

A Simple Approach for Modelling Reaction Rates in Shocked Multi-Component Solid Explosives

B.S. Ermolaev,

N.Semenov Institute of Chemical Physics, RAS, Moscow, Russia

B.A Khasainov*, H.-N. Presles and P. Vidal

Laboratoire de Combustion et de Détonique, UPR CNRS 9028, ENSMA, Poitiers, France

Abstract

We present a theoretical model to analyze experimental data on low velocity detonation (LVD) and to estimate the chemical conversion rate for ammonium nitrate and its mixtures with aluminium in the 0.5 – 3 GPa pressure range. When applied to high explosives, our model results in a Z-shaped curve for the velocity – curvature relationship. This leads to a dependence of the detonation velocity on the charge diameter in the form of two separate branches for low-velocity and normal detonations, which can be used to explain the transient delayed processes observed under shock initiation, including the so-called delayed detonation phenomenon.

Introduction

Many multi-component explosives and rocket propellants have so low reactivity under nominal conditions that tests used to evaluate their response to unforeseen stimuli or attacks are not suitable because of too small device sizes. On the other hand, large-scale experiments are often too expensive. Multi-dimensional numerical modeling can be thought as an alternative to money and labor-consuming experiments. However, its predictive ability remains limited. One of the reasons is the lack of information on chemical conversion rates of explosive compositions and their components, especially in the 0.5 – 10 GPa pressure range which plays a key role in shock-induced transient processes.

In this work, we attempt to infer these data from a numerical analysis of low velocity detonations (LVD). This process is a special mode of the supersonic propagation of exothermic reaction in an energetic material, showing the same properties as a normal Chapman-Jouguet (CJ) detonation except that the LVD velocity is several times smaller than the CJ detonation. Information on LVD is available for many condensed energetic materials. LVD attracts attention because its characteristics (with propagation velocities from 0.8 up to 2.5 km/s) fall in the aforementioned pressure range. Besides, LVD properties can be easily studied or changed by varying the charge diameter and confinement in laboratory-scale tests.

Our analysis describes LVD as a particular kind of steady nonideal detonations. A model of LVD is developed in the quasi-one-dimensional approximation for weekly diverging flow in multiphase reactive medium. We assume that chemical reaction proceeds in the surface burning mode. This model is used here for numerically interpreting several series of experimental data. We have analyzed our measurements of LVD in AN prills confined in thin steel tubes of different diameters [1], as well as the data of other authors available in the literature on LVD and nonideal detonations in AN in confined and unconfined charges

[2,3]. By comparing calculations with experimental results, we have estimated the chemical conversion rate of AN and its pressure dependence in the 0.5 – 3 GPa pressure range. We also have analyzed the literature data on LVD and nonideal detonations in mixtures of AN with aluminium (Al) [3] and found out that AN burns faster in the mixtures.

We have conducted a similar analysis for loose-packed Tetryl. This high explosive, in contrast to AN and its mixtures, exhibits a dependence of its detonation velocity on the charge diameter in the form of two separate branches for low-velocity and normal detonations [4]. We have estimated the chemical conversion rate of Tetryl and obtained a Z-shaped velocity – curvature relationship. We also have conducted an illustrative dynamical calculation to demonstrate that the Z-shaped relationship can explain some transient delayed effects observed in shock initiation of high explosives, such as delayed detonation which, to date, had no clear interpretation [5].

Specific Objectives

This study aims at obtaining information on the chemical conversion rates of solid explosive compositions and their components under dynamic pressures of several GPa. The lack of information on chemical conversion rates significantly restricts the predictive ability of multi-dimensional numerical modeling. The base of our study is a theoretical analysis of experimental data on LVD. The model, developed to analyze LVD, can be useful for studying safety aspects of modern explosive compositions and propellants.

Model

Our model describes LVD as a particular steady nonideal detonation with effective energy losses due to lateral expansion. We use the classical quasi-one-dimensional approximation [6] for a weekly diverging flow in multiphase reactive medium [7]. We consider the steady-state reaction zone in a self-sustaining

* Corresponding author : Boris.Khasainov@lcd.ensma.fr
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detonation wave that propagates with a constant velocity D along the axis of a cylindrical charge of explosive material of radius r^* . The reactive medium is a mixture of the initial components and of the products of their final chemical conversion. For instance, in the case of an explosive composition made of AN and Al, the reactive mixture includes 4 species: (1) the solid AN, (2) the products of AN chemical conversion, (3) the solid Al and (4) the products of Al burning in the AN products. The state of the reactive medium is determined by the mass fractions of these 4 species (η_i) with the mixture density and internal energy calculated from the species density (ρ_i) and internal energy (e_i) by means of the usual additivity rule. We assume that the species are in local mechanical equilibrium (identical pressure P and material velocity U), and that species (2) and (4) have identical temperature which, however, differs from the temperatures of the solid species.

The chemical reaction is assumed to proceed in the surface burning mode. In this case the conversion rate could be expressed as the product of the specific surface and of the burning rate. However, in the LVD context these two parameters are unknown. Therefore, in our model, the chemical conversion rate is described as the function of species contents and pressure, which includes a scaling factor applied as a fitting parameter. The intensity of the chemical conversion of the explosive material per unit volume is thus given by

$$M_1 = \rho_{10}(\eta_1/\eta_{10})^{2/3} G_1 (P/P_r)^n [1 + F(P/P_r)^k] \quad (1)$$

where ρ_{10} and η_{10} designate the TMD and the initial mass fraction of the species (1). The pressure parameter P_r is introduced for convenience in order to express the burning coefficient G_1 in sec^{-1} irrespective of the pressure exponent n and is set to 1 GPa. We assume a degressive burning with exponent set to 2/3. Usually, (except for mentioned cases), the coefficient F is set to zero, hence, only two parameters G_1 and n are varied to obtain the best agreement between calculations and experiment.

The relation used to calculate the intensity of Al burning is

$$M_3 = \rho_{30} G_3 (\eta_3/\eta_{30})^{2/3} (P/P_r)^r \left(\frac{C_{ox}}{1 + \eta_4/\eta_2} \right)^s \quad (2)$$

where the exponent for Al content is again set to 2/3. The coefficient C_{ox} designates the oxygen content in gas components of the species (2) that participate into Al oxidation. The multiplier comprising C_{ox} accounts for the dependence of the Al burning rate on concentration of oxidizing gases. In the case of the AN + Al mixtures we have set $C_{ox} = 0.71$ and $s = 1.0$. Because there is no definitive agreement on the pressure effect on the Al burning rate at elevated pressures, we have set the pressure exponent r to 0. Thus, only the coefficient G_3 of the Al burning rate is used as an adjustable parameter.

The surface burning follows the chemical reaction initiation at hot spots generated in the shock wave front. Theoretical considerations [8] demonstrate that the hot spot ignition delay under shock loadings of a porous

high explosive quickly drops with increase of the difference between the front pressure P_f and the threshold pressure P_{ign} of the chemical reaction initiation. Thus, one can assume that burning begins just after the shock wave front, so the ignition delay can be ignored. However, the amount η_{2f} of high explosive consumed during the hot spot process needs to be estimated. Here, we assume that this amount is very small and therefore set η_{2f} to 0.01 in our calculations for LVD in AN. Note that a non-zero value for η_{2f} parameter eliminates the initial point singularity. Preliminary calculations made with different η_{2f} have shown no significant effect on the integration results.

The equations of the model include the balance equations for mass, momentum and energy of the mixture, EOS, balance equations of mass and energy of species, and relationships connecting the internal energy and density of the mixture with the internal energy, density and mass fraction of species. By means of the steady-state assumption, these equations have been reduced to an ODE system relating the space derivatives of the variables and the local values of these variables. To explain the properties of these ODEs, we can consider the only equation for particle velocity in the simple case of a two-species reactive medium composed by a high explosive without additive:

$$\frac{dU}{dx} = \frac{M_1 \frac{[\kappa_2(1 - \rho_2/\rho_1) + \Gamma_2(e_1 - e_2)]}{(\kappa_2\rho_2 + P\Gamma_2)} - \frac{UdS}{Sdx}}{(1 - U^2/C^2)} \quad (3)$$

where $x \geq 0$ is the distance from the shock front in the direction normal to this front and Γ_i is the Gruneisen coefficient. The functions $\kappa_i = \partial P / \partial \rho_i |_{e_i}$ ($i=1,2$) are obtained by differentiating the EOS of the species, S is the cross section area of the flow tube and C designates the sound speed:

$$C = \left\{ \rho^2 \left[\frac{\eta_1}{\rho_1(\rho_1\kappa_1 + P\Gamma_1)} + \frac{\eta_2}{\rho_2(\kappa_2\rho_2 + P\Gamma_2)} \right] \right\}^{-1/2} \quad (4)$$

The EOS for solid explosive and its detonation products are chosen in the form

$$P = \Gamma_1 \rho_1 (e_1 - e_{01}) + B_1 \left[\left(\frac{\rho_1}{\rho_{10}} \right)^l \left(1 - \frac{\Gamma_1}{l-1} \right) - \Gamma_1 - 1 + \frac{\rho_1}{\rho_{10}} \frac{\Gamma_1 l}{l-1} \right] \quad (5)$$

$$P = \Gamma_2 \rho_2 (e_2 - e_{02}) + B_2 \rho_2^m (1 - \Gamma_2 / (m-1)) \quad (6)$$

The coefficients in (5) are determined from the literature data on Hugoniot and Gruneisen coefficient. The coefficients in (6) are adjusted to get a best approximation of the CJ detonation parameters. These were calculated with the TDS thermodynamic code [9] using BKWC EOS, the initial densities of explosive material having been varied to get the detonation pressure range expected in the LVD process.

The numerator in (3) comprises two terms: the first one is responsible for the burning effect and the second one for the flow divergence effect. To approximate the latter, we have used two different estimates, depending

on whether the explosive charge is confined or unconfined. For unconfined charges, we assume that the wave velocity in the direction normal to the front is a function of the local front curvature (K), that the radius of the front curvature is much greater than the reaction zone length, and that the flow in the reaction zone can locally be approximated as a spherically symmetric one. These assumptions yield the following result [10]:

$$d(\ln S)/dx = (D/U - 1)K/(1 - Kx/2) \quad (7)$$

For charges confined in steel tubes the latter assumptions are not suitable. The first reason is that the sound speed in steel is higher than the LVD velocity, a case for which a theoretical steady-state solution does not exist. However, the experimental observations [11] show that the process is governed by inertial and strength properties of confinement. The second reason is that steel confinement considerably reduces the charge diameters in which LVD is observed. As a result, the ratio of the reaction zone length to the charge diameter can not be considered as a small quantity. Thus, for confined charges, we assume that the front curvature effect is small and can be ignored, and the flow divergence is defined by radial expansion of the tube channel. It yields

$$d(\ln S)/dx = 2V_r/(RD) \quad (8)$$

where R is the local channel radius. V_r is the radial velocity of the channel expansion that can be calculated by integrating the equation:

$$\rho_W H_c D (R_c + H_c/2) \frac{d}{dx} (V_r) = \begin{cases} R(P - P_W) \\ 0, & P < P_W \end{cases} \quad (9)$$

where H_c and R_c are the wall thickness and the initial radius of the channel, ρ_W and Y are the density and yield of tube material, and $P_W = Y \ln(1 + H_c/R_c)$ is the strength of the tube. Equation (9) is obtained using the assumptions that ideal plasticity controls the tube deformation and that the effects of tube material compressibility and decrease in tube strength P_W during the tube deformation can be ignored.

The above-described system of equations is integrated from the shock front ($x=0$) up to the singular sonic point where the numerator and the denominator of the right part of the equation (3) must simultaneously become equal to zero. For charges confined in steel tubes, the unique solution which begins at $x=0$ and passes through this singular point, defines the LVD velocity D as an eigenvalue of the problem. The solution is obtained by numerical "shooting". For unconfined charges, the equation system involves the additional parameter K (the front curvature). There, the solution includes two steps. In the first step, the system of equations is solved by "shooting" so as to obtain the value of K as an eigenvalue of the problem associated with a given value of D . Importantly, D must be here interpreted as the wave velocity D_n normal to the front in a local point of this front. As a result, an evolution equation $K(D_n)$ for the detonation shock front is obtained. In the second step, one determines the front shape and the charge diameter d_{ch} associated with the given detonation velocity along the charge axis D .

Simple geometry consideration for a steady axially-symmetric, curved wave [10] leads to the following differential equation for the shock shape $Y(r)$

$$Y'' = (1 + Y'^2)[K(1 + Y'^2)^{1/2} - Y'/r] \quad (10)$$

with boundary condition at the charge axis:

$$r = 0: Y = Y' = 0 \quad (11)$$

In (10), K is the function $K(D_n)$ obtained in the first step and D_n is related to D by the identity

$$D_n = D/(1 + Y'^2)^{1/2} \quad (12)$$

Equation (10) is integrated from $r = 0$ up to a point r^* , where the particle velocity at the wave front becomes equal to the sound speed:

$$C_f^2 = U_f^2 + D^2/(1 + Y'^{-2}) \quad (13)$$

This point defines the charge diameter as $d_{ch} = 2r^*$.

The $K(D_n)$ relation can also be used to model some dynamical (i.e., unsteady) effects. For example, we have considered the behavior of a diverging spherical detonation with initial values of the front radius $R = R_0$ and detonation velocity $D = D_0$ at the initial time $t = 0$. A suitable structure for the evolution equation, suggested by [12], is

$$\delta D/\delta t = 2C_r^2/[K(D) - \zeta] \quad (14)$$

where C_r is the characteristic speed of the disturbance propagation on the front surface (considered as a known function of D), and ζ is the spherical front total curvature ($2/R$).

As a whole, our model, despite its simplified gas-dynamical character, includes all the key physical components of the processes under consideration. It enables one to easily study the behavior and properties of LVD. We shall later assess the precision of our model calculations (presented below) by means of the two-dimensional computer code described in [13].

Results and Discussion

Fig.1 compares the experimental data [1] and numerical results for LVD in NA prills confined in thin-walled steel tubes. The size of the prills is 1 – 2 mm, the charge density is 690 kg/m³, and the CJ detonation velocity is nearly 3.3 km/s. The coefficients used in the numerical modelling are: $e_{10} = -4.56$ MJ/kg, $\Gamma_1 = 0.9$, $l = 6.8$, $B_1 = 1.22$ GPa, $e_{20} = -6.16$ MJ/kg, $\Gamma_2 = 0.58$, $m = 3.46165$, $B_2 = 0.05434$, $Y = 0.22$ GPa and $\eta_{2f} = 0.01$. The best fit is obtained with the reaction rate coefficient $G_1 = 0.032 \mu s^{-1}$ and $n=1.1$. The results calculated with ± 10 % variation of G_1 and n are also shown in Fig.1. When the velocity increases from 1.1 to 1.9 km/s, the reaction zone length and the amount of burnt AN increase from 17 to 21 mm and from 0.37 to 0.68, respectively, and the tube expansion decreases from 30% to 16 %. The calculations cover the 0.5 - 3 GPa pressure range. The lower value corresponds to a LVD in the critical diameter (12 mm) charge and yields an evaluation of the threshold pressure P_{ign} for hot spot ignition in the considered AN.

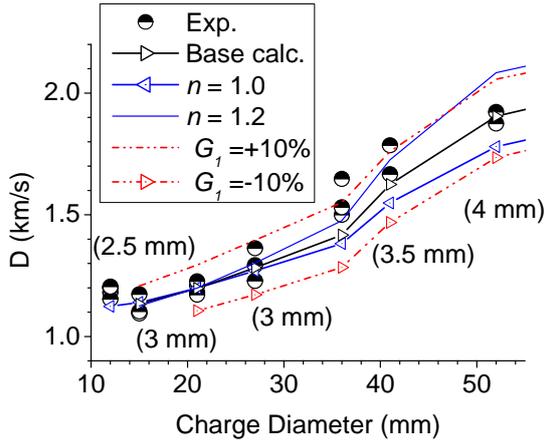


Fig.1. Charge diameter effect for LVD in AN prills confined in thin-walled steel tubes. Comparison of calculation results (lines + symbols) to experimental data (symbols). Figures in brackets give the wall thickness, which slightly varies with the tube diameter.

We then have estimated the reaction rates in other kinds of AN. The calculations are able to reproduce the effect of the wall thickness of steel confinement on the nonideal detonation velocity in AN prills of density 850 kg/m^3 observed experimentally in 100-mm diameter charges [2]. The pressure exponent was set to $n = 1.1$ and the best fit was then obtained with $G_1 = 0.0162 \mu\text{s}^{-1}$. Calculations were also conducted for simulating nonideal detonations in crystalline AN (sieved fraction with mean particle size of nearly 0.4 mm) of density 1040 kg/m^3 in unconfined 160 – 460 mm diameter charges [3]. These led to $G_1 = 0.0112 \mu\text{s}^{-1}$. We conclude that the reaction rate of AN decreases when the charge density increases. This effect is rather significant. For example, our calculations indicate that the charges made out of the two last kinds of AN, and confined in thin-wall steel tubes, yield a detonation failure in the charge diameters less than 50 mm. Only higher reactivity of AN prills studied in our experiments provides LVD in these charge diameters. Additional investigations are required to determine the reason of this effect, which, perhaps, could be either the decrease in the specific surface of AN or a difference in the reactivity of the different kinds of AN.

To investigate LVD in AN + Al mixtures we have analyzed the experimental data presented in [3]. The unconfined charges of density 1050 kg/m^3 were made by mixing the crystalline AN considered above with a polydisperse Al of two different sieved fractions (below 80μ and between 80 and 500μ). The experimental results demonstrate the strong influence of Al content on the charge diameter effect observed up to 8 % Al. A further increase of Al content up to 15% has no influence. The change of fine Al by coarse Al results in increasing the charge diameters. Our numerical modelling of these experiments is based upon the aforementioned coefficients for AN and upon the following ones for Al and its burning products: $s = 1$,

$C_{ox} = 0.71$, $r = 0$, $e_{30} = -0.415 \text{ MJ/kg}$, $\Gamma_3 = 1.7$, $l_3 = 6.8$, $B_3 = 10.74 \text{ GPa}$, $e_{40} = -9.0 \text{ MJ/kg}$, $\Gamma_4 = 0.28$, $m_4 = 3.19$ and $B_4 = 0.0428$. Fig.2 compares our calculation results to the experimental data.

The calculations conducted for the AN + 2%Al mixture with $G_1 = 0.0112 \mu\text{s}^{-1}$ (value obtained above for crystalline AN) and $G_3 = 0.035 \mu\text{s}^{-1}$ show that the Al additive would decrease the charge diameter by nearly 1.5 times at the same detonation velocities. However, the experimental effect turns out to be much larger.

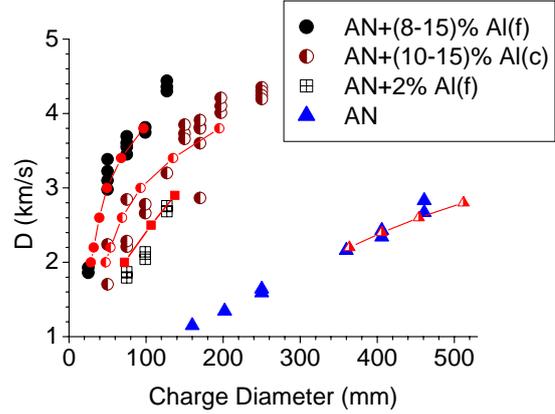


Fig.2. Charge diameter effect for low-velocity and nonideal detonations in unconfined charges of crystalline AN and the AN + Al mixtures. Comparison of experimental data [3] (symbols) to numerical modelling (red lines + symbols). Al(f) denotes the Al fraction sieved below 80μ and Al(c) the Al fraction sieved between 80 and 500μ .

We have conducted calculations with G_3 set to larger values such as $0.05 \mu\text{s}^{-1}$ (and more). However this increase has no effect because of the decrease in the concentration of oxidizing products (see Equation (2)). Thus, the only way to reproduce the experimental curve turns out to increase nearly 3 times the AN burning rate coefficient, i.e., $G_1 = 0.032 \mu\text{s}^{-1}$. Also, to reproduce the experimental data for the mixture with 8 % Al, the coefficient G_1 has been again increased about nearly 2 times up to $0.065 \mu\text{s}^{-1}$. Finally, to reproduce the effect observed when fine Al was replaced by coarse one, we have set G_3 to $0.001 \mu\text{s}^{-1}$ which corresponds to the 1-ms burning time of the Al particles. Taking into account the difference in the particle size, this time can be compared with the burning time of $50 - 70 \mu\text{s}$ for the $5\text{-}\mu$ Al particles evaluated in [14] for a detonation in Nitromethane + Al mixtures.

The results of the analysis thus show that a shift of the detonation velocity- charge diameter curves towards small diameters due to the Al additive observed in experiments can be explained only by assuming that AN in the AN + Al mixtures burns faster than without additive. As for the Al burning rate, reliable evaluations require additional experimental data on LVD in mixtures with Al powders of narrow fraction.

Fig. 3 shows experimental data on detonation velocity for coarse-grained Tetryl of three different

fractions in unconfined loose-packed charges of different diameters [4]. In this case, in contrast to AN and its mixtures, the results for LVD and normal detonation split into two different branches with an intermediate range of detonation velocities where detonation is not realized. The analysis shows that such behavior can be obtained only by assuming the two-term pressure dependence of the reaction rate (1). The results calculated with $n=0.8$, $F=0.26$, $k=1.7$ and $G_I = 0.11 \mu\text{s}^{-1}$ for the 0.4 – 0.63 mm grain size fraction are also presented in Fig.3. To reproduce the grain size effect, we have decreased the coefficient G_I in 1.3 and 1.7 times for the 0.63-1 mm fraction and the 1-1.6 mm fraction, respectively. Note that the two-term pressure dependence of the burning rate results in the Z-shaped $K(D_n)$ relationship shown in Fig. 4. Clearly, the intermediate branch of this Z-shaped $K(D_n)$ relationship, as well as the associated portion of the S-shaped relation between the velocity and the charge diameter (Fig.3) corresponds to an unstable process. This explains why experiments do not allow for steady detonations between the normal detonation and LVD branches.

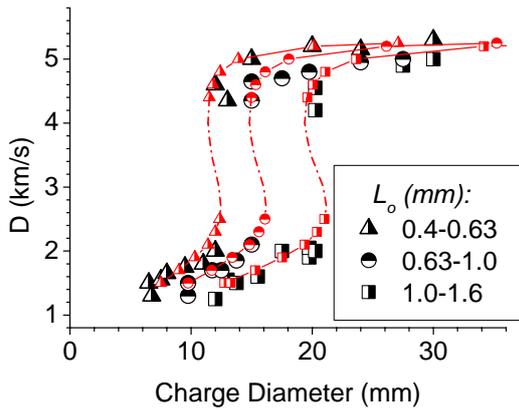


Fig.3. Low-velocity and nonideal detonations in coarse-grained Tetryl of three different fractions of grain size (L_o). Comparison of experimental data [4] (symbols) to results of numerical modelling (red lines + symbols).

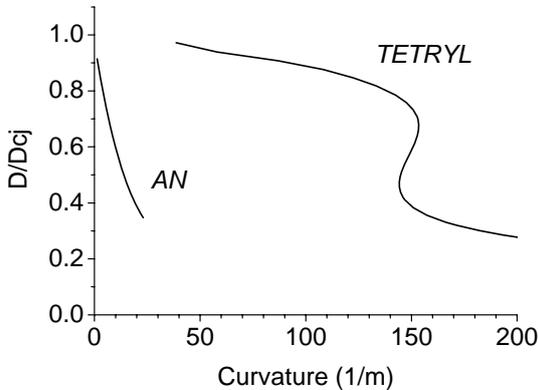


Fig.4. Calculated velocity – curvature relationship showing a monotonically decaying curve for AN and a Z-shaped curve for Tetryl with grain size 1.0 – 1.6 mm.

The Z-shaped velocity – curvature curve has interesting consequences for shock-induced transient processes, and, especially, in long delay for detonation transition. This effect can be directly used to explain such effect as delayed detonation [5] which, to date, had no clear interpretation. As an example, we consider the evolution of a diverging detonation in a spherical charge of PBX. To extract information on the burning rate, we have analyzed the experimental data on LVD in strongly confined charges of 16 mm i.d. of X1 (HMX/binder 96/4, density 1.823 g/cm^3 , detonation velocity 8.77 km/s and critical detonation diameter 2 mm) [15]. To simulate this data we have used properties of HMX with porosity 4%. The coefficients used in our calculations are: $e_{10} = 0.25 \text{ MJ/kg}$, $\Gamma_1 = 0.77$, $l = 8$, $B_1 = 1.194 \text{ GPa}$, $e_{20} = -5.15 \text{ MJ/kg}$, $\Gamma_2 = 0.55$, $m = 3.429$, $B_2 = 0.0795$, $\eta_{2f} = 10^{-4}$, $\rho_w = 7800 \text{ kg/m}^3$ and $Y = 500 \text{ MPa}$.

To obtain both the low-velocity and normal-velocity branches, we have used the two-term relation (1) with coefficients $G_I = 0.0012 \mu\text{s}^{-1}$, $n=1.0$, $F=0.3$ and $k=2.5$. The calculated Z-shaped velocity – curvature curve is shown in Fig.5.

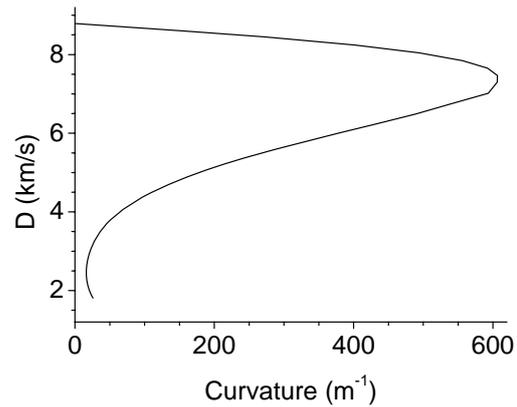


Fig.5. The Z-shaped velocity – curvature curve calculated for a model PBX

The calculations for diverging spherical detonation have been conducted for different values of D_o (hence, for different values of the pressure amplitude P_o in the initial wave) assuming that $C_{tr}^2 = 0.6D^2$, a good representative average value. The results calculated for $R_o = 32 \text{ mm}$ are shown in Fig.6. We found a qualitative agreement with the experiments in [5], specifically, the change of the prompt transition mode to a mode with $60\text{-}\mu\text{s}$ delay in the narrow 6.7 - 6.44 GPa interval of initiation pressures, a low-velocity stage propagating with velocity about 2500 m/s and a jump-like transition from the low velocity to the normal-detonation mode.

In order to confirm the above results, we have currently began modeling transient processes and shock initiation with a two-dimensional gas-dynamic code developed earlier and based on the visco-plastic model of pore collapse describing hot spot ignition of porous high explosives [13]. These calculations use the data

obtained above on the chemical conversion rates. A problem of shock initiation of detonations in a cylindrical unconfined charge of an explosive material of loose-packed density and a problem of divergent detonation initiated in the center of a spherical charge of a high density explosive material will be considered.

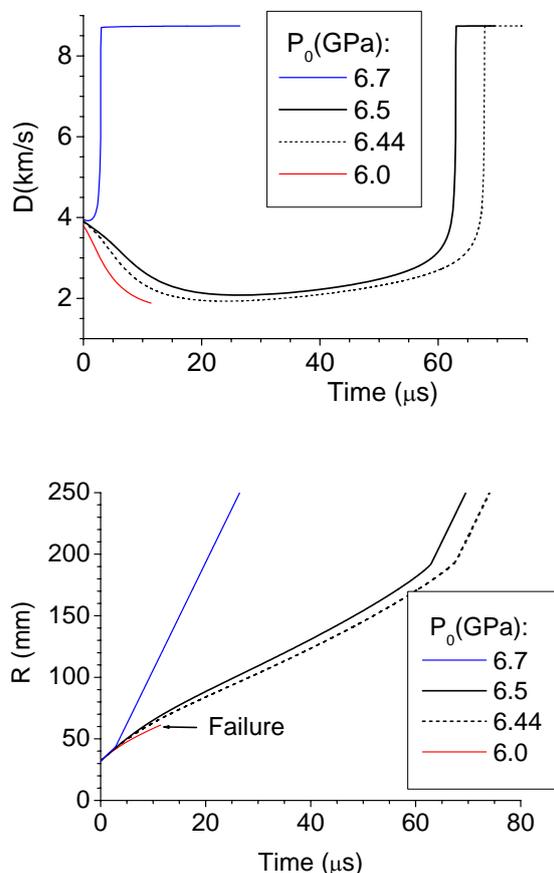


Fig 5. An illustrative example of a diverging spherical detonation in an unconfined charge of a model PBX with initial radius 32 mm depending on pressure amplitude P_0 of initial detonation wave. Evolution of the detonation velocity (top) and trajectory of the front (bottom).

Conclusions

The chemical conversion rate and its pressure dependence in ammonium nitrate and its mixtures with aluminium in the GPa-range of shock loadings have been estimated by means of a theoretical analysis of experimental data on LVD. In the case of pure AN, the pressure exponent of the chemical conversion rate equals 1.1, and the rate coefficient (attributed to 1 GPa) depending on the AN kind (or initial density) differs by a factor 3.

The shift of the detonation velocity – charge diameter curve towards smaller diameters obtained in experiment by the addition of Al to ammonium nitrate can be only explained if one assumes that AN in the mixture burns faster.

In the case of high explosives, the chemical reaction which occurs in LVD results in the Z-shaped velocity -

curvature relationship. This Z-shaped relation yields the dependence of detonation velocity on charge diameter with two separate branches for low-velocity and normal detonations observed experimentally in unconfined high explosives. Besides, the Z-shaped relation enables one to explain some delayed transient effects (including delayed detonation), which have been observed in shock initiation of explosive materials, but had not received clear interpretation yet.

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