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LECTURES ON MATHEMATICAL COMBUSTION

Lecture 1: Pre-Asymptotic Combustion Revisited

Technical Report No. 146

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Contents

Lecture 1

PRE-ASYMPTOTIC COMBUSTION REVISITED

The description of reacting systems can be simplified when the so-called activation energy is large; the notion is an old one, but its full power is only released by modern singular perturbation theory. More than forty years ago, Frank-Kamenetskii introduced approximations based on large activation energy to construct a thermal theory of spontaneous combustion, and we shall start there. His problem, which neglects the fluid-mechanical effects of main concern to us, focuses attention on the reaction and thereby acts as a precursor for the lectures that follow. The problem and its generalizations have been the happy hunting grounds of mathematical analysts for many years, but it was not until quite recently that a complete description of the ignition and explosion processes was made available by Kapila and Kassoy (working separately) through activation-energy asymptotics, the main theme of these lectures.

1. Ignition

Let us suppose that a combustion system has a characteristic temperature T_c and that the heat generated by reaction can be expressed as a function of T_c in the Arrhenius form

$$qe^{-\theta/T_c}. \tag{1}$$

This function has an inflection point at $T_c = \theta/2$ and its second derivative is positive for smaller values, where the graph is accordingly concave upwards (figure 1). Suppose also that the heat loss by conduction and convection has the linear form

$$k(T_c-T_f) \tag{2}$$

where T_f is the ambient temperature.

The system can only be in equilibrium if the heat generated (1) is equal to the heat lost (2). The parameters q and k do not then play independent roles but, rather, it is

$$\mathcal{D} = q/k \qquad (3)$$

that is relevant. This ratio, which will be called the Damköhler number (cf. later lectures), can be altered by changing the parameters of the system.

It is apparent from figure 1 that, for $\theta > 4T_f$, there is either one solution or there are three solutions, depending on the value of $\mathcal{D}$. If $\mathcal{D}$ is increased, the straight line rotates about its end point in a clockwise direction; and we can identify the transitions from 1 solution (cold) to 3 solutions to 1 solution (hot). It is this second transition that is our concern in this lecture. The state of the system, represented by a cold point such as C, moves towards I as the Damköhler number is increased, and then must jump to a hot point such as H at the transition. This jump is called ignition.

Ignition is ubiquitous in combustion systems; it can generally be attributed to the nonlinear dependence of heat generation on temperature and the essentially linear dependence of heat loss. The precise nature of the phenomenon can only be determined by detailed analysis, though the results of different calculations carried out by activation-energy asymptotics often bear a strong family resemblance. They are characterized by the following elementary example, introduced by Frank-Kamenetskii.

2. Spontaneous Combustion

Consider the boundary-value problem

$$d^2T/dx^2 = -\mathcal{D}e^{-\theta/T} \text{ for } |x| \leq 1,\ T = T_f \text{ at } x = \pm 1. \qquad (4)$$

Heat conduction in the infinite slab is balanced by heat generation due to the reaction. Depletion of the reactant has been ignored, so that the reaction rate depends only on temperature, as in section 1.

Such models have been used for many years to explain spontaneous combustion, the auto-ignition that occurs, for example, in large volumes of damp organic material. Nondimensionalization of the heat-conduction Laplacian makes the Damköhler number $\mathcal{D}$ proportional to a^2, where a is a length characteristic of the volume (here the semi-thickness of the slab). Thus, an increase in $\mathcal{D}$ may be achieved by increasing the volume and, as we shall see, this can lead to ignition.

We seek a solution of the problem (4) that, as $\theta \to \infty$, deviates only by $O(\theta^{-1})$ from the uniform state, i.e.

$$T = T_f + \theta^{-1}T_f^2\phi + \ldots \text{ with } \phi = (1/T)_1. \tag{5}$$

This leads to

$$d^2\phi/dx^2 = -\delta e^{\phi} \text{ with } \delta = \mathcal{D}\theta e^{-\theta/T_f}/T_f^2, \tag{6}$$

an equation first obtained by Frank-Kamenetskii, and the boundary conditions

$$\phi = 0 \quad \text{at} \quad x = \pm 1. \tag{7}$$

Here δ, the scaled Damköhler number, is assumed to be $O(1)$.

The perturbation ϕ achieves its maximum (ϕ_m) at the midpoint $x = 0$ and so may be written

$$\phi = 2\,\ln[e^{\phi_m/2}\operatorname{sech}(cx)] \text{ with } c^2 = \tfrac{1}{2}\delta e^{\phi_m}. \tag{8}$$

The boundary conditions then imply that

$$\sqrt{\delta/2} = e^{-\phi_m/2}\cosh^{-1}(e^{\phi_m/2}), \tag{9}$$

which defines the maximum temperature in terms of the parameter δ. The result is shown in figure 2, displaying the phenomenon of ignition. For δ less than the critical value

$$\delta_c = 0.878, \tag{10}$$

there is a steady solution, in fact there are two solutions; whereas for $\delta > \delta_c$ there is no solution of the type (5), and in fact none at all. The absence of a steady state for supercritical values of δ implies that, with unsteady effects included, the temperature will increase without bound when, for example, the system is initially in a uniform state $T = T_f$. In practice, the increase is limited by depletion of the reactant, an effect that is ignored here. Of the two solution branches for $\delta < \delta_c$, the upper one is believed to be unstable, though this has never been proved.

It is a general characteristic of ignition that it is associated with $O(\theta^{-1})$ perturbations of the frozen solution, i.e. the solution obtained for $\mathcal{D} = 0$. This is certainly true for the diffusion flames treated in lectures 8 and 9 (see section 8.6); the details differ from those presented here, but the essential ideas do not.

3. Homogeneous Explosion

So far we have inferred ignition from a steady-state theory. The phenomenon itself is inherently unsteady, and certain aspects of the unsteadiness deserve examination. To that end, it is useful to consider first the spatially homogeneous initial value problem

$$dT/dt = \mathcal{D}\, e^{-\theta/T}, \quad T = T_f \text{ for } t = 0. \tag{11}$$

There is no value of $\mathcal{D}$ for which a steady state can be attained; the problem is always supercritical. The physical reason is that no heat-loss mechanism, such as conduction to the boundaries (section 2), exists.

In terms of the exponential integral

$$Ei(y) = \oint_{-\infty}^{y} \frac{e^u}{u}\, du, \tag{12}$$

this has the exact solution

$$\mathcal{D}t = \theta[f(T_f)-f(T)] \text{ with } f(y) = Ei(\theta/y)-(y/\theta)e^{\theta/y} \tag{13}$$

for any value of θ. Since the function f has the symptotic expansion

$$f(y) = (y/\theta)^2 e^{\theta/y} +\ldots \text{ as } \theta \to \infty, \tag{14}$$

it follows that

$$\tilde{t} = \tilde{t}_e-(T/T_f)^2 e^{\theta/T-\theta/T_f}+\ldots \text{ with } \tilde{t} = \delta t; \tag{15}$$

here

$$\tilde{t}_e = 1. \tag{16}$$

Deviations from the initial state of the form (5) are therefore described by

$$\phi = -\ln(\tilde{t}_e-\tilde{t}), \tag{17}$$

so that t_e is the time to explosion (i.e. the time that T takes to deviate from its initial value T_f by more than $O(\theta^{-1})$). The behavior of T in some small neighborhood of t_e is called thermal runaway.

Thermal runaway terminates what is known as the induction phase, the problem with which pre-asymptotic theory was almost exclusively concerned. To go substantially further, modern asymptotic methods must be used, as will be discussed in these lectures. To analyze the so-called explosion phase that follows induction, we return to the expansion (15), and introduce a fast time τ given by

$$e^{-\theta\tau} = \tilde{t}_e-\tilde{t}. \tag{18}$$

This identifies an exponentially small neighborhood of t_e within which the expansion can be written in the form

$$1/T - 1/T_f = -\tau + 2\theta^{-1}\ln(T_f/T) + \ldots; \qquad (19)$$

so that, to leading order,

$$T = T_f/(1-\tau T_f). \qquad (20)$$

Starting at the value T_f for $\tau = 0$, the temperature increases without bound as τ increases to $1/T_f$. The unboundedness is a consequence of our failure to account for reactant depletion; if that is remedied, T increases towards a burnt value T_b (see equation (25)), entering the so-called relaxation phase when it is $O(\theta^{-1})$ away. The relaxation phase lasts an exponentially short time also.

These features are shown in figure 3 and, so long as $T - T_b = O(1)$, the problem without depletion provides a qualitatively accurate description of them. In particular, the fast time τ is still relevant.

The results can also be obtained directly, without recourse to the exact solution. Thus, if the expansion (5a) is substituted into the problem (11), we find

$$d\phi/d\tilde{t} = e^{\phi}, \ \phi = 0 \ \text{ for } \ t = 0, \qquad (21)$$

with solution (17); and introducing the fast time τ into equation (11a) yields

$$dT/d\tau = T_f^2 e^{\theta(1/T_f - 1/T - \tau)}, \qquad (22)$$

with solution (19).

4. Inhomogeneous Explosion

We now combine unsteadiness with spatial inhomogeneity by considering the slab problem

$$\partial T/\partial t - \partial^2 T/\partial x^2 = (T_f^2 \delta / T_f \theta)(T_b - T) e^{\theta/T_f - \theta/T}, \tag{23}$$

$$T = T_f \text{ for } t = 0 \text{ and at } x = \pm 1. \tag{24}$$

To account for reactant depletion, $\mathcal{D}$ has been replaced by $\mathcal{D}Y/Y_f$, where Y is the mass fraction (i.e. concentration) of the reactant and Y_f its initial value. The Shvab-Zeldovich relation

$$T + Y = T_f + Y_f \equiv T_b \tag{25}$$

(cf. section 2.2) is then used to eliminate Y, thereby ensuring conservation of total enthalpy (the sum of thermal enthalpy, represented by T, and chemical enthalpy, represented by Y).

During the induction phase, the $O(\theta^{-1})$ departures of T from T_f expressed by the expansion (5) satisfy

$$\partial\phi/\partial t - \partial^2\phi/\partial x^2 = \delta e^{\phi}, \ \phi = 0 \text{ for } t = 0 \text{ and } x = \pm 1. \tag{26}$$

Reactant depletion plays no role during this initial evolution of the temperature. When the system is subcritical, i.e. for $\delta < \delta_c$, the perturbation ϕ tends to the steady state (8) with the smaller ϕ_m, as $t \to \infty$. But, for $\delta > \delta_c$, the absence of a steady-state solution implies that ϕ will increase without limit, and indeed thermal runaway is found (numerically) to occur after a finite time.

Further progress depends on a description of this runaway, which by symmetry must take place in the neighborhood of $x = 0$. Since the spatial derivatives must play a role, they have to be increasingly large in order to be comparable to the ever-increasing time derivative. It follows that the region in which runaway occurs, called a hot-spot, must continually shrink; this self-focusing is an essential feature of the process.

The appropriate variables for the runaway are $\tilde{t}$ and

$$\eta = x/(t_e - t)^{\frac{1}{2}} \tag{27}$$

(see figure 4), where $t_e(\delta)$ is the runaway time, to be determined numerically. The use of η is suggested by the form of equation (26) and by the focusing discussed above, though Kassoy's achievement in identifying it is in no way diminished by such <u>a posteriori</u> observations. Now we have

$$\partial\phi/\partial\tilde{t} - (\tilde{t}_e - \tilde{t})^{-1}[\partial\phi^2/\partial\eta^2 - \tfrac{1}{2}\eta\partial\phi/\partial\eta] = e^{\phi} \tag{28}$$

and we seek an asymptotic expansion

$$\phi = -\ell n(\tilde{t}_e - \tilde{t}) + \psi(\eta) + \dots \text{ as } \tilde{t} \uparrow \tilde{t}_e \text{ with } \eta \text{ fixed,} \tag{29}$$

finding

$$\psi'' - \tfrac{1}{2}\eta\psi' + e^{\psi} = 1 \text{ with } \psi'(0) = 0 \tag{30}$$

(as a symmetry condition). Another boundary condition is needed to complete the problem for ψ, and this comes from matching with the solution outside the shrinking hot spot, i.e. as $\eta \to \infty$ with x fixed. Thus, since ψ cannot become exponentially large, it has the asymptotic form

$$\psi = -2\ell n\eta + A + \dots \text{ as } \eta \to +\infty, \tag{31}$$

corresponding to

$$\phi = -2\ell nx - \ell n\delta + A + \dots \text{ as } \tilde{t} \uparrow \tilde{t}_e \text{ with } x \text{ fixed.} \tag{32}$$

Numerical solutions of the supercritical problem (26) exhibit the behavior (32), and hence determine the constant $A(\delta)$ that is needed for the second boundary condition

$$\lim_{\eta\to\infty} (\psi + 2\ell n\eta) = A(\delta). \tag{33}$$

The problem (30), (33) uniquely determines the function ψ, which can be readily calculated numerically.

Following the initiation of thermal runaway, the temperature rises at an increasing (exponential) rate, so that self-focusing continues. The variable η still plays a role during this process but now the appropriate time variable is the fast one (18); in terms of η, τ equation (23) becomes

$$\partial T/\partial\tau + \theta(\tfrac{1}{2}\eta\partial T/\partial\eta - \partial^2 T/\partial\eta^2) = (T_f^2/Y_f)(T_b - T)\exp(\theta/T_f - \theta/T - \theta\tau). \qquad (34)$$

Note that $x = \delta^{-\frac{1}{2}}\eta e^{-\theta\tau/2}$ with $\eta = O(1)$ provides a measure of rapid focusing.

The homogeneous result (20) is, to leading order, the solution here also; otherwise the reaction plays no role. The perturbation is found to be

$$T_1 = T_f^2\{\psi(\eta) - \ln[(1-T_f\tau)(Y_f - T_fT_b\tau)/Y_f]\}/(1-T_f\tau)^2, \qquad (35)$$

where ψ satisfies the equation (30a). It must, in fact, be identical to the runaway function just constructed, since otherwise there would be a mismatch as $\tau \to 0$.

The hot-spot evolves so rapidly that the temperature outside has no time to change, i.e.

$$T = T_f + \theta^{-1}T_f^2(-2\ln x - \ln\delta + A) + \ldots \text{ as } x \to 0, \qquad (36)$$

and this does not match the hot-spot expansion, even to leading order. The reason is clear: the expansion (36) breaks down at points inside the hot-spot when it is thickest (τ small), and such points can be well outside once the focusing is under way (τ moderate). The shrinking hot spot leaves behind an intermediate structure, which turns out to be stationary (i.e. independent of τ); to leading order it is described by

$$T = T_f/(1-T_f\chi) \text{ with } \chi = -2\theta^{-1}\ln x. \qquad (37)$$

For the homogeneous problem of section 2, with no reactant depletion, the temperature increases indefinitely as $\tau \to 1/T_f$; but, with depletion, figure 3 shows that T is limited by the value T_b, which is approached within $O(\theta^{-1})$ as $\tau \to Y_f/T_f T_b$ (an earlier time). The latter is true here also, but there can no longer be the single relaxation phase shown in figure 3 as being described on the scale

$$\bar{\tau} = (\tilde{t}-\tilde{t}_e)/\varepsilon \text{ with } \varepsilon = (Y_f\theta/T_f^2)e^{-\theta Y_f/T_f T_b}, \tag{38}$$

since now the temperature is close to T_b only in the vicinity of $x = 0$. Instead, there is a transition phase at the hot-spot, described in terms of $\bar{\tau}$ and the spatial variable

$$\bar{x} = \delta^{\frac{1}{2}}x/\varepsilon^{\frac{1}{2}}, \tag{39}$$

followed by propagation and, finally, relaxation phases. Figure 5 is the result of numerically integrating the limit problem for the transition phase, during which the focusing of the hot-spot is opposed by reactant depletion, thereby forming an incipient deflagration wave (section 2.4). Once formed the wave propagates rapidly through the right side of the slab burning up the reactant, after which the relaxation phase takes over. (Of course, similar remarks apply to the left side of the slab too.) Buckmaster & Ludford (1982, p. 236) give the details, following Kapila.

5. Ignition by External Agencies

So far we have confined the discussion to ignition due to self-heating, but it can also be caused by an external agency. As an example, suppose the half-space $x < 0$ is filled with a combustible material subject to a prescribed heat flux at its surface. The mathematical problem is

$$\partial T/\partial t - \partial^2 T/\partial x^2 = \mathcal{D}e^{-\theta/T}, \tag{40}$$

$$\partial T/\partial x = T_s' > 0 \quad \text{at} \quad x = 0, \; T = T_f \text{ for } t = 0 \text{ and as } x \to -\infty. \quad (41)$$

Here the constant T_s' is the dimensionless heat flux and we shall suppose that

$$\mathcal{D} = e^{\theta/T_r} \text{ with } T_r > T_f \qquad (42)$$

The parameter T_r characterizes the reactivity of the material: for temperatures below T_r, the reaction is negligibly weak in the limit $\theta \to \infty$. A pre-exponential factor (even depending on θ) can be given to $\mathcal{D}$, but this is equivalent to changing T_r slightly, provided nothing is added to the exponential growth of $\mathcal{D}$ with θ.

During an initial (finite) time interval, the material is colder than the reactivity temperature so that the reaction is frozen (i.e. exponentially weak) and the heat equation governs. Because of the heat flux T_s' the temperature rises, its maximum value occuring at the surface $x = 0$. Ignition occurs when the surface temperature reaches T_r. The subsequent process of thermal runaway, hot-spot development and deflagration-wave formation has been discussed by Kapila. Here we shall mention only the mathematical problem involved in the thermal runaway.

The rise in temperature is much more rapid than that for spontaneous combustion, extending only over a period $O(\theta^{-1})$. If conduction is to rival the temporal changes, spatial gradients must therefore be $O(\theta^{\frac{1}{2}})$ so that, at corresponding distances from the surface, the temperature has dropped by $O(\theta^{-\frac{1}{2}})$ and there is no reaction. (In the limit $\theta \to \infty$, reaction only occurs at temperatures $O(\theta^{-1})$ away from T_r.) The thermal runaway is therefore governed by the heat equation

$$\partial\tilde{T}_2/\partial\tilde{\tau} - \partial^2\tilde{T}_2/\partial\tilde{\chi}^2 = 0 \qquad (43)$$

and its cause lies in the nonlinear boundary conditions

$$\partial\tilde{T}_2/\partial\tilde{\chi} = e^{\tilde{\tau}+\tilde{T}_2} \text{ for } \tilde{\chi} = 0, \;\; \tilde{T}_2 = o(1) \text{ as } \tilde{\chi} \to -\infty \text{ and as } \tau \to -\infty. \qquad (44)$$

Here $\tilde{\chi}$ and $\tilde{\tau}$ are appropriately transformed space and time variables while

$\tilde{T}_2$ represents the second perturbation in an expansion of the temperature.

The problem (42,43) should be compared with that for spontaneous combustion, given by equations (26). Clearly they have entirely differently forms.

6. Ignition by an Externally Generated Hot-Spot

Suppose some portion of an infinite combustible material is burnt very rapidly so that there is a local rise in temperature and depletion of reactant. That is, we create a hot spot (using a spark, for example) somewhat like the one that develops in auto-ignition. The hot-spot can have one of two fates: either it decays by diffusion, so that after a certain time the temperature is essentially uniform once again (and constant until a homogeneous explosion occurs because of self-heating); or it acts as a ignition source, producing a deflagration wave that sweeps across and consumes the fresh material. If the hot-spot is very small, temperature gradients will be very large and the resultant cooling will eliminate it. On the other hand, the results for auto-ignition suggest that a sufficiently large hot-spot will ignite the material. In general, the initial-value problem that must be solved to determine the fate of a particular hot-spot is difficult. We shall therefore consider a very special configuration from which plausible conclusions can be drawn quite easily.

Consider the spherically symmetric form

$$\frac{\partial T}{\partial t} - \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial T}{\partial r}\right) = \mathcal{D}\left(\frac{T_b - T}{Y_f}\right)e^{-\theta/T} \qquad (45)$$

of equation (23). This has an exact, stationary solution

$$T = \begin{cases} T_b \\ T_f + Y_f r_*/r \end{cases} \quad \text{for} \quad r \lessgtr r_* \qquad (46)$$

in the limit $\theta \to \infty$, provided

$$r_* = Y_f^{3/2}\,\theta e^{\theta/2T_b}/\sqrt{2D}\,T_b^2. \qquad (47)$$

Thus, the reaction term is asymptotically zero on either side of the flame at $r = r_*$, where the temperature gradient takes a jump

$$\partial T/\partial r\Big|_{r_*+0} = -\sqrt{2D/Y_f}\,T_b^2\,\theta^{-1}e^{-\theta/2T_b} \qquad (48)$$

(cf. the deflagration-wave solution in section 2.4).

This combustion field is now subjected to spherically symmetric disturbances. A straightforward stability analysis shows that the perturbation of the flame radius grows like e^{t/Y_f}, which corresponds to a collapsing or growing hot spot depending on whether the flame is displaced inwards or outwards initially. The result suggests that the radius (46) is critical: larger hot-spots will grow and smaller ones will collapse.

REFERENCE

Buckmaster, J. & Ludford, G.S.S. (1982). Theory of Laiminar Flames. Cambridge: University Press.

Figure Captions

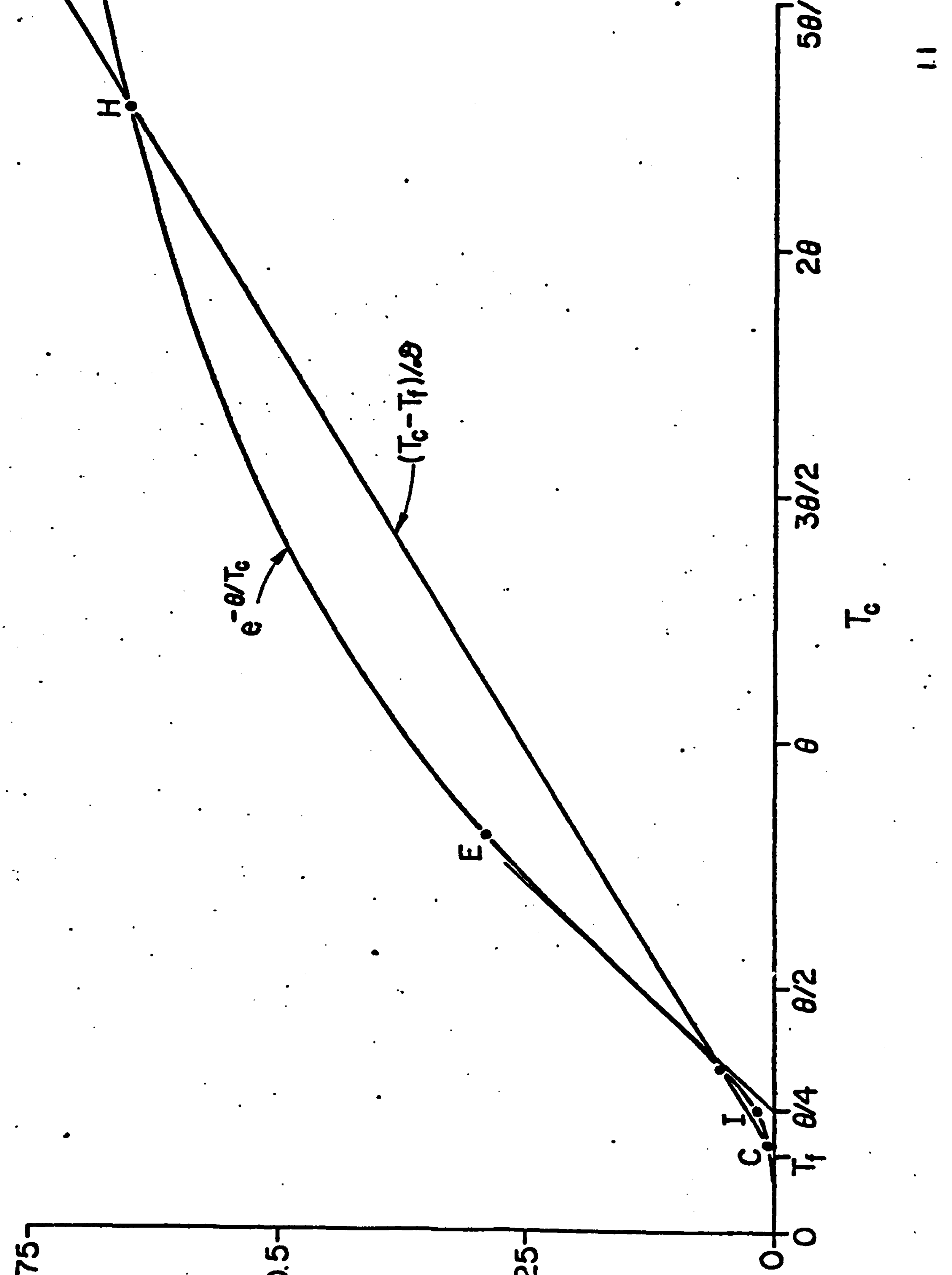
1.1 Heat-generation and heat-loss curves.

1.2 Steady-state response for slab with surface maintained at initial uniform temperature, as determined by equation (9).

1.3 Temperature history for homogeneous explosion with reactant depletion.

1.4 Parabolas η = const.

1.5 Temperature profiles during transition phase. (Courtesy A.K. Kapila.)



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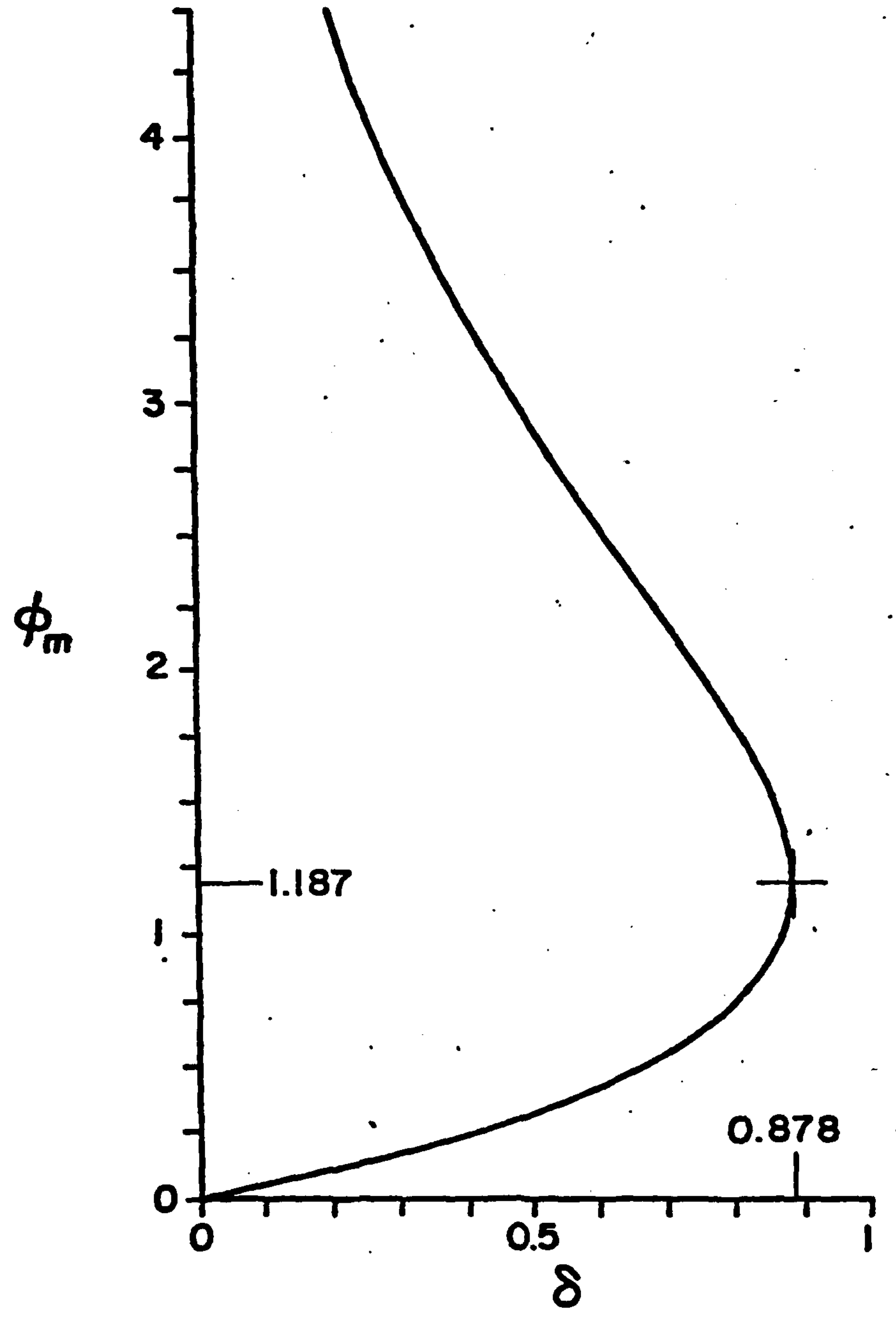

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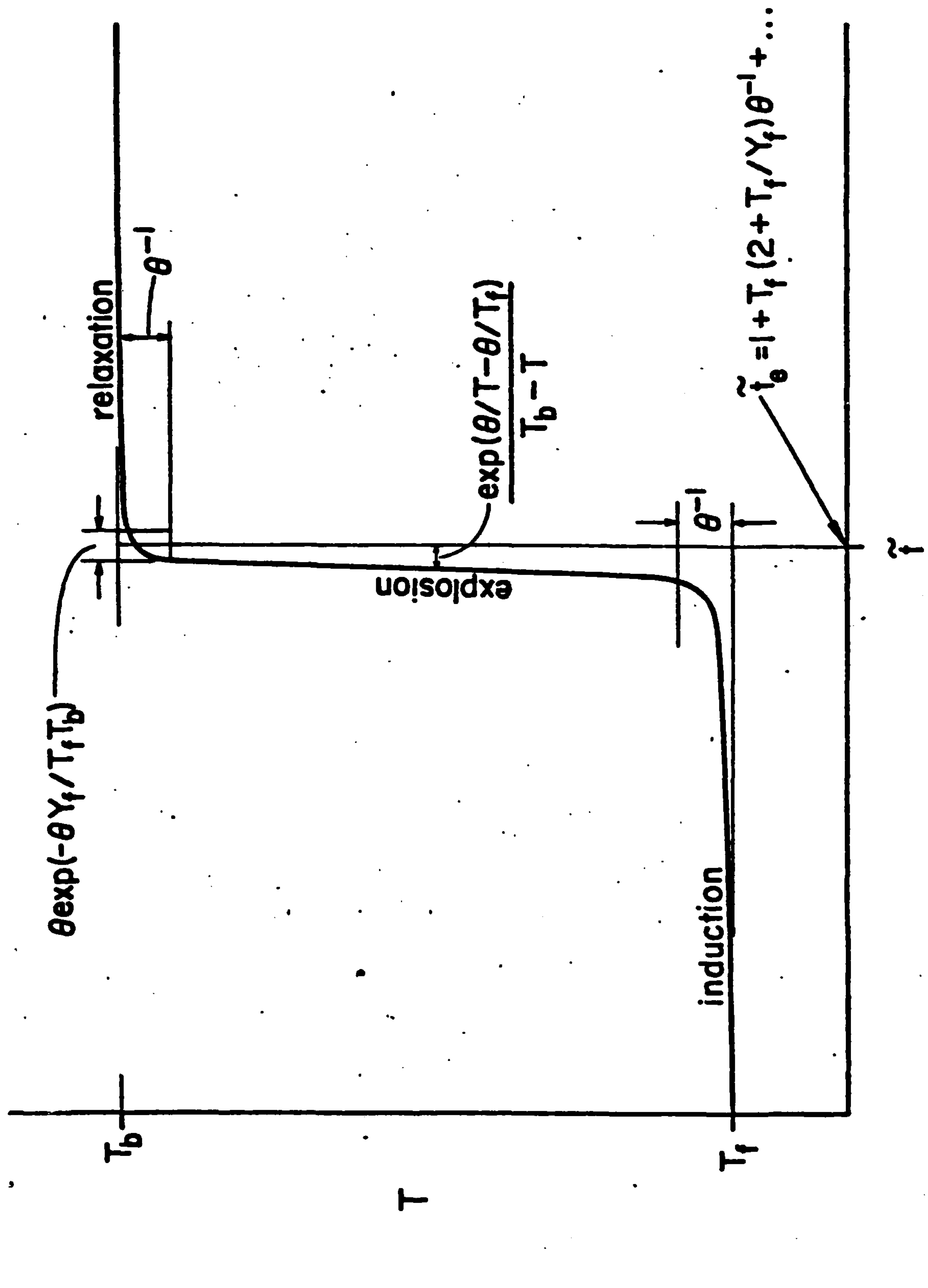
relaxation
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$\theta \exp(-\theta Y_f / T_f T_b)$
$\dfrac{\exp(\theta/T - \theta/T_f)}{T_b - T}$
explosion
θ^{-1}
$\tilde{t}_\theta = 1 + T_f(2 + T_f/Y_f)\theta^{-1} + \ldots$
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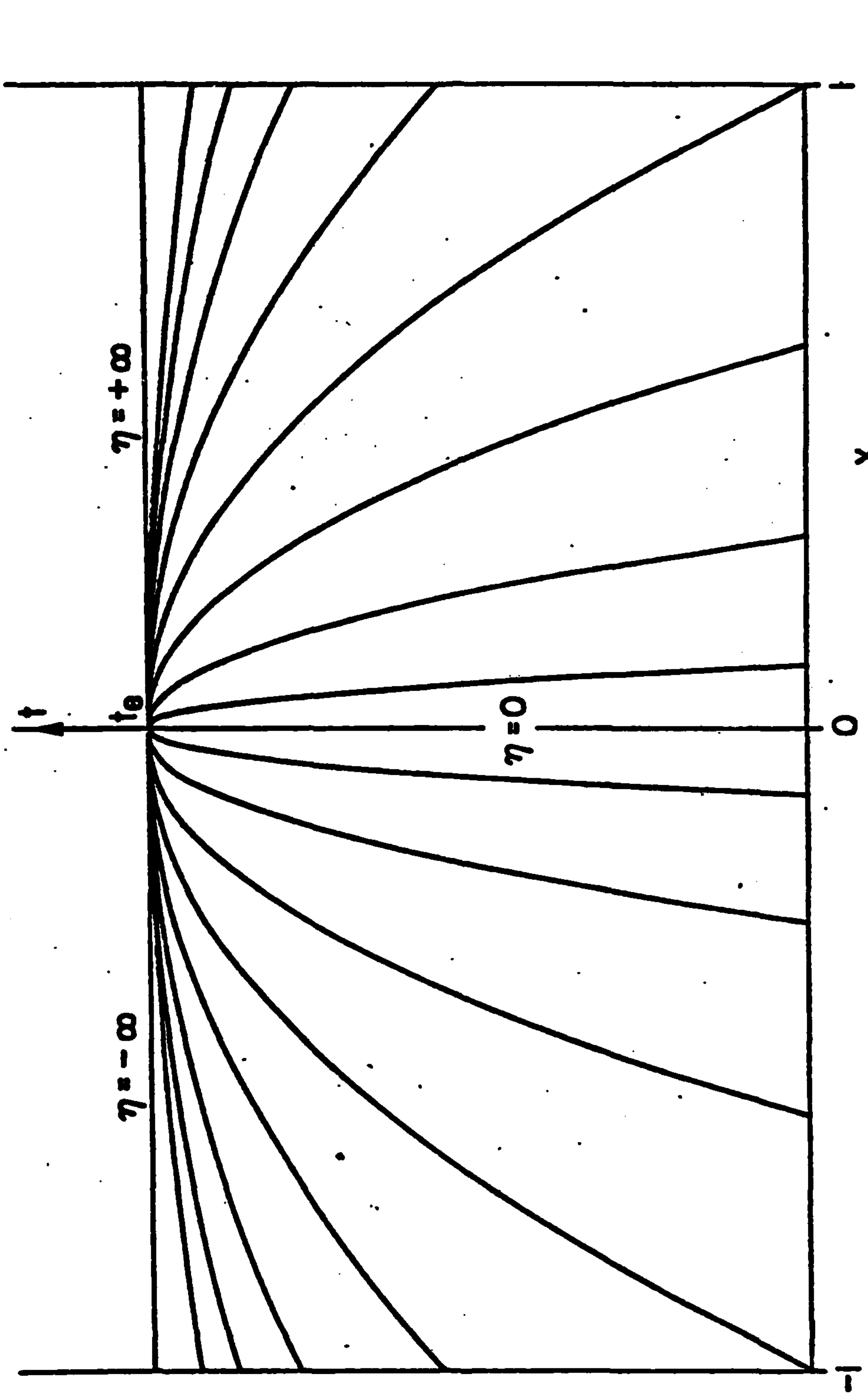
1.3

4.1



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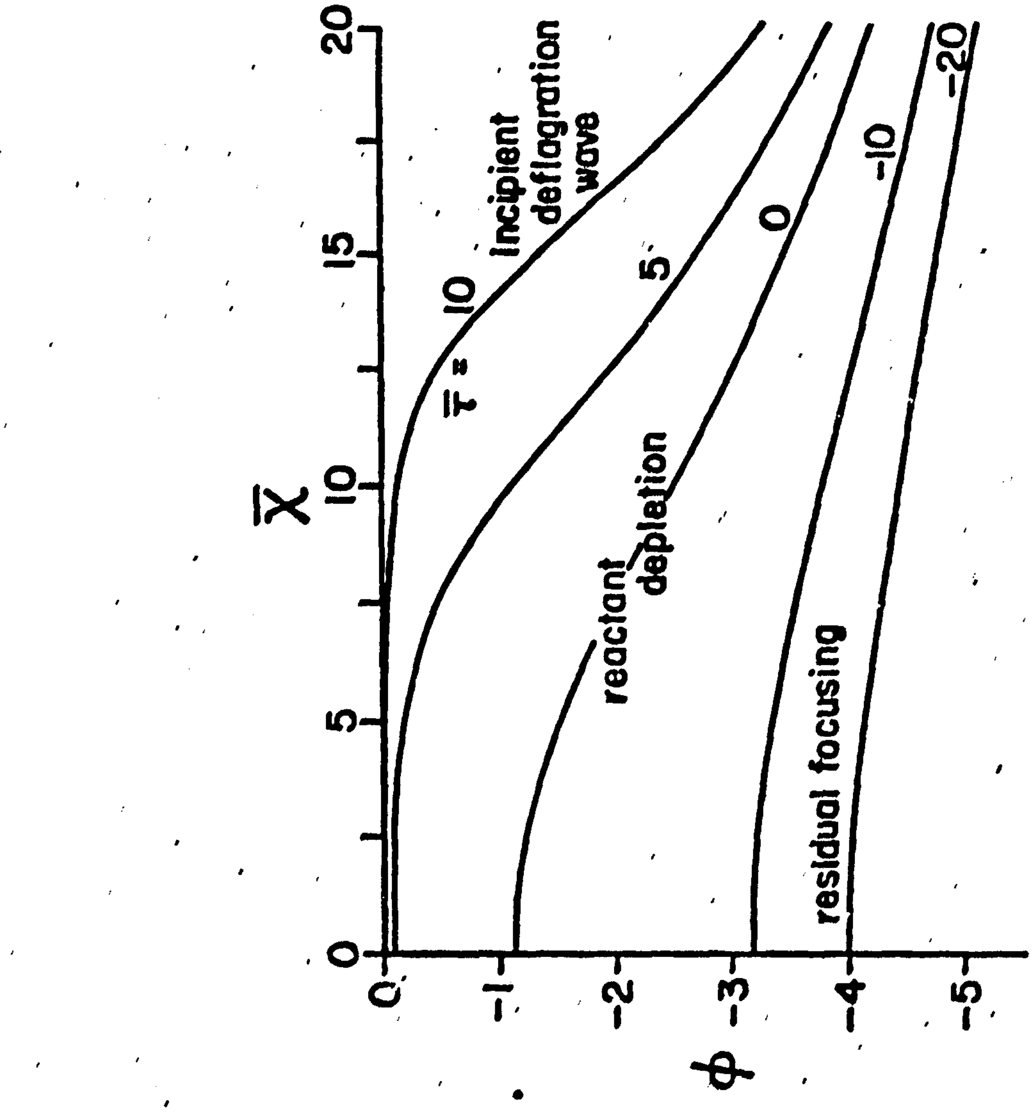

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Incipient deflagration wave
reactant depletion
residual focusing

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19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The description of reacting systems can be simplified when the so-called activation energy is large; the notion is an old one, but its full power is only released by modern singular perturbation theory. More than forty years ago, Frank-Kamenetskii introduced approximations based on large activation energy to construct a thermal theory of spontaneous combustion, and we shall start there. His problem, which neglects the fluid-mechanical effects of

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered) main concern to us, focuses attention on the reaction and thereby acts as a precursor for the lectures that follow. The problem and its generalizations have been the happy hunting grounds of mathematical analysts for many years, but it was not until quite recently that a complete description of the ignition and explosion processes was made available by Kapila and Kassoy (working separately) through activation-energy asymptotics, the main theme of these lectures.