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Preparation, Crystal Structure and Properties of a New Crystal Form of Diammonium 5,5'-bistetrazole-1,1'-diolate

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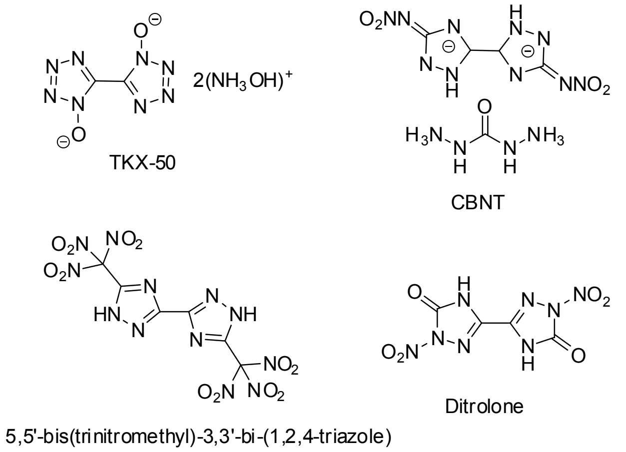
A new crystal form of diammonium 5,5'-bistetrazole-1,1'-diolate (**1**) was prepared by two novel methods and fully characterized by using IR, NMR spectroscopy, elementary analysis, single crystal X-ray crystallography and thermal gravity/differential thermal analysis (TG/DTA). Crystalline **1** was found as monoclinic and space group of *P*21/*c* (14). The TG/DTA analysis showed that the decomposition temperature of **1** was 287.8 ℃ with a mass loss of 91.2% in the range of 220—300 ℃ at a heating rate of 5 ℃/min. The sensitivities test towards impact, friction of **1** indicated that **1** has much lower sensitivities than those of RDX/HMX and is comparable to those of TNT, which suggested that **1** could be used as a good candidate of new insensitive energetic compound.

Keywords energetic material, diammonium 5,5'-bistetrazole-1,1'-diolate, crystal form, thermal decomposition, explosive performances

Introduction

Energetic materials (explosives, propellants and pyrotechnics) are used extensively both for civil and military applications. Recently, research in the field of energetic materials faced a profound change because of many catastrophic explosions resulting from unintentional initiation of munitions by aboard ships, aircraft carriers, munitions trains, impact or shock.[1,2] Additionally, concerns about the environmental impact of energetic nowadays have caused a renaissance in the synthesis of so-called green energetic materials, which are based on the high nitrogen content that release mainly environmentally benign N_2 after decomposition.[3-6] Whereas, traditional energetic materials decompose into a significant amount of toxic products after degradation, the use of nitrogen-rich azoles such as triazole and tetrazole derivatives avoid such drawbacks.[7] Meanwhile, it can also increase the heat of formation caused by C—N and N—N bonding in nitrogen-rich azoles molecules.[8] Recent studies on C—C connected heterocycles like bistetrazoles revealed excellent characteristics regarding stability and detonation properties,[9-11] as most extensively reported by Klapötke *et al.* on the synthesis and characterization of a series of nitrogen-rich salts of 5,5-bistetrazolates and 3,3-bistriazolates,[12-14] such as carbonic dihydrazidinium bis[3-(5-nitroimino-1,2,4-triazolate)], dihydroxylammonium (TKX-50), (CBNT) *etc.* (see in Scheme 1).

Scheme 1 The structure of nitrogen-rich salts of 5,5-bistetrazolates and 3,3-bistriazolates



Diammonium salt of 5,5'-bistetrazole-1,1'-diolate (**1**), one potential nitrogen-rich azole bearing C—C connected bistetrazoles was firstly reported by Klapötke *et al.*[15] with good explosive performances, such as denotation velocity 8817 m/s and thermal decomposition temperature 290 ℃. It has appeared promising application value in high energy explosive with low sensitivities.

Hence, a new crystal form of **1** was prepared by new synthetic procedures and the obtained crystalline was fully characterized in this study. To evaluate its energetic performances, the explosive properties and ther-

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mal behavior of **1** are also presented. As expected, **1** was believed to be an excellent candidate of new insensitive high energetic compound.

Experimental

Caution! Diazidoglyoxim, 1-*H*-5,5'-bistetrazole-1,1'-diol and diammonium salt of 5,5'-bistetrazole-1,1'-diolate are energetic with increased sensitivities towards shock and friction. Therefore, proper safer precautions (safety glass, face shield, earthed equipment and shoes, gloves, and ear plugs) have to be applied when synthesizing and handling the described compounds.

Infrared spectra were measured with a Bruker Spectrum One FTIR spectrometer as KBr. ^{1}H NMR, ^{13}C NMR were recorded with a Bruker instrument. The chemical shifts quoted in the text refer to typical standards such as tetramethylsilane. To measure elemental analyses, Vario EL III was employed. To determine the thermolysis performance of target compounds, SHIMADZU DTG-60 (heating rate 5 ℃/min) was used.

Glyoxime (**3**): In a 500 mL round-bottomed-neck flask equipped with a thermometer and a dropping funnel, NaOH (55.0 g, 1.4 mol) was dissolved in water (150 mL) and the solution was cooled to 0—5 ℃ in a salt ice bath. Hydroxylammonium chloride (139 g, 2.0 mol) was added while stirring. To the above mixed solution, glyoxal (145 g, 1.0 mol, 40% w/w in H_2O) was added dropwise while the temperature was kept below 10 ℃ over 30 min. After complete addition of the glyoxal, the solution was further stirred for 4 h at room temperature. The solid was removed by suction filtration and washed with a little ice water to remove the remaining sodium chloride. The white residue was recrystallized from methanol to afford glyoxime as white crystals (69.0 g, 78.0 %); m.p. 175—177 ℃ (lit.[18] 172—174 ℃); ^{1}H NMR (300 MHz, DMSO-d_6) δ: 11.4 (s, 2H), 7.71 (s, 2H); ^{13}C NMR (300 MHz, DMSO-d_6) δ: 145.1.

Method A

Diazidoglyoxime (**4**): Chlorinated agent (114 mmol) was added to a stirring solution of glyoxime (5.0 g, 57.0 mmol) in DMF (100 mL) at 0 ℃. The reaction mixture was stirred at room temperature for 2 h. After the reaction was complete, the solution was cooled to 0 ℃ in a salt ice bath, 1.0 mol•L^{-1} sodium azide solution was added. The precipitate was filtered, washed with 100 mL of water and air-dried. The solid was recrystallized from ethanol to afford diazidoglyoxime as colorless crystals (8.1 g, 84.0 %).

DTA-TG (5 ℃/min): 170 ℃ (dec.). IR (KBr) v: 3210 (w), 2169 (w), 2124 (w), 1623 (w), 1402 (w), 1363 (w), 1288 (m), 1013 (vs), 931 (s), 920 (s), 855 (s), 734 (s) cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6) δ: 12.1; ^{13}C NMR (300 MHz, DMSO-d_6) δ: 136.2; EA (found, calcd for $C_2H_2N_8O_2$, M_r=170.09): C (14.14, 14.12), H (1.39, 1.19), N (66.15, 65.88).

5,5'-Bistetrazole-1,1'-diol dehydrate (**5**): Diazidoglyoxime (3.0 g, 17.6 mmol) was suspended in diethyl ether (60 mL), which was cooled to 0—5 ℃ in a salt ice bath, HCl gas was bubbled through the reaction mixture at 0 ℃ while stirring for 1 h and then the flask was sealed and stirred at room temperature for 24 h. The HCl overpressure was released carefully, then the solvent was removed under vacuum on a rotary. After most of the diethyl ether had evaporated, water (100 mL) was added to result in a solution. A remaining colorless precipitate indicated uncompleted conversion of diazidoglyoxime to 5,5'-bistetrazole-1,1'-diol. The water was evaporated again on a rotary to remove the remaining HCl and diethyl ether, thus yielding crude as colorless solid, which could be purified by recrystallization from water, yield 3.4 g (16.5 mmol, 93.6%).

DTA-TG (5 ℃/min): 210 ℃(dec.). IR (KBr) v: 3327 (m), 2439 (w), 1668 (m), 1412 (w), 1377 (w), 1310 (w), 1208 (w), 1143 (m), 995 (s), 916 (w), 807 (s), 714 (w) cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6) δ: 7.78; ^{13}C NMR (300 MHz, DMSO-d_6) δ: 135.6; EA (found, calcd for $C_2H_6N_8O_4$, M_r=206.12): C (11.49, 11.65), H (2.41, 2.93), N (53.82, 54.36).

Diammonium 5,5'-bistetrazole-1,1'-diolate (**1**): 5,5'-Bistetrazole-1,1'-diol dehydrate (4.1 g, mmol) was dissolved in 100 mL of water, aqueous ammonia (4 mol/L, 20 mL) was added to the above solution while stirring. The mixture was maintained for 1 h at room temperature, the colorless crystalline residue was then filtered and air dried, which could be purified by recrystallization in water, yield 2.9 g (14.2 mmol, 71.1%).

DTA-TG (5 ℃/min): 287.8 ℃ (dec.). IR (KBr) v: 3187 (s), 3049 (s), 2153 (w), 1818 (w), 1668 (m), 1433 (s), 1353 (s), 1310 (w), 1230 (s), 1043 (m), 997 (m), 726 (m) cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6) δ: 7.12; ^{13}C NMR (300 MHz, DMSO-d_6) δ: 134.1; EA (found, calcd for $C_2H_8N_{10}O_2$, M_r=204.15): C (11.59, 11.77), H (3.82, 3.95), N (68.55, 68.61).

Method B

Sodium 5,5'-bistetrazole-1,1'-diolate tetrahydrate (**6**): Chlorinated agent (114 mmol) was added to a stirring solution of glyoxime (5.0 g, 57 mmol) in DMF (100 mL) at 0 ℃. The reaction mixture was stirred at room temperature for 6 h. After the reaction was complete, the solution was cooled to 0 ℃ in a salt ice bath, sodium azide (8.0 g, 123 mmol) was added. The suspension was stirred for 1 h at 0 ℃. HCl gas was bubbled through the reaction mixture at 0 to 5 ℃ while stirring for 1 h, then the flask was sealed and stirred at room temperature for 12 h. The HCl overpressure was released carefully, and the solution was alkalized with sodium hydroxide solution to pH=7—8. The reaction mixture was refluxed for 0.5 h and cooled to room temperature, the precipitate was filtered, crude solid could be purified by recrystallization from hot water, yield 4.5 g (15.6 mmol, 92%).

DTA-TG (5 ℃/min): 383 ℃ (dec.). IR (KBr) v: 3487 (s), 3305 (s), 2193 (w), 1676 (m), 1424 (s), 1359 (s), 1239 (s), 1178 (s), 1068 (m), 1007 (m), 997 (m), 756 (m) cm^{-1}; EA (found, calcd for $C_2H_8N_8Na_2O_6$, M_r =286.11): C (8.32, 8.40), H (2.79, 2.82), N (39.04, 39.16).

Diammonium 5,5'-bistetrazole-1,1'-diolate (**1**): Sodium 5,5'-bistetrazole-1,1'-diolate tetrahydrate (2.8 g, 10 mmol) was dissolved in 100 mL of warm water, ammonia chloride (1.6 g, 30 mmol) was added to the above solution while stirring. The mixture was maintain for 1 h, the solution was cooled to room temperature, the colorless crystalline residue was filtered and air dried, which could be purified by recrystallization in water, yield 1.8 g (8.8 mmol, 88.2%).

DTA-TG (5 ℃/min): 287 ℃ (dec.). IR (KBr) *v*: 3187 (s), 3049 (s), 2153 (w), 1818 (w), 1668 (m), 1433 (s), 1353 (s), 1310 (w), 1230 (s), 1043 (m), 997 (m), 726(m) cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ: 7.12; ^{13}C NMR (300 MHz, DMSO-d_6) δ: 134.1; EA (found, calcd for $C_2H_8N_{10}O_2$, M_r=204.15): C (11.59, 11.77), H (3.82, 3.95), N (68.55, 68.61).

Results and Discussion

Synthesis

Glyoxal (**2**) was converted to glyoxime (**3**) following the literature procedure,[16] while **1** was prepared by our modified synthetic routes from Fischer *et al.*[17] in two different methods (Scheme 2). Notably, in comparison with Fischer's step-by-step synthetic route, much higher yield of **6** was fulfilled in one pot from glyoxime (90% in method B), solving the headache of isolation of high friction and impact sensitive intermediates dichloroglyoxime and diazidoglyoxime. Additionally, we utilized new chlorinated reagent to replace Cl_2, not only greatly increasing the chlorination efficiency but also simplifying the synthetic progress.

Crystal structure

The crystal of compound **1** was grown in methanol solution by slow evaporation at room temperature. A colorless crystal of **1**, irregularly shaped, of approximate dimensions 0.22 mm×0.21 mm×0.20 mm, was used for crystal and intensity data collection. The crystal of **1** was measured on a Bruker-AXS smart APEX2 CCD detector using a Mo-Kα radiation wavelength of λ= 0.71073 Å. The structures were measured at 296 K. Bond distances, bond angles, torsion angles, hydrogen bond, and atomic positional parameters for non-hydrogen atoms are shown in Table S2—S6. The molecular structure and packing in the crystal lattice was illustrated in Figures 1 and 2, respectively. Additionally, the normalized XRD patterns of **1** measured and simulated by single crystal data were also shown in Figure S2, where the main sharp peak at 13.46 can be attributed to **1** crystallite based on the Debye-Scherrer equation.

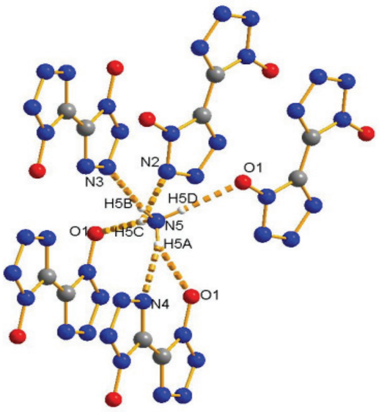
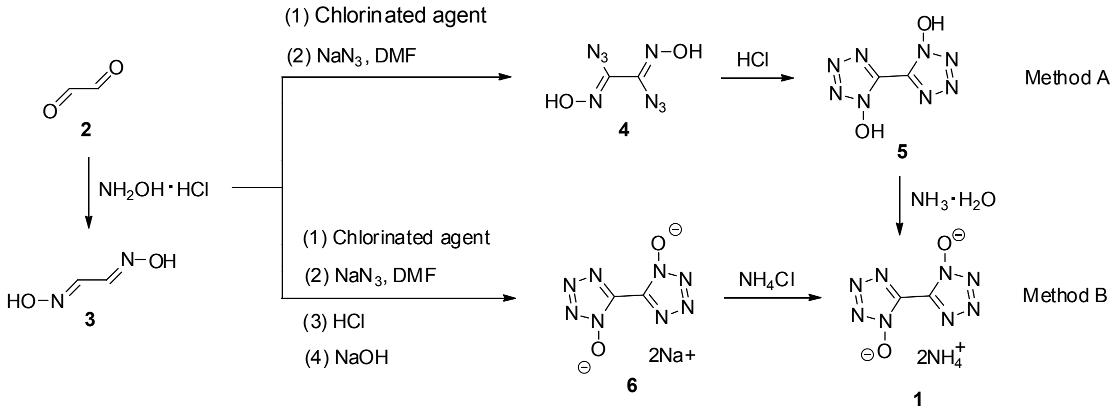


Figure 1 Coordination of the NH_4^+ cations in **1**. Thermal ellipsoids are drawn at the 50% probability level and hydrogen atoms are shown as small spheres or arbitrary radii. Symmetry operators: (i) −1+*x*, *y*, *z*; (ii) −1+*x*, 0.5−*y*, 0.5+*z*; (iii) 1–*x*, −0.5+*y*, −0.5–*z*; (iv) −1+*x*, 0.5–*y*, −0.5+*z*; (v) *x*, 0.5–*y*, −0.5+*z*. Grey C; blue N; red O; white H.

Interestingly, **1** crystallizes as monoclinic space group *P*21/*c* with 2 molecules in the unit cell (Figure 2), and has density of 1.765 g/cm^3 (at 296 K) (Table 1), which is different from the reported orthorhombic face-centered space group *I*222.[15] Comparing with

Scheme 2 The two synthetic methods for preparation of **1**



Klapötke's result, crystal **1** in our work has five bistetrazoles coordinating with one ammonium ion, while one bistetrazole coordinating with two ammonium ion was presented in literature. The different crystalline form is possibly attributed to the polarity of recrystallization solvent. In addition, the C-N and N-N bonds of the tetrazole ring are in the range of formal C-N and N-N single and double bonds (C-N: 1.47 Å, 1.22 Å; N-N: 1.48 Å, 1.20 Å).[27] The C(1)-N(1)-N(2) angle of N-oxide has a value of 108.41(97)° as compared to 111.47(57)° for neutral compound **5**. As expected, the bond length of N(1)-O(1) is shortened to 1.31 Å upon deprotonation [1.34 Å in **5**]. The tetrazole ring [torsion angle N(1)-N(2)-N(3)-N(4) = 0.12(14)°] is planar, which is a condition of the 6π aromaticity, and oxygen groups [O(1)-N(1)-N(2)-N(3)=79.48(10)°] is also almost planar out of this ring.

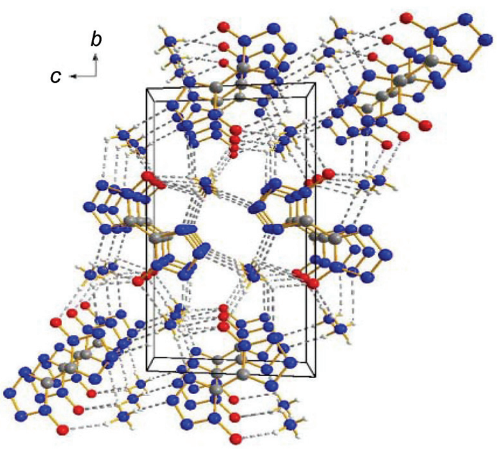


Figure 2 The perspective view of the unit cell of **1** along the *a*-axis. Grey C; blue N; red O; white H.

Due to the planarity of both cation and anion, the crystal structure of **1** is built up by planes that are kept together by a strong network of hydrogen bonding, in contrast with the highly disordered crystalline in Klapötke's result. As shown in Figure 2, each BTO^{2-} anion is surrounded by five ammonium cations via strong hydrogen bonds towards the N atoms of the tetrazole ring. It is remarkable to note that all accessible nitrogen atoms and the *N*-oxide O(1) act as acceptors for hydrogen bonds, which is merely possible due to the several N—H groups of the ammonium cations. Meanwhile, the strong π-π interactions were also found between the BTO rings, which could facilitate the molecular stability.

Characterization of thermal behavior

The thermal behavior of **1** was studied by thermogravimetric differential analysis (DTA-TG) in Al_2O_3 containers with a hole in the lip for gas release and a nitrogen flow of 20 mL/min at a heating rate of 5 ℃/min. As shown in Figure 3, decomposition of **1** was started at 220 ℃ and reached a peak at 287.8 ℃, indicating that **1** is thermally stable at least up to 220 ℃.

Table 1 Crystal date and experimental details of **1**

Formula	$C_2H_8N_{10}O_2$
Color	Colorless
Crystal system	Monoclinic
Space group	*P*21/*c*
a/Å	3.6786(9)
b/Å	14.506 (3)
c/Å	7.3456(16)
α/(°)	90
β/(°)	101.373[5]
γ/(°)	90
Volume/Å^3	384.29(15)
Density/(g•cm^{-3})	1.765
Z	2
μ/mm^{-1}	0.150
θ range/(°)	2.81—25.12
F(000)	212
Index range	$-4<h<3$, $-16<k<17$, $-8<l<8$
R(int)	0.0155
Refl. collected	2404
Refl. unique	687
R_1, wR_2 [$I>2\sigma(I)$]	0.0278, 0.0816
R_1, wR_2 (all data)	0.0299, 0.0845
GOOF	1.068
CCDC	1057866

Additionally, a significant mass loss of 91.2% was found in the range over 220—300 ℃. Both studies suggested that **1** had a high thermal stability, which most likely could be attributed to the extensive hydrogen bonding observed in the crystal structures.

Sensitivities

By applying the China National Military Standard, the impact and friction sensitivities of **1** were determined.[18] The impact sensitivities of **1** were determined with a fall hammer apparatus. The test was performed after drying the sample for 6 h at 60 ℃. During the test, the room temperature was 23 ℃ and relative humidity was 65%. A sample of **1** (30 mg) was placed between two steel poles and hit with a 5.0 kg drop hammer from a starting height of 25 cm. The probability detonating of **1** was 0—8%, whereas that of HMX at class 1, was 100%. The determination of the 50% firing height (h_{50}) was performed with a 5.0 kg drop hammer. The results for **1**, TNT, RDX, HMX and Comp. B are 89.4, 157, 22.3±0.3, 26.1±0.1 and 48.7±0.1.[19-21] From these data, **1** appears to be less sensitive to impact than RDX, HMX and Comp. B. In the case of friction sensitivity test, **1** was measured with the MGY-1 pendular friction sensitivities apparatus according to the standard procedure using 20 mg of sample. After **1** was compressed between two steel poles with mirror surfaces at a pressure of 3.92 MPa, it was then hit by 5 kg hammer at a

90° angle relative to the horizontal, the resulting firing rate was obtained to be 0%. Therefore, **1** was proved to be as insensitive as TNT to friction. The test results are summarized in Table 2, which is as expected because the extensive hydrogen bonding in the molecule benefits the stabilization of material.[22]

Table 2 Hydrogen bonds for **1** (Å and deg)

D−H···A	*d*(D−H)	*d*(H···A)	*d*(D−A)	∠DAH
N5-H5C-N2	0.85(1)	2.61(1)	3.11(2)	117.42
N5-H5B-N3	0.85(1)	2.21(1)	3.06(2)	172.41
N5-H5A-N4	0.85(1)	2.19(1)	3.01(2)	161.97
N5-H5C-O1	0.85(1)	2.09(1)	2.91(1)	178.41
O1-H5D-N5	0.85(1)	2.52(1)	3.01(2)	117.47

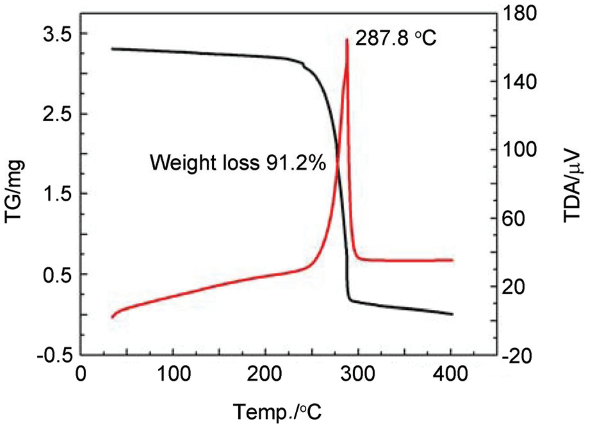


Figure 3 DTA-TG curves of **1**.

By comparing the density of **1** with that of other important explosives in Table 2, the density value measured from the crystal of **1** at 296 K is 1.765 g/cm^3, which is higher than that of TNT and Comp. B, but slightly lower than that of RDX. The overall results indicated that **1** could be used as good candidate of new insensitive high explosives. Meanwhile, it also could get a conclusion that different crystal form of **1** resulted in different explosive properties, especially on the aspect of impact sensitivities and detonation velocity (Table 3).

Conclusions

The diammonium 5,5'-bistetrazole-1,1'-diolate (**1**) was synthesized by two novel methods using inexpensive and readily available starting materials. The new crystal form of compound **1** was found as monoclinic space group *P*21/*c* with 2 molecules in the unit cell from water solution. The intensive intermolecular hydrogen bonding and the planar structure of bistetrazole ring resulted in the high thermal stability of the overall molecule, which was further proved by DTA-TG analysis. **1** shows the high thermal stability of 287.8 ℃ and outstanding insensitivity towards friction (H_{50}=89.1 cm) and impact (P=0%). In conclusion, **1** could become an excellent candidate as insensitive explosive in various warheads and ammunitions in the future.

Table 3 Explosive properties of **1** compared with some other important explosives

Explosive	Impact		Friction	Density/	D^e/	T_{dec}/	Ref.
	P^b/%	50%^{c}/cm	P^d/%	(g•cm^{-3})	(m•s^{-1})	℃	
1	4±4	89.4	0	1.765 (296K)	8762	287.8	
1$^{Ref.}$	8±4	82.5	0	1.779 (293K)	8817	290	[15]
TNT	0	157±0.3	0	1.684 (298K)	6950	285	[23]
RDX	80±8	23.3±0.3	76	1.806 (298K)	8754	210	[24]
HMX	100	26.1±0.3	100	1.890 (298K)	9100	280	[25]
Comp. B^a	36±4	48.7±0.1	32	1.720	—	—	[26]

^{a} An explosive composition with 39.5% of TNT, 59.5% of RDX, and 1% of wax; ^{b} Explosion probability; ^{c} Firing height; ^{d} Explosion probability; ^{e} Detonation velocity.

Acknowledgement

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