EXTINCTION OF A POWDER WITH A CHANGE IN PRESSURE

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Existing theories with respect to unsteady-state conditions for the combustion of powders [1-6] use dependences of the mass combustion rate m on the pressure p and the initial temperature of the charge T_∞:

$$m = m(p, T_\infty) \tag{0.1}$$

of the surface temperature of charge T_1, more accurately the temperature at the internal boundary of the reaction zone in the condensed phase T_{1p}, on the combustion rate and the pressure or the initial temperature:

$$T_1 = T_1(m, p) = T_1(m, T_\infty) \tag{0.2}$$

or, sometimes, of the temperature of the determining flame state T_2 on the combustion rate and the pressure:

$$T_2 = T_2(m, p) \tag{0.3}$$

obtained under steady-state conditions. However, in unsteady-state processes the parameter of a powder may take on values falling outside the intervals within which the dependences (0.1)-(0.3) were determined experimentally. Therefore, to describe extinction and ignition processes, these dependences must be supplemented by criteria which determine their limits of applicability. An exception is the case of a constant surface temperature of the powder, discussed by Ya. B. Zel'dovich [1, 2]. In this case, extinction manifests itself as the impossibility of agreement between the temperature distributions in the gas and condensed phases. The present work establishes a criterion for the case when the surface temperature of the powder depends only on the combustion rate:

$$T_1 = T_1(m) \tag{0.4}$$

Such a dependence was noted by B. V. Novozhilov [7] and was obtained directly in experiments with powder H by A. A. Zenin.† A dependence of this type is characteristic for a Q-model [8], in which it is assumed that the breakdown of the surface of the powder is determined by the depth of the conversion in some reaction taking place in the condensed phase. For a reaction of the general form (0.2), the determining stage is obviously the fume-gas zone. It is shown that the criteria for the extinction of a flame with a stepwise decrease in the pressure and the ignition of a powder are determined by laws analogous to those obtained earlier by Ya. B. Zel'dovich for the case of a constant surface temperature of the powder.

1. For a clear picture of transitional conditions with the combustion of a powder, following [1] it is convenient to use a graphical representation of the steady-state dependences (0.1)-(0.3) in the form of dependences on the mass combustion rate (Fig. 1). In addition to the dependences of T_∞, T_1, and T_2 (curves 1-3, respectively), there is shown also the change in the maximal possible temperature $T_3 = (c_1 T_\infty + q_1 + q_2)/c_2$ (line 4). Here q_1, c_1, q_2, and c_2 are the thermal effects and the heat capacities in the condensed and gas phases, respectively. Curves 1-4 are plotted for the case of a constant temperature of the charge.

For two values of the pressure, p and p*, plots are also given of the dependences of the temperature of the flame $T_2 = T_2(m, T_\infty)$ with a variable temperature of the charge (curves 5 and 6, respectively); these can be regarded as the laws governing combustion in the gas phase at the surface of the powder. On these curves there must exist limiting points corresponding to the limits of stable combustion of the powder; these

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† A. A. Zenin, Doctoral Dissertation, Institute of Chemical Physics (1972).

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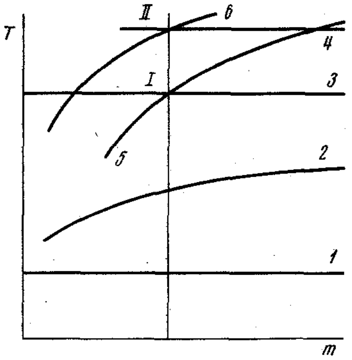

Fig. 1

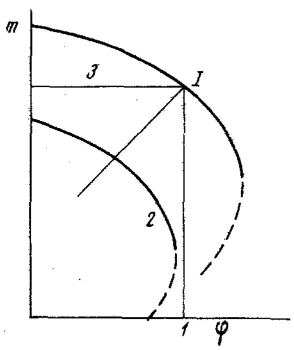

Fig. 2

are not considered here. With an instantaneous decrease in the pressure from p to p*, following Ya. B. Zel'dovich we shall assume that the processes taking place in the gas phase are inertialess. This means that the point representing the combustion process in the gas phase must lie on curve 6. On the other hand, by virtue of the thermal inertia of the condensed phase, the rate of gassification at the surface of the powder, determined by relationship (0.4), remains unchanged. Thus, point II, obtained from the intersection of curve 6 and a line m = const, passing through the initial point I, determines the conditions at the surface of the powder immediately after the decrease in the pressure. The maximal possible temperature of the flame at a surface with the temperature T_1 is equal to $T_3 = (c_1T_1 + q_2)/c_2$. Therefore, if point II lies above curve 4, there is extinction of the flame at the surface of the powder. Under these circumstances, the rate of gassification exceeds the maximal combustion rate, and the flame cannot maintain itself at the surface and is carried downstream. Another clear picture of this process can be obtained in the coordinates $\varphi_1 = \partial T_1/\partial x$, m (Fig. 2). A process taking place with an instantaneous decrease in the pressure under the condition $T_1 =$ const [2] is represented in these coordinates by the line $\varphi_1 =$ const, passing through the initial point I. Extinction corresponds to a decrease in the pressure down to a level where there is no intersection of the curve corresponding to steady-state combustion at the pressure p*. Here and in what follows an asterisk denotes critical values.

In the case under consideration, the process is represented by the line m = const, i.e., the straight line 3, and there is extinction even in the case when there is no intersection of this line and curve 2. We note that, near the point $\varphi_1 = 0$, steady-state combustion is unstable [1] and extinction may set in with a smaller change in the pressure than follows from the condition $\varphi_1{}^* = 0$.

2. For the critical value of the pressure p*, at which the flame is extinguished, the following equality is satisfied:

$$m\,(T_3,\ p^*) = m\,(T_2{}^\circ,\ p) \tag{2.1}$$

Here the degree sign denotes the steady-state value. Using relationships (0.1) and (0.2) in the form

$$m = A_2 p^\nu \exp\,(-E_2\ /\ RT_2) \tag{2.2}$$

$$m = A_1 \exp\,(-E_1\ /\ RT_1) \tag{2.3}$$

and also

$$T_2{}^\circ = (c_1T_1 + q_2)\ /\ c_2 - \rho\varkappa_2\varphi_2\ /\ m \tag{2.4}$$

$$T_3 = (c_1T_1 + q_2)\ /\ c_2 \tag{2.5}$$

$$T_1{}^\circ = T_\infty + q_1\ /\ c_1 + \rho\varkappa_1\varphi_1\ /\ m \tag{2.6}$$

(here ρ is the density of the substance; $\varkappa$ is the coefficient of thermal diffusivity), we obtain

$$(p^*\ /\ p)^\nu = \exp\,(-E_2\,(1\ /\ T_2{}^\circ - 1\ /\ T_3)\ /\ R) \approx \exp\,(-E_2\ /\ R\ (T_2{}^\circ)^2)$$

Using the definition

$$\beta = \left(\frac{\partial \ln u}{\partial T_\infty}\right)_p = E_2\,\frac{\partial T_1}{\partial T_\infty}\ \Big/\ R\,(T_2{}^\circ)^2$$

we obtain

$$(p^*\ /\ p)^\nu = \exp\,(-c_2\beta\,(T_1{}^\circ - T_\infty - q_1\ /\ c_1)\ /\ c_1) \tag{2.7}$$

a relationship which coincides with the result of Ya. B. Zel'dovich [1].

If there is no extinction of the flame, the change in the combustion rate can be calculated in accordance with the theory of B. V. Novozhilov [9]; to this end, it is necessary only to pass over from the quantities T_1 and φ_1 to the quantities T_{1p} and φ_{1p}. If the thermal effect of the reaction in the condensed phase is known, this can be done using dependences (2.4)-(2.6) and analogous relationships containing φ_{1p}.

With a finite rate of change in the pressure, conditions (2.1) may not be satisfied at the initial moment of time, but later. It must be noted that conditions (2.1) are the conditions for extinction of the flame at the surface of the powder; therefore, the possibility of repeated ignition cannot be excluded.

The condition for the stability of combustion in the model of Ya. B. Zel'dovich, $\partial\varphi_1/\partial m < 0$, can be interpreted as the existence of a negative feedback, stabilizing small deviations in the value of φ_1 from the equilibrium value. Actually, with a random rise in the value of φ_1, there is a drop in the flame temperature, determined by the relationship

$$T_2 = T_2^\circ + (\rho\varkappa_1\varphi_1 / m)^\circ - (\rho\varkappa_1\varphi_1 / m)$$

This leads to a decrease of the combustion rate in the gas phase, to a movement of the flame away from the surface, and to a decrease in the value of φ_1. For a combustion model in which relationship (0.4) is satisfied, the analogous criterion will be

$$\left(\frac{\partial m_1}{\partial T_1}\right)_p > \left(\frac{\partial m_2}{\partial T_1}\right)_p \tag{2.8}$$

Relationship (2.8) means that, with a random increase in the temperature of the surface, there is also a change in the combustion conditions; under these circumstances, the flame moves away from the surface. This takes place as a result of the fact that the rate of gassification m_1 increases more strongly than the rate of propagation of the flame m_2. Movement of the flame away from the surface leads to a decrease in in the value of φ_1 and, by the same token, to a decrease in the surface temperature. Using relationships (2.3), (2.4), (2.6) and the definition of the quantities β and $r = (\partial T_1/\partial T_\infty)_p$, criterion (2.8) can be converted to the form

$$E_2 / RT_2^2 > r\beta c_2 / c_1$$

3. Let us examine the possible mechanisms for the combustion of a powder, which will permit an explanation of the different transitional processes, under the conditions $\varphi_1 = \text{const}$ and $m = \text{const}$. It is known [1, 2] that models with a constant surface temperature correspond to the combustion of volatile compositions, for which the combustion rate is determined by vaporization or some other type of phase transition. The main special characteristic of such a model is the dependence of the amount of substance vaporizing from the surface on the value of the heat flux arriving at the surface. If l is the heat of the phase transition and λ is the coefficient of thermal conductivity, then

$$m = (\lambda_2\varphi_2 - \lambda_1\varphi_1) / l \tag{3.1}$$

Here subscripts 1 and 2 refer to values at the surface, from the side of the condensed phase and of the gas, respectively. By virtue of thermal inertia, the value of φ_1 varies continuously, even with a stepwise change in the combustion rate. Precisely in view of this, a process with $\varphi_1 = \text{const}$ is a transitional process.

Under steady-state conditions and taking account of the fact that

$$\varphi_1 = m\,(T_1 - T_\infty) / \rho_1\varkappa_1 \tag{3.2}$$

relationship (3.1) can be rewritten in the form

$$m = \lambda_2\varphi_2 / (l + c_1\,(T_1 - T_\infty)) \tag{3.3}$$

Under the condition $T_1 = \text{const}$, relationship (3.3) means that the value of the combustion rate depends only on the value of the heat flux.

A dependence of the combustion rate only on the surface temperature can be observed if the rate of gasification of the condensed phase η is determined by the degree of completion of some chemical reaction taking place in the condensed phase. This postulation is known as a Q-model [8, 10]. Let the rate of the chemical reaction be $W(T, \eta)$. Then, the value of $\eta = \eta^\circ$ at the surface, under steady-state combustion conditions, is determined by the relationship

$$\eta^\circ = \int_{-\infty}^{t} W\,(T\,(t'), \eta\,(t'))\,dt' \tag{3.4}$$

Here $T(t')$ and $\eta(t')$ are the temperature and depth of conversion at the point of the charge which, at the moment of time t, falls on the surface of the condensed phase. Here, diffusion in the condensed phase is neglected.

Going over to a system of coordinates connected with the surface, we obtain a relationship for the linear rate of motion of the surface of the condensed phase:

$$u = \int_{-\infty}^{0} W(T(x), \eta(x))\, dx / \eta^\circ \tag{3.5}$$

We make the assumption that the rate of the chemical reaction is described by an Arrhenius dependence with a large activation energy. This is equivalent to the assumption of the inertialess nature of the reaction in the condensed phase, since the whole reaction zone will be located in a narrow zone at the surface of the powder, where maximal temperatures are observed. As a result of the strong temperature dependence of the reaction rate, the dependence of the latter on the depth of conversion η may be neglected. Using an expansion of Frank-Kamenetskii's exponent and determining the value of φ_1 from relationship (2.6), we obtain

$$u^2 = \frac{A_1 \varkappa_1 R T_1^2 \exp(-E_1 / RT_1)}{\eta^\circ E_1 (T_1 - T_\infty - q_1 / c_1)} \tag{3.6}$$

In this respect, the determining factor is the dependence of the combustion rate on the temperature of the surface; the dependence on the initial temperature and the thermal effect is only slightly noted.

Under unsteady-state conditions, let the depth of the reaction remain constant (we note the analogy with Ya. B. Zel'dovich's postulation with respect to the constancy of the surface temperature); then, the combustion rate is determined as the rate of motion of the surface

$$\eta(x, t) = \eta^\circ, \text{ i.e., } u = -\frac{\partial \eta}{\partial t} \Big/ \frac{\partial \eta}{\partial x}$$

Here the derivatives must be calculated in a laboratory system of coordinates. Consequently,

$$\partial \eta / \partial t = W(T, \eta)$$

and the value of $\partial \eta / \partial x$ is determined from the solution of the equation of conservation of mass. With a given distribution of $\eta(x, 0)$ and under the boundary condition $\eta(0, t) = \eta^\circ$, by virtue of its inertialess nature, the quantity η varies continuously. This implies continuity of the combustion rate.

From the position of curves 1 and 3 (Fig. 2) it follows that in the general case (dependence (0.2) for the surface temperature) the transitional process develops along a line occupying an intermediate position. In this case, cessation of the combustion is not very probable. This case obviously corresponds to combustion under conditions when the zone which determines the process is the fume-gas zone. In this case, the most important special characteristic is the continuity of the distribution of the parameters over the combustion zone. This is completely analogous to the case of the laminar combustion of gases when, to attain extinction, the heat losses from the flame front must be taken into consideration. Using steady-state dependences, there are no such losses, and consequently there is no extinction.

4. It is also possible to use relationship (3.4) to describe the process of the ignition of a powder, with given laws governing the external heat flux to the surface of the condensed phase. In this case, gasification at the surface starts only when the required depth of conversion has been attained at the surface of the powder. With rapid heating, the temperature of the surface at the moment of the start of gasification will be higher than with slow heating. If, after the start of gasification, the heat flux does not decrease as the result of flow of the gases away from the surface, such a high rate of gasification may be obtained that a flame does not appear at the surface of the powder. As before, the critical condition will be condition (2.1), which is conveniently represented in the form

$$m(T_3, p) = m^*$$

Thus, the combustion model under consideration, in spite of the considerable difference between dependence (0.4) and that proposed by Ya. B. Zel'dovich, has properties which are close to the properties of a model with a constant surface temperature [11].

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