CALCULATION OF ONE TRANSIENT MODE
OF COMBUSTION OF A CONDENSED SYSTEM

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A one-dimensional process representing the combustion of a powder sample on a metal substrate at constant pressure is considered on the basis of a two-phase model of the thermal decomposition of a condensed system. The results of numerical computer calculations are presented. Qualitative comparison is made with experiment.

The burning of cylindrical samples of N powder on a copper plate was studied experimentally in [1, 2]. A state of combustion was observed in which the stage of ignition was followed by one of steady propagation, and then a cessation of burning as the combustion front approached the plate, this latter stage being attributed to the outflow of heat from the combustion zone to the plate, which had a far greater thermal conductivity than the powder. It was found that, after burning had ceased, a thin layer of unconsumed powder remained on the plate, its thickness depending on the pressure and initial temperature; experimental data were obtained as to the relationship between these three quantities.

An approximate theory of the phenomenon based on a model of combustion proposed by Ya. B. Zel'dovich [4] was developed in [2, 3]. In this paper we shall attempt a more detailed theoretical description of the mode of combustion studied in [1, 2]. Using an electronic computer we shall obtain a numerical solution to the problem of the transient combustion of a layer of typical condensed composition on a metal plate. As our model of combustion, we use one with an extended zone of dispersion, as proposed in [5, 6], with a continuous transition from the condensed to the gas phase.

1. Presentation of the Problem. Model of Combustion. Mathematical Formulation. In accordance with the experimental conditions, we consider the one-dimensional propagation of the zone of combustion along a flat layer of k phase, the thickness of which at the initial instant of time $t=0$ equals L. The thermal conductivity of copper and the total heat capacity of the copper disc are very high, and we may therefore consider that, in burning, the temperature at the powder/metal interface ($x=0$) remains constant and equal to that of the surrounding medium T_0. At the initial instant the sample temperature equals T_0. Let us assume that at the initial instant the temperature of the surface of the powder ($x=L$) becomes equal to the temperature of the flame T_* and retains this value the whole time, although the combustion front (leading edge) will move away from this point on account of the consumption of the k phase.

We must here remember that, in the absence of combustion, if at the initial instant the temperature takes the values T_0 and T_* at the points $x=0$ and $x=L$, respectively, the region $0 \leq x \leq L$ starts heating, and this process ends by the establishment of a linear steady-state temperature distribution. However, the process of heating takes place very much more slowly in the absence of heat evolution, and its influence appears after extinction.

We shall start from the combustion model proposed in [5, 6].

In this model it is assumed that the transformation of the k phase into gaseous reaction products takes place in such a way that there is no sharp k phase/gas interface. The gas is evolved within the vol-

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ume of the k phase in the form of small bubbles, the number and size of which increase continuously, the k phase swells, and the mean density of the material diminishes. The zone of transformation of the k phase into gas thus occupies an infinite region. The model gives an accurate description for the combustion of certain condensed systems and enables the steady-state rate of burning to be established and the steady-state concentration and temperature profiles to be determined [5, 6]; it also reflects certain characteristic features of the burning of N powder.

We assume that, when burning occurs in a condensed system, there is an irreversible chemical reaction of the first order. The reaction transforms condensed substance A_1 into gaseous products A_2 and is characterized by a thermal effect Q. The reaction velocity obeys the Arrhenius law with a frequency factor B and an activation energy E. The original substance does not contain any bubbles and consists entirely of the k phase, its density being equal to ρ_0.

The gas bubbles appearing in the k phase during the chemical reaction are small and are on average uniformly distributed throughout the mass of condensed material. The gaseous products are insoluble in the k phase and satisfy the equation of state of an ideal gas.

Let ρ_1, ρ_2 be the effective densities of substances A_1 and A_2, calculated on the assumption that both materials are distributed uniformly over the whole volume at each point in space. Then the average density of the condensed system is

$$\rho = \rho_1 + \rho_2 \tag{1.1}$$

The mass proportions of substances A_1, A_2 are

$$a_1 = \rho_1 / \rho, \qquad a_2 = 1 - a_1 \tag{1.2}$$

Let us express the average density ρ in terms of the mass proportion a_1, the pressure p, and the temperature T. We assume that the pressure is the same in all the gas bubbles and equal to the external pressure, while heat transfer takes place so quickly between the gas and the k phase that the gas and the k phase have the same temperature at every point.

The equation of state of the gas in the bubbles takes the form

$$p = M_2^{-1} \rho_2^* RT \tag{1.3}$$

Here R is the universal gas constant, M_2 is the molecular weight of the gas, ρ_2^* is the true gas density.

The effective density ρ_2 is related to the true density by

$$\rho_2^* = \frac{\rho_2}{1 - \rho_1/\rho_0} \tag{1.4}$$

It follows from (1.1)-(1.4) that

$$\rho = \rho_0 \left[\frac{RT}{M_2 p} \rho_0 (1 - a_1) + a_1\right]^{-1} \tag{1.5}$$

The local thermophysical properties of the system are characterized by the average values of the specific heat c and the thermal conductivity λ. The specific heat of the system is given by

$$c\rho = c_1 \rho_1 + c_2 \rho_2 \tag{1.6}$$

where c_1 is the specific heat of the k phase and c_2 that of the gas.

It follows from (1.6) and (1.2) that

$$c = c_1 a_1 + c_2 (1 - a_1) \tag{1.7}$$

The thermal conductivity λ, according to [7], is given by

$$\lambda (V_1 + V_2) = \lambda_1 V_1 + \lambda_2 V_2 \tag{1.8}$$

Here λ_1 is the thermal conductivity of the k phase, V_1 is the specific volume of the k phase, V_2 is the specific volume of the gas, and λ_2 is the thermal conductivity of the gas.

Clearly

$$\rho_0 V_1 = \rho_1 (V_1 + V_2), \quad \rho_0 V_2 = (V_1 + V_2)(\rho_0 - \rho_1)$$

Hence

$$\lambda = \lambda_1\left[a_1 + \frac{\lambda_2}{\lambda_1}\frac{RT}{M_2 p}\rho_0(1-a_1)\right]\left[a_1 + \frac{RT}{M_2 p}\rho_0(1-a_1)\right]^{-1} \tag{1.9}$$

We assume that λ_1 and λ_2 are independent of temperature and pressure.

There is no representation of a burning surface in the model of [5, 6]. When considering steady-state problems this has no effect on the results, since in the steady-state case all the points on the temperature and concentration profiles move at the same velocity, the steady-state velocity of combustion. These profiles resemble each other at different moments of time. In transient problems the temperature and concentration distributions no longer possess this property, and the concept of a combustion velocity becomes indeterminate. In order to eliminate this indeterminacy, we take the velocity of the plane on which the relative volumetric proportion of the gaseous combustion products is constant as the transient combustion velocity, i.e., the degree of dispersion $\alpha \equiv V_2/V_1 = \text{const}$. This quantity may be taken as equal to 0.5 in the case of the combustion of N powder, which approximately corresponds to the degree of dispersion on the burning surface.

Let us write down the equation and boundary conditions describing the transient process of the combustion of the powder on a metal substrate for the model in question.

The equation of conservation of mass is

$$\frac{\partial\rho}{\partial t} + \frac{\partial m}{\partial x} = 0, \qquad m = \rho u \tag{1.10}$$

Here m is the mass velocity and u the linear velocity of the substance. The change in density described by Eq. (1.10) takes place as a result of the gradual transition of some of the condensed material into the gaseous state.

The equation of conservation of the reacting substance is

$$\rho\frac{\partial a_1}{\partial t} = -m\frac{\partial a_1}{\partial x} - B\rho a_1 \exp\frac{-E}{RT} \tag{1.11}$$

The equation of energy conservation is

$$\rho\frac{\partial cT}{\partial t} = \frac{\partial}{\partial x}\left(\lambda\frac{\partial T}{\partial x}\right) - m\frac{\partial cT}{\partial x} + BQ\rho a_1 \exp\frac{-E}{RT} \tag{1.12}$$

The expansion of the condensed system as a result of the formation of gaseous products may be regarded as inertialess; hence, the equation of motion takes the form p = const.

The boundary conditions are

$$\begin{gathered} x = 0, \quad T(0, t) = T_0, \quad a_1(0, t) = 1, \quad m(0, t) = 0 \\ x = L, \quad T(L, t) \equiv T_* = Q/c_2 + c_1 T_0/c_2 \end{gathered} \tag{1.13}$$

The initial conditions are

$$t = 0, \quad T(x, 0) = T_0, \quad a_1(x, 0) = 1, \quad T(L, 0) = T_* \tag{1.14}$$

In accordance with the definition introduced in the foregoing, the coordinate of the surface of combustion x_s is given by the equation

$$\alpha = \frac{V_2(x_s)}{V_1(x_s) + V_2(x_s)} \tag{1.15}$$

In dimensionless variables the system of equations (1.10)-(1.12) and the conditions (1.13)-(1.15) may, after allowing for the relations (1.5), (1.7), and (1.9), be written in the form

$$\frac{\partial\varphi}{\partial\tau} + \frac{\partial\omega}{\partial\xi} = 0 \tag{1.16}$$

$$\varphi\frac{\partial a_1}{\partial\tau} + \omega\frac{\partial a_1}{\partial\xi} + \frac{\gamma\sigma\varphi a_1}{2(\beta+1)}\exp\frac{\gamma(\vartheta-1)}{\vartheta+\beta} = 0 \tag{1.17}$$

$$\varphi\Delta\frac{\partial\vartheta}{\partial\tau} - \frac{\partial}{\partial\xi}\left(\Lambda\frac{\partial\vartheta}{\partial\xi}\right) + \omega\Delta\frac{\partial\vartheta}{\partial\xi} - \frac{\gamma\sigma\varphi a_1(1-\vartheta+\vartheta\delta)}{2(1+\beta)}\exp\frac{\gamma(\vartheta-1)}{\vartheta+\beta} = 0 \tag{1.18}$$

$$\xi = 0,\ \vartheta(0, \tau) = \omega(0, \tau) = 0,\ a(0, \tau) = 1,\ \xi = \xi_0,\ \vartheta(\xi_0, \tau) = 1 \tag{1.19}$$

$$\tau = 0,\ \vartheta(\xi, 0) = \omega(\xi, 0) = 0,\ a(\xi, 0) = 1,\ \vartheta(\xi_0, 0) = 1 \tag{1.20}$$

$$\alpha = \frac{r(\vartheta + \beta)(1 - a_1)}{(1 + \beta)a_1 + r(\vartheta + \beta)(1 - a_1)} \tag{1.21}$$

Here

$$\varphi = \frac{r(1 + \beta)}{a_1(1 + \beta) + r(\vartheta + \beta)(1 - a_1)},\ \Lambda = \frac{l(1 + \beta)a_1 + r(1 - a_1)(\vartheta + \beta)}{(1 + \beta)a_1 + r(1 - a_1)(\vartheta + \beta)}$$

$$\tau = \frac{m_0{}^2 c_2 RT_*}{\lambda_2 p M_2}\, t,\quad \xi = \frac{m_0 c_2}{\lambda_2}\, x,\quad \omega = \frac{m}{m_0},\quad \delta = \frac{c_1}{c_2}$$

$$\vartheta = \frac{T - T_0}{T_* - T_0},\quad \xi_0 = \frac{m_0 c_2}{\lambda_2}\, L,\quad l = \frac{\lambda_1}{\lambda_2},\quad r = \frac{\rho_0 RT_*}{M_2 p}$$

$$\gamma = \frac{E}{RT_*},\quad \sigma = \frac{Q}{c_2(T_* - T_0)},\quad \beta = \frac{T_0}{T_* - T_0}$$

$$m_0{}^2 = 2\lambda_2 M_2 pBT_* Q^{-1} E^{-1} \exp(-E / RT_*),\quad \Delta = 1 - a_1 + a_1\delta$$

In passing from Eq. (1.12) to Eq. (1.18) we have used Eq. (1.11). As m_0 we choose the steady-state velocity of propagation of the combustion front defined by the Zel'dovich–Frank–Kamenetskii formula. Equations (1.16)-(1.18), together with conditions (1.19) and (1.2), determine the functions $\vartheta(\xi, \tau)$, $\omega(\xi, \tau)$, $a_1(\xi, \tau)$. Equation (1.21) serves to find the transient motion of the combustion surface $\xi_S = \xi_S(\tau)$.

2. Method of Solution. The problem (1.16)-(1.21) is a nonlinear boundary problem for three equations in partial derivatives. The solution may be obtained by numerical integration on a computer. For solving Eq. (1.18) we use an implicit four-point scheme with space and time steps respectively equal to h and k. The scheme is absolutely stable for the heat-conduction equation with constant coefficients, whatever the value of k/h^2. The computing region $(0, \xi_0, \tau)$ is divided up in the following way:

$$\xi_n = nh\ (n = 0, 1, 2, \ldots, N),\ \tau_m = mk\ (m = 0, 1, 2, \ldots)$$

The values of the functions at the point (nh, mk) are subsequently denoted by an upper index m and a lower index n.

The derivative with respect to time is written

$$\frac{\partial\vartheta}{\partial\tau} \approx \frac{\vartheta_n^{m+1} - \vartheta_n^m}{k} \tag{2.1}$$

The first term on the right-hand side of Eq. (1.18) takes the form

$$\frac{\partial}{\partial\xi}\left(\Lambda\, \frac{\partial\vartheta}{\partial\xi}\right) \approx \frac{\Lambda_{n+1/2}^m(\vartheta_{n+1}^{m+1} - \vartheta_n^{m+1}) - \Lambda_{n-1/2}^m(\vartheta_n^{m+1} - \vartheta_{n-1}^{m+1})}{h^2}$$

$$\Lambda_{n\pm 1/2}^m = \Lambda(a_{1,\, n\pm 1/2}^m,\ \vartheta_{n\pm 1/2}^m) \tag{2.2}$$

$$a_{1,\, n\pm 1/2}^m = \frac{a_{1n}^m + a_{1,\, n\pm 1}^m}{2},\quad \vartheta_{n\pm 1/2}^m = \frac{\vartheta_n^m + \vartheta_{n\pm 1}^m}{2}$$

The derivative with respect to the coordinate ξ may be written

$$\frac{\partial\vartheta}{\partial\xi} \approx \frac{\vartheta_n^{m+1} - \vartheta_{n-1}^{m+1}}{h} \tag{2.3}$$

Expressing all the coefficients and also the fourth term in Eq. (1.18) in terms of the values of the variables in the m-th layer, we obtain a difference equation which approximates the differential equation, with a residual term $O(h + \tau)$ linear with respect to the values of ϑ in the m+1-th layer,

$$A_n^{m+1}\vartheta_{n+1}^{m+1} - 2B_n^{m+1}\vartheta_n^{m+1} + C_n^{m+1}\vartheta_{n-1}^{m+1} = D_n^{m+1} \tag{2.4}$$

with the conditions $\vartheta_0^{m+1} = 0$, $\vartheta_N^{m+1} = 1$.

Here

$$A_n^{m+1} = \frac{\Lambda_{n+1/2}^m}{h^2},\quad 2B_n^{m+1} = \frac{\Lambda_{n+1/2}^m + \Lambda_{n-1/2}^m}{h^2} + \frac{\omega_{n-1/2}^m \Delta_{n-1/2}^m}{h} + \frac{\varphi_n{}^m \Delta_n{}^m}{k}$$

$$C_n^{m+1} = \frac{\Lambda_{n-1/2}^m}{h} + \frac{\omega_{n-1/2}^m \Delta_{n-1/2}^m}{h}$$

$$D_n^{m+1} = -\vartheta_n{}^m \frac{\varphi_n{}^m \Delta_n{}^m}{k} - \frac{\sigma\gamma}{2(1+\beta)} [1 + (\delta - 1)\vartheta_n{}^m] \exp\left[\gamma \frac{(\vartheta_n{}^m - 1)}{\vartheta_n{}^m + \beta}\right] \varphi_n{}^m a_{1,n}^m$$

We solve Eqs. (2.4) by asymptote fitting.

In solving Eq. (1.17) we use an implicit three-point scheme. We write

$$\frac{\partial a_1}{\partial \xi} \approx \frac{a_{1,n}^{m+1} - a_{1,n-1}^{m+1}}{h}, \quad \frac{\partial a_1}{\partial \tau} \approx \frac{a_{1,n}^{m+1} - a_{1,n}^m}{k} \tag{2.5}$$

Expressing the coefficients and the last term on the right-hand side in terms of the value of the function in the m-th layer, we obtain a recurrence formula for a_n^{m+1},

$$a_{1,n}^{m+1} = a_{1,n-1}^{m+1} S_n{}^m + F_n{}^m; \quad S_n{}^m = \frac{\omega_{n-1/2}^m}{h}\left[\frac{\varphi_n{}^m}{k} + \frac{\omega_{n-1/2}^m}{h}\right]^{-1} \tag{2.6}$$

$$F_n{}^m = \varphi_n{}^m a_{1,n}^m \left[\frac{1}{h} - \frac{\sigma\gamma}{2(1+\beta)} \exp \frac{\gamma(\vartheta_n{}^m - 1)}{\vartheta_n{}^m + \beta}\right]\left(\frac{\varphi_n{}^m}{k} + \frac{\omega_{n-1/2}^m}{h}\right)^{-1}$$

with the condition $a_{1,0}^{m+1}$.

As we now know the values of ϑ and a_1 in the m+1-th layer, we may find the values of φ_n^{m+1} from the equation approximating (1.16).

We obtain

$$\omega_n^{m+1} = \omega_{n-1}^{m+1} - h(\varphi_n^{m+1} - \varphi_n{}^m) k^{-1} \tag{2.7}$$

with the condition $\omega_0^{m+1} = 0$.

After determining the values of ϑ, a_1, ω in the m+1-th layer, we may pass to the m+2-th layer, and so on.

In order to find the law governing the motion of the surface on which condition (1.21) is satisfied, we carry out a linear interpolation between the space nodes of the network for $\xi_n \leq \xi \leq \xi_{n+1}$

$$\vartheta^n(\xi_*{}^n) = \frac{\vartheta_{m+1}^n - \vartheta_m{}^n}{h} \xi_*{}^n + \vartheta_{n+1}^m - n(\vartheta_{n+1}^m - \vartheta_n{}^m) \tag{2.8}$$

$$a_1{}^n(\xi_*{}^n) = \frac{a_{1,m+1}^n - a_{1,m}^n}{h} \xi_*{}^n + a_{1,m}^n - n(a_{1,n+1}^m - a_{1,n}^m) \tag{2.9}$$

Solving Eqs. (2.8), (2.9), and (1.21), we obtain a quadratic equation in $a_1{}^n(\xi_*{}^n)$. Taking the root lying between 0 and 1, we obtain the values of $a_1{}^n(\xi_*{}^n)$, $\vartheta^n(\xi_*{}^n)$, $\xi_*{}^n$. The validity of this difference scheme was verified by making a control calculation with different values of k. The results agreed very closely.

A calculation was carried out for a number of initial temperatures $T_0 = 275$, 300, 325, 350°K with a pressure of $p = 0.44$ cal/cm^3 (19 atm), and for a number of pressures $p = 0.44$, 0.66, 0.88 (cal/cm^3) at $T_0 = 300$°K.

In the case of $p = 0.44$ cal/cm^3 and $T_0 = 300$°K the values of the dimensionless parameters equal $\delta = 0.85$, $l = 2.43$, $\sigma = 1.09$, $\gamma = 14.27$, $r = 176.67$, $\beta = 0.61$. These values in particular correspond to the following physicochemical characteristics of the model combustion system: $E = 22{,}200$ cal/mole, $Q = 180$ cal/g, $\nu = 0.66$, $c_1 = 0.29$ cal/g·deg, $c_2 = 0.34$ cal/g·deg, $\lambda_1 = 3.65 \cdot 10^{-4}$ cal/sec·cm·deg, $\lambda_2 = 1.5 \cdot 10^{-4}$ cal/sec·cm·deg, $\alpha = 0.5$, $M_2 = 30$ g/mole.

3. Results. Figures 1-3 illustrate the space–time picture of the process in hand. Figure 1 shows the temperature distribution for various values of the dimensionless time, Fig. 2 the concentration distribution of mass velocity. Numbers 1-15 in the figures correspond to the following instants of time τ_n: 26, 104, 234, 286, 387, 488, 589, 690, 791, 993, 1094, 1299, 1325, 1429, 1455.

It follows from the choice of dimensionless parameters that all the quantities with the dimensions of length are referred to the thickness of the steady-state thermal layer in the gas.

We see from the graphs that the whole process may be divided into three stages: ignition, steady propagation, and extinction.

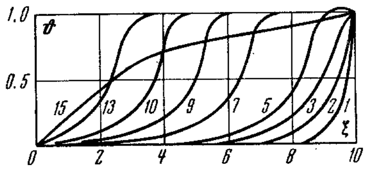

Fig. 1

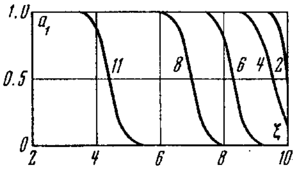

Fig. 2

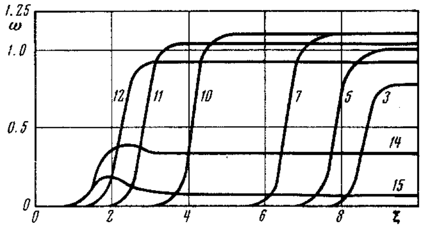

Fig. 3

At the first stage there is a rise in temperature and a fall in the concentration of the reacting substance close to the igniting surface. The medium starts moving. The temperature rise is here associated with two factors: heat evolution due to the chemical reaction, and the inflow of heat from the igniting surface. The second factor explains the slight rise in the local temperature of this zone above T_*. The combustion surface ($\alpha = 0.5$) first moves in a nonuniform manner, then at a distance of $\xi = \xi_0$ from the surface, equal to $\xi_0 - \xi \approx 1.8$, the steady-state mode sets in. The transient mode is replaced by the steady state at approximately the instant $\tau = \tau_6$. The distribution of the parameters in the combustion zone becomes similar at different moments of time (second stage).

The dimensionless mass velocity of combustion is here approximately equal to unity, in agreement with the results obtained in [6] when considering the steady-state propagation of a combustion front. At a distance from the metal–powder contact surface approximately equal to the thickness of the thermal layer of the k phase the thermal interaction of the combustion zone with the cold metal plate becomes substantial, the outflow of heat from the k phase increases, the reaction zone is "frozen," and the velocity of the burning surface falls (third stage). This causes the sharp spatial temperature front to vanish and reduces the slope of the whole temperature profile. The reaction velocity diminishes and the propagation of the burning front ceases. Subsequently the temperature distribution asymptotically tends toward the steady state.

In order to compare the results of the calculation with the experimental data of [1, 2], we must relate the thickness of the unburnt layer of k phase to the pressure and initial temperature. As the thickness of the unburnt residue s we take the minimum distance within which the combustion surface $\alpha = 0.5$ approaches the surface $\xi = 0$. We assume that the velocity constant of the chemical reaction depends on the pressure as $B = B_0 p^n$. Then the pressure dependence of the steady mass velocity of combustion may be written $m_0 \sim p^\nu$, $\nu = (n+1)/2$.

The results of the calculations relating to the thickness of the unburnt layer at different pressures were approximated to a fair accuracy by the formula

$$\ln s = \text{const} - \nu \ln p \quad (T_0 = \text{const})$$

The results of the calculations of s for different initial temperatures may be approximated by the formula

$$(\partial \ln s / \partial \ln m_0)_p = -1$$

The foregoing considerations show that, despite the relatively simple nature of the original combustion model, the results provide an excellent description of the process of powder combustion. The form of the $s = s(p, T_0)$ relationship obtained by the numerical solution of the problem agrees with experimental data. For a more precise quantitative analysis it is first essential to have a more detailed knowledge of the physicochemical properties of the powder; secondly, it is essential to perfect the combustion model, allowing for the occurrence of several reactions in the condensed and gas phases, and also diffusion in the gas.

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