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TECHNICAL MEMORANDUM 1598

EFFECTS OF PRESSURE ON THE STABILITY OF COMBUSTION IN POROUS EXPLOSIVES

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JUNE 1965

TRANSLATED BY U. S. JOINT PUBLICATION RESEARCH SERVICE
FROM ZH. PRIKL. MEKHAN. I TEKHN. FIZ. NO. 5: 117-120 (1963)

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Zhurnal prikladnoy mekhaniki i tekhnicheskoy fiziki, Moscow, No 5, 1963,
pp 117-120.

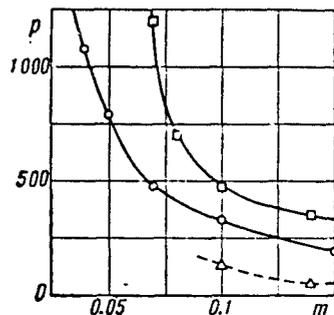
It was noted in [1] that in a porous explosive the penetration of combustion causes a rapid increase in the combustion surface and in the rate of gas generation, and thereby can result in an explosion.

K. K. Andreyev [2] showed that in the combustion of porous explosives the explosion occurs when a sufficiently high pressure is attained in the combustion process.

In the following we present quantitative data concerning the pressures at which the stability of combustion is disrupted in the case of pressed charges of RDX, PETN and TNT (the size of the initial powders being approximately 10 to 20 μ). The charges varied in porosity, $m = 1 - \rho/\rho_{\max}$. Here ρ is the density of the charge, and ρ_{\max} is the density of the monocrystal. The value of m indicates the proportion of the volume occupied by pores.

In the tests we burned in a manometric bomb significantly heavy charges of different porosity, in the shape of rods 10 mm in diameter. The rise in pressure was recorded piezoelectrically. So long as layer-to-layer combustion remained stable, the pressure versus time curve rose at an evenly increasing rate. A sharp break in the pressure versus time curve (up to the appearance of the compression shock) was an indication that layer-to-layer combustion had been disrupted, and that combustion had penetrated the pores. The pressure at which this took place was recorded directly.

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Some of the results we obtained are shown in the diagram. The abscissa represents the porosity m ; and the ordinate, the pressure p at which the combustion penetrates the pores. This pressure is measured in bars, and one bar is equal to 1.02 kg/cm^2 . The small squares represent TNT; the small circles, PETN; and the small triangles, RDX.

With rough approximation we may write $pm = A$. Here A is a constant that depends on the properties of the material, on the nature of its porosity, and on the test conditions. From a more detailed analysis of the test results it is evident that the vertical asymptote of the curves is $m \neq 0$. In the case of PETN, $p(m - 0.02) = A_1$; and for TNT, $p(m - 0.05) = A_2$. In practice this means that at $m = 0.02$ the combustion of PETN ($\rho = 1.735$) can be stable even at pressures measured in thousands of atmospheres. This was established [3] even for PETN of slightly lower density. At the same porosity (for example, 0.1) combustion is the most stable (and the pressure at which explosion occurs the highest) in the case of TNT, and the least stable in the case of RDX.

Taylor [4] noted that a molten layer must form in the combustion of PETN and RDX (and the more so in the case of TNT), and that this layer stabilizes the combustion until the thickness of the layer (decreasing with rising pressure) becomes smaller than the size of the pores. At an identical pressure, this layer must be the thickest for TNT; and the thinnest, for RDX.

In the case of PETN,* the thickness x of the molten layer can be expressed as

$$x \approx \frac{\lambda}{c\rho u} \ln \frac{T_* - T_0}{T^0 - T_0} \quad (1)$$

Here λ is the heat conductivity of the liquid, c is the specific heat, $u\rho$ is the mass combustion rate [5], T_* is the critical temperature of PETN, T^0 is the melting point, and T_0 is the initial temperature.

For PETN, under the conditions of the experiment, at $p = 330$ bars and $m = 0.1$,

$$\frac{\lambda}{c} \sim 10^{-8} \frac{\text{e}}{\text{cm}^2 \text{sec}}, \quad u\rho \approx 6.4 \frac{\text{e}}{\text{cm}^2 \text{sec}}, \quad T_* \sim 580^\circ, \quad T^0 \sim 140^\circ, \quad T_0 \sim 20^\circ \text{C}$$

*For rough approximation we may take as a limit $\lambda/\rho cu$ (the characteristic length of the heat wave), the distance at which the temperature dependence drops e times.

Substituting these values in (1), we find that x equals a few microns (approximately 2-3 μ). Let us assume that at the boundary the diameter d of the pores is equal to the thickness of the molten layer, $d \approx x$. (If $d > x$, the combustion penetrates the pores; if $d < x$, there is no penetration.) Thus we come to the conclusion that in the case of PETN, at a pressure p approximately equal to 330 bars, the combustion is able to penetrate pores that are a few microns in size.* It is possible to evaluate also the mean hydraulic pore size [6], $D \sim \sqrt{k/m}$, where k is the gas permeability. For PETN when $m = 0.1$, we find that $D \sim 0.01-0.1 \mu$, which is one to two orders of magnitude smaller than d . The explanation of the obtained relationship ($d \geq D$) is natural. The gas permeability is determined for all pores, including the smallest ones. In the initial stage, the combustion penetrates the largest pores (or the one largest pore). Obviously, the distribution of pores by size is such that the largest pores ($d \sim x$) are larger by one to two orders of magnitude than the mean hydraulic pore size D .

With rising pressure, the velocity u increases ($u \sim p$), the thickness x of the molten layer drops, and the combustion is able to penetrate pores of gradually smaller size:

$$d \sim x \sim \frac{1}{u} \sim \frac{1}{p}, \quad \text{or} \quad pd = c \approx \text{const}$$

This last relationship is satisfied approximately. With increasing pressure, the size of the pores or voids that the combustion can penetrate will always become smaller, $d \sim 1/p$. In various materials, the combustion penetrates the pores at different pressures. But if the combustion is stabilized by the molten layer, and if the charges are identical in structure, then the thickness of the molten layer should be similar. Computations show that in the case of RDX (at $p = 130$ bars) as well as in the case of PETN (at 330 bars), the thickness of the molten layer is a few microns. In the case of TNT (at 480 bars) the layer is thicker, and the combustion should be penetrating the pores at approximately twice that pressure (~ 800 bars). Obviously, at the same porosity ($m = 0.1$), the structure of the TNT charge is somewhat different. By measuring the gas permeability it was directly established that the mean hydraulic pore size is 1.5 times larger in the case of TNT than in the case of PETN (at the same porosity, $m = 0.1$). Evidently, the largest pores of TNT are twice larger than the largest pores in the case of PETN. Incidentally, at lower porosities (0.07-0.08) the charges of PETN and TNT are more similar in structure.

These data show good agreement with Taylor's assumption [4] concerning the stabilizing action of the molten layer. Adreyev's opposite assumption [7] -- i.e., that the self-turbulence of the molten layer may be disrupting the stability of combustion in the case of PETN -- was not evident in our

*If the actual thickness of the molten layer is more than the thickness computed on the basis of equation (1), the critical pore size will be larger.

tests. (It must be admitted, however, that our experimental conditions were basically different.)

Let us examine the case when the combustion of a porous explosive does not form a molten layer. It would be false to assume that in the case of a nonmelting explosive the combustion will penetrate the pores at any (small) pressure. In such cases the combustion will be accompanied by the flow of combustion products into the pores where these products will ignite the material. Both these factors will depend on the conditions of combustion and on the distance x' between the surface of the material and the zone of intensive reaction in the gaseous or fume-gas phase. This distance

$$x' \approx \frac{\lambda'}{c'(u\rho)} \ln \frac{T^* - T_0}{T^0 - T_0} \quad \left(\frac{\lambda'}{c'} \sim 5 \cdot 10^{-4} \frac{s}{cm \cdot sec} \right) \quad (2)$$

Here λ' and c' are the heat conductivity and specific heat of the gaseous phase, T^0 is the surface temperature, T^* is the temperature of intensive reaction in the gaseous phase (in most cases T^* will not be the maximum combustion temperature). The other symbols are the same as in equation (1). In a rough approximation, the value of x' is somewhat smaller than the thickness x of the molten layer. Furthermore, the viscosity of the gas is considerably lower than the viscosity of the fluid, which should enhance gas penetration of the pores. In sum, the penetration of combustion should be easier in the absence of the molten layer. (At identical porosities, the combustion should penetrate at pressures several times lower; at identical pressures, it should penetrate pores of d' that is several times smaller.) In a rough approximation, the condition that $pd' = (\text{const})'$ also should be satisfied, while $(\text{const})' < \text{const}$.

Exploratory tests with nonmelting explosives showed that these conclusions generally hold true, and specifically that the combustion penetrates the pores more easily than in the case of melting explosives. It should be noted that the break in the pressure versus time curve (an indication that the combustion has penetrated the pores) is somewhat blurred and less pronounced.

As noted in the dissertation* of the author of [8], pore penetration should be particularly easy during the combustion of mercury fulminate (a primary explosive). In fact, if at a pressure $p \approx 100$ bars, a pore size d of a few microns is needed for penetration in the case of PETN, then the value of d for mercury fulminate must be at most in the order of 0.1μ . The value of d must be even smaller in the case of lead azide. At one time [8], on the basis of indirect data, the combustion rate of lead azide was assumed to be so high that the combustion itself (without penetration) causes the considerable compression shock, and thereby the detonation. In their article [9], K. K. Andreyev and B. N. Kondrikov gave a more substantiated esti-

* A. F. Belyayev, Mekhanizm goreniya vzryvchatykh veshchestv (Mechanism of Combustion in Explosives), doctorate dissertation at the Institute of Chemical Physics, Soviet Academy of Sciences, 1946.

mate of the combustion rate for lead azide. According to their estimate, the combustion rate of lead azide is only two to three times faster than the combustion rate of mercury fulminate. To explain the extremely sudden detonation of lead azide (it detonates upon ignition) at this rate of combustion, we must assume that at the moment of ignition (at any pressure) the combustion immediately penetrates the charge, and that actually the combustion surface is considerably larger.

Here we should note that if the combustion mechanism of lead azide is similar to that of mercury fulminate, then at low pressure (and partially at atmospheric pressure) the combustion must be able to penetrate the lead azide pores whose size is in the order of 1.0μ . Such pores can form between particles even when the lead azide is compressed to a high density. Furthermore, in the case of lead azide crystals in the order of one micron, there may be voids (microscopic pores and gaps) between crystals. To this we must add that, according to direct observations by Bowen and his associates [10], defects in the crystals may arise or increase even during combustion. Finally, in the combustion of lead azide the dynamic rise in temperature must be very significant. All these factors indicate that in the case of lead azide there are many possibilities for the intensive penetration of combustion, for a sharp increase in the combustion surface, i.e., for an instantaneous explosion that immediately goes over into a detonation.

K. K. Andreyev [11] noted that in the case of primary explosives combustion must be able to penetrate the pores more readily, mainly because the explosion reaction is more complete, and the combustion temperature is much higher. The value of the combustion temperature is unquestionably essential (it is one of the factors that determine the combustion rate), but the main reason why combustion is able to penetrate the pores of primary explosives more readily is the small distance between the surface of the explosive and the zone of intensive reaction. Under otherwise identical conditions, this distance is considerably smaller in case of primary explosives than in the case of secondary ones. The high temperature of the combustion products unquestionably enables combustion to penetrate the pores more easily, but more important in this respect is the value of the temperature gradient dT/dx at the surface. In primary explosives this temperature gradient is two to three times higher than in secondary explosives, basically because of the smaller heat generating zone, the size of which in the final outcome depends on the nature of the combustion reaction and on the corresponding constants of the explosive.

Returning to an overall evaluation of the observed phenomenon, we wish to note that combustion penetration of the pores is a very complex problem, and that here it was examined in a simplified form. Combustion penetration of the pores and the nature of this combustion depend on the physical and chemical properties of the materials (our experiments were made with explosives that react predominantly in the gaseous phase [12]), on the conditions

of combustion, on the gas dynamics of the combustion products, and on the type of porous structure. For example, if we take an explosive consisting of fairly large crystals, its gas permeability will be higher, and the size of the largest pores will increase even more significantly. In sum, the penetration of combustion will be easier. As we have noted above, the tests were made in a manometric bomb, at significant ratios of dp/dt and considerable pressure differentials dp/dx .

Under other test conditions, the results may prove different. For example, in a bomb of constant pressure, the stability of combustion will increase: the pores will be filled with compressed inert gas. Nevertheless, this simplified investigation has enabled us to obtain useful results.

Institute of Chemical Physics
Soviet Academy of Sciences

Submitted: 25 April 1963

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4. DESCRIPTIVE NOTES <i>(Type of report and inclusive dates)</i> Translation of Russian Journal Article		
5. AUTHOR(S) <i>(Last name, first name, initial)</i> Belyayev, A. F. Sulimov, A. A. Korotkov, A. I.		
6. REPORT DATE June 1965	7a. TOTAL NO. OF PAGES 9	7b. NO. OF REFS 12
8a. CONTRACT OR GRANT NO. b. PROJECT NO. c. d.	9a. ORIGINATOR'S REPORT NUMBER(S) Technical Memorandum 1598	
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