

INITIAL PHASE OF DETONATION INITIATION IN PRESSED TROTYL

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A focal mechanism is now generally accepted for the excitation of detonation by weak shock waves in physically inhomogeneous solid explosives. However, there have been open questions concerning the causes of the reaction foci, the time and place of nucleation, the dimensions and concentration of the foci, the course of the reaction at a focus, and the interactions of foci. Unfortunately, it is difficult to examine by experiment the micromechanisms responsible for nucleation and development of foci of fast reactions, and until now we have had only indirect information obtained by various integral methods.

There are generally available methods of determining the critical parameters of shock waves that can initiate reactions in explosives, which are capable of strengthening the propagating waves under certain conditions up to a detonation level, which enables one to compare the susceptibilities of various explosives to shock-wave excitation. Internal methods can be employed such as the electromagnetic measurement of mass velocity or pressure recording with manganin transducers, which means that the average gasdynamic parameters of the flow behind a planar shock wave can be used to judge the start and place of the energy release [1-3] and in some cases even the rate of decomposition [4, 5].

In studies in this area, one usually examines the evolution of the mass-velocity profile u or the profile of the pressure p in the shock wave up to the point where the normal detonation front is produced; one uses a large number of measurements in various sections of the charge for a given intensity of the initiating shock wave. The wave remains planar only for a short period on account of the unloading effect of the side surface of the specimen, so one commonly employs shock waves of intensity considerably in excess of the critical value.

Measurements on the shock compressibility of explosives [6, 7] as a rule are performed with large steps in the shock-wave intensity, and in some cases in the range of pressures exceeding the detonation-initiation limit on the assumption that the explosive does not have time to decompose substantially at the shock front. There has been no detailed study of the behavior of an explosive under shock-wave compression that causes chemical reaction, which hinders elucidation of the initiation mechanism. Here we examine the initial stages of the explosive transformation of pressed trotyl near the limits of ignition initiation and detonation in response to planar shock waves of stepped profile.

Figure 1 shows the system. The plane-wave shock-wave generator was an explosive gun (internal diameter of the barrel 1 is 50 mm, and 2 is the piston). The speed of the duralumin striker 3 of thickness 9.2 mm was measured with the two contact transducers 4 with an error of not more than 0.5%. The velocity can be predicted with an error of 3%. A pressure pulse of initial amplitude from 10 to 25 kbar was applied to the specimen 6 via the duralumin screen 5 of thickness 5 mm. The specimens were of density 1.56 g/cm^3 and were prepared by pressing crushed trotyl, which was a polydisperse powder with particle sizes from 1 to $400 \mu\text{m}$ (individual particles). The two manganin pressure transducers 7 were installed one at the screen-specimen boundary and the other within the specimen at a distance of 6 mm from the screen in order to examine the behavior of the shock wave. The transducers were made by photoetching from manganin foil of MNMtsAZh3-12-0.25-0.2 grade of thickness 0.03 mm. The sensitive element in the transducer occupied an area of $4 \times 4 \text{ mm}$, and the initial resistance was 3.5Ω . The transducers were isolated from the specimen and the screen with PTFE films of thickness $60 \mu\text{m}$. The recording circuit has been described in detail [8]. The pressure profiles were drawn up from the pressure dependence of the relative change in resistance of the manganin [9].

Figure 2 shows typical waveforms. The amplitude of the initiating shock wave was determined from the pressure profile at the screen-specimen boundary as extrapolated to $t=0$. There was not more than 3% spread in the pressures at the front for a given collision velocity as determined from five experiments. The error in determining the absolute value of the pressure was about 5-10%, which included the error in determining the calibration curve. The pressure fall corresponds to the arrival of the decompression wave from the rear side of the striker. The first transducer regularly recorded the elastic precursor in the duralumin, whose ampli-

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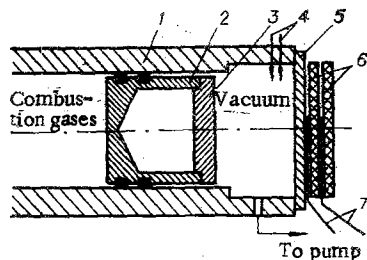


Fig. 1

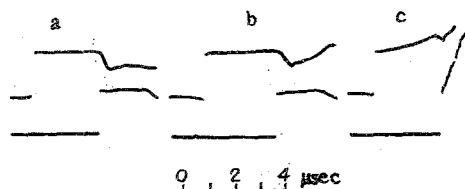


Fig. 2

Fig. 1. Method of recording the shock-wave evolution in pressed trotyl.

Fig. 2. Waveforms for pressure profiles at the boundary between screen and specimen (upper) and at 6 mm from the screen (lower) for pressures (kbar) of: a) 11; b) 16; c) 25.

tude was 1-3 kbar, which was 0.1-0.15 μsec ahead of the plastic wave, which agrees with the known longitudinal velocity of sound in duralumin and with the dimensions of the screen. The second transducer did not record any characteristic features in the structure of the wave front, which indicates that the first pressure step at the front dies away rapidly as the wave propagates, and the wave does not split up in trotyl in the range 10-25 kbar. Apart from these pressure measurements at two points, the experiments allowed us to determine the shock wave speed D averaged over the baseline and the speed of the decompression wave.

We constructed the shock-compression curve for pressed trotyl in p - u coordinates from the measured speed of the striker, the pressure at the shock front at the screen-specimen boundary, and the known shock-wave adiabatics for the striker and screen [10]. The weak-wave approximation was used in constructing the unloading isentrope for the screen, and the intersection of this with the isobar corresponding to the initial amplitude of the shock wave gave the point on the shock-wave adiabetic for the pressed trotyl.

The shock-wave adiabetic constructed from these points is shown in Fig. 3, curve 1, which has a slight S-shaped character, and correspondingly may be divided into three parts: the lower branch up to 13 kbar, the transition region 13-21 kbar, and the upper branch above 21 kbar. This figure also shows the pressure profiles in two sections for four different shock-wave intensities.

Some valuable conclusions can be drawn from the parts of the shock-wave adiabetic and the corresponding pressure profiles. The profiles for the lower part of the curve are always of rectangular form with equal amplitudes in both sections, which shows that there are no chemical reactions in the trotyl capable of amplifying the wave during the observation time, i.e., the lower part of the curve corresponds to shock compression of an inert substance. Above some particular pressure, in the present case 13 kbar, the start of the transitional part, the wave begins to be amplified from section to section. This at once means that the flow behind the front is reacting and that a fast chemical reaction has been initiated in the trotyl. In [11], the onset of chemical reaction in pressed trotyl at about the same shock-wave density was specified as the initiation limit for TNT.

More detailed consideration of the pressure profile corresponding to the transition part of the curve indicates that there are two modes of shock-wave amplification: increase in the pressure amplitude at the front from section to section and formation of a compression step in a broad zone behind the front, which catches up with and strengthens the head part of the wave. At low shock-wave intensities, this compression step is produced at a considerable distance from the front or may be blocked by the decompression wave arriving from the rear side of the striker, in which case the rising pressure at the front cannot be explained by energy transfer from the compression wave to the front. The rise at the front can be explained only by assuming that there is energy production in the region directly adjoining the shock front. In Fig. 3 the pressure amplitude at the front increases from section to section for a shock-wave intensity of 15 kbar without appreciable distortion of the pressure profile behind the front. The compression step is not observed on account of the smallness of the display time, and also it is blocked by the rear decompression wave, so amplification of the wave from behind is unlikely. Therefore, the amplification of the wave at the front is due to reaction in the material in a time comparable with the transit time of the front width and less than the time resolution of the method (<100 nsec). If this is so, the measured shock-wave adiabetic is an indirect reflection of the extent of chemical reaction.

If we assume a monotone shock-compression curve for an inert substance, it is reasonable to extrapolate the lower branch of curve 1 to higher parameters by treating it as a hypothetical shock-wave adiabetic for

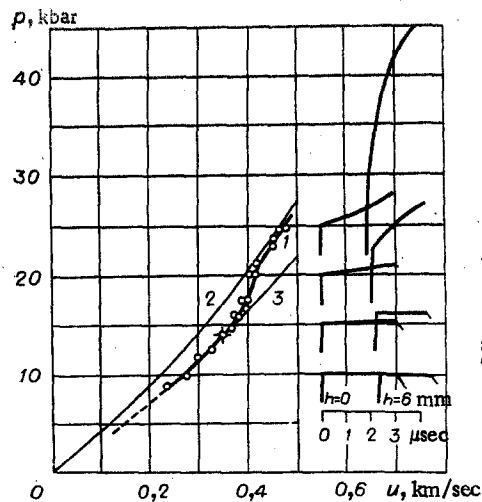


Fig. 3

Fig. 3. The $p-u$ relationship for trotyl and evolution of shock waves of different intensities; 1) observed shock-wave adiabat for pressed TNT with $\rho_0 = 1.56 \text{ g/cm}^3$; 2) shock-wave adiabat for cast trotyl with $\rho_0 = 1.614 \text{ g/cm}^3$; 3) supposed shock-wave adiabat for pressed trotyl with frozen reactions.

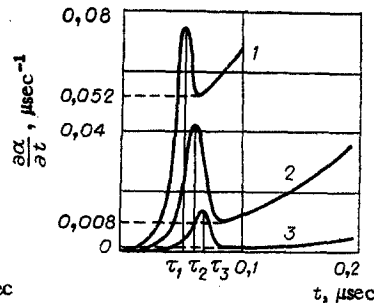


Fig. 4

Fig. 4. Scheme for the initial stage of decomposition in TNT.

pressed trotyl with the chemical reactions frozen (curve 3 of Fig. 3). Curve 3 may be described by the linear relation $D = (1.92 + 1.7 u) \text{ km/sec}$, while curve 2 is described by $D = (2.3 + 2.15 u) \text{ km/sec}$ by averaging the data of [2, 7]. Curve 2 is given as an example of a shock-wave adiabat for a substance (cast trotyl) whose detonation-initiation limit lies considerably above the working pressure range. The shapes of curves 1-3 indicate that the differences in position of curves 1 and 3 will correspond to the energy release at the step on the basis that the measured shock-wave adiabat provides information on the conversion step at the shock front.

As the shock-wave intensity increases, the compression step begins to be produced closer and closer to the head of the shock wave, and the initiation is determined in the main by the rate of energy production throughout the region behind the front, not in the region directly by the front (Fig. 3, $p_{\text{sw}} = 20 \text{ kbar}$). When we reach the upper part of the curve ($p_{\text{sw}} > 21 \text{ kbar}$), we find that the compression step begins to be formed directly behind the shock front, which leads to very rapid acceleration of the wave. No detailed study was made of the final stage of conversion of an initiating shock wave to a detonation wave. In [11], the same pressure range ($p > 17$) was called the detonation-initiation limit for pressed trotyl ($\rho_0 = 1.56 \text{ g/cm}^3$).

The pressure profiles and the shock-wave adiabat for pressed TNT provide at least two important conclusions. Firstly, there are two modes of wave amplification, and secondly there is a nonmonotone heat release by chemical reaction at the shock front. In the latter case, the heat is produced in a very narrow zone comparable in width with the front. Here it is reasonable to consider what physical meaning attaches to the term shock front. In a physically inhomogeneous explosive, a shock front is substantially three-dimensional, in contrast to a homogeneous explosive, and the width can be estimated to a first approximation as proportional to the size of the explosive particles. This zone has the maximum gradients in the velocity, pressure, density, and temperature, and the directions of these may not coincide with the direction of the material flow. For this reason there may be marked local heating, with the temperature rise greatly exceeding the mean value over the flow.

The two-stage rise in the gasdynamic parameters should correspond to a two-stage reaction in the macrokinetic sense. If we assume that the inhomogeneity causes local temperature rises at the shock front on account of viscous friction between the grains, then the thickness of the heated layer will be determined primarily by the friction mechanism, which in turn is dependent on the physical properties of the explosive and the shock-wave intensity. The narrow zone with high gradients exists for a very short time, and we can therefore show that the temperature rise at the points of contact is adiabatic. If the temperature in these foci is sufficient at the given shock-wave intensity to cause ignition during the time the wave is multidimensional, then there will be amplification of the shock wave. If this is not so, there will be thermal dissipation of the focus.

TABLE 1

No. of curve in Fig. 4	P_{sw} , kbar	τ_r , μsec	α_1 , %	$\left(\frac{\partial\alpha_{II}}{\partial t}\right)_0$, μsec^{-1}
1	25	0,060	0,2	0,052
2	19	0,062	0,1	0,008
3	10	0,071	0,01	0,0014

By increasing the intensity of the initiating shock wave, one can always attain the state where there is thermal ignition of the individual hot spots within the characteristic time of the multidimensional front. In the present case this intensity is about 13 kbar. There is then burning in the layer heated by the friction at a high rate (explosive burning), after which there is contact between the burning front and the cold core of the grain, which results in extinction or retardation of the reaction to a rate that in turn is limited by heat transfer.

Another factor governing the determination of the reaction in the first stage may be gasdynamic unloading of the focus, i.e., wave pulsations spreading through the grains produce cracks in the zone of the focus, through which the hot decomposition products escape, thereby reducing the temperature and pressure at the focus. The number and temperature of the foci increase with the shock-wave intensity, and this is associated with a reduction in the ignition delay. The proportion of the explosive decomposing, and therefore the heat production going to amplify the front, will increase, while the probability of complete extinction of a focus on arrival of the unloading wave will be reduced (Fig. 3, $p = 13$ -20 kbar, where curve 1 increasingly diverges from curve 3). When the pressure in the initiating shock wave attains 20-22 kbar, the concentration of foci becomes so large that they interact before the unloading wave arrives, i.e., the shock front ignites virtually the entire surface of the explosive particles.

Any further rise in the shock-wave intensity produces little further increase in the rate of heat production in the zone, and therefore the divergence of curves 1 and 3 (Fig. 3) becomes slower, because the concentration of foci has become maximal, and the increase in heat production in the foci is determined by the increase in the parameters in the foci themselves and probably also by crushing. This will explain the second inflection on curve 1.

Further chemical reaction and the main heat production occur in a broad zone behind the shock front, which develops from the reactive foci, i.e., foci in which the reaction has not died away completely but continues as burning of the grains from the surface, with the rate limited by heat transfer. Fairly convincing evidence of this mechanism has been given in [11, 12]. If the concentration of foci is less than maximal, there is an expansion of the burning surface within a focus and ignition of new surfaces by expanding hot products with some delay. When the burning extends to the entire surface area of the explosive grains (directly behind the shock front at the maximum focal concentration or after some time behind the front at a lower concentration), one gets burning of the particles from the surface in an essentially closed volume equal to the initial volume of a particle if we neglect the residual porosity behind the front and the combustion of the layer of explosive in the first stage. Two competing factors then influence the decomposition rate: the growth in pressure in the closed volume and the reduction in the burning surface as the decomposition proceeds. Of course, all this has been discussed in the absence of a decompression wave arriving from the surface of the charge, which may cause partial or complete suppression of the reaction and may retard or inhibit the development of detonation in accordance with the rate and extent of the decomposition.

The above is illustrated by Fig. 4 and Table 1, where τ_r is the reaction time in the first stage. It is impossible to determine the combustion time of the heated layer of trotyl at a hot focus even approximately, so τ_r was defined as the time required for the shock wave to pass through a layer of explosive of thickness 100 μm and return of the unloading wave at the corresponding shock-wave intensity; α_1 is the degree of reaction of the substance in the first stage, which was calculated as the increment in the specific internal energy on going from the state on curve 3 to the state on curve 1 in Fig. 3:

$$\Delta E_1 - \Delta E_3 = 1/2 \cdot p_1(v_0 - v_1) - 1/2 \cdot p_3 \cdot (v_0 - v_3).$$

There $(\partial\alpha_{II}/\partial t)_0$ is the initial rate of decomposition of TNT in stage II as calculated by the method of [13] from the observed shock-wave pressure profile at the contact boundary and the time dependence of the shock-wave speed.

Figure 5 shows $(\partial\alpha_{II}/\partial t)_0$ in relation to shock-wave intensity. The values given for the decomposition parameters in Table 1 are crude estimates and appear to reflect only the qualitative side of the process. The

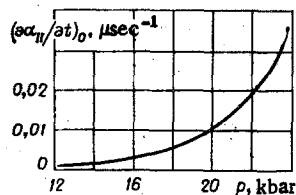


Fig. 5. Effects of shock-wave amplitude on the initial rate of decomposition of trotyl behind the front.

curves for the decomposition rate of TNT in the first stage have been drawn up on the basis of an exponential dependence of the reaction rate on time in such a way that the area under the curve was equal to the extent of decomposition in the first stage. The initial part of the curve for the decomposition rate in the second stage has been constructed from published data on TNT [5].

There is reasonable agreement between the initial rate of decomposition behind the shock front for the range $p > 21$ kbar and the rate of decomposition calculated from the linear rate of combustion of TNT as derived from an extrapolation formula in [14], $u = 0.25 + 45 p$ mm/sec, where p is in kbar. Figure 5 gives $(\partial\alpha_{II}/\partial t)_0 = 0.043 \mu\text{sec}^{-1}$ for $p_{\text{sw}} = 24$ kbar. The decomposition rate determined from the combustion rate for spherical particles burning over the entire surface is $0.051 \mu\text{sec}^{-1}$ for particles of size $50 \mu\text{m}$, as against $0.03 \mu\text{sec}^{-1}$ for particles of size $100 \mu\text{m}$.

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