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Fig. 1

Figure 1: Fig. 1

Abstract

Full Text

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ON THE IGNITION OF PARTICLES OF EXPLOSIVE SUBSTANCES IN A HEATED GAS

(Presented by Academician N. N. Semenov, 20 IV 1964)

Numerous works have been devoted to the study of evaporation and combustion of liquid droplets, as well as to heterogeneous combustion of solid particles (see, for example, ([1])). Along with this, the study of the behavior of particles of homogeneous explosive substances (e.s.) in a heated gas is of undoubted interest, since it makes it possible: a) to clarify a number of questions connected with various regimes of exothermic transformations of e.s. (thermal self-ignition, ignition, transition from self-ignition to ignition ([2])); b) to investigate the possibility of using ignition as a method for studying the macrokinetics of chemical reactions at high temperatures; c) to obtain information on the lifetime and laws of transformation of dispersed particles formed during the combustion of "nonvolatile" condensed explosive systems ([3],[4]), etc.

In the present work, using barium azide (BaN_6) as an example, an experimental study was carried out of the regularities of ignition of particles of homogeneous e.s. in a heated gas.

For this purpose the following apparatuses were used. A vertically mounted glass tube is heated by an electric coil arranged so that visual observation and photography of the ignition of particles could be carried out (Fig. 1A). Constancy of temperature along the length of the tube is monitored by thermocouples. At the top of the tube there is a small opening for introducing particles. The fall of the particle in the tube can be regulated by creating a counterflow of heated gas, and its rate of motion is determined by the force of gravity and the resistance of the medium. The apparatus was used to study ignition of particles at long delay times.

Fig. 1. A: 1 –glass tube; 2 –heating coil; 3 –opening for introducing particles;

4 –heater; 5 –thermocouples; 6 –EPP-09 potentiometer. **B:** 1 –RS-5 rotameter; 2 –furnace; 3 –quartz tube; 4 –winding of the thermal screen; 5 –thermocouples; 6 –EPP-09 potentiometer; 7 –camera; 8 –opening for introducing particles

For short ignition-delay times an apparatus was used consisting of a horizontally arranged glass (quartz) tube through which gas, heated in an electric furnace, is passed (Fig. 1B). The tube has a lateral opening for introducing particles. This apparatus is similar to that proposed by V. A. Fedoseev for the study of heterogeneous combus-

formation and evaporation of particles (5). To eliminate the axial and radial temperature gradients present in this setup, which, in the case of ignition of homogeneous explosive particles, may substantially affect the delay time, the tube was fitted with a heat-shield winding heated by an electric current. The velocity of the gas flow in the tube is determined from the volumetric flow rate in the cold section, measured by a rotameter, with subsequent recalculation to the gas temperature in the tube. An explosive particle is introduced through a side opening in the tube and enters the stream of heated gas, which carries it along as it moves. The ignition delay time can be calculated from the transcendental equation:

$$t_{\text{ign}} = -\frac{2}{9}\frac{r_0^2\rho_0}{\eta}\ln\left[1 - \frac{9}{2}\frac{\eta}{r_0^2\rho_0}\left(t_{\text{ign}} - \frac{L}{v_0}\right)\right],$$

where t_{ign} is the ignition delay time; r_0 is the particle radius; ρ_0 is the particle density; η is the dynamic viscosity of the medium; v_0 is the velocity of the gas flow in the tube; L is the distance traveled by the particle up to the time t_{ign}. The equation was derived under the assumption that Stokes drag forces act. In the general case, an undoubted difficulty is the evaluation of heat exchange between the particle and the surrounding medium under the experimental conditions. However, for regimes $\text{Re} = vd/\nu \ll 1$, which occur for small particle sizes and low gas-flow velocities, the heat transfer between the particle and the gas may be taken as conductive (6), and, for the corresponding case,

$$\text{Nu} = \frac{\alpha_{\text{cond}}d}{\lambda_{\text{med}}} = 2 \qquad \left(\alpha_{\text{cond}} = \frac{\lambda_{\text{med}}}{r_0}\right).$$

Radiative heat exchange between the particle and the tube walls may be characterized by the quantity

$$\alpha_{\text{rad}} = \left[1 + \frac{T}{T_0} + \left(\frac{T}{T_0}\right)^2 + \left(\frac{T}{T_0}\right)^3\right]\varepsilon_1\sigma T_0^3$$

or, in the cases of interest to us, during heating and pre-explosive heating of the particle,

$$\alpha_{\text{rad}} = \psi\varepsilon_1\sigma T_0^3,$$

where $\psi = 1 \div 4$.

Thus, the total heat exchange will be determined by the quantity

$$\alpha = \alpha_{\text{cond}} + \alpha_{\text{rad}}.$$

In this case the Biot criterion, characterizing the ratio between internal and external heat exchange, is equal to

$$\text{Bi} = \frac{\alpha r_0}{\lambda} = \frac{\lambda_{\text{med}}}{\lambda}\left(1 + \frac{\psi\varepsilon_1\sigma T_0^3 r_0}{\lambda_{\text{med}}}\right).$$

Here v is the velocity of flow around the particle; d is the particle diameter; ν is the kinematic viscosity of the gas; α, α_{cond}, α_{rad} are heat-transfer coefficients; λ is the thermal-conductivity coefficient of the particle; λ_{med} is the thermal-conductivity coefficient of the gas; T_0 is the gas temperature; T is the particle temperature; ε_1 is the degree of blackness; σ is the Stefan–Boltzmann constant.

For the explosive–gas system, $\text{Bi} \simeq 0.05 \div 0.2$, i.e., it may be assumed that there is no temperature distribution within the particle. Calculations show that for flow velocities of about 2–3 m/sec, which occurred in our setup, the limiting value of the particle diameter for which $\text{Re} \ll 1$ is approximately 200 μ.

In the experiments, particles were selected under a microscope and introduced into the gas by means of a special needle.

Under thermal action in the given conditions, ignition of an explosive particle, depending on its size and the temperature of the surrounding medium, may proceed either in a self-ignition regime or in an ignition regime. The different ignition regimes are illustrated in Fig. 2. Self-ignition is characterized by a "round" flash, while ignition is characterized by a "track."

To the article by Yu. M. Grigor' ev, E. I. Maksimov, and A. G. Merzhanov

Fig. 2. Photographs of the ignition of spherical particles: **A** –pyroxylin ($T_0 = 300^\circ$ and $r_0 = 150\,\mu$) and **B** –barium azide ($T_0 = 300^\circ$ and $r_0 = 81\,\mu$) in the self-ignition regime; **V** –pyroxylin ($T_0 = 550^\circ$ and $r_0 = 150\,\mu$) in the ignition regime. In the photographs the winding of the thermal screen is visible.

To the article by E. N. Arnatskaya and T. N. Nesmeyanova, p. 1486

Fig. 1. Growth of nerve fibers through the region of transection of the dorsal root. Ganglion on the right. Silver impregnation. 100×

Fig. 3

Figure 2: Fig. 3

Fig. 4

Figure 3: Fig. 4

We studied the self-ignition of spherical* particles of barium azide in the temperature range 260-650°C. The thermal decomposition of barium azide at temperatures of 100-150°, studied in detail in works (7, 8), proceeds according to the equation:

$$BaN_6 = Ba + 3N_2 - 6.32 \text{ kcal.}$$

The activation energy of this process is 21-23 kcal/mole. However, because of its endothermicity, this reaction cannot lead to ignition. According to the investigations of K. K. Andreev (9), the decomposition of barium azide may proceed according to another overall equation:

$$4BaN_6 = Ba + Ba_3N_2 + 11N_2 + 71.0 \text{ kcal.}$$

The activation energy of this reaction is not given. In all probability, a similar process occurs under ignition conditions at high temperatures. The results of the experimental determination of the ignition delay times of barium azide particles are shown in Fig. 3. Each point is the average of 15-20 measurements. The reproducibility of the results for barium azide particles is fairly good (the greatest deviation from the mean value does not exceed 10%). The working medium was heated air. Special experiments carried out with heated nitrogen showed that the ignition delay times of barium azide in air and in nitrogen, all other conditions being equal, are practically identical. As can be seen from Fig. 3, the curves of the dependence of ignition delay time on temperature for particles of different size intersect; moreover, in the region of higher temperatures the delay times are the greater, the larger the particle size. The character of the dependence of ignition delay time on the temperature of the medium and on the particle diameter is in agreement with the conclusions of work (2), which considers ignition of the system with allowance for the heating stage.

Fig. 3. Dependence of the ignition delay time of barium azide particles of different size in the temperature range 280-650°

Fig. 4. Dependence $T_{0cr}(r_0)$.

a–$\varepsilon_1 = 1$, b–$\varepsilon_1 = 0$

Table 1 gives the critical self-ignition temperatures of barium azide particles of different size. The critical temperature T_{0cr} was taken as the mean value between

the temperatures of explosive and nonexplosive decomposition; the accuracy in determining the division is $\pm 2.5^\circ$.

The value of the critical self-ignition temperatures corresponding to different particle sizes makes it possible to determine the activation energy and the product of the thermal effect of the reaction by the preexponential factor of the overall exothermic decomposition of barium azide. For this purpose, taking into account the absence of a temperature distribution in the particle,

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* The spherical shape was imparted to the crystals by a special device, by blowing the crystals with air against fine emery paper.

one can use the thermal-explosion condition of N. N. Semenov [10]

$$\frac{Q}{\alpha S/V}\,\frac{E}{RT_{0\ \mathrm{cr}}^2}\,k_0 e^{-E/RT_{0\ \mathrm{cr}}} = \frac{1}{e},$$

where Q is the heat effect of the reaction; S/V is the ratio of the surface area to the volume of the particle; E is the activation energy; k_0 is the pre-exponential factor;

Table 1

$d,\ \mu$	50	75	87.5	125	162
$T_{0\ \mathrm{cr}}$, °C	302.5	292.5	284	272	260

R is the universal gas constant; e is the base of natural logarithms. Hence, taking into account that

$$\alpha = \frac{\lambda_{\mathrm{av}}}{r_0}\left(1 + \frac{\psi\varepsilon_1\sigma T_0^3 r_0}{\lambda_{\mathrm{av}}}\right), \qquad \frac{S}{V} = \frac{3}{r_0},$$

we obtain

$$\ln\frac{T_{0\ \mathrm{cr}}^2}{r_0^2}\left(1 + \frac{\psi\varepsilon_1\sigma T_{0\ \mathrm{cr}}^3 r_0}{\lambda_{\mathrm{av}}}\right) = \ln\frac{Qk_0Ee}{3R\lambda_{\mathrm{av}}} - \frac{E}{R}\,\frac{1}{T_{0\ \mathrm{cr}}}.$$

Figure 4 gives the dependence* $T_{0\ \mathrm{cr}}(r_0)$ in semilogarithmic coordinates, from which it follows that in the temperature range 260-305 $E = 35\,000$ cal/mole and $Qk_0 = 4 \cdot 10^{15}$ cal/cm^3 · sec (in the calculation $\lambda_{\mathrm{av}} = 10^{-4}$ cal/cm · sec · deg and $\psi = 4$ were used).

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* The results presented in Fig. 4 show that E and Qk_0 practically do not depend on the degree of blackness ε_1, i.e., in these experiments radiation has practically no effect on the critical conditions.

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