USE OF THE METHOD OF INTEGRAL RELATIONS TO SOLVE PROBLEMS IN IGNITION THEORY

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The method of integral relations is used to solve an unsteady system of equations of thermal explosion. With the help of this method several problems of ignition theory are solved.

As is known [1], the first averaged equations of thermal explosion were obtained, on the basis of physical considerations, by Todes. They were used later in [2, 3]. Averaging of the starting equations of the unsteady theory of thermal explosion was developed as a mathematical technique in [4], where two methods of averaging were given. In using the first of these it is necessary, as a preliminary, to solve the nonlinear boundary problem to determine δ_* and to make two assumptions, while in using Khudyaev's method it is necessary to solve the appropriate linear boundary value problem and to make one assumption. This paper uses the equations of the first approximation of the method of integral relations [5, 6], whose solutions in our case require only one assumption.

The unsteady system of equations of thermal explosion has the form

$$\frac{\partial \Theta}{\partial \tau} = \varphi(\eta) \exp \frac{\Theta}{1+\beta\Theta} + \frac{1}{\delta} \nabla^2 \Theta, \tag{1}$$

$$\frac{\partial \eta}{\partial \tau} = \gamma\varphi(\eta) \exp \frac{\Theta}{1+\beta\Theta}. \tag{2}$$

The system (1), (2) is solved under the following initial and boundary conditions:

$$\Theta(x, y, z, 0) = 0, \quad \eta(x, y, z, 0) = 0, \quad \Theta|_{\Gamma} = 0. \tag{3}$$

We shall assign the profile $f(x, y, z, \Theta_0)$ which satisfies the boundary conditions (3), and, as far as possible, correctly reflects the variation of $\Theta(x, y, z, \tau)$ in space. The chosen profile must include the totality of prior information about the behavior of $\Theta(x, y, z, \tau)$, which we may obtain from physical reasoning, for example, without recourse to the numerical solution of (1) and (2) with conditions (3). The quantity $\Theta_0 = \Theta_0(\tau)$ in $f(x, y, z, \Theta_0)$ has everywhere below the meaning of maximum temperature. Substituting $f(x, y, z, \Theta_0)$ into (1) and (2) in place of $\Theta(x, y, z, \tau)$, and integrating the result over the region G, we obtain a system of two nonlinear differential equations of first order to determine $\Theta_0(\tau)$ and $\bar{\eta}(\tau)$, in deriving which our sole assumption has been

$$\int_G \varphi(\eta) \exp \frac{\Theta}{1+\beta\Theta} dV = \varphi(\bar{\eta}) \int_G \exp \frac{\Theta}{1+\beta\Theta} dV. \tag{4}$$

Since as x, y, z, τ varies, η varies in the narrow range $0 \leq \eta \leq 1$, and considerably more slowly than $\exp[\Theta/(1+\beta\Theta)]$, it may be considered, according to [7], that the assumption (4) does not give a large error. The accuracy of the method may be increased if, following [5, 6], we introduce in place of the single free parameter i parameters depending on τ and integrate the result with respect to i over the subregions of region G. We then obtain 2i ordinary nonlinear differential equations of first order for the 2i free parameters and $\bar{\eta}_i = \bar{\eta}_i(\tau)$, where $\bar{\eta}_i$ is the mean value of η over the i-th subregion. With increase of the number of subregions, the error of assumption (4) decreases, and we may expect that the final results will be more accurate, although the labor of the method increases considerably. In examining examples we shall set $\beta = 0$ to facilitate integration, i.e., we shall use the Frank-Kamenetskii expansion [8] for $\exp(-E/RT)$. However, if we take dimensionless temperature in the form $\Theta = RT/E$, this expansion is not required.

We shall examine thermal explosion in an infinite cylinder. In this case, owing to symmetry, the boundary and initial conditions have the form

$$\left.\frac{\partial \Theta}{\partial \xi}\right|_{\xi=0} = 0, \quad \Theta(1, \tau) = 0, \quad \Theta(\xi, 0) = 0. \tag{5}$$

The function $f(x, y, z, \Theta_0)$ in this case may conveniently be taken to be

$$f = \Theta_0 - 2\ln(1 + a\xi^2). \tag{6}$$

Substituting (6) into (1) and (2), multiplying both sides of (2) by ξ, and integrating the result with respect to ξ from 0 to 1, we have the system of equations

$$\frac{da}{d\tau} = \frac{a^2[\delta\varphi(\bar{\eta})(1+a)^2 - 8a]}{2\delta[(1+a)\ln(1+a) - a]},$$

$$\frac{d\bar{\eta}}{d\tau} = \frac{\gamma\varphi(\bar{\eta})(1+a)}{2} \tag{7}$$

with initial conditions

$$a(0) = 0, \quad \bar{\eta}(0) = 0. \tag{8}$$

It may be assumed, a priori, that the system (7) is more accurate than the system (5.1) of [4] for thermal explosion in a cylinder, since the spatial features of this problem are taken into account not only by the structure of Eqs. (1) and (2), but also by the choice of profile (6). For the zero-order reaction $\gamma = 0$, and there remains only the first of Eqs. (7) with $\varphi(\bar{\eta}) \equiv 1$, which may easily be integrated, yielding, as $a \to \infty$,

$$\tau_0 = 2\delta \int_0^\infty \frac{(1+a)\ln(1+a) - a}{a^2[\delta(1+a)^2 - 8a]} da. \tag{9}$$

If the induction period τ_0 is finite, explosion of the reacting system occurs. Therefore, in this substitution the problem of thermal explosion in a cylinder reduces to the problem of the stability, in Lagrange's sense [9], of the solution of the first equation of (7) with $\varphi(\bar{\eta}) \equiv 1$. The quantity τ_0 tends to ∞, or does not exist at all, if the denominator of the integrand in (9) vanishes, i.e., if $\delta = 8a/(1+a)^2$. The limiting value $\delta = \delta_*$ at which the denominator again vanishes is reached for $a_* = 1$, and is equal to 2. Therefore, with $\delta > 2$ we have $\tau_0 < \infty$ and explosion occurs, while for $\delta \leq 2$ there is no explosion. The value $\delta_* = 2$ agrees with the corresponding exact value obtained in [8] with the aid of the steady theory of thermal explosion. Transforming (9), we have for the induction period the formula

$$\tau_0 = 2\delta \int_0^1 \{[(1+x)\ln(1+x) - x + x^3\{(1+x)\ln(1+x^{-1}) - 1\}] \times [x^2\{\delta(1+x)^2 - 8x\}]^{-1}\} dx. \qquad (10)$$

The values of τ_0, τ_{01}, τ_{02} for various values of δ are shown below:

δ	2.2	2.4	2.6	2.8	3	3.5,
τ_0	3.288	2.282	1.865	1.631	1.483	1.302,
τ_{01}	3.774	2.701	2.221	1.945	1.769	1.512,
τ_{02}	4.419	3.159	2.572	2.272	2.062	1.758.

It follows from this that we expect also that the value of τ_0 is more accurate than the value of τ_{02} satisfying the averaged system (5.1) of [4], while the accuracy of τ_0 (which is particularly important) is the greater, the closer δ is to 2.

In the case of a plate it is convenient to take

$$f = \Theta_0 - 2\ln \operatorname{ch} sx \qquad (11)$$

as $f(x, y, z, \Theta_0)$. Using (11), we may obtain by analogous considerations the value $\delta_* = 0.88$, which agrees with the exact value found in [8]. The chief difficulty in using the method of integral relations lies in integration of Eqs. (1) and (2), following the substitution in them of $f(x, y, z, \Theta_0)$. However, if we use the Grey and Harper [3] approximation,

$$\exp\Theta \approx 1 + 0.72\Theta + \Theta^2, \qquad (12)$$

this difficulty drops out. For example, it is easy, using (12), to solve the problem of thermal explosion in a parallelepiped. As $f(x, y, z, \Theta_0)$ we take

$$f = \Theta_0(1 - x^2)(1 - y^2p^{-2})(1 - z^2q^{-2}). \qquad (13)$$

The characteristic dimension here is equal to half the length of the smallest side. Because of the symmetry of the problem in question we integrate not over the whole region G, but over the part $0 \leq x \leq 1$, $0 \leq y \leq p$, $0 \leq z \leq q$. Substituting (13) into (1) and (2), using (12), and integrating the result over the above part of region G, we obtain

$$\frac{d\Theta_0}{d\tau} = \varphi(\bar{\eta})(3.375 + 0.72\Theta_0 + 0.512\Theta_0^2) - \frac{3\Theta_0 d}{\delta},$$

$$\frac{d\bar{\eta}}{d\tau} = \gamma\varphi(\bar{\eta})(1 + 0.2133\Theta_0 + 0.2731\Theta_0^2) \qquad (14)$$

with the conditions

$$\Theta_0(0) = 0, \quad \bar{\eta}(0) = 0. \qquad (15)$$

For the zero-order reaction there remains the first of Eqs. (14), which is easily integrated, and for the induction period we obtain the expression

$$\tau_0 = \frac{2\delta}{\sqrt{\Delta}}\left(\frac{\pi}{2} - \operatorname{arc\,tg}\frac{0.72\delta - 3d}{\sqrt{\Delta}}\right). \qquad (16)$$

From the condition $\Delta = 0$ we easily find the limiting value,

$$\delta_* = 0.896(1 + p^{-2} + q^{-2}). \qquad (17)$$

Expression (17) may also be obtained in the same way in which $\delta_* = 2$ was found for the infinite cylinder. For $\delta < \delta_*$ the integral determining τ_0 does not exist as an improper integral, but we may find its Cauchy principal value [9],

$$\tau_0 = \frac{\delta}{\sqrt{-\Delta}} \ln \frac{3d - \sqrt{-\Delta} - 0.72\delta}{3d + \sqrt{-\Delta} - 0.72\delta}. \qquad (18)$$

From (18) we obtain $\tau_0 < 0$, which is physically unreal. Thus, for $\delta > \delta_*$, $0 < \tau_0 < \infty$ and explosion occurs, while for $\delta \leq \delta_*$ it does not. From (17), with $p = q = 1$ we find $\delta_* = 2.69$; with $p = 1$, $q \to \infty$ we have for a cylinder with a square base $\delta_* = 1.79$; for a plate with $p \to \infty$, $q \to \infty$ we have $\delta_* = 0.896$. The exact value of δ_* for a cube is 2.53 [11]. When δ is little different from δ_*, formula (16) for τ_0 gives fair results, but with $\delta \gg \delta_*$ it gives greatly overestimated values of τ_0, in view of the fact that in its derivation we reduced considerably the source function $\exp\Theta$. For example, for a plate with $\delta = 0.968$ $\tau_0 = 4.83$, and with $\delta = 1.32$ $\tau_0 = 3.85$, while the accurate results, found with the aid of [4], are equal to 4.14 and 2.06, respectively.

We shall examine the problem of thermal explosion in an infinite cylinder with an arbitrary time of dependence of the cylinder wall temperature. A similar problem with linear growth with time of the external temperature was examined on the basis of the quasi-steady theory of thermal explosion in [12]. This is a problem of special interest in connection with storage of explosives. We shall assume that a zero-order reaction occurs. In this case we have only the single equation (1) with $\varphi(\bar{\eta}) \equiv 1$, along with the conditions

$$\left.\frac{\partial\Theta}{\partial\xi}\right|_{\xi=0} = 0, \quad \Theta(1, \tau) = \alpha\psi(k\tau), \quad \Theta(\xi, 0) = 0. \qquad (19)$$

With the aid of the substitution $w = \Theta - \alpha\psi(k\tau)$ we obtain the equation

$$\frac{\partial w}{\partial\tau} = \frac{1}{\delta\xi}\frac{\partial}{\partial\xi}\left(\xi\frac{\partial w}{\partial\xi}\right) + \exp[w + \alpha\psi(k\tau)] - \alpha k\psi'(k\tau) \qquad (20)$$

with homogeneous boundary and initial conditions, similar to conditions (5). Substituting the profile (6) into

(20) in place of w and integrating the result with respect to ξ from 0 to 1, we have the ordinary differential equation

$$\frac{da}{d\tau} = a^2\{\delta(1+a)^2\exp[\alpha\psi(k\tau)] - \delta\alpha k\psi'(k\tau)(1+a) - 8a\} \times \{2\delta[(1+a)\ln(1+a) - a]\}^{-1} \quad (21)$$

with the first of conditions (8). We shall examine the case when $\psi'(k\tau) > 0$ with $0 \leq \tau \leq \infty$, i.e., when $\psi(k\tau) \to \infty$ with $\tau \to \infty$. We note that the curve defined by the equation

$$\frac{d\underline{a}}{d\tau} = \frac{\underline{a}^2(1+\underline{a})\{\delta\exp[\alpha\psi(k\tau)] - \delta\alpha k\psi'(k\tau) - 2\}}{2\delta[(1+\underline{a})\ln(1+\underline{a}) - \underline{a}]}, \quad (22)$$

along with the first of conditions (8), lies below the curve $a(\tau)$ with $0 \leq \tau \leq \infty$, according to [13], since the right side of (21) is greater than the right side of (22) for any value of τ. Equation (22) is easily integrated:

$$\delta\int_0^\tau \exp[\alpha\psi(k\tau)]\,d\tau = 2\delta\int_0^{\underline{a}} \frac{\ln(1+a) - a}{a^2(1+a)^2}\,da + 2\tau + \delta\alpha\psi(k\tau). \quad (23)$$

It is easy to see that when $a \to \infty$, Eq. (23) has the solution $\overline{\tau}_0 < \infty$, since when $\tau = 0$ the right side of (23) is greater than the left, but as τ increases, the left side of (23) increases faster than the right side, and it is evident that the two curves intersect for $\overline{\tau}_0 < \infty$. Since $\underline{a}(\tau) \to \infty$ when $\tau \to \overline{\tau}_0 < \infty$, it is easy to see that $a(\tau) \to \infty$ when $\tau \to \tau_0 < \overline{\tau}_0$, and therefore explosion occurs for any values $\delta > 0$. Now let the external temperature increase, as $\alpha\psi(k\tau)$, until time $\tau_1 < \tau_0$, and remain constant at $\alpha\psi(k\tau_1)$ from time τ_1 on. On the basis of the foregoing analysis it may be asserted that when $0 < \tau < \tau_1$ explosion does not occur, and it makes sense to study (21), beginning from time τ_1, to which the value a_1 corresponds. In this case (21) is integrated:

$$\tau = \tau_1 + 2\delta\int_{a_1}^{a} \frac{(1+a)\ln(1+a) - a}{a^2\{\delta(1+a)^2\exp[\alpha\psi(k\tau_1)] - 8a\}}\,da. \quad (24)$$

If $a_1 \leq 1$, then the limiting value $\delta = \delta_*$ for which the denominator of the integrand in (24) vanishes, is reached with $a_* = 1$ and $\delta_* = 2\exp[-\alpha\psi(k\tau_1)]$. Thus an explosion limit exists in this case, and explosion occurs with $\delta > 2\exp[-\alpha\psi(k\tau_1)]$.

We shall examine the problem of auto-ignition of a viscous, incompressible reacting fluid moving in an infinite cylindrical tube. A similar problem, with no allowance for heat transfer and friction heat, was solved previously in [14]. We shall assume that the fluid flow follows the Poiseuille law, and that all the thermophysical properties are constant. Mathematically, the problem reduces to solution of the system of the following differential equations,

$$\nu\left(\frac{\partial^2 v}{\partial r^2} + \frac{1}{r}\frac{\partial v}{\partial r}\right) = -\frac{1}{\rho}\frac{\partial P}{\partial z_1}, \quad (25)$$

$$v\frac{\partial T}{\partial z_1} = \frac{\mu}{\rho c_p}\left(\frac{\partial v}{\partial r}\right)^2 + \varkappa\left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial z_1^2}\right) + \frac{Qk_0(1-\eta)^n}{c_p\rho}\exp\left(-\frac{E}{RT}\right), \quad (26)$$

$$v\frac{\partial\eta}{\partial z_1} = D\left(\frac{\partial^2\eta}{\partial r^2} + \frac{1}{r}\frac{\partial\eta}{\partial r} + \frac{\partial^2\eta}{\partial z_1^2}\right) + (1-\eta)^n k_0\exp\left(-\frac{E}{RT}\right) \quad (27)$$

with the boundary conditions

$$v(r_0, z_1) = 0, \quad \left.\frac{\partial v}{\partial r}\right|_{r=0} = 0, \quad T(r, 0) = T_0, \quad T(r, \infty) = T_0,$$

$$T(r_0, z_1) = T_0, \quad \left.\frac{\partial T}{\partial r}\right|_{r=0} = 0, \quad \eta(r, 0) = 0,$$

$$\eta(r, \infty) = 1, \quad \left.\frac{\partial\eta}{\partial r}\right|_{r=0,\,r_0} = 0. \quad (28)$$

The system of Eqs. (25)–(27) with boundary conditions (28) describes the regimes of auto-ignition and combustion in the tube. If attention is restricted to the auto-ignition regime, which may sometimes be realized, according to [15], in rocket motors, the original system may be appreciably simplified. In this case, as is usual, we may neglect burnup of the reacting substance right up to the time of ignition. Then (27) may be omitted, and the system reduces to (25) and (26). In addition, we neglect conductive heat transfer along the tube in comparison with convective, which is quite justified [14]. Equation (25) is easily integrated, and gives the well-known Poiseuille profile [16] for v, substitution of which into (26), and reduction of the result to dimensionless form using the Frank-Kamenetskii transformation [18] for exp(−E/RT) gives the equation

$$\frac{\partial}{\partial\xi}\left(\xi\frac{\partial\Theta}{\partial\xi}\right) + g\xi^3 - m\sqrt{\delta}\,\xi(1-\xi^2)\frac{\partial\Theta}{\partial z} + \delta\xi\exp\Theta = 0 \quad (29)$$

with the boundary conditions

$$\Theta(\xi, 0), \quad \Theta(1, z), \quad \left.\frac{\partial\Theta}{\partial\xi}\right|_{\xi=0} = 0. \quad (30)$$

By substituting profile (6) in (29) in place of Θ, allowing for the fact that then $a = a(z)$, we have

$$\frac{da}{dz} = \frac{a^3[2\delta(1+a)^2 + g(1+a) - 16a]}{2m\sqrt{\delta}\,[2(1+a)^2\ln(1+a) - a(2+3a)]} \quad (31)$$

with an initial condition similar to (8). Equation (31) is easily integrated, and for the dimensionless distance from the edge of the tube $z = z_0$ at which ignition of the reacting mixture occurs, we obtain

$$z_0 = 2m\sqrt{\delta}\int\limits_0^\infty \frac{2(1+a)^2\ln(1+a) - a(2+3a)}{a^3[2\delta(1+a)^2 + g(1+a) - 16a]}\,da. \qquad (32)$$

It is easy to see that the limiting value $\delta = \delta_*$ is reached with $a_* = (16 + g)/(16 - g)$ and, equally, $\delta_* = (16 - g)^2/128$. Therefore, the higher the intensity of dimensionless friction heat g, the lower the ignition limit δ_*. When $g = 0$ the limiting value coincides, as might be expected, with the corresponding value $\delta_* = 2$ for thermal explosion in the cylinder. When $\delta > \delta_*$ $z_0 < \infty$ auto-ignition of the reacting mixture also occurs, while when $\delta \le \delta_*$ it does not. It may be seen from (32) that with increase of $\bar{v}$, and therefore of m, the quantity z_0 increases, since the influence of friction heat is relatively small.

The examples presented, while they do not exhaust the whole scope of problems which may be solved with the aid of the method of integral relations, do indicate the effectiveness of this method for qualitative and quantitative investigation of ignition theory problems.

NOTATION

$\Theta = \dfrac{E(T-T_0)}{RT_0^2}$ —dimensionless temperature; $\tau = \dfrac{QEk_0t}{c_V\rho RT_0^2}\exp\left(-\dfrac{E}{RT_0}\right)$ —dimensionless time; $x = \dfrac{x_1}{r_0}$, $y = \dfrac{y_1}{r_0}$, $z = \dfrac{z_1}{r_0}$ — dimensionless coordinates; $\gamma = \dfrac{c_V\rho RT_0^2}{E}$ —a dimensionless parameter; $\delta = \dfrac{QEk_0r_0^2}{\lambda RT_0^2}\exp\left(-\dfrac{E}{RT_0}\right)$ —Frank-Kamenetskii criterion [8]; $\beta^{-1} = E/RT_0$—relative activation energy; $\varphi = 1$, $(1-\eta)$, $(1-\eta)^2$ — for the reaction of zeroth, first, and second order, respectively; Γ—boundary of region G in which the system (1), (2) is defined; $\bar{\eta}$—mean value of η over region G; $\xi = r/r_0$—dimensionless variable radius; T—absolute temperature in reaction vessel; T_0—temperature of vessel walls; R—universal gas constant; E—activation energy; c_V—specific heat at constant volume; Q—thermal effect of reaction; ρ—density; t—time; k_0—preexponent; r_0—characteristic dimension; x_1, y_1, z_1, r—dimensionless coordinates; λ—thermal conductivity; $a = \exp(\Theta_0/2) - 1$; $s = \mathrm{Arch}\exp(\Theta_0/2)$ $p = l_2/r_0$, $q = l_3/r_0$—half lengths of the second and third edges of parallelepiped, expressed as a fraction of the characteristic dimension; $d = 1 + p^{-2} + q^{-2}$; $\Delta = 6.3936\delta^2 + 4.32\delta d - 9d^2$; τ_{10} — induction period as found from formula (4.1) of [4], with $\beta = 0$, $\gamma = 0$, $\beta \to \infty$ and $n = 1$; τ_{02} — induction period, as found from (4.1) of [4], with $\beta = 0$, $\gamma = 0$ and $f_2 \equiv 1$; $\alpha = \alpha_1 E/RT_0^2$—dimensionless parameter; α_1—a parameter with dimension °K; $k = RT_0^2\rho k_1c_V/qEk_0\exp(-E/RT_0)$—dimensionless parameter: k_1—parameter with dimension $\sec^{-1}$; ν—kinematic viscosity; $\varkappa$—thermal diffusivity; v—longitudinal component of flow velocity; P—pressure; c_p—specific heat at constant pressure; n—order of the reaction; $g = 16\mu\bar{v}^2E/\rho c_pRT_0^2$—dimensionless parameter describing the intensity of friction heat; $\bar{v} = \bar{Q}/\pi r_0^2$—mean flow velocity; $m = \dfrac{2\bar{v}}{\varkappa}\left[\dfrac{\lambda RT_0^2}{QEk_0}\times\exp\left(\dfrac{E}{RT_0}\right)\right]^{1/2}$—dimensionless parameter; $\bar{Q}$—mass flow of the fluid per second; μ—viscosity; D—diffusion coefficient.

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