THE DETERMINATION OF IGNITION CONDITIONS

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Zhurnal prikladnoi mekhaniki i tekhnicheskoi fiziki, No. 1, pp. 68-75, 1965

A simple method of determining the conditions for ignition, based on linearization of the resulting nonlinear equations, is presented. Good agreement is obtained between the exact conditions and the approximate conditions given by the linearization method. A physical interpretation of the new method is offered.

1. Equations of the Steady-State Theory of Thermal Explosion

In the general case, problems of the steady-state theory of thermal explosion [1-2] lead to the solution of the equation:

$$\frac{\partial^2\Theta}{\partial x^2}+\frac{\partial^2\Theta}{\partial y^2}+\frac{\partial^2\Theta}{\partial z^2}+\delta e^{\Theta}=0 \qquad (1.1)$$

$$\Theta=\frac{(T-T_0)\,E}{RT_0^2}, \qquad \delta=\frac{qL^2}{kRT_0^2}A\exp\left(-\frac{E}{RT_0}\right)$$

where Θ is the dimensionless temperature, δ is a dimensionless kinetic parameter, x, y, and z are dimensionless coordinates referred to the characteristic linear dimension L of the region D occupied by the reactant, q is the thermal effect of the reaction per unit volume, E is the activation energy, k is the thermal conductivity, T (x, y, z) is the absolute temperature at an arbitrary point in the region, T_0 is the absolute temperature of the medium surrounding the reacting system, R is the universal gas constant, and A is the coefficient of the exponential function in the expression for the reaction rate:

$$W(T_0)=A\exp\left(-\frac{E}{RT_0}\right)=\frac{1}{t_p} \qquad \left(t_p=\frac{1}{W(T_0)}\right).$$

Here t_p is the time of total combustion for a constant rate and the initial temperature.

At the boundary of the region:

$$\left(\frac{\partial\Theta}{\partial n}+\gamma\Theta\right)\Big|_{\Gamma}=0 \qquad \left(\gamma=\frac{\alpha L}{k}\right) \qquad (1.2)$$

where γ is the Biot number, α is the coefficient of heat transfer from the reacting surface to the surrounding medium, and n is the normal to the boundary of the region Γ.

In formulating Eq. (1.1) it was assumed that the relative heating of the reacting system is small $(T-T_0)/T_0 \ll 1$, and the expansion of exp (−E/RT) was used. For intense heat transfer and large values of the Biot number $\gamma \to \infty$, condition (1.2) becomes

$$\Theta|_{\Gamma}=0\,. \qquad (1.3)$$

At small values of the Biot number $\gamma \to 0$ the temperature at all points of the reacting system is almost the same, and the given problem is equivalent to the problem of autoignition formulated by N. N. Semenov [3].

At certain values of the parameter $\delta = \delta_*$ the boundary problem (1.1), (1.2) ceases to have a real solution. These values of δ_* are critical, i.e., values of δ for which thermal autoignition is achieved, and the problem of the steady-state theory of thermal explosion consists in determining these values of $\delta = \delta_*$. From the mathematical point of view, δ_* are branch points of the boundary problem (1.1), (1.2), since, according to [2, 4] and the results of the next section, the two solutions of the problem merge.

2. One-Dimensional Problems of the Steady-State Theory of Thermal Explosion

For the subsequent discussion it is necessary to solve one-dimensional problems of the steady-state theory of thermal explosion. With the aid of the Frank-Kamenetskii integral for the boundary conditions

$$\frac{d\Theta}{dx}\Big|_{x=0}=0 \quad \left(\frac{d\Theta}{dx}+\gamma\Theta\right)\Big|_{x=1}=0 \qquad (2.1)$$

it is easy to find the critical value

$$\delta_*=\frac{2s_*^2}{\mathrm{ch}^2\,s_*}\exp\left(-\frac{2}{\gamma}s_*\,\mathrm{th}\,s_*\right) \qquad (2.2)$$

$$(s = \sqrt{{}^1/_2\,\delta \exp \Theta_0}) \qquad (2.2 \text{ cont.})$$

for a plate, where s_* is the root of the equation

$$\gamma = (\gamma + 1)\, s_* \operatorname{th} s_* + s_*^2 \operatorname{sch}^2 s_* . \qquad (2.3)$$

The critical temperature profile is determined from the formula

$$\Theta_* (x) = \Theta_{0*} - 2 \ln \operatorname{ch} s_* x \qquad (2.4)$$

From formula (2.4) it is easy to find for x = 1 the wall temperature of a plane reaction vessel, and the critical pre-explosion heating

$$\Theta_{0*} = \ln (2 s_*^2 / \delta_*) . \qquad (2.5)$$

In Fig. 1 the solid line represents the relation between δ_* and $\ln \gamma$, calculated from formula (2.2), while in Fig. 2 it represents the relation between Θ_{0*} and Θ_* (1) and $\ln \gamma$. It can be seen from Fig. 2 that $\Theta_{0*} \to 1$, $\Theta_*(1) \to 1$ when $\gamma \to 0$.

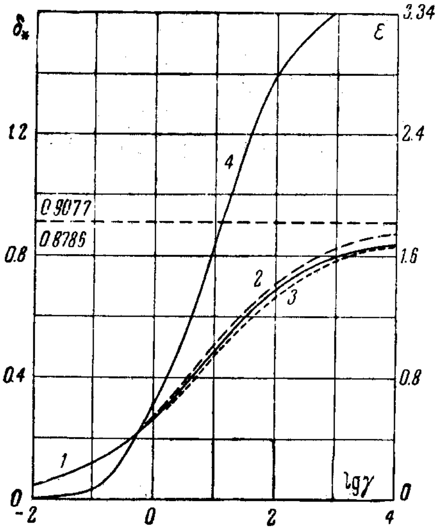

Fig. 1.

The problem of a thermal explosion with boundary conditions of the third kind has been solved in [5] for the case of an infinite cylinder. The quantity δ_* can be expressed in the form:

$$\delta_* = \frac{8 m_*}{(1 + m_*)^2} \exp \left[- \frac{4 m_*}{\gamma (1 + m_*)} \right], \qquad m_* = 2\gamma^{-1} (\sqrt{1 + {}^1/_4 \gamma^2} - 1) . \qquad (2.6)$$

The maximum temperature $\Theta_{0*} = \ln (8 m_* / \delta_*)$, and the critical temperature profile

$$\Theta_* (x) = \Theta_{0*} - 2 \ln (1 + m_* x^2) . \qquad (2.7)$$

In Fig. 3 the solid line 1 represents the relation between δ_* and $\ln \gamma$, and Fig. 4 gives the graph of Θ_{0*} and $\Theta_*(1)$ as a function of $\ln \gamma$. Hence it can be seen that when $\gamma \to 0$ both quantities tend to 1.

For a spherical vessel we have the equation

$$\frac{d^2 \Theta}{dx^2} + \frac{2}{x} \frac{d \Theta}{dx} + \delta e^{\Theta} = 0 . \qquad (2.8)$$

By means of the substitution

$$\xi = x \sqrt{{}^1/_2 \exp \Theta_0}, \quad \varphi = \Theta - \Theta_0$$

we have

$$\frac{d^2 \varphi}{d \xi^2} + \frac{2}{\xi} \frac{d \varphi}{d \xi} + 2 e^{\varphi} = 0 . \qquad (2.9)$$

According to [6], the solution of Eq. (2.8), satisfying the conditions of symmetry, can be written in the form:

$$\Theta = \Theta_0 - 2 \int_0^{\xi} x^{-2} \left(\int_0^{x} \xi^2 \exp [\varphi (\xi)]\, d\xi \right) dx . \qquad (2.10)$$

Subjecting Eq. (2.10) to the boundary condition (2.1) for x = 1, we have an equation for determining the value of δ at which there exists a solution of the boundary problem (1.1), (1.2) in the spherical case:

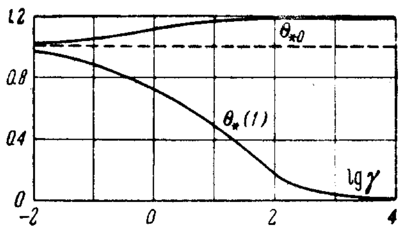

Fig. 2.

$$-\frac{2}{s} \int_0^s x^2 \exp [\varphi (x)]\, dx + \gamma \left[\Theta_0 - 2 \int_0^s \frac{1}{x^2} \left(\int_0^x \xi^2 \exp [\varphi (\xi)]\, d\xi \right) dx \right] = 0 . \qquad (2.11)$$

Differentiating Eq. (2.11) with respect to Θ_0, we have an equation for determining the value of $s = s_*$, at which the maximum value of $\delta = \delta_*$ is reached:

$$\frac{1}{s_*} (1 - \gamma) \int_0^{s_*} x^2 \exp [\varphi (x)]\, dx + \gamma - s_*^2 \exp [\varphi (s_*)] = 0 . \qquad (2.12)$$

Here and above $\varphi(\xi)$ is the solution of Eq. (2.9) for the conditions

$$\varphi'(0) = \varphi(0) = 0. \tag{2.13}$$

According to [4], with the change of variables

$$\psi = 2 + \xi d\varphi/d\xi, \qquad p = \xi^2 \exp[\varphi(\xi)]. \tag{2.14}$$

Eq. (2.9) can be reduced to the first-order equation

$$\frac{d\psi}{dp} = \frac{2(1-p)-\psi}{\psi p} \tag{2.15}$$

The graph of $\psi = \psi(p)$ is plotted qualitatively in [4]. An equation of the type (2.15) was integrated numerically by Emden [7]. The quantities z_1 and y given in [7] are related to the quantities ψ and p in the following manner:

$$\psi = -y, \qquad z_1 = \ln 2p. \tag{2.16}$$

Substituting (2.14) in (2.11), after certain transformations we have:

$$\delta = 2p \exp[(\psi - 2)/\gamma], \tag{2.17}$$

and instead of (2.12) we have the equation

$$\psi(p) = 2(p-1)/(\gamma-1). \tag{2.18}$$

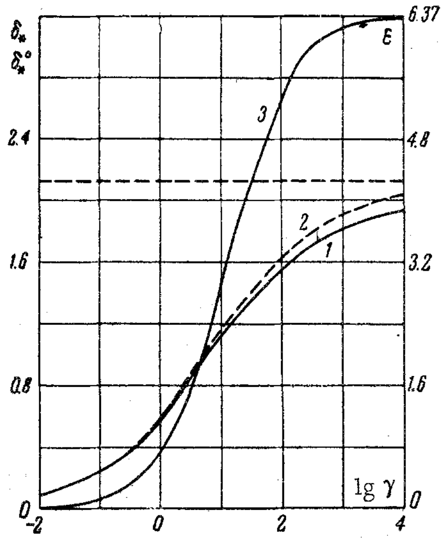

Fig. 3.

Having determined the values of ψ_* and p_* from Eq. (2.18) as the first point of intersection of the curve $\psi(p)$ and the straight line $\psi = 2(p-1)/(\gamma-1)$, we find δ_* from formula (2.17) for any corresponding value of γ. It is easy to show that when $\psi(p)$ is varied the quantity s always increases. Since, according to [4], for p = 1 equation (2.15) has a singular point of the focus type, there exist an infinite number of points of intersection of the curve $\psi(p)$ and the straight line $\psi = 2(p-1)/(\gamma-1)$ and, consequently, there exist an infinite number of values of $s = s_{i*}$, at which an extremum of $\delta = \delta_{i*}$ is reached, i.e., the curve $\delta = \delta(s)$ has an infinite number of maxima and minima which decrease in absolute value with increase in s and, asymptotically approach the value $\delta = 2\exp(-2/\gamma)$ as $s \to \infty$. The first maximum $\delta = \delta_{1*}$, attained at the minimum, compared with the rest, value of $s = s_{1*}$, is the greatest; then for $\delta > \delta_{1*}$ there is no real solution of the boundary problem (2.8), (2.1), and this value $\delta = \delta_{1*}$ should be considered critical.

For $\gamma = 0.2$ and 0.4 we found $\delta_{1*} = 0.212$ and 0.4072, respectively, with the aid of [7]. By expanding the solution of the boundary problem (2.8), (2.1) in powers of ξ we found that when $\gamma = 0$ the temperature at the wall of the vessel $\Theta_*(1)$ and Θ_{0*} tend to 1.

Note that the first two boundary problems for $\delta < \delta_*$ have two solutions, one of which has supercritical and the other subcritical pre-explosion heating; for $\delta = \delta_*$ these two solutions merge. For $\delta_{2*} < \delta \leq \delta_{1*}$, where δ_{1*} and δ_{2*} are two consecutive maxima of the curve $\delta(s)$, boundary problem (2.8), (2.1) also has two solutions, which merge when $\delta = \delta_{1*}$. Different numbers of solutions are possible for other values of δ, and when $\delta = 2\exp(-2/\gamma)$ there are an infinite number of solutions.

3. Method of Linearization

By means of the corresponding Green function it is possible to obtain, instead of the nonlinear boundary problem (1.1), (1.2), an equivalent nonlinear integral equation, whose branch point will also be $\delta = \delta_*$. For this nonlinear integral equation, by means of the method developed in [8], we can construct a linear integral equation whose eigenvalue will be $\delta = \delta_*$. Reverting from this linear integral equation to the linear boundary problem, we have

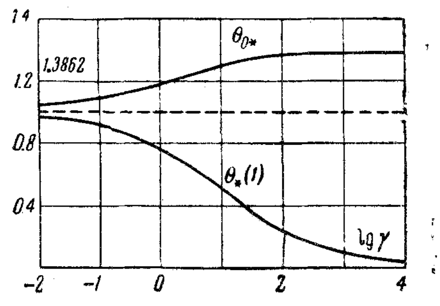

Fig. 4.

$$\Delta v + \delta e^{\Theta_*(x,y,z)} v = 0, \qquad (\partial v/\partial n + \gamma v)|_\Gamma = 0, \tag{3.1}$$

where v is the limit of the difference of the two solutions of the nonlinear boundary problem (1.1), (1.2) when $\delta \to \delta_*$.

Note that the determination of δ_* as the eigenvalue of the boundary problem (3.1) is, according to [8], necessary but not sufficient. The sufficiency of this condition can be shown easily for simple reaction vessels (plane, cylindrical, spherical).

For a plane reaction vessel, in place of equation (3.1), we have the equation:

$$\frac{d^2v}{dx^2} + \frac{2s^2}{\mathrm{ch}^2\, sx} v = 0, \quad v'(0) = 0, \quad \left(\frac{dv}{dx} + \gamma v\right)\Big|_{x=1} = 0. \tag{3.2}$$

The general solution of Eq. (3.2), according to [6], has the form:

$$v = c_1 \,\mathrm{th}\, sx + c_2 (1 - sx \,\mathrm{th}\, sx) \tag{3.3}$$

By subjecting (3.3) to boundary conditions (3.2) we obtain an equation that coincides with Eq. (2.3). The critical solution of the corresponding nonlinear problem is assumed to be known; therefore, knowing Θ_0 and determining the root s_* from Eq. (2.3), it is easy to find $\delta_* = 2s_*^2 \exp(-\Theta_{0*})$.

For the cylindrical case we have the equation

$$\frac{d^2v}{dx^2} + \frac{1}{x}\frac{dv}{dx} + \frac{8m}{(1+mx^2)^2} v = 0 . \tag{3.4}$$

The general solution of Eq. (3.4) can be found in the form

$$v = c_1 \left[\frac{2 + (1 - mx^2)\ln x}{1 + mx^2} \right] + c_2 \frac{1 - mx^2}{1 + mx^2} . \tag{3.5}$$

Here and above c_1 and c_2 are arbitrary constants. By making Eq. (3.5) satisfy boundary conditions (3.2) we obtain an equation for m_*, by solving which we find m_* in the form (2.6). Since Θ_{0*} is assumed to be known, $\delta_* = 8 m_* \cdot$ $\cdot \exp(-\Theta_{0*})$.

For a spherical reaction vessel Eq. (3.1), according to [7], has the solution:

$$v = c_1 (\xi d\varphi / d\xi + 2) \tag{3.6}$$

which satisfies the first of boundary conditions (3.2). By subjecting (3.6) to the second boundary condition (3.2), we obtain equation (2.12). Since Θ_{0*} is known, $\delta_* = 2 s_*^2 \exp(-\Theta_{0*})$.

Note that the sufficiency of determining δ_* as the eigenvalue of the boundary problem (3.4), (3.5) was previously shown in [9] for $\gamma \to \infty$.

It follows from the results of §2 that Θ_* (x, y, z) ~ 1 when $\gamma \sim 0$; therefore for small values of γ in place of Eq. (3.1) we can write the simpler equation:

$$\nabla^2 u + \delta e u = 0 . \tag{3.7}$$

Thus, for small values of γ we can determine δ_* for any shapes with the aid of the formula

$$\delta_*^\circ = \mu_1 / e \tag{3.8}$$

and assume that the error in calculating δ_* is small.* Here μ_1 is the first eigenvalue of the boundary problem (3.7), (3.1). Moreover, as examples show [9], at any rate for all the known exact analytic solutions, on the average e^{Θ_*} differs little from e = 2.718... for any value of γ, which, according to [10], guarantees fair accuracy in determining the eigenvalue of the exact linearized boundary problem of (3.1) with the aid of the first eigenvalue of the approximate linearized boundary problem (3.7), (3.1).

Thus, for a plane reaction vessel we have

$$K_1 = \int_0^1 e^{\Theta_*} dx = \frac{\exp\Theta_{0*}\,\mathrm{th}\, s_*}{s_*} \qquad \begin{pmatrix} K_1 \to e = 2.718 & \text{for } \gamma \to 0 \\ K_1 \to 2.31 & \text{for } \gamma \to \infty \end{pmatrix}. \tag{3.9}$$

Correspondingly, for the case of an infinite cylinder

$$K_2 = \int_0^1 e^{\Theta_*} dx = \frac{\exp\Theta_{0*}}{2} \left(\frac{1}{1 + m_*} + \frac{\mathrm{arc\,tg}\sqrt{m_*}}{\sqrt{m_*}} \right) \quad \text{for} \quad \gamma \to \infty \tag{3.10}$$

* Note that formula (3.8) was also obtained, from other reasoning, in [11] for $\gamma = \infty$. However, there is no discussion of the accuracy of this formula in this paper.

$$m_* = 1 \text{ and } K_2 = 2.57, \qquad K_2 \to e = 2.718\ldots \quad \text{for } \gamma \to 0 .$$

The general solution of Eq. (3.7) has the form

$$u = c_1 \cos (x \sqrt{\delta e}) + c_2 \sin (x \sqrt{\delta e}) \tag{3.11}$$

for a plane reaction vessel;

$$u = c_1 J_0 (x \sqrt{\delta e}) + c_2 V_0 (x \sqrt{\delta e}) \tag{3.12}$$

for a cylindrical reaction vessel;

$$u = c_1 x^{-1} \cos (x \sqrt{\delta e}) + c_2 x^{-1} \sin (x \sqrt{\delta e}) \tag{3.13}$$

for a spherical reaction vessel.

Here and above, c_1 and c_2 are arbitrary constants, and J_0 and V_0 are the corresponding zero-order Bessel functions of the first and second kind. By making (3.11)-(3.13) satisfy boundary conditions (3.1), we obtain the equations for determining δ_*°:

$$\gamma = \sqrt{\delta e}\ \mathrm{tg} \sqrt{\delta e} \quad \text{for a plane vessel} \tag{3.14}$$

$$\gamma J_0 (\sqrt{\delta e}) = \sqrt{\delta e} J_1 (\sqrt{\delta e}) \quad \text{for a cylindrical vessel} \tag{3.15}$$

$$\gamma = 1 - \sqrt{\delta e}\ \mathrm{ctg} \sqrt{\delta e} \quad \text{for a spherical vessel} \tag{3.16}$$

where J_1 is a first-order Bessel function of the first kind. The roots of Eqs. (3.14)-(3.16) were found by a trial method and refined by Newton 's method.

Figures 1 and 3 present plots of $\delta_*(\ln \gamma)$ (curve 1) and $\delta_*^\circ (\ln \gamma)$ (curve 2) for a plane and cylindrical vessel, respectively, together with the relative error ε of the quantity δ_*° (curves 3 and 4). Points in Fig. 1 represent the interpolation formula [5]:

$$\delta = \frac{0.88\gamma}{0.88e + \gamma} . \tag{3.17}$$

For a spherical vessel with $\gamma = 0.2$ we found $\delta_*^\circ = 0.212$, which is almost identical with the value of δ_* found earlier, but for $\gamma = 0.4$ we have $\delta_*^\circ = 0.408$, for $\gamma \to \infty$, $\delta_*^\circ = 3.63$, which exceeds the exact value of δ_* obtained in [2] by 9.3 %. As a simple interpolation formula we can use the expression [5]:

$$\delta_* = \frac{3.32\gamma}{e + \gamma} . \tag{3.18}$$

4. Interpretation of the Results

As follows from the results of § 2, when $\delta < \delta_*$ several solutions of the nonlinear boundary problem (1.1), (1.2), exist.

Let us consider the stability of the solutions of the boundary problem (1.1), (1.2). We shall assume that the solution of the steady-state problem (1.1), (1.2) differs little from the solution of the nonsteady-state equation of thermal explosion

$$\frac{\partial \vartheta}{\partial t} = \Delta \vartheta + \delta e^{v} \qquad \left(t = \frac{k}{cL^2}\tau\right) \tag{4.1}$$

with boundary conditions (1.2), i.e.,

$$v = \Theta + f(x, y, z, t) \tag{4.2}$$

where ϑ is the solution of the nonsteady-state problem; the function $f \ll 1$; t is the dimensionless time, and c is the heat capacity per unit volume. Substituting (4.2) in (4.1) and discarding small terms of the second order, we have for the perturbation of $f(x, y, z, t)$ the boundary problem

$$\frac{\partial f}{\partial t} = \Delta f + \delta e^{\Theta} f, \quad \left(\frac{\partial f}{\partial n} + \gamma f\right)\Big|_{\Gamma} = 0 . \tag{4.3}$$

We solve problem (4.3) by the method of separation of variables, putting

$$f(x, y, z, t) = v(x, y, z) \exp(-\lambda t) . \tag{4.4}$$

Substituting (4.4) in (4.3), we have

$$\Delta v + (\lambda + \delta e^{\Theta}) v = 0, \qquad \left(\frac{\partial v}{\partial n} + \gamma v\right)\Big|_{\Gamma} = 0 . \tag{4.5}$$

It follows from formula (4.4) that if for boundary problem (4.5) $\lambda_1 > 0$, where λ_1 is the first eigenvalue, then any initial temperature distribution is resolved with time; when $\lambda_1 = 0$ this is no longer so. Finally, when $\lambda_1 < 0$ there is an infinite increase in temperature with time. In the latter case the steady-state temperature distribution is unstable, i.e., autoignition of the reacting mixture takes place.

Note that if $\Theta = \Theta_*(x, y, z)$, then boundary problem (4.5) has a first eigenvalue $\lambda_1 = 0$, since in this case boundary problem (4.5) coincides with the boundary problem (3.1) investigated earlier, i.e., when $\vartheta = \Theta_*$ and $\delta = \delta_*$ the limiting condition for ignition, $\lambda_1 = 0$, is satisfied. We shall now show, without making detailed calculations, that solutions of boundary problem (1.1), (1.2), for which maximum heating is above critical, are unstable, while solutions for which maximum heating is below critical are stable. These calculations are given in [6] for $\gamma \to \infty$. For small values of γ the function $\Theta_*(x,y,z)$ tends to the Semenov temperature distribution $\Theta_*(x,y,z) \sim 1$. Then, at least for small values of γ, the equation

$$\nabla u + (\lambda + \delta e) u = 0 \tag{4.6}$$

can be investigated instead of boundary problem (4.5), with the same boundary conditions. It follows from the results of §3 that Eq. (4.6) can be used for any values of γ, since on the average $\exp \Theta_*$ differs little from e = 2.718....

Then the limiting condition for autoignition of the reacting mixture, $\lambda_1 = 0$, will be satisfied if δ is found from formula (3.8).

For $\delta > \delta_*^\circ$ we have $\lambda_1 < 0$, and any initial temperature distribution will increase with time and lead to an explosion, but for $\delta < \delta_*^\circ$ this is not so.

Using (3.8) and developing the expression for δ given at the beginning of §1, the condition for thermal explosion (3.8) can be reduced to the form:

$$t_p \leqslant e \frac{E}{RT_0} \frac{q}{cT_0} t_e . \tag{4.7}$$

The "equals" sign corresponds to the ignition boundary. Further reduction in t_p, i.e., an increase in reaction rate and heat release, always leads to thermal explosion. The quantity t_e is the thermal relaxation time during which heating decreases e = 2.718 times.

In the absence of heat release from the reaction, q = 0 and $\delta = 0$, the boundary problem (4.6), (4.5) describes the cooling of a nonreacting, initially hot body of the same shape and size, and with the same heat capacity and thermal conductivity as the reacting system, for the same boundary conditions (4.5). Then λ_1 determines the thermal relaxation time and

$$t_e = \frac{cL^2}{k\lambda_1} . \tag{4.8}$$

A physical interpretation of the condition of autoignition in the form (4.7) was previously given by one of the authors [12] for the case of thermal explosion analyzed by N. N. Semenov ($\gamma \to 0$). As may be seen from the analysis presented, this relation remains approximately valid for all cases of thermal explosion, whatever the system geometry.

It follows from the results of §3 that δ_*° is always somewhat greater than the true value δ_*. Thanks to the exponential increase in δ with temperature, this corresponds to a very small overestimate of the critical autoignition temperature T_0, by approximately 1-3°C.

The result obtained (4.7) permits a considerable simplification of the problem of finding the critical conditions of thermal autoignition for a body of arbitrary shape under mixed cooling conditions (1.2).

Moreover, in many cases the quantity t_e is more simply determined experimentally, by measuring the regular cooling regime [13] on models of a nonreacting substance with similar geometric and thermal parameters.

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