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Some High Nitrogen Derivatives of Nitrotetrazolyl-imidazole as New High Performance Energetic Compounds

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Abstract: This work introduces important properties of some new derivatives of nitrotetrazolyl-imidazole as high nitrogen energetic compounds, which are evaluated and discussed using some reliable models. The predicted properties are also compared with 1,3,5,7-tetranitro-1,3,5,7-tetraazacyclooctane (HMX). It is shown that some of these compounds can be seen as interesting organic explosives with relatively high performance and low sensitivity, which could be used for important industrial applications. Since some of the new compounds have a relatively good oxygen balance, the calculated specific impulses confirm that these compounds can be considered as suitable oxidizers in solid propellants.

Keywords: nitrotetrazolyl-imidazole, high energy material, performance, safety, thermochemical property

1 Introduction

The prediction of physicochemical properties of energetic materials is of great importance prior to their actual synthesis. An ideal energetic material must

have high performance and thermal stability but low sensitivity to external stimuli. Detonation velocity, detonation pressure and heat of detonation are three important parameters for the assessment of the performance of high explosives. In addition the specific impulse can be used as an important factor for determining the performance of propellants. In recent years, the synthesis of nitrogen rich compounds has been of significance because these compounds usually have a high heats of formation, density and oxygen balance. They are also considered suitable ingredients in low smoke propellant charges [1-4]. It has been shown that the existence of high levels of nitrogen compounds in the combustion products can reduce gun barrel erosion, because of the formation of iron nitride instead of iron carbide on the inner surface of the barrel [5, 6]. There are several reports concerning the development efforts in the synthesis of new energetic compounds [7-12].

Polynitroimidazole derivatives are some of the most attractive high nitrogen energetic materials. It is found that they are insensitive high performance explosives. There are several studies describing investigations of the properties of imidazole derivatives [13-25]. Among several derivatives, 2,4-dinitroimidazole (DNIMD) is more interesting due to its insensitivity and relatively high performance. Its impact sensitivity is greater than 100 cm, whilst its performance is within about 0.8X that of 1,3,5,7-tetranitro-1,3,5,7-tetraazacyclooctane (HMX). However, it has a higher performance than 1,3,5-triamino-2,4,6-trinitrobenzene (TATB) [19]. In contrast to DNIMD, 1,4-dinitroimidazole is thermodynamically unstable, and can be easily converted to DNIMD by heating [20]. Jadhav *et al*. synthesized several derivatives of 2,4,5-trinitroimidazole (TNIMD) [17]. They also calculated the performances of these derivatives and compared them with other insensitive explosives. Li *et al*. calculated the detonation properties of several imidazole derivatives through quantum mechanical methods [22]. Su *et al*. studied some properties of nitroimidazole, polynitroimidazole and their methyl derivatives [23]. They believed that 1-methyl-2,4,5-trinitroimidazole is a good candidate for use as a melt castable explosive due to its low melting point, *i.e*. 82 ºC [23]. Hou *et al*. have also synthesized several N-substituted derivatives of dinitroimidazole [24].

The purpose of the present work is to introduce some derivatives of imidazole as high performance, high nitrogen energetic compounds. Figure 1 shows the molecular structures, names and abbreviations of these new derivatives. Reliable methods have been used to study various significant aspects of these compounds. Crystal density, condensed phase heat of formation, detonation temperature, impact sensitivity, electric spark sensitivity, deflagration temperature, power and heat of detonation, as well as detonation pressure and velocity are important

properties for explosive users, and will be investigated here. The predicted results will be discussed and compared with those of HMX.

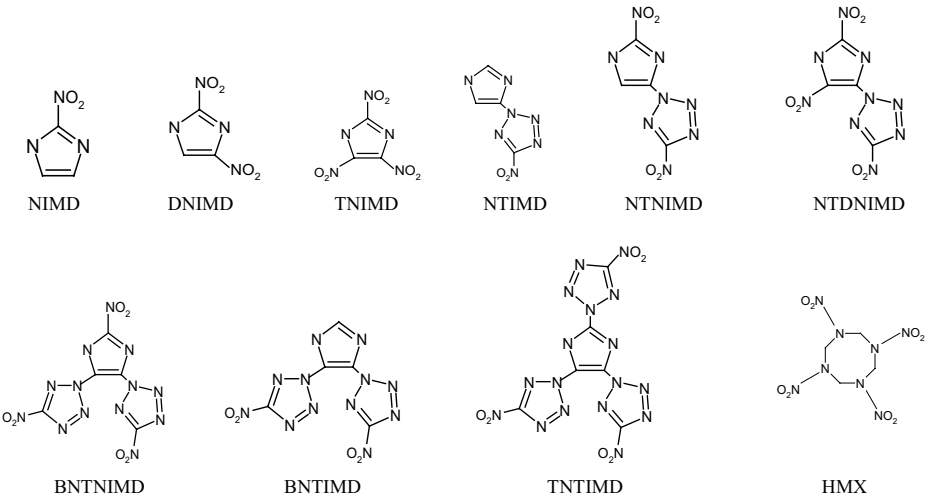

Figure 1. Molecular structures of NIMD: 2-nitroimidazole; DNIMD: 2,4-dinitroimidazole; TNIMD: 2,4,5-trinitroimidazole; NTIMD: 4-(5-nitrotetrazolyl)-imidazole; NTNIMD: 4-(5-nitrotetrazolyl)-2-nitroimidazole; NTDNIMD: 4-(5-nitrotetrazolyl)-2,5-dinitroimidazole; BNTNIMD: 4,5-bis(5-nitrotetrazolyl)-2-nitroimidazole; BNTIMD: 4,5-bis(5-nitrotetrazolyl)-imidazole; TNTIMD: 2,4,5-tris(5-nitrotetrazolyl)-imidazole; HMX: 1,3,5,7-tetranitro-1,3,5,7-tetraazacyclooctane.

2 Results and Discussion

2.1 Crystal density

Since detonation performance of energetic materials has a great dependence on their crystal densities, the prediction of crystal density is of utmost importance for chemists who synthesize energetic materials. To predict the densities of new derivatives of imidazole, a suitable model for evaluating the crystal density of nitroaromatic energetic compounds with the general formula $C_aH_bN_cO_d$ has been used [26], and is given in Equation 1:

$$\rho_0 = 10.57\left(\frac{a}{Mw}\right) + 0.1266\left(\frac{b}{Mw}\right) + 30.38\left(\frac{c}{Mw}\right) + 35.18\left(\frac{d}{Mw}\right) \quad (1)$$

where ρ_0 is the core crystal density in g/cm^3, which can be revised for the presence of some functional groups or molecular fragments, and *Mw* is the molecular weight of the energetic compound. The predicted results are given in Table 1. As seen in Table 1, replacement of hydrogen atoms in NIMD by nitro and nitrotetrazolyl groups can increase the density. Thus, the presence of these molecular moieties may enhance molecular packing.

It was indicated that the densities of polynitro heteroarenes are somewhat higher than the corresponding carbocyclic nitroaromatics [1]. The densities of all of the compounds in Table 1 are ordered in the sequence TNIMD ~ NTDNIMD ~ BNTNIMD ~ TNTIMD > BNTIMD > HMX ~ NTNIMD ~ DNIMD > NTIMD > NIMD. Replacement of hydrogen atoms by nitrotetrazolyl groups or nitro groups does not seem to have a significant effect in increasing the density.

2.2 Condensed phase heat of formation

The estimation of the condensed phase heat of formation or enthalpy of formation of a new energetic material, with a specified molecular structure, is important because it is required for the evaluation of the detonation pressures and velocities using various computer codes or empirical methods [27, 28]. A reliable model [29] is used here for the evaluation of the condensed phase heat of formation of energetic materials. This provides more reliable results compared to complex quantum mechanical methods [30]. It can be expressed as:

$$\begin{aligned}\Delta_f H^{\theta}(kcal\ mol^{-1}) &= 7.829a - 8.117b + 16.52c - 27.80d + 29.82n_{NO_2} \\ &- 15.56n_{Ar-NH} - 22.38n_{OH} - 48.34n_{COOH} + 3.241(n_{Ar} - 1) + 29.02n_{-N=N-} \\ &+ 53.34n_{cyclo\ N-O-N}\end{aligned} \quad (2)$$

The parameters in Equation (2) are related to the number of some specific functional groups or molecular fragments in the molecule of the energetic material [29]. As seen in Table 1, the introduction of nitrotetrazolyl groups into imidazole derivatives is more effective than nitro groups in increasing the calculated heat of formation. The condensed phase heat of formation is raised by increasing the number of nitrotetrazolyl substituents in the imidazole derivatives, *i.e.* TNTIMD > BNTIMD > NTIMD.

2.3 Impact sensitivity

The reliable estimation of sensitivity parameters of energetic materials is of great

Table 1. Summary of various properties of the imidazole derivatives

Properties	NIMD	DNIMD*	TNIMD	NTIMD	NTNIMD	NTDNIMD	BNTNIMD	BNTIMD	TNTIMD	HMX*
Crystal density [g·cm^{-3}]	1.70	1.86 1.77 [a]	1.949	1.791	1.884	1.947	1.946	1.904	1.945	1.89 [b]
Heat of formation in solid phase [kJ/mol]	216.106	167.468	140.394	825.193	669.911	566.199	821.18	1041.979	1116.265	74.9 [c]
Impact sensitivity [cm]	1150	137 > 100 [d]	32	213	53	21	16	25	14	29 [c]
Impact sensitivity [J]	287.5	34.25 > 25 [d]	8	53.25	13.25	5.25	4	6.25	3.5	7.25 [c]
Electric spark sensitivity [J]	17.59	9.88	8.27	16.86	9.14	7.54	6.81	6.12	6.07	2.65
Heat of detonation [kJ/g] [for H_2O (l)]	3.486	4.086	4.794	4.052	4.507	5.069	5.518	5.004	6.056	6.197 [c]
Detonation temperature [K]	3679.1	4538.4	4930.4	4030.3	4967.2	5436.9	5850.2	5565.5	6216.9	4406
Deflagration temperature [K]	503.42	491	478.6	516.5	504.1	491.7	504.7	517.2	517.8	560 [c]
Velocity of detonation [km/s]	7.79	8.58	9.11	8.47	9.02	9.46	9.80	9.44	9.62	9.10 [c]
Detonation pressure [GPa]	26.72	34.51	39.9	31.15	36.57	40.94	42.03	38.68	43.12	39.0 [b]
Power relative to TNT as a standard 100	113.1	123.1	133.1	119.1	129.1	139.1	145.1	135.1	151	150 [e]
Isp [N·s/g]	2.34	2.45	2.55	2.51	2.61	2.72	2.89	2.78	3.06	2.61

* Experimental values of listed properties for HMX and DNIMD, when available, were taken from [a][47], [b][42], [c][48], [d][21] and [e][49].

importance in the synthesis and use of novel energetic molecules because safe handling of these materials is of utmost importance. Some stimuli, such as impact, friction, heat and electric spark, can cause detonation. It is believed that hot spots in the energetic materials contribute to initiation by external stimuli. It was found that the nanosizing of explosives leads to a reduction in their sensitivity to external stimuli, because of the reduction in the formation of hot spots [31]. Impact stimuli are closely related to many accidents which have occurred during the production, handling and storage of energetic materials. Experimentally, impact sensitivity is measured by subjecting a sample to the impact of a standard weight falling from various heights and recording evidence of reaction or no reaction. Thus, a height of 50% probably in causing an explosion (h_{50}) is measured by impacting the sample with a falling hammer, typically of 2.5 kg weight. For the prediction of the impact sensitivities of imidazole derivatives, a suitable correlation has been used, which was introduced to estimate the impact sensitivities of nitro-heterocyclic aromatic compounds of general formula $C_aH_bN_cO_d$ [32] as:

$$\log h_{50} = \frac{46.29a + 35.63b - 7.701c + 7.942d + 44.42n_{-CNC-} + 102.3n_{-CNNC-}}{Mw} \quad (3)$$

where $\log h_{50}$ is the impact sensitivity in cm, and n_{-CNC-} and n_{-CNNC-} are the numbers of –CNC– and –CNNC– moieties in the molecule. As can be seen in Table 1, the calculated value of h_{50} for NIMD is higher than that for the other compounds. However, the replacement of hydrogens by nitro groups or nitrotetrazolyl groups has an appreciable effect in increasing the sensitivity and decreasing safety. The values of h_{50} for imidazole derivatives are raised by increasing the number of these substituents, *i.e.* NIMD > DNIMD > TNIMD and BNTIMD > TNTIMD.

2.4 Electric spark sensitivity

The electric spark or electrostatic sensitivity of an energetic compound can be defined as the degree of sensitivity to electrostatic discharge, which can be determined by subjecting the explosive to a high-voltage discharge from a capacitor. In order to study the electrostatic sensitivity, a reliable correlation has been used for predicting the electrostatic sensitivity of nitroaromatic energetic compounds [33], which can be given as:

$$E_{ES}(J) = 4.60 - 0.733a + 0.724d + 9.16r_{b/d} - 5.14C_{R,OR} \quad (4)$$

where $r_{b/d}$ is the ratio of hydrogen to oxygen atoms and $C_{R,OR}$ is the presence of certain groups such as alkyl (R) or alkoxy (OR) groups attached to an aromatic ring. As seen in Table 1, the electric spark sensitivity of HMX is greater than that

of all the other compounds. It seems that the effect of introducing either nitro or nitrotetrazolyl groups is similar in decreasing the electric spark sensitivity of the imidazole derivatives, *i.e*. NIMD ~ NTIMD and BNTNIMD ~ TNTIMD.

2.5 Heat of detonation

The evaluation of the performance of an explosive depends on the amount of energy which is released during detonation and the rate of its release. A new approach has recently been introduced to estimate the heat of detonation of aromatic energetic compounds [34], which is expressed as:

$$Q(kJ/g) = 2.129 + 0.178c + 0.874r_{d/a} + 0.160r_{b/d} + 0.965C_{SFG} \quad (5)$$

where $r_{d/a}$ and $r_{b/d}$ the ratios of oxygen to carbon and hydrogen to oxygen atoms, respectively, and $C_{SFG} = -1$ for aromatic energetic compounds that have some specific functional groups [34]. The predicted results of the imidazole derivatives are given in Table 1. As seen in Table 1, the heat of detonation of TNTIMD is comparable to that of HMX. However, the substitution of a hydrogen atom by a nitrotetrazolyl group is more effective in increasing the heat of detonation than a nitro group, *i.e*. BNTIMD > DNIMD and TNTIMD > TNIMD.

2.6 Detonation temperature

The detonation temperature is measured experimentally from the brightness of the detonation front as it proceeds towards the detector. However, its measurement is difficult. A reliable model has been recently introduced for predicting the detonation temperature [35], which gives good predictions compared to one of the best available equations of state, *i.e*. BKWC-EOS [36], as:

$$T(K) = 5136 - 190.1a - 56.4b + 115.9c + 148.4d - 466.0r_{d/a} - 700.8r_{b/d} - 282.9n_{NH_x} \quad (6)$$

where n_{NH_x} is the number of NH_2 and NH_4^+ groups in the energetic compound; $r_{d/a}$ and $r_{b/d}$ are as defined above. The predicted detonation temperatures of the new compounds are given in Table 1. As shown in Table 1, the detonation temperature is raised by increasing the number of nitro or nitrotetrazolyl groups, *i.e*. TNTIMD > BNTIMD > NTIMD and TNIMD > DNIMD > NIMD. Also the introduction of nitrotetrazolyl groups to the imidazole derivatives is more effective in increasing the calculated detonation temperature than nitro groups, *i.e*. TNTIMD > TNIMD and BNTIMD > DNIMD. Among the energetic compounds given in Table 1, TNTIMD has the highest detonation temperature.

2.7 Detonation pressure and velocity

The performance of an explosive is mainly defined in terms of the velocity of detonation and the detonation pressure. Of course, both the detonation pressure and the velocity of detonation of an explosive are strong functions of its density. Thus, these properties can be enhanced by increasing the value of the loading density [37, 38]. Empirical methods, or computer codes with various equations of state, can be used to predict the detonation pressure and velocity [27, 28]. Two reliable methods were used to predict the detonation pressure and velocity of the new proposed energetic compounds, the results of which are given in Table 1 [39, 40]. The model used to predict the detonation velocity is given by Equation (7):

$$D(km/s) = 1.644 + 3.593\rho_0 - 0.132a - 0.0034b + 0.120c + 0.0442d - 0.2768n_{NRR'} \quad (7)$$

where $n_{NRR'}$ is the number of specific groups in the explosive, including NH_2, NH_4^+ and  groups. For the prediction of the detonation pressure, the following correlation was used:

$$P(kbar) = 22.32 + 104.0\rho_0^2 - 10.98a - 1.997b + 5.562c + 5.539d - 23.68n_{NH_x} - 154.1n_1^0 \quad (8)$$

where n_1^0 has the value of one for an energetic compound that satisfies the condition $d > 3(a + b)$, and for other cases has a value of zero. Since the number of moles of gaseous detonation products per unit weight of an energetic material is an important factor in enhancing its detonation performance, the resulting products, based on the Kistiakowsky-Wilson rules [41], are shown in Table 2. Higher values of the crystal density in an explosive can also provide higher values of the detonation velocity and pressure. As can be seen in Table 1, the detonation performance of BNTNIMD has the highest values relative to the other imidazole derivatives and HMX.

Table 2. Detonation products of the newly proposed imidazole derivatives

Energetic material	Molecular formula	Detonation products
NIMD	$C_3H_3N_3O_2$	2 CO + $C_{(S)}$ + 1.5 H_2 + 1.5 N_2
DNIMD	$C_3H_2N_4O_4$	3 CO + H_2O + 2 N_2
TNIMD	$C_3HN_5O_6$	2.5 CO_2 + 0.5 CO + 0.5 H_2O + 2.5 N_2
NTIMD	$C_4H_3N_7O_2$	2 CO + 2 $C_{(S)}$ + 1.5 H_2 + 3.5 N_2
NTNIMD	$C_4H_2N_8O_4$	4 CO + H_2 + 4 N_2
NTDNIMD	$C_4HN_9O_6$	1.5 CO_2 + 2.5 CO + 0.5 H_2O + 4.5 N_2
BNTNIMD	$C_5HN_{13}O_6$	0.5 CO_2 + 4.5 CO + 0.5 H_2O + 6.5 N_2
BNTIMD	$C_5HN_{12}O_4$	4 CO + 0.5 $C_{(S)}$ + 0.5 H_2 + 6 N_2
TNTIMD	$C_6HN_{17}O_6$	6 CO + 0.5 H_2 + 8.5 N_2
HMX	$C_4H_8N_8O_8$	4 CO + 4 H_2O + 4 N_2

2.8 Deflagration temperature

The deflagration point is defined as the temperature at which a small sample of the explosive, placed in a test tube and externally heated, bursts into flame, decomposes rapidly or detonates violently [42]. It can be determined by heating 0.02 g of sample in a glass tube in a Wood's metal bath at a heating rate of 5 °C min^{-1}. However a new method has been recently developed to predict the deflagration temperature of new proposed energetic compounds [43], which is shown as:

$$DT(K) = 476.6 + 13.08a - 6.21d + 103.7F^{+}_{non-add} - 103.7F^{-}_{non-add} \quad (9)$$

where the functions $F^{+}_{non-add}$ and $F^{-}_{non-add}$, show increasing and decreasing contributions of non-additive structural parameters, respectively [43]. The predicted deflagration temperatures of the new imidazole derivatives are also given in Table 1. As shown in Table 1, the deflagration temperature of all of the compounds in Table 1 are ordered in the sequence HMX > TNTIMD ~ BNTIMD ~ NTIMD > BNTNIMD ~ NTNIMD ~ NIMD > NTDNIMD ~ DNIMD > TNIMD. The presence of a nitrotetrazolyl group has a greater effect in increasing the deflagration temperature than a nitro group.

2.9 Power

The power or strength of an explosive is a measure of its ability to do useful work. This is also termed the potential of an explosive and is the total quantity of heat given off by an explosive at constant volume [37]. The strength of a high explosive can be correlated with its detonation performance through the heat

of detonation and volume of gaseous detonation products [44]. The ballistic mortar provides a relative measure of the strength or power of an explosive, usually by comparison with TNT as the standard explosive [42]. In the ballistic mortar test, a heavy steel mortar is attached to a pendulum. About 10 g of the explosive charge is initiated in the mortar cavity, which is enclosed by a steel projectile. The mortar is swung from its initial position when the projectile is ejected out of the mortar. The maximum swing of the mortar is recorded to determine the power of the tested explosive. In order to evaluate the strength of the new proposed imidazole derivatives, a reliable simple method was used [45], which is given as:

$$EP(\%TNT) = 113.0 - 5.16a + 2.786c + 3.612d - 46.18OCF \quad (10)$$

where *OCF* is the correcting function [45].

The predicted results are given in Table 1. As seen, TNTIMD has the highest strength amongst the energetic compounds which are introduced in Table 1.

2.10 Specific impulse of imidazole derivatives as monopropellants

The specific impulse is used to compare performances of rocket propellants [41]. It is defined as the thrust per unit weight flow rate of propellant. Its value is of utmost significance for the determination of the propellant mass necessary to meet the ballistic requirements [37]. It is also related to flame temperature, the number of moles of gas produced per unit mass of propellant and the average molecular mass of the gaseous products of combustion [37]. However, the specific impulse of an energetic compound depends on its structural parameters. In order to predict the specific impulse of the imidazole derivatives, a simple method can be used that is based on the structural parameters [46]. This method is expressed by Equation (11) [46]:

$$I_{SP} = 2.421 - 0.0740a - 0.0036b + 0.0237c + 0.400d - 0.1001n_{NH_2,NH} + 0.1466(n_{Ar} - 1) \quad (11)$$

where I_{sp} is the specific impulse in $N \cdot s \cdot g^{-1}$, $n_{NH_2,NH}$ is the number of NH_2 and NH groups, and n_{Ar} is the number of aromatic rings in aromatic explosives. The calculated specific impulses of the proposed new imidazole derivatives are given in Table 1, and are ordered in the sequence TNTIMD > BNTNIMD > BNTIMD > NTDNIMD > HMX = NTNIMD > TNIMD > NTIMD > DNIMD > NIMD.

3 Conclusions

The properties of several new nitrotetrazolyl-imidazole derivatives as high nitrogen explosives were evaluated and compared with HMX. It was shown that some of them have relatively high performance, low sensitivity and good thermochemical properties. The results of this paper can be of help in designing or developing ideal energetic compounds. Among the different derivatives, TNIMD and NTDNIMD are suitable as oxidizers in solid propellants, because they contain the excess oxygen necessary for converting some of the carbon atoms to carbon dioxide. Three derivatives, NTDNIMD, BNTNIMD and TNTIMD, have relatively high crystal densities, strength, detonation velocities and pressure with respect to the other derivatives, so that these compounds are good candidates as high performance explosives. Moreover, the sensitivities of NIMD, DNIMD, NTIMD and NTNIMD are relatively low compared to the other imidazole derivatives.

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