

ONE LIMITING SCHEME FOR THE PROPAGATION OF A PULSATING  
EXOTHERMIC REACTION FRONT IN A CONDENSED MEDIUM

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In connection with studies on the synthesis of various solid substances in a combustion wave (self-propagating high-temperature synthesis) [1-3] a model is proposed for the unsteady combustion of gasless compositions which describes the relaxation mechanism of the propagation of an exothermic chemical reaction front in a condensed system, forming refractory products. The period of the oscillations in the combustion rate, the movement of the reaction front during one oscillation, and other characteristics of the process are determined. A comparison is made with the results of numerical calculations.

1. Relaxation Mechanism of the Propagation of an Exothermic Chemical Reaction Front in a Condensed Medium. Numerical calculations [1] have showed that in a wide region of the determining parameters a steady mode of combustion, responsible for the propagation of a reaction front with a constant velocity, is unstable, and the reaction front propagates in a self-oscillatory manner. Analytical studies [4, 5] of the stability of the stationary mode of combustion of gasless compositions by the method of small perturbations also showed the existence of an unstable region. The phenomenon of a self-oscillating mode of propagation of a reaction front was detected experimentally in [2].

A study of the structure of the pulsations in the combustion rate, carried out in [1] through the numerical solution of a nonstationary system of equations, made it possible to establish that the amplitude of the pulsations in the combustion rate increases in proportion to the departure from the curve of neutral stability into the depths of the unstable region, while the oscillations themselves have more and more of a relaxation nature; prolonged depressions in the combustion rate alternate with its considerable outbursts. In this case the space-time distributions of the temperature and concentration indicate that the prolonged heating of a certain part of the initial material is replaced by its rapid ignition and burn-up. Numerical calculations [1, 2] show that such a combustion mode for gasless compositions is established even at a small distance from the boundary of stability and is typical. The time dependence of the coordinates of the reaction front obtained by numerical calculations and presented in Fig. 1 is taken from [2] (curves 1 and 2 correspond to different distances from the boundary of stability).

A limiting model for the propagation of a pulsating exothermic reaction front in a condensed material is proposed below (a brief description of it was given earlier in [6]). In it we assume that following the burnup of the next portion of the initial material the heating of a new portion of it takes place over some time interval until the ignition condition is reached. After this the heated material rapidly burns up, as a result of which the reaction front moves a certain distance. The problem consists in the determination of the values of the movement of the reaction front and the period of the oscillations.

2. Derivation of the Integral Equation Determining the Temperature in the Chemical Reaction Zone. We will assume that during the entire combustion process the width of the chemical reaction zone because of the sharp dependence of the chemical reaction rate on the temperature is considerably less than the thickness of the heating zone (this assumption is confirmed by numerical calculations [1, 2]).

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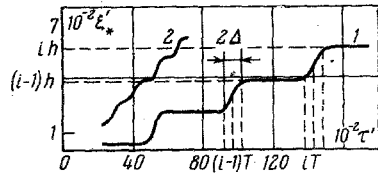


Fig. 1

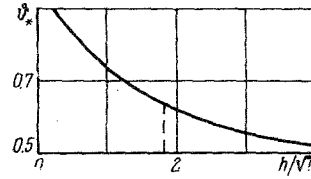


Fig. 2

Then the chemical reaction zone in which the complete conversion of the material takes place can be considered as a surface separating the initial material and the combustion products. Let us introduce a coordinate system connected with this surface (the initial material is in the region  $x < 0$ ).

The unsteady process of heat propagation in the condensed material to the left and to the right from the chemical reaction zone is described by the following equations and boundary and initial conditions:

in region I ( $x < 0$ )

$$\frac{\partial \theta_1}{\partial \tau} = \frac{\partial^2 \theta_1}{\partial \xi^2} - v(\tau) \frac{\partial \theta_1}{\partial \xi} \quad (2.1)$$

$$\tau = \tau_0, \theta_1 = \theta_1^0(\xi), \xi = -\infty, \theta_1 = 0, \xi = 0, \theta_1 = \vartheta(\tau)$$

in region II ( $x > 0$ )

$$\frac{\partial \theta_2}{\partial \tau} = \frac{\partial^2 \theta_2}{\partial \xi^2} - v(\tau) \frac{\partial \theta_2}{\partial \xi} \quad (2.2)$$

$$\tau = \tau_0, \theta_2 = \theta_2^0(\xi), \xi = 0, \theta_2 = \vartheta(\tau), \xi = +\infty, \theta_2 = 1$$

$$\theta = \frac{T' - T_0'}{T'_a - T_0'}, \xi = x \frac{u_a}{\kappa}, \tau = t \frac{u_a^2}{\kappa}, v(\tau) = \frac{u(t)}{u_a}$$

where  $x$  is the spatial coordinate,  $t$  is the time,  $T'$  is the temperature,  $T_0'$  is the initial temperature of the material,  $T'_a = T_0' + Q/c$  is the adiabatic combustion temperature ( $Q$  is the calorific effect, and  $c$  is the heat capacity),  $\kappa$  is the thermal diffusivity,  $u_a^2 = \kappa(RT'_a{}^2/E(T'_a - T_0'))k_0 \exp(-E/RT'_a)$  is the propagation rate of the exothermic reaction front corresponding to a steady solution ( $E$  is the activation energy,  $R$  is the gas constant, and  $k_0$  is the preexponent), and  $u(t)$  is the rate of movement of the reaction front in the laboratory coordinate system. The functions  $\theta_1^0$  and  $\theta_2^0$  describe the initial temperature distributions in each of the regions. The temperature  $\vartheta(\tau) = (T'(t, 0) - T_0')/(T'_a - T_0')$  in the reaction zone is an unknown function and is subject to determination.

By analogy with [7], where an integral equation for the unsteady combustion rate of a powder was derived, we apply a Fourier transformation to (2.1) and (2.2), assuming for problem (2.1) that  $\theta = 0$  when  $\xi > 0$ , and for problem (2.2) that  $\theta = 0$  when  $\xi < 0$ . We introduce

$$F_1(k, \tau) = \int_{-\infty}^0 \theta_1(\tau, \xi) e^{-ik\xi} d\xi \quad (2.3)$$

$$F_2(k, \tau) = \int_0^{+\infty} [\theta_2(\tau, \xi) - 1] e^{-ik\xi} d\xi \quad (2.4)$$

From (2.1) we have

$$dF_1/d\tau + (ikv + k^2)F_1 = \varphi_1 + (ik - v)\vartheta \quad (\varphi_1 = \partial\theta_1/\partial\xi|_{\xi=0})$$

$$\tau = \tau_0, F_1(k, \tau_0) = \int_{-\infty}^0 \theta_1^0(\xi) e^{-ik\xi} d\xi \quad (2.5)$$

From (2.2) it follows that

$$dF_2 / d\tau + (ikv + k^2)F_2 = -\varphi_2 + (ik - v)(1 - \vartheta) \quad (2.6)$$

$(\varphi_2 = \partial\theta_2 / \partial\xi|_{\xi=0})$   
 $\tau = \tau_0, \quad F_2(k, \tau_0) = \int_0^{\infty} [\theta_2^{\circ}(\xi) - 1] e^{-ik\xi} d\xi$

The solutions of the differential equations (2.5) and (2.6) with the indicated initial conditions have the respective forms

$$F_1(k, \tau) = \int_{\tau_0}^{\tau} \{\varphi_1(\tau') + [ik - v(\tau')] \vartheta(\tau')\} \exp[-k^2(\tau - \tau') - ikI] \times d\tau' + F_1(k, \tau_0) \exp[-k^2(\tau - \tau_0) - ikJ] \quad (2.7)$$

$$I = \int_{\tau'}^{\tau} v(\tau'') d\tau'' = \xi_*(\tau) - \xi_*(\tau')$$

$$F_2(k, \tau) = \int_{\tau_0}^{\tau} \{-\varphi_2(\tau') + [ik - v(\tau')][1 - \vartheta(\tau')]\} \exp[-k^2(\tau - \tau') - ikI] d\tau' + F_2(k, \tau_0) \exp[-k^2(\tau - \tau_0) - ikJ], \quad J = \int_{\tau_0}^{\tau} v(\tau'') d\tau'' \quad (2.8)$$

Here  $\xi_*(\tau)$  is the coordinate of the reaction front in the laboratory coordinate system at the time  $\tau$ . Applying the inverse transformation

$$\theta_1(\xi, \tau) = \frac{1}{2\pi} \int_{-\infty}^{\infty} F_1(k, \tau) e^{ik\xi} dk$$

$$\theta_2(\xi, \tau) - 1 = \frac{1}{2\pi} \int_{-\infty}^{\infty} F_2(k, \tau) e^{ik\xi} dk$$

to (2.7) and (2.8) and substituting the value  $\xi = 0$  into the equations obtained, we obtain, respectively,

$$\vartheta(\tau) = \frac{1}{\sqrt{\pi}} \left\{ \int_{\tau_0}^{\tau} \left[ \varphi_1 - v\vartheta + \frac{\vartheta I}{2(\tau - \tau')} \right] \exp\left[-\frac{I^2}{4(\tau - \tau')}\right] \times \right. \quad (2.9)$$

$$\left. \times \frac{d\tau'}{\sqrt{\tau - \tau'}} + \frac{1}{\sqrt{\tau - \tau_0}} \int_{-\infty}^0 \theta_1^{\circ}(u) \exp\left[-\frac{(u + J)^2}{4(\tau - \tau_0)}\right] du \right\}$$

$$\vartheta(\tau) - 1 = \frac{1}{\sqrt{\pi}} \left\{ \int_{\tau_0}^{\tau} \left[ -\varphi_2 + v(\vartheta - 1) - \frac{(\vartheta - 1)I}{2(\tau - \tau')} \right] \times \right. \quad (2.10)$$

$$\left. \times \exp\left[-\frac{I^2}{4(\tau - \tau')}\right] \frac{d\tau'}{\sqrt{\tau - \tau'}} + \frac{1}{\sqrt{\tau - \tau_0}} \int_0^{+\infty} [\theta_2^{\circ}(u) - 1] \exp\left[-\frac{(u + J)^2}{4(\tau - \tau_0)}\right] du \right\}$$

Since we seek a periodic solution not dying down with time and describing a mode of established oscillations, we should go to the limit as  $\tau_0 \rightarrow -\infty$  in Eqs. (2.9) and (2.10). Then the terms on the right sides of (2.9) and (2.10) which describe the effect of the initial conditions disappear. Combining the resulting equations, we obtain

$$2\vartheta - 1 = \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\tau} \left[ \varphi_1(\tau') - \varphi_2(\tau') - v(\tau') + \frac{I}{2(\tau - \tau')} \right] \exp\left[-\frac{I^2}{4(\tau - \tau')}\right] \frac{d\tau'}{\sqrt{\tau - \tau'}} \quad (2.11)$$

Let us now use the connection of the heat fluxes entering the reaction zone. Adding the heat-conduction equation, written with allowance for heat release and integrated over  $\xi$ , and the equation of consumption of the reacting material and reducing the width of the chemi-

cal reaction zone toward zero, we obtain

$$\varphi_1 - \varphi_2 = v - \lim_{\delta_x \rightarrow 0} \int_{-\delta_x}^{+\delta_x} \frac{\partial}{\partial \tau} (\theta + a) d\xi \quad (2.12)$$

Here  $a$  is the concentration of the reagent, and  $\delta_x = \delta_x / (v/u_a)$  is the dimensionless half-width of the chemical reaction zone.

For the combustion mode examined in Sec. 3, connected with sharp changes in the temperature of the chemical reaction zone, the integral on the right side of (2.12) has a maximum value during the "jump" of the reaction front, equal in order of magnitude to the ratio of the two small parameters  $\delta_x$  and  $\Delta_t = \Delta v / u_a^2$  ( $2\Delta$  is the burnup time for the next portion of the initial material; see Fig. 1), which is small in comparison with the other terms of the equation in virtue of the estimate  $\delta_x / \Delta_t \ll h / \Delta_t = \bar{u}$  ( $\bar{u}$  is the mean rate of propagation of the reaction front in the transition section). Substituting the connection of the heat fluxes (2.12) with allowance for this remark into (2.11) and using (2.7), we obtain

$$2\theta - 1 = \frac{1}{2\sqrt{\pi}} \int_{-\infty}^{\tau} \frac{[\xi_*(\tau) - \xi_*(\tau')]}{(\tau - \tau')^{3/2}} \exp\left\{-\frac{[\xi_*(\tau) - \xi_*(\tau')]^2}{4(\tau - \tau')}\right\} d\tau' \quad (2.13)$$

The periodicity of the function  $\theta(\tau)$  determined from this equation follows from the periodicity of the function  $v(\tau)$ . Equation (2.13) also shows that the dimensionless temperature of the chemical reaction zone is determined only by the law of motion of the reaction front. In particular, when the reaction front propagates with a constant velocity, i.e.,  $\xi_*(\tau) = \tau$ , it follows from (2.13) that  $\theta = 1$ . Below we will examine a mode of relaxation oscillations accompanied by prolonged heating of the inert material, although the integral equation (2.13) is derived only on the assumption of narrowness of the chemical reaction zone and therefore is valid for an arbitrary law of motion of the reaction front describing a mode of combustion over long times.

3. Determination of Ignition Temperature in the Case of Propagation of a Chemical Reaction Front "by Jumps." With the help of Eq. (2.13) we can determine the temperature of the chemical reaction zone at the moment of ignition for a combustion mode consisting in the alternation of the prolonged heating of some part of the initial material and its relatively rapid burnup. Let us assign the time dependence  $\xi_*(\tau)$  of the coordinate of the reaction front responsible for the indicated mode and illustrated graphically in Fig. 1 in the form

$$\begin{aligned} \xi_*(\tau) &= (i-1)h + \Phi_i(\Delta, \tau), (i-1)T \leq \tau \leq iT, i = -\infty, \dots, n \\ \Phi_i(\Delta, \tau) &= -\alpha h f_1^i(\Delta, \tau) + \beta h f_2^i(\Delta, \tau) \\ f_1^i(\Delta, \tau) &= f_1\left(\frac{\tau - (i-1)T}{\Delta}\right) \\ f_2^i(\Delta, \tau) &= f_2\left(\frac{iT - \tau}{\Delta}\right), \quad \alpha + \beta = 1, \quad \Delta \ll T \\ f_j(0) &= 1, f_j = 0 \text{ for } \tau \in [(i-1)T + \Delta, iT - \Delta] (j = 1, 2) \\ f_1'(0) &= f_2'(0) \end{aligned} \quad (3.1)$$

The transition function  $\Phi_i(\Delta, \tau)$  introduced here in the most general form describes the law of motion of the reaction front during the burnup of the heated portion of the initial material, which is accompanied by intense heat release. The functions  $f_1$  and  $f_2$  are monotonic and continuous in their physical sense.

Let us substitute the expression for  $\xi_*(\tau)$  into the integral equation (2.13) and determine the temperature in the chemical reaction zone during the time interval  $(n-1)T + \Delta \leq \tau \leq nT - \Delta$ . Since the function  $\xi_*(\tau')$  is periodic, it is convenient to replace the integral on the right side of (2.13) with a sum of integrals each of which is taken over the period. Calculating each integral with the accuracy of small values of order  $\Delta/T$ , with which all further calculations are conducted, we obtain

$$\begin{aligned}
2\theta - 1 = & \sum_{i=-\infty}^{n-1} \left\{ \operatorname{erf} \left[ \frac{h(n-i)}{2\sqrt{\tau-iT}} \right] - \operatorname{erf} \left[ \frac{h(n-i)}{2\sqrt{\tau-(i-1)T}} \right] \right\} + \\
& + \frac{1}{2\sqrt{\pi}} \int_{(n-1)T-\Delta}^{(n-1)T} \frac{h[1-\beta f_2^{n-1}(\Delta, \tau')]}{(\tau-\tau')^{3/2}} \exp \left\{ -\frac{h^2[1-\beta f_2^{n-1}(\Delta, \tau')]^2}{4(\tau-\tau')} \right\} \times \\
& \times d\tau' + \frac{1}{2\sqrt{\pi}} \int_{(n-1)T}^{(n-1)T+\Delta} \frac{\alpha h f_1^n(\Delta, \tau')}{(\tau-\tau')^{3/2}} \exp \left\{ -\frac{\alpha^2 h^2 [f_1^n(\Delta, \tau')]^2}{4(\tau-\tau')} \right\} d\tau' \quad (3.2)
\end{aligned}$$

For the calculations conducted in Sec. 4 it is convenient to reckon the time from the moment the chemical reaction front stops, i.e., to set  $\tau = \tau - (n-1)T - \Delta$ . Then Eq. (3.2) takes the form

$$\begin{aligned}
2\theta - 1 = & \sum_{i=-\infty}^{n-1} \left\{ \operatorname{erf} \left[ \frac{h(n-i)}{2\sqrt{\tau+(n-i-1)T}} \right] - \operatorname{erf} \left[ \frac{h(n-i)}{2\sqrt{\tau+(n-i)T}} \right] \right\} + \frac{1}{2\sqrt{\pi}} \int_{(n-1)T-\Delta}^{(n-1)T} \times \\
& \times \frac{h[1-\beta f_2^{n-1}(\Delta, \tau')]}{((n-1)T+\Delta+\bar{\tau}-\tau')^{3/2}} \exp \left\{ -\frac{h^2[1-\beta f_2^{n-1}(\Delta, \tau')]^2}{4((n-1)T+\Delta+\bar{\tau}-\tau')} \right\} d\tau' + \\
& + \frac{1}{2\sqrt{\pi}} \int_{(n-1)T}^{(n-1)T+\Delta} \frac{\alpha h f_1^n(\Delta, \tau')}{((n-1)T+\Delta+\bar{\tau}-\tau')^{3/2}} \exp \left\{ -\frac{\alpha^2 h^2 [f_1^n(\Delta, \tau')]^2}{4((n-1)T+\Delta+\bar{\tau}-\tau')} \right\} d\tau' \quad (3.3)
\end{aligned}$$

Note that the first term on the right side of (3.2), representing a converging series containing positive terms, arises as a result of the integration over the segments  $[(i-1)T+\Delta, iT-\Delta]$ . One can show that the integrals over the transition sections  $[(i-1)T, (i-1)T+\Delta]$  and  $[iT-\Delta, iT]$  ( $i = \infty, \dots, n-1$ ) make a contribution on the order of  $\Delta/T$  to the value of  $\theta$ , except for the integrals over the last transition section  $[(n-1)T-\Delta, (n-1)T+\Delta]$ , which are also written on the right side of (3.2). These integrals make a finite contribution to  $\theta$ , depending on the behavior of the smoothing functions  $f_1$  and  $f_2$ , only at the very start of the period of cooling of the chemical reaction zone when  $\tau - (n-1)T \sim \Delta$ . For times  $\tau - (n-1)T \gg \Delta$  the value of the integrals on the right side of (3.2) is on the order of  $\Delta/T$ , and the time dependence of the temperature in the reaction zone has the following form with the accuracy of small terms of order  $\Delta/T$ :

$$2\theta - 1 = \sum_{i=-\infty}^{n-1} \left\{ \operatorname{erf} \left[ \frac{h(n-i)}{2\sqrt{\tau-iT}} \right] - \operatorname{erf} \left[ \frac{h(n-i)}{2\sqrt{\tau-(i-1)T}} \right] \right\} \quad (3.4)$$

Thus, the function  $\theta$  falls sharply at the start of the period (in times on the order of  $\Delta$ ) from some value exceeding the adiabatic combustion temperature (the amount of the excess depends on the law of motion of the reaction front in the transition section) to a lower value and then varies smoothly in accordance with (3.4). Such behavior of the function  $\theta(\tau)$  is explained by the fact that after the burnup of the next portion of the initial material the temperature in the reaction zone is strongly increased, while the heat fluxes into the zone of reaction products and into the zone of initial material become very large, which results in a sharp decrease in the temperature.

Substituting the value  $\tau = nT - \Delta$  into (3.4), we obtain the temperature  $\theta_*$  in the reaction zone at which ignition occurs,

$$2\theta_* - 1 = \sum_{i=-\infty}^{n-1} \left\{ \operatorname{erf} \left[ \frac{h}{2\sqrt{T}} \sqrt{n-i} \right] - \operatorname{erf} \left[ \frac{h}{2\sqrt{T}} \sqrt{n-i+1} \right] \right\} \quad (3.5)$$

The dependence of the ignition temperature on the parameter  $h/\sqrt{T}$  is presented in Fig. 2. This curve is universal, since  $\theta_*$  does not depend on the other parameters of the problem.

4. Additional Relationships Determining the Size of the Jumps and the Period of the Pulsations. Following the ignition and rapid burnup of the next portion of the initial material the reaction front stops, and the temperature in the zone of chemical conversion increases considerably. With this the outflow of heat into the region of initial material

increases sharply, which leads to its warming due to the heat supplied from the zone of reaction products. As soon as the heated layer in the unreacted material becomes large enough ignition occurs.

In order to assign the condition of ignition let us use the concepts of the theory of the ignition of a combustible material by an incandescent surface developed by Zel'dovich [8, 9]. At the start of the period of cooling of the chemical reaction zone, when the temperature gradient at  $\xi = -0$  is very steep, one can neglect the heat being released in the reaction zone in comparison with the heat being drawn off into the interior of the initial material, and calculate its heating as the heating of an inert material with an assigned temperature at the boundary which varies with time in accordance with Eq. (3.3). The heat flow into the region  $\xi < 0$  decreases with time, while the thickness of the reacting layer increases, so that the heat being released in the reaction zone becomes comparable with the heat being drawn off into the initial material. Following [9], let us assign the condition of ignition (of reaching the ignition temperature) as the equality of the heat flux  $\varphi_1$  from the reaction zone into the region  $\xi < 0$  to the critical value  $\varphi_*$ . The flux  $\varphi_*$  corresponds to the smallest heat flux for which a stationary temperature distribution still exists in the reacting layer of the material. Thus, the condition of ignition has the form

$$\varphi_1 = l\varphi_* = l\sqrt{2} \frac{\varepsilon + \vartheta_*}{\varepsilon + 1} \exp \frac{\vartheta_* - 1}{2\gamma(\vartheta_* + \varepsilon)} \quad \left( \varepsilon = \frac{cT_0}{Q}, \gamma = \frac{RT_a}{E} \right) \quad (4.1)$$

In (4.1) we introduce an additional correction coefficient  $l$ , which has a value on the order of unity and can be chosen from a comparison of the results of the theory with experimental data or with the results of computer calculations for purposes of a more exact description of the effect when it deviates from the ideal scheme considered in the article.

For the determination of the heat flux into the interior of the initial material, neglecting the heat release in the chemical reaction zone, one must solve the heat-conduction problem:

$$\begin{aligned} \partial\theta / \partial\bar{\tau} &= \partial^2\theta / \partial\xi^2 \\ \bar{\tau} = 0, \theta &= \theta_0(\xi), \xi = 0, \theta = \vartheta(\bar{\tau}), \xi = -\infty, \theta = 0 \end{aligned} \quad (4.2)$$

Here  $\theta_0(\xi)$  is the temperature distribution in the initial material at the moment the reaction front stops. The solution of (4.2) has the form

$$\begin{aligned} \theta(\xi, \bar{\tau}) &= \frac{2}{\sqrt{\pi}} \int_{-\xi/2\sqrt{\bar{\tau}}}^{+\infty} \vartheta\left(\bar{\tau} - \frac{[\xi^2]}{4r^2}\right) e^{-r^2} dr + \\ &+ \frac{1}{2\sqrt{\pi\bar{\tau}}} \int_0^{\infty} \theta_0(y) \left[ \exp\left(-\frac{(\xi+y)^2}{4\bar{\tau}}\right) - \exp\left(-\frac{(\xi-y)^2}{4\bar{\tau}}\right) \right] dy \end{aligned} \quad (4.3)$$

Calculating from this the heat flux from the reaction zone at the moment of ignition, we obtain

$$\varphi_1 = \frac{\partial\theta}{\partial\xi}(0, T) = \frac{1}{\sqrt{\pi}} \left\{ \frac{\vartheta(0)}{\sqrt{T}} - \lim_{\xi \rightarrow 0} \xi \int_{-\xi/2\sqrt{T}}^{+\infty} \vartheta' \frac{e^{-r^2}}{r^2} dr - \frac{1}{2\sqrt{T}} \int_0^{\infty} y \exp\left(-\frac{y^2}{4T}\right) \theta_0(y) dy \right\} \quad (4.4)$$

As the numerical calculations of [1, 2] show, the initial distribution  $\theta_0(\xi)$  is such that the thickness of the heated layer at the initial time is small compared with the thickness of the layer heated by the time of ignition. This allows one to neglect the last term on the right side of (4.4), which describes the effect of the initial temperature distribution. Let us now substitute the expression for  $\vartheta(\bar{\tau})$  from (3.3) into (4.4) and calculate the heat flux  $\varphi_1$  with the accuracy of small values of order  $\Delta/T$ , keeping in mind that the contribution to  $\varphi_1$  from the integrals on the right side of (3.3), describing the effect of the transition section, is negligibly small because of their rapid dying out (in times on the order of  $\Delta$ ). As a result we obtain

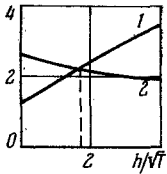


Fig. 3

$$\varphi_1 = \frac{1}{2\sqrt{\pi T}} [1 + g(z)] \quad (z = h/2\sqrt{T}) \quad (4.5)$$

$$g(z) = \sum_{i=-\infty}^{n-1} \left\{ \operatorname{erf} \left( \frac{z(n-i)}{\sqrt{n-i-1}} \right) - \operatorname{erf} (z\sqrt{n-i}) + \right.$$

$$+ \frac{1}{\sqrt{n-i+1}} \exp \left( -\frac{z^2(n-i)^2}{(n-i+1)} \right) \operatorname{erf} \left( z\sqrt{\frac{n-i}{n-i+1}} \right) -$$

$$\left. - \frac{1}{\sqrt{n-i}} \exp(-z^2(n-i)) \operatorname{erf} \left( z\sqrt{\frac{n-i}{n-i-1}} \right) \right\}$$

Now substituting  $\varphi_1$  from (4.5) into (4.1), we obtain the ignition condition in the form

$$\frac{1}{2\sqrt{\pi T}} \left[ 1 + g \left( \frac{h}{2\sqrt{T}} \right) \right] = l\sqrt{2} \frac{\varepsilon + \vartheta_*}{\varepsilon + 1} \exp \frac{\vartheta_* - 1}{2\gamma(\varepsilon + \vartheta_*)} \quad (4.6)$$

We note that the ignition condition  $\varphi_1 = \varphi_*$  is used in the theory of the ignition of a material by an incandescent surface at which a constant temperature is maintained. In the problem being considered here the ignition function  $\vartheta(\tau)$ , describing the law of the time variation of the temperature at the surface of the igniting material, varies sharply from some value at  $\tau = 0$  which exceeds the adiabatic combustion temperature to a considerably smaller value after a time on the order of  $\Delta$ , and then varies more slowly (close to a constant) in accordance with the behavior of the series on the right side of (3.3) which depends on the combination  $h/\sqrt{T}$ . Because of this circumstance one can use the condition (4.1) with good accuracy.

The value  $h$  by which the reaction front is shifted during each of its movements can be approximately determined from the temperature gradient near the reaction zone which is known at the moment of ignition, i.e.,

$$h = m \frac{\vartheta_*}{\varphi_*} = m \frac{\vartheta_*}{\sqrt{2}} \frac{\varepsilon + 1}{\varepsilon + \vartheta_*} \exp \frac{1 - \vartheta_*}{2\gamma(\varepsilon + \vartheta_*)} \quad (4.7)$$

Here  $m$  is a correction coefficient on the order of unity. We note that correction coefficients of the type introduced in Eqs. (4.1) and (4.7) are usually used in an approximate theory of ignition (see [9, 10], for example) for better quantitative agreement of the data of physical or numerical experiments with the theoretical dependences, which reproduce well all the qualitative relationships.

5. Calculation Procedure. Comparison of Results with the Data from a Numerical Solution of the Problem on a Computer. For the determination of  $h$  and  $T$  one must solve Eqs. (3.5), (4.6), and (4.7) relative to these values. It is convenient to proceed as follows. Multiplying together the right and left sides of Eqs. (4.6) and (4.7) we obtain (all further calculation is conducted for  $l = m = 1$ )

$$\frac{h}{\sqrt{T}} [1 + g(h/2\sqrt{T})] = 2\sqrt{\pi}\vartheta_* (h/\sqrt{T}) \quad (5.1)$$

The left (curve 1) and right (curve 2) sides of Eq. (5.1) are presented in Fig. 3. The point of intersection of the curves determines the sought value  $h/\sqrt{T} = 1.89$  (if the coefficients  $l$  and  $m$  are different from unity the value of the ratio  $h/\sqrt{T}$  naturally changes). Now one can determine from Fig. 2 the ignition temperature  $\vartheta_* = 0.625$ . Note that the indicated values of  $h/\sqrt{T}$  and  $\vartheta_*$  are universal for all modes (when  $l = m = 1$ ) corresponding to the propagation of a chemical reaction front "by jumps." Thus, the entire calculation is reduced to the determination of  $h$  from Eq. (4.7) for the given values of the parameters  $\varepsilon$  and  $\gamma$  and with  $\vartheta_* = 0.625$  and the subsequent calculation of the pulsation period

$$T = (h/1.89)^2 \quad (5.2)$$

and the mean velocity  $\omega = h/T$  of propagation of the chemical reaction front.

We note that the ignition temperature  $\vartheta_* = 0.625$  does not differ greatly from the value  $\vartheta_* = 0.5$  which is obtained in the solution of the self-similar problem on the decay of a thermal step ( $\theta_1(\xi, 0) = 0$ ,  $\theta_2(\xi, 0) = 1$ ). However, the difference which exists, produced by the effect of all the preceding cycles of oscillations, is very important in the determin-

ation of the critical conditions of ignition. If calculations are conducted according to (4.7) and (5.2) using  $\phi_* = 0.5$ , the results will differ by several orders of magnitude from the values obtained by computer calculations.

Let us make a comparison with the numerical calculations performed in [2], the results of which are presented in Fig. 1 in the dimensionless variables  $\xi_*' = xu_a'/\kappa$  and  $\tau' = tu_a'^2/\kappa$ , adopted in that work ( $u_a'$  is the steady propagation velocity of the reaction front at the boundary of stability, which corresponds to  $\gamma = 0.1619$ ).

For mode 1 (curve 1), which corresponds to  $\gamma = 0.0952$  and  $\varepsilon = 0.05$ , we find  $h' = h(u_a'/u_a) = 180$ ,  $T' = T(u_a'/u_a)^2 = 4600$ , and  $\omega' = \omega(u_a'/u_a) = 0.039$ . On the other hand, it follows from Eqs. (4.7) and (5.2) that  $h' = 145$ ,  $T' = 5810$ , and  $\omega' = 0.025$ . For mode 2 (curve 2), corresponding to  $\gamma = 0.1048$  and  $\varepsilon = 0.05$ , the numerical calculation gives  $h' = 125$ ,  $T' = 1450$ , and  $\omega' = 0.086$ , while calculations from Eqs. (4.7) and (5.2) give  $h' = 64$ ,  $T' = 1150$ , and  $\omega' = 0.056$ .

Thus, our calculations show that with a decrease in the parameter  $\gamma$  (an increase in the activation energy) the period of the pulsations in the combustion rate and the amount of movement of the reaction front during one oscillation increase, while the mean velocity decreases.

The above comparison with the results of numerical calculations indicates that the model of the pulsating combustion of condensed systems proposed in the article conveys the main aspects of the process qualitatively correctly and is in satisfactory quantitative agreement with the results of computer calculations.

In conclusion, we note that other schemes of the relaxation oscillations in the combustion rate of smokeless powders have been analyzed in [11, 12].

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