



A Novel Correlation for Predicting the Density of Tetrazole–*N*-oxide Salts as Green Energetic Materials through Their Molecular Structure

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Abstract: In this study, a new and reliable model is derived for predicting the density of tetrazole *N*-oxide salts using molecular structure descriptors through multiple linear regression methods (*MLR*). The proposed model is based on the elemental, functional group and type of molecule descriptors. The coefficient of determination (R^2) of the model was 0.9321 for 36 energetic tetrazole *N*-oxides with different molecular structures as an exploratory set. The predictive ability of this model has been checked using a cross validation method ($Q^2_{\text{LOO}} = 0.9325$ and $Q^2_{\text{LMO}} = 0.9324$). The new correlation had a root mean square deviation (*RMSD*) of $0.033 \text{ g}\cdot\text{cm}^{-3}$ and an average absolute deviation (*AAD*) of $0.025 \text{ g}\cdot\text{cm}^{-3}$. This correlation also gave good predicted results for a further 10 energetic tetrazole *N*-oxides as a test set. The new reliable model can also be applied for designing novel energetic tetrazole *N*-oxides.

Keywords: tetrazole-*N*-oxide energetic compounds, density, correlation, multiple linear regression method, molecular structure.

1 Introduction

Nitrogen-rich compounds are interesting energetic compounds for chemists and chemical engineers, especially for those who research energetic compounds. Because of the presence of the N-N and C-N bonds in these compounds, they can form a large number of hydrogen bonds and then show special properties such as high positive heats of formation, low sensitivities, good thermal stabilities,

and relatively high densities [1-4]. Since the major gas released on combustion of nitrogen-rich compounds is nitrogen, which is a non-toxic and inherently cooler gas, they are considered as initial candidates for “green” energetic materials [5].

Compared to non-ionic molecules, nitrogen-rich energetic salts have some advantages, for example:

- (i) they have a very low vapour pressure, which essentially reduces the risk of exposure of personnel to toxic vapour;
- (ii) due to the existence of coulombic forces for forming ordered and dense lattice structures in the salts, they often have a higher density than non-ionic nitrogen rich compounds [6-10];
- (iii) designing new salts with optimized characteristics is possible only through the appropriate combination of different cations and anions [2, 11].

The organic nitrogen-containing heterocycle fragments, such as pyrazole-, imidazole-, triazole-, tetrazole- and pentazole-moieties, can be either cationic or anionic in nitrogen rich energetic salts [12-20]. Among these energetic ions, the tetrazolium (positively charged tetrazole) or tetrazolide (negatively charged tetrazole) rings (see Figure 1) furnish an important area of chemistry because of the practical and theoretical significance of these unique compounds and because they tend to have a reduced environmental impact [21].

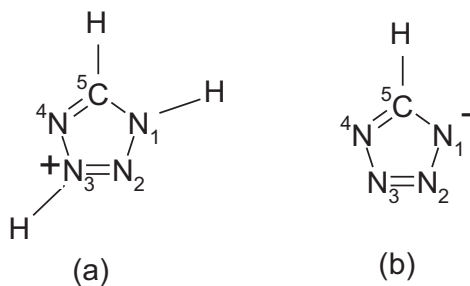


Figure 1 Molecular formulae of (a) tetrazolium cation, and (b) tetrazolide anion

Recently, several research groups have reported that tetrazole-containing compounds are improved through the formation of the tetrazolate-*N*-oxide anion. This can be generally made possible by the oxidation of the N1 (Figure 2a) or N2 (Figure 2b) atoms and formation of an N→O group. The insertion of oxygen increases the oxygen balance of tetrazole *N*-oxide salts. Also, tetrazole-*N*-oxides have a higher density and higher performance in comparison with the corresponding non-oxidized tetrazole derivatives. In addition, research has shown that the energetic tetrazole *N*-oxide salts have lower friction, impact and shock sensitivities than the non-oxidized tetrazole derivatives [22].

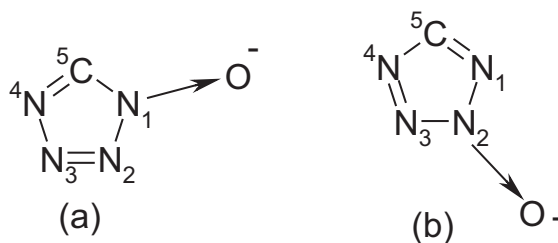


Figure 2. Molecular formulae of the two type of tetrazole N-oxide

Because of the existence of a correlation between the detonation velocity and pressure of an explosive and its density, the prediction of density of a new energetic compound can help chemists to design novel ideal energetic materials. As shown in the empirical Kamlet-Jacobs equations [23], Equations 1 to 3, the increasing density of an energetic material can produce increases in its performance [24].

$$P = 1.558\rho^2\phi \quad (1)$$

$$D = 1.01\phi^{1/2}(1 + 1.3\rho_0) \quad (2)$$

$$\phi = 0.4889N(\bar{M}Q)^{1/2} \quad (3)$$

where P is the predicted detonation pressure in GPa, D is the detonation velocity in $\text{km}\cdot\text{s}^{-1}$ and ρ is the density of a compound in $\text{g}\cdot\text{cm}^{-3}$, N is the number of moles of gaseous detonation products *per* gram of explosive, \bar{M} is the mean molecular mass of the detonation products, and Q is the detonation energy in $\text{cal}\cdot\text{g}^{-1}$.

There are several methods for predicting the density of a compound, and quantum mechanical methods and group additivity methods are two important procedures [25-28]. On the basis of Ammon's method [26], the crystal density of an atom and functional group volumes and densities were determined for organic compounds with the elements H, C, B, N, O, F, P, S, Cl, Br, I, and halide anions. Although group additivity is a relatively simple and rapid method, it does not consider the effects of isomerization, conformation and crystal polymorphism. Politzer *et al.* used the electrostatic potential on the cationic and anionic surfaces for predicting the density of energetic ionic compounds [30]. In the method proposed by Rice *et al.* for predicting the density of energetic compounds, the molecular volume or formula unit volumes have been used. In this method, the volumes within the 0.001 electron·Bohr⁻³ isosurface of electron density for the constituent isolated neutral and ionic molecules are calculated to define the molecular volume or formula unit volumes used

in predicting the crystal density [31]. Quantum mechanical methods are often complex, time consuming procedures and need advanced computing systems. A quantitative structure-property relationship (QSPR) is another method used to evaluate the physicochemical properties of an energetic compound. The molecular descriptors of the compounds are correlated with their various properties, such as density. The multiple linear regression (MLR) technique is the method used for estimating the coefficients involved in the model equation. This technique works on a least-squares procedure [32-34].

Recently several simple correlations have also been proposed for estimating the density of various classes of energetic compounds, such as nitroaliphatics, nitrate esters and nitramines [35-46]. The effect of some functional groups, such as $-\text{NO}_2$, $-\text{CN}$, $-\text{N}_3$, $-\text{NF}_2$ and $-\text{NH}_2$, on the density of tetrazolium nitrate salts has been investigated through the multiple linear regression method [47]. Furthermore, in previous work we developed a model for predicting the density of azole-based energetic compounds through QSPR [48].

The purpose of the present work was to develop a correlation for predicting the density of tetrazole N-oxide salts through the multiple linear regression method. In this method the various molecular descriptors that may have an effect on the density of these compounds are investigated. The new reliable correlation proposed in this work can help chemists design novel ideal tetrazole-N-oxide salts as green energetic materials.

2 Model development

2.1 Derived new correlation

For investigating the effects of various descriptors on the density of tetrazolium nitrate salts, the relationship between density and their structural parameters was studied through multiple linear regression methods. For this purpose, various structural parameters were selected and tested, such as elemental composition, specific functional groups, types of substitutions on the tetrazole ring, as well as their positions.

The fourth column of the Table 1 contains experimental density values for the 36 tetrazole-N-oxide salts used as the exploratory set, which were obtained from Ref. [49] and were used in deriving Equation 4. This study has shown that it is possible to express the density of these compounds as a function of several structural parameters. Multiple linear regression methods were used to obtain the relationship between density and molecular structure in Equation 4 as [50]:

$$\rho = 1.514 + \omega - 0.039 n_{H_2O} + 0.075 TET + 0.171 \rho^+ - 0.118 \rho^- \quad (4)$$

$$\omega = -0.047 n_C - 0.025 n_H + 0.028 n_N + 0.073 n_O \quad (5)$$

where ρ is the crystal density of the tetrazole *N*-oxide salts in $\text{g}\cdot\text{cm}^{-3}$, n_{H_2O} is the number of the H_2O molecules in the crystal of the salt. The *TET* parameter for tetrazole 1-*N*-oxide salts is 1.0 and for those salts which contain a 2-*N*-oxide fragment it is zero. ω is the optimum elemental composition in Equation 5, the n_C , n_H , n_N and n_O are the numbers of carbon, hydrogen, nitrogen and oxygen atoms in the salt, respectively. ρ^+ and ρ^- are the positive and negative non-additive structural parameters, which are defined as follows:

(a) Definition of ρ^+ :

If there is a 2-hydroxytetrazole fragment in a tetrazole *N*-oxide energetic molecule, the value of ρ^+ is 1.0.

b) Definition of ρ^- :

b1. For ammonium tetrazolate-1-*N*-oxide salts, $\rho^- = 1.0$.

b2. If the cation of the tetrazole *N*-oxide salt is amino-nitroguanidinium the value of ρ^- is 1.0.

2.2 Variables of the model

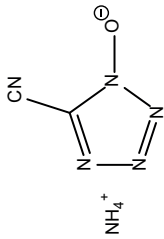
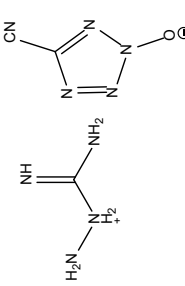
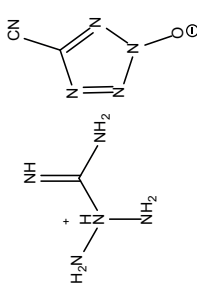
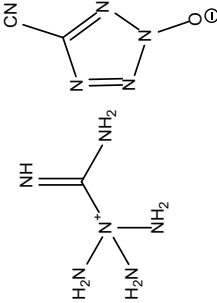
The study showed that of the various structural descriptors that were tested for in deriving the best model, the number of carbon, hydrogen, nitrogen and oxygen atoms, the number of H_2O molecules in the crystal salts and the tetrazole *N*-oxide type (*TET*) participate most in the correlation. For estimating the coefficients of the variables of Equation (4), the least-squares procedure was used. The least-squares method minimises the sum of the squares of the deviations of the predicted values from the experimental data.

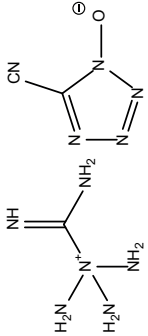
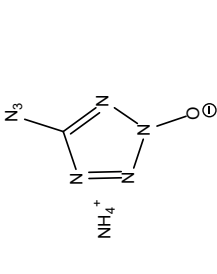
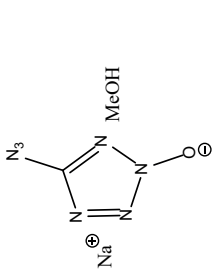
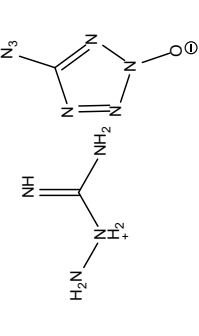
As expected, the sign of n_N and n_O is positive, which confirms that increasing the number of nitrogen and oxygen atoms can increase the density of the tetrazole *N*-oxide salts. Based on previous reports, we knew that the introduction of certain substituents, such as NF_2 , NO_2 and N_3 , in the molecular structure of nitrogen rich compounds, can enhance their density due to the probable formation a strong π bond between the tetrazole ring and the substituent [47].

Based on the definition of the *TET* parameter in Equation 4, tetrazole-1*N*-oxide salts have a higher density compared with the tetrazole-2*N*-oxide salts. So in designing a new tetrazole-*N*-oxide salt, this observation ought to be considered. The negative sign of the n_{H_2O} term in Equation 4, confirms that the presence of water in the crystal structure of a salt can decrease its density.

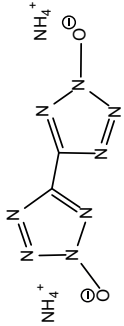
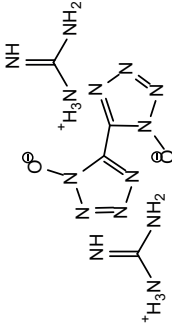
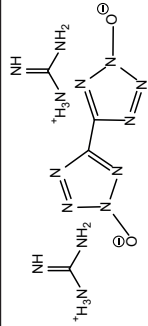
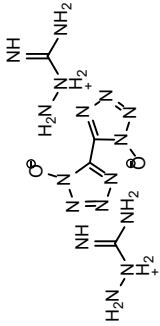
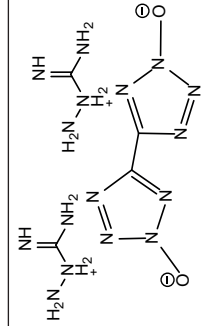
Table 1. Comparison of the predicted densities with experimental data as an exploratory set

No.	Name	Structure	$\rho^{(exp)}$ [g·cm ⁻³] [49]	$\rho^{(cal)}$ [g·cm ⁻³]	Dev.	Ref. [44]	Dev	Ref. [37]	Dev.
1	Hydroxylammonium 5-nitrotetrazole-2 <i>N</i> -oxide		1.84	1.827	0.013	1.87	-0.03	2.04	-0.20
2	5-Nitro-2-hydroxytetrazole		1.94	1.972	-0.032	2.15	-0.21	2.05	-0.11
3	Triaminoguanidinium 5-nitrotetrazolate-2 <i>N</i> -oxide		1.639	1.722	-0.083	1.82	-0.19	1.96	-0.32
4	Ammonium 5-cyanotetrazolate-2 <i>N</i> -oxide		1.554	1.561	-0.007	1.90	-0.34	1.85	-0.30

No.	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	$\rho(\text{cal})$ [g·cm ⁻³]	Dev.	Ref. [44]	Dev	Ref. [37]	Dev.
5	Ammonium 5-cyanotetrazolate-1 <i>N</i> - oxide		1.526	1.518	0.008	1.90	-0.37	1.85	-0.33
6	Aminoguanidinium 5-cyanotetrazolate-2 <i>N</i> - oxide		1.504	1.523	-0.019	1.85	-0.34	1.83	-0.33
7	Diaminoguanidinium 5-cyanotetrazolate-2 <i>N</i> - oxide		1.534	1.526	0.008	1.84	-0.30	1.84	-0.31
8	Triaminoguanidinium 5-cyanotetrazolate-2 <i>N</i> - oxide		1.55	1.529	0.021	1.83	-0.28	1.85	-0.31

No.	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	$\rho(\text{cal})$ [g·cm ⁻³]	Dev.	Ref. [44]	Dev.	Ref. [37]	Dev.
9	Triaminoguanidinium 5-cyanotetrazolate-1 <i>N</i> - oxide		1.583	1.604	-0.021	1.83	-0.25	1.85	-0.28
10	Ammonium 5-azidotetrazolate-2 <i>N</i> -oxide		1.689	1.664	0.025	1.74	-0.05	2.006	-0.32
11	Sodium 5-azidotetrazolate- 2 <i>N</i> -oxide monohydrate		1.609	1.623	-0.014	1.63	-0.02	1.47	0.13
12	Aminoguanidinium 5-azidotetrazolate-2 <i>N</i> -oxide		1.608	1.626	-0.018	1.66	-0.05	1.94	-0.33

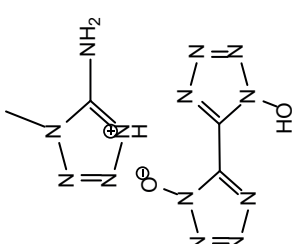
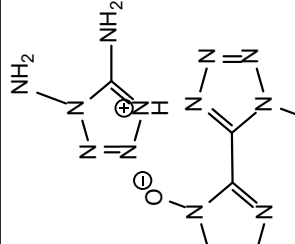
No.	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	Dev.	Ref. [44]	Dev.	Ref. [37]	Dev.
13	Hydroxyaminotetrazole		1.695	0.015	2.04	-0.34	1.95	-0.26
14	Hydroxylammonium 5-aminotetrazolate-1 <i>N</i> - oxide(orthorhombic)		1.664	-0.042	1.74	-0.07	1.96	-0.30
15	Hydroxylammonium 5-aminotetrazolate-1 <i>N</i> - oxide(monoclinic)		1.735	0.029	1.74	0.00	1.96	-0.23
16	Ammonium 5-aminotetrazolate-1 <i>N</i> - oxide		1.53	0.015	1.74	-0.21	1.92	-0.40
17	Bisammonium 5,5'-bis(tetrazolate-1 <i>N</i> - oxide)		1.8	0.079	1.84	-0.04	1.93	-0.14

No.	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	Dev.	Ref. [44]	Dev.	Ref. [37]	Dev.
18	Bisammonium 5,5'-bis(tetrazolate-2N-oxide)		1.664	0.018	1.84	-0.17	1.93	-0.27
19	Bisguanidinium 5,5'-bis(tetrazolate-1N-oxide)		1.639	0	1.81	-0.17	1.86	-0.22
20	Bisguanidinium 5,5'-bis(tetrazolate-2N-oxide)		1.633	0.069	1.81	-0.18	1.86	-0.23
21	Bisaminoguanidinium 5,5'-bis(tetrazolate-1N-oxide)		1.596	-0.049	1.80	-0.20	1.87	-0.28
22	Bisaminoguanidinium 5,5'-bis(tetrazolate-2N-oxide)		1.637	0.067	1.80	-0.16	1.87	-0.24

No.	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	$\rho(\text{cal})$ [g·cm ⁻³]	Dev.	Ref. [44]	Dev	Ref. [37]	Dev.
23	5,5'-bis(1-hydroxytetrazole) dihydrate		1.81	1.783	0.027	1.96	-0.15	2.72	-0.91
24	Ammonium 1,4-bis(2- <i>N</i> -oxidotetrazolate)-1,2,4,5-tetrazine monohydrate		1.627	1.616	0.011	1.74	-0.11	2.56	-0.94
25	5,5'-Bis(2-hydroxytetrazole)		1.953	1.911	0.042	2.20	-0.24	1.96	-0.02
26	Bishydroxylammonium 5,5'-bis(tetrazolate-1 <i>N</i> -oxide); TKX-50		1.918	1.867	0.051	1.83	0.09	1.97	-0.06

No.	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	$\rho(\text{cal})$ [g·cm ⁻³]	Dev.	Ref. [44]	Dev.	Ref. [37]	Dev.
27	Bishydroxylammonium 5,5'-(tetrazolate-2 <i>N</i> -oxide)		1.822	1.792	0.03	1.83	0.00	1.97	-0.15
28	Bis(dihydrazinium)5,5'- bis(tetrazolate-1 <i>N</i> -oxide)		1.725	1.727	-0.002	1.82	-0.10	1.94	-0.22
29	Diaminoguanidinium 1-hydroxy-1 <i>H</i> ,1 <i>H</i> - 5,5-bitetrazol-1-olate monohydrate		1.729	1.717	0.012	1.91	-0.18	1.91	-0.19
30	Triaminoguanidinium 1-hydroxy-1 <i>H</i> ,1 <i>H</i> - 5,5-bitetrazol-1-olate monohydrate		1.749	1.72	0.029	1.81	-0.06	1.92	-0.18

No.	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	$\rho(\text{cal})$ [g·cm ⁻³]	Dev.	Ref. [44]	Dev	Ref. [37]	Dev.
31	1-Amino-3-nitroguanidinium 1-hydroxy-1 <i>H</i> ,1 <i>H</i> -5,5-bitetrazol-1-olate dihydrate		1.778	1.779	-0.001	1.91	-0.13	2.35	-0.58
32	Diaminouronium 1 <i>H</i> ,1- <i>H</i> -5,5-bitetrazole-1,1-diolate monohydrate		1.8	1.787	0.013	1.81	-0.01	1.93	-0.13
33	Bis(oxalyl)dihydrazidinium 1 <i>H</i> ,1 <i>H</i> -5,5-bitetrazole-1,1-diolate		1.847	1.843	0.004	1.81	0.04	3.31	-1.46
34	5-Aminotetrazolium 1-hydroxy-1 <i>H</i> ,1 <i>H</i> -5,5-bitetrazol-1-olate		1.839	1.833	0.006	2.16	-0.32	1.94	-0.11

No.	Name	Structure	$\rho^{(\text{exp})}$ [g·cm ⁻³] [49]	$\rho^{(\text{cal})}$ [g·cm ⁻³]	Dev.	Ref. [44]	Dev	Ref. [37]	Dev.
35	1-Methyl-5-aminotetrazolium 1-hydroxy-1H,1H-5,5-bitetrazol-1-olate		1.762	1.736	0.026	2.06	-0.30	1.88	-0.12
36	1,5-Diaminotetrazolium 1-hydroxy-1H,1H-5,5-bitetrazol-1-olate		1.828	1.836	-0.008	2.14	-0.31	1.95	-0.12
rms deviation [g·cm ⁻³]			-	-	0.033	-	-	-	-
average absolute deviation [g·cm ⁻³]			-	-	0.025	-	-	-	-

2.3 Reliability of the correlation and its validation

The R^2 value or the coefficient of determination of Equation 4 was 0.932. As seen in the fifth column of Table 1, the predicted density of tetrazole *N*-oxide salts has a root mean square (rms) value and average absolute deviation of 0.033 and 0.025 g·cm⁻³, respectively. Table 2 lists the statistical parameters of Equation 4, which allows a comparison of the relative weights of the variables in Equation 4. The standard errors give a measure of the precision of the estimation of a coefficient, and can determine the precision over repeated measurements. The P value can assess the significance of an observed effect or variation. For a P value less than 0.05, it 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, *i.e.* 0.932, confirm that the predicted results of these new methods are in good agreement with experimental values.

Table 2. Standardized coefficients and some statistical parameters of Equation 4

Variable	Coefficient	Standard Error	P -Value	Lower bound (95%)	Upperbound (95%)
<i>Intercept</i>	1.5140	0.02430	1.09E-30	1.4640	1.5639
n_C	-0.0466	0.0110	0.000223	-0.0691	-0.02416
n_H	-0.0248	0.0040	1.06E-06	-0.0329	-0.0167
n_N	0.02858	0.0048	2.46E-06	0.0187	0.0384
n_O	0.07338	0.0060	1.52E-12	0.0610	0.0856
	-0.03878	0.0112	0.001816	-0.0617	-0.0158
<i>TET</i>	0.07538	0.0138	8.52E-06	0.0471	0.1036
ρ^+	0.1710	0.0328	1.71E-05	0.1037	0.2382
ρ^-	-0.11850	0.0245	4.7E-05	-0.1688	-0.0683

Cross validation methods were used for checking the predictive ability of the developed models through the QSPR approach. The leave-one-out cross-validation method (Q^2_{LOO}), which is defined by Equation 6 below, was chosen to explain the internal validation [41, 42].

$$Q^2_{LOO} = 1 - \frac{\sum_{i=1}^n (y_i - \hat{y}_{ic})^2}{\sum_{i=1}^n (y_i - \bar{y}_i)^2} \quad (6)$$

where y_i , \bar{y}_i , and \hat{y}_i are the experimental density or ρ_{exp} value of an energetic azole-based energetic compound for the i -th entry of each equation's data set, the mean value of the ρ_{exp} of all compounds, and the estimated ρ_{exp} of the i -th compound

by a model which is obtained without using the present model, respectively. The term n refers to the number of compounds in the exploratory set.

In addition, the leave-many-out cross validation method (Q^2_{LMO}) was also used for checking the model validity of the model. Parts of the data, more than one, were repetitively excluded from the exploratory set and evaluated in the validation of the model. For small sample sets (less than 25 samples), the leave-one-out cross-validation method is usually used to assess the predictive ability of the model [44]. In the present work, the (leave-(20%)-out) cross validation method was performed to validate the new model and the average value of the cross validation coefficient was derived as Q^2_{LMO} .

Table 3 contains a further 10 tetrazole N-oxide salts with various structures, which have been used for external validation of the proposed correlation. As seen in Table 3, the new proposed method gives relatively good results and confirms its reliability.

As shown in Figure 3, the predicted densities of tetrazole N-oxide salts by Equation 4, which are listed in Tables 1 and 3, have a good linear relation to the experimental data.

The predicted densities of the energetic tetrazole N-oxide salts by the new proposed correlation, which are listed in Table 1 and Table 3, are compared with two of the previously reported models [37, 44].

The reliability and validation test results of this regression model are summarized in Table 4. As seen in this Table, the small differences between Q^2_{LMO} , Q^2_{EXT} and R^2 in Equation 4 show that the correlation obtained is a good model and has good predictive power [33, 34].

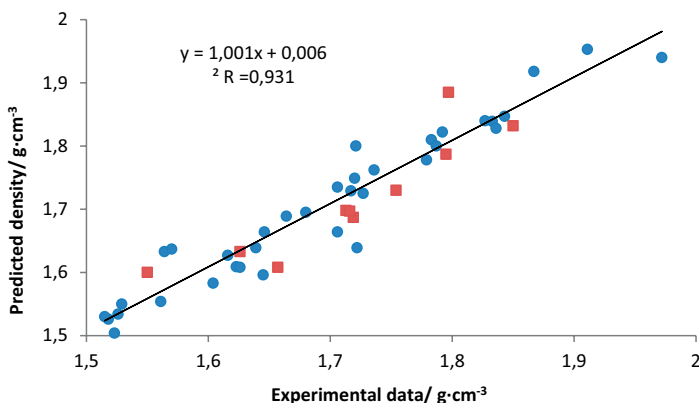
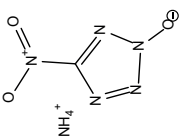
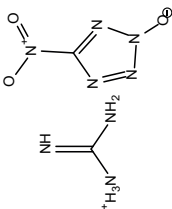
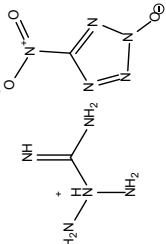
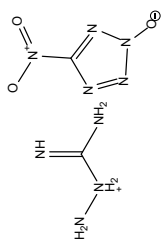


Figure 3. The predicted density of energetic tetrazole-*N*-oxide salts vs. experimental data, which are listed in Table 1 and Table 3. The circles and squares refer to exploratory and test set data, respectively

Table 3. Comparison of the predicted densities with computational data as a test set

No	Name	Structure	$\rho(\text{exp})$ [g·cm ⁻³] [49]	$\rho(\text{cal})$ /cm ⁻³	Dev.	Ref. [44]	Dev.	Ref. [37]	Dev.
1	Ammonium 5-nitrotetrazolate-2 <i>N</i> -oxide		1.73	1.754	-0.024	1.88	-0.15	2.02	-0.29
2	Guanidinium 5-nitrotetrazolate-2 <i>N</i> -oxide		1.698	1.713	-0.015	1.85	-0.15	1.94	-0.25
3	Diaminoguanidinium 5-nitrotetrazolate-2 <i>N</i> -oxide		1.687	1.719	-0.032	1.83	-0.14	1.96	-0.27
4	Aminoguanidinium 5-nitrotetrazolate-2 <i>N</i> -oxide		1.697	1.716	-0.019	1.84	-0.14	1.95	-0.25

No	Name	Structure	$\rho^{(exp)}$ [g·cm ⁻³] [49]	$\rho^{(cal)}$ /cm ⁻³	Dev.	Ref. [44]	Dev.	Ref. [37]	Dev.
5	Bis(5-amino-1- <i>H</i> -tetrazolium) 5,50-bis(tetrazole-2-oxide) dihydrate		1.633	1.744	-0.111	1.92	-0.28	2.32	-0.68
6	Oxalyldihydrazidinium 1 <i>H</i> ,1 <i>H</i> -5,5-bitetrazole-1,1- diolate dihydrate		1.885	1.797	0.088	1.74	0.15	2.34	-0.46
7	3,6-Bishydrazino-1,2,4,5- tetrazinium 1 <i>H</i> ,1 <i>H</i> -5,5- bitetrazole-1,1-diolate		1.787	1.795	-0.008	2.00	-0.21	1.92	-0.13

No	Name	Structure	$\rho^{(exp)}$ [g·cm ⁻³] [49]	$\rho^{(cal)}$ [cm ⁻³]	Dev.	Ref. [44]	Dev.	Ref. [37]	Dev.
8	1 <i>H</i> ,1 <i>H</i> -5,5-Bitetrazole-1,1-diol-2(5-amino-2-methyl-2 <i>H</i> -tetrazole)		1.608	1.657	-0.049	2.06	-0.45	3.11	-1.50
9	Bis(3-amino-1-nitroguanidinium) 5,5-bis(tetrazole-2-oxide)		1.832	1.85	-0.018	1.89	-0.06	1.96	-0.13
10	Bis(triaminoguanidinium) 5,5-bis(tetrazole-2-oxide) dihydrate		1.6	1.55	0.05	1.68	-0.08	2.24	-0.64
rms deviation [g·cm ⁻³]			-	-	0.039	-	-	-	-
average absolute deviation [g·cm ⁻³]			-	-	0.031	-	-	-	-

3 Conclusions

A new simple and reliable correlation has been developed for predicting the densities of tetrazole N-oxide salts. The effects of several molecular descriptors on the apparent crystal density of tetrazole N-oxide salts was investigated. It was found that optimum elemental composition, number of water molecules which exists in the crystal structure, and the type of tetrazole N-oxide, are the most effective descriptors on the densities of these compounds, as well as several non-additive structural parameters. It was confirmed that the density of tetrazole-1N-oxide salts is higher than that of salts whose oxidation occurred on nitrogen 2 (N-2) in the tetrazole ring. Furthermore, as expected, increasing the number of the nitrogen and oxygen atoms can increase the density of a salt. Because of the satisfactory statistical results ($R^2 = 0.9321$, $Q^2_{LMO} = 0.9324$ and $Q^2_{LOO} = 0.9325$), the new proposed model has good performance in predicting the density of new tetrazole N-oxide salts. The best correlation between density and molecular structure of tetrazole N-oxide salts was developed through the multiple linear regression (MLR) method. The novel proposed model can be used for designing novel energetic tetrazole N-oxide salts with the ideal performance parameters as green energetic materials.

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