Detonation propagation along percolating cluster in composite explosives

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Abstract. The detonation performance of melt-cast plastic-bonded explosives (PBXs) based on high melting explosive octogen (HMX) was studied in the paper. It has been found that the detonation velocity strongly depends from the dispersion distribution of HMX particles: it changes from 7800 to 8700 m/s. We explain this by the possibility of detonation propagation in PBX through different mechanisms, including detonation front propagation along a percolating cluster formed by filler particles. Thus, varying the particle size distribution can bring about one detonation mechanism or another and hence control the energy release dynamics of melt-cast PBXs to attain high efficiency in practice. Experimental results confirm the assumptions.

1 Introduction

Plastic-bonded explosives (PBX) structurally represent mixed high-order explosives with a polymeric binder. Due to heterogeneity PBXs can be defined as a ‘solid-suspension’ in which explosive crystals (solid phase) are distributed in a continuous polymeric matrix (dispersed phase).

Depending on the volume polymer fraction, PBX charges can be shaped by pressing, extrusion, or casting: the charge shape is secured (cured) by chemical cross-linking or transition to the high-elasticity state upon cooling (in case thermoplastic polymers are used). There is also a curing system in use that is based on dissolving the polymer in a plasticizer to generate a high-viscosity matrix (plastisol PBX). The hexogen (RDX) and high melting explosive octogen (HMX) are frequently used as the high-order filler of conventional PBXs. The polymeric binder is typically a homogeneous multicomponent system comprising a polymeric base, plasticizers, processing and performance additives, etc. Many PBXs contain aluminum as the energetic additive [1-3].

All polymeric binders used in PBX compositions can be classified into two types: inert binders (inert polymer binder – IPB) not involving in exothermic explosion reactions and active binders (active polymer binder – APB) capable of self-explosion and those being oxidizers towards PBX components. This classification is conditional: fluorinated polymers in PBXs containing no metal fuel are inert, whereas those in aluminized PBXs should be referred to the APB because their decomposition products when interacting with Al contribute considerably to the explosion energy output. PBXs are widely used because they have superb mechanical properties, a low porosity, are able to fill in complex forms, and exhibit a reduced sensitivity to mechanical stimuli. The listed advantages are mostly inherent in melt-cast PBXs that are characterized by a relatively high content of the binder. Their sensitivity is low because the polymeric matrix is continuous and elastic and is capable of uniformly distributing external mechanical energy, thereby preventing the detonation progression.

The global tendency to mitigating the risks when handling and using explosives determines the increasing proportion of PBXs in the nomenclature of practical formulations. For instance, fluorinated matrices filled with powdered triaminotriphenylene (TATB) are modern standards of safe explosives such as PBX 9502 (95 wt.% TATB, 5% fluorinated binder) and LX-17-0 (92.5% TATB and 7.5% chlorofluoropolymer binder) [1-2].

The further improvement in explosive formulations is associated with the development of melt-cast PBXs based on an APB. New high-energy-density polymers and plasticizers have now emerged that contain nitrate groups (Poly-GLIN, poly-NIMMO, PGN), azide groups (GAP, BAMO, AMMO) [4], azole moieties [5], etc. PBXs with a high content of the APB can ensure safety and casting properties with a minimum reduction in the detonation performance compared to more powerful pressed formulations.

The approach to developing melt-cast PBXs are typically attributed to minimizing the binder content where a possibility remains to shape charges by the casting process, and the practice demonstrates that the greater the content of the high-order filler, the better the detonation performance.
The casting properties of the suspension are governed by its viscosity which depends on the amount, shape and size of filler particles [6], interfacial energy, etc. The known method for reducing the viscosity at a high filler content is to use the bimodal grain size of the solid phase [6-7]. Thus, the maximum filler content and acceptable viscosity can be optimized under other equal conditions by down-selecting a filler fractional composition which will eventually be determinate for one or another system. In doing so, the practical efficiency attributes such as brisance, acceleration ability and blast effect are accepted de facto and considered a-priori to be maximum for the thus-optimized system.

In our previous studies on melt-cast HMX-based PBX composites with an APB [8-12] presented herein, we did not aim to provide the maximum filler content and confined it to no less than 25 vol.%. The purpose on this paper is to show the relationships between detonation velocity, acceleration ability, and filler particle size while preserving casting properties.

2 Experimental part

2.1 Materials

We examined a melt-cast PBXs based on an APB – a homogeneous explosive system – which is a plasticized methylpolyvinyltetrazole polymer (CHNO-type) having a density of about 1.5 g/cm³. The binder’s own detonation velocity was 7200 m/s for a charge with diameter of 25 mm in a steel tube; the critical diameter of detonation was over 20 mm. The filler was HMX (Russian production) with different particle sizes, whose calculated threshold of the detonation velocity was 9160 m/s [8] and the density was 1.91 g/cm³. Micron-sized aluminum powders (Al, brand «ASD, RUSAL, Russia) with spherical particles were used as the energetic additive. A curing agent in the concentration of 0.5 vol.% was incorporated into all composite explosives. The explosive blends were mixed by hand, then cured, and loaded into plastic tubes and kept under specified conditions until they were completely cured.

2.2 Experimental setup

The detonation velocity \( D \) was measured by discrete point system, based on the short circuit of ionization sensors, the signal from which is transmitted to a recording oscilloscope with a frequency of 50 MHz. Shellless or plastic encapsulated charges of 20-25 mm in diameter and 50-100 mm long were used. Distance between two sensors was 20-25 mm. A booster charge with the same diameter and a height of 10 mm was used for initiation. The booster charge’s own detonation velocity was 7500 m/s. The first sensor was at a distance of no more than 25 mm from the initiation plane. The measurement error was ±50 m/s. Typically, at least three parallel tests were run, and the average of three was the measurement result.

The acceleration ability was determined by the end-face throwing method in which a 25-mm diameter steel plate of 2.5 mm in thickness and 9.35±0.05 g in weight was thrown from the end face of a plastic shell-enclosed charge of 25-mm in diameter and 50 mm long. The acceleration ability was expressed as the plate flying velocity \( \omega \) (m/s) measured by discrete point system, similar to that described above. The distance between the contact sensors was 300 mm to minimize the influence of the overclocking area on the result of measuring. The measurement system error was not in excess of ±50 m/s. Two to three parallel measurements were done and the results were taken into account only from those tests in which the thrown plate did not break down, as was evidenced by the way how the contact target was breached. The measurement schematic is illustrated in Figure 1.

![Fig. 1. Schema for the measurement of acceleration ability.](image)

3 Results and discussion

3.1 Relationship between detonation velocity and HMX particle size

In initial experiments, the detonation velocity \( D \) was examined as a function of the HMX particle size in binary PBXs such as HMX/APB. Two types of HMX was used: a coarse fraction (fraction 1) and a fine fraction (fraction 2) at a different ratio thereof. Measurements were done on charges of 25 mm in diameter. The test composite explosives and detonation velocity measurement results are summarized in Table 1. The average HMX particle size for fine \( (d_1) \) and coarse \( (d_2) \) fractions are given therein. All the composites under test retained casting properties.

It can be seen in Table 1 that the detonation velocity is dependent heavily on the component particle sizes: the obtained detonation velocity values ranged from 7800 to 8760 m/s. The following general tendencies can be deduced from the findings. When the single-fraction filler is used, the detonation velocity increases with increasing particle size (compare the test results for compositions Nos 1-3 and 6 in Table 1). Probably, this tendency is extremal in nature: if the particle size exceeds some limit, the detonation velocity declines (compositions Nos 10-11 compared to Nos 3 and 6). Further, the highest detonation velocity is achieved by using two-fraction HMX when the mean fine fraction size is below 50 µm and the mean coarse fraction size is 200–300 µm, in which case the optimum coarse/fine
fraction ratio for this system is ~70/30 % (compositions Nos 5 and 8-9).

Table 1. Detonation velocity of HMX/APB composites depending on HMX particle size.

| #  | Concentration (%) and size of HMX powder in PBX compositions | APB, % | \( D, \, m/s \) |
|----|-------------------------------------------------------------|--------|----------------|
| 1  | Fraction 1 \( \% \) \( d_1, \mu m \) Fraction 2 \( \% \) \( d_2, \mu m \) |        |               |
| 2  | – – 80.0 85 20.0 7800                                      |        |               |
| 3  | – – 80.0 125 20.0 7760                                     |        |               |
| 4  | 32.2 500 45.8 125 22.0 8300                                |        |               |
| 5  | 54.6 315 23.4 <50 22.0 8560                                |        |               |
| 6  | 78.0 200 – – 22.0 8400                                     |        |               |
| 7  | 54.6 500 23.4 125 22.0 8200                                |        |               |
| 8  | 54.6 200 23.4 <50 22.0 8460                                |        |               |
| 9  | 54.6 200 23.4 <50 22.0 8760                                |        |               |
| 10 | 40.0 950 40.0 125 20.0 8150                                |        |               |
| 11 | 30.0 950 50.0 125 20.0 8300                                |        |               |

We explain the obtained results as follows: when the polymeric matrix is mixed with crystalline HMX, the crystals are separated by the polymeric interlayer and randomly oriented in space. The propagation of detonation along the explosive charge is defined by the filler’s volume fraction \( \varphi \) and the interlayer ability to transmit detonation from the crystal to crystal [8-10]. The propagation velocity in local areas is not monotonically increasing.

\[
D = \sum_{i=1}^{n} \varphi_i \left( \frac{\varphi_i}{D_{0i}} \right)^{-1} \tag{1}
\]

where \( \varphi_i \) is the volume fraction of the \( i \)-th component; \( D_{0i} \) is the detonation velocity (front transmit velocity) along the \( i \)-th component.

Fig. 2. Propagation of detonation in PBX.

A high filler content in the polymeric matrix may result in an HMX particle ‘chain’, the detonation through which may outrun the mean detonation velocity defined by Eq. (1). This leading detonation will consequently give rise to a ‘nonsmooth’ detonation front, which is recorded by the end-face imprint left by the PBX charge on the steel disk. The detonation propagation pattern is schematically displayed in Figure 3.

Fig. 3. Detonation propagating in a monocomponent explosive (left) and in a PBX along the percolating cluster (right), and end traces of detonation on metal plates.

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\[
D = \sum_{i=1}^{n} \varphi_i \left( \frac{\varphi_i}{D_{0i}} \right)^{-1} \tag{1}
\]

where \( \varphi_i \) is the volume fraction of the \( i \)-th component; \( D_{0i} \) is the detonation velocity (front transmit velocity) along the \( i \)-th component.
acceleration ability when HMX or RDX contained 10-15% Al.

The calculated and experimental results [20-21] show that 10-15% Al introduced into HMX enhance the acceleration ability to the limit: the projectile velocity increases by 2-4%, whereas the heat of explosion has a 25-30% increase.

The use of percolation effect in PBXs as the way to control the detonation velocity, according to the commonly accepted considerations on \( \omega = \sqrt{\rho \cdot D} \), predetermines the possibility of substantially improving the acceleration ability of PBXs.

The acceleration ability of PBX formulations comprised of HMX/APB/Al was examined herein as a function of the filler particle size by the procedure shown in Figure 1.

Varying the particle size of HMX and Al in PBXs with the component ratio and composition density being unchangeable was found to allow the acceleration ability to be effectively controlled from the minimum value of 2140 m/s to maximum 2600 m/s. The latter value is achieved when the formulation contains 10 wt.% Al, 20 wt.% polymer binder, and two-fraction HMX at a ratio of coarse (315-700 \( \mu \)m) to fine fraction (specific surface area 3000 cm\(^2\)/g) of about 5 to 2 [22]. Note that the acceleration ability of a maximally pressed HMX charge comprising 2.5% binder does not exceed 2350 m/s when tested by the same procedure.

The possibility of enhancing the acceleration ability by varying the solid particle size in PBXs under examination is explained from our point of view by the particular structure of such explosives and by the mechanism through which the energy is released and transmitted to the projectile body. The structure is peculiar because it has two physically heterogeneous systems – an APB and HMX capable of self-detonation – and because the metal fuel particles are in close contact with the APB.

In the context of assumptions that the detonation wave propagates along the percolating cluster, the acceleration process can qualitatively be described by the following postulates.

The projectile body can accelerate as long as the gas pressure behind the body is higher than that before it, that is, the acceleration is defined by both the detonation product pressure and detonation product action time.

The detonation products of composite explosives do not intermix directly in the detonation wave and in the area nearest to it [23-24], and the reaction zone represents a kinetically unbalanced volume in which mass velocity gradients of a few hundred meters per second occur [25]. The propagation of detonation wave will bring the detonation plume out to the interface with the projectile body before all reactions within the explosive are complete, thereby providing the starting acceleration of the body. This starting acceleration will obviously be smaller than that upon approach of a flat detonation front, but the projectile body will persist in the action zone of products of so-called secondary reactions whereby the time the projectile body is exposed to detonation products will increase. The mass velocity gradients originating in the detonation wave promote a fast oxidation of the metal fuel both due to the protective oxide films being separated mechanically from the particle surface and due to the unreacted products intermixing intensively.

### 3.3 Analysis of potential energy of PBX conversion into acceleration performance

The conversion coefficient of the energy potential into acceleration performance in cylinder tests [14] can be estimated by the equation:

\[
K_m = \frac{E}{q}
\]  
(2)

where \( K_m \) is the conversion coefficient of the energy potential into the acceleration ability of the explosive; \( E \) is the kinetic energy of the projectile body (kJ):

\[
E = \frac{M \omega^2}{2}
\]  
(3)

where \( M \) and \( \omega \) are the weight (kg) and velocity (m/s) of the projectile body, respectively;

\[
q = Q \cdot C
\]  
(4)

\( C \) is the weight of the charge to throw the body (kg); \( Q \) is the energy potential of the explosive (kJ/kg).

| Composition, (wt,%) | \( \rho \) g/cm\(^3\) | \( Q_{max} \) kJ/kg | \( q \) kJ | \( E \) kJ | \( K_m = \frac{E}{q} \) |
|---------------------|------------------|-----------------|---------|--------|------------------|
| HMX (100)           | 1.91             | 6270            | 37.6    | 22.5   | 0.60             |
| HMX/Al (88/12)      | 1.97             | 7920            | 49.1    | 24.2   | 0.49             |
| CL-20 (100)         | 2.04             | 6700            | 43.0    | 25.7   | 0.60             |
| CL-20/Al (90/10)    | 2.10             | 8040            | 53.1    | 26.7   | 0.50             |
| HMX/IPB 98/2        | 1.84             | 6400            | 45.9    | 25.8   | 0.56             |
| HMX/APB (78/22)     | 1.77             | 6150            | 45.3    | 22.2   | 0.49             |
| HMX/APB/Al (70/20/10)| 1.84             | 7540            | 56.7    | 31.6   | 0.56             |

Note: APB – active polymeric, IPB – inert polymeric binder.

The evaluation of \( q \) by the end-face throwing technique requires the knowledge of the ‘active’ explosive charge weight, the notion of which is given in [14]:

\[
C_a = \frac{2}{3} \pi r_0 \rho_0 l = \frac{l}{12} \pi d^3 \rho_0,
\]  
(5)

where \( C_a \) is the active weight, \( r_0 \) is the radius, \( l \) is the length and \( \rho_0 \) is the density of the explosive charge.

Eq. (5) is applicable only to a shell-free charge because a massive shell requires that its strength properties are taken account of.

As the energy potential \( Q \) of the explosive, it is reasonable to use the maximum heat of explosion \( Q_{max} \) – the energy quantity that corresponds to the most highly
thermic reactions upon decomposition of the explosive and is equivalent to the maximum entropy. \( Q_{\text{max}} \) is an explosive constant because it is defined only by the chemical composition of an explosive. Equations for estimating \( Q_{\text{max}} \) are given in [14].

The estimation results for \( k_{\text{max}} \) by Eqs. (2–5), as reported in [26], are summarized in Table 2. The potential energy of mono-component explosives can be converted into acceleration performance to the greatest possible extent of 60%, whereas the explosion energy conversion into acceleration of the projectile body decreases to 50% and downwards with added Al. The energy intake by the projectile body can be enhanced to 56% by optimizing the particle size distribution in PBXs.

4 Conclusion

In this study on detonation performance of PBXs, the detonation velocity was found to depend considerably on the filler particle size – within more or less 500 m/s of the mean value. It is assumed to be due to different possible mechanisms for detonation front propagation in PBXs: if the filler particle sizes do not reach the critical diameter of detonation, the PBX detonates as a homogeneous explosive to generate a classical ‘smooth’ front. As the filler particles increase in size, they can produce percolating clusters, along which the leading detonation forms a complex front pattern. In-depth studies of the described phenomenon open the door to controlling the detonation performance of PBXs and also make it possible:

- to control the detonation velocity of melt-cast PBX composites up to \( \pm 10\% \) of the mean value by only varying the filler particle size, without modifying the explosive formulation;
- to develop approaches to compounding melt-cast composite explosives that can increase the energy conversion of aluminized explosives into acceleration performance from 45-50 to 56%;
- to design composite explosives 10-12% superior to all the known counterparts in acceleration ability.

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