A. G. MERZHANOV, V. V. BARZYKIN, V. T. GONTKOVSKAYA

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Abstract

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A. G. MERZHANOV, V. V. BARZYKIN, V. T. GONTKOVSKAYA

THE PROBLEM OF A FOCAL THERMAL EXPLOSION

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The question of a focal thermal explosion has been widely discussed in the literature ([1]-[5]) in connection with the problem of sensitivity to mechanical actions; however, the authors confined themselves only to separate estimates and did not carry out a theoretical analysis of a focal explosion.

As is known, in a focal explosion the reaction does not occur throughout the entire mass of the substance, but in local places—reaction foci. It is essential that the medium surrounding the focus is not inert and that, as the focus develops, the adjacent layers of the substance are drawn into the reaction. In the present work, in the simplest formulation* the problem of a thermal explosion caused by a local focus of heating is solved. The focus is specified by a Π-shaped temperature profile at the initial moment of time in a spherical coordinate system. We assume that the dimensions of the focus are much smaller than the dimensions of the main mass of the substance. The initial differential equation in dimensionless variables has the form

$$\frac{\partial\theta}{\partial\tau} = e^{\theta/(1+\beta\theta)} + \frac{1}{\delta}\left(\frac{\partial^2\theta}{\partial\xi^2} + \frac{2}{\xi}\frac{\partial\theta}{\partial\xi}\right)$$

$$0 \le \xi < \infty, \qquad \tau \ge 0.$$

The initial and boundary conditions are specified:

$$\tau = 0, \quad \theta = 0 \text{ for } \xi \le 1; \quad \theta = -\theta_0$$

$$\text{for } \xi > 1; \quad \tau \ge 0, \quad \theta = -\theta_0$$

$$\text{for } \xi = \infty.$$

Fig. 1. Temperature at the center of the focus as a function of time for various δ at $\theta_0 = 10.35$

Figure 1

Figure 1: Figure 1

Here

$$\theta = \frac{E}{RT_0^2}(T - T_0); \qquad \xi = \frac{x}{r}; \qquad \tau = \frac{QEk_0}{c\rho RT_0^2} e^{-E/RT_0} t;$$

$$\delta = \frac{QEr^2 k_0}{\lambda RT_0^2} e^{-E/RT_0}; \qquad \beta = \frac{RT_0}{E}; \qquad \theta_0 = \frac{E}{RT_0^2}(T_0 - T_1);$$

x is the radial coordinate (cm); t is time (sec.); $T(x,t)$ is temperature (°K); T_0 is the initial temperature of the focus (°K); T_1 is the temperature of the mass of substance far from the focus (°K); r is the initial radius of the focus (cm); Q is the thermal effect of the reaction (cal/cm^3); k_0 is the pre-exponential factor (1/sec.); E is the activation energy (cal/mole); λ is the coefficient of thermal conductivity (cal/cm · sec · deg); c is the heat capacity (cal/g · deg); ρ is the density (g/cm^3). The parameters δ and β have the usual meaning for the theory of thermal explosion, while the parameter θ_0 characterizes the temperature head of the focus.

The initial equation was solved on an electronic computer; the temperature distribution was found as a function of time and of the system parameters $\theta = \theta(\xi, \tau, \delta, \theta_0)$. In all main calculations β was taken—

* The conditions of focus formation, phase transitions, etc., are not considered.

was taken equal to 0.03. The temperature head θ_0 was varied within the limits $4 < \theta_0 < 25$. For $\theta_0 < 4$ the formulation of the problem in this setting is incorrect, since the reaction in the bulk of the substance at temperature T_1 becomes comparable with the reaction in the hot spot.

The hot-spot problem has two aspects: explosion of the hot spot itself and hot-spot initiation of a self-propagating process in the bulk of the substance. The development of the explosion of the hot spot in time is most conveniently observed by the temperature at the center (Fig. 1). As is seen from Fig. 1, at small values of δ there is rapid cooling of the hot spot without heating up. At larger δ, the temperature at the center first increases and then decreases. At still larger δ, the hot spot does not have time to dissipate and the temperature grows in an explosive manner. The condition for explosion corresponds to values of the parameter δ greater than a certain critical value δ_{cr}. Figure 2 presents the dependence of δ_{cr}, as well as of the critical induction period τ_{cr} and the pre-explosion heating $\theta_{\text{max cr}}$, on the temperature head of the hot spot θ_0. As is seen from the figure, all these dependences are weak. For convenience of use, we represent the dependence $\delta_{\text{cr}}(\theta_0)$ by the approximate formula*

$$\delta_{\mathrm{cr}} \simeq 12.1(\ln\theta_0)^{0.6}$$

or, in dimensional variables,

$$r_{\mathrm{cr}} \simeq 3.48\,T_0\sqrt{\frac{\lambda R}{k_0 Q E}}\,e^{E/2RT_0}\left[\ln\frac{E}{RT_0^2}(T_0 - T_1)\right]^{0.3}.$$

For rough estimates one may take

$$\delta_{\mathrm{cr}} \simeq 20; \qquad \tau_{\mathrm{cr}} \simeq 2; \qquad \theta_{\mathrm{max\ cr}} \simeq 4.$$

Fig. 2. Dependence of $\delta_{\mathrm{cr}}, \tau_{\mathrm{cr}}$, and $\theta_{\mathrm{max\ cr}}$ on θ_0

Table 1

Dependence of hot-spot characteristics on β at $\theta_0 = 10.35$

β	δ_{cr}	τ_{cr}	$\theta_{\mathrm{max\ cr}}$
0.01	20.7	1.64	4.3
0.03	20.1	1.74	3.8
0.05	19.5	1.96	3.8

The dependence of the induction period on δ and θ_0 under supercritical conditions was also calculated. The calculation shows that as δ increases, the induction period at any θ_0 drops sharply and rapidly approaches the adiabatic value. (Thus, for example, at $\theta_0 = 10.35$ and $\delta/\delta_{\mathrm{cr}} = 1.4$, $\tau/\tau_{\mathrm{ad}} = 1.03$, whereas $\tau_{\mathrm{cr}}/\tau_{\mathrm{ad}} = 1.63$.) The dependence of the hot-spot characteristics on β, as was to be expected, proved weak (see Table 1).

To clarify the role of burnout of the substance in the course of the reaction, the system of heat-conduction and chemical-kinetics equations was solved for a first-order reaction:

$$\frac{\partial\theta}{\partial\tau} = e^{\frac{\theta}{1+\beta\theta}}(1-\eta) + \frac{1}{\delta}\left(\frac{\partial^2\theta}{\partial\xi^2} + \frac{2}{\xi}\frac{\partial\theta}{\partial\xi}\right),$$

$$\frac{\partial\eta}{\partial\tau} = \gamma e^{\frac{\theta}{1+\beta\theta}}(1-\eta),$$

$$0 \le \xi < \infty; \qquad \tau \ge 0; \qquad 0 \le \eta \le 1.$$

Table 2

Dependence of hot-spot characteristics on γ at $\theta_0 = 10.35$ and $\beta = 0.03$

γ	δ_{cr}	τ_{cr}	$\theta_{\max\text{ cr}}$
0.01	20.4	1.81	3.9
0.005	20.3	1.78	3.9
0.001	20.2	1.78	3.8
Zero order	20.1	1.74	3.8

* Accuracy of determining δ: 2%; accuracy of the approximation: within the accuracy of the calculation.

Initial and boundary conditions

$$\tau = 0;\ \theta = 0, \quad \eta = 0 \text{ for } \xi \leqslant 1; \qquad \theta = -\theta_0,\ \eta = 0 \text{ for } \xi > 1; \qquad \tau \geqslant 0,$$

$$\theta = -\theta_0 \text{ for } \xi = \infty.$$

Here η is the depth of conversion.

In problems taking burnout into account, an additional parameter is introduced,

$$\gamma = \frac{c\rho}{\theta}\frac{RT_0^2}{E}.$$

Table 2 gives the results of calculations for various γ. As we see, burnout has practically no effect on the characteristics of the process.

For clarifying the role of reaction in the surrounding medium during the development of a hot spot, cases were calculated in which heat release in the medium was assumed to be zero at all temperatures (a hot spot in an inert medium). For this purpose the initial conditions for η were specified as follows: $\eta = 0$ for $\xi \leqslant 1$ and $\eta = 1$ for $\xi > 1$. As a result of the calculation it was found that the reaction in the surrounding medium has no influence on the laws governing explosion of the hot spot. The characteristics of a hot spot in a reacting medium and in an inert one coincided (to within 1.5%). Moreover, it turned out that if, instead of a hot spot, one considers a vessel with thermostated walls, i.e., specifies a temperature constant in time at the boundary, then in this case too the critical values of the parameter δ and the other characteristics differ little from those considered above (Table 3).

Fig. 3. Temperature distributions at different times for $\theta_0 = 10.35$

The increase of δ_{cr} over the entire range of θ_0 is 15-25% (i.e., the critical temperature changes by several degrees). Thus, the calculations performed showed

Fig. 3. Temperature distributions at different times for $\theta_0 = 10.35$

Figure 2: Fig. 3. Temperature distributions at different times for $\theta_0 = 10.35$

that, in essence, there is no specificity of focal explosion associated with the presence of a reaction-capable environment and the absence of boundary conditions on the surface of the hot spot.

Table 3

Dependence of the hot-spot characteristics on the conditions in the surrounding medium. $\theta_0 = 10.35,\ \beta = 0.03,\ \gamma = 0.005$

	δ_{cr}	τ_{cr}	$\theta_{\text{max cr}}$
Hot spot in a reacting medium	20.1	1.74	3.8
Hot spot in an inert medium	20.2	1.74	4
Thermostated vessel	23.4	1.73	4.4

The presence of a reaction-capable environment manifests itself only in the second stage, when a self-propagating process is excited in the bulk of the substance. The study of the space-time development of a focal explosion is conveniently carried out by analyzing nonstationary temperature distributions (a typical picture is shown in Fig. 3). The initial store of heat in the hot spot warms the adjacent layers of the substance, and as a result strong cooling of the peripheral layers of the hot spot occurs. The temperature drop at the boundary ($\xi = 1$) rapidly decreases by approximately a factor of e, and subsequently changes only slightly over the entire induction period (by $\sim 10\%$). It is of interest to trace the change in the radius of the hot spo-

···over time at various temperature levels (Fig. 4). As is seen from Figs. 3 and 4, at temperature levels $-\frac{e-1}{e}\theta_0 < \theta < 0$, a narrowing of the source occurs during the induction period*. The size at the level $\theta = -\frac{e-1}{e}\theta_0$ (corresponding to a decrease in the difference by a factor of e) practically does not change.

For $-\theta_0 < \theta < -\frac{e-1}{e}\theta_0$, the radius of the source increases. After the expiration of the induction period, an explosive expansion of the source occurs at all temperature levels. The explosive rise in temperature originates at the center and then propagates in the radial direction, encompassing ever new layers, first

Fig. 4

Figure 3: Fig. 4

of the heated and then of the "cold" substance. Thus, we see that at temperature levels corresponding to the greatest reaction, the dimensions of the source decrease during the induction period. In light of this result, the conclusion that the presence of a reaction-capable environment has no influence on the characteristics of the explosion of the source becomes understandable. From the calculated data one may estimate the initial velocity of propagation of the process (see Fig. 4). In the first approximation, near the limit $(d\xi/dt)_{\text{init}} \simeq b/\delta$, where the quantity b depends only weakly on θ_0 and δ, so that

$$(dx/dt)_{\text{init}} = (2 \div 3)\, 10^3 a/d,$$

where a is the thermal diffusivity (cm^2/sec), and d is the initial diameter of the source (cm). By this formula one may estimate the initial velocity of propagation of the process in the bulk of the substance, if the initial diameter of the source is known. Thus, for example, for $d = 10^{-4}$ cm and $a = 10^{-3}$ cm^2/sec, $(dx/dt)_{\text{init}} = (200 \div 300)$ m/sec. The dependence of the initial propagation velocity on the diameter is apparently connected with the nonstationary character of the initiation of the process.

Fig. 4. Change of the radius of the source over time at various temperature levels: 1—at the level $\theta = 0$; 2—$\theta = -\frac{e-1}{e}\theta_0$; 3—$\theta = -0.95\theta_0$

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Institute of Chemical Physics
Academy of Sciences of the USSR

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* The quantity ξ_1, up to which the narrowing of the source occurs during the induction period, at the level of the initial temperature depends only weakly on θ_0 and increases with increasing δ. For example, for $\theta_0 = 10.35$, at $\delta = 20.5$, $\xi_1 = 0.37$; at $\delta = 25$, $\xi_1 = 0.5$; at $\delta = 35$, $\xi_1 = 0.6$; at $\delta = 100$, $\xi_1 = 0.79$.

Note: Figure translations are in progress. See original paper for figures.