{\partial \tau}{\partial \theta})_{\xi = \eta}$ is of great significance in the early stages of the process, where the temperature at the bubble surface falls very rapidly. Neglect of this term would lower the order of the equations; hence the problem is suited to the methods of singular perturbation theory.

The parameter α_2 represents the ratio of energy transfer by inertia forces to that by thermal effects and has magnitude of order unity.

The introduction of a velocity U, defined by

$$U = \frac{K}{C_p \rho_L R_o} = \frac{K}{C_p \rho_L} \left\{ \frac{Q f e^{-E/RT_1}}{K(T_1 - T_o)} \right\}^{1/2}$$

(18)

and analogous to the laminar flame speed enables α_3 to be written

$$\alpha_3 = \frac{p_1}{\frac{1}{2}\rho_L U^2} = 1 \tag{19}$$

which may then be interpreted as the ratio of internal energy to kinetic energy of disturbance.

Similarly

$$\eta_1 = \frac{\mathcal{R}_1 U}{\dfrac{K}{c_p \rho_L}} \tag{20}$$

has the form of a Peclet number based on this velocity U.

The assumptions (17) imply a value for $\mathcal{R}_o$ of order 10^{-4} cms., which is of the desired magnitude. T_1 and T_o are assumed to be approximately $1000^{\circ}K$ and $300^{\circ}K$ respectively.

2.4.   Solution by profile method

On multiplying the energy conservation equation (13) by ξ^2 and integrating over the space variable from η to ∞, we obtain

$$\frac{d}{d\theta} \int_{\eta}^{\infty} \xi^2 \tau \, d\xi = -\eta^2 \left(\frac{\partial \tau}{\partial \xi}\right)_{\xi=\eta} + \int_{\eta}^{\infty} \xi^2 \tau^n \, d\xi \tag{21}$$

since $\left(\dfrac{\partial \tau}{\partial \xi}\right)_{\xi=\infty} = 0.$

In order to evaluate the integrals in (21) we assume a profile of the form

$$\tau = \tau_s \, f(\frac{\xi}{\eta}, a) \quad \text{with } f(1,a) = 1$$

where τ_s, a, η are functions of the time variable θ only. Such a profile must admit the possibility of either positive or negative $(\frac{\partial \tau}{\partial \xi})_{\xi=\eta}$, this being the reason for the introduction of the time-dependent parameter a.

Further, we may combine the integrated energy conservation equation and the boundary condition at the interface, also making use of (12), the momentum equation, to obtain a conservation principle for the total energy

$$\frac{d}{d\theta} \left\{ \int_{\eta}^{\infty} \xi^2 \tau \, d\xi + \alpha_1 \tau_s + \alpha_2 \eta^3 (\frac{1}{3} + \dot{\eta}^2) \right\}$$

$$= \int_{\eta}^{\infty} \xi^2 \tau^n \, d\xi \, . \qquad (22)$$

This expression equates the rate of change of total energy, both thermal and inertial in origin, to the rate of increase in energy due to chemical reaction. Written in terms of the profile assumption we have

$$\frac{d}{d\theta} \left\{ \tau_s (\eta^3 A_1(a) + \alpha_1) + \alpha_2 \eta^3 (\frac{1}{3} + \dot{\eta}^2) \right\} = \tau_s^n \eta^3 A_n(a) \qquad (23)$$

where
$$A_1(a) = \int_1^\infty y^2 f(y,a)\,dy$$

$$A_n(a) = \int_1^\infty y^2 [f(y,a)]^n\,dy.$$

This, together with the equations

$$\frac{\tau_s}{\eta} f'(1,a) = \alpha_1 \dot{\tau}_s + \alpha_2 \dot{\eta}\, \pi_s \tag{24}$$

$$\pi_s = 3\dot{\eta}^2 + 2\eta\ddot{\eta} + 1 \tag{25}$$

$$\tau_s = \frac{\pi_s \eta^3 - 1}{\eta_1^3 - 1} \tag{26}$$

determines the whole system. We shall eliminate the pressure variable π_s at this stage using (25) and use only three equations henceforth.

We have at $\Theta = 0$

$$\tau_s = 1, \qquad \dot{\eta} = \ddot{\eta} = 0,$$
$$\eta = \eta_1 \tag{27}$$

which, together with a suitable initial condition for a defines the initial conditions.

It is convenient, at this stage, to specify the profile assumption to be used. This is given by

$$f(\frac{\xi}{\eta}, a) = (\frac{\xi}{\eta})\left\{(a+1) - a\frac{\xi}{\eta}\right\}, \qquad 1 \leq \frac{\xi}{\eta} \leq 1 + \frac{1}{a},$$

$$= 0 \qquad\qquad \frac{\xi}{\eta} \geq 1 + \frac{1}{a}. \qquad (28)$$

This gives positive, zero, or negative temperature gradient at the bubble interface according as to whether a < 1, a = 1, a > 1 respectively. To obtain the desired initial rectangular temperature distribution, a must be chosen to be infinite initially. It is worth noting that in our former use of profile methods no attempt was made at simulating the initial rectangular profile. For small values of time, corresponding to a >> 1, the choice of profile assumption is not crucial. Thus, a different profile, for example $\tau = \tau_s (\frac{\xi}{\eta})^{-a}$, $\xi \geq \eta$ with a $\geq$ 0, where 'a' has a very different significance leads to similar equations for the profile parameters. This form of profile, however, does not permit the possibility of positive temperature gradient at the bubble interface.

2.5. Solution for small values of time

Due to the initial stepped temperature distribution it is expected that the early stages of the process will be dominated by thermal conduction effects. Accordingly let us assume a power series expansion for each of the three time-dependent parameters in powers of $\theta^{1/2}$, as is usual in processes of pure conduction. The constants may then be determined by comparing coefficients. In order to achieve self-consistency the expansions must be of the form

$$\tau_s = 1 + \lambda_1 \theta^{1/2} + \lambda_2 \theta + \ldots$$

$$a = \mu_1 \theta^{-1/2} + \mu_2 + \ldots \qquad (29)$$

$$\eta = \eta_1 + \nu_1 \theta^{5/2} + \nu_2 \theta^3 + \ldots$$

We also assume that $a \gg 1$ and hence we may take

$$A_1(a) \simeq \frac{1}{2a} + \frac{1}{2a^2} .$$

Comparison of powers of $\theta^{1/2}$ then yields

$$\tau_s = 1 - \frac{\eta_1^2}{\alpha_1} \theta^{1/2} + \frac{2}{3} \frac{\eta_1^4}{\alpha_1^2} \theta + \ldots$$

$$a = \frac{\eta_1}{2} \theta^{-1/2} + (1 - \frac{1}{6} \frac{\eta_1^3}{\alpha_1}) + \ldots \qquad (30)$$

$$\eta = \eta_1 - \frac{2}{15} \frac{1}{\alpha_1 \eta_1^2} (\eta_1^3 - 1)\theta^{5/2} + \frac{1}{18} \frac{(\eta_1^3 - 1)}{\alpha_1^2} \theta^3 + \ldots$$

Equations (30) are an exact series representation for the time-dependent parameters τ_s, a, η. Due to the extremely small magnitude of α_1, it can however be seen that they are valid only for very small values of the time variable Θ. Consequently, in accordance with the methods of singular perturbation theory it is necessary to scale the time variable suitably. We therefore make the substitution

$$\Theta_1 = \frac{\Theta}{\alpha_1{}^2} \ . \tag{31}$$

Equations (30) then indicate that the perturbation solution is of the form

$$\tau_s = \tau_s^{(o)}(\Theta_1) + \alpha_1 \, \tau_s^{(1)}(\Theta_1) + \ldots$$

$$a = \frac{1}{\alpha_1} \, b(\Theta_1) + c(\Theta_1) + \ldots \tag{32}$$

$$\eta = \eta_1 + \alpha_1{}^4 \eta^{(o)}(\Theta_1) + \alpha_1{}^5 \eta^{(1)}(\Theta_1) + \ldots$$

Substitutions of these expressions into equations (23)-(26), and comparison of coefficients of powers of α_1 yields

$$\tau_s^{(o)}(\Theta_1)(\eta_1{}^3 - 1) + \frac{1}{?} = \eta_1{}^3 [2\eta_1 \, \ddot{\eta}^{(o)}(\Theta_1) + 1] \tag{33}$$

$$\tau_s^{(o)}(\Theta_1) \left\{ \frac{\eta_1{}^3}{2b(\Theta_1)} + 1 \right\} = 1 \tag{34}$$

$$- \tau_s^{(o)}(\theta_1) b(\theta_1) \eta_1 = \dot{\tau}_s^{(o)}(\theta_1) \tag{35}$$

where differentiation is now with respect to θ_1. It should be noted that equations (34) and (35) could be obtained by ignoring inertia terms in the original equations ($\dot{\eta} = \ddot{\eta} \equiv 0$) and taking the parameter a large and equal to b. This first perturbation solution is thus equivalent to a 'frozen flow' approximation where all motion within the fluid is 'frozen' and, in effect, a hot gas inclusion in a solid medium is studied; this is, of course, a valid problem in its own right.

The above equations (33)-(35) yield

$$\frac{1}{\tau_s^{(o)}(\theta_1)} - 1 + \log \tau_s^{(o)}(\theta_1) = \frac{\eta_1^b}{2} \theta_1 \tag{36}$$

but no explicit relations may be obtained for $b(\theta_1)$, $\eta^{(o)}(\theta_1)$. Consideration of the second perturbation terms yields the following simple results

$$\tau_s^{(1)}(\theta_1) \equiv 0 ,$$

$$c(\theta_1) \equiv 1 , \tag{37}$$

$$\eta^{(1)}(\theta_1) \equiv 0 .$$

The use of the variable θ_1 'stretched' the time scale so that for small times $\theta \sim \alpha_1$, it is possible to take $\theta_1 \to \infty$. Consequently as $\theta_1 \to \infty$, we have, considering only lower order terms in α_1,

$$\tau_s \to 0$$
$$\partial \to 1 \qquad\qquad (38)$$
$$\eta \to \eta_1 .$$

The above analysis shows that the bubble temperature initially falls very rapidly to the ambient value. It should be noted that the activation energy parameter n does not feature in the perturbation solutions obtained; the chemical reaction rate thus plays no part in the initial stages of the process, which is consequently one of pure conduction. The extreme rapidity of the process is due to steep temperature gradients at the bubble interface and also due to the very small thermal capacity of the gas bubble as compared to that of an equal volume of fluid.

The above investigation represents an 'inner' expansion valid for small values of time.

2.6. Numerical solution for 'outer' expansion

In order to extend the 'inner' expansion to describe the process at larger values of the time variable, it is necessary to find an 'outer' solution. The full set of equations to be solved may be written

$$\frac{d}{d\theta}\left\{ \tau_s(\eta^3 A_1(a)+\alpha_1) + \alpha_2 \eta^3(\tfrac{1}{3} + \dot{\eta}^2) \right\} = \tau_s^n \eta^3 A_n(a) \tag{39}$$

$$- \tau_s(a-1)\eta = \alpha_1 \dot{\tau}_s + \alpha_2 \eta^2 \dot{\eta}(3\dot{\eta}^2 + 2\eta\ddot{\eta} + 1) \tag{40}$$

$$\tau_s(\eta_1^3 - 1) + 1 = \eta^3(3\dot{\eta}^2 + 2\eta\ddot{\eta} + 1) . \tag{41}$$

At this stage, steep temperature gradients no longer exist and it is possible to neglect the term in α_1 completely. This assumption does however lower the order of the equations; to compensate we assume that the parameter a remains at a mean value of unity. These two additional assumptions imply that $\dot{\eta}$ must remain small.

Using the properties of the 'inner' solution we take the following initial conditions

$$\theta = 0 \;:\; \qquad \eta = \eta_1$$

$$\dot{\eta} = 0$$

$$\tau_s = \tau_{s_0} , \qquad \text{some starting value,} \tag{42}$$

$$\ddot{\eta} = \frac{\eta_1^3 - 1}{2\eta_1^4}(\tau_{s_0} - 1)$$

Equations (39)-(41), together with the two additional assumptions were solved numerically using a forward integration process; as before the calculations were performed on an IBM 7090 electronic computer. Typical integral curves are shown in Figs. 12, 13, 14; the parameter n was assumed to be equal to 10 throughout the calculations. Examination shows that the integral curves of τ_s versus θ (see Fig. 12) are basically of two kinds; for large initial τ_s it is found that the central temperature rises indefinitely with time; as τ_{s_o} decreases, oscillations begin to appear but the bubble temperature still ultimately rises. For small values of τ_{s_o} sustained oscillations appear.

It will further be seen that the somewhat large variations in η are not consistent with the assumption of a mean value for a of unity, as $\dot{\eta}$ is not now very small. In addition, the sustained oscillations do not represent an equilibrium state as one might expect for small values of τ_{s_o}. This apparent inability to achieve equilibrium may be partially due to the neglect of the term proportional to α_1; this term represents the rate of change of internal energy of the bubble. If this term is taken to be zero, the internal energy of the bubble remains

constant, only inertial and conduction effects being taken into account; an incessant interchange of these two kinds of energy occurs, producing oscillatory integral curves.

Although these computations represent an outer solution of the problem, there is no means of matching the two solutions. The parameter α_1 does not feature in the outer region; hence powers of α_1 may not be compared. For this reason and those mentioned above, the outer solution as derived here should be viewed with some scepticism.

3. Hot gas bubble initially surrounded by a thermal layer

Finally, we consider the behaviour of a hot inert gas bubble surrounded by a heated thermal layer whose thickness is of the same order of magnitude as the bubble radius. It is assumed that a temperature distribution is already established in this layer.

The problem to be solved is then defined by equations (39)-(41), together with the initial conditions

$$\theta = 0 \quad : \quad \begin{aligned} \tau_s &= 1 \\ \eta &= \eta_1 \\ \dot{\eta} &= \ddot{\eta} = 0 \\ a &= 1 \end{aligned} \tag{43}$$

i.e. the temperature gradient at the interface of the
bubble and the thermal layer is taken to be zero initially.
This assumption is analogous to our previous use of profile
methods.   In this case the problem is no longer one of
pure conduction, and attempts to consider power series
expansions in powers of $\Theta^{1/2}$ fail.   It is however possible
to find power series expansions of the form

$$\tau_s = 1 + \lambda_1 \Theta^2 + \lambda_2 \Theta^3 + \ldots$$

$$a = 1 + \mu_1 \Theta + \mu_2 \Theta^2 + \ldots \qquad (44)$$

$$\eta = \eta_1 + \nu_1 \Theta^4 + \nu_2 \Theta^5 + \ldots$$

Comparison of coefficients of powers of Θ in (39)-(41),
then yields

$$\tau_s = 1 - \frac{A_n(1)\,\eta_1}{2\alpha_1 A_1'(1)}\,\Theta^2 + O\left(\frac{\Theta^3}{\alpha_1^2}\right) \qquad (45)$$

$$a = 1 + \frac{A_n(1)}{A_1'(1)}\,\Theta + O\left(\frac{\Theta^2}{\alpha_1}\right) \qquad (46)$$

$$\eta = \eta_1 - \frac{A_n(1)}{\alpha_1 A_1'(1)}\,\frac{(\eta_1^3 - 1)}{48\,\eta_1^3}\,\Theta^4 + O\left(\frac{\Theta^5}{\alpha_1}\right) \qquad (47)$$

where it is assumed that $A_1(a)$, $A_n(a)$ may be expanded in a

Taylor series about the value a = 1. These expansions represent an exact solution of the problem valid for very small values of the time.

In accordance with the methods of singular perturbation theory it is therefore necessary to scale the time variable suitably. Further inspection of the expansions (45)-(47) indicates the choice of new variable. We put

$$\theta_1 = \frac{\theta}{\alpha_1}. \tag{48}$$

Then the perturbation solutions must be of the form

$$\tau_s = 1 + \alpha_1 \tau_s^{(0)}(\theta_1) + \alpha_1^2 \tau_s^{(1)}(\theta_1) + \cdots$$

$$a = 1 + \alpha_1 a^{(0)}(\theta_1) + \alpha_1^2 a^{(1)}(\theta_1) + \cdots \tag{49}$$

$$\eta = \eta_1 + \alpha_1^3 \eta^{(0)}(\theta_1) + \alpha_1^4 \eta^{(1)}(\theta_1) + \cdots$$

Substitution of these expansions into equations (39)-(41) and comparison of coefficients of powers of α_1, once more assuming expansions for $A_1(a)$, $A_n(a)$ about a = 1, yields

$$\tau_s^{(0)}(\theta_1) = \frac{A_n(1)}{A_1(1)} \left[\theta_1 + \frac{A_1{}'(1)}{A_1(1)\eta_1} \left(1 - \exp\left(\frac{A_1(1)}{A_1'(1)} \eta_1\theta_1\right)\right) \right] \tag{50}$$

$$a^{(0)}(\theta_1) = -\frac{A_n(1)}{A_1(1)\eta_1} \left[1 - \exp\left(\frac{A_1(1)}{A_1'(1)} \eta_1\theta_1\right)\right] \tag{51}$$

$$\eta^{(o)}(\theta_1) = \left(\frac{\eta_1^3 - 1}{2\,\eta_1^4}\right)\left(\frac{A_n(1)}{A_1(1)}\right)\left[\frac{\theta_1^3}{6} + \frac{A_1'(1)}{A_1(1)\eta_1}\frac{\theta_1^2}{2} + \left(\frac{A_1'(1)}{A_1(1)\eta_1}\right)^2\theta_1\right.$$

$$\left. + \left(\frac{A_1'(1)}{A_1(1)\eta_1}\right)^3\left(1 - \exp\left(\frac{A_1(1)}{A_1'(1)}\eta_1\theta_1\right)\right)\right] \qquad (52)$$

It is not possible to obtain simple expressions for the second order perturbation solutions.

Since $A_1'(1) < 0$, it may be seen that as θ_1 increases, $\tau_s^{(o)}$ increases, $a^{(o)}$ decreases and $\eta^{(o)}$ increases. Now by definition $a \geq 0$ and hence an additional restriction on the validity of these expansions is obtained. This is of the form

$$\frac{\alpha_1 A_n(1)}{A_1(1)\eta_1} \leq 1 .$$

Since $\alpha_1 \ll 1$, $A_n(1) \ll 1$ this does not represent a serious restriction.

Consequently this inner expansion defines an ignition regime and it is unnecessary to seek an outer solution. It will be noted that the characteristic time of this process is larger than that for the initial stepped temperature distribution although still small. Nevertheless the change

in initial conditions produces a distinctly different
sequence of events; the process is not characterised by
pure conduction, in fact there exists no expansion in
powers of $\theta^{1/2}$.

4.   Discussion

The early stages of the process in the case of the
initial stepped temperature distribution are characterised
by the very rapid fall to zero of the bubble temperature;
during this time the bubble size remains fixed and chemical
reaction plays no part.   A process of pure conduction takes
place due to steep temperature gradients.   It has not been
adequately established whether this phase is followed by
another during which the temperature rises again; this
would perhaps yield a critical condition for ignition.

For the problem of the bubble surrounded by a thermal
layer the situation is very different.   Steep temperature
gradients do not exist at the bubble interface and both
chemical reaction and inertia effects play a part.   Ignition
takes place inasmuch as the bubble expands and its temperature
rises.   The characteristic time for this process, although
small, is one order of magnitude in α_1 larger than in the
first case.

It is worth remembering that the bubbles considered are very small; the assumptions $\alpha_3 = \eta_o = 1$ imply small initial bubble radii. The effect of surface tension has been neglected in the above analysis; since the surface tension coefficient decreases rapidly with rising temperature this assumption is partially justified. The inclusion of this effect would leave the energy and momentum equations unaltered, while the equation of state and the boundary condition at the bubble interface would be adjusted by an additional $2\sigma/R$, where σ is the surface tension coefficient, added to the ordinary ~~bubble~~ *liquid* pressure. In making the equations dimensionless it would be necessary to introduce an additional dimensionless parameter. It is expected that the result of including these extra terms in the problem with a thermal layer would be to prevent the bubble from expanding initially and hence could perhaps yield some critical condition for ignition.

The inherent difficulties of the problem are several, namely:

1) Non-linearity introduced by chemical reaction rate and inertia terms.

2) Initial stepped temperature distribution implies $a \to \infty$ initially; for small a, on the other hand, the terms $A_n(a)$, $A_1(a)$ become very large.

3) The introduction of the small parameter α_1; this makes it impossible to solve the equations numerically in their complete form.

4) The large number of parameters, and hence effects involved.

Further, since we are unaware of any experimental observations on problems of this nature, we have no means of verifying assumptions or checking results.

It is felt, however, that the use of profile methods cannot be extended further, as the choice of profile assumption implies some foreknowledge of the physical characteristics of the process which may not always be known.

Although this problem is by no means complete, some insight into the mechanisms governing it has been obtained.

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FIG.1. VARIATION OF HEAT-EVOLUTION AND HEAT-REMOVAL RATES (q) WITH TEMPERATURE T.
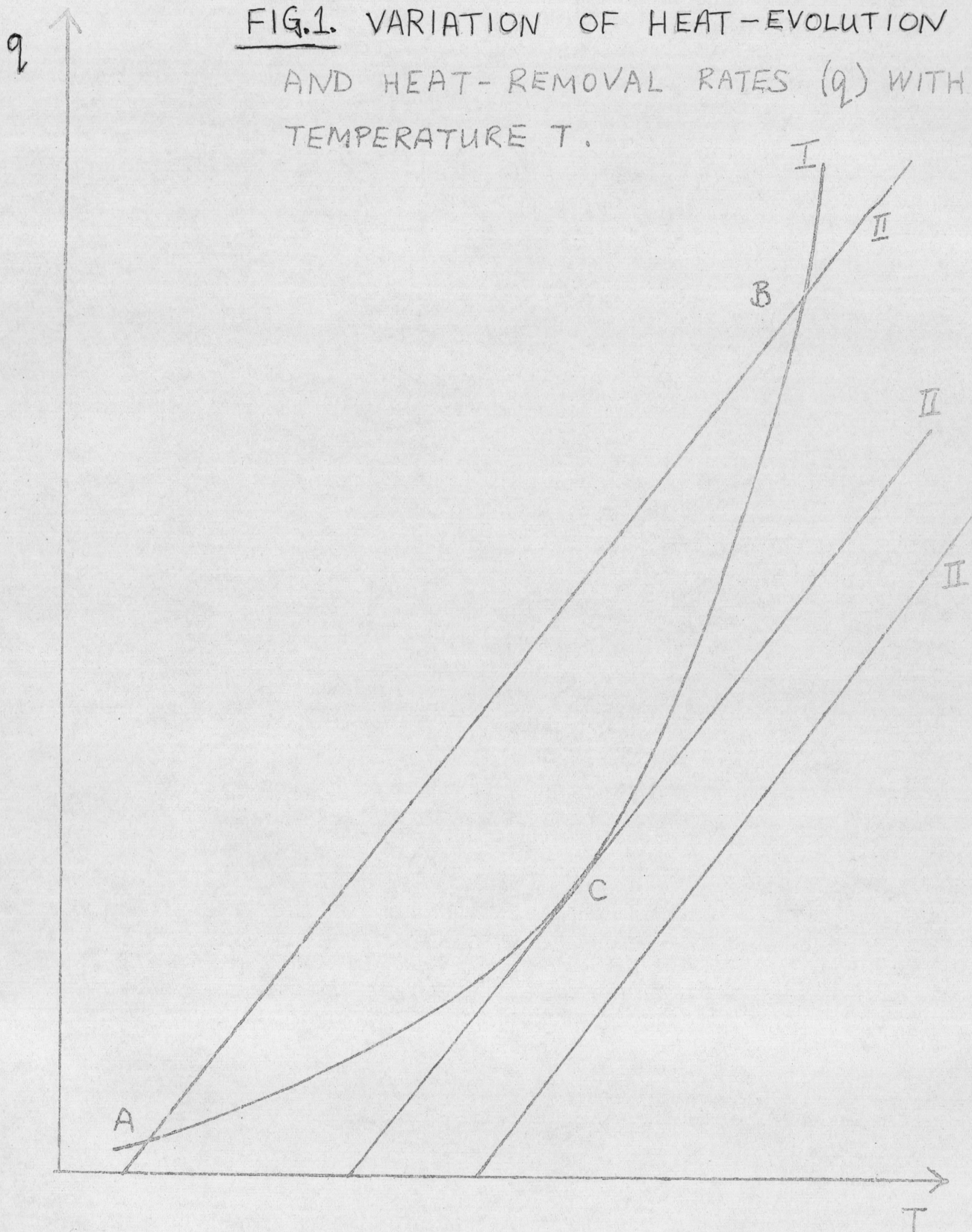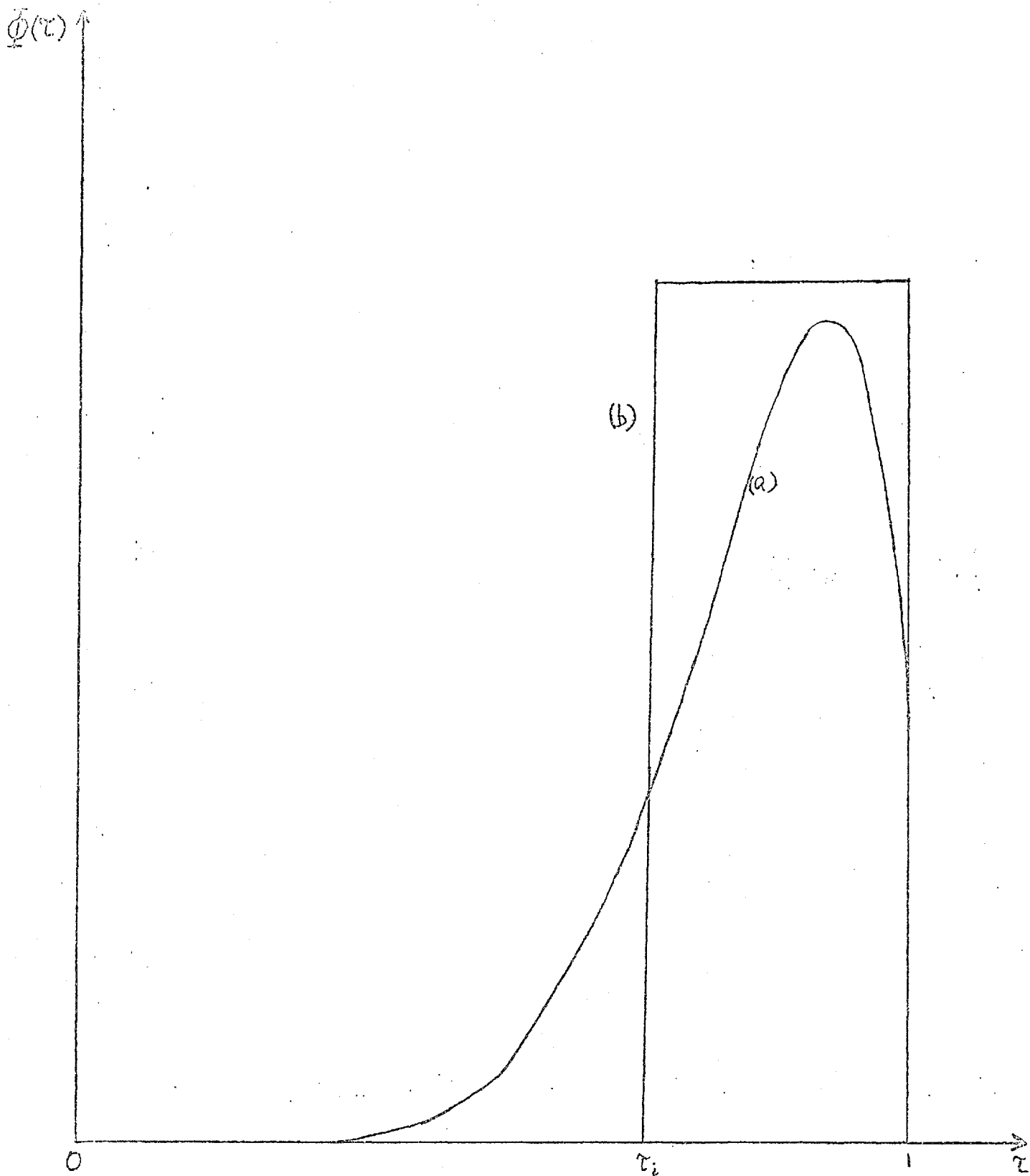
FIG. 2.　a) TYPICAL REACTION RATE FUNCTION

b) STEP-FUNCTION REACTION RATE
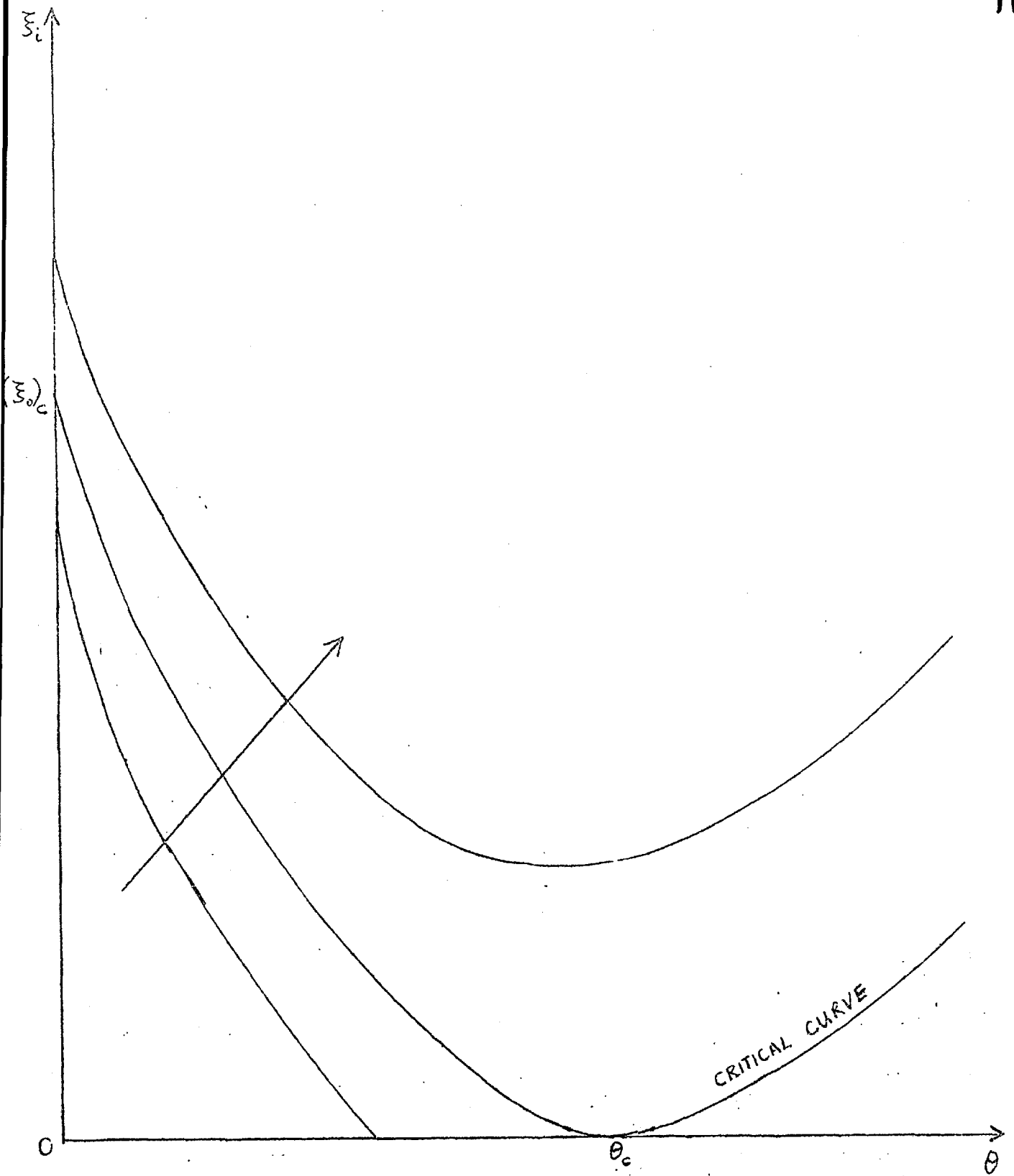
FIG.3. QUALITATIVE BEHAVIOUR OF FLAME RADIUS ξ_i FOR

FIXED τ_i AND INCREASING VALUES OF ξ_o.

FIG. 4. CRITICAL RADIUS OF HOT GAS BUBBLE $(\xi_0)_c$ VERSUS IGNITION TEMPERATURE τ_i.
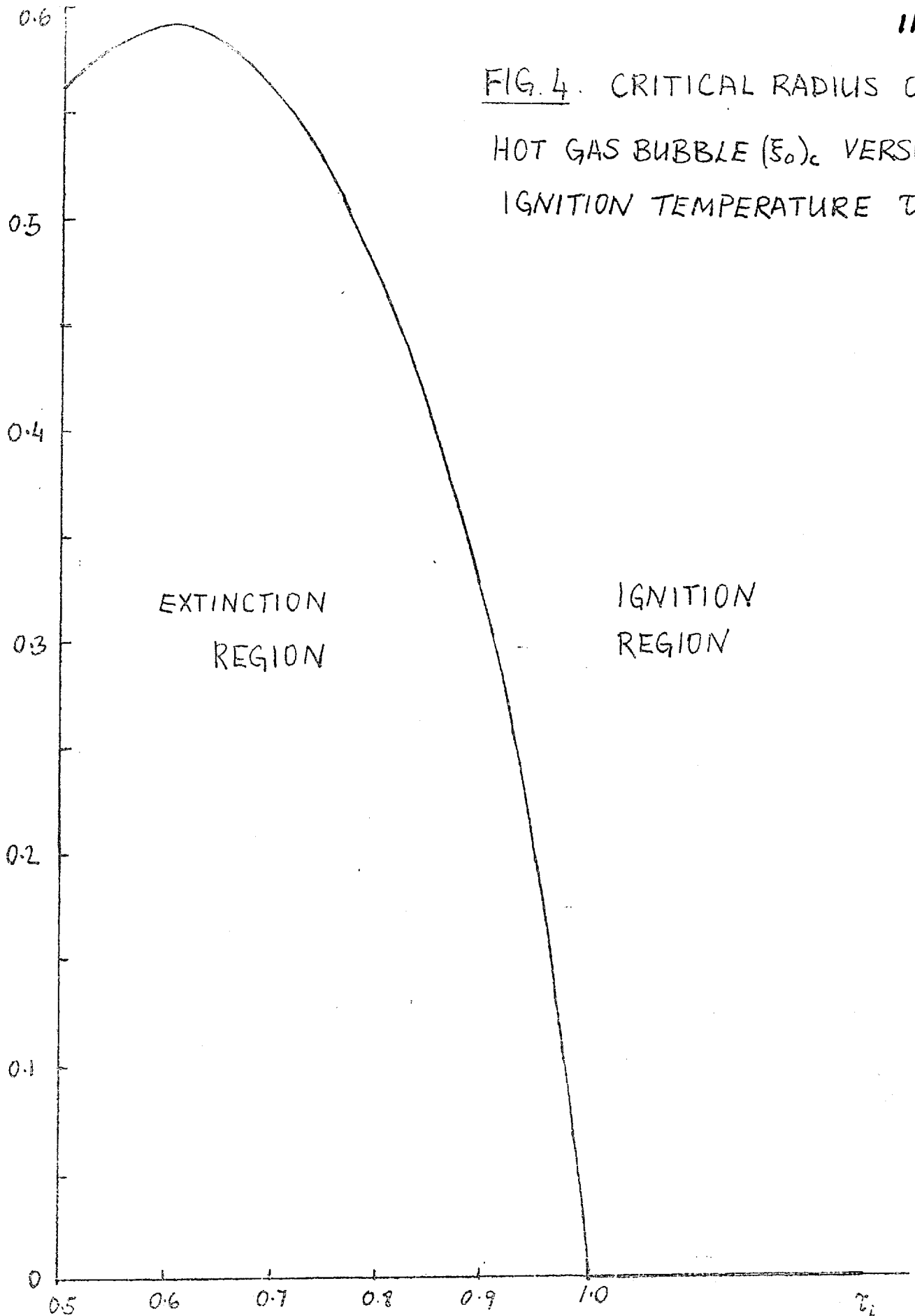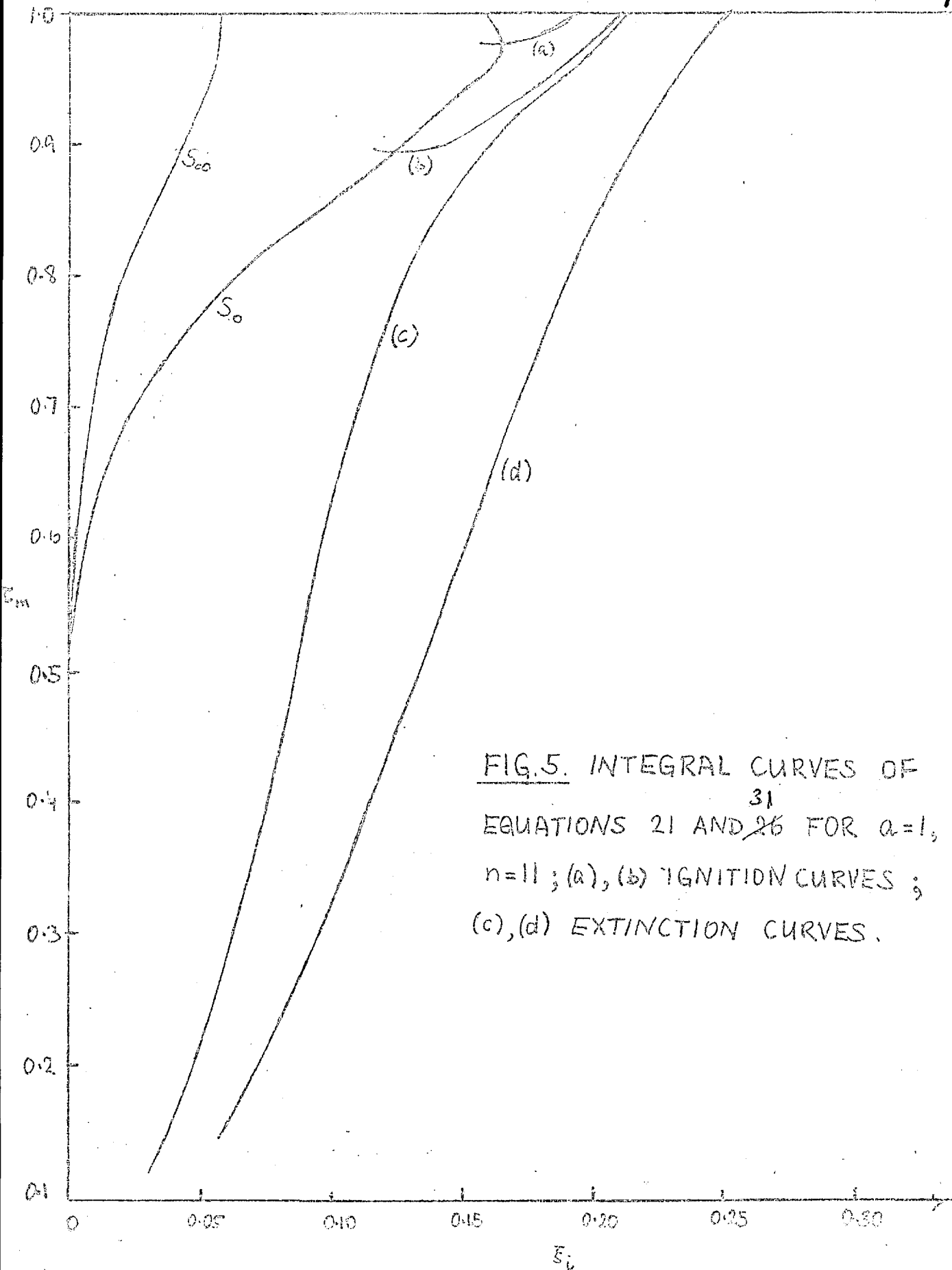
FIG.5. INTEGRAL CURVES OF EQUATIONS 21 AND 26 FOR $a=1$, $n=11$; (a), (b) IGNITION CURVES; (c), (d) EXTINCTION CURVES.

FIG.6. CRITICAL VALUES δ_c VERSUS PARAMETER a, FOR $n = 11, 20$.
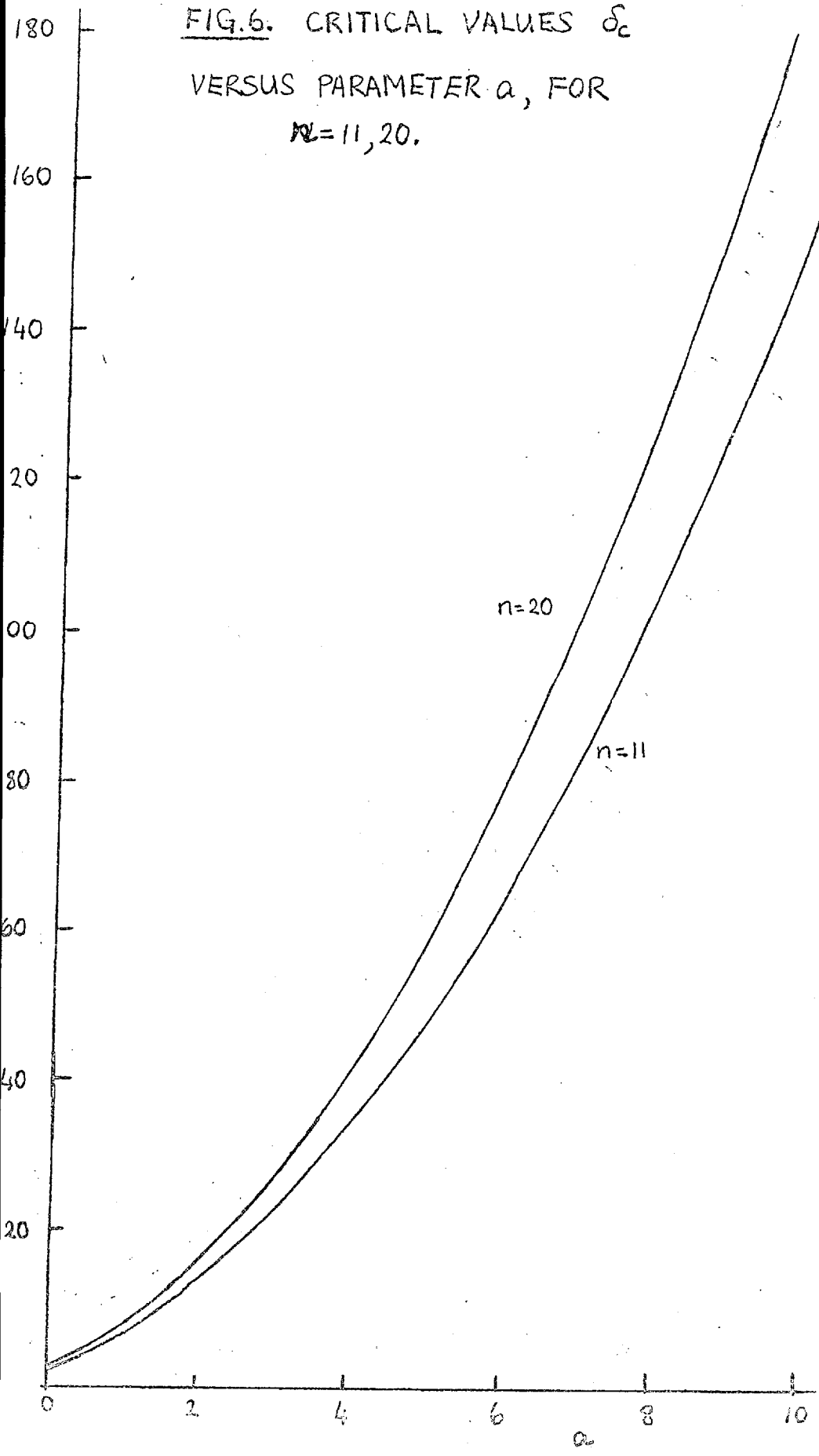
FIG. 7. VARIATION OF (r_c/r_c^*) WITH PARAMETER a, FOR $n = 11, 20$.
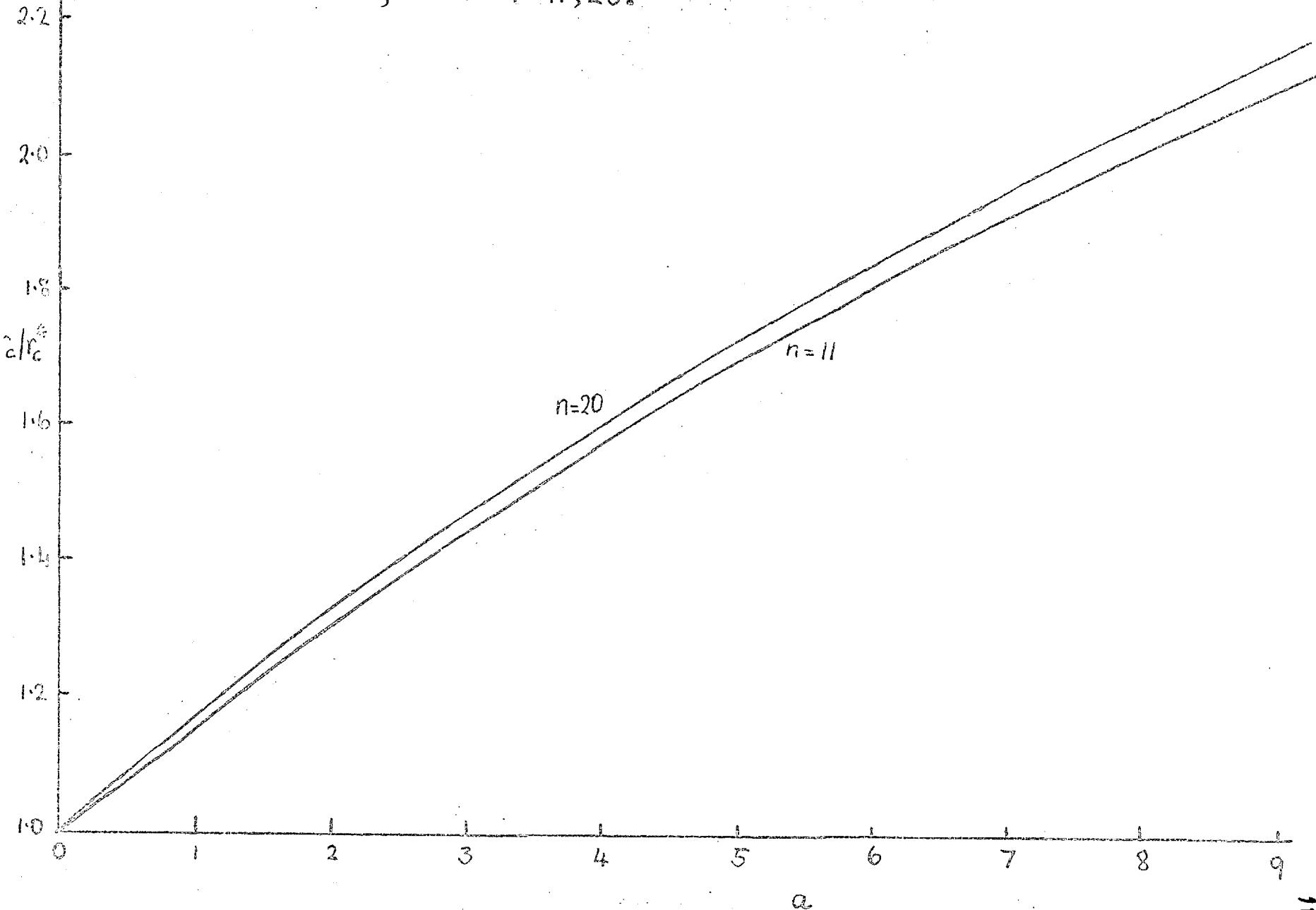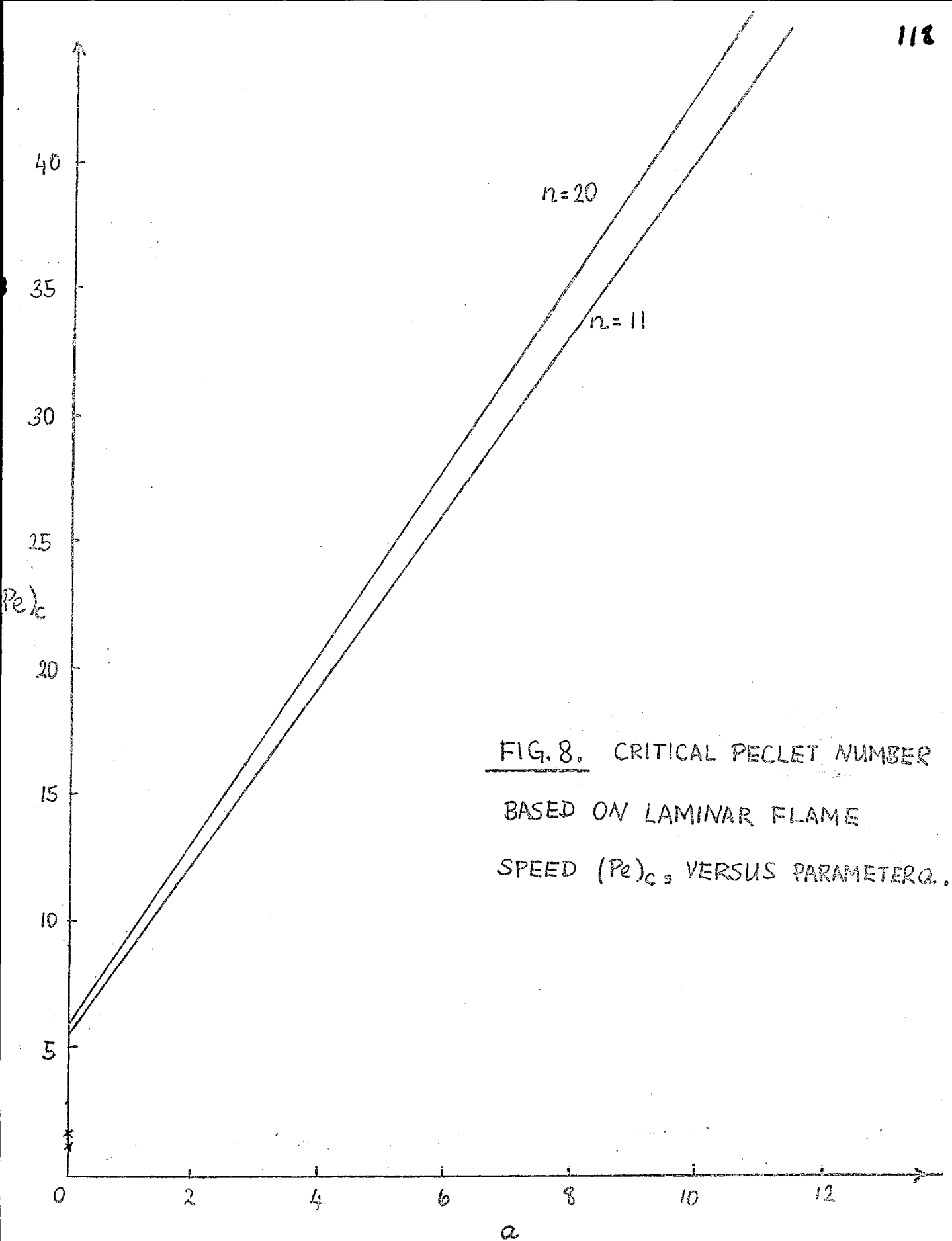
FIG. 8. CRITICAL PECLET NUMBER BASED ON LAMINAR FLAME SPEED $(Pe)_c$, VERSUS PARAMETER a.

FIG.9 VARIATION OF A(σ)

WITH σ INDICATING

LOCAL MAXIMUM
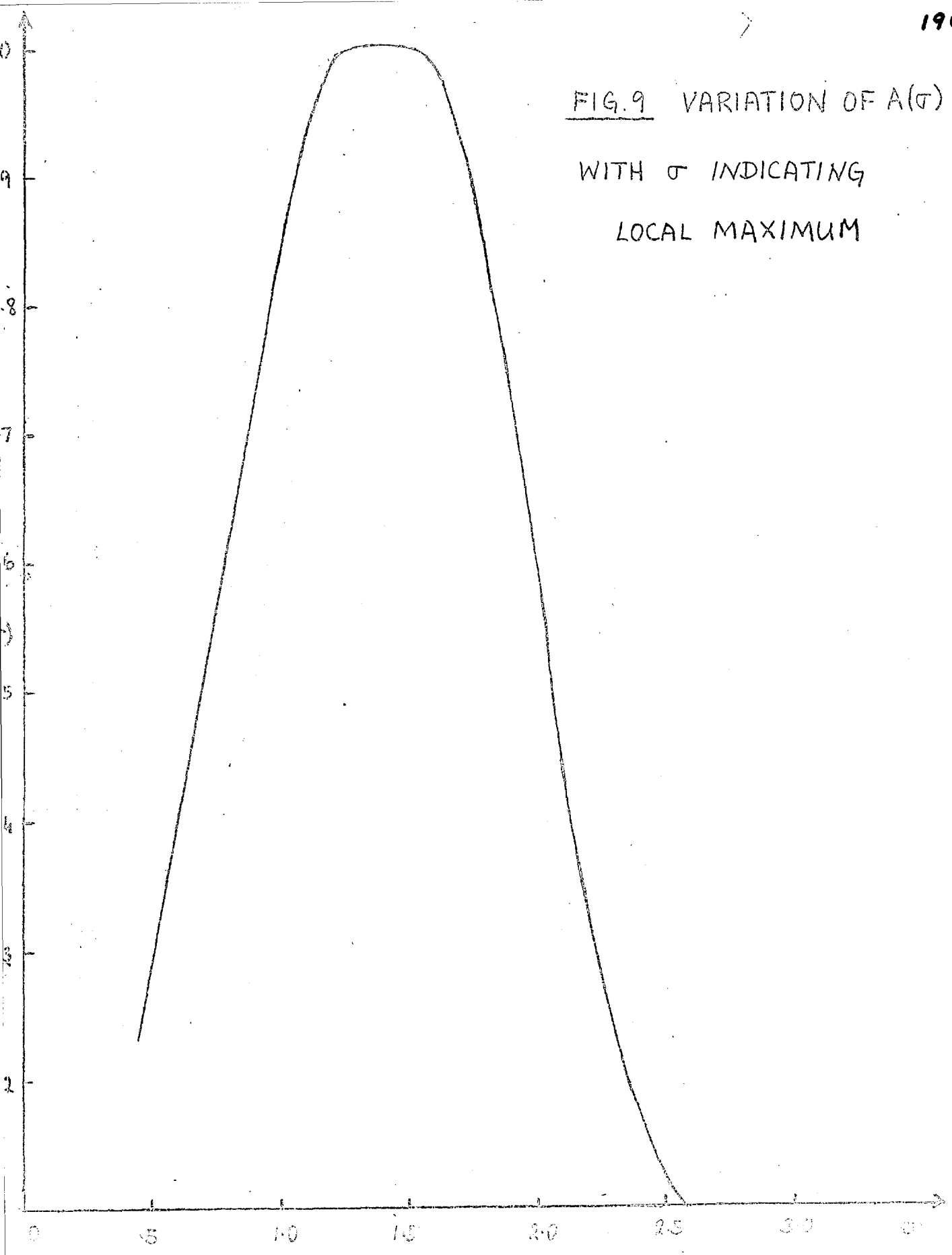
FIG. 16. COMPARISON OF CRITERIA FOR CRITICAL
SPHERICAL HOT SPOTS.
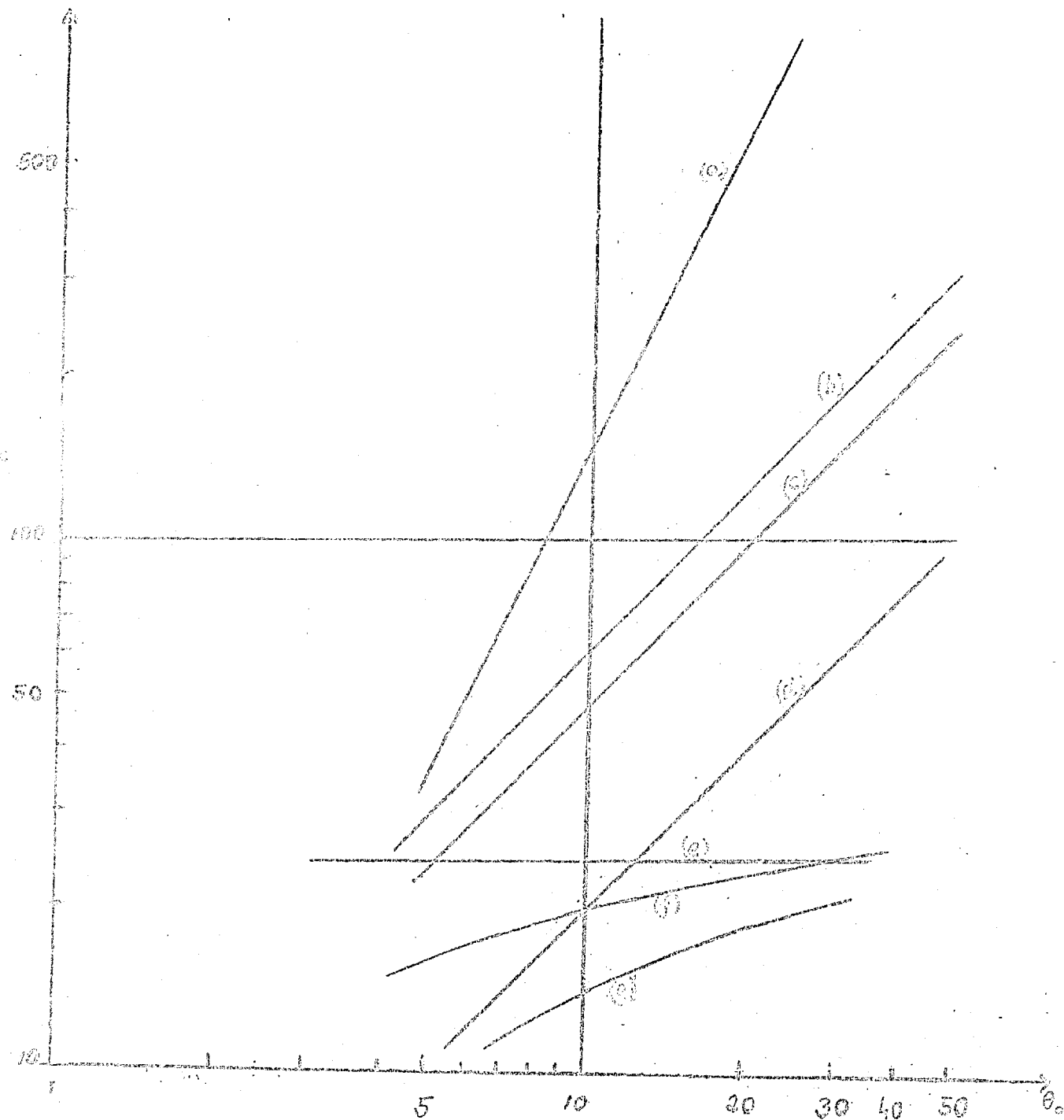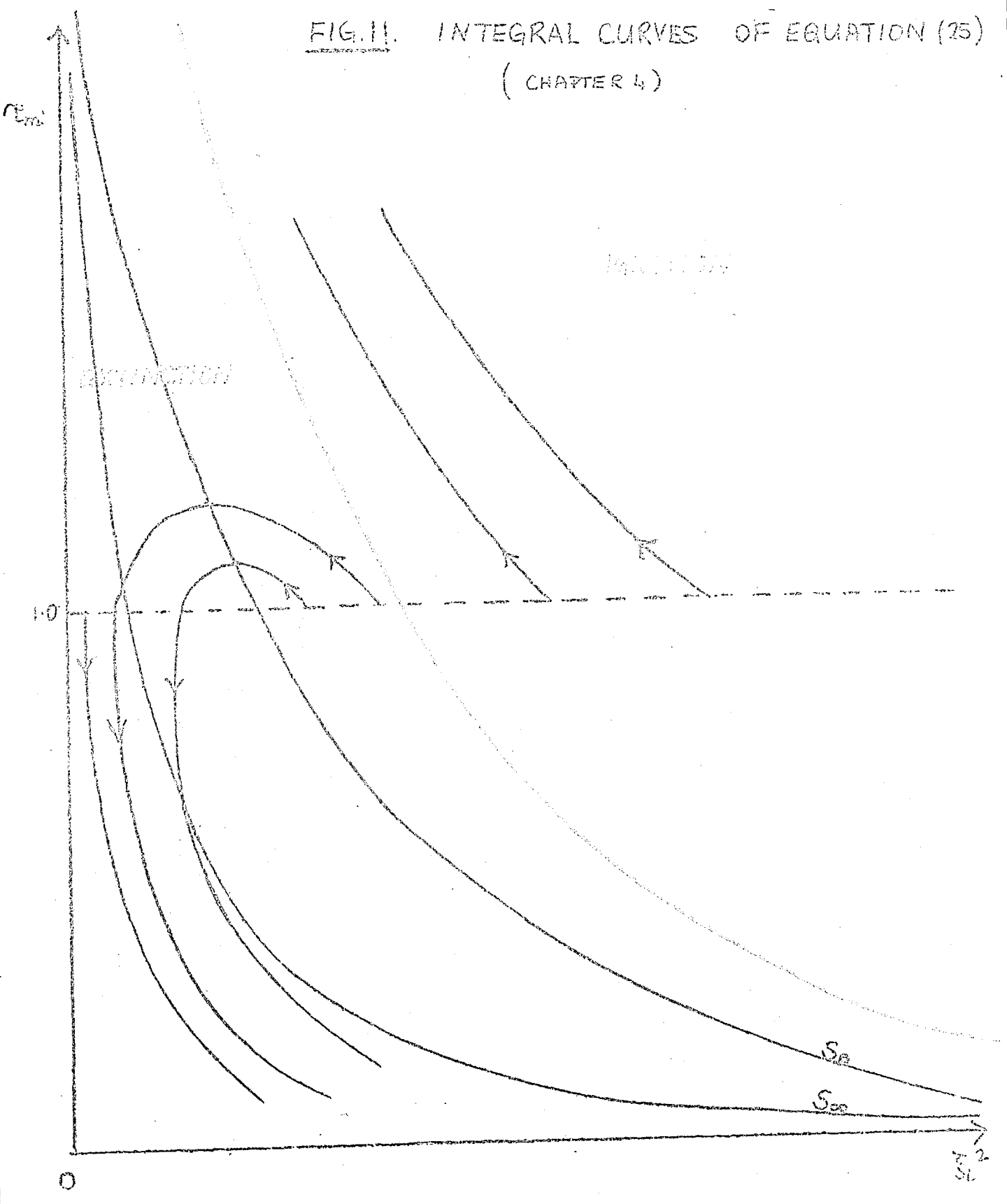

a) BODDINGTON
b) EGN (34)
c) FRIEDMAN/FUJITA
d) THOMAS
e) ZINN
f) MERZHANOV
g) BODDINGTON-
ZINN-THOMAS

FIG.II. INTEGRAL CURVES OF EQUATION (25)
(CHAPTER 4)

— — — CRITICAL CURVE

FIG. 12. VARIATION OF τ_s WITH θ, FOR VARIOUS INITIAL τ_0

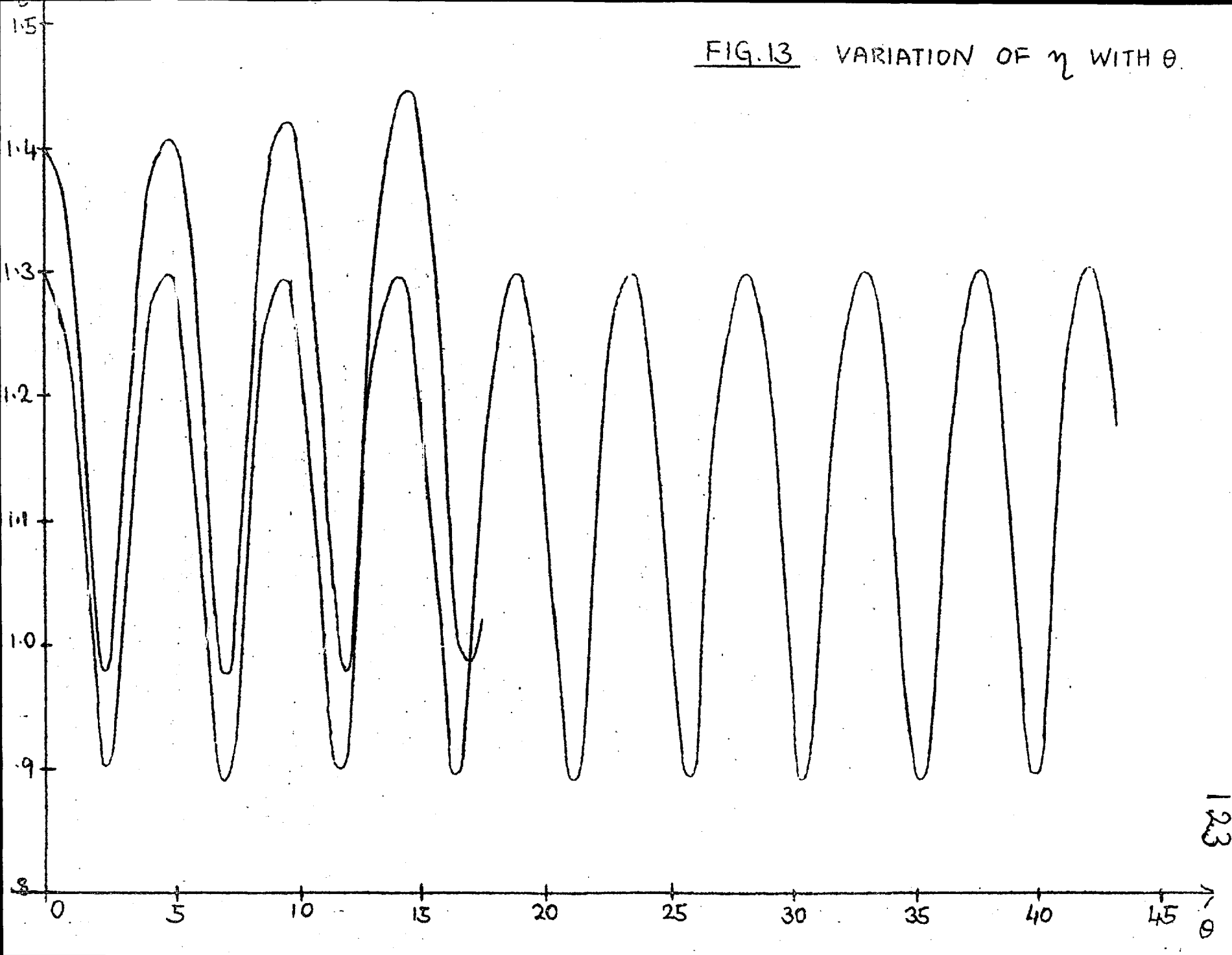$d_2 = 1$, $\alpha = 1$, $n = 10$,
$\eta_1 = 1.3, 1.4$.
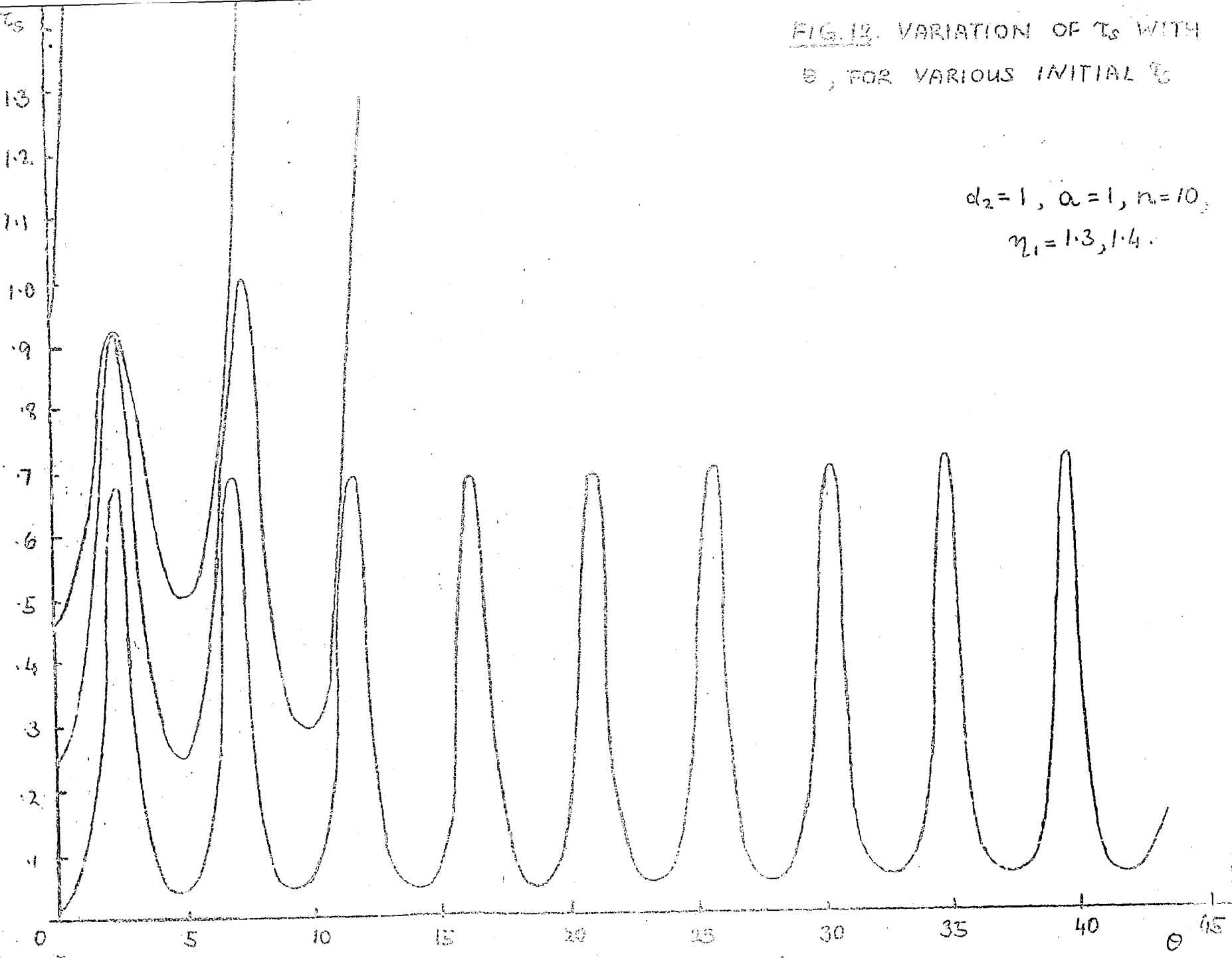
FIG. 13   VARIATION OF η WITH θ.

123

FIG. 14   VARIATION OF $\dot{\eta}$ WITH θ.

124