



## A New Method for Predicting the Friction Sensitivity of Nitramines

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**Abstract:** This study presents a new simple correlation between friction sensitivity of nitramines and their molecular structures. This novel correlation can help chemists and chemical engineers to predict the friction sensitivity of new nitramines without using any experimental data, which is important for safety in industrial processes. The new correlation can also help to elucidate the mechanism of initiation of energetic materials by frictional stimuli. This new method assumes that friction sensitivity of a nitramine of general formula  $C_aH_bN_cO_d$  can be expressed as a function of the optimized elemental composition and the contributions of specific molecular structural parameters. The new correlation has root mean square and average deviations of 7.64 and 6.44 J, respectively, for 20 nitramines with different molecular structures. The proposed new method was also tested for 11 nitramines containing complex molecular structures.

**Keywords:** friction sensitivity, nitramine, correlation, safety

### 1 Introduction

Organic explosives are substances containing metastable molecules capable of undergoing very rapid and highly exothermic reactions [1]. A major goal

within the energetic materials community has been to predict various aspects of a notional energetic compound including sensitivity, performance, physical and thermodynamic properties. The study of explosives by various empirical methods has provided a considerable insight into the understanding of factors affecting their behaviour [2]. However, a significant effort has been made world-wide to understand better the relationship between the molecular structures of energetic compounds and their sensitivities [3].

Nitramines are one of the most important classes of energetic materials having high energy content and high crystal densities [4]. The evaluation of friction sensitivity of these compounds is very important from a safety point of view. It is essential to avoid unintended initiation or explosion of an energetic compound during its handling, storage and use. Therefore, we should know or be able to predict the value of its sensitivity parameters. Most reports in recent years have focused on the shock and impact sensitivities of these compounds. However, a few studies have been reported concerning the friction sensitivity of energetic materials. Experience has shown that the experimental values of the friction sensitivity of energetic compounds can be heavily influenced by human variability [5].

Some relationships have been demonstrated between the sensitivity parameters of energetic compounds and their related variables, *e.g.* molecular structure [6-8], detonation velocity [9-13], detonation pressure [12-15], molecular electronic properties [16, 17], and particle size [18], as well as their thermal decomposition parameters [19-22]. Jungova *et al.* [23] investigated the relation between the friction sensitivity of nitramines and their thermal decomposition parameters. Friedl *et al.* [24] studied the relation between friction sensitivity of nitramines and their surface electronic potentials. Jungova *et al.* [5] also compared the friction sensitivity of nitramines with their impact sensitivities and heats of fusion.

The purpose of the present work is to introduce a new correlation between the friction sensitivity of nitramines and their molecular structures. The new correlation confirms that the sensitivity parameters of energetic compounds are primarily due to their chemical characteristics [5]. It will be shown that elemental composition and several structural parameters can be used to derive a novel correlation. Moreover, the new correlation can help chemists to design new, safe, energetic materials containing N-NO<sub>2</sub> groups.

## 2 Results and Discussion

### 2.1 Materials and methods

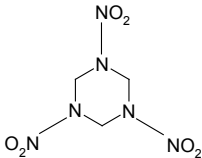
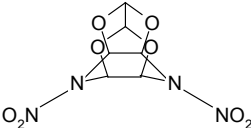
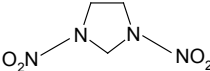

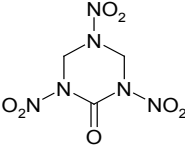
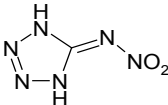
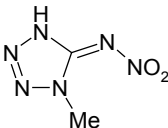
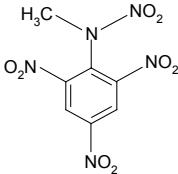
Table 1 contains experimental data for the friction sensitivity of nitramine compounds, which were collected from various references. These data were obtained from BAM friction or Julius Peters friction apparatus. The explosive sample is held between a porcelain plate and a porcelain peg under a given load. Frictional forces are applied by the horizontal movement of the porcelain plate. The relative sensitivity to friction is indicated by the lowest load, expressed in Newtons, that leads to ignition, crackling or explosion [25]. The study of various nitramines has shown that the friction sensitivity of nitramines can be related to their molecular structure as:

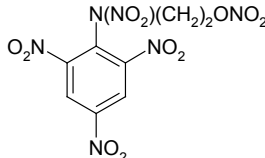
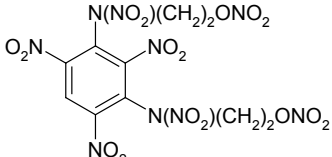
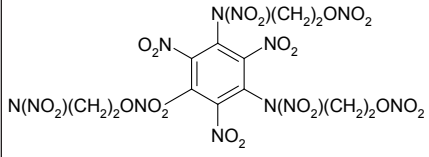
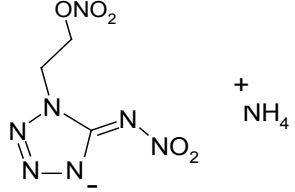
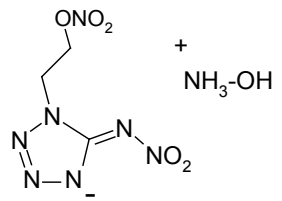
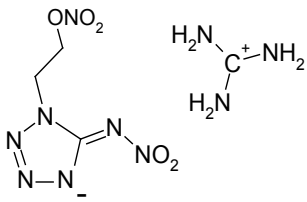
$$FS(N) = 600.8 - 2428.6 (n_H/Mw) - 6481.4 (n_N/Mw) - 9560.9(n_O/Mw) + 54.5 P_{FS}^+ - 77.8 P_{FS}^- \quad (1)$$

where  $FS$  is the friction sensitivity in N,  $n_H$ ,  $n_N$  and  $n_O$  are the number of hydrogen, nitrogen, and oxygen atoms in the molecular formula of the nitramine, respectively, and  $Mw$  is the molecular weight of the nitramine. It was found that the number of hydrogen, nitrogen, and oxygen atoms in the molecular formula of the explosive gives suitable variables for the elemental composition in finding a good correlation. The contribution of the number of carbon atoms is small enough to be neglected. A conventional variable has also been defined that was divided into  $P_{FS}^+$  and  $P_{FS}^-$  on the basis of the molecular structure of the nitramines. The adjustable coefficients are found from the experimental data given in Table 1.

**Table 1.** Comparison of the predicted friction sensitivity with experimental data

Name	Compound	FS (exp.) <sup>a</sup> [N]	FS (calc.) [N]	Dev.
2,4,6-Trinitro-2,4,6-triazaheptane (ORDX)	$\begin{array}{ccccccc} \text{H}_3\text{C} & - & \text{N} & - & \text{CH}_2 & - & \text{N} & - & \text{CH}_2 & - & \text{N} & - & \text{CH}_3 \\ & &   & & & &   & & & &   & & \\ & & \text{NO}_2 & & & & \text{NO}_2 & & & & \text{NO}_2 & & \end{array}$	147.7	148.83	-1.13
2,4-Dinitro-2,4-diazapentane (OCPX)	$\begin{array}{ccccccc} \text{H}_3\text{C} & - & \text{N} & - & \text{CH}_2 & - & \text{N} & - & \text{CH}_3 \\ & &   & & & &   & & \\ & & \text{NO}_2 & & & & \text{NO}_2 & & \end{array}$	74.9	91.06	-16.16
2,5-Dinitro-2,5-diazahexane (DMEDNA)	$\begin{array}{ccccccc} \text{H}_3\text{C} & - & \text{N} & - & \text{CH}_2 & - & \text{CH}_2 & - & \text{N} & - & \text{CH}_3 \\ & &   & & & & & &   & & \\ & & \text{NO}_2 & & & & & & \text{NO}_2 & & \end{array}$	57.9	64.96	-7.06

Name	Compound	FS (exp.) <sup>a</sup> [N]	FS (calc.) [N]	Dev.
1,3,5-Trinitro-1,3,5-triazacyclohexane (RDX)		148.5	156.09	-7.59
4,10-Dinitro-2,6,8,12-tetraoxa-4,10-diazaisowurtzitane (TEX)		161.3	154.29	7.00
1,3-Dinitro-1,3-diazacyclopentane (CPX)		57.7	56.40	1.30
1,4-Dinitro-1,4-diazacyclohexane (DNDC)		122.3	125.81	-3.51
1,3,5-Trinitro-2-oxo-1,3,5-triazacyclohexane (keto-RDX)		108 <sup>b</sup>	111.27	-3.27
5-Nitriminotetrazole		8 <sup>c</sup>	19.95	-11.95
1-Methyl-5-nitriminotetrazole		160 <sup>c</sup>	157.74	2.26
1-Methylnitramino-2,4,6-trinitrobenzene (TETRYL)		353 <sup>d</sup>	342.57	10.43

Name	Compound	FS (exp.) <sup>a</sup> [N]	FS (calc.) [N]	Dev.
1-(2-Nitratoethylnitramino)-2,4,6-trinitrobenzene		324 <sup>e</sup>	326.10	-2.10
1,3-Bis(2-nitratoethylnitramino)-2,4,6-trinitrobenzene		192 <sup>e</sup>	199.01	-7.01
1,3,5-Tris(2-nitratoethylnitramino)-2,4,6-trinitrobenzene		126 <sup>e</sup>	134.59	-8.59
Ammonium 1-(2-nitratoethyl)-5-nitriminotetrazolate		120 <sup>f</sup>	123.45	-3.45
Hydroxylammonium 1-(2-nitratoethyl)-5-nitriminotetrazolate		60 <sup>f</sup>	51.40	8.60
Guanidinium 1-(2-nitratoethyl)-5-nitriminotetrazolate		120 <sup>f</sup>	108.34	11.66

Name	Compound	FS (exp.) <sup>a</sup> [N]	FS (calc.) [N]	Dev.
Aminoguanidinium 1-(2-nitratoethyl)- 5-nitriminotetrazolate		108 <sup>f</sup>	103.14	4.86
Diaminoguanidinium 1-(2-nitratoethyl)- 5-nitriminotetrazolate		108 <sup>f</sup>	98.45	9.55
1,1'-dinitro-3,3'-azo- 1,2,4-triazole		80 <sup>g</sup>	78.69	1.31
rms deviation, [J]				7.64
Average absolute deviation, [J]				6.44

<sup>a</sup> All experimental data is reported from Jungova *et al.* [5], except for those specified;

<sup>b</sup> Ref. [31]; <sup>c</sup> Ref. [32]; <sup>d</sup> Ref. [25]; <sup>e</sup> Ref. [33]; <sup>f</sup> Ref. [34]; <sup>g</sup> Ref. [35].

## 2.2 The values of $P_{FS}^+$ and $P_{FS}^-$

- Acyclic nitramines: for acyclic nitramines with more than two repetitive  $[-CH_2 N(NO_2)-]$  units, the value of  $P_{FS}^+$  equals 1.0, whilst the value of  $P_{FS}^-$  is 0.5 for the presence of the molecular fragment  $-N(NO_2)-CH_2-CH_2-N(NO_2)-$ . For those acyclic nitramines containing nitramine groups of the form  $N(NO_2)-C=$ , the value of  $P_{FS}^+$  depends on the number of separate molecular  $N(NO_2)-C=$  ( $n_{N(NO_2)-C=}$ ) moieties, *i.e.*  $P_{FS}^+ = 5 - 2 n_{N(NO_2)-C=}$ , except for  $n_{N(NO_2)-C=} \geq 3$  when  $P_{FS}^+ = 0$ .
- Cyclic nitramines: for cyclic nitramines in which equal numbers of nitramine and methylene groups exist, the value of  $P_{FS}^+$  equals 1.0. For cyclic nitramines containing five membered rings, the values of  $P_{FS}^-$  are 0.75 and 1.25 for the presence of two and one  $-N(NO_2)-$  groups per ring, respectively.
- The attachment of a  $-N(NO_2)-$  group to a triazole ring: for the 5-nitriminotetrazole salts, the values of  $P_{FS}^+$  and  $P_{FS}^-$  equal 0.5 in the

presence of ammonium and hydroxylammonium cations, respectively. For 5-nitriminotetrazole and its methyl derivatives, the values of  $P_{FS}^-$  and  $P_{FS}^+$  are 1.25 and 0.5, respectively.

Experimental data from different sources for friction sensitivity, which are given in Table 1, were used to derive Equation (1) through a multiple linear regression method [26]. Although the presence of various factors affect the experimental data of friction sensitivity, especially human variability [5], fortunately the R-squared value or the coefficient of determination of Equation (1) was 0.989.

### 2.3 Comparison of the predicted results with experimental data

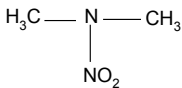
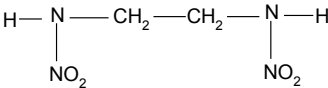
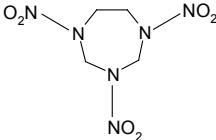
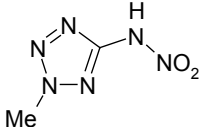
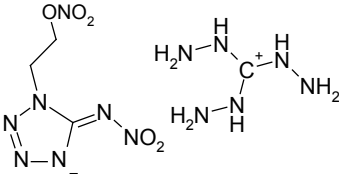
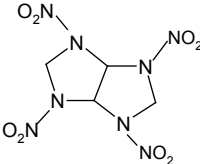
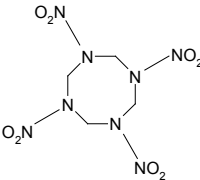
As seen in Table 1, the predicted friction sensitivities of nitramines have a root mean square (rms) and an average absolute deviation of 7.64 and 6.44 J, respectively. Moreover, the deviation of the estimated friction sensitivity exceeds 12.0 J for only five nitramines. Table 2 gives the statistical parameters of Equation (1), which allows the relative weights of the variables in the model to be compared. The standard error gives a measure of the precision of the estimated coefficient. This can determine the precision after repeated measurements. The P value can assess the significance of an observed effect or variation. A P value of less than 0.05 may confirm that the observed effect is not due to random variations and that the effect is significant. Thus, suitable statistical parameters and a relatively good  $R^2$  value show that the predicted results of new method are in good agreement with experimental values.

The application of the new method has been checked for 11 nitramines containing complex molecular structures, as shown in Table 3. The predicted results have also been compared with the experimental data. As seen in Table 3, the new method gives relatively good results, which confirms the reliability of the new method.

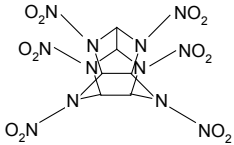
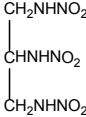
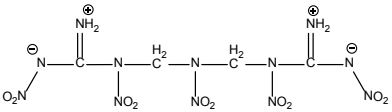
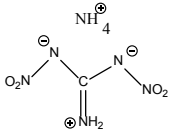
**Table 2.** Standardized coefficients and some statistical parameters of Equation (1)

Variable	Coefficient	Standard error	P-value	Lower limit (95%)	Upper limit (95%)
Intercept	600.8	43.56	0.0000	510.51	691.16
$n_H/MW$	-2428.6	189.81	0.0000	-2822.26	-2034.99
$n_N/MW$	-6481.4	614.98	0.0000	-7756.85	-5206.03
$n_O/MW$	-9560.9	930.88	0.0000	-11491.4	-7630.38
$P_{FS}^+$	54.5	2.66	0.0000	49.00	60.02
$P_{FS}^-$	-77.8	5.04	0.0000	-88.21	-67.30

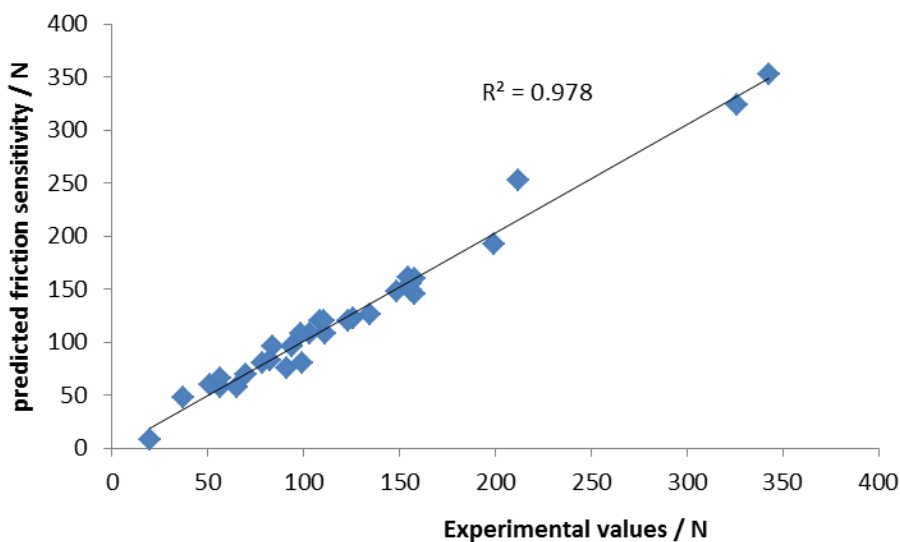
**Table 3.** Comparison of the predicted friction sensitivity of some nitramines containing complex molecular structures.

Name	Compound	FS (exp.) [N]	FS (calc.) [N]	Dev.
2-Nitro-2-azapropane (DMNA)		82.4	82.40	0.00
1,4-Dinitro-1,4-diazabutane (EDNA)		47.4	36.96	10.44
1,3,5-Trinitro-1,3,5-triazacycloheptane (HOMO)		119.96	110.62	9.34
2-Methyl-5-nitraminetetrazole		145 <sup>c</sup>	157.74	-12.74
Triaminoguanidinium 1-(2-nitratoethyl)-5-nitriminetetrazolate		96 <sup>f</sup>	94.19	1.81
<i>cis</i> -1,3,4,6-tetranitrooctahydroimidazo[4,5-d]imidazol (BCHMX)		66.12	56.36	9.76
1,3,5,7-Tetranitro-1,3,5,7-tetraazacyclooctane (HMX)		154.4	156.09	-1.69



Name	Compound	FS (exp.) [N]	FS (calc.) [N]	Dev.
2,4,6,8,10,12-Hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane (HNIW)		69	69.67	-0.67
1,3-Dinitramino-2-nitratopropane		96 <sup>h</sup>	84	12
1,7-Diamino-1,7-dinitrimino-2,4,6-trinitro-2,4,6-triazaheptane (APX)		80 <sup>i</sup>	98.9	18.9
Ammonium dinitroguanidine (ADNQ)		252 <sup>i</sup>	211.9	40.1

<sup>h</sup> Ref. [36]; <sup>i</sup> Ref. [37].



**Figure 1.** The predicted friction sensitivities of nitramines versus their experimental values given in Tables 1 and 3.

Figure 1 shows the relation between the predicted and experimental friction

sensitivity values of the nitramine compounds which were introduced in Tables 1 and 3.

Equation (1) can also be programmed for designing new nitramine energetic compounds on the basis of the  $P_{FS}^+$  and  $P_{FS}^-$  correction factors [27, 28]. Although quantum mechanical methods have also been used to study the thermal decomposition and electrostatic sensitivity of some energetic compounds [29, 30], there is no need to use high speed computers and special computer codes in the present method.

### 3 Conclusions

It was shown that there is a simple reliable correlation between friction sensitivity of nitramines and their molecular structures, which may be interesting for chemists and the chemical industry. The new correlation assumes that the friction sensitivity of a nitramine with the general formula  $C_aH_bN_cO_d$  can be expressed as a function of the optimized elemental composition and the contribution of correction factors, *i.e.*  $P_{FS}^+$  and  $P_{FS}^-$ . The present method can help to elucidate the mechanism of initiation of energetic materials by frictional stimuli because it confirms that the initiation reactivity of energetic compounds is intimately related to their chemical character and molecular structures. Furthermore, this new relationship can be used to design new cyclic and acyclic nitramines.

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