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THE DYNAMIC STABILITY OF CONFINED, EXOTHERMICALLY REACTING FLUIDS

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Abstract—The importance of convective effects in exothermic chemical reactions taking place in the gaseous or liquid phase in a parallel-plate geometry is investigated by means of a linear stability analysis of the temperature distribution predicted by the classical conduction theory of thermal ignitions, in which the reactions are taken to be of zero order. The critical Rayleigh numbers for the onset of convection are determined by the numerical solution of an algebraic eigenvalue problem derived from the stability analysis using a 'selected points' method. The relationship of the theory to the observations of convective effects in experimental systems with spherical geometry, for which no equilibrium solutions of the purely conductive theory exist, is discussed.

NOMENCLATURE

A, pre-exponential factor in Arrhenius' Law;
d, radius of reaction vessel;
E, activation energy of reaction;
g, acceleration due to gravity;
$\boldsymbol{k}$, wave vector, see equation (25);
p, dimensionless pressure;
Pr, Prandtl number, ν/κ;
Q, exothermicity of reaction;
R, gas constant;
Ra, Rayleigh number, $gd^3\alpha RT_0^2/\kappa\nu E$;
$Ra^{(T)}$, modified Rayleigh number, $gd^3\alpha\Delta T/\kappa\nu$, see equation (40);
T, temperature;
$\boldsymbol{u}$, dimensionless velocity;
$\hat{\boldsymbol{z}}$, unit vector in vertical direction.

Greek symbols
α, coefficient of cubical expansion of fluid;
β, RT_0/E;
δ, $QEd^2A\exp(-E/RT_0)/\kappa RT_0^2$;
ΔT, $T - T_0$;
κ, thermal conductivity of fluid;
ν, kinematic viscosity of fluid;
ρ, density of fluid;
ϕ, dimensionless temperature.

Subscripts
CR, value at ignition limit;
$\boldsymbol{k}$, denotes dependence on wave vector $\boldsymbol{k}$;
min, minimum value as $\boldsymbol{k}$ varies;
0, value at wall of vessel;
x,y,z Cartesian components.

Superscripts
$\bar{}$, conduction-only solution;
$'$, perturbation quantity.

1. INTRODUCTION

In this paper we are concerned with the importance of convection in the heat transfer processes involved when exothermic chemical reactions take place in a fluid medium. Such reactions may be described by the theory of thermal ignitions, which has many practical applications to situations in which combustion and explosion occur. Following the initial contribution of Semenov [1], the classicial theory of thermal ignition described by Frank-Kamenetskii [2] was formulated, chiefly by Todes and Frank-Kamentskii. In this theory heat transfer is assumed to occur by means of conduction alone. However, for reactions taking place in the gaseous or liquid phase, convection may sometimes be expected to be a significant heat transfer mechanism, and the predictions of the classical theory may need to be modified accordingly. Indeed, of the three cases discussed in detail in [2], namely those in which the reacting medium is confined (i) between infinite horizontal planes, (ii) inside a circular cylinder and (iii) inside a sphere, only (i) can exhibit, for a fluid medium, the truly stable temperature distribution predicted by the classical theory, since in (ii) and (iii) the classical temperature distribution implies the existence of temperature gradients perpendicular to the direction of the gravity vector. Here we examine the stability of this classical conduction solution to small perturbations in order to predict when convection effects will become significant. The importance of convection effects has been noted experimentally by Tyler [3] and Ashmore, Tyler and Wesley [4] in spherical vessels, and Merzhanov and Shtessel [5] in parallel-plate vessels.

The purely conductive theory is governed by the steady-state diffusion equation for the temperature field, with a heat source term derived by assuming that the combustion reaction is of zero order, and that its rate depends on the temperature in accordance with Arrhenius' Law. The assumption of a zero order reaction may be regarded as an approximation which neglects the consumption of reactants in the course of the reaction. Frank-Kamenetskii's non-dimensionalisation scheme leads to an equation whose solutions are found to depend on two dimensionless parameters δ and β. A measure of the relative rates of supply and removal of heat, and hence of the likelihood of the system to explode, is provided by δ, whilst β characterises the explosive properties of the reacting medium, for a given temperature T_0 of the environment. Analytical solutions are presented by Frank-Kamenetskii [2] for the simpler form of the equation obtained in the limit $\beta \to 0$; numerical solutions for some non-zero values of β were found by Parks [6]. The aim of both investigations was to determine the ignition limit; this is the interpretation given to the critical value $\delta_{CR}(\beta)$ of δ above which no solutions of the steady-state equation exist. In Section 3 of this paper the 'selected points', or 'collocation', technique of Lanczos [7] is used to solve the non-linear conduction equation for several values of β. The results are compared with those given in [2] and [6]. Although attention is confined here to the parallel-plate geometry the other geometries considered in [2] are readily treated using the same technique.

In Section 4 we study convective effects in the case of parallel-plate geometry, by seeking critical Rayleigh numbers below which the predicted conduction solutions are stable to small disturbances. This we achieve by using linear stability theory in conjunction with the full fluid mechanical equations for the gaseous or liquid medium. The application of linear stability theory, of which a full description is given by Chandrasekhar [8], leads to an eigenvalue problem for a system of ordinary differential equations. Linear stability analyses of situations which differ from that considered here in the form of internal heat generation have been made by several authors; in the classical Bénard problem (see Rayleigh [9] and Pellew and Southwell [10]) there is no internal heat generation, whilst Roberts [11] solves the case in which there is uniform heat generation.

Here we represent the solution of the system of differential equations arising from the stability analysis by a finite series of Chebychev polynomials. Equations for the coefficients in the series are found by means of the 'selected points' method described in [7]. In this way the eigenvalue problem is transformed in Section 5 into an algebraic eigenvalue problem, for the solution of which many techniques are available. We adopt one of the so-called 'power' methods described, for example, by Wilkinson [12].

For each value of δ, the critical value $Ra_{\min}$ of the Rayleigh number thus calculated may be regarded as the smallest Rayleigh number at which convection. effects are significant. Hence for lower values of the Rayleigh number the predictions of the classical condition theory should be valid. However, for values above the critical value convection effects may be expected to be significant; considerable modification of the temperature profiles calculated from the purely conductive theory may then result. Furthermore, the ignition limit δ_{CR} will also be dependent upon the Rayleigh number. The enhancement of heat transfer processes when convection occurs will lead to higher values of δ_{CR} than those derived from the classical theory.

The critical Rayleigh numbers calculated here for the parallel-plate geometry are close to those at which convection was first observed to be significant in a spherical apparatus in [3] and [4]. Since, however, in a fluid medium the temperature profiles predicted by the purely conductive theory cannot exist in equilibrium in a spherical geometry, the experimental values may be regarded as stability limits at best only in a 'quasi-steady' sense. This possibility would be realised if convection were unimportant relative to conduction in establishing the basic profile, so that the convective effects which inevitably occur in the experimental situation could be considered to arise from perturbations of the type with which we are concerned here.

Unfortunately, although the experiments described in [5] were performed in a parallel-plate vessel, the minimum values of the Rayleigh number leading to noticeable convective effects were not determined.

2. THE GOVERNING EQUATIONS

In order to formulate the equations governing the dynamics and thermodynamics of the chemically reacting system studied, we must represent the heat production of the reaction by a spatial distribution of heat sources. If the exothermicity of the reaction is Q, and if the rate of reaction depends on the temperature T according to Arrhenius' Law, then the density q of heat sources is given by

$$q = QAf(\boldsymbol{c})\exp(-E/RT), \tag{1}$$

where A is the 'pre-exponential factor', E is the activation energy of the reaction, and $f(\boldsymbol{c})$ is a function of the reactant concentrations $\boldsymbol{c}$. Here we represent the combustion reaction by a zero-order reaction, so that $f(\boldsymbol{c}) = 1$. If distances are non-dimensionalised with respect to a length d typical of the vessel containing the reacting medium, the adoption of Frank-Kamenetskii's dimensionless temperature ϕ, defined by

$$\phi = \frac{E}{RT_0^2}(T - T_0), \tag{2}$$

where T_0 is a temperature typical of the system, allows us to write the continuity, momentum and energy equations governing the system in the non-dimensional forms

$$\nabla . \boldsymbol{u} = 0, \tag{3}$$

$$\frac{\partial \boldsymbol{u}}{\partial t} + \frac{1}{Pr}(\boldsymbol{u} . \nabla)\boldsymbol{u} = -\nabla p + \nabla^2 \mathbf{u} + Ra\phi\hat{\mathbf{z}}, \tag{4}$$

and

$$Pr\frac{\partial \phi}{\delta t} + (\boldsymbol{u} . \nabla)\phi = \nabla^2\phi + \delta\exp\{\phi/(1 + \beta\phi)\} \tag{5}$$

respectively. Here velocities are measured in units of κ/d, where κ is the thermal diffusivity, time in units of d^2/ν, where ν is the kinematic viscosity, and pressure in units of $\kappa\nu\rho_0/d^2$. In deriving the equations (3)–(5) it has been assumed that $|T - T_0| \ll T_0$ so that the Boussinesq approximation may be made; that is, the density ρ has been assumed to be related to the temperature by

$$\rho - \rho_0 = -\alpha\rho_0(T - T_0) \tag{6}$$

in the buoyancy term of (4) but elsewhere the density and all other physical parameters of the reacting fluid have been assumed constant. The dimensionless parameters characterizing the system are the Prandtl number

$$Pr = \nu/\kappa, \tag{7}$$

the Rayleigh number

$$Ra = gd^3\alpha RT_0^2/\kappa\nu E, \tag{8}$$

Frank-Kamenetskii's parameter

$$\delta = QEd^2A\exp(-E/RT_0)/\kappa RT_0^2, \tag{9}$$

and

$$\beta = RT_0/E. \tag{10}$$

The steady-state equations governing the classical conduction theory are obtained from (3)–(5) by setting $u \equiv 0$ and omitting the explicit time derivative terms. This gives

$$\nabla\bar{p} = Ra\bar{\phi}\hat{z}, \tag{11}$$

and

$$\nabla^2\phi + \delta\exp\{\bar{\phi}/(1 + \beta\bar{\phi})\} = 0, \tag{12}$$

where we use an overbar to denote solutions of the equations governing the purely conductive theory. In the next Section we consider the solution of these equations.

3. CONDUCTION-ONLY STATE

Solutions of equation (12) governing the temperature field in the steady-state purely conductive theory, and values of the ignition limit δ_{CR} obtained from it have been presented by several authors. When $\delta < \delta_{CR}$ a solution of (12) exists, but for $\delta > \delta_{CR}$ no solutions of the steady-state equation exist and according to this theory the system is expected to explode. Analytical solutions of (12) for the case of parallel-plate geometry with $\beta = 0$ are given by Frank-Kamenetskii [2], together with the ignition limit $\delta_{CR} = 0{\cdot}88$. Parks [6] uses standard finite-difference methods to integrate (12) for several non-zero values of β and tabulates δ_{CR} as a function of β. Na and Tang [13] transform the two-point boundary value problem posed by (12) and the associated boundary conditions for ϕ into an initial value problem in order to be able to employ a Runge–Kutta technique.

In Section 4 of this paper we shall be concerned solely with the stability of a system contained between infinite horizontal plates separated by a distance $2d$ and held at a constant temperature T_0. Here, therefore, we shall only discuss solutions of (12) appropriate to such a situation, although the method of solution described is also applicable to cases of cylindrical and spherical geometries when the temperature depends on the distance from the centre alone. Hence we consider

$$\frac{d^2\bar{\phi}}{dz^2} = -\delta\exp\{\bar{\phi}/(1 + \beta\bar{\phi})\}, \tag{13}$$

subject to the boundary conditions

$$\bar{\phi} = 0 \text{ on } z = \pm 1. \tag{14}$$

We solve the non-linear equation (13) iteratively, using a quasilinearized form of the equation at each stage of the iteration. Thus, if $\bar{\phi}^{(n)}$ denotes the nth iterate for $\bar{\phi}$, we neglect terms of second and higher orders in $(\bar{\phi}^{(n)} - \bar{\phi}^{(n-1)})$ to obtain the linear equation

$$\frac{d^2\bar{\phi}^{(n)}}{dz^2} + \frac{\delta\exp\{\bar{\phi}^{(n-1)}/(1 + \beta\bar{\phi}^{(n-1)})\}}{(1 + \beta\bar{\phi}^{(n-1)})^2}\bar{\phi}^{(n)} = -[\delta\exp\{\bar{\phi}^{(n-1)}/(1 + \beta\bar{\phi}^{(n-1)})\}] \times [1 - \bar{\phi}^{(n-1)}/(1 + \beta\bar{\phi}^{(n-1)})^2] \tag{15}$$

for the current iterate $\bar{\phi}^{(n)}$, which must satisfy the boundary conditions

$$\bar{\phi}^{(n)} = 0 \quad \text{on} \quad z = \pm 1. \tag{16}$$

We represent $\bar{\phi}^{(n)}$ by the series

$$\bar{\phi}^{(n)}(z) = \sum_{j=1}^{N} b_j T_{j-1}(z), \tag{17}$$

where T_j is the Chebychev polynomial of degree j. The coefficients b_j are determined by employing the 'selected points', or 'collocation', method of Lanczos [7]. Thus the boundary conditions (16) provide two equations for these coefficients and the remaining $(N - 2)$ equations required are obtained by satisfying (15) exactly at the points

$$\bar{z}_i = \cos\{(i-1)\pi/(N-3)\} \quad i = 1, \ldots, N-2. \tag{18}$$

The resulting set of N simultaneous linear equations for $b_1, \ldots, b_N$ is now solved using a standard numerical algorithm, and $\bar{\phi}^{(n)}$ then calculated from (17). The iteration cycle is repeated until successive iterates for $\bar{\phi}$ differ by less than a prescribed tolerance, which in our integrations was chosen to give 4 significant figures for $\bar{\phi}$.

When the choice (18) is made for the selected points, the errors in $\bar{\phi}^{(n)}$ arising from the truncation of the series (17) after N terms may be expected to be distributed fairly uniformly over the entire range $|z| \leqslant 1$. The coefficients b_j are found to decrease steadily in absolute value as j increases; consequently, since $|T_j(z)| \leqslant 1$ when $|z| \leqslant 1$ for all j, an estimate of the truncation error is available from the magnitude of the last few coefficients. The truncation error estimated in this way was 0 (10^{-9}) in our calculations for $N = 20$.

Temperature profiles for a range of values of β between 0·01 and 0·1 are presented in Fig. 1 for $\delta = 0{\cdot}3$ and $\delta = 0{\cdot}7$. These results were obtained using $N = 20$ in (17). The corresponding profiles obtained analytically for $\beta = 0$ by Frank-Kamenetskii are also plotted for comparison. Our profiles for $\beta = 0$ agree with those of Frank-Kamenetskii to at least 4 significant figures.

In Fig. 2 the behaviour of the ignition limit δ_{CR} as a function of β is shown, together with the corresponding results from Parks' calculations. Frank-Kamenetskii found the limiting value $\delta_{CR} = 0{\cdot}88$ when $\beta = 0$.

It must be emphasized that the equation (13) governing the purely conductive theory is appropriate to a fluid medium only when that medium is at rest. In the next Section we shall examine the stability of this solution, for which the velocity field is zero, when small perturbations of the velocity and temperature fields are

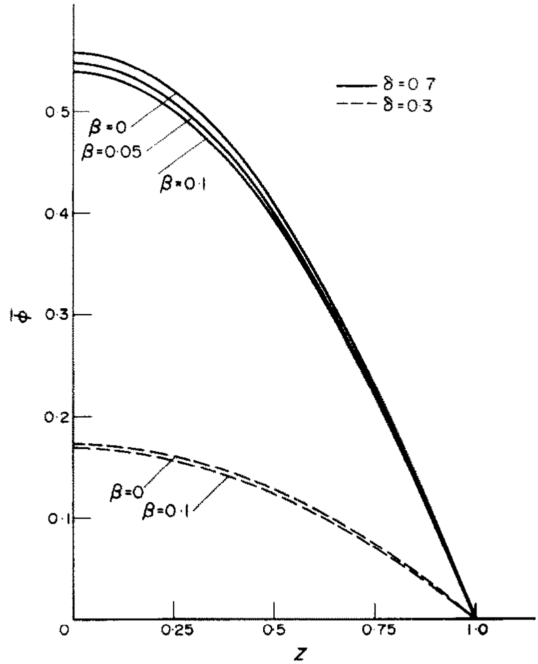

FIG. 1. Temperature profiles from the steady-state purely conductive theory.

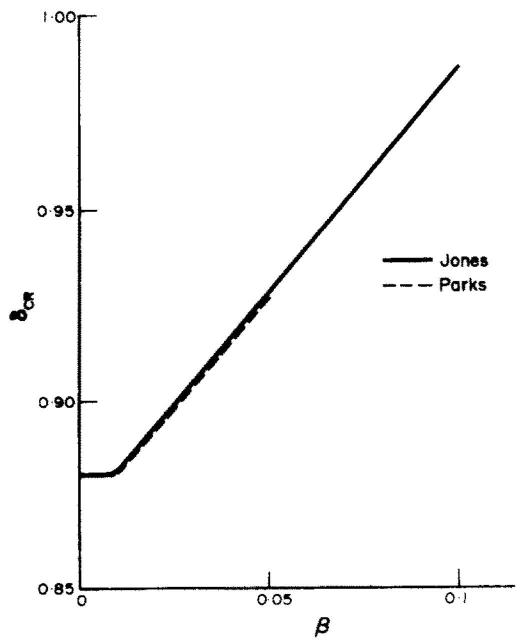

FIG. 2. Variation of the ignition limit δ_{CR} with β.

made. It will be found that for each set of values of β and δ for which $\delta < \delta_{CR}(\beta)$ a critical value $Ra_{\min}$ of the Rayleigh number exists, for values below which such perturbations will decay, but above which there will be some perturbations which do not decay. Thus for Rayleigh numbers below the critical value the conduction theory of this Section will be valid, whereas for values greater than $Ra_{\min}$ an appreciable modification may result from convective effects. For example, the ignition limits δ_{CR} for situations in which convection occurs will be higher than the corresponding limits for purely conductive systems in general.

4. LINEAR STABILITY ANALYSIS

In order to investigate the hydrodynamic stability of the conduction solutions discussed in Section 3 we examine the behaviour of small temperature and velocity perturbations of these solutions. We suppose that ϕ, p and $\boldsymbol{u}$ result from perturbations ϕ, p' and $\boldsymbol{u}'$ of the base state, so that

$$\phi = \bar{\phi} + \phi',$$
$$p = \bar{p} + p' \tag{19}$$
and
$$\boldsymbol{u} = \bar{\boldsymbol{u}} + \boldsymbol{u}'.$$

Then ϕ, p and $\boldsymbol{u}$ must satisfy equations (3)–(5) and the boundary conditions

$$\phi = 0 \quad \text{and} \quad \boldsymbol{u} = 0 \quad \text{on} \quad z = \pm 1, \tag{20}$$

for all time. By neglecting terms of second and higher orders in the perturbation quantities we obtain the linear equations

$$\nabla . \boldsymbol{u}' = 0, \tag{21}$$

$$\frac{\partial \boldsymbol{u}'}{\partial t} = -\nabla p' + \nabla^2 \boldsymbol{u}' + Ra\phi'\hat{\boldsymbol{z}}, \tag{22}$$

and

$$Pr\frac{\partial \phi'}{\partial t} + (\boldsymbol{u}'.\nabla)\bar{\phi} = \nabla^2\phi' + \frac{\delta \exp\{\bar{\phi}/(1+\beta\bar{\phi})\}}{(1+\beta\bar{\phi})^2}\phi', \tag{23}$$

since $\bar{\boldsymbol{u}} = 0$. The boundary conditions for ϕ' and $\boldsymbol{u}'$ are

$$\phi' = 0 \quad \text{and} \quad \boldsymbol{u}' = 0 \quad \text{on} \quad z = \pm 1. \tag{24}$$

It may be noted here that similar stability analyses have been applied by other authors to problems which differ from that under consideration only in the form of the internal heat generation term. For the classical Bénard problem, discussed by Rayleigh [9] and Pellew and Southwell [10] amongst others, this term is zero, while in the case of uniform heat generation considered by Roberts [11] the last term in (5) is replaced by unity.

We now follow the familiar procedure, described in detail by, for example, Chandrasekhar [8], of representing the general perturbation we have introduced as an integral over all possible wave vectors. In view of the linearity of the problem we may examine the stability of the mode associated with each wave vector separately. A typical mode may be written

$$u'_z = U_{\boldsymbol{k}}(z)\exp\{i(\boldsymbol{k}.\boldsymbol{x}) + \omega_{\boldsymbol{k}}t\},$$
and
$$\phi' = \Phi_{\boldsymbol{k}}(z)\exp\{i(\boldsymbol{k}.\boldsymbol{x}) + \omega_{\boldsymbol{k}}t\}, \tag{25}$$

where the wave vector $\boldsymbol{k}$ has cartesian components $(k_x, k_y, 0)$ and u'_x, u'_y may be expressed in terms of $U_{\boldsymbol{k}}(z)$ using (21). Substitution of the forms (25) into equations (22) and (23) yields the equations

$$(D^2 - k^2)^2 U_{\boldsymbol{k}} - \omega_{\boldsymbol{k}}(D^2 - k^2)U_{\boldsymbol{k}} = Rak^2\Phi_{\boldsymbol{k}}, \tag{26}$$

and

$$(D^2 + F(z) - k^2 - \omega_{\boldsymbol{k}})\Phi_{\boldsymbol{k}} = (D\bar{\phi})U_{\boldsymbol{k}} \tag{27}$$

for the functions $U_{\boldsymbol{k}}(z)$ and $\Phi_{\boldsymbol{k}}(z)$, where

$$F(z) = \delta\exp\{\bar{\phi}/(1+\beta\bar{\phi})\}/(1+\beta\bar{\phi})^2 \tag{28}$$

is a known function of dz $D \equiv d/dz$, and $k^2 = |\boldsymbol{k}|^2$. The boundary conditions on $z = \pm 1$ are

$$\Phi_{\boldsymbol{k}} = U_{\boldsymbol{k}} = DU_{\boldsymbol{k}} = 0. \tag{29}$$

The last condition may be derived from (24) using the continuity equation (21).

The boundary between stability and instability for a given wave vector $\boldsymbol{k}$ is given by the smallest value of Ra for which a non-trivial solution of (26) and (27) subject to the conditions (29) exists when $Re(\omega_{\boldsymbol{k}}) = 0$. At this stage in the application of linear stability analysis to the Bénard problem, for which the base profile $\bar{\phi}$ is linear in z, it is possible to establish the Principle of Exchange of Stabilities, which states that if $Re(\omega_{\boldsymbol{k}}) = 0$ then $\mathrm{Im}(\omega_{\boldsymbol{k}}) = 0$ also. Here, however, as in the case of uniform heat addition [11], we are unable to obtain such a Principle, but there seems no reason to suppose that it is violated. Accordingly, we assume $\omega_{\boldsymbol{k}} = 0$ in (26) and (27) for our marginal stability analysis. Then, if we eliminate $\Phi_{\boldsymbol{k}}$ the critical Rayleigh number we seek is, for each $\boldsymbol{k}^2$, the smallest positive eigenvalue of the sixth order equation

$$(D^2 - k^2)^2(D^2 + F(z) - k^2)U_{\boldsymbol{k}} = Rak^2(D\bar{\phi})U_{\boldsymbol{k}}, \tag{30}$$

subject to the boundary conditions

$$U_{\boldsymbol{k}} = DU_{\boldsymbol{k}} = (D^2 - k^2)^2 U_{\boldsymbol{k}} = 0 \text{ on } z = \pm 1. \tag{31}$$

For given values of β and δ, the minimum, $Ra_{\min}$, of these eigenvalues Ra as k^2 varies is the Rayleigh number at which we may expect convection to occur first, as the Rayleigh number is increased gradually from zero.

In Section 5 we describe the conversion of the differential eigenvalue problem into an algebraic eigenvalue problem and the numerical method adopted to solve the latter.

5. THE ALGEBRAIC EIGENVALUE PROBLEM

The eigenvalue problem defined by (30) and (31) may be solved by one of several techniques, for example variational or 'shooting' methods. However, the suitability of the 'selected points' method for the solution of two-point boundary value problems allows the explicit formulation of the problem as an algebraic eigenvalue problem, for which powerful solution techniques are available. Hence, as for the conduction solutions, we represent $U_{\boldsymbol{k}}(z)$ by a finite Chebychev series

$$U_{\boldsymbol{k}}(z) = \sum_{j=1}^{M} a_j T_{j-1}(z), \tag{32}$$

and find relationships between the coefficients a_j using the 'selected points' method. If $\boldsymbol{a}$ is the vector $\begin{pmatrix} a_1 \\ \vdots \\ a_M \end{pmatrix}$, the six boundary conditions (31) give six linear relationships between the components of $\boldsymbol{a}$. We may write these six equations in the form

$$\boldsymbol{g}^{(m)} . \boldsymbol{a} = 0, \tag{33}$$

where

$$[\boldsymbol{g}^{(1)}]_j = T_{j-1}(z)|_{z=1},$$

$$[\boldsymbol{g}^{(2)}]_j = DT_{j-1}(z)|_{z=1},$$

and

$$[\boldsymbol{g}^{(3)}]_j = (D^2 - k^2)^2 T_{j-1}(z)|_{z=1}, \quad \text{for } j = 1, \ldots, M \tag{34}$$

and the remaining $\boldsymbol{g}^{(m)}$s are similarly obtained from the boundary conditions at $z = -1$.

By evaluating (30) at (M-6) selected points z_i we may derive another (M-6) relationships between the components of $\boldsymbol{a}$. These may be written

$$\boldsymbol{d}^{(i)} . \boldsymbol{a} = Ra\ \boldsymbol{e}^{(i)} . \boldsymbol{a} \qquad i = 1, \ldots, M - 6 \tag{35}$$

where

$$[\boldsymbol{d}^{(i)}]_j (D^2 - k^2)^2 (D^2 + F(z) - k^2) T_{j-1}(z)|_{z=z_i} \tag{36}$$

and

$$[e^{(i)}]_j = k^2(D\bar{\phi}(z))T_{j-1}(z)\big|_{z=z_i} \quad \text{for } i = 1,\ldots, M-6 \quad \text{and } j = 1,\ldots, M. \tag{37}$$

It is, of course, preferable that the selected points z_i be a subset of the $\bar{z}_i$ defined by (18). Combining (33) and (35) we must solve

$$\begin{pmatrix} d^{(1)} \\ \vdots \\ d^{(M-6)} \\ \hline g^{(1)} \\ \vdots \\ g^{(6)} \end{pmatrix} a = Ra \begin{pmatrix} e^{(1)} \\ \vdots \\ e^{(M-6)} \\ \hline 0 \end{pmatrix} a. \tag{38}$$

Since the matrix on the left-hand side of (38) is non-singular we may multiply both sides of (38) by its inverse and so write the equation in the form

$$Aa = \mu a \tag{39}$$

where $\mu = 1/Ra$ and A is a known $M \times M$ matrix.

Thus we require the largest positive eigenvalue μ (corresponding to the smallest positive eigenvalue Ra) of the equation (39). When this is the sole requirement, the most appropriate technique of solution is one of the 'power' methods described, for example, by Wilkinson [12]. These rely on the observation that repeated multiplication of an arbitrary vector by a matrix will result in a vector dominated by eigenvectors of the matrix corresponding to its eigenvalue of largest modulus, so that the ratio of successive iterates will tend to this eigenvalue as the iteration proceeds. In practice it was found that the 'inverse iteration' variant was the most efficient for our problem: the required first approximations to the eigenvalues at given

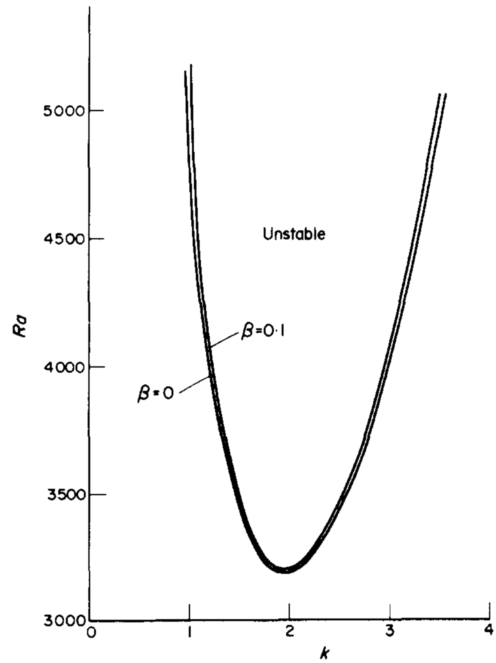

FIG. 3. Marginal stability curves for $\delta = 0{\cdot}3$.

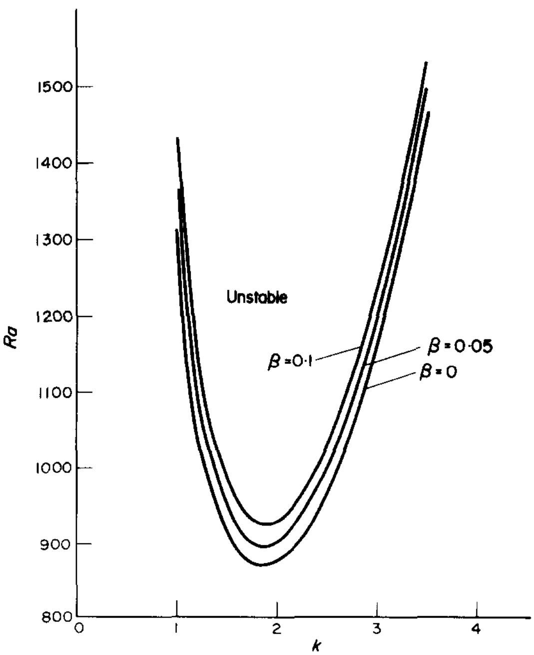

FIG. 4. Marginal stability curves for $\delta = 0{\cdot}7$.

values of δ, β and k were readily provided by the eigenvalues at neighbouring values of these parameters.

Figures 3 and 4 show the variation of the critical Rayleigh number with the wavenumber k for the basic profiles presented in Fig. 1. All of the results to which reference is made here relate to the case $Pr = 1$. The results plotted in Figs. 3 and 4, which are correct to 6 significant figures, were obtained by taking $M = 12$ in (32). The use of $M = 24$ enables us to calculate critical Rayleigh numbers correct to 7 significant figures.

A check on this method of calculation of critical Rayleigh numbers was made by applying it to the problems solved by Roberts [11] and

Table 1. Critical Rayleigh numbers for other problems—a comparison of results

	Roberts	Pellew and Southwell	Present work $M = 12$	$M = 24$
Uniform internal heat generation	2772·28	—	2772·27	2772·274
Zero internal heat generation	—	1707·8	1707·76	1707·763

Pellew and Southwell [10] using other techniques. A comparison of the values calculated for the minimum, $Ra_{\min}$, of the marginal stability curve in each case is made in Table 1.

6. CONCLUDING REMARKS

In Fig. 5 the variation with δ of the calculated values of $Ra_{\min}$ for several values of β is displayed. These values may be compared with Frank-Kamenetskii's estimate of 10^4 for $Ra_{\min}$. Since $Ra_{\min}$ does not tend to zero as δ approaches the ignition limit δ_{CR} predicted by the classical theory it appears that according to our stability analysis in this particular geometry it is possible, for sufficiently small values of the Rayleigh number, to proceed to an explosion without convective effects ever becoming significant. However, in such a situation some features of our model, for example, the Boussinesq approximation, may no longer be appropriate. As already indicated, the onset of convection at Rayleigh numbers above $Ra_{\min}$ should be expected to lead to modifications in the ignition limits δ_{CR} from the values predicted by the classical theory. The improvement of heat transfer processes which will accompany the occurrence of convection should allow the existence of a non-explosive regime for some values of β and δ which would otherwise give rise to an explosion.

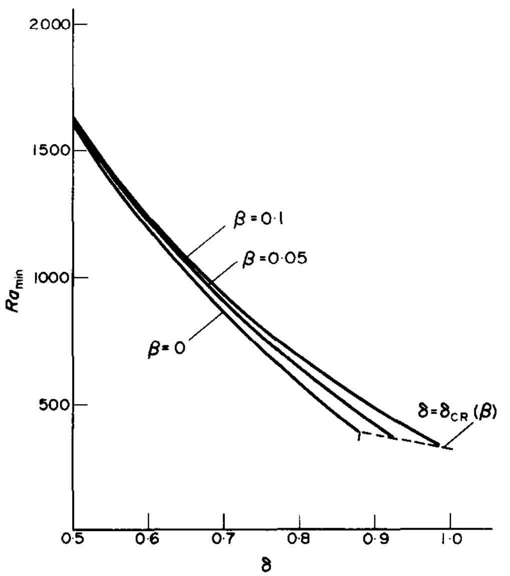

FIG. 5. Variation of the critical Rayleigh number with δ.

Tyler [3] and Ashmore, Tyler and Wesley [4] adopt a definition of the Rayleigh number which is different from that used throughout this paper. Their definition is

$$Ra^{(T)} = gd^3\alpha\Delta T/\kappa\nu, \tag{40}$$

where $\Delta T = T - T_0$. Thus we have

$$Ra^{(T)} = \phi|_{z=0} . Ra. \tag{41}$$

If we denote by $Ra^{(T)}_{\min}$ the minimum, for given δ and β, of $Ra^{(T)}$ as k^2 varies, then

$$Ra^{(T)}_{\min} = \phi|_{z=0} . Ra_{\min}. \tag{42}$$

Figure 6 shows the dependence of $Ra^{(T)}_{\min}$ on δ.

In [3] and [4] convection effects were observed to be significant for $Ra^{(T)} \gtrsim 600$. However, as was emphasised in Section 1, for a fluid medium enclosed in a spherical or cylindrical vessel the temperature profiles predicted by the purely conductive theory cannot exist in equilibrium.

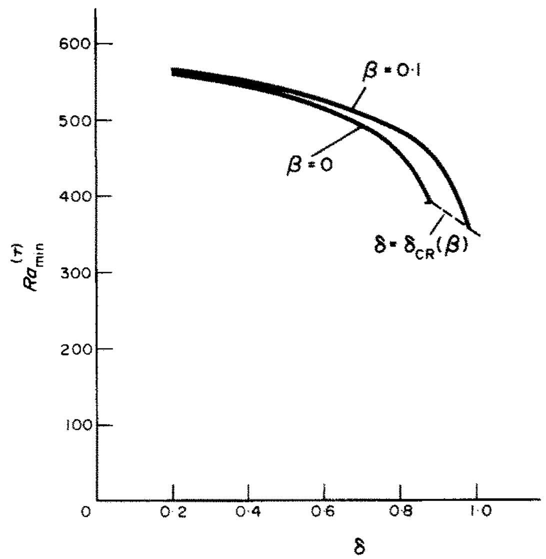

FIG. 6. Variation of the critical modified Rayleigh number with δ.

Nevertheless, perturbation effects of the type discussed here may be directly relevant to the experimental situation if a quasi-steady temperature profile develops, principally by conduction mechanisms, whilst the inevitable convection effects remain small. Further light could be shed on this problem by numerical integrations of the full equations (3)–(5) in geometries corresponding to the experimental system.

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LA STABILITE DYNAMIQUE DE FLUIDES CONFINES EN REACTION EXOTHERMIQUE

Résumé—L'importance des effets convectifs dans les réactions chimiques exothermiques ayant lieu dans la phase gazeuse ou liquide dans une géométrie à plans parallèles est étudiée à l'aide de l'analyse linéaire de stabilité de la distribution de température prédite par la théorie de conduction classique d'ignition thermique dans laquelle les réactions sont prises d'ordre zéro. Les nombres critiques de Rayleigh pour l'établissement de la convection sont déterminés par la solution numérique d'un problème à valeurs propres dérivé de l'analyse de stabilité qui utilise une méthode "à points sélectionnés". On discute la relation entre la théorie et les observations des effets convectifs dans des systèmes expérimentaux à géométrie sphérique pour laquelle existent des solutions hors équilibre de la théorie purement conductive.

DIE DYNAMISCHE STABILITÄT VON BEGRENZTEN FLUIDEN MIT EXOTHERMER CHEMISCHER REAKTION

Zusammenfassung— Die Bedeutung der konvektiven Effekte bei exothermen chemischen Reaktionen, die in gasförmiger oder flüssiger Phase zwischen parallelen Platten stattfinden, wurde mittels einer linearen Stabilitätsanalyse der Temperaturverteilung untersucht. Die Temperaturverteilung wurde durch die klassische Leitungstheorie der thermischen Zündung bestimmt; die Reaktionen wurden als solche nullter Ordnung angenommen. Die kritische Rayleigh-Zahl für das Einsetzen der Konvektion wurde durch die numerische Lösung eines algebraischen Eigenwertproblems gelöst. das von der Stabilitätsanalyse mittels einer Methode der "ausgewählten Punkte" abgeleitet wurde. Es wurde der Zusammenhang der Theorie mit den Beobachtungen konvektiver Effekte in Experimenten mit Kugelgeometrie diskutiert, für die keine Gleichgewichtslösungen für das reine Leitungsproblem existieren.

ДИНАМИЧЕСКАЯ УСТОЙЧИВОСТЬ ЭКЗОТЕРМИЧЕСКИ РЕАГИРУЮЩИХ ЖИДКОСТЕЙ В НЕБОЛЬШОМ ОБЪЕМЕ

Аннотация—Влияние конвекции при экзотермических реакциях в жидкой или газовой фазах в плоско-параллельных каналах оценивалось по устойчивости линейного профиля температур, расчитанного по классической теории теплопроводности для теплового воспламенения при реакциях нулевого порядка. Критическое число Рэлея, характеризующее возникновение конвекции, определялось из численного решения алгебраической задачи о собственных значениях, вытекающей из анализа устойчивости, с помощью метода отдельных точек. Обсуждается соответствие теории с наблюдаемыми конвективными эффектами при экспериментальном исследовании сферических объемов, для которых чистая теории теплопроводности не дает равновесных решений.