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Convective stability of the horizontal reacting liquid layer in the presence of various complicating factors

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Abstract—The convective stability of a horizontal chemically active liquid layer is studied within which an exothermal zero-order reaction takes place in the presence of a number of complicating factors: warming of the layer, different thermal conductivities of the boundaries and transverse motion of the reactant. Using the Runge–Kutta method, numerical investigation of the linear spectral stability problem of the steady-state regimes of heat transfer determined by solving the generalized non-linear problem of thermal explosion is carried out. The results obtained demonstrate a substantial influence of the above-listed complicating factors on the critical conditions for the thermal explosion and on the limit of the incipience of the reactant convective motion.

1. INTRODUCTION

GENERALLY, the chemical reaction proceeding in a liquid or gas leads to the liberation (absorption) of heat and to the formation of a product, the density of which differs from that of the reactant. The non-uniformities of the density thus produced in the gravity field may induce convection in a moving reacting mixture. The interest in the studies of such situations is justifiable from different viewpoints, in particular in connection with the elucidation of the effect which convective motion exerts on the rate of reaction. In those cases when the character of the density non-uniformity and the force field are such that a mechanical equilibrium is possible, the problem of the stability of the latter suggests itself. The non-uniform composition of the mixture and the reaction-induced temperature stratification lead to peculiar convective instability with the chemical activity of the medium serving both as the main reason for the instability and as a strong additional factor. Basically, there may well be, and are in the literature, different statements of the stability problems in accord with the type of reaction, relative role of the thermal effect, conditions on the boundaries of the region, etc. A review of some of the early relevant works can be found in ref. [1].

Pertaining to the kinetics of a chemical process, the most simple situation is the problem of the occurrence of convection in a horizontal layer of liquid in which an exothermal reaction takes place with an appreciable thermal effect. It is then possible to neglect the formation of the product and the dependence of heat release on concentration (model reaction of zero order); in this case the strength of the inner heat sources increases exponentially with temperature. At constant and identical temperatures of the boundaries, the temperature distribution across the layer is maximal in the middle of the layer. The steady-state heat conducting regime is characterized by the Frank-Kamenetskii dimensionless parameter Fk. Such a regime is possible when $Fk < Fk_{CR}$ where Fk_{CR} is a certain characteristic value; when $Fk > Fk_{CR}$ the steady-state regime is impossible, since thermal explosion is initiated. However, even when $Fk < Fk_{CR}$ the steady-state regime may be disturbed due to convective instability. The problem of convective stability of such a system and of the influence of developed convection on the thermal explosion threshold Fk_{CR} was formulated for the first time in the monograph by Frank-Kamenetskii [2]. The parameter that determines the convective stability is the Rayleigh number Ra. Its critical value (in the sense of convection occurrence), Ra_*, depends on the Frank-Kamenetskii parameter. The first good estimate of this dependence was obtained in the work of Merzhanov and Shtessel [3]. The solution of the linear problem of stability was made by Jones [4] who numerically calculated the function $Ra_*(Fk)$ for this case. The critical value of Ra_* decreases monotonically with the rise of Fk and has a final value at the point $Fk = Fk_{CR}$.

The linear approach allows the determination of the limit of convective stability depending on the reaction parameters. There naturally arises another question, namely about the effect of the originating convection on the limit of existence of the steady-state regime. The elucidation of this problem requires an essentially non-linear approach. It turns out that the increase in the intensity of heat removal due to developed convection substantially increases the limiting values of the Frank-Kamenetskii parameter Fk_{CR}. Of the theoretical studies of this kind note should be made of numerical calculations by the method of grids in a square domain [5], a horizontal circular cylinder [6] and in a horizontal layer [7], which made it possible to determine the functions $Fk_{CR}(Ra)$.

The present paper is concerned with a review of

NOMENCLATURE

a	integration constant
Bi	Biot number, $\alpha_i d/\kappa$ ($i = 1, 2$)
c_p	specific heat at constant pressure
d	layer thickness
E	activation energy
$\mathbf{e}$	unit vector directed vertically upward
Fk	Frank-Kamenetskii parameter, $QEd^2k_0 \exp(-E/RT_0)/\kappa RT_0^2$
g	gravity acceleration
k	wave number, $\sqrt{(k_1^2+k_2^2)}$
k_0	pre-exponential factor
p	convective contribution to pressure
Pe	Peclet number, $v_0 d/\chi$
Pr	Prandtl number, ν/χ
Q_R	thermal effect of reaction
R	universal gas constant
Ra	analogue of Rayleigh number, $g\alpha RT_0^2 d^3/E\nu\chi$
T	absolute temperature
T_0, T_1	absolute temperatures of layer boundaries
t	time
$\mathbf{v}$	velocity of convective motion
$\mathbf{v}_0$	velocity of transverse motion of reactant
W	chemical reaction rate
w	amplitude of velocity perturbation
x, y, z	Cartesian coordinates.

Greek symbols

α	coefficient of thermal expansion, $(-\partial\rho/\partial T)_p/\rho_0$
α_i	heat transfer coefficient on boundaries
β	RT_0/E
Θ	$T-T_0$
Θ_0	equilibrium value of temperature
Θ_{0m}	maximum value of Θ_0
θ	amplitude of temperature perturbation
κ	thermal conductivity, $\chi\rho_0 c_p$
λ	decrement of perturbations, $\lambda_r + i\lambda_i$
μ_n	hydrodynamic levels of the spectrum of decrements
ν	kinematic viscosity
ν_n	thermal levels of the spectrum of decrements
ρ_0	mean density
τ	$\lvert T_0 - T_1\rvert E/RT_0^2$
χ	thermal diffusivity.

Subscripts

*	minimum critical values of Rayleigh number and of the corresponding wave number
CR	critical values of the Frank-Kamenetskii parameter
i	boundary of the layer
n	levels of the spectrum of decrements
+	values of the Frank-Kamenetskii parameter up to which the transverse temperature gradient retains constant sign throughout the layer.

Superscript

$'$	derivative with respect to vertical coordinate.

the results of linear investigation into the convective stability of steady-state regimes of heat transfer in a horizontal reacting liquid layer in which an exothermal zero-order reaction takes place in the presence of various external effects. The influence of the heating of the layer from above and below, of different regimes of heat removal through the layer boundaries, and of transverse homogeneous reactant motion through the permeable layer boundaries on the conditions of convection initiation is considered.

The solution of the problem regarding the possible mechanical equilibrium of a chemically active liquid exposed to the influence of the above-listed complicating factors is reduced to the study of various generalizations of the classical problem of thermal explosion, the results of which are also presented in this paper.

2. EQUATIONS OF CONVECTION IN REACTING MEDIA

The convective motion of a reacting medium is represented by a system of convection equations for a chemically inert liquid [8] with additional terms characterizing the action of inner thermal and concentrational sources, the specific form of which is governed by the reaction kinetics. The case considered is an exothermal reaction occurring throughout the entire layer corresponding to the zero-order reaction model. In this case, the reaction rate does not depend on the reactant concentration, and increases exponentially with temperature according to the Arrhenius law, $W = k_0 \exp(-E/RT)$. Such a model can be applied to fast reactions with great release of heat when thermal processes develop against the background of virtually invariable concentration of a reactant.

The system of dimensionless equations of free thermal convection in an incompressible medium with zero-order reaction in the Boussinesq approximation [8] has the form

$$\frac{\partial \mathbf{v}}{\partial t} + \frac{1}{Pr}(\mathbf{v}\nabla)\mathbf{v} = -\nabla p + \Delta\mathbf{v} + Ra\,\Theta\mathbf{e}$$

$$Pr\frac{\partial\Theta}{\partial t} + \mathbf{v}\nabla\Theta = \Delta\Theta + Fk\exp\frac{\Theta}{1+\beta\Theta}$$

$$\operatorname{div} \mathbf{v} = 0. \tag{1}$$

The quantities d, d^2/ν, χ/d, RT_0^2/E and $\rho_0\nu\chi/d^2$ were selected as the units for measuring the distance, velocity, temperature and pressure. Besides the parameters Ra and Fk mentioned above, equations (1) involve the Prandtl number, $Pr = \nu/\chi$, and the small dimensionless parameter $\beta = RT_0/E$.

The inner heat generation in the layer and the removal of the heat through the boundaries results in non-uniform heating of the reacting liquid and in the incipience of convection. However, there are special conditions in a horizontal layer under which steady-state heat transfer is possible in it; then the convective motion is absent and all the variables are independent of time. It can be shown [8] that in this case the temperature gradient must be vertical. As regards the velocity of the medium, it is either equal to zero (mechanical equilibrium), or there is homogeneous transverse constant-velocity motion in the layer due to vertical injection of liquid through one permeable boundary of the layer and suction through the other. It follows from equations (1) that in the first case the temperature distribution in the layer is governed by the well known equation of Frank-Kamenetskii [2]

$$\Theta_0'' + Fk \exp\frac{\Theta_0}{1+\beta\Theta_0} = 0 \tag{2}$$

and in the second case by its correlation

$$\Theta_0'' - Pe\,\Theta_0' + Fk \exp\frac{\Theta_0}{1+\beta\Theta_0} = 0. \tag{3}$$

The dimensionless Peclet number, $Pe = v_0 d/\chi$, characterizes the intensity of the transverse motion of the reactant.

The solution of equations (2) and (3) subject to the corresponding boundary conditions that ensure the verticality of the temperature gradient makes it possible to find steady-state temperature profiles and the areas of the parameter values in which they exist.

In order to elucidate the problem of convective stability of these steady-state heat transfer regimes, small unsteady-state perturbations, periodic in the plane (x, y), will be considered according to the linear theory of stability:

$$v_z = w(z) \exp[-\lambda t + i(k_1 x + k_2 y)]$$
$$\Theta = \theta(z) \exp[-\lambda t + i(k_1 x + k_2 y)] \tag{4}$$

where $\lambda = \lambda_r + i\lambda_i$ is the complex decrement of perturbations; k_1 and k_2 are the wave numbers; the stability limit is determined by the condition $\lambda_r = 0$; the quantity λ_i characterizes the frequency of fluctuations. After the substitution of equation (4) into system (1), and linearization and elimination of the pressure, a system of ordinary differential equations is obtained for the amplitudes of normal disturbances w and θ:

$$-\lambda(w'' - k^2 w) + \frac{Pe}{Pr}(w''' - k^2 w') = (w^{IV} - 2k^2 w'' + k^4 w) - Ra\,k^2\Theta$$

$$-\lambda\, Pr\,\theta + Pe\,\theta' + w\Theta_0' = (\theta'' - k^2\theta) + Fk\,\theta\frac{\exp[\Theta_0/(1+\beta\Theta_0)]}{(1+\beta\Theta_0)^2}. \tag{5}$$

In the present work, the convective stability of a reacting liquid is investigated for the following versions of the problem statement that differ in boundary conditions and in the effect of complicating factors:

(1) The layer is bounded by solid, impermeable infinitely heat-conducting planes having the same temperature.

(2) Solid impermeable boundaries of the layer are maintained at different temperatures; the cases of heating from below and from above are considered.

(3) The boundaries of the layer have an arbitrary thermal conductivity.

(4) In the layer with the isothermal boundaries, a uniform transverse pumping of the reactant is made at a constant speed.

It is necessary to formulate the corresponding boundary conditions for the amplitudes of disturbances. In all cases the velocity of convective motion $\mathbf{v}$ on the boundaries of the layer disappears, and the general conditions are

$$w(0) = 0, w'(0) = 0, w(1) + 0, w'(1) = 0. \tag{6}$$

Two types of boundary conditions are considered for temperature disturbances. In the case of infinitely conducting boundaries (versions 1, 2 and 4) the temperature disturbances on them disappear:

$$\theta(0) = 0, \theta(1) = 0. \tag{7}$$

Heat removal through the boundaries of arbitrary thermal conductivity (version 3) is represented by the conditions

$$\theta'(0) = Bi_1\,\theta(0), \theta'(1) = -Bi_2\,\theta(1) \tag{8}$$

where the dimensionless Biot number $Bi_i = \alpha_i d/\kappa$ characterizes heat transfer at the boundaries.

The solution of this boundary-value problem allows the determination of the critical values of the parameters and forms of disturbances at which the convective instability of the stationary heat transfer originates and develops in the layer.

3. STEADY-STATE HEAT TRANSFER REGIMES

As mentioned earlier, steady-state heat transfer in a horizontal layer of a reacting liquid is possible when the temperature depends only on the vertical coordinate, and the distributions $\Theta_0(z)$ are the solutions of equation (2) or (3) with the corresponding boundary conditions. For the versions of the problem considered these conditions have the form:

(1) for the layer with boundaries of identical temperature (versions 1 and 4):

$$\Theta_0(0) = 0, \Theta_0(1) = 0; \quad (9)$$

(2) for the layer with boundaries of different temperatures (version 2) two cases are investigated:

(a) heating from below $\Theta_0(0) = 0$, $\Theta_0(1) = -\tau$

and

(b) heating from above $\Theta_0(0) = -\tau$, $\Theta_0(1) = 0$

$$(10)$$

where $\tau = |T_0 - T_1|E/RT_0^2$ is the dimensionless difference of the temperatures of the boundaries;

(3) for the layer with the final thermal conductivity of the boundaries (version 3):

$$\Theta_0'(0) = Bi_1\,\Theta_0(0), \Theta_0'(1) = -Bi_2\,\Theta_0(1). \quad (11)$$

First, the base problem will be considered in which the reactant is at rest under steady-state heat transfer conditions, and the boundaries of the layer have identical temperatures (version 1). In this case, the boundary-value problem (2), (9) is reduced to the classical problem of thermal explosion.

It is known [2, 4] that this boundary-value problem has solutions in a certain range of the values of the Frank-Kamenetskii parameter, $0 \leqslant Fk \leqslant Fk_{CR}$. For a plane layer at $\beta = 0$, the parameter $Fk_{CR} = 3.514$ increases little with increasing β [4], which does not exceed 0.1 for actual processes. A detailed analysis of the effect of β on the thermal explosion threshold is contained in ref. [9]; the most precise calculation of the value of Fk_{CR} was made in ref. [10]. When $Fk < Fk_{CR}$, the problem has two solutions, low-temperature and high-temperature solutions, with the latter always being unstable with respect to plane-parallel temperature perturbations. An example of the resulting equilibrium distributions $\Theta_0(z)$ being symmetric about the middle of the layer where there is a maximum value Θ_{0m} is given in Fig. 1(a) as a dashed line. The corresponding function $\Theta_{0m}(Fk)$ is depicted as curve 1 in Fig. 2.

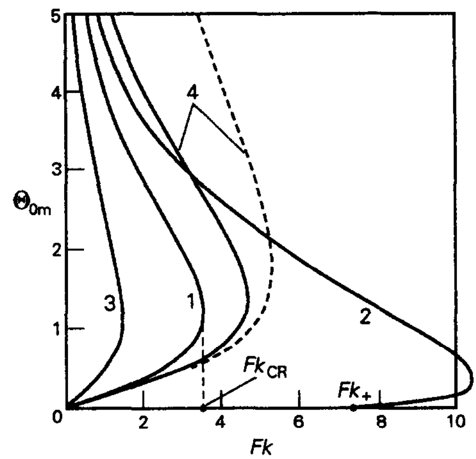

FIG. 2. The maximum temperature Θ_{0m} as a function of Fk. (1) For boundaries with identical temperatures. (2) For boundaries of different temperatures at $\tau = 2.5$. (3) For boundaries of finite thermal conductivity at $Bi_1 = 1$ and $Bi_2 = \infty$. (4) For the layer with transverse pumping of the reactant at $Pe = 4$. Solid lines are constructed at $\beta = 0$; dashed lines at $\beta = 0.1$.

Now, the influence of the complicating factors on the form of the stationary solutions and the thermal expansion threshold will be considered.

The effect of the transverse temperature gradient on the conditions for the occurrence of thermal explosion was first analysed in ref. [11]. The cooling of one of the layer walls (it is natural here to take the initial temperature reading from the hot wall, T_0) leads to an increase in heat removal from the reaction zone and, consequently, to the rise of the thermal explosion threshold. As a result of the analytical solution of equation (2) for the threshold values of Fk, the following relation was obtained in ref. [11]:

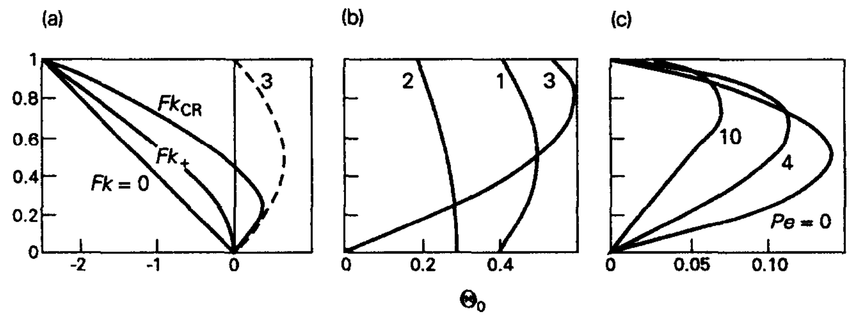

FIG. 1. Stationary distributions of temperature $\Theta_0(z)$ at $\beta = 0$. (a) In the layer heated from below at $\tau = 2.5$ and different values of Fk (solid lines) and at $\tau = 0$, $Fk = 3$ (dashed line). (b) In the layer with boundaries of different thermal conductivities at $Bi = 1$ for three versions of the ratios of the thermal conductivities of the boundaries at (1) $Fk = 0.5$; (2) $Fk = 0.125$; (3) $Fk = 1.25$. (c) In the layer with transverse motion of a reactant at different values of Pe.

$$Fk = \frac{2}{a}(\text{arcosh}\sqrt{(a\exp\tau)} + \text{arcosh}\sqrt{a})^2. \quad (12)$$

At a fixed value of τ, the critical condition for the thermal explosion Fk_{CR} is governed by the maximum of the right-hand side of equation (12) as a function of the integration constant a.

The numerical solution of the boundary-value problem (2), (10) by the Runge–Kutta technique made in ref. [12] shows, according to equation (12), that the transverse gradient drastically increases the explosion threshold. This dependence is shown in Fig. 3 and, naturally, is common for boundary conditions (10a) and (10c). When $\tau \neq 0$ and $Fk < Fk_{CR}$, the problem has two solutions (just as in the case of isothermal boundaries ($\tau = 0$), the upper of which is unstable with respect to temperature disturbances. The dependence of the maximum temperature Θ_{0m} on the Frank-Kamenetskii parameter for a fixed value of τ is presented in Fig. 2 (curve 2). At values of Fk smaller than a certain value of Fk_+ ($Fk_+ < Fk_{CR}$), the temperature of the hot wall is maximal for the lower equilibrium regime in the layer; when $Fk_+ < Fk < Fk_{CR}$ the maximum temperature is displaced into the layer resulting in a substantial change in liquid density stratification. The value of Fk_+ is determined from the condition of the derivative $d\Theta_0/dz$ being equal to zero on the hot boundary. The corresponding function $Fk_+(\tau)$ is shown in Fig. 3.

Figure 1(a) depicts the low-temperature equilibrium distributions $\Theta_0(z)$ for the layer heated from below at constant differences of temperatures of the boundaries and different values of Fk. Similar temperature profiles for heating from above are obtained by means of replacement of z by $1-z$.

Strictly speaking, the assumption about the constant temperature at the interface between the heat generating substance and the walls of the layer (infinite thermal conductivity of the boundaries) is not realizable for liquid reactants, the heat capacity of which is often comparable with that of solid bodies. The boundary-value problem (2), (11) corresponds to the condition of heat transfer of general type. According to ref. [13], the qualitative picture of the solution—the existence of two equilibrium regimes and their stability at the subexplosive values of the parameter Fk—is similar to that already described. The form of the low-temperature equilibrium distributions $\Theta_0(z)$ is displayed in Fig. 1(c) for three most interesting specific cases:

(1) Both boundaries have the same thermal conductivity, i.e. $Bi_1 = Bi_2 = Bi$.

(2) One of the boundaries (for example, the lower) is thermally insulated, the other has an arbitrary thermal conductivity: $Bi_1 = 0$, $Bi_2 = Bi$.

(3) One of the boundaries (the upper) has an infinite thermal conductivity, while the thermal conductivity of the other is arbitrary: $Bi_1 = Bi$, $Bi_2 = \infty$. An example of the function $\Theta_{0m}(Fk)$ for the latter case is given in Fig. 2 (curve 3).

The equilibrium heat and mass transfer regimes in such a system exist at values of Fk smaller than $Fk_{CR}(Bi_{1,2})$, some values of which are presented in Table 1 according to the data of ref. [13]. The interchange of the upper and lower boundaries certainly does not influence the values of $Fk_{CR}(Bi)$.

The steady-state heat transfer in the case of uniform transverse motion of the reactant (the boundary-value problem (3), (9)) depends greatly on the intensity of this motion. According to ref. [14], at $Pe \neq 0$, just as in the aforementioned versions of the problem of the reacting liquid's mechanical equilibrium, two heat-conducting regimes are possible in the layer for $Fk < Fk_{CR}$, and of these only the low-temperature regime is stable. As the intensity of injection increases, the symmetry of the stationary temperature profiles is violated, and the zone of greatest heating displaces to the boundary $z = 1$ (see Fig. 1(c)). Figure 2 (curve 4) shows the functions $\Theta_{0m}(Fk)$ at a fixed value of Pe and two values of β. It is seen that the value of β should be taken into account only for the high-temperature (unstable) regime.

The dependence of the critical value of the Frank-Kamenetskii parameter on Peclet number is given in Fig. 3. It follows from the plot that the injection substantially increases the thermal explosion threshold. The function $Fk_{CR}(Pe)$ at $Pe < 4$ and $\beta = 0$ can be approximated accurate to 3% by the polynomial

$$Fk_{CR} = 3.514 + 0.0485Pe^2 + 0.0015Pe^4.$$

For large values of Pe, the function $Fk_{CR}(Pe)$ is linear.

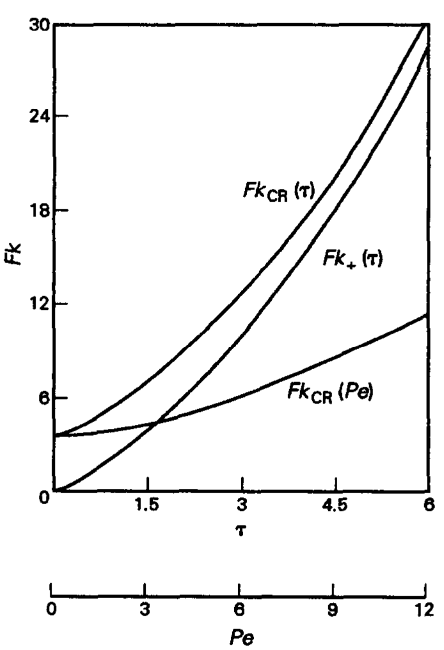

FIG. 3. The functions $Fk_{CR}(\tau)$ and $Fk_+(\tau)$ for the heated layer and $Fk_{CR}(Pe)$ for the layer with transverse motion of a reactant at $\beta = 0$.

Table 1. Dependence of the critical value of the Frank-Kamenetskii parameter Fk_{CR} on Bi for three versions of the thermal conductivity ratios of boundaries

	$Bi_1 = Bi_2 = Bi$	$Bi_1 = 0, Bi_2 = Bi$	$Bi_1 = Bi, Bi_2 = \infty$
Bi	Fk_{CR}	Fk_{CR}	Fk_{CR}
0	0.00000	0.00000	0.8785
0.05	0.03648	0.01810	0.9144
0.10	0.07237	0.03559	0.9496
0.20	0.1424	0.06890	1.018
0.50	0.3390	0.1567	1.208
1	0.6269	0.2707	1.479
2	1.083	0.4209	1.888
5	1.889	0.6194	2.513
10	2.478	0.7290	2.931
∞	3.514	0.8785	3.514

The given function $Fk_{CR}(Pe)$ is also preserved in the case of the reversal of injection ($Pe < 0$). Here, the temperature profiles are obtained by the replacement of z by $1-z$. However, the distributions of the reactant density are very different in these cases.

In connection with the analysis of the mechanisms underlying the effect on the explosion threshold, mention should be made of ref. [15], in which the problem of thermal explosion was investigated for the general case when a reactant was injected into a layer having boundaries of different temperatures, with the injection being made alternatively on both (cooler and hotter) sides of the layer.

The analysis of the existence of steady-state heat transfer in the layer of a reacting liquid and the determination of the critical conditions for thermal explosion show that the presence of the complicated factors does not lead to qualitative changes in the solution of the problem. However, these factors strongly influence the region of existence of steady-state regimes (in other words, the thermal explosion threshold) and the form of the corresponding temperature distributions that determine the liquid density stratification in the layer and thus the conditions for the incipience of free convection.

4. CONVECTIVE STABILITY OF STEADY-STATE HEAT TRANSFER REGIMES

4.1. *A layer with isothermal boundaries*

The boundary-value stability problem formulated for an incompressible fluid in which a homogeneous exothermal zero-order reaction takes place is the most general one. In cases when mechanical equilibrium of fluid is possible (i.e. at $Pe = 0$), the system of equations (5) is simplified. Its solution was obtained [4] for boundary conditions (6), (7) that correspond to solid, ideally heat-conducting walls of the layer with identical temperatures. It was assumed for the solution that the disturbances leading to the violation of equilibrium and to the development of convection were monotonous and that the limit of stability was determined by the condition $\lambda = 0$. As a result, a set of neutral curves $Ra(k)$ were constructed at different values of the parameters Fk and β for a low-temperature equilibrium solution stable in a motionless reactant. The calculated data are generalized in the form of the dependence of the minimum critical Rayleigh number Ra_* on the Frank-Kamenetskii parameter (hereafter this relationship will often be referred to for comparison).

It is shown in ref. [4] that at small values of Fk the parameter β has virtually no influence on the limit of convective stability and at $Fk \approx Fk_{CR}$ even the value $\beta = 0.1$ increases this limit by no more than 5–7%. In the majority of further calculations this allows an assumption that $\beta = 0$ [2].

The assertion about the monotonous character of critical disturbances [4] is not obvious and requires analysis. In general, the decrements of normal disturbances, which are the eigenvalues of problem (5)–(7), are complex, and the limit of stability is determined by the condition $\lambda_r = 0$; the values $\lambda_r > 0$ correspond to stable conditions, $\lambda_r < 0$ to unstable conditions. At $Ra = 0$ and $Fk = 0$, equation (5) becomes independent, and the boundary-value problem becomes a self-conjugated one signifying the monotonous character of disturbances. Generally, one fails to prove the monotonicity of disturbances.

In order to find the eigenvalues at $Ra \neq 0$ and $Fk = 0$, the Runge–Kutta method was used [16] with automatic selection of the integration step depending on the accuracy required [17]. The equations of the boundary-value problem were presented as a system of 12 ordinary differential first-order equations for the real and imaginary parts of the amplitudes of disturbances, and three linearly independent solutions satisfying the boundary conditions at the initial point of integration $z = 0$ were constructed numerically. The requirement for the existence of the non-trivial solution to the problem that would satisfy the conditions at the end of the integration interval $z = 1$ makes it possible to obtain the characteristic relation which determines the eigenvalues of the problem.

The behaviour of disturbances at different Rayleigh numbers is characterized by the spectrum of disturbance decrements $\lambda(Ra)$. Figure 4 presents the real parts of the lower levels of the spectrum of decrements

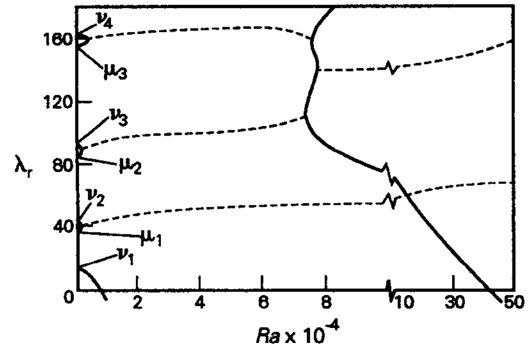

FIG. 4. Monotonous (solid lines) and vibrational (dashed lines) branches of the spectrum of the real parts of decrements at $Fk = 2.8$, $k = 3$, $Pr = 1$ and $\beta = 0$.

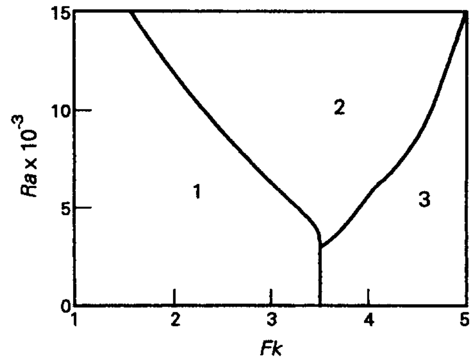

FIG. 5. The chart of the regimes on the plane (Fk, Ra) for $Pr = 1$, $k = 3.5$ and $\beta = 0$.

of the thermal (ν_n) and hydrodynamic (μ_n) perturbations of the low-temperature equilibrium solution at fixed values of Fk, k and β, and at $Pr = 1$ (reacting gases) as functions of the Rayleigh number. It follows from the plot that within the range of Ra variation of the Ra values there are two levels of the spectrum that lead to monotonous instability. The critical value of the most interesting first level of instability coincides with that obtained in ref. [4]. The spectrum of the decrements shows that in the system investigated vibrational regimes are possible with $\lambda_i \neq 0$ (dashed curves in Fig. 4) which, however, do not lead to instability. The spectra for Fk, k and Pr have similar forms. The eigenvalues of the problem are very sensitive to changes in the Prandtl number, whereas the critical values of Ra of monotonous instability do not depend on Pr.

The calculations show that though oscillating disturbances are possible in the system, vibrational instability is absent in the region of parameters studied [16] (the value of the Prandtl number varied from 1 to 20, the Frank-Kamenetskii parameter from 1 to 3.51 and the wave number from 1 to 4), and it is monotonous disturbances that lead to the crisis of equilibrium. This result confirms the basic assumption made in ref. [4].

The analysis of the spectrum of decrements for the high-temperature equilibrium solution, which is unstable with respect to thermal disturbances, shows that the allowance for hydrodynamic disturbances does not lead to stabilization, and this regime remains unstable at all Rayleigh numbers.

The solution of the full non-linear convection equations was obtained in ref. [7] with allowance for the results of linear investigation of stability for the statement of the problem under consideration. Figure 5 shows the chart of the regimes on the plane (Fk, Ra). In region 1 the reacting liquid is in mechanical equilibrium corresponding to the low-temperature regime. In region 2, convective motion is developing. The boundary between regions 1 and 2 practically coincides with that determined by linear theory. In region 3, a thermal explosion takes place. Of interest is the fact that the convective motion, which creates an additional transverse heat transfer, considerably shifts the thermal explosion threshold to the side of the higher values of the Frank-Kamenetskii parameter (the boundary between regions 2 and 3).

4.2. *The layer with the transverse temperature gradient*

Now, the problem will be considered dealing with the convective stability of the reactant equilibrium in the layer whose boundaries have different temperatures (boundary-value problem (5)–(7), equilibrium problem (2), (10)).

When $\tau = 0$, the wall temperatures of the layer are the same, and the problem of stability is reduced to that considered in Section 4.1. In the other limiting case when $Fk = 0$ ($\tau \neq 0$), the internal heat generation is absent and equations (5) with boundary conditions (6), (7) describe the well known Rayleigh problem [8].

The solution of this boundary-value problem in its complete formulation was made in ref. [12] with the help of the above-described numerical procedure. Just as in the two limiting situations, $\tau = 0$, $Fk \neq 0$ and $\tau \neq 0$, $Fk = 0$, the monotonous disturbances with $\lambda_i = 0$ turn out to be responsible for the lower level of instability, i.e. the neutral disturbances are determined by the condition $\lambda = 0$. The results of the numerical solution are reported below. First, consider the version when the lower boundary of the layer is heated, and the equilibrium temperature distributions are described by boundary-value problem (2), (10a).

The boundary of the convective stability $Ra(k)$ is presented in Fig. 6 for a fixed difference of temperatures and different values of the Frank-Kamenetskii parameter (the regions of instability are located above the curves). The curve $Fk = 0$ is the solution of the Rayleigh problem. As already noted, the augmentation of the exothermal chemical reaction (the increase in Fk) leads to a noticeable change in liquid density stratification. Thus, when $Fk > Fk_+$, the rise of the values of the Frank-Kamenetskii parameter is

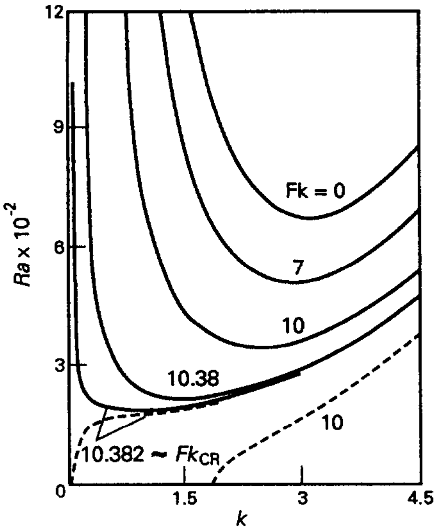

FIG. 6. The neutral curves $Ra(k)$ at $\beta = 2.5$ and different values of Fk for the low-temperature (solid lines) and high-temperature (dashed lines) stationary solutions.

accompanied by a decrease in the thickness of the region with the potentially unstable distribution of the density in the upper portion of the layer. As the power of internal heat generation increases, the convective stability of the system is lowered; at $Fk = Fk_{CR}$, the value of $Ra_* \equiv \min Ra(k)$ is finite. When $Fk < Fk_+$, the transverse temperature gradient is directed downward across the entire layer, whereas the liquid density stratification differs weakly from the Rayleigh stratification. This explains the slight decrease in stability observed at the initial stage of the rise in the value of Fk. When $Fk < Fk_+$ the wave numbers k_* that correspond to Ra_* and determine the dimensions of the critical convective cells are also close to the value $k \approx 3.1$ which is characteristic of the Rayleigh problem. As Fk increases in the region with $Fk > Fk_+$, the contribution of the internal heat generation into the system instability becomes substantial and leads to a sharp decrease in the value of Ra_*.

The behaviour of k_* in this region of variation of Fk is of interest. For values of Fk close to $Fk_{CR}(\tau)$, the length of the waves of critical disturbances increases sharply (correspondingly, the values of k_* decrease). This result seems to be associated with the specific nature of heat generation in reacting systems. Due to the non-linear dependence of the chemical reaction rate on the temperature, the problem of stability of the steady-state heat-conducting regimes in a quiescent liquid occurs. The stability of solutions at $Ra = 0$ (in this case the motion equation in system (5) has a trivial solution) was investigated in ref. [14], where it was shown that the most dangerous disturbances were those with $k = 0$. This distinction of the plane-parallel disturbances in reacting systems also influences the picture of stability in the general case when $Ra \neq 0$.

The dashed lines in Fig. 6 represent the neutral curves relating to the high-temperature equilibrium regime. In this case the distributions $\Theta_0(z)$ are similar to those shown in Fig. 1(a) for Fk_{CR}. Here, to the stable state of the liquid there correspond regions under the $Ra(k)$ curves. Just as in the previous statement of the problem [4, 16], the neutral curves become greatly distorted in transition to the high-temperature equilibrium solution, and in the zone of long-wave disturbances there appears a section of absolute convective instability (ensuing at $Ra = 0$), naturally due to the action of the mechanism of thermal instability in a quiescent liquid. With a decrease in Fk, which is accompanied by the rise of Θ_{0m} in this regime, the region of absolute instability expands.

The results of investigations can be conveniently presented as the dependence of the minimum critical Rayleigh number Ra_* on the parameters of the problem. The function $Ra_*(Fk)$ for various values of τ is given in Fig. 7. In the absence of heating ($\tau = 0$), the density inhomogeneities disappear in the layer with a decrease in Fk, and the values of Ra_* tend to infinity. As already noted, at $\tau \neq 0$ and $Fk = 0$, the boundary-value problem is reduced to the Rayleigh problem, the main features of which are also preserved at small values of the Frank-Kamenetskii parameter. The increase in internal heat generation leads to a decrease in stability, but the values of $Ra_*(Fk)$ are finite at all values of τ. All the curves $Ra_*(Fk)$ have end points, due to the fact that in the case of $Fk > Fk_{CR}$ the equilibrium regimes are absent and the problem of their stability does not occur.

Now, the problem will be elucidated regarding the possibility for the setting-up of convection when the layer is heated from above. With this variant of the boundary conditions the equilibrium distributions of temperature are found from the boundary-value prob-

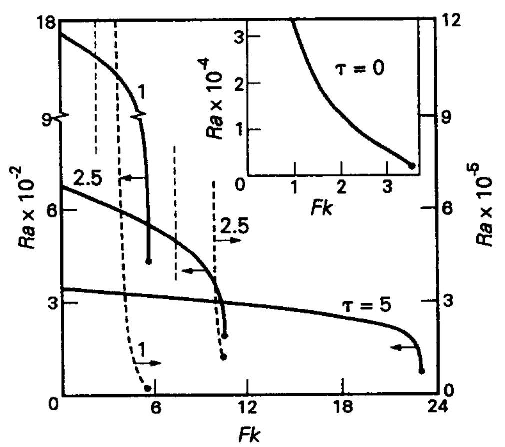

FIG. 7. The minimum critical value of the Rayleigh number Ra_* as a function of Fk for different values of τ with heating of the layer from below (solid lines) and from above (dashed lines).

lem (2), (10c). If there is no internal heat generation in the layer ($Fk = 0$), the transverse temperature gradient directed upward produces a stable stratification of density, and convective motion of the reactant is impossible. The stabilizing effect of the transverse temperature gradient is also preserved in the presence of the exothermal chemical process up to $Fk = Fk_+$. When $Fk > Fk_+$, the maximum temperature is shifted toward the inside of the layer and a narrow region of the reactant with potentially unstable density distribution develops near the upper boundary. In contrast to the previous case, for the boundary conditions considered, two mechanisms of liquid stratification compensate one another.

In Fig. 7 the functions $Ra_*(Fk)$ are presented for two values of τ. Just as in the case of heating from below, at $Fk = Fk_{CR}$ these relations have end points. As Fk decreases, the values of Ra_* increases rapidly, and the functions $Ra_*(Fk)$ approach the asymptotes shown in Fig. 7 by vertical dashed lines. The location of the asymptotes is dictated by the values of $Fk_+(\tau)$ found from the solution of the equilibrium problem; when $Fk < Fk_+$, the equilibrium state of the reactant is absolutely stable.

The results presented in Fig. 7 show that while the transverse temperature gradient greatly expands the region of existence of equilibrium regimes of heat transfer, it considerably decreases (with heating from below) and increases (with heating from above) their convective stability as compared with the case of $\tau = 0$ [4], also depicted in Fig. 7.

4.3. *A layer with boundaries of arbitrary thermal conductivity*

Now, the effect of the finite thermal conductivity of the boundaries of a horizontal reactant layer on the convective stability will be considered on the example of the same three particular cases of the relationship between the thermal conductivities of the upper and lower surfaces, which were discussed above in connection with the problem of thermal explosion (2), (11). As Bi varies from 0 to ∞, in the first case ($Bi_1 = Bi_2 = Bi$) Fk_{CR} varies within the range from 0 to 3.514, in the second case ($Bi_1 = 0$, $Bi_2 = Bi$) from 0 to 0.8785, and in the third case ($Bi_1 = Bi$, $Bi_2 = \infty$) from 0.8785 to 3.514. The third case at $Bi = 0$ coincides with the second case at $Bi = \infty$, and at $Bi = \infty$ the third and the first cases are equivalent.

The numerical solution of the boundary-value problem (5), (6), (8) determines the limits of convective stability of such steady-state regimes of heat transfer (it is understood here that $Pe = 0$). For the low-temperature steady-state regime, which is stable in a quiescent medium, Fig. 8 presents a family of relationships between the minimal critical values of the Rayleigh number Ra_* and the Frank-Kamenetskii parameter, which corresponds to the three considered versions of the relationships between Bi_1 and Bi_2 and to different values of the Biot number Bi. At $Fk = Fk_{CR}$ all the curves $Ra_*(Fk)$ have final points

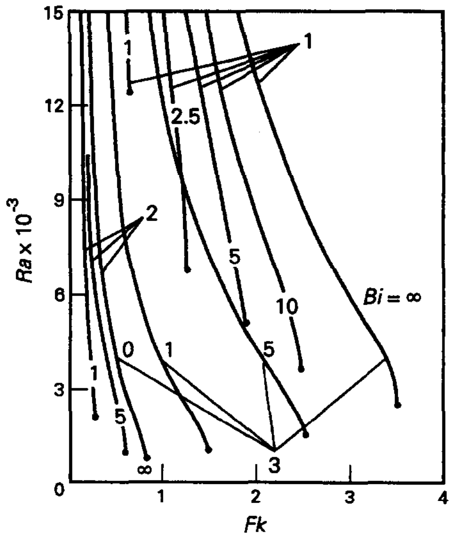

FIG. 8. The minimum critical value of the Rayleigh number Ra_* as a function of Fk at different values of Bi for three versions of thermal conductivity ratios.

due to the absence of equilibrium heat transfer regimes for $Fk > Fk_{CR}(Bi)$. The region of convective instability of the system is located above the curves $Ra_*(Fk)$.

The decrease in the values of the parameter Fk corresponds to weaker internal heat generation which is responsible for reactant density stratification. This leads to a sharp increase in the convective stability of the reactant, and when $Fk \to 0$ (absence of reaction), $Ra_* \to \infty$. For the relationship between the thermal conductivities of boundaries 1.3 at $Bi = \infty$, the problem is reduced to the well known investigation with perfectly conducting boundaries, and here the results of calculation coincide with those obtained in ref. [4].

The behaviour of the critical wave number k_* (corresponding to Ra_*) with increasing Fk is typical for such problems. At any values of Bi in all of the considered versions of heat conducting properties of the boundaries the length of the waves of critical disturbances increases for values of Fk close to $Fk_{CR}(Bi)$, i.e. k_* decreases from the characteristic values $k_* \sim 3.5$ to $k_* \sim 1$.

Note that in contrast to the problem of thermal explosion, the reversal of the boundaries in the study of convective stability is reduced to the equivalent statement only for the first particular case of the relationship between Bi_1 and Bi_2; for the second case this reversal leads to a stably stratified state of the reactant in which the convection does not originate; for the third case it leads to a separate problem of stability.

Allowance for the finite conductivity of the layer boundaries alters very significantly both the explosion threshold and the convective stability of the reactant. The determination of the conditions for the origination of convection in the general case of the arbitrary values of Bi_1 and Bi_2 does not lead to additional computational difficulties; the complexity here consists of the representation of the results for such a multi-parametric problem.

4.4. *A layer with transverse pumping of a reactant*

Now, the effect of the transverse motion of the reactant on the convective stability of the system will be considered. It is assumed that the layer boundaries are maintained at a constant temperature T_0. The reactant is uniformly injected at a velocity v_0 through the lower boundary and is sucked at the same velocity through the upper boundary. This leads to the existence of a non-disturbed cross-section in the layer with a uniform vertical velocity v_0. The infinite steady-state heat transfer in such a system was considered in Section 3.

In a moving reacting medium, the stability of the low-temperature steady-state regime of convection may be violated due to the setting-in of convection.

The behaviour of disturbances in such a medium is described by boundary-value problem (5)–(7). The equilibrium temperature distributions $\Theta_0(Pe, Fk, Z)$ are the solutions of equilibrium problem (3), (9), and they exist in the region $Fk \leqslant Fk_{CR}(Pe)$.

At $Ra = 0$ (no buoyancy force), boundary-value problem (5)–(7) is reduced to the problem of the stability of non-convective heat transfer processes against temperature disturbances in the reactant in uniform lateral motion. When $Pe = 0$, the problem coincides with that considered in ref. [4].

The calculations carried out in ref. [18] show that the spectra of the decrements $\lambda_r(Ra)$ have a very complex structure, resembling that depicted in Fig. 4 for the case $Pe = 0$. However, just as in the cases already considered, the responsibility for convective instability rests with monotonous disturbances. The presence of blowing leads to the appearance of the dependence of the monotonous instability limit ($\lambda = 0$) on Prandtl number. Note that, in contrast to the corresponding problem of thermal explosion [14], the picture of convective instability in the presence of blowing is not invariant with respect to the reversal of the transverse motion of the reactant.

The increase in the intensity of blowing at a fixed value of the Frank-Kamenetskii parameter for the lower regime decreases the heating of the liquid due to internal heat generation and narrows the region of unstable stratification of density near the upper boundary of the layer (Fig. 1(c)). These circumstances lead to a considerable increase in the convective stability of the medium with an increasing parameter Pe and to the displacement of the critical wave numbers to the side of short-wave disturbances.

The results of the solution of the stability problem are presented in Fig. 9, representing the dependence of the minimum critical Rayleigh number Ra_* of the basic instability level on the remaining parameters.

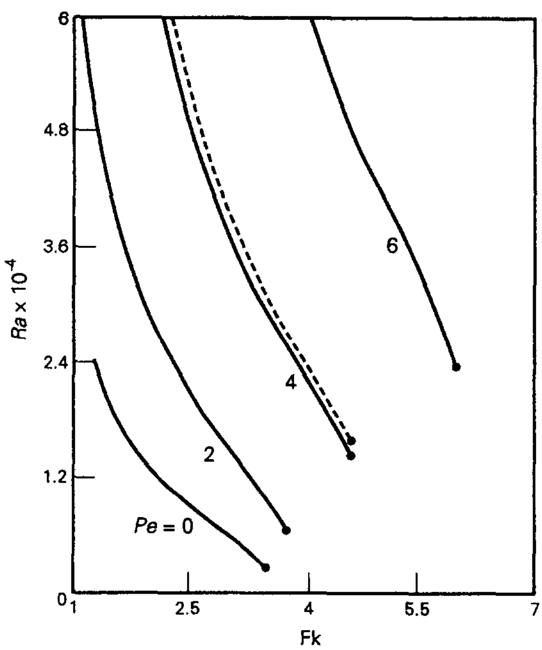

FIG. 9. The minimum critical Rayleigh number Ra_* as a function of Fk at different values of Pe for $Pr = 1$ (solid lines) and $Pr = 20$ (dashed line).

Figure 9 presents the function $Ra_*(Fk)$ at a fixed value of Pr and different values of Pe. At $Fk = Fk_{CR}$ the curves $Ra_*(Fk)$ have end points, the locations of which are determined by the Peclet number. The enhancement of blowing exerts a stabilizing effect. With an increase in the Frank-Kamenetskii parameter, the power of chemical internal heat sources increases as well as the warming of the reactant, and this leads to a decrease in convective stability. The increase in the values of the Prandtl number for liquid reactants (for liquid explosives $Pr \sim 20$) is accompanied by a weak increase in the threshold of the setting-up of convection. The function $Ra_*(Fk)$ for $Pr = 20$ is represented by a dashed line.

The critical values of the Rayleigh number Ra_* rise monotonically with the reactant rate; at all values of the Frank-Kamenetskii parameter the increase in convective stability turns out to be very notable. Thus, in a 1 cm thick layer of reactant with properties close to those of water, the transverse motion at a velocity of 0.005 cm s^{-1} increases the stability four times.

The dependence of the convective stability boundary on the Prandtl number has a rather complex form and was analysed in detail earlier [18]. According to the results obtained, as Pr increases, the stability of the system rises slightly in the region $Pr > 1$, and the curves $Ra_*(Pr)$ approach the asymptotes corresponding to $Pr = \infty$. At $Pr = \infty$, the values of Ra_* exceed Ra_* at $Pr = 1$ by only 4–5%. For the majority of chemically active liquids high values of Pr

are typical, and therefore in the solution of boundary-value problem (5)–(7) in the equation of motion, the terms that describe the pumping of the reactant can be neglected. In this asymptotic case the dependence of Ra_* on Pr disappears.

At $Pr \sim 1$, the functions $Ra_*(Pr)$ have minima. When $Pr < 1$ (reacting gases), a sharp increase in the stability threshold is observed with decreasing Pr, and as $Pr \to 0$, at all the values of the parameters, $Ra_* \to \infty$ [18].

The aforegoing results indicate that transverse blowing of the reactant, at the temperature of the layer boundaries, makes it possible to substantially increase the convective stability of the system.

5. CONCLUSIONS

The following basic results were obtained in the study of free convection in an infinite horizontal layer of a moving incompressible reactant in the presence of a homogeneous exothermal zero-order reactio

(1) The complicating factors, such as heating o 1085
layer from below or from above, different thermal conductivities of the layer boundaries and unif transverse motion of the reactant, substant 1085
change heat removal from the reaction zone and the density stratification of the chemically active liquid.

(2) The study of the specific features of heat transfer in such a system is reduced to the generalization of the classical problem of thermal explosion. The factors listed above do not vary the well known qualitative picture of the solution: when $Fk < Fk_{CR}$, two steady-state heat transfer regimes are possible in the layer: the low- and high-temperature regimes; when $Fk > Fk_{CR}$, thermal explosion takes place. The high-temperature regime always turns out to be unstable with respect to plane-parallel disturbances.

(3) The influence of the complicating factors listed above in the equilibrium statement of the problem shows up in the determination of the dependence of Fk_{CR} on the parameters of the problem. The functions $Fk_{CR}(\tau)$, $Fk_{CR}(Bi)$ and $Fk_{CR}(Pe)$ obtained indicate the possibility for a substantial increase and decrease in the explosive threshold as compared with the well known value for the layer with perfectly heat-conducting boundaries.

(4) Even though vibrational regimes are possible in the system, in all cases the monotonous disturbances were responsible for the incipience of convective motion.

(5) The dependencies of the minimum critical Rayleigh number on the parameters of the problem, $Ra_*(\tau)$, $Ra_*(Bi)$ and $Ra_*(Pe)$, characterize a very strong influence of the mechanisms studied on the critical conditions for the incipience of convection. In the limiting cases, correspondence is established between the results obtained and the solutions of the Rayleigh [8] and Jones [4] problems.

Thus, three effective mechanisms for the control of the threshold phenomena of thermal explosion and of convective instability in reactive media have been analysed. The aforegoing investigation of the stability is limited by the possibilities of the linear theory which, in particular, does not allow one to describe the effect of developed convection on the critical conditions of thermal explosion.

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STABILITE CONVECTIVE D'UNE COUCHE LIQUIDE HORIZONTALE REACTIVE EN PRESENCE DE DIFFERENTS FACTEURS COMPLICATIFS

Résumé—La stabilité convective d'une couche liquide horizontale et chimiquement active est étudiée quand une réaction exothermique d'ordre zéro prend place en présence de facteurs complicatifs : chauffage de la couche, conductivités thermiques différentes des frontières et mouvement transversal du réactant. Utilisant la méthode de Runge–Kutta, une étude numérique du problème de stabilité linéaire spectrale des régimes de transfert thermique est conduite en résolvant le problème non linéaire généralisé de l'explosion thermique. Les résultats obtenus montrent une influence notable des facteurs complicatifs (précédemment désignés) sur les conditions critiques de l'explosion thermique et sur la limite de la naissance du mouvement convectif du réactant.

KONVEKTIVE STABILITÄT EINER HORIZONTALEN REAGIERENDEN FLÜSSIGKEITSSCHICHT IN ANWESENHEIT VERSCHIEDENER STÖRENDER EINFLÜSSE

Zusammenfassung—Die konvektive Stabilität einer horizontalen, chemisch aktiven Flüssigkeitsschicht wird untersucht. In der Schicht findet eine exotherme Reaktion nullter Ordnung statt, wobei eine Anzahl störender Einflüsse vorhanden ist: Aufwärmung der Schicht, unterschiedliche Wärmeleitfähigkeiten der Berandungen und Querströmung im Reaktanden. Unter Verwendung des Runge–Kutta Verfahrens wird das lineare spektrale Stabilitätsproblem der stationären Wärmeübertragung numerisch untersucht. Dies erfolgt durch Lösen des verallgemeinerten nichtlinearen Problems der thermischen Explosion. Die Ergebnisse zeigen einen wichtigen Einfluß der oben genannten störenden Faktoren auf die kritischen Bedingungen für eine thermische Explosion und auf die Grenze für das Einsetzen einer Konvektionsströmung im Reaktanden.

КОНВЕКТИВНАЯ УСТОЙЧИВОСТЬ ГОРИЗОНТАЛЬНОГО СЛОЯ РЕАГИРУЮЩЕЙ ЖИДКОСТИ ПРИ НАЛИЧИИ РАЗЛИЧНЫХ ОСЛОЖНЯЮЩИХ ФАКТОРОВ

Аннотация—Изучается конвективная устойчивость горизонтального слоя химически активной жидкости, во всем объеме которой протекает экзотермическая реакция нулевого порядка, при воздействии ряда осложняющих факторов: подогрева слоя, различной теплопроводности границ, поперечного движения реагента. Методом Рунге–Кутта численно исследуется линейная спектральная задача устойчивости стационарных режимов теплопереноса, найденных в результате решения обобщенной нелинейной задачи теплового взрыва. Полученные результаты показывают значительное влияние перечисленных осложняющих факторов на критические условия теплового взрыва и границу возникновения конвективного движения реагента.