



Performance and Detonation Characteristics of Polyurethane Matrix Bonded Attractive Nitramines

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Abstract: Several cast-cured plastic bonded explosives (PBXs) based on cyclic nitramines bonded by a polyurethane matrix have been prepared and studied. The nitramines were ϵ -CL20 (ϵ -2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane, ϵ -HNIW), BCHMX (bicyclo-HMX, *cis*-1,3,4,6-tetranitro-octahydroimidazo-[4,5-d]imidazole), RDX (1,3,5-trinitro-1,3,5-triazacyclohexane) and HMX (1,3,5,7-tetranitro-1,3,5,7-tetraazacyclooctane). The detonation velocities were measured experimentally. The brisance of the prepared compositions was determined by the Kast method. The penetration performance of shaped charges filled with the prepared compositions was measured experimentally. The detonation parameters of the studied compositions and the individual explosives were calculated using the EXPLO5 thermodynamic code. It was concluded that CL20-HTPB has the highest detonation characteristics and performance of all of the prepared PBXs. BCHMX-HTPB is an interesting PBX with performance and detonation characteristics higher than those of RDX-HTPB. A linear relationship between the detonation pressures of the prepared PBXs and their performances in terms of the explosive brisance was observed; while the penetration depths formed by the shaped charge jets depended on the Gurney velocity of the studied PBXs samples.

Keywords: BCHMX, HNIW, detonation, shaped charge, brisance

1 Introduction

There is a growing body of literature that recognizes the importance of plastic bonded explosives in commercial blasting, demolition, and insensitive munitions (IM). Traditionally, it has been argued that polymer bonded explosives are particulate composites containing a high volume fraction of stiff crystalline

explosive particles in a compliant rubberlike viscoelastic binder [1]. To date, several studies have investigated the effects of explosive type [2, 3], explosive particle size [4-6], binder system [7-9] and their physical and/or chemical compatibility [10, 11] on energetic performance, safety and cost of plastic bonded explosives.

Plastic bonded explosives (PBXs) based on ϵ -HNIW (2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane, ϵ -CL-20) have been intensively investigated, as ϵ -CL-20 has excellent explosive performance [12, 13], high energy density, and good thermal stability [14].

Previous studies have reported that hydroxyl-terminated polybutadiene (HTPB) is regarded as the state of the art workhorse binder for such applications. However, a recent trend is to use energetic binders such as poly(3,3-bis(azidomethyloxetane) [poly(BAMO)], poly(3-azidomethyl-3-methyloxetane) [poly(AMMO)], nitrated HTPB (NHTPB), poly(NIMMO) and poly(GLYN) for PBXs in order to obtain better performance [1].

PBX compositions based on 1,3,5,7-tetranitro-1,3,5,7-tetraazacyclooctane (HMX), 2,4,6-triamino-1,3,5-trinitrobenzene (TATB), and 2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane (ϵ -CL-20) tend to be significantly more expensive than other conventional ones based on 1,3,5-trinitro-1,3,5-triazacyclohexane (RDX). A relatively new explosive *cis*-1,3,4,6-tetranitro-octahydroimidazo-[4,5-d]imidazole (bicyclo-HMX or BCHMX) [15, 16], which may become a useful and cost effective alternative to HMX and ϵ -HNIW has been introduced. It has been studied as a PBX based on a sylgard binder [17] and polymethyl methacrylate [18].

The fabrication of some plastic bonded explosive formulations was performed according to the procedure of Daniel [19]. A plastic bonded explosive containing BCHMX was prepared along with its RDX, HMX and ϵ -HNIW analogs to evaluate and compare the performance of such compositions. The detonation velocities of the prepared samples were experimentally determined and the main detonation characteristics were further calculated using the EXPLO5 computer code [20, 21].

The main goal was to determine the usability of BCHMX as a plastic bonded explosive and to compare it to other powerful PBXs based on RDX, HMX and ϵ -HNIW. In addition, the explosive performance was confirmed using small caliber shaped charges of the PBXs and therefore exhibited the explosive performance in terms of the observed penetration depth into mild steel targets. The brisance of BCHMX-HTPB was compared with those of the other studied PBXs.

2 Experimental

2.1 Materials

2.1.1 Polyurethane matrix system

The polyurethane matrix used in this study consisted of a hydroxyl-terminated polybutadiene (HTPB, R-45M from ARCO Co.) as the prepolymer, having a hydroxyl value of 0.84 meq/gram, and hexamethylenediisocyanate (HMDI, Shandong YuchengYiao Technology Co.Ltd., China) as the curing agent, with an NCO equivalence value of 11.83 meq/gram. The chemical bonding agent tris-1-(2-methylaziridinyl)phosphine oxide (MAPO, Hangzhou Yuhao Chemical Technology Co. Ltd., China) and plasticizer dioctyl adipate (DOA, Island Pyrochemical Industries, China) were used as received.

2.1.2 Military high explosives

RDX and HMX were obtained from Eurenco, Paris, France, with average particle sizes of about 64 μm and 42 μm , respectively. BCHMX was prepared in our laboratories by a two-stage synthetic process according to the Czech patent method [15], with an average particle size of about 18 μm . Technical-grade α -HNIW was a product of Heliopolis Company pilot plant. It was recrystallized in our department as detailed by Elbeih *et al.* [22, 23] with an average particle size of about 24 μm .

2.2 Preparation of cast cured PBXs

PBXs were prepared in a 4 L vertical sigma mixer. All liquid ingredients, HTPB, MAPO and DOA except the curing agent, were added and mixed for 20 min at 40 °C followed by vacuum mixing for a further 20 min to drive out entrapped air. The nitramine was then added in three portions during 30 min. Finally, the curing agent, HMDI, was added at 55 °C and the ingredients were mixed for a further 30 min. The mixture was then cast and cured at 60 ± 2 °C for seven days under vacuum. The cured HTPB binder system was prepared with an NCO/OH ratio of 1.3. MAPO was 0.5 wt.%, while DOA was 20 wt.% of the total weight of the binder system. The prepared PBXs contained 82 wt.% explosive and 18 wt.% binder system, and were designated as RDX-HTPB, HMX-HTPB, BCHMX-HTPB and CL20-HTPB.

2.3 Heat of combustion determination

The heat of combustion of the studied samples was determined as dry matter using an automatic adiabatic combustion calorimeter Laget MS 10 A under excess

oxygen pressure [24]. Once the samples were totally burned in the calorimetric vessel, the temperature increase values were converted to the recommended calculated heat of combustion values. The heats of combustion of the samples are reported in Table 1.

2.4 Velocity of detonation (VOD) measurements

The detonation velocities were measured using the instrument “Explomet-Fo-2000 Multichannel” (Kontinitro AG, Geneva, Switzerland) [24]. The tested samples were prepared in the form of sheets of 20 mm thickness, 40 mm width and 300 mm length. Three optical sensors (probes) were placed in each charge, with the first sensor (probe) being placed at a distance of 100 mm from the surface containing the detonator. Each of the other two probes were placed at a distance of 80 mm from the previous one. Charges were set off using an electrical detonator no. 8. The mean values of three measurements for each sample (max. $\pm 114 \text{ m}\cdot\text{s}^{-1}$) are reported in Table 1.

2.5 Detonation characteristics calculations

The theoretical detonation characteristics (detonation velocity, D , heat of detonation, Q , detonation pressure, P) of all of the studied compositions and the individual explosives were calculated using the EXPLO5 thermodynamic code [20, 21]. The BKWN set of parameters for the Becker-Kistiakowsky-Wilson (BKW) equation of state (EOS) was applied; these parameters are: $\alpha = 0.5$, $\beta = 0.298$, $\kappa = 10.50$, $\Theta = 6620$. The results of these calculations are presented in Table 1.

2.6 Brisance measurements (Kast method)

Brisance, measured by the Kast method produced by OZM Company, was used to determine the relative performance of the prepared compositions [25]. The deformation of a standard copper crusher (7 mm diameter and 10.5 mm high) was used to estimate the brisance of each composition. Each composition (2 g) was cast in an aluminum tube of height 30 mm, 12 mm internal diameter and 4 mm wall thickness. The aluminum tube was then placed on a solid steel cylinder, which served as a counter weight for the copper crusher. The charges were initiated by an electric detonator. The results are reported in Table 1, relative to TNT (as a reference explosive).

2.7 Shaped charge jet penetration performance

The shaped charges used had an internal diameter of 15 mm, length 55 mm, with casing made from aluminum alloy F28. The shaped charges were filled with the

prepared PBXs and a conical liner was pressed against them. The liner was made of copper-tellurium alloy, and had apex angle 60°, diameter 14.9 mm, length 23.5 mm and conical liner wall thickness 0.5 mm. The mass of each composition used to fill the charge was 4 g. The charges were filled with the prepared PBXs and left to cure in the presence of the liner. Semi-infinite mild steel plates were used as targets. All of the prepared charges were detonated over the target at a distance of 45 mm (three times the diameter of the liner, 3D stand-off distance). An electric detonator, containing 0.3 g lead azide and 0.4 g RDX, was used for the initiation of the prepared charges. The penetration performance of the charges was represented by the penetration depth formed in the target as a result of the jet penetration. The results are reported in Table 1 and photographs of the charge used and the experimental arrangement are presented in Figure 1.

Table 1. Measured and calculated parameters

Samples	Measured parameters					EXPLO5 code calculations			
	Loading density [g·cm ⁻³]	Experimental detonation velocity, D [m·s ⁻¹]	Internal energy of combustion (J·g ⁻¹)	Relative brisance (%TNT)	Depth of jet penetration [mm]	Detonation velocity [m·s ⁻¹]	$\frac{D_{\text{calc}}-D_{\text{exp}}}{D_{\text{exp}}/100}$ [%]	Detonation pressure [GPa]	Detonation heat [J·g ⁻¹]
RDX cryst.	1.76	8750 ^a	9522	-	-	8718	- 0.4	32.12	6085
HMX cryst.	1.90	9100 ^a	9485	-	-	9225	+ 1.4	38.00	6075
BCHMX cryst.	1.79	8650 ^b	9124	-	-	8840	+ 2.2	33.95	6447
ε-HNIW cryst.	1.98	9473 ^c	8311	-	-	9410	- 0.7	41.74	6465
RDX-HTPB	1.52	7526	14162	122	42	7449	- 1.0	20.06	5487
HMX-HTPB	1.57	7812	14118	125	47	7627	- 2.3	21.34	5453
BCHMX-HTPB	1.56	7746	13798	123	45	7593	- 1.9	21.23	5744
HNIW-HTPB	1.63	8167	13255	129	53	7919	- 3.0	23.73	5786

^a Taken from Ref. [8];

^b Taken from Ref. [16], measured for BCHMX phlegmatized with 3% Viton B;

^c Taken from Ref. [26].

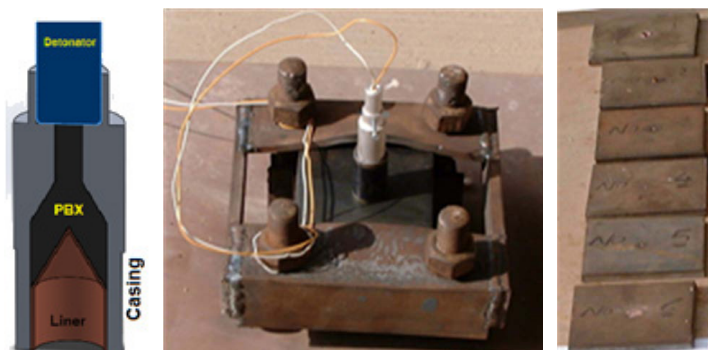


Figure 1. Arrangement for the shaped-charge jet penetration test

3 Results and Discussion

From the experimental detonation velocity measurements, a relationship between the charges densities and their detonation velocities was observed, as shown in Figure 2. It is clear that CL-20 has the highest detonation velocity of all of the studied samples and its PBX is the highest of all of the prepared PBXs. The new PBX based on BCHMX has a detonation velocity slightly lower than that of HMX-HTPB and higher than that of RDX-HTPB. There is no experimental measurement of the detonation velocity for pure BCHMX in the literature due to its high sensitivity; as a result it has been measured after phlegmatizing with 3% Viton B [16].

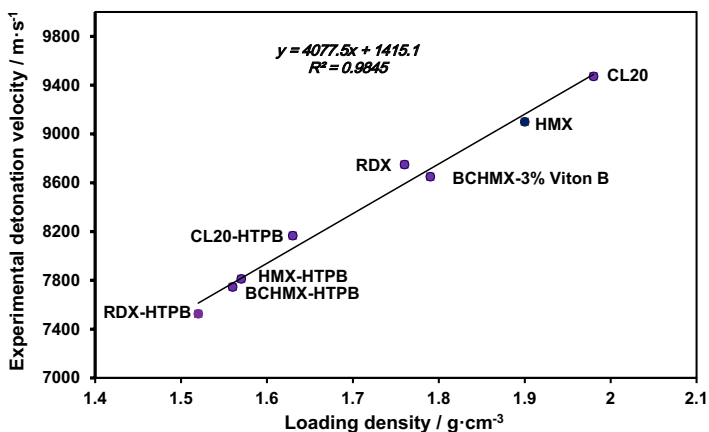


Figure 2. Relationship between the experimental detonation velocity and the loading density of the studied explosives

The experimentally measured detonation velocities, D , are plotted against the calculated ones in Figure 3. It is evident that the majority of the points lie below the line representing a perfect match. This means that the calculated results are slightly lower than the experimental ones. A good match was found for the calculated results (maximum deviations of 3% from experiment).

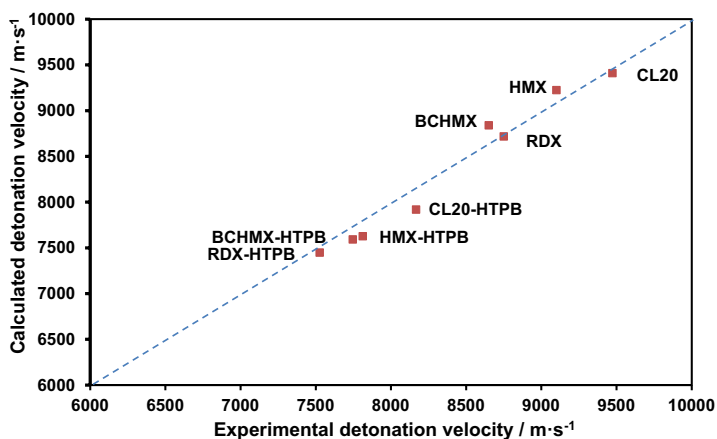


Figure 3. A comparison of the calculated detonation velocities with those measured

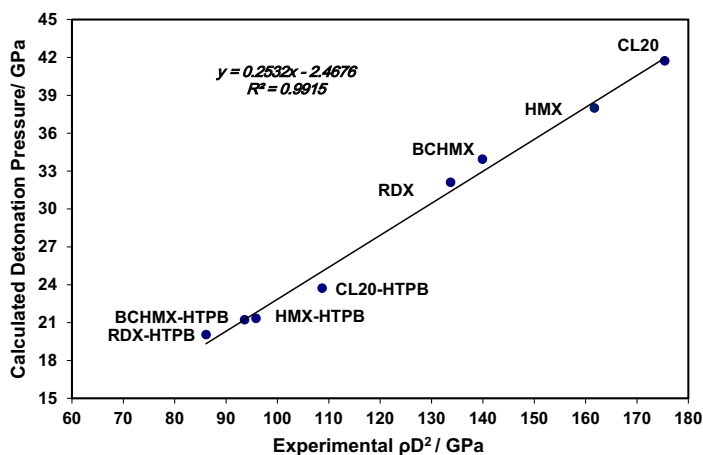


Figure 4. Relationship between the calculated detonation pressure and the square of the experimental detonation velocity multiplied by the loading density

According to explosion physics, the detonation pressure can be calculated

with respect to the detonation velocity as described in the literature [27]. An empirical equation exists based on multiplication of the initial density with the square of the detonation velocity and a constant. In order to check the compatibility of the calculated results with the experimental ones, values of the square of the detonation velocity and the initial density were plotted against the detonation pressure, as shown in Figure 4. A linear relationship was observed which included both the results of the pure explosives and their PBXs, and confirms the compatibility of the calculated results with the measured ones.

An inverse relationship between the values of the heat of detonation and the experimental heats of combustion of the studied samples is presented in Figure 5. It shows that the polyurethane matrix acts as a fuel in the prepared samples and increased the calorific value of the pure explosives, but decreased the detonation heat.

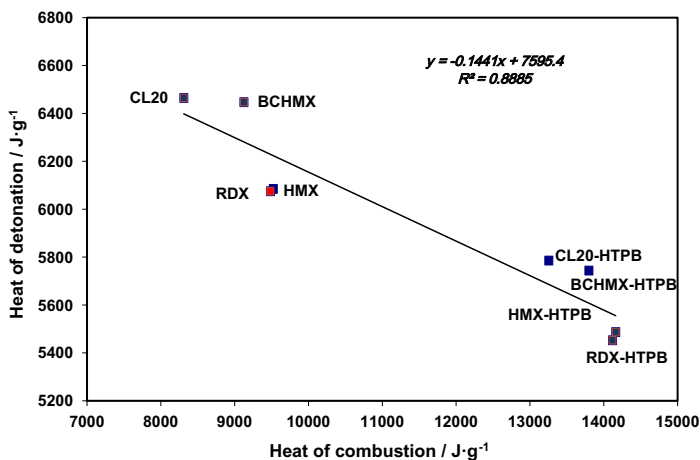


Figure 5. Relationship between the heat of detonation and heat of combustion of the studied explosives

The performance of the new BCHMX-HTPB PBX was compared with the other prepared PBXs using the relative brisance as measured by the Kast method. Figure 6 presents a comparison between the relative brisance of the prepared PBXs and the experimental ρD^2 . It is clear that CL20-HTPB has the highest brisance of the studied PBXs, while BCHMX-HTPB is higher than RDX-HTPB and lower than HMX-HTPB. The relation proved that the brisance of an explosive is strongly affected by its detonation pressure.

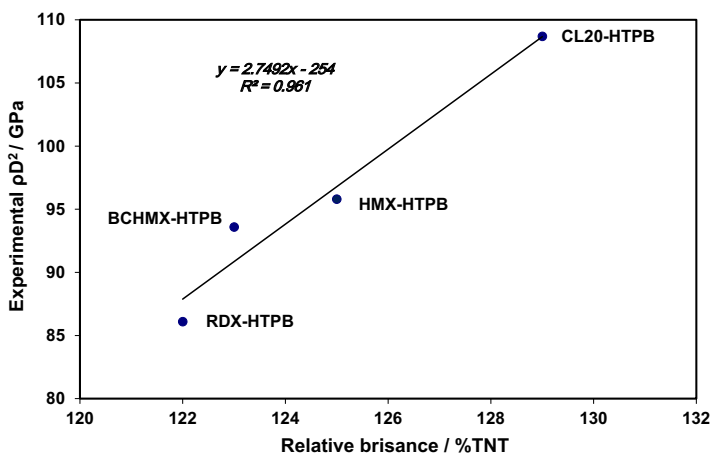


Figure 6. Relationship between the experimental ρD^2 (representing detonation pressure) and the relative brisance of the prepared PBXs

Initiation and detonation of a shaped charge filled with CL20-HTPB produced the largest penetration depth of all of the studied PBXs, with a penetration depth of 53 mm into the mild steel target. It had a higher penetration performance than HMX-HTPB by nearly 13%. BCHMX-HTPB had a higher penetration performance than RDX-HTPB when used in shaped charges. This can be attributed to the variation of the detonation velocity of the different PBX formulations, which in turn gives different values for the Gurney velocity ($\sqrt{2E}$). According to the fundamentals of shaped charges and the unsteady state PER theory, it was reported that all shaped charge jet velocities (*i.e.* stagnation, flow and jet velocities) represent an explicit function of the collapse velocity, which depends mainly on the Gurney velocity and the explosive metal geometry and explosive to metal mass ratios [28]. On the other hand, the Gurney velocity has been approximated more simply by Koch *et al.* [29], where they correlated the relation between the Gurney velocity and the detonation velocity of an explosive by the simple relation:

$$\sqrt{2E} = \frac{D}{3.08} \quad (1)$$

Locking [30] studied the relationships between the Gurney theories and the hydrodynamic work energy. He presented a general equation derived from both Cooper's and Koch's Gurney theories, which is:

$$VG_x = D_x / f_x \quad (2)$$

where $V_{G_x}(\sqrt{2E})$ is the Gurney velocity of an explosive, $D_x(D)$ is the detonation velocity of an explosive and f_x is a function of an explosive material (3.08 according to Koch). He stated that:

$$f_x = 18.0467 (1 + 1.3 \rho_x / 1000) / \rho_x^{0.4} \quad (3)$$

where ρ_x is the loading density of an explosive in $\text{kg}\cdot\text{m}^{-3}$. Locking [30] stated that varying the coefficient (f_x) gives a slightly improved Gurney velocity prediction when compared with both the Cooper and Koch theories. It can be observed from both theories that the higher the detonation velocity of an explosive, the higher the Gurney velocity and the jet tip velocity, and therefore the larger the penetration depth into the target material [31].

This phenomena has been investigated by a linear relationship between the jet penetration depth into a semi-infinite mild steel target and the corresponding calculated Gurney velocity of the PBX formulation by both the Koch and Locking equations, as shown in Figure 7. This relationship confirms that the explosives having high detonation velocities are the best choice for filling shaped charges and produce the largest jet penetration depth into the target material. BCHMX-HTPB has a penetration ability higher than RDX-HTPB.

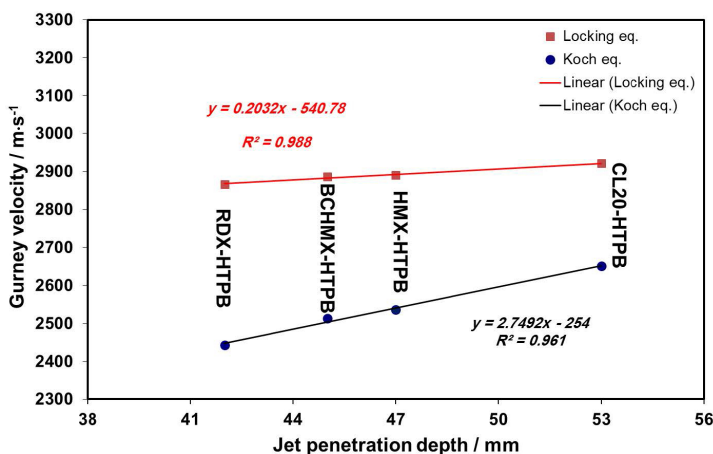


Figure 7. Relationship between the jet penetration depths obtained from the shaped charges and the calculated Gurney velocity obtained by both Locking, Equation 3, and Koch, Equation 1

The Gurney velocity, calculated by Locking, Equation 3, is higher than that calculated by Koch, Equation 1, where the difference between the results increases as the loading density of the studied explosive decreases. It seems from the relation that the calculated Gurney velocity by both the Locking and Koch equations will be close to each other for explosives at high loading densities.

4 Conclusions

The measured detonation velocities can be arranged in the following decreasing order: HNIW-HTPB > HMX-HTPB \geq BCHMX-HTPB > RDX-HTPB. It was shown that the detonation velocities, D , calculated by the EXPLO5 code are close to the experimental values. The correlation between the detonation pressures (represented by the experimental ρD^2) of the studied PBXs and their shattering effect (brisance) was confirmed. Small scale shaped charges filled with CL20-HTPB had the highest penetration ability of the studied PBXs, and a correlation between the penetration ability of the studied explosives and the detonation velocity (represented by the Gurney velocity) was presented. BCHMX-HTPB has a detonation heat of the same level as CL20-HTPB and higher than the other PBXs studied. Addition of a polyurethane matrix to explosives increases their heat of combustion and decreases their detonation heat. BCHMX-HTPB has detonation characteristics and performance higher than RDX-HTPB, while CL20-HTPB is the most powerful PBX of all of the prepared samples.

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