CRITICAL CONDITIONS FOR THERMAL EXPLOSION IN CONDUCTIVE HEAT TRANSFER IN THE REACTION ZONE AND THE SURROUNDING MEDIUM

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The critical conditions for thermal explosion are examined for plane, sphere, and cylinder in an unbounded medium with convective heat transfer in the internal and external regions. Criteria are used in integro-differential equations in the form of a Duhamel integral, which allow the conjugate problem to be reduced to a boundary one. Critical conditions are calculated by computer, with analysis for the cases of greatest practical interest.

NOTATION

c is the thermal capacity; ρ is the density; λ is the thermal conductivity; a is the thermal diffusivity: Q is the heat of reaction; E is the activation energy; k_0 is the preexponential factor; η is the extent of reaction; T is the temperature; T_0 is the temperature of surrounding medium; r_0 is the characteristic dimension of body. Symbols with subscript 1 refer to the surrounding medium.

1. The problem of thermal explosion may be formulated as follows in terms of dimensionless quantities for conductive heat transfer from a body in an unbounded medium:

a) Equation of conduction in the body:

$$\gamma \frac{\partial\theta}{\partial\tau} = \varphi(\eta) \exp\frac{\theta}{1+\theta\beta} + \frac{1}{\delta}\left(\frac{\partial^2\theta}{\partial\xi^2} + \frac{n}{\xi}\frac{\partial\theta}{\partial\xi}\right) \quad (\xi \leqslant 1); \qquad (1.1)$$

b) Equation of conduction in the medium:

$$\omega_a\gamma \frac{\partial\theta}{\partial\tau} = \frac{1}{\delta}\left(\frac{\partial^2\theta}{\partial\xi^2} + \frac{n}{\xi}\frac{\partial\theta}{\partial\xi}\right) \quad (\xi \geqslant 1); \qquad (1.2)$$

c) Kinetic equation:

$$\frac{\partial\eta}{\partial\tau} = \varphi(\eta) \exp\frac{\theta}{1+\beta\theta}. \qquad (1.3)$$

Initial conditions:

$$\tau = 0, \quad \eta = 0, \quad \theta = 0. \qquad (1.4)$$

Boundary conditions:

$$\partial\theta / \partial\xi = 0, \quad \xi = 0; \quad \theta \to 0, \quad \xi \to \infty. \qquad (1.5)$$

Conditions for junction of solutions:

$$\theta|_{\xi=1-0} = \theta|_{\xi=1+0}, \quad \omega_\lambda \frac{\partial\theta}{\partial\xi}\Big|_{\xi=1-0} = \frac{\partial\theta}{\partial\xi}\Big|_{\xi=1+0}. \qquad (1.6)$$

Here θ is temperature, τ is time, and ξ is coordinate:

$$\theta = \frac{E}{RT_0^2}(T - T_0), \quad \tau = k_0 \exp\left(-\frac{E}{RT_0}\right)t, \quad \xi = \frac{x}{r_0}.$$

Dimensionless parameters:

$$\delta = \frac{Qk_0Er_0^2}{\lambda RT_0^2} \exp\left(-\frac{E}{RT_0}\right), \quad \gamma = \frac{c\rho RT_0^2}{QE},$$

$$\beta = \frac{RT_0}{E}, \quad \omega_a = \frac{a}{a_1}, \quad \omega_\lambda = \frac{\lambda}{\lambda_1}. \qquad (1.7)$$

The number n characterizes the symmetry: n = 0, planar case; n = 1, cylindrical; n = 2, spherical; $\varphi(\eta)$ is the kinetic function. In the case of a first-order reaction, $\varphi(\eta) = 1 - \eta$.

This problem has been solved by computer for wide ranges in the parameters.

2. The initial conjugate problem may be divided into two parts: an internal one ($\xi \leq 1$), where there are continuously distributed heat sources, and an external one ($\xi \geq 1$), where we have a classical problem in the theory of heat conduction, which may be formulated as follows as an independent problem:

$$\frac{\partial\theta}{\partial\tau} = \frac{1}{\nu}\left(\frac{\partial^2\theta}{\partial\xi^2} + \frac{n}{\xi}\frac{\partial\theta}{\partial\xi}\right) \quad (\nu = \delta\gamma\omega_a) \qquad (2.1)$$

$$\tau = 0, \quad \theta = 0; \quad \xi = 1 + 0,$$

$$\theta = \theta(\tau); \quad \xi \to \infty, \quad \theta \to 0. \qquad (2.2)$$

The solution to this characterizes the external heat transfer of a body in an unbounded medium. Solutions to many such problems are well known, mainly for particular cases where the temperature at the surface of the body remains constant ($\theta = 1$).

Then for the above three cases we have [1]

$$n = 0, \quad N = \frac{1}{\sqrt{\pi F}} \quad \left(N = -\frac{\partial\theta}{\partial\xi}\Big|_{\xi=1}\right)$$

$$n = 1, \quad N = \frac{4}{\pi^2}\int_0^\infty \exp(-u^2F)\frac{du}{u\,[I_0^2(u) + Y_0^2(u)]}$$

$$n = 2, \quad N = 1 + \frac{1}{\sqrt{\pi F}} \quad \left(F = \frac{\tau}{\nu}\right). \qquad (2.3)$$

Here N is Nusselt's number, F is the Fourier number, and $I_0(u)$ and $Y_0(u)$ are Bessel functions of zero order and of the first and second kinds.

Then $N \to 0$ for $F \to \infty$ for a plane or cylinder, i.e., these do not allow of a steady state in external heat transfer, since the temperature in the external medium tends to equalize. $N \to 1$ for $F \to \infty$ in the case of a sphere.

Then for F large, namely

$$1 / \sqrt{\pi F} \ll 1,$$

the deviation from stationary thermal conditions may be neglected.

If the surface temperature varies, with $\theta|_{\xi=1+0} = \theta(\tau)$ a function of arbitrary form, the external conduction problem may be solved by the methods of operational calculus, the solution giving the varying heat flow through the surface of the body. Then use of Eq. (1.6) allows one to reduce the conjugate problem to a boundary one, the boundary condition for $\xi = 1$ being provided by integro-differential relations in the form of a Duhamel integral.

For instance, for the above three cases

$$n = 0, \quad \frac{\partial\theta}{\partial\xi} = -\frac{\sqrt{\delta}}{\sigma}\frac{d}{d\tau}\int_0^\tau \frac{\theta(z)}{\sqrt{\tau - z}}\,dz \quad \left(\sigma = \left[\frac{\pi\omega_\lambda^2}{\gamma\omega_a}\right]^{1/2}\right) \quad (2.4)$$

$$n = 1, \quad \frac{\partial\theta}{\partial\xi} = -\frac{4}{\pi^2\omega_\lambda}\frac{d}{d\tau}\int_0^\tau \theta(\tau - z) \times$$

$$\times \int_0^\infty \exp\left(-\frac{u^2\sigma^2 z}{\pi\delta\omega_\lambda^2}\right)\frac{du}{u\,[J_0^2(a) + Y_0^2(u)]}\,dz \quad (2.5)$$

$$n = 2, \quad \frac{\partial\theta}{\partial\xi} = -\frac{\sqrt{\delta}}{\sigma}\frac{d}{d\tau}\int_0^\tau \frac{\theta(z)}{\sqrt{\tau - z}}\,dz - \frac{\theta}{\omega_\lambda}. \quad (2.6)$$

Another boundary condition is the symmetry condition of (1.5).

The problem is thus reduced to one described by Eqs. (1.1) and (1.3), the initial condition (1.4), and the boundary conditions (1.5) and (2.4)–(2.6).

Given the form of $\theta(\tau)$, these integro-differential relations allow one to write down the expression for Nusselt's number

$$N = -\frac{1}{\theta}\frac{\partial\theta}{\partial\xi}\Big|_{\xi=1}$$

for a variable surface temperature in each particular case.

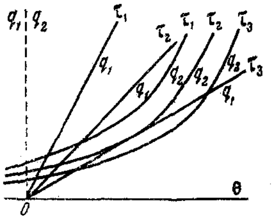

Fig. 1. Semenov diagram: q_1) direct heat loss, q_2) rate of heat release.

The form of $\theta(\tau)$ is not known in advance in problems of thermal explosion; this is to be determined. All the same, the Duhamel integral simplifies the analysis and the data processing.

3. The first two shapes of body do not provide a steady-state temperature distribution in the medium, and $N \to 0$ for $\tau \to \infty$, so critical conditions for thermal explosion for these bodies can exist only as a result of consumption of the material in a decomposition reaction of order higher than zero. Explosive decomposition will occur for any values of the parameters in the case of a zero-order reaction.

This is conveniently illustrated by Semenov's diagram [2] (Fig. 1).

As $N \to 0$ as $\tau \to \infty$, the direct loss q_1 will decrease $(\tau_1 < \tau_2 < \tau_3)$. The heat release is represented by a single curve for a zero-order reaction, so the heat output exceeds the heat loss in the course of time for any given values of the parameters, which inevitably leads to spontaneous ignition; critical conditions do not exist. A nonzero-order reaction, whose rate is dependent on the concentrations, will have q_1 falling as a result of consumption of material during the pre-explosion period, so the reaction may occur explosively or otherwise, in accordance with the relations between the parameters, and critical conditions do exist.

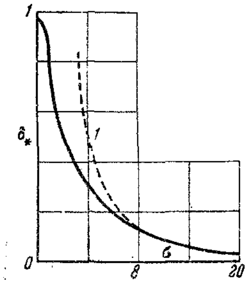

Fig. 2. The relation $\delta_* = f_0(\sigma)$ for $\gamma = 0.005$ and $\beta = 0.03$; curve 1 is $\delta_* = 7.8/\sigma_2$.

$N \to 1$ and $\tau \to \infty$ in the case of a sphere, and critical thermal-explosion conditions will exist for a reaction of any order.

Four parameters determine the critical values of δ_* (Frank-Kamenetskii's parameter [3]), which define the boundary between the regions of explosive and nonexplosive reaction: $\delta_* = f_n(\omega_\lambda, \omega_a, \gamma, \beta)$.

We assume, as usual,† that the β and γ of (1.1) have little effect on δ_*; we employ the new dimensionless parameter σ (obtained by reducing the problem to a boundary one) to simplify the relations:

$$n = 0, \quad \delta_* = f_0(\sigma)$$
$$n = 1, \quad \delta_* = f_1(\sigma, \omega_\lambda)$$
$$n = 2, \quad \delta_* = f_2(\sigma, \omega_\lambda).$$

The problem thus reduces to that of finding the forms of the functions f_n, which are substantially dependent on the geometry.

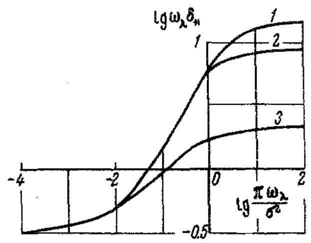

Fig. 3. The relation $\omega_\lambda\delta_* = F(\pi\omega_\lambda/\sigma^2)$ for $\gamma = 0.005$, $\beta = 0.03$, and ω_λ of: 1) 10; 2) 5; 3) 1.

4. Numerical calculations have been performed for the conjugate and boundary problems; the results agree to ~10%.

a) The δ_* for a plane is a function of σ alone; Fig. 2 shows $\delta_* = f_0(\sigma)$.

Here there are two limiting regions to consider. For $\sigma \to 0$, it follows from (2.4) that $\theta|_{\xi=1} \to 0$, i.e., we have conditions such that rapid external heat transfer causes the temperature at $\xi = 1$ to remain

†See [4–6] on the dependence of δ_* on β and γ.

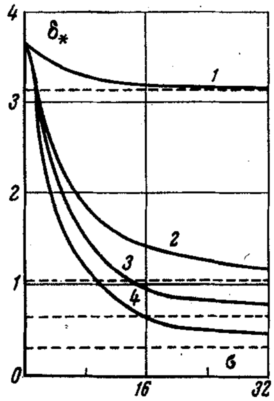

Fig. 4. The relation $\delta_* = f_1(\sigma, \omega_\lambda)$ for $\gamma = 0.005$, $\beta = 0.03$, and ω_λ of: 1) 0.1; 2) 1; 3) 2; 4) 4. The dashed lines represent the asymptotic values of δ_* given by the steady-state theory.

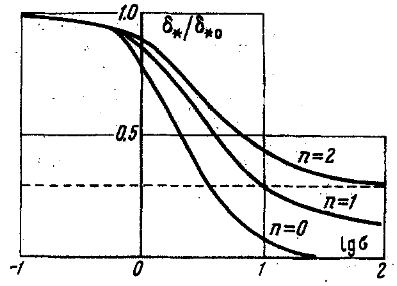

Fig. 5. Relation of δ_*/δ_{*0} to σ for plane, cylinder, and sphere for $\gamma = 0.005$ and $\beta = 0.03$; $\omega_\lambda = 1$ for n = 1 and n = 2.

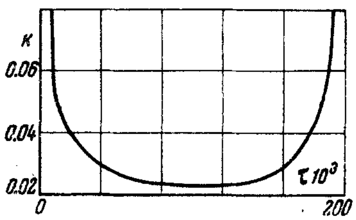

Fig. 6. The relation of $k = \gamma d\theta/d\tau$ to τ for $\gamma = 0.005$ and $\delta/\delta_* = 1.13$.

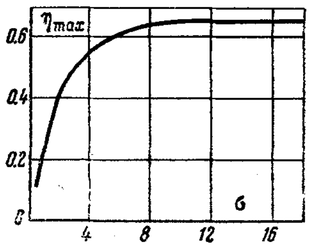

Fig. 7. Relation of η_{max} (degree of decomposition corresponding to maximum temperature rise) to σ for $\delta/\delta_* = 0.87$, $n = 0$, $\gamma = 0.005$, and $\beta = 0.03$.

constant at T_0 (boundary conditions of the first kind). Then δ_*, as corrected for $\gamma \neq 0$ and $\beta \neq 0$ [6], coincides with the δ_* for a plane calculated in [3].

As σ increases, (2.4) shows that the heat flow through the surface decreases, and δ_* falls; $\partial\theta/\partial\xi|_{\xi=1} \to 0$ for $\sigma \to \infty$, and δ_* also tends to 0.

For σ large, the low rate of dispersal of heat in the medium (relative to the body) means that there will be no temperature gradient in the reaction zone.

The following equations allow us to represent the problem via (2.4) if the temperature gradients in the reaction zone are neglected:

$$\gamma \frac{d\theta}{d\tau} = \varphi(\eta) \exp\frac{\theta}{1+\beta\theta} - \frac{1}{\Omega}\frac{d}{d\tau}\int_0^\tau \frac{\theta(z)}{\sqrt{\tau - z}}dz \qquad (\Omega = \sqrt{\delta}\sigma)$$

$$\frac{d\eta}{d\tau} = \varphi(\eta)\exp\frac{\theta}{1+\beta\theta}, \quad \tau = 0, \quad \theta = 0, \quad \eta = 0. \qquad (4.1)$$

For the critical conditions, neglecting the effects of β and γ, we have Ω_* = constant, so $\delta_* = f_0(\sigma)$ for $\sigma \to \infty$ is

$$\delta_* = c_1/\sigma^2.$$

The c_1 given by the solution of (4.1) is 7.35 ± 0.5, which agrees with the $c_1 = 7.8$ derived from $\delta_*(\sigma)$ (Fig. 2) for σ large.

The calculations show that $\sigma < 0.7$ and $\sigma > 8$ correspond to within ~5% with the limiting conditions for ideal heat transfer at the surface (boundary conditions of the first kind) and zero temperature gradient in the reaction zone respectively.

b) From Eq. (2.5), we have [1] for a cylinder for $\sigma \to 0$, $\omega_\lambda \neq 0$, and $z \neq 0$ that

$$I = \int_0^\infty \exp\left(-\frac{u^2\sigma^2 z}{\pi\omega_\lambda{}^2\delta}\right)\frac{du}{u\,[I_0^2(u) + Y_0^2(u)]} \approx \frac{\pi^2}{4}\frac{\omega_\lambda\sqrt{\delta}}{\sigma\sqrt{z}}.$$

Then

$$\frac{\partial\theta}{\partial\xi}\Big|_{\xi=1} \approx -\frac{\sqrt{\delta}}{\sigma}\frac{d}{d\tau}\int_0^\tau\frac{\theta(z)}{\sqrt{\tau-z}}dz, \qquad \lim_{\sigma\to 0}\theta|_{\xi=1} = 0,$$

i.e., the result for σ small is as for a plane, and $\delta_* = f_1(\sigma, \omega_\lambda) \approx f_1(\sigma)$.

We can determine δ_* in this range from the calculated results for a plane, with $\delta_* \approx f_1(\sigma) \approx 2.27\, f_0(\sigma)$.

For $\sigma \to \infty$, $\omega_\lambda \neq 0$, and $z \neq 0$, we have

$$I \approx \frac{\pi^2}{4}\left[\ln\frac{4z\sigma^2}{\pi\delta\omega_\lambda{}^2}\right]^{-1}, \quad \frac{\partial\theta}{\partial\xi}\Big|_{\xi=1} \approx -\frac{2}{\omega_\lambda}\frac{d}{d\tau}\int_0^\tau\frac{\theta(\tau - z)}{\ln 4z\sigma^2/\pi\delta\omega_\lambda{}^2}dz \to 0.$$

Hence $\delta_* \to 0$. The temperature gradient in the reaction zone is small when the conditions deviate widely from those of the first kind, as for a plane.

Then (2.5) becomes for a cylinder

$$\gamma\frac{d\theta}{d\tau} = \varphi(\eta)\exp\frac{\theta}{1+\beta\theta} - \frac{8}{\pi^2\omega_\lambda\delta}\frac{d}{d\tau}\int_0^\tau\theta(\tau - z)\times$$

$$\times\int_0^\infty\exp\left(-\frac{u^2\sigma^2 z}{\pi\omega_\lambda{}^2\delta}\right)\frac{du}{u\,[I_0^2(u) + Y_0^2(u)]}dz,$$

$$\frac{d\eta}{d\tau} = \varphi(\eta)\exp\frac{\theta}{1+\beta\theta}, \quad \tau = 0, \quad \theta = 0, \quad \eta = 0. \quad (4.2)$$

The functional relation between the parameters for critical conditions is then

$$\omega_\lambda\delta_* = \Phi\left(\frac{\pi\omega_\lambda}{\sigma^2}\right).$$

Figure 3 shows the form of this relationship. The region of nearly zero temperature gradient in the reaction zone is reached the earlier (at lower σ) the greater ω_λ.

c) Figure 4 shows $\delta_* = f_2(\sigma, \omega_\lambda)$ for a sphere.

We have $\delta_* \to 3.32$ (with correction for γ and β) for $\sigma \to 0$ (or $\omega_\lambda \to 0$). The effects of σ become slight (saturation) as σ increases, and δ_* in this range is dependent only on ω_λ. Physically, this means that the nonstationary external transfer can be replaced by quasi-stationary transfer, with $\alpha = \lambda_1/r_0$ [7] the expression for the coefficient of external heat transfer. Here $1/\omega_\lambda$ has the meaning of Biot's number [see Eq. (2.6)]. The calculations show that the deviation from stationary heat transfer can be neglected (with an error of ~10% in δ_*) for $\sigma \geq 50$ and $\omega_\lambda \leq 4$.

If ω_λ is large and $\sigma \to \infty$, the temperature gradient in the reaction zone is small, and δ_* can be found by Semenov's method [8]: $\delta_* = 3/\omega_\lambda e$. The gradient cannot be neglected for ω_λ small, and the boundary problem should be used to find δ_* [9]:

$$\delta_* = \frac{1.66}{\omega_\lambda^2}\left(\sqrt{1+4\omega_\lambda{}^2} - 1\right)\exp\left(\sqrt{1+4\omega_\lambda{}^2} - 2\omega_\lambda - 1\right).$$

The two methods give the same results (to ~5%) for this limiting case for $\omega_\lambda \geq 2$, i.e., the gradient can be neglected in this range.

This formulation of the problem is thus the most general in the case of spherical symmetry. It implies as limiting cases solutions known from the theory of thermal explosion: Frank-Kamenetskii's [3] ($\sigma \to 0$ or $\omega_\lambda \to 0$), Semenov's [2] ($\sigma \to \infty$ and $\omega_\lambda \to \infty$), and the boundary theory [9] ($\sigma \to \infty$).

5. The results allow us to examine the conditions for thermal explosion for n of 0, 1, and 2 in various real cases.

Ignition of spherical particles of explosive in a hot gas is characterized by $\omega_\lambda \approx 4$, $\sigma \approx 10^3$; so we can use the stationary approach (since $\sigma > 50$) to determine the critical conditions and can neglect the temperature gradient in the particles ($\omega_\lambda > 2$), i.e., we have Semenov's case [2].

For explosion mixture in a thin-walled container, $\omega_\lambda \approx 0.05$ and $\sigma \approx 0.2$, and all three shapes have boundary conditions of the first kind (Frank-Kamenetskii's case [3]).

For condensed material in molten lead or Wood's metal ($\omega_\lambda \approx 0.01$, $\sigma \approx 2.5$), we have boundary conditions of the first kind for cylinder and sphere ($\omega_\lambda \approx 0$), but not for a plane (Fig. 2).

For condensed material in a glass block ($\omega_\lambda \approx 0.2$, $\sigma \approx 10$), for a suspension ($\omega_\lambda \approx 0.4$, $\sigma \approx 12$), and for hot spots in a reactive mass ($\omega_\lambda \approx 1$, $\sigma \approx 18$), etc., it is essential to consider the specific features of the external conductive heat transfer; the critical conditions for thermal explosion may be derived from the present results.

The thermophysical parameters of [10, 11] have been used in deriving the dimensionless parameters.

6. Thermal explosion in the presence of conductive heat transfer has the following features:

a) The precise trends are very much dependent on the geometry. Only the spherical case produces a steady state under certain conditions and thus allows the classical approach to the critical conditions (steady-state theory). The other cases, except the limiting case of ideal heat transfer at the surface, have the critical conditions essentially linked to consumption of the material. The effects of consumption are in the nature of a correction in the case of convective external transfer.

It has been shown [6, 9] that δ_*/δ_{*0} (in which δ_{*0} is the value for boundary conditions of the first kind) for convective heat transfer from the surface is the same for all three cases, all conditions being the same. This is so in the present problem only for σ small; the δ_*/δ_{*0} for the various shapes may differ substantially if σ is large (Fig. 5).

b) The relation of internal transfer to external transfer is independent of size for all three shapes (the size does not appear in the basic parameters σ and ω_λ), and so the size does not affect the transition from ideal transfer to absence of temperature gradient in the reaction zone. The principal parameter for convective transfer is Bi, which does depend on the size.

c) A nonautocatalytic reaction near the self-ignition limit can take a quasi-stationary course in the case of conductive transfer to the medium, because $N(\tau)$ represents a time dependence. The quasi-stationary theory of thermal explosion deals with conditions such that the balance between heat production and heat loss gradually shifts on account of change in some quantity. An autocatalytic reaction [12] produces this shift by isothermal increase in the reaction rate, while under dynamic heating conditions [13, 14] the cause is increase in the temperature of the medium (in this case, reduction in the effective heat-transfer coefficient). Figure 6 illustrates the quasi-stationary condition for this problem by reference to the function [12] $k = \gamma d\theta/d\eta$, which characterizes the ratio of the heat-accumulation rate to the heat-loss rate. If $k \ll 1$, the state is quasi-stationary, and one consequence of this state is the greater extent of pre-explosion reaction characteristic of these conditions (Fig. 7).

An approximate quasi-stationary treatment for σ large in the planar case gives us the system of equations

$$\gamma \frac{d\theta}{d\tau} = \varphi(\eta) \exp \frac{\theta}{1+\beta\theta} - \frac{\theta}{\Omega} \frac{1}{\sqrt{\tau}}$$

$$\frac{d\eta}{d\tau} = \varphi(\eta) \exp \frac{\theta}{1+\beta\theta}, \quad \tau = 0, \quad \theta = 0, \quad \eta = 0 \qquad (6.1)$$

(as $\theta(z)$ varies little relative to $\sqrt{\tau - z}$, it can be extracted from the integral and the derivative).

We have $\Omega_* = \text{const}$ for critical conditions. Numerical integration gives $\Omega_* = 1.37 \pm 0.12$ for $\gamma = 0.005$ and $\beta = 0.03$, while integration of (4.1) for the same γ and β gives $\Omega_* = 2.7 \pm 0.1$. The approximate approach gives a result different from that for the exact approach on account of the long time needed to reach the quasi-stationary state in this case.

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