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Explosive Properties of a High Explosive Composition Based on Cis-1,3,4,6-tetranitrooctahydroimidazo-[4,5-d] imidazole and 1,1-Diamino-2,2-dinitroethene (BCHMX/FOX-7)

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Abstract: In this work, preparation and characterization of a plastic bonded explosive (PBX) based on a mixture of cis-1,3,4,6-tetranitrooctahydroimidazo-[4,5-d]imidazole (BCHMX) and 1,1-diamino-2,2-dinitroethene (DADNE or FOX-7) bonded by poly-dimethyl siloxane binder (PDMS) were investigated. Impact and friction sensitivities were determined. Detonation velocity was measured and the detonation characteristics were determined by the EXPLO 5 thermodynamic code. Relative explosive strength was determined using the ballistic mortar test. For comparison, four individual cyclic nitramines, RDX (1,3,5-trinitro-1,3,5-triazinane), β-HMX (β-1,3,5,7-tetranitro-1,3,5,7-tetrazocane), BCHMX and ε-CL20 (ε-2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane, ε-HNIW) in addition to their PBXs bonded by the same polymeric matrix were studied. Also traditional plastic explosives such as Czech plastic explosive (SEMTEX 10) and Egyptian Plastic Explosive (EPX-1) were included in the study. The results confirmed that BCHMX/FOX7-Sil has the lowest impact and friction sensitivity of all the studied samples and its detonation velocity and pressure are higher than the traditional PETN plastic explosives.

Keywords: BCHMX · FOX-7 · nitramines · sensitivity · detonation characteristics

1 Introduction

The increased interest in novel energetic materials and mixtures with high performance and lower sensitivity to external stimuli became a passion for developers and researchers. However, increasing the explosive performance is usually accompanied by an increase in the sensitivity [1–4]. Licht [2] has pointed out that this statement cannot be proved by any theory. Therefore, exceptions are possible, i.e. explosives which approach the optimum explosive compositions (high explosive strength combined with high safety), are exemplified in Refs.[2, 4, 5]. cis-1,3,4,6-Tetranitrooctahydroimidazo-[4,5-d]imidazole (BCHMX) is an interesting explosive which has been prepared by our team in a two-stage synthesis method according to the patents [6, 7]. It has a high performance compared with traditional explosives but its sensitivity is relatively high – on the level of penterythritol tetranitrate (PETN) [8]. Some aspects of this problem were documented and studied using the sensitivity and performance data of different cyclic nitramines [9, 10] and their plastic bonded explosives (PBXs) [3, 4, 11, 12]; these explosives are based on 1,3,5-trinitro-1,3,5-triazinane (RDX), β-1,3,5,7-tetranitro-1,3,5,7-tetrazocane (β-HMX), ε-2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane (ε-HNIW, ε-CL-20) and BCHMX. Different publications presented the effect of several polymeric matrices on the performance of the explosives such as Viton A 200 [3, 11, 13], polydimethylsiloxane [3, 11], plasticized acrylonitrile-butadiene rubber (NBR) [3, 11, 14, 15], plasticized polyisobutylene (PIB) [11, 16] and plasticized poly-(methyl methacrylate) (PMMA) [16] and HyTemp acrylate [17] Regarding to the recent study [11], it was concluded that the polydimethyl-siloxane(PDMS) matrix represents the optimum binder to be used with explosives: it does not affect the thermal stability of the nitramines studied, it has roughly the same influence on the thermochemistry of detonation (according to the volume heat of explosion) as those obtained with PBXs bonded by Fluorel binder. Moreover, this PDMS matrix should increase the safety of explosives against mechanical impulses by adding 1,1-diamino-2,2-dinitroethene (FOX-7) [18, 19]. Therefore, in the framework of this study of the new BCHMX applications we have inserted in our attention a modified plastic explosive on its base with FOX-7 bonded by PDMS binder which is a topic of this paper. There is no information in the literature about explosive mixture based on BCHMX with FOX-7. As a result, sensitivities, detonation characteristics and performance of

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this composition are studied in this paper. Explosive characteristics of the new explosive mixture are compared with those of plastic explosives based on PETN (Czech explosive SEMTEX 10 and new Egyptian explosive EPX-1) and with analogous PBXs, filled by RDX, β-HMX, BCHMX and ε-CL20 and also with individual Explosives: PETN, BCHMX, ε-CL20, β-HMX and RDX.

2 Experimental Work

2.1 Materials and Preparation Method

BCHMX is a white crystalline powder. It was prepared at the Institute of Energetic Materials as reported in Refs.[6, 7] and it was recrystallized using solvent/antisolvent technique by acetone/heptane to obtain crystals with uniform shape and small particle size. FOX-7 is a yellow crystalline substance with a density of 1.88 $g\,cm^{-3}$ and it was prepared in our laboratories by the nitration of 2-methylpyrimidine-4,6-diol and intermediate 2-(dinitromethylene)dihydro-5,5-dinitropyrimidine-4,6(1H,5H)-dione, which hydrolyses to FOX-7 and CO_2 in the presence of dinitromethane [20–23].

Polydimethylsiloxane binder (PDMS, Si-binder) was used as a thermoplastic polymeric matrix for bonding of the prepared explosive mixture. Two types of these PDMS polymers were used. The first had an average molecular mass of 37.5 $kg\,mol^{-1}$ and a polydispersity index of 1.62 (Wacker® AK 10000). The second type had a molecular mass of 72.9 $kg\,mol^{-1}$ and a polydispersity index of 1.57 (Wacker® AK 60000). These two types were used in a ratio of 1:1 to obtain the desired average molecular mass and viscosity of the polymeric matrix. Using computerized mixer plastograph BRABENDER, the solid explosive ingredients were mixed together (BCHMX 44 mass%: FOX-7 44 mass%) then the polymeric matrix (12 mass%) was added at 25 °C for 90 min kneading time. The prepared plastic explosive was extruded to form cylindrical explosives with 21 mm diameter.

2.2 Elemental Analysis

Fisons – EA-1108 CHNS-O elemental analyzer was used for the determination of C, H, and N in the prepared PBX. The results of the elemental analysis were recalculated to match the nitrogen content to the individual explosive. By this way, the calculated formula was used as if it was an individual explosive and it was used as an input data for the EXPLO5 code to calculate the detonation parameters.

2.3 Heat of Combustion Determination

An automatic high pressure Bomb calorimeter, model BCA 500, OZM Company, Czech Republic, was used to measure the heat of combustion of the BCHMX/FOX7-Sil. The sample was placed in a closed bomb filled with an excess of oxygen [24] and ignited. The output data was used for calculation of the enthalpy of formation of the BCHMX/FOX7-Sil, which was then used for determining the detonation characteristics.

2.4 Sensitivity to Impact and Friction

The standard impact tester with exchangeable drop weight of BAM impact sensitivity instrument [25] was used ; the amount of substance tested was 50 mm^3, and drop hammers of 2 and 5 kg weight were used. The probity analysis was used to determine the probability levels of the initiation. Only the 50% probability of initiation (H50) is used and is reported in Table 1. A BAM friction test apparatus was used to determine the sensitivity to friction using the standard test conditions [25]. The sensitivity to friction was determined by spreading about 0.01 g of the studied explosive on the surface of the porcelain plate in the form of a thin layer. Different loads were used to change the normal force between the porcelain pistil and the plate. Sample initiation was observed through sound, smoke appearance, or by the characteristic smell of the decomposition products. Using the Probit analysis [26], only the normal force at which 50% of initiations occurred is reported in Table 1 as the friction sensitivity.

2.5 Detonation Velocities Measurements

The detonation velocity was measured by an EXPLOMET-FO-2000 produced by KONTINITRO AG. The composition tested was prepared in the form of cylinders with 21 mm diameter and 200 mm length. Three optical sensors were placed in each charge, with the first sensor being placed at a distance of 50 mm from the surface containing the detonator. Each of the other two sensors was placed at a distance of 60 mm from the previous one. Charges were set off using a no. 8 detonator. Three measurements were performed for the composition and the mean value (max. $\pm 68\ m\,s^{-1}$) is reported in Table 2.

2.6 Calculation of the Detonation Characteristics

The theoretical detonation characteristics (detonation velocity, D, heat of detonation, Q, and detonation pressure, P) of the prepared compositions, as well as those of the individual explosives, were calculated by the EXPLO5 code version 5.04 [30]. The following Becker-Kistiakowsky-Wilson equation of state (BKW EOS) was used with BKWN set of parameters $\alpha = 0.5$, $\beta = 0.176$, $\kappa = 14.71$, $\Theta = 6620$ [30]. The detonation heat is the heat released in a constant volume explosion and is determined by subtracting the heats of for-

Table 1. Results of the experimental measurements on the samples studied.

Code designation	Summary formula	Heat of combustion [$J\,g^{-1}$]	Enthalpy of formation [$kJ\,mol^{-1}$]	Impact sensitivity [J]	Friction sensitivity [N]
BCHMX	$C_4H_6N_8O_8$	9124	236.5	3.2 [a]	88[a]
β-HMX	$C_4H_8N_8O_8$	9485	77.3	6.4[a]	95[a]
RDX	$C_3H_6N_6O_6$	9522	66.2	5.6[a]	120[a]
RSε-CL20$_{(10.8)}$ [b]	$C_6H_6N_{12}O_{12}$	8311	397.8	10.8[a]	69[a]
RSε-CL20$_{(11.2)}$ [b]		8311	397.8	11.2[c]	84[c]
ε-CL20		8311	397.8	4.1[a]	64[a]
FOX-7	$C_2H_4N_4O_4$	8749	−63.75	23.7	> 360
PETN	$C_5H_8N_4O_{12}$	8182[d]	−538.7[d]	2.9[e]	46[e]
BCHMX-Sil.	$C_{5.02}H_{9.18}N_8O_{8.47}Si_{0.54}$	11328	−27.16	24.3[a]	232[a]
HMX-Sil.	$C_{5.05}H_{11.14}N_8O_{8.5}Si_{0.54}$	11647	−172.1	27.6 [a]	228[a]
RDX-Sil.	$C_{3.78}H_{8.53}N_6O_{6.37}Si_{0.406}$	11664	−129.3	31.9 [a]	254[a]
ε-CL20-Sil.	$C_{7.54}H_{10.87}N_{12}O_{12.77}Si_{0.8}$	10672	27.4	26.0 [a]	192 [a]
BCHMX/FOX7-Sil.	$C_{4.52}H_{9.82}N_6O_{7.36}Si_{0.446}$	11863	−286.5	42.3	> 360
EPX-1	$C_{7.88}H_{12.36}N_4O_{12.59}$	11528[d]	−666.5[d]	13.9[e]	176[e]
SEMTEX10	$C_{8.05}H_{12.64}N_4O_{12.37}$	11942[b]	−646.8[b]	15.7[d]	204[d]

a) [11]; b) product with reduced sensitivity prepared in the sense of patent [27]; c) measured in framework of this paper d) [28]; e) [29]

Table 2. Performance and detonation parameters of the samples studied.

Studied sample No.	Code of samples	Experimental Density ρ [$g\,cm^{-3}$]	Relative explosive strength (% of TNT)	Calculated detonation parameters by Explo5 Detonation velocity [$m\,s^{-1}$] D_{exp}	$D_{cal.}$	Error %	Detonation pressure P [GPa]	Detonation heat Q [$kJ\,kg^{-1}$]
1.	BCHMX	1.79[a]	–	8650	8840	+2.19	33.9	6447
2.	β-HMX	1.90[b]	–	9100	9225	+1.37	38.0	6075
3.	RDX	1.76[b]	–	8750	8718	−0.40	32.1	6085
4.	RSε-CL20$_{(10.8)}$	1.98	–	9473	9407	−0.60	41.7	6455
5.	RSε-CL20$_{(11.2)}$	1.98	–	9473	9407	−0.60	41.7	6455
6.	ε-CL20	1.98[b]	–	9473	9407	−0.60	41.7	6455
7.	FOX-7	1.78[c]	–	8325[c]	8260	−0.78	27.7	4942
8.	PETN	1.70[d]	–	8400[d]	8318	−0.97	28.5	6160
9.	BCHMX sil.	1.62[e]	137.5	7994[e]	7742	−2.82	24.2	6245
10.	HMX sil.	1.64[e]	136.4	8083[e]	7781	−3.73	24.3	5994
11.	RDX Sil	1.58[e]	136.0	7822[e]	7560	−3.34	22.2	5958
12.	ε-CL20 sil.	1.74[e]	141.1	8267[e]	8023	−2.95	27.6	6292
13.	BCHMX/FOX7-Sil.	1.59	123.5	7889	7438	−5.71	21.3	5507
14.	EPX-1	1.55[f]	–	7636[f]	7398	−3.10	21.17	5742
15.	Semtex10	1.52[d]	132.8	7486[d]	7370	−1.50	20.89	5708

a [8], b [32], c [33], d [28], e [11], f [34]

mation of the explosive (reactants) from the sum of the heats of formation of the detonation products. The theoretical calculation and the error between the experimental results are listed on Table 2.

2.7 Relative Explosive Strength Measurements

A ballistic mortar test was used for the determination of the relative explosive strength of the samples studied, using TNT as a reference [25,31]. A fixed amount of a tested explosive (10 g was wrapped in polypropylene foil and inserted into the mortar enclosed by a steel projectile and fired using a nonelectric detonator (No. 8). The determination is based on measuring the swing angle of the pendulum and by comparing the measurement with a calibration curve for the standard explosive (TNT) at different masses. The explosive strength of the tested explosive is thus expressed relative to TNT (relative explosive strength, RS as % of TNT) [31]. For each measurement, a part of the non-electric detonator is inserted in the plastic sample and fired by a match. Three measurements were made for each sample and the mean values are reported in Table 2.

3 Results and Discussion

The performance of explosive material can be correlated with its impact and friction sensitivity which was represented in recently accepted literature [4,35], Generally, the high performance of an explosive material is accompanied by an increase of explosive sensitivity. However, modern techniques are used to combine the high values of performance accompanied by the reduction of sensitivity. Therefore, the plastic explosive mixture is one of the methods to combine the performance and the sensitivity reduction. In order to study the sensitivities of the studied samples, a comparison between the impact sensitivity (initiation due to uniaxial compression) and the friction sensitivity (initiation due to shear slide of a fixed volume) is shown in Figure 1, of all the studied samples which is in agreement with the results of papers [4,36,37] (relatively close semilogarithmic relationships between these sensitivities of nitramine explosives).

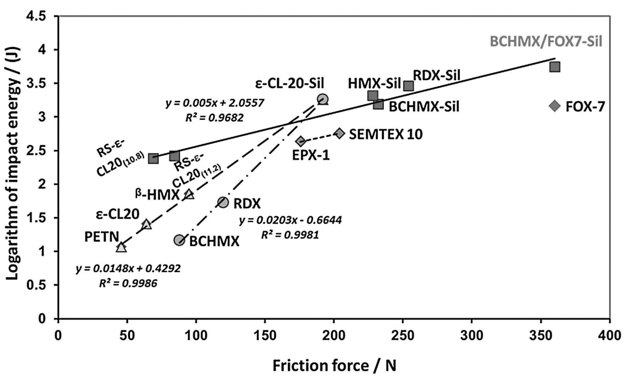

Figure 1. Semi-logarithmic relationship between friction and impact sensitivities – numbers in parentheses near RS-ε-CL20 mean impact sensitivity in J (more about this type of relationship is in Ref [4,37]).

It was observed that the samples could be divided into three groups; the first group includes the pure explosives PETN, technical ε-CL-20, β-HMX and technical ε-CL-20 incorporated into PDMS. The second group with the less sensitive to friction: BCHMX, RDX and again technical ε-CL-20 incorporated into PDMS. The third group contains PBXs based on PDMS (including the new BCHMX/FOX7-Sil), at which the sensitivity data correlate with two kinds of crystalline ε-CL-20 with reduced sensitivity (RS [27]).

The good crystal structure of the RS-ε-CL-20, in comparison with its technical quality, is the reason of this correlation. The effect of the silicone matrix on the sensitivity of the studied nitramines appeared clearly with the third group, where the impact sensitivity of all the studied individual explosives bonded by this matrix is in the range of 24–42 J and their friction sensitivity in the range of 232–360 J (taken without crystalline RS-ε-CL-20), which also clarifies the effect of the silicone matrix to decrease the sensitivities compared with the traditional plastic explosives (Semtex10 and EPX-1). The addition of the low sensitivity FOX-7 to BCHMX-Sil affects sensitivity and forms a new low sensitivity plastic explosive BCHMX/FOX7-Sil with an impact sensitivity 42.3 J and a friction sensitivity >360 N which are lower than all the studied PBXs. The new plastic explosive could be considered as a candidate for Insensitive munitions (IMs) or Low Vulnerable Ammunition (LOVA) and might have applications in warheads. In order to check the performance of this new plastic explosive; the detonation velocity, the relative strength (compared with TNT) and the detonation characteristics were determined.

An interesting relationship between volume heat of explosion (the product of loading density and the detonation heat $\rho.Q/\mathrm{MJ\,m^{-3}}$) compared with the logarithm of impact sensitivity [4,37] is presented in Figure 2.

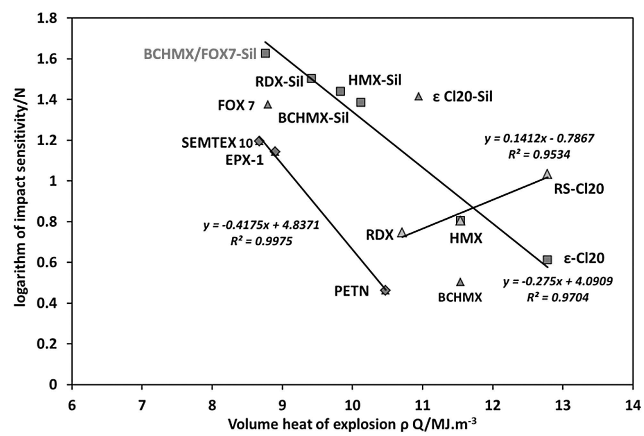

Figure 2. Semi-logarithmic relationship between impact sensitivity and volume heat of explosion (more about this type of relationship is in Ref [4]).

Also three groups are presented on this figure. The first group with a positive slope includes the pure nitramines which are sensitive compared with the other studied samples but have a high volume heat of explosion, in this case a partial relationship has opposite running as is discussed in papers [4]; the second group includes all the PBX samples based on the nitramines and the new BCHMX/FOX7-Sil except CL20-Sil.

This result was discussed in paper [38] where the detonation chemistry of PBXs with the CL20 filler can be different compared with other plastic explosives, which are filled by other nitramines. All the PBXs have low impact sensitivity (due to the effect of the silicone binder) compared with the individual explosives. The pure PETN and its plastic bonded explosives, EPX1 and SEMTEX 10, formed the third group which have higher sensitivity to impact than the other studied PBXs. The new BCHMX/FOX7-Sil has the lowest sensitivity to impact but with low performance compared with the other nitramines' PBXs. This figure confirms the general finding that an increase in the performance (volume heat of detonation) is usually accompanied by an increase in the sensitivity of explosives (with exception for RDX, HMX and pure ε-CL-20 – see in paper [4]).

The measured densities and detonation velocities were recorded in Table 2 and the results are presented in Figure 3 where the well-known relationship between the density and the detonation velocity was confirmed [39]. From this figure it is clear that ε-CL-20 has the highest detonation velocity of the studied samples and as predicted the silicone matrix decreased the detonation velocity of each individual explosive. The new BCHMX/FOX7-Sil has a detonation velocity slightly lower than BCHMX-Sil (by 105 $m\,s^{-1}$) but clearly higher than the traditional plastic explosives and even higher than RDX-Sil. It means that FOX-7 has a slightly negative effect of the detonation velocity of BCHMX-Sil but a great positive effect on its sensitivity.

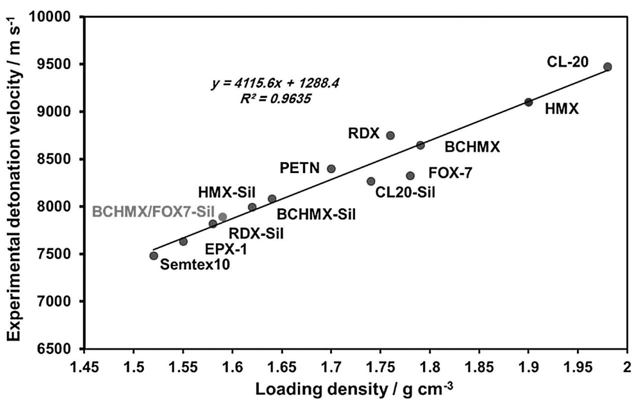

Figure 3. A relationship between the density and the detonation velocity of all the studied explosives.

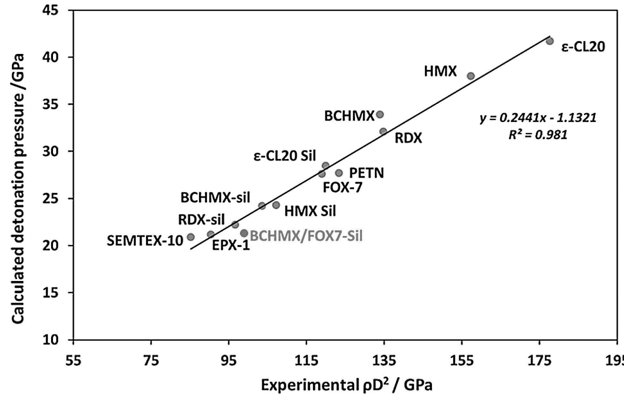

Figure 4. Relationship between calculated detonation pressure and the square of the experimental detonation velocity multiplied by the loading density.

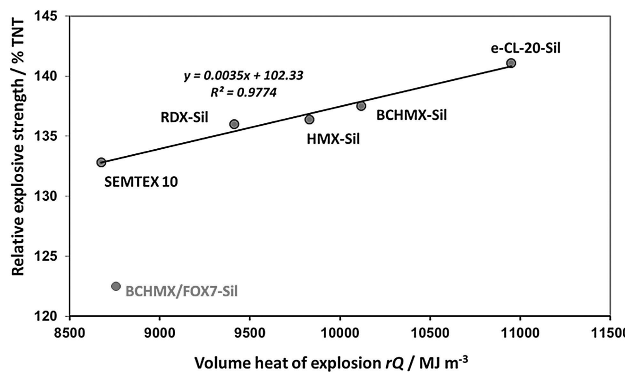

Figure 5. Relationship between relative explosive strength and volume heat of explosion (Q in $kJ\,kg^{-1}$ was calculated by the EXPLO5 code) of the studied PBXs.

Regarding to the physics of explosion, the detonation pressure of the explosives could be correlated with the products of the square of the detonation velocities and the loading density. Figure 4 presents a comparison between the calculated detonation pressures (by EXPLO5 code) with the products of the square of the experimental detonation velocities and the loading densities. A linear relationship between these data was observed with correlation coefficient of $R^2 = 0.981$ which confirms the agreement of the calculated results by EXPLO5 with the experimental measurements. Also in order to check the performance practically, the ballistic mortar was used to determine the explosive strength of the new PBX and to compare the result with other PBXs.

Figure 5 presents a relationship between the volume heat of explosion, calculated by EXPLO5 (here Q in $kJ\,kg^{-1}$) and the relative explosive strength (%TNT) measured by ballistic mortar.

From Figure 5, the reasonable dependence of the relative explosive strength on the heat of detonation was confirmed. BCHMX/FOX7-Sil has a relative strength of 123% TNT which confirm the high performance of the new PBX but it has the lowest result of all the studied PBX. Actually we didn't find the relative explosive strength of EPX-1 but it is clear that Semtex 10 (PETN based) has a higher explosive strength, it means that FOX7 decreased the explosive strength of BCHMX-Sil but still BCHMX/FOX7-Sil is higher than the traditional TNT. The overall results confirm that mixing of FOX7 with BCHMX in PBX bonded by Silicone matrix reduced the sensitivity of BCHMX. The detonation characteristics are higher than the traditional plastic explosives.

4 Conclusion

A relatively sensitive explosive cis-1,3,4,6-tetranitrooctahydroimidazo-[4,5-d]imidazole (BCHMX) can be improved against mechanical impact by incorporation into a polydimethylsiloxane matrix (PDMS). Another increase of resistance against these impacts is reached by incorporation of 1,1-diamino-2,2-dinitroethene (FOX-7) into the obtained plastic bonded explosives (PBX), named BCHMX/FOX7-Sil. Admixture of FOX7 decreases the explosive strength of original BCHMX-PDMS mixture which is, however, in the case of BCHMX/FOX7-Sil still higher than the traditional TNT. The overall results confirm that mixing of FOX7 with BCHMX in PBX, bonded by PDMS, changes the sensitivity of BCHMX to a range to be considered for IM explosives. The detonation velocity and pressure of this new PBX are higher than the traditional PETN plastic explosives. The calculated detonation characteristics of this PBX by the EXPLO5 thermodynamic code are in good agreement with the experimental values.

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