

# ON THE MECHANISM OF DEFLAGRATION TO DETONATION TRANSITION IN GAS-PERMEABLE HIGH EXPLOSIVE

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*Main stages of deflagration to detonation transition in porous PETN are considered, including gas penetrative (convective) burning (CB) and low velocity detonations (LVD). The propagation velocity of the process is shown to be determined substantially by the pressure in a lead zone and exhibited only a slight dependence on the rate of pressure rise. The critical pressures of normal burning-CB-LVD-normal detonations transitions are given. Using streak camera and pressure gauges the CB and LVD processes are studied, and the conditions of the steady-state regimes are determined. The propagation velocity is found to be highly dependent on the pressure in lead zone. Experimental results are compared with the theoretical models.*

## INTRODUCTION

Deflagration in porous high explosives confined in metal tubes is known to tend to accelerate when the charge is ignited with a relatively weak ignition source. This acceleration eventually results in detonation if the charge is long enough. The main transient stages between the normal layer-by-layer combustion and the normal detonation are convective burning (CB) and low velocity detonation (LVD). The mechanisms of the latter two are still far from complete understanding because of the poor reproducibility and stability of the process and its strong dependence on the experimental conditions. CB and LVD differ in the mechanism of the reaction initiation. Combustion products in the case of CB penetrate through pores into the unreacted explosive overtake the flame front and preheat the walls of pores up to the ignition point (1). The initiation of the reaction in LVD is due to collapse of pores and to the plastic flow in the compression (shock) wave, which leads the LVD (1). However there is one common feature which makes CB and LVD different from normal burning and normal detonations, that is a rather small part of the total energy released in the vicinity of the front (less than

10-20%). The rest of the heat is released in an extended burn-up zone due to surface burning of the grains or cavities formed in the explosive and ignited at the first stage.

The intention in undertaking this study was to determine the conditions under which CB or LVD could be stabilized (at least approximately and in charges of limited sizes) and could be investigated in details in order to elucidate the basic characteristics of these transient (in a general case) processes. It has been shown experimentally that the quasi-steady CB and LVD could not be observed over the whole range of possible propagation velocities. For instance in the case of PETN these ranges are 1-10 m/sec for CB and 1000-3300 m/sec for LVD.

## CRITICAL PRESSURES

It is convenient to depict these transient processes in the  $W - P_m$  plane, where  $W$  is the propagation velocity and  $P_m$  the pressure in the lead zone. In the case of CB this zone corresponds to a narrow region adjacent to the ignition front, and  $P_m$  could be

chosen as the maximum pressure in the convective burning wave. For the LVD it is reasonable to take  $P_m$  as the pressure immediately behind the lead compression wave. The  $W - P_m$  plane is convenient because one can point out definite critical pressures on it:  $P_{tr}$ ,  $P_{min}$ , and  $P_{cr}$ . Region  $P_{tr} \leq P_m \leq P_{min}$  corresponds to CB and  $P_{min} \leq P_m < P_{cr}$  corresponds to LVD. These critical pressures are functions of the nature of the explosive, its structure (gas permeability porosity and grain sizes), of the charge diameter, and confinement parameters.

Experimental values of  $W$  can be plotted as a single curve  $W(P_m)$  for a certain range of experimental conditions and for a given explosive. This curve represents the evolution of the explosion in a porous high explosive and usually consists of several sections with markedly different sensitivity of the process to pressure variations. The effective pressure exponent of  $W$  calculated as  $\nu = d \ln W / d \ln P_m$  is about 2-3 near  $P_{tr}$  and then decreases to 1.5 (and even lower in the region of nonsteady regimes) when  $P_m$  is increasing. In the LVD region  $\nu = 4$  nearby  $P_{min}$  and then drops abruptly to 0.25 when the wave becomes stronger. The high value of  $\nu$  is one of the causes of the poor stability of CB and LVD. Generally speaking  $W$  is a function not only of  $P_m$ , but of the time derivative of  $P_m$  as well. However, the experimental observations show that  $dP_m/dt$  has only a minor influence on the critical pressure  $P_{tr}$ , and that the generation of the normal detonation is also determined mainly by the pressure in the wave and is practically independent of the wave profile in a reasonably limited range (3). This relative weak sensitivity of the CB process with respect to the pressure-time history could be understood from approximate theoretical analysis given in (4). According to (4)

$$W \sim \sqrt{r} P_m^{3/2} \quad (1)$$

when the pressure in the combustion zone is proportional to  $\exp(rt)$ . This relation is valid when  $1/r$  is assumed to be of the order of and/or less than the characteristic time of the filtration zone (that is the time of the pressure decay in the preignition region) yet much greater than the characteristic times of the preheating and ignition zones. Relation (1) shows a weak dependence of  $W$  on  $r$ . The dependence of  $W$  on  $P_m$  for PETN with the initial porosity 0.1 is illustrated in Fig. 1.

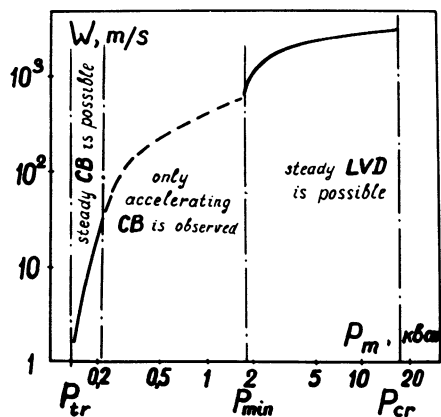


Fig. 1. Velocity of convective burning and low velocity detonation as a function of pressure (PETN,  $\phi_0 = 0.1$ , grain size  $d_0 = 0.5$  mm).

Thus even from those scarce experimental and theoretical results reported one can conclude that the evolution of the explosion in porous high explosives is determined mainly by the pressure level rather than by the rate of pressure change. Certainly this is true in the case of quasi-steady stages of the process when the pressure changes caused by external sources are decoupled from the pressure changes due to CB or LVD themselves. However, the rate of pressure changes is very important in estimating the actual pressure (or velocity)-time histories of the deflagration to detonation transition but not the gross characteristics of the quasi-steady stages of the process.

Measurements of the critical pressures have been carried out for pressed PETN having a relatively low initial porosity ( $\phi_0 = 0.03 - 0.2$ ). The PETN with a narrow distribution of grain sizes around  $d_0 = 0.5$  mm was pressed in pellets. The lateral surfaces of the pellets were covered with a thin layer of epoxy. The charge was mounted in metal or plexiglass tubes. The constant volume bomb technique (1,2) has been used in order to measure  $P_{tr}$ . Critical pressures  $P_{min}$  and  $P_{cr}$  (for initiation of LVD and of normal detonation, respectively) have been measured using the conventional technique of shock wave initiation. Shock waves from the donor charge passed through an inert plate into the test charge. The intensity of the compression wave was regulated by changing the thickness of the plate. The experimental results are shown in Table 1.

TABLE 1

Critical Pressures for PETN  
(Charge diameter 5 mm)

| Initial Porosity ( $\phi_o$ ) | $P_{tr}$ (kbar) | $P_{min}$ (kbar) | $P_{cr}$ (kbar) |
|-------------------------------|-----------------|------------------|-----------------|
| 0.2                           | 0.09            | 2.5              | 5.5             |
| 0.1                           | 0.13            | 2.5              | 8.5             |
| 0.025                         | 0.45            | 3.0              | 17.0            |

Unlike  $P_{tr}$  and  $P_{cr}$ , the value of  $P_{min}$  is only slightly dependent on porosity. As a consequence of this fact  $P_{min}$  is several times less than  $P_{cr}$  at lower porosities, however, the difference decreases with increasing  $\phi_o$ .

CONVECTIVE BURNING

Nomenclature:

- a – nondimensional coefficient dependent on the geometry of junction of small and large pores
- c – specific heat
- E – see Eq. (7)
- g – see Eq. (4)
- k – gas permeability
- l – averaged spacing of large pores
- P – pressure
- Q – heat of combustion
- R – universal gas constant divided by molecular weight
- $T_i$  – ignition temperature of the explosive
- $T_o$  – initial temperature
- $T_{ad}$  – adiabatic flame temperature
- u – gas velocity
- $u_n$  – rate of normal burning
- W – velocity of propagation of the ignition front
- t – time
- x – spatial coordinate
- $\gamma$  – specific heats ratio
- $\delta$  – diameter of pores
- $\lambda$  – thermal conductivity
- $\mu$  – viscosity
- $\nu$  – pressure exponent,  $d \ln W / d \ln P$
- $\rho$  – density
- $\phi$  – porosity
- $\xi_f$  – thickness of the filtration zone

Indexes: s – solid phase, g – gas phase, 1 – larger pores, 2 – smaller pores, o – boundary between ignition and filtration zones, m – lead zone

Experiments were carried out in a transparent combustion chamber made of plexiglass. Photographic technique and piezoelectric gauges were applied in order to monitor the propagation velocity W and the pressure profiles at different points along the charge. The charge was ignited at the open end of the tube by a fast burning igniter. The igniter created a sharp jump of pressure, and then the pressure in the combustion products was maintained at a constant level within 10%. The CB proved to propagate with an approximately constant velocity when the following conditions were fulfilled:

- a) porosity and gas permeability of the charge lower than certain threshold values,
- b) the maximum pressure in the combustion zone and that in the combustion products maintained constant,
- c) a subsonic flow of the combustion products.

Figure 2 shows W as a function of  $P_m$  for PETN with porosity 0.1. As one can see the average value of  $\nu$  in the tested region is 2.7. At higher velocities and

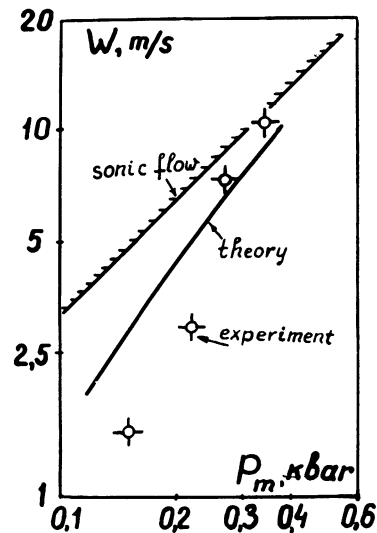


Fig. 2. Experimental (points) and theoretical dependencies of W on  $P_m$  for PETN. Shaded line corresponds to sonic flow of the combustion products.

pressures the CB accelerates very fast. Rough estimations made using the streak camera and pressure records suggest the value of  $\nu$  between 1 and 1.5 in this region.

A typical streak camera record of the quasi-steady CB is presented in Fig. 3

It follows from this record that CB is non-one-dimensional and pulsating in its nature. The periodical decrease in the light emission at the flame front and dark striations in the combustion products are evidence of an intensive fragmentation of the explosive in the combustion zone. Such a behavior of the deflagration wave is presumably caused by advanced penetration of the hot gases through larger pores and by a fast pressure rise inside the latter. Measured depths of the pulsations (several mm) are longer than the size of the preheat zone but less than the size of the filtration zone. The frequency is proportional to  $W$  and is of the order of a hundred Hz.

An analytical model is suggested which gives an approximate (at least qualitative) description of the steady CB process. The model is based on the experimental evidence that CB is led by hot gases penetrating through larger pores. This fact was confirmed by analysis of the quenched specimens. Size distribution curves obtained by direct microscopic measurements of pores at cross-sections of pressed charges demonstrated that pores with sizes ten times higher than the average exist, and that their volume is not more than 10% of the total volume of voids. The estimated values of relative volumes of larger pores are given in Table 2.

The estimations are made using the data on gas permeability ( $k_{exp}$ ) for different porosities in charges

TABLE 2

*Estimated Volumes of Large Pores in Pressed PETN*

|  |      |      |      |      |
|--|------|------|------|------|
| Porosity $\phi_0$                            | 0.45 | 0.3  | 0.25 | 0.1  |
| Gas permeability ratio $k_{theor.}/k_{exp.}$ | 1    | 25   | 60   | 1100 |
| Volume of large pores $\phi_1$               | 0.45 | 0.12 | 0.07 | 0.01 |

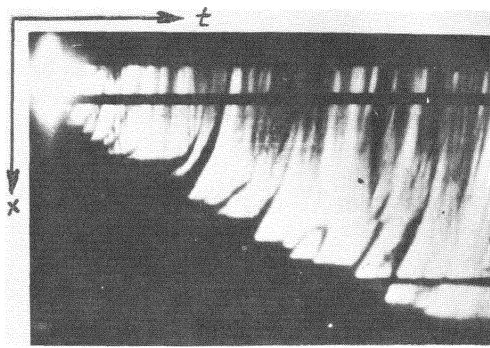


Fig. 3. Streak camera photograph of convective burning of pressed PETN.

of pressed PETN (1,2). The difference between the experimental  $k_{exp}(\phi_0)$  curve and that calculated from the Cozeni-Karman relation is assumed to be a consequence of the fact that the volumetric portion of larger pores which determine the effective gas permeability of a real charge is small compared to  $\phi_0$ . Smaller pores have high filtration resistance, ignition of the explosive inside those pores is much less probable than in larger ones because of the very small amount of heat stored in the gas which fills up the pore. Thus the smaller pores form an effective sink of the gas from the larger pores. This means that the main objection against the possibility of steady one dimensional regimes of CB with advanced gas penetration (13) can be avoided. Kuo, and Summerfield (6), and Dubovitsky, et al. (7) have obtained steady-state solutions for the regimes where the flow rate of gases did not exceed the velocity of the ignition front, i.e., the solid was preheated in their case by another mechanism than the advanced filtration of combustion products.

In the suggested model the gas is allowed to flow through the ignition front into the unreacted substance in the steady-state case if all the gas that has penetrated into the unreacted explosive through large pores is accumulated in smaller pores. For convenience we consider a system with two types of pores: larger ones having diameter  $\delta_1$ , gas permeability  $k_1$ , and porosity  $\phi_1$  and smaller ones with the relevant parameters  $\delta_2$ ,  $k_2$ ,  $\phi_2$ . It is assumed that  $\delta_1 \gg \delta_2$ ,  $k_1 \gg k_2$ ,  $\phi_1 \ll \phi_2$ . The basic principles and equations for filtration in a medium having dual porosity are given in (5). According to Barenblatt et al. (5) the two values of pressure in gas phase are referred to each spatial point  $P_1$  and  $P_2$  for large and small pores, respectively. The equations which describe

the filtration in a system with dual porosity can be written as follows:

$$\frac{d^3\sigma}{dz^3} + \frac{d}{dz} \left( \sigma \frac{d\sigma}{dz} \right) - \frac{d\sigma}{dz} = 0 \quad (2)$$

$$\pi^2 = \sigma^2 + \frac{d\sigma}{dz}. \quad (3)$$

Here the zero point of the coordinate system is traveling at the velocity of CB at the boundary between the filtration zone and the preheat zone. The boundary conditions are:

$$\text{at } z = -\infty: \quad \sigma = \frac{d\sigma}{dz} = \frac{d^2\sigma}{dz^2}$$

$$\text{at } z = 0: \quad \pi = \pi_o, \quad \frac{d\pi}{dz} = \left( \frac{d\pi}{dz} \right)_o$$

where

$$\pi = P_1/P_x$$

$$\sigma = P_2/P_x$$

$$z = \xi/\xi_x$$

$$\xi = Wt - x$$

$$\xi_x = \ell \sqrt{k_1/ak_2}$$

$$P_x = gW$$

$$g = 2\mu_g\phi_2\ell/\sqrt{ak_1k_2}. \quad (4)$$

The initial pressure in the pores is neglected when writing the boundary conditions. Equations (2) and (3) are to be solved for the filtration zone only, since it is much longer than the preheat zone and the amount of gas accumulated in smaller pores can be neglected in the preheat zone. Thus the preheat zone and the subsequent ones are described by the relations derived in (4) for the case of monoporous explosives. Using the expression for the mass flow of the filtrating gas given in (4) one gets:

$$\left( \frac{d\pi}{dz} \right)_o = \frac{\phi_1 u_o}{2\phi_2 W} \quad (5)$$

where  $u_o = u_{z=0}$ . Furthermore it was shown in (4) that

$$\frac{u_o}{W} = 1 + \frac{E}{P_o} \quad (6)$$

where

$$E = \left( \frac{T_i - T_o}{T_{ad} - T_o} \right)^2 \frac{c_s \lambda_s \rho_s (\gamma - 1) T_o}{c_p \mu_g \gamma}. \quad (7)$$

The solution of Eqs. (2) and (3) can be expressed analytically in terms of parameter  $q = d\sigma/dz$

$$\sigma^2 = -2 [\ln(1 - q) + q] \quad (8)$$

$$\pi^2 = -2 [\ln(1 - q) + q/2]. \quad (9)$$

At  $z = 0$  this gives

$$\pi_o^2 = -2 [\ln(1 - q_o) + q_o/2] \quad (10)$$

$$\sigma_o^2 = -2 [\ln(1 - q_o) + q_o] \quad (11)$$

$$\left( \frac{d\pi}{dz} \right)_o = \sigma_o (1 + q_o) / 2\pi_o. \quad (12)$$

The value of  $q_o$  varies over the range of 0 - 1. Excluding  $u_o/W$  from Eqs. (5), (6), and (12) one finds that

$$W = \frac{E}{g[\sigma_o(1 + q_o)\phi_2/\phi_1 - \pi_o]}. \quad (13)$$

Equations (13), (4), (10), and (11) give a parametric relation between  $W$  and  $P_o$  in terms of  $q_o$ . The analytical expression for  $W(P_o)$  can be obtained if  $q_o \ll 1$ . Expansion of  $\ln(1 - q_o)$  in series up to the  $q_o^2$  term yields a simple formula:

$$W = \frac{\phi_2 P_o^2}{\phi_1 g(E + P_o)}. \quad (14)$$

This expression is valid for

$$[(1 + E/P_o)(\phi_1/\phi_2)]^2 \ll 1.$$

In order to replace an uncertain parameter  $P_o$  for the  $P_{\max}$  (or  $P_m$ ) measured experimentally it is reasonable to use the equation for  $P_m$  derived in (4):

$$P_m^2 = P_o^2 + (8,6 + f)\gamma RT_{ad} \left[ \rho_s W \frac{\lambda_s}{\lambda_g} \left( \frac{T_i - T_o}{T_{ad} - T_o} \right)^2 \right]^2 \quad (15)$$

where

$$f = \mu_g \sqrt{4, 2(\phi_1/k_1)/\rho_s u_n} \cdot \quad (16)$$

Equation (15) in most practical cases can be simplified, since usually  $P_{max} - P_o \ll P_o$ . If  $q_o \ll 1$ , the characteristic size of the filtration zone is expressed as follows:

$$\xi_f = 2\ell \sqrt{k_1/ak_2} \cdot \quad (17)$$

Equation (17) predicts substantially thinner filtration zones than those in the case of monoporous charges. For pressed PETN with grain size of 0.5 mm experimental values of  $\xi_f$  (several cm) can be obtained if  $l$  is assumed to be of the order of several grain sizes.

An example of a calculated  $W(P_{max})$  curve shown in Fig. 2 (solid line) is illustrated further in Table 3.

TABLE 3

Calculated Parameters of Convective Burning of PETN

| $P_m$ bar                   | 100 | 200   | 300  | 400  |
|-----------------------------|-----|-------|------|------|
| $W$ m/sec                   | 1.6 | 4.3   | 7.2  | 10   |
| Length of pre-heat zone, cm | 0.3 | 0.5   | 1.4  | 2.0  |
| $P_1/P_2$ at $z = 0$        | 0.2 | 0.155 | 0.14 | 0.13 |

## LOW VELOCITY DETONATIONS

As the amplitude of the nonelastic wave increases the more probable becomes the initiation of the reaction in hot spots. Eventually it results in transition of CB to LVD. Special experiments with metal diaphragms which separated two parts of a charge have demonstrated that the gas filtration is dominant

until  $W$  exceeds the level of 700-800 m/sec. The process propagating at higher velocities can be easily transmitted through a metal diaphragm and should be regarded as LVD. For this stage of transient process there also exist certain conditions where LVD propagates at nearly constant velocity.

Experiments were carried out with charges of PETN ( $\rho_s = 1.73$  g/cc) confined in steel tubes (9,10). The tubes were of 5 mm i.d. and of different wall thickness. A pyrotechnic igniter or a weak shock wave were used to initiate LVD. The propagation velocity was measured using a photographic technique. The results are shown in Fig. 4 (points). Because of an insufficient frequency response of the applied transducer the pressure  $P_m$  was measured indirectly in special experiments point  $P_m$  is assumed to be approximately equal to the maximum pressure which the tube with a given wall thickness can withstand when a charge of explosive is burned up in the tube. The rate of pressure rise in these experiments was about 0.5 kbar/ $\mu$ sec. Velocity of LVD grows from 1300 to 3300 m/sec when a stronger confinement (higher pressure in the compression wave) is used. In a certain region  $W$  is much less than the longitudinal acoustic velocity of the solid phase (2250 m/sec).

The experimental data on the propagation of LVD (1,9) and on the compressibility of PETN in weak shock waves (10) show that the velocity of LVD depends on profiles of the compression wave in the explosive and its amplitudes. Transition from elastic deformation to a nonelastic flow is very important in determining the conditions under which LVD arises. For pressed high explosives the plastic yield strengths lie in the range of 1-3 kbar. When  $P > P_T$  ( $P_T$  is the yield strength) the compression wave consists of two waves (elastic and plastic), which may propagate with different velocities. The reaction starts in the nonelastic wave which can propagate much slower than the elastic (acoustic) precursor. In order to find the conditions where both waves are separated experiments were made with pressed TNT ( $\rho = 1.6$  g/cc) using the conventional technique of electromagnetic measurements. A two wave configuration was observed for pressures lower than 7 kbar, at higher pressures the pressure profile corresponded to a single shock wave. It was demonstrated in (11) and (12) that LVD propagates at a supersonic speed with respect to the two-phase medium behind the elastic precursor, and hence it could be called a detonation phenomenon.

Figure 4 (solid curve) shows the calculated dependence of velocity on pressure. The calculations were made using simple estimations of wave parameters given in (12). These estimations account for the bifurcation of the wave and collapse of pores in the plastic wave.

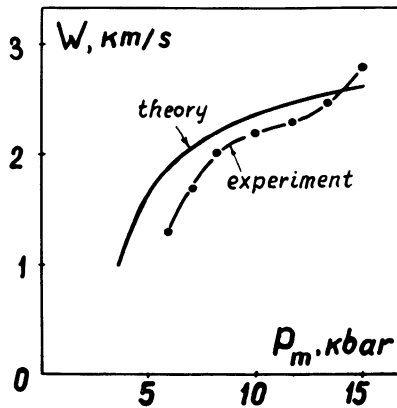


Fig. 4. Velocity of low velocity detonation in pressed PETN as a function of pressure in the compression wave. Calculated values—solid line, experimental results—points. Charge diameter 5 mm, confinement—steel tubes. ( $b = 0.03$ )

When comparing the calculations with the experimental results the following values of parameters were used:  $\phi_0 = 0.1$ ;  $\phi_1 = 0.01$ ; grain size 0.5 mm;  $k_1 = 10^{-11}$  cm<sup>2</sup>;  $E = 8 \cdot 10^7$  g/cm sec<sup>2</sup>;  $g = 2 \cdot 10^6$  g/cm<sup>2</sup> sec. The range of reasonable values of  $E$  and  $g$  calculated using the available literature data is  $4 - 15 \cdot 10^7$  g/cm sec<sup>2</sup> and  $1 - 6 \cdot 10^6$  g/cm<sup>2</sup> sec. As mentioned before the length of the filtration zone is several cm. Noteworthy is the high sensitivity of the CB velocity to pressure variations, the exponent  $\nu$  is everywhere higher than 1. The results of calculations are in reasonable agreement with the experimental data. The agreement could be made better if one accounts for the possible turbulent filtration of the gas through pores.

The model of a steady-state CB considered is not unique. There could be found other nondimensional effects which would result in effective gas losses upstream to the ignition front. For instance, Margolin and Margulis (8) have shown that U-shaped pores generate a swirled filtration of the gas and make the spontaneous penetration of a flame into pores easier. The role of this process in a forced CB is to be elucidated.

Besides the high pressure exponent of  $W$  there is another destabilizing factor in CB which restricts the region of steady regimes at high propagation velocities. This factor is the appearance of a region of sonic flow in the combustion products (flow choking) and, as a consequence, generation of compression waves. The critical condition of flow choking is calculated by integrating the steady-state conservation equations for a two-phase reacting flow. Suppose that the starting point for integration is  $P = P_{\max}$ ,  $u_s = u_g = 0$ ;  $\phi = \phi_m$ ;  $T_g = T_{ad}$ ;  $T_s = T_0$ , and the terminal point is  $\phi = 1$ ,  $P = P_{\infty}$ . Assuming that flow tubes have a constant cross-section and neglecting the interaction of the flow with the confinement wall one can come to the expression:

$$\frac{P_{\infty}}{P_m} = \frac{1}{\gamma + 1} \pm \sqrt{\left(\frac{\gamma}{\gamma + 1}\right)^2 - \frac{b}{P_m}} \quad (18)$$

$$b = \rho_s(1 - \phi_m)W \sqrt{2\gamma RT_{ad}/(\gamma + 1)}. \quad (19)$$

The condition  $b \geq P_m \gamma / (\gamma + 1)$  means that the process becomes nonstationary (accelerating). The calculated upper limiting values of  $W$  for steady regimes of CB are shown in Fig. 2 (shaded line). This effect together with the strong pressure dependence of  $W$  explain why CB in pressed PETN charges under confinement is highly unstable, and that CB propagating at velocities greater than 10 m/sec is impossible.

Acceleration of CB up to velocities of 100 m/sec and higher results in formation of a nonelastic compression wave in the unreacted explosive. This nonelastic wave is expected to affect the CB process in different ways:

- to decrease the porosity,
- to change the ratio of large and small pores,
- to preheat the gas and create hot spots in the solid,
- to change the conditions for fragmentation.

The analytical models which account for the effects of fragmentation of the solid in a CB wave and of the compression of the unreacted porous explosive would complete the description of the transient process, if they were worked out. However, this is still a problem for future studies.

A theoretical approach to the problem of stabilization of LVD (14) has shown that lateral expansion of confinement wall provides the energy losses needed for making the process steady. However, the analysis given in (14) has to be further developed if one wants to understand the mechanism of the transition from low velocity detonation to normal detonations. Extremely important in this process is the influence of heat release in the burn up zone on the flow parameters nearby the LVD front. This problem is under investigation at the present time.

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