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***Research paper***

# A Simple Approach for Predicting the Density of High Nitrogen Organic Compounds as Materials for Providing Clean Products and Enormous Energy Release

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**Abstract:** High nitrogen organic compounds (N>50 wt.%) are important for chemical industries because they can provide clean products with generally low-molecular weight product gases and enormous energy release. The density of these materials at or near room temperature is an important physical property for the assessment of their detonation and combustion performances. A novel method is introduced here for the prediction of the density of various classes of organic compounds, including different derivatives of triazole, tetrazole, triazine, tetrazine, furazan, and some organic nitrogen-containing chains. The core model is based on elemental composition, where its reliability has been improved by considering some molecular fragments including specific functional groups. The high reliability of these simple model has been compared with the output from two complex quantum mechanical approaches. For 91 high nitrogen compounds, the values of the standard deviation (*SD*) of the core and improved correlations were 0.076 and 0.047 g·cm$^{-3}$. For a further 32 materials, the values of *SD* were 0.057 and 0.042 g·cm$^{-3}$ for the core and improved correlations, respectively. These data are close to core and improved quantum mechanical methods, *i.e.* 0.056 and 0.042 g·cm$^{-3}$, respectively, where the calculated data from complex quantum mechanical approaches were available.

**Keywords:** density, high nitrogen organic compounds, molecular structure, clean product, enormous energy release

### List of Important Symbols

| | |
|---|---|
| $a$ | The number of carbon atoms |
| $b$ | The number of hydrogen atoms |
| $c$ | The number of nitrogen atoms |
| $d$ | The number of oxygen atoms |
| $Mw$ | Molecular weight of the desired high nitrogen compound |
| $Mw_{molecule}$ | Molecular mass of the molecule |
| $\rho$ | Density |
| $\rho_{core}$ | Core density |
| $\rho^{+}_{non\text{-}add}$ | Increasing factor for non-additive function of density |
| $\rho^{-}_{non\text{-}add}$ | Decreasing factor for non-additive function of density |
| $AD$ | Average deviation |
| $AD_{\max}$ | Absolute maximum deviation |
| $R^2$ | Coefficient of determination |
| $SD$ | Standard deviation of model |
| $sd$ | Standard deviation of each individual coefficient |

## 1 Introduction

Nitrogen-rich compounds liberate high energy because they usually have high positive heats of formation. Since the presence of N–N and N=N bonds in nitrogen-rich compounds, with the average energies for N–N bonds (160 kJ/mol) and N=N bonds (418 kJ/mol) being much lower than N≡N (954 kJ/mol), nitrogen-rich compounds can provide enormous energy release [1, 2]. For detonation and combustion processes, the main product of nitrogen-rich compounds is $N_2$ gas [3, 4]. Therefore, they can provide clean products with, generally, low-molecular weight gases [5-7]. Since nitrogen-rich compounds have low numbers of carbon and hydrogen atoms, they have higher densities and good oxygen balance, which are further advantages [8, 9].

The density of an energetic compound is a very important parameter for an assessment of its detonation velocity and pressure [10-13]. Moreover, it is desirable to use nitrogen-rich compounds with high densities as propellants in order to obtain an effective combustion process [14]. Common high detonation performance explosives, such as hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX), octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX), and 2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane (CL-20), contain nitrogen contents up to 38 wt%. High nitrogen organic compounds, defined as those with more than 50 wt% nitrogen in their molecular structures [15],

often contain derivatives of triazole, tetrazole, triazine, tetrazine, furazan, and organic nitrogen-containing chains up to 88 wt.% nitrogen in their structures. Most high nitrogen organic materials are derivatives of five or six membered cycles or nitrogen-containing chains, which have a variety of nitrogen-containing fragments such as $–NH_2$, $–NO_2$, $–N_3$, –C≡N, –N=N–, –C=N– and $–N=N^+O^-–$. Due to the importance of high nitrogen organic compounds, they are suitable candidates for insensitive high explosives [1], gun propellants [16-18], and clean gas generators in vehicle airbags [5, 19].

Different methods have been developed for predicting the density of different classes of organic compounds containing energetic functional groups. These include group additivity, quantum mechanics [20-25], quantitative structure-property relationship (QSPR) methods [26, 27], and molecular fragments including some specific functional groups [13]. Quantum mechanical calculations need expensive and intensive computations with specific software, as well as expert operators. Thus, they can only be used for relatively small molecules. In contrast to quantum mechanical methods, group additivity methods are simple to use but they cannot be used for the compounds containing unusual chemical structures and complex molecular structures, because these may give large deviations from experimental data. QSPR models usually require complex molecular descriptors and different computational tools, such as multiple linear regression (MLR), non-linear regression (NLR), partial least squares (PLS), artificial neural networks (ANN), genetic algorithms (GA), and support vector machines (SVM) [13, 28]. For some classes of organic compounds and ionic liquids containing energetic functional groups, it was shown that molecular fragments including some specific functional groups can also be used to predict their densities with good reliability [28-39]. These methods should be used only for specific classes of organic compounds, because they may give large deviations for other classes of organic materials.

The purpose of the present work was to introduce a new approach for predicting the density of high-nitrogen organic compounds, at or near room temperature, containing triazoles, tetrazoles, triazines, tetrazines, furazans, and some organic chains with more than 50 wt.% nitrogen. The core correlation was derived, based on experimental data that have been collected from different sources, by considering their elemental compositions. Its reliability is improved by two correcting factors. To test the reliability of the new correlations, the predicted results for further high nitrogen organic compounds is compared with two complex quantum mechanical methods, where the output for high nitrogen organic compounds was available [22]. It is shown that application of the new correlations for different complex high nitrogen molecules is straightforward

and very simple. Moreover, the new method will provide predictions as good as those reported by complex quantum mechanical approaches.

## 2 Development of the New Model

Previous studies have confirmed that elemental composition has an important role in predicting the densities of different classes of organic compounds containing energetic functional groups [28-35, 37, 38], as well as ionic liquids [36, 39]. Moreover, some molecular fragments have significant effects on the densities of organic molecules. The study of experimental values of the densities of high nitrogen compounds, at or near room temperature, showed that it is possible to correlate their densities with elemental composition, as well as specific functional groups and molecular fragments, under certain conditions. Table 1 lists the experimental data of densities for 91 high nitrogen compounds, which have been collected from different sources.

These data have been used to construct the new model by the MLR method [83]. It was found that the ratio of elemental composition to molecular weight of organic compounds containing energetic functional groups could improve the correlation coefficient significantly [32-35, 37, 38]. Thus, the core correlation as the additive part for high nitrogen organic materials with the empirical formula of $C_aH_bN_cO_d$ was derived as follows:

$$\rho_{core} = 2.216 - 9.098(a/Mw) - 5.202(b/Mw) - 5.569(c/Mw) \quad (1)$$

where $\rho_{core}$ is the core density in $g \cdot cm^{-3}$; $a$, $b$, $c$, and $d$ are the numbers of carbon, hydrogen, nitrogen, and oxygen atoms; $Mw$ is the molecular weight of the high nitrogen compound. The coefficient of determination in the MLR method ($R^2$) [83] measures the proportion of variation in the dependent variable that can be predicted from the set of independent variables. The value of $R^2$ for Equation 1 was 0.706. Since the presence of the ratio of oxygen atoms ($d$) to $Mw$ cannot improve the value of $R^2$, the contribution of $d/Mw$ has been neglected. It was found that the presence of some specific functional groups and molecular fragments leads to large deviations of the predicted results of Equation 1 from experimental data. Table 2 lists these functional groups and molecular fragments. Thus, the reliability of Equation 1 can be improved by considering non-additive correcting functions as:

**Table 1.** Comparison of the predicted results from Equations 1 and 2 for 91 high nitrogen organic compounds with experimental data ($g \cdot cm^{-3}$)

| Name | Composition | Exp. | N wt% | Equation 1 | Dev | Equation 2 | Dev |
|---|---|---|---|---|---|---|---|
| 3,3'-Bis(4-(5-methyl-1*H*-1,2,4-triazol-3-yl)-4,4'-azofurazan | $C_{10}H_8N_{12}O_2$ | 1.81 [40] | 51.2 | 1.608 | –0.202 | 1.817 | 0.007 |
| Tris[2-(5-amino-1*H*-tetrazol-1-yl)ethyl]amine | $C_9H_{18}N_{16}$ | 1.492 [41] | 63.98 | 1.46 | –0.032 | 1.451 | –0.041 |
| (Z,Z,Z)-*N*,*N*',*N*''-(1,1',1''-(Nitrilotris(ethane-2,1-diyl)) tris(1*H*-tetrazole-1(4*H*)-yl-5(4*H*)-ylidene))trinitramide | $C_9H_{15}N_{19}O_6$ | 1.616 [42] | 54.84 | 1.668 | 0.0519 | 1.669 | 0.053 |
| (1R,2R)-1,2-Bis(5-amino-1*H*-tetrazol-1-yl)cyclohexane | $C_8H_{14}N_{10}$ | 1.453 [41] | 55.98 | 1.411 | –0.042 | 1.401 | –0.052 |
| (1R,4R)-1,4-Bis(5-amino-1*H*-tetrazol-1-yl)cyclohexane | $C_8H_{14}N_{10}$ | 1.46 [41] | 55.98 | 1.411 | –0.049 | 1.401 | –0.059 |
| 3,3',5,5'-Tetra(azidomethyl)-4,4'-azo-1,2,4-triazole | $C_8H_8N_{20}$ | 1.534 [43] | 72.9 | 1.628 | 0.0938 | 1.553 | 0.019 |
| (5-Azido-1*H*-tetrazol-1-yl)-[6-(5-azido-1*H*-imino) cyclohexenyl]amine | $C_8H_8N_{16}$ | 1.65 [44] | 68.28 | 1.595 | –0.055 | 1.573 | –0.077 |
| *N*,*N*'-Bis(5-azido-1*H*-tetrazol-1-yl)-1,4-diiminocyclohexane | $C_8H_8N_{16}$ | 1.63 [44] | 68.28 | 1.595 | –0.035 | 1.573 | –0.057 |
| 3,3'-Bis(4-amino-1*H*-1,2,4-triazol-5(4*H*)-one)-4,4'-azofurazan | $C_8H_6N_{14}O_4$ | 1.68 [40] | 54.13 | 1.713 | 0.0331 | 1.703 | 0.023 |
| 3,3'- Bis[(1*H*-1,2,4-triazol-5-amine)-diyl]-4,4'-azofurazan | $C_8H_6N_{14}O_2$ | 1.79 [40] | 59.38 | 1.664 | –0.126 | 1.814 | 0.024 |
| 3,6-Bis-(2-(4,6-diazido-1,3,5-triazin-2-yl)-hydrazinyl)-1,2,4,5-tetrazine | $C_8H_4N_{26}$ | 1.755 [45] | 78.43 | 1.702 | –0.053 | 1.675 | –0.080 |
| Di(5-nitramino-1,2,4-triazoly)-azo-furazan | $C_8H_4N_{16}O_6$ | 1.873 [46] | 53.32 | 1.781 | –0.092 | 1.880 | 0.007 |
| 3, 3'-Bis(5-amino-1,2,4-oxadiazol-3-yl)-4,4'-azo-furazan | $C_8H_4N_{12}O_4$ | 1.85 [40] | 50.59 | 1.733 | –0.117 | 1.840 | –0.010 |
| 3,3'-Bis (5-amine-1,3,4-oxadiazol-yl)-4,4'-azofurazn | $C_8H_4N_{12}O_4$ | 1.83 [40] | 50.59 | 1.733 | –0.097 | 1.840 | 0.010 |
| 3,6-bis-(2-(4,6-diazido-1,3,5-triazin-2-yl)-diazenyl)-1,2,4,5-tetrazine | $C_8N_{26}$ | 1.763 [45] | 79.12 | 1.743 | –0.02 | 1.711 | –0.052 |
| 2,5,8-Triazido-s-heptazine | $C_6N_{16}$ | 1.823 [47] | 75.66 | 1.73 | –0.093 | 1.698 | –0.125 |
| (Z,Z)-*N*,*N*'-(1,1'-(Butane-1,4-diyl)bis(1*H*-tetrazole-1(4*H*)-yl-5(4*H*)-ylidene))dinitramide | $C_6H_{10}N_{12}O_4$ | 1.579 [42] | 53.49 | 1.663 | 0.0844 | 1.665 | 0.086 |
| 1,3,5-Tri(1-methyltetrazol-5-yl)pentazadiene | $C_6H_9N_{17}$ | 1.532 [48] | 74.59 | 1.601 | 0.0691 | 1.584 | 0.052 |
| 1,3,5-Tri(2-methyltetrazol-5-yl)pentaza-1,4-diene | $C_6H_9N_{17}$ | 1.524 [48] | 74.59 | 1.601 | 0.0771 | 1.584 | 0.060 |
| 1-(2-(1*H*-Pyrazol-1-yl)ethyl)-1*H*-tetrazol-5-amine | $C_6H_9N_7$ | 1.39 [49] | 54.73 | 1.431 | 0.0415 | 1.418 | 0.028 |

| Name | Composition | Exp. | N wt% | Equation 1 | Dev | Equation 2 | Dev |
|---|---|---|---|---|---|---|---|
| (E)-1-(2-(1*H*-Pyrazol-1-yl)ethyl)-5-(hydroxydiazenyl)-1*H*tetrazole | $C_6H_8N_8O$ | 1.5 [49] | 53.83 | 1.539 | 0.0391 | 1.529 | 0.029 |
| 1-(2-(3,5-Diazido-4-nitro-1*H*-pyrazol-1-yl)ethyl)-1*H*-tetrazol-5-amine | $C_6H_6N_{14}O_2$ | 1.63 [50] | 64.04 | 1.681 | 0.0506 | 1.667 | 0.037 |
| 1-(2-(4-Azido-3,5-dinitro-1*H*-pyrazol-1-yl)ethyl)-1*H*-tetrazol-5-amine | $C_6H_6N_{12}O_4$ | 1.7 [50] | 54.18 | 1.723 | 0.0234 | 1.719 | 0.019 |
| Tris(triazolo)benzene | $C_6H_3N_9$ | 1.77 [51] | 62.67 | 1.617 | –0.153 | 1.778 | 0.008 |
| 1,2-Bis(5-azido-4-nitro-1*H*-imidazol-1-yl)diazene | $C_6H_2N_{14}O_4$ | 1.77 [52] | 58.67 | 1.788 | 0.0177 | 1.776 | 0.006 |
| 3,3'-Bis(1*H*-tetrazol-5-yl)-4,4'-azofurazan | $C_6H_2N_{14}O_2$ | 1.69 [40] | 64.89 | 1.742 | 0.0524 | 1.722 | 0.032 |
| 3,3'-Dicyano-4,4'-azofurazan | $C_6N_8O_2$ | 1.62 [40] | 51.84 | 1.757 | 0.1368 | 1.681 | 0.061 |
| 1,4-Bis(azidomethyl)-3,6-dinitro-1,4-dihydropyrazolo[4,3-c]pyrazole | $C_6H_4N_{12}O_4$ | 1.71 [53] | 54.53 | 1.754 | 0.044 | 1.694 | –0.016 |
| 4,8-Bis(azidomethyl)-4*H*,8*H*-bis([1,2,5]oxadiazolo)[3,4-b:3',4'-e]pyrazine | $C_6H_4N_{12}O_2$ | 1.68 [53] | 60.86 | 1.7 | 0.0205 | 1.684 | 0.004 |
| 1,3-Bis(5-amino-1*H*-tetrazol-1-yl)propan-2-ol | $C_5H_{10}N_{10}O$ | 1.585 [41] | 61.93 | 1.538 | –0.047 | 1.533 | –0.052 |
| 1,3-Bis(5-amino-1*H*-tetrazol-1-yl)propane | $C_5H_{10}N_{10}$ | 1.567 [41] | 66.65 | 1.486 | –0.081 | 1.476 | –0.091 |
| 1,2-Bis(4,5-dihydro-5-nitroimino-1*H*-tetrazol-1-yl)propane | $C_5H_8N_{12}O_4$ | 1.658 [42] | 55.99 | 1.703 | 0.0447 | 1.704 | 0.046 |
| 1-[1,3-Diazido-2-(azidomethyl)-2-propyl]-3-nitrourea | $C_5H_8N_{12}O_3$ | 1.54 [54] | 59.14 | 1.674 | 0.1337 | 1.535 | –0.005 |
| 1-(2-(5-Amino-3-nitro-1*H*-1,2,4-triazol-1-yl)ethyl)-1*H*-tetrazol-5-amine | $C_5H_8N_{10}O_2$ | 1.66 [50] | 58.32 | 1.621 | –0.039 | 1.616 | –0.044 |
| 1,3-Bis((Z)-5-(nitroimino)-4,5-dihydro-1*H*-tetrazol-1-yl)propan-2-yl nitrate | $C_5H_7N_{13}O_7$ | 1.759 [42] | 50.41 | 1.788 | 0.0293 | 1.795 | 0.036 |
| 1-(2-(3-Nitro-1*H*-1,2,4-triazol-1-yl)ethyl)-1*H*-tetrazol-5-amine | $C_5H_7N_9O_2$ | 1.58 [50] | 55.99 | 1.629 | 0.049 | 1.623 | 0.043 |
| (Z)-*N*-(1-(2-(3-nitro-5-(nitroamino)-1*H*-1,2,4-triazol-1-yl)ethyl)-1,4-dihydro-5*H*-tetrazol-5-ylidene)nitramide | $C_5H_6N_{12}O_6$ | 1.8 [50] | 50.9 | 1.781 | –0.019 | 1.785 | –0.015 |
| (Z)-*N*-(1-(2-(3-nitro-1*H*-1,2,4-triazol-1-yl)ethyl)-1*H*-tetrazol-5(4*H*)-ylidene)nitramide | $C_5H_6N_{10}O_4$ | 1.69 [50] | 51.84 | 1.725 | 0.0354 | 1.725 | 0.035 |

| Name | Composition | Exp. | N wt% | Equation 1 | Dev | Equation 2 | Dev |
|---|---|---|---|---|---|---|---|
| 1,2-Bis(5-amino-1*H*-tetrazol-1-yl)ethane | $C_4H_8N_{10}$ | 1.567 [41] | 71.41 | 1.534 | –0.033 | 1.522 | –0.045 |
| 4,4',5,5'-Tetraamino-3,3'-bi-1,2,4-triazole | $C_4H_8N_{10}$ | 1.68 [55] | 71.41 | 1.534 | –0.146 | 1.688 | 0.008 |
| 1,1'-Ethylenebis(oxy)bis(5-nitroiminotetrazole) | $C_4H_6N_{12}O_6$ | 1.81 [56] | 52.82 | 1.793 | –0.017 | 1.799 | –0.011 |
| 1,2-Bis(4,5-dihydro-5-nitroimino-1*H*-tetrazol-1-yl)ethane | $C_4H_6N_{12}O_4$ | 1.858 [42] | 58.73 | 1.746 | –0.112 | 1.746 | –0.112 |
| 5,5'-Diamino-4,4'-dinitramino-3,3'-bi-1,2,4-triazole | $C_4H_6N_{12}O_4$ | 1.756 [57] | 58.73 | 1.746 | –0.01 | 1.746 | –0.010 |
| *N*,*N*'-Bis(5-amino-1*H*-tetrazol-1-yl)oxalamide | $C_4H_6N_{12}O_2$ | 1.65 [58] | 66.13 | 1.687 | 0.0365 | 1.679 | 0.029 |
| 1-(2-(5-Nitro-2*H*-tetrazol-2-yl)ethyl)-1*H*-tetrazol-5-amine | $C_4H_6N_{10}O_2$ | 1.7 [50] | 61.94 | 1.67 | –0.03 | 1.664 | –0.036 |
| 3,3'-Diamino-4,4'-azo-1,2,4-triazole | $C_4H_6N_{10}$ | 1.61 [59] | 72.15 | 1.58 | –0.03 | 1.563 | –0.047 |
| 4-Amino-3,5-di(azidomethyl)-1,2,4-triazole | $C_4H_6N_{10}$ | 1.563 [43] | 72.15 | 1.58 | 0.0172 | 1.563 | 0.000 |
| 3-amino-4-(5-amino-1,2,4-triazol-3-yl)furazan | $C_4H_5N_7O$ | 1.596 [60] | 58.67 | 1.609 | 0.0127 | 1.598 | 0.002 |
| 2,2'-(Oxybis(methylene))bis(5-nitro-1*H*-tetrazole) | $C_4H_4N_{10}O_5$ | 1.846 [61] | 51.46 | 1.801 | –0.045 | 1.803 | –0.043 |
| 3,3'-Diamino-5,5'-dinitro-4,4'-bis(1,2,3-triazole) | $C_4H_4N_{10}O_4$ | 1.85 [62] | 54.68 | 1.775 | –0.075 | 1.774 | –0.076 |
| 5,5'-Dinitro-2*H*,2'*H*-3,3'-bi(1,2,4-triazole)-2,2'-diamine | $C_4H_4N_{10}O_4$ | 1.83 [63] | 54.68 | 1.775 | –0.055 | 1.774 | –0.056 |
| 2,3'-Diamino-5,5'-dinitro-4,4'-bis(1,2,3-triazole) | $C_4H_4N_{10}O_4$ | 1.84 [62] | 54.68 | 1.775 | –0.065 | 1.774 | –0.066 |
| 2,2'-Diamino-5,5'-dinitro-4,4'-bis(1,2,3-triazole) | $C_4H_4N_{10}O_4$ | 1.85 [62] | 54.68 | 1.775 | –0.075 | 1.774 | –0.076 |
| 3,3'-Diamino-4,4'-azoxyfurazan (DAAF) | $C_4H_4N_8O_3$ | 1.745 [3] | 52.82 | 1.736 | –0.009 | 1.733 | –0.012 |
| 5-(5-Amino-1*H*-1,2,4-triazol-3-yl)-3-nitro-1*H*-1,2,4-triazole | $C_4H_4N_8O_2$ | 1.61 [64] | 57.13 | 1.697 | 0.0866 | 1.688 | 0.078 |
| 5-(5-Nitrimino-1,3*H*-1,2,4-triazol-3-yl)-3-nitro-1*H*-1,2,4-triazole | $C_4H_3N_9O_4$ | 1.7 [64] | 52.27 | 1.792 | 0.092 | 1.790 | 0.090 |
| 1,2-Bis(3-nitro-1*H*-1,2,4-triazol-1-yl)diazene | $C_4H_2N_{10}O_4$ | 1.8 [52] | 55.11 | 1.812 | 0.0123 | 1.807 | 0.007 |
| 5-(5-Azido-1*H*-1,2,4-triazol-3-yl)-3-nitro-1*H*-1,2,4-triazole | $C_4H_2N_{10}O_2$ | 1.68 [64] | 63.05 | 1.754 | 0.0741 | 1.739 | 0.059 |
| Urea 2-methyl-5-nitraminotetrazole | $C_3H_8N_8O_3$ | 1.56172 [65] | 54.89 | 1.66 | 0.0979 | 1.670 | 0.108 |
| 1,3-Bis(5-amino-1*H*-tetrazol-1-yl)urea | $C_3H_6N_{12}O$ | 1.65 [58] | 74.33 | 1.661 | 0.0112 | 1.651 | 0.001 |
| 3,4,5-Triamino-1-tetrazolyl-1,2,4-triazole (TATT) | $C_3H_6N_{10}$ | 1.685 [66] | 76.91 | 1.588 | –0.097 | 1.693 | 0.008 |
| 3,6,7-Triamino-7*H*-[1,2,4]triazolo[4,3-b][1,2,4]triazole | $C_3H_6N_8$ | 1.725 [67] | 72.71 | 1.547 | –0.178 | 1.701 | –0.024 |
| 1-(2-Azidoethyl)-5-aminotetrazole polymorph II | $C_3H_6N_8$ | 1.4763 [68] | 72.71 | 1.547 | 0.0703 | 1.534 | 0.058 |
| 1-Methyl-5-(methylnitramino)-1*H*-tetrazole | $C_3H_6N_6O_2$ | 1.522 [47] | 53.15 | 1.634 | 0.112 | 1.513 | –0.009 |
| 1-(2-Azidoethyl)-5-nitriminotetrazole | $C_3H_5N_9O_2$ | 1.64088 [68] | 63.31 | 1.696 | 0.0552 | 1.692 | 0.051 |

| Name | Composition | Exp. | N wt% | Equation 1 | Dev | Equation 2 | Dev |
|---|---|---|---|---|---|---|---|
| 1,1'-Methylenebis(oxy)bis(5-nitroiminotetrazole) | $C_3H_4N_{12}O_6$ | 1.9 [56] | 55.26 | 1.838 | –0.062 | 1.844 | –0.056 |
| 5,5'-(Hydrazonomethylene)bis(1*H*-tetrazole) | $C_3H_4N_{10}$ | 1.71 [69] | 77.76 | 1.639 | –0.071 | 1.620 | –0.090 |
| 5-(5-Nitrimino-1,4*H*-1,2,4-triazol-3-yl)tetrazol-1-ol | $C_3H_3N_9O_3$ | 1.85 [70] | 59.15 | 1.779 | –0.071 | 1.776 | –0.074 |
| 5-(5-Nitrimino-1,2,4-triazol-3-yl)-tetrazole | $C_3H_3N_9O_2$ | 1.7 [70] | 63.95 | 1.744 | 0.0436 | 1.735 | 0.035 |
| 3-Nitro-1-(2*H*-tetrazol-5-yl)-1*H*-1,2,4-triazol-5-amine | $C_3H_3N_9O_2$ | 1.77 [71] | 63.95 | 1.744 | –0.026 | 1.735 | –0.035 |
| Di(1*H*-tetrazol-5-yl)methanone oxime | $C_3H_3N_9O$ | 1.74 [69] | 69.6 | 1.702 | –0.038 | 1.687 | –0.053 |
| 5-Azido-4-nitro-1*H*-imidazol-1-amine | $C_3H_3N_7O_2$ | 1.65 [72] | 57.98 | 1.731 | 0.0812 | 1.725 | 0.075 |
| 5-Azido-3,4-dinitro-1*H*-pyrazol-1-amine | $C_3H_2N_8O_4$ | 1.82 [73] | 52.33 | 1.831 | 0.0114 | 1.832 | 0.012 |
| 5-(3-Nitro-1*H*-1,2,4-triazol-5-yl)tetrazol-1-ol | $C_3H_2N_8O_3$ | 1.86 [70] | 56.56 | 1.8 | –0.06 | 1.796 | –0.064 |
| 5-(5-Nitro-1,2,4-triazol-3-yl)-tetrazole | $C_3H_2N_8O_2$ | 1.7 [70] | 61.53 | 1.764 | 0.0638 | 1.754 | 0.054 |
| 5-Nitrotetrazol-2-ylacetonitrile | $C_3H_2N_6O_2$ | 1.72169 [74] | 54.53 | 1.754 | 0.0323 | 1.746 | 0.024 |
| 5-Amino-1-methoxy-1*H*-tetrazole | $C_2H_5N_5O$ | 1.503 [41] | 60.86 | 1.589 | 0.0862 | 1.487 | –0.016 |
| 5,5'-Hydrazinebistetrazole (HBT) | $C_2H_4N_{10}$ | 1.86195 [75] | 83.32 | 1.652 | –0.21 | 1.826 | –0.036 |
| 1-Methoxy-5-nitroiminotetrazole | $C_2H_4N_6O_3$ | 1.66 [56] | 52.49 | 1.763 | 0.1032 | 1.668 | 0.008 |
| 3-Nitro-1*H*-1,2,4-triazole-1,5-diamine | $C_2H_4N_6O_2$ | 1.76 [73] | 58.33 | 1.713 | –0.047 | 1.716 | –0.044 |
| 2,4-Diamino-5-nitro-1,2,3-triazole | $C_2H_4N_6O_2$ | 1.73 [76] | 58.33 | 1.713 | –0.017 | 1.716 | –0.014 |
| 3,4-Diamino-5-nitro-1,2,3-triazole | $C_2H_4N_6O_2$ | 1.71 [76] | 58.33 | 1.713 | 0.0029 | 1.716 | 0.006 |
| 5,5'-Bis(1*H*-tetrazolyl)amine | $C_2H_3N_9$ | 1.83404 [77] | 82.34 | 1.667 | –0.167 | 1.838 | 0.004 |
| 3-Nitro-1*H*-1,2,4-triazol-1-amine | $C_2H_3N_5O_2$ | 1.68 [73] | 54.26 | 1.738 | 0.0579 | 1.741 | 0.061 |
| 1-Nitroamino-1,2,3-triazole | $C_2H_3N_5O_2$ | 1.8 [78] | 54.26 | 1.738 | –0.062 | 1.812 | 0.012 |
| 4-Nitroamino-1,2,4-triazole | $C_2H_3N_5O_2$ | 1.82 [78] | 54.26 | 1.738 | –0.082 | 1.812 | –0.008 |
| 1-(2*H*-Tetrazol-5-yl)-5-nitraminotetrazole | $C_2H_2N_{10}O_2$ | 1.77 [79] | 70.7 | 1.79 | 0.02 | 1.781 | 0.011 |
| Nitroaminoguanidine | $CH_5N_5O_2$ | 1.71 [12] | 58.82 | 1.687 | –0.023 | 1.704 | –0.006 |
| 1,5-Di(nitramino)tetrazole | $CH_2N_8O_4$ | 1.93 [80] | 58.94 | 1.879 | –0.051 | 1.887 | –0.043 |
| 5-Azido-1*H*-tetrazole | $CHN_7$ | 1.67039 [81] | 88.28 | 1.736 | 0.0653 | 1.713 | 0.043 |
| 5-Nitro-2-hydroxytetrazole | $CHN_5O_3$ | 1.90469 [82] | 53.43 | 1.894 | –0.011 | 1.902 | –0.002 |
| 1-Methyl-5-(methylnitramino)-1*H*-tetrazole | $C_3H_6N_6O_2$ | 1.522 [47] | 53.15 | 1.634 | 0.112 | 1.513 | –0.009 |

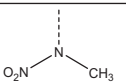
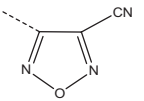
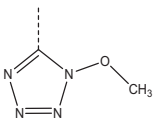
$$\rho = 2.278 - 10.58(a/Mw) - 4.775(b/Mw) - 6.763(c/Mw) + 0.1188\rho^{+}_{non\text{-}add} - 0.1046\rho^{-}_{non\text{-}add} \quad (2)$$

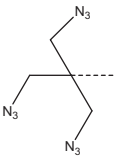
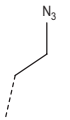
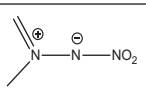
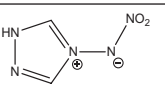
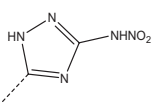
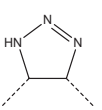
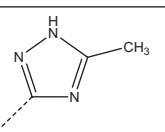
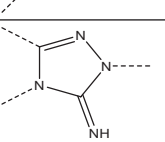
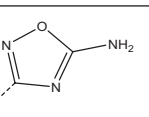
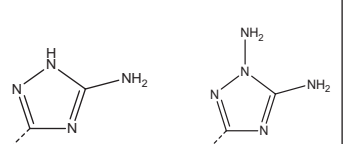
where $\rho^{+}_{non\text{-}add}$ and $\rho^{-}_{non\text{-}add}$ are two non-additive correcting functions, which can increase and decrease the density of a high nitrogen compound, respectively. Table 2 provides different values of $\rho^{+}_{non\text{-}add}$ and $\rho^{-}_{non\text{-}add}$, which have been derived by tedious work on the basis of the following steps:

1) *Specification of common functional groups and molecular fragments between different high nitrogen organic compounds*. The effects of common functional groups and molecular fragments, confirmed in previous studies [32-35, 37, 38], which may give large deviations from those obtained by the contributions of elemental composition.
2) *Adjustment of the best values for* $\rho^{+}_{non\text{-}add}$ *and* $\rho^{-}_{non\text{-}add}$. These data were obtained by regression of different quantities for $\rho^{+}_{non\text{-}add}$ and $\rho^{-}_{non\text{-}add}$ to give the highest value of $R^2$ and lowest the root-mean-square deviation (*RMSD*).

Thus, the functional groups and molecular fragments listed in Table 2 were specified for some derivatives of high nitrogen organic compounds that show large positive or negative deviations from Equation 1. Details of the derivation method have been illustrated elsewhere [84]. For those high nitrogen organic compounds that do not contain the functional groups listed in Table 2, the values of $\rho^{+}_{non\text{-}add}$ and $\rho^{-}_{non\text{-}add}$ are zero.

**Table 2.** The values of $\rho^{+}_{non\text{-}add}$ and $\rho^{-}_{non\text{-}add}$

| | $\rho^{-}_{non\text{-}add}$ | | |
|---|---|---|---|
| Molecular moieties | $\rho^{-}_{non\text{-}add}$ | Examples | Comments |
| –N(R)($NO_2$ or NO) | 1.2 |  | – |
| –CN | 0.5 |  | The attachemnt of –CN to heterocyclic ring. |
| –OR | 1.1 |  | The attachment of –OR to tetrazole ring. |

| | | | |
|---|---|---|---|
| $-N_3$ | 1.3 |  | The attachment of three $(CH_2)_n$-$N_3$ to tertiary carbon. |
| | 0.5 |  | The attachment of more than two $-(CH_2)_n-N_3$ to heterocyclic rings. |
| | | $\rho^+_{non\text{-}add}$ | |
| Molecular moieties | $\rho^+_{non\text{-}add}$ | Examples | Comments |
|  | 0.6 |  | – |
| $-NH-NO_2$ | 0.9 |  | The attachment of more than one -NH-$NO_2$ to triazole rings without other substituents. |
| –NH– | 1.6 |  | The presence of more than two –NH– groups without substituents in ring case. |
| –R | 1.9 |  | The attachment of more than one –R groups to triazole rings. |
| =NH (cyclic) | 1.0 |  | – |
| $-NH_2$ | 1.0 |  | The attachment of more than two $-NH_2$ groups to oxadiazole rings. |
| | 1.4 |  | The attachment of more than one $-NH_2$ group to triazole rings without other substituents or –N=N– between two triazole rings. |

# 3 Results and Discussion

## 3.1 Statistical assessment of Equation 2

Due to the uncertainty of experimental data, the value of $R^2$ for Equation 2 was 0.878, which shows an improvement with respect to Equation 1. The standard deviation (*SD*) of the new model estimates the deviation of predictions with respect to the experimental values. The value of *SD* for Equation 2 was equal to 0.047 g·cm$^{-3}$, where $F$ = 165.1 and the significance of $F$ = 5.14E–51, which is lower than the value calculated for Equation 1, *i.e.* 0.076 g·cm$^{-3}$.

Table 3 provides the statistical parameters for five variables of Equation 2. The values of the standard deviations (*sd*) for these variables indicate whether or not the individual variables are significant in predicting the density. Thus, the contribution of a specified variable is significant if its standard deviation is small relative to the coefficient [85]. The *P*-value is another statistical parameter that gives the probability that a parameter estimated from the measured data should have the value that was determined. If the *P*-value for a variable is less than 0.05, the effect of that variable is significant and the observed effect is not due to random variations. As indicated in Table 3, all of the five variables given in Equation 2 have suitable statistical parameters because they have a highly significant impact in the development of Equation 2.

**Table 3.** Regression coefficients, standard deviations (*sd*), *P*-values, and confidence intervals of the best linear regression model

| | Coefficient | *sd* | *P*-value | Lower limit (95%) | Upper limit (95%) |
|---|---|---|---|---|---|
| *Intercept* | 2.278 | 0.035 | 1.79E–92 | 2.208 | 2.348 |
| *a*/*Mw* | –10.58 | 0.810 | 1.49E–24 | –12.19 | –8.981 |
| *b*/*Mw* | –4.775 | 0.322 | 1.41E–28 | –5.412 | –4.137 |
| *c*/*Mw* | –6.763 | 0.651 | 2.93E–18 | –8.053 | –5.473 |
| $\rho^{+}_{non\text{-}add}$ | 0.1188 | 0.011 | 2.55E–19 | 0.0970 | 0.1405 |
| $\rho^{-}_{non\text{-}add}$ | –0.1046 | 0.018 | 7.06E–08 | –0.1405 | –0.0686 |

## 3.2 Comparison of the reliability of the new model with two complex quantum mechanical approaches

A suitable method using quantum-mechanically determined molecular volumes ($V_M$) of an isolated molecule has been developed for estimating the densities of neutral molecules as [20]:

$$\rho = Mw_{molecule}/V_M \quad (3)$$

where $Mw_{molecule}$ is the molecular mass of the molecule and $V_M$ is the volume inside the 0.001 a.u. isosurface of electron density surrounding the molecule, calculated using density functional theory (DFT) at the B3LYP/6-31G** level [86] with the Gaussian program package [87]. It was indicated that adding corrections for electrostatic interactions can be done to better represent the intermolecular interactions in neutral CHNO crystals as [88]:

$$\rho = \alpha_1(Mw_{molecule}/V_M) + \beta_1(v\sigma^2_{tot}) + \gamma_1 \quad (4)$$

where $\sigma^2_{tot}$ is the total variance of the electrostatic potential on the 0.001 a.u. molecular surface; the parameter $v$ quantifies the degree of balance between the positive and negative potentials on the molecular surface; the three parameters $\alpha_1$, $\beta_1$ and $\gamma_1$ were found through least squares fitting to experimental information.

Table 4 compares the predicted results from Equations 1 and 2 with the output from the quantum-mechanical approaches of Equations 3 and 4, which have been computed by Rice and Byrd [22] for a further 32 high nitrogen organic compounds as a test set. Table 5 provides the different statistical parameters of Equations 1 and 2 as compared to the complex methods given in Equations 3 and 4. Five statistical parameters are listed in Table 5, *viz* the average deviation (*AD*), *SD*, absolute maximum deviation ($AD_{max}$), average absolute deviation (*AAD*), absolute maximum deviation ($AD_{max}$) and average absolute relative deviation (*AARD%*). *AD* measures accuracy, which defines the value of bias and systematic errors. As mentioned before, *SD* is a measure of precision, which can be used to determine the spread of data around the regression model. $AD_{max}$ is another measure of precision because it determines the worst error in the predictions. *AAD* is a linear measure of errors, which show the average size of errors. *AARD%* provides an easy way of judging the extent, or importance of errors, which facilitates relative comparisons among various methods. If the ratio of *SD*/*AAD* approaches 1.25, it shows that the errors are distributed in a normal pattern. For a heavily-tailed distribution of errors, the *SD*/*AAD* ratio approaches 1.71 [89]. The values of the *SD*/*AAD* ratio for Equation 1, Equation 2, Equation 3 and Equation 4 were 1.26, 1.29, 1.19 and 1.2, respectively. All of the statistical parameters in Table 5 confirm that Equation 2 is an improvement over Equation 1, where the results are unbiased and free from systematic errors. Equations 2 and 4 can predict the density of high nitrogen compounds with adequate accuracy and precision.

**Table 4.** Comparison of the predicted results of Equations 1 and 2 for high nitrogen organic compounds with experimental data ($g \cdot cm^{-3}$), as well as the output from the quantum-mechanical approaches of Equations 3 and 4, which have been computed by Rice and Byrd [22]

| Name | Composition | Exp. [47] | N% | Equation 1 | Dev | Equation 2 | Dev | Equation 3 | Dev | Equation 4 | Dev |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 3-Amino-6-(3,5-dimethylpyrazol-1-yl)-1,2,4,5-tetrazine | $C_7H_9N_7$ | 1.449 | 51.29 | 1.433 | –0.016 | 1.417 | –0.032 | 1.414 | 1.367 | 1.452 | 0.003 |
| 1-Phenyl-5-(azido)-1*H*-tetrazole | $C_7H_5N_7$ | 1.515 | 52.39 | 1.528 | 0.0126 | 1.501 | –0.014 | 1.487 | 1.460 | 1.463 | –0.052 |
| 4,4',6,6'-Tetra-azido-2,2'-azo-1,3,5-triazine (triclinic beta) | $C_6N_{20}$ | 1.674 | 79.53 | 1.744 | 0.0702 | 1.713 | 0.039 | 1.713 | 1.713 | 1.696 | 0.022 |
| 4,4',6,6'-Tetra-azido-2,2'-azo-1,3,5-triazine (orthorhombic alpha) | $C_6N_{20}$ | 1.704 | 79.53 | 1.744 | 0.0402 | 1.713 | 0.009 | 1.718 | 1.718 | 1.689 | –0.015 |
| 4,4',6,6'-Tetra-azido-2,2'-hydrazo-1,3,5-triazine | $C_6H_2N_{20}$ | 1.649 | 79.08 | 1.718 | 0.0685 | 1.689 | 0.040 | 1.703 | 1.697 | 1.678 | 0.029 |
| 1,4-Bis-[1-methyltetrazol-5-yl]-1,4-dimethyl-2-tetrazene | $C_6H_{12}N_{12}$ | 1.473 | 66.65 | 1.486 | 0.0132 | 1.476 | 0.003 | 1.458 | 1.410 | 1.433 | –0.040 |
| *N*,*N*-Dimethyl-*N*'-(5-methyltetrazol-1-yl)formamidine | $C_5H_{10}N_6$ | 1.328 | 54.53 | 1.366 | 0.0376 | 1.361 | 0.033 | 1.306 | 1.241 | 1.297 | –0.031 |
| *N*,*N*-Bis(1-*H*-3-methyl-tetrazol-5-yl)-methylamine | $C_5H_9N_9$ | 1.4901 | 64.6 | 1.485 | –0.005 | 1.474 | –0.016 | 1.458 | 1.412 | 1.441 | –0.049 |
| *N*,*N*-Bis(1-*H*-3-methyl-tetrazol-5-yl)-amine | $C_4H_7N_9$ | 1.50783 | 69.6 | 1.537 | 0.0288 | 1.523 | 0.015 | 1.495 | 1.456 | 1.482 | –0.026 |
| 2,2'-Dimethyl azotetrazolate | $C_4H_6N_{10}$ | 1.48 | 72.15 | 1.58 | 0.1002 | 1.563 | 0.083 | 1.520 | 1.489 | 1.517 | 0.037 |
| 2,2'-Dimethyl-5,5'-azotetrazole | $C_4H_6N_{10}$ | 1.515 | 72.15 | 1.58 | 0.0652 | 1.563 | 0.048 | 1.522 | 1.491 | 1.520 | 0.005 |
| 4,5-Dicyano-1*H*-1,2,3-Triazole | $C_4HN_5$ | 1.554 | 58.8 | 1.632 | 0.0782 | 1.545 | –0.009 | 1.505 | 1.497 | 1.544 | –0.010 |
| 1-Hydroxyethyl-5-aminoterazole | $C_3H_7N_5O$ | 1.511 | 54.25 | 1.506 | –0.005 | 1.510 | –0.001 | 1.437 | 1.383 | 1.499 | –0.012 |

| Name | Composition | Exp. [47] | N% | Equation 1 | Dev | Equation 2 | Dev | Equation 3 | Dev | Equation 4 | Dev |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1-Methyl-5-(methylamino)-1*H*-tetrazole | $C_3H_7N_5$ | 1.381 | 61.93 | 1.406 | 0.0246 | 1.402 | 0.021 | 1.330 | 1.268 | 1.378 | –0.003 |
| 1-Azidoethyl-5-aminotetrazole | $C_3H_6N_8$ | 1.4763 | 72.71 | 1.547 | 0.0703 | 1.534 | 0.058 | 1.466 | 1.427 | 1.501 | 0.025 |
| 1-Methyl-5-(methylnitrosoamino)-1*H*-tetrazole | $C_3H_6N_6O$ | 1.444 | 59.14 | 1.568 | 0.1244 | 1.441 | –0.003 | 1.485 | 1.443 | 1.446 | 0.002 |
| 5,6-Dihydro-1*H*-imidazo[1,2-d] tetrazole | $C_3H_5N_5$ | 1.524 | 63.05 | 1.485 | –0.039 | 1.472 | –0.052 | 1.462 | 1.417 | 1.497 | –0.027 |
| 7-Nitro-5,6-dihydro-7*H*-imidazolo[1,2-d]tetrazole | $C_3H_4N_6O_2$ | 1.69 | 53.84 | 1.693 | 0.0032 | 1.692 | 0.002 | 1.667 | 1.641 | 1.715 | 0.025 |
| 7-Nitroso-5,6-dihydro-7*H*-imidazolo[1,2-d]tetrazole | $C_3H_4N_6O$ | 1.647 | 59.99 | 1.634 | –0.013 | 1.625 | –0.022 | 1.594 | 1.565 | 1.628 | –0.019 |
| 2,4,6-Triazido-1,3,5-triazine | $C_3N_{12}$ | 1.736 | 82.34 | 1.754 | 0.0184 | 1.724 | –0.012 | 1.690 | 1.690 | 1.648 | –0.088 |
| *N*-Methyl-1*H*-tetrazol-5-amine | $C_2H_5N_5$ | 1.479 | 70.69 | 1.488 | 0.009 | 1.481 | 0.002 | 1.397 | 1.346 | 1.463 | –0.016 |
| 1-Methyl-5-nitriminotetrazole | $C_2H_4N_6O_2$ | 1.716 | 58.33 | 1.713 | –0.003 | 1.716 | 0.000 | 1.638 | 1.610 | 1.658 | –0.058 |
| 2-Methyl-5-nitriminotetrazole | $C_2H_4N_6O_2$ | 1.667 | 58.33 | 1.713 | 0.0459 | 1.716 | 0.049 | 1.617 | 1.589 | 1.606 | –0.061 |
| 3,6-Diamino-1,2,4,5-tetrazine | $C_2H_4N_6$ | 1.611 | 74.99 | 1.569 | –0.042 | 1.556 | –0.055 | 1.506 | 1.470 | 1.524 | –0.087 |
| 5-Azido-1-methyl-2*H*-tetrazole | $C_2H_3N_7$ | 1.533 | 78.39 | 1.633 | 0.1005 | 1.615 | 0.082 | 1.520 | 1.496 | 1.494 | –0.039 |
| 1,2,4-Triazole | $C_2H_3N_3$ | 1.456 | 60.85 | 1.484 | 0.0278 | 1.469 | 0.013 | 1.377 | 1.334 | 1.432 | –0.024 |
| s-Tetrazine | $C_2H_2N_4$ | 1.501 | 68.28 | 1.595 | 0.0943 | 1.573 | 0.072 | 1.470 | 1.446 | 1.435 | –0.066 |
| 3,6-Diazido-1,2,4,5-tetrazine | $C_2N_{10}$ | 1.719 | 85.36 | 1.765 | 0.0462 | 1.736 | 0.017 | 1.686 | 1.686 | 1.659 | –0.060 |
| *N,N'*-Diaminourea | $CH_6N_4O$ | 1.518 | 62.22 | 1.52 | 0.0024 | 1.541 | 0.023 | 1.395 | 1.328 | 1.454 | –0.064 |
| 5-Nitriminotetrazole | $CH_2N_6O_2$ | 1.894 | 64.61 | 1.809 | –0.085 | 1.811 | –0.083 | 1.765 | 1.750 | 1.841 | –0.053 |
| 1*H*-1,2,3,4-Tetrazole | $CH_2N_4$ | 1.529 | 79.99 | 1.619 | 0.0899 | 1.604 | 0.075 | 1.486 | 1.457 | 1.563 | 0.034 |
| Tetrazole azide | $CHN_7$ | 1.67 | 88.28 | 1.736 | 0.0657 | 1.713 | 0.043 | 1.628 | 1.619 | 1.701 | 0.031 |

Thus, the reliability of the simple model of Equation 1 is close to Equation 3. Moreover, the improved correlations 2 and 4 can make predictions that are more reliable than Equations 1 and 3. Comparison between the predicted values from the simple models of Equations 1 and 2, as well as from the complex quantum mechanical approaches of Equations 3 and 4, with experimental data is also indicated in Figure 1.

**Table 5.** Comparison of the statistical parameters of Equations 1 and 2 with the output from the quantum-mechanical approaches of Equations 3 and 4, which have been computed by Rice and Byrd [22] for 32 high nitrogen organic compounds

| Model | *AD* [g·cm$^{-3}$] | *SD* [g·cm$^{-3}$] | $AD_{max}$ [g·cm$^{-3}$] | *AAD* [g·cm$^{-3}$] | *AARD%* |
|---|---|---|---|---|---|
| Equation 1 | 0.032 | 0.057 | 0.124 | 0.045 | 2.903 |
| Equation 2 | 0.013 | 0.042 | 0.083 | 0.032 | 2.057 |
| Equation 3 | 0.035 | 0.056 | 0.129 | 0.047 | 3.007 |
| Equation 4 | –0.021 | 0.042 | 0.088 | 0.035 | 2.191 |

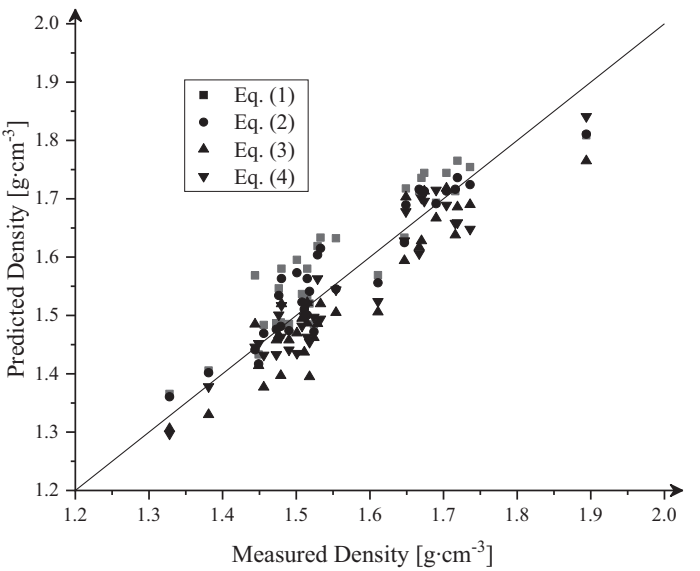



**Figure 1.** Comparison between the predicted values from the simple models of Equations 1 and 2, as well as the complex quantum mechanical approaches of Equations 3 and 4, with experimental data

## 4 Conclusions

A reliable approach was introduced for the prediction of the density of high nitrogen organic compounds (>50 wt.% N) at or near room temperature. Equation 1 is a core correlation, which is based on elemental composition, for the prediction of the density, and its reliability was improved by considering $\rho^{+}_{non\text{-}add}$ and $\rho^{-}_{non\text{-}add}$ in Equation 2. These equations can be applied for different types of high nitrogen organic compounds, including triazole, tetrazole, triazine, tetrazine, furazan, and some of organic nitrogen-containing chains. The reliability of these equations was compared with the output from two complex quantum mechanical models. The former facilitate the calculation of densities of different high nitrogen compounds, without the need to use complicated expensive computer programs, as well as expert operators. Thus, the present method provides the simplest and easiest procedure and at the same time gives reliable results relative to complex quantum mechanical methods.

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## References

[1] Talawar, M.B.; Sivabalan, R.; Asthana, S.N.; Singh, H. Novel Ultrahigh-energy Materials. *Combust., Explos., Shock Waves* **2005**, *41*(3): 264-277.

[2] Eremets, M.I.; Trojan, I.A.; Gavriliuk, A.G.; Medvedev, S.A. Synthesis of High-Nitrogen Energetic Material. In: *Static Compression of Energetic Materials.* (Peiris, S.M.; Piermarini, G.J.; Eds.), Springer, **2008**, pp. 75-97.

[3] Klapötke, T.M. *Energetic Materials Encyclopedia*. Walter de Gruyter GmbH & Co KG, **2018**.

[4] Keshavarz, M.H. *Combustible Organic Materials: Determination and Prediction of Combustion Properties*. Walter de Gruyter GmbH & Co KG, **2018**.

[5] Shreeve, J.M.; Wei, H.; Zhang, J. Synthesis, Characterization, and Energetic Properties of 6-Amino-tetrazolo[1,5-b]-1,2,4,5-tetrazine-7-*N*-oxide: a Nitrogen-Rich Material with High Density. *Chem. Asian J.* **2015**, *10*(5): 1130-1132.

[6] Lee, K.-Y.; Gilardi, R.; Hiskey, M.A.; Stine, J.R. ANTA and Its Oxidation Products. In: *Decomposition, Combustion, and Detonation Chemistry of Energetic Materials* (Brill, T.B.; Russell, T.P.; Tao, W.C.; Wardle, R.B., Eds.), Symp. Proc., vol. 418, Materials Research Society (MRS), Boston, Massachusetts, USA, **1995**.

[7] Hiskey, M.A.; Goldman, N.; Stine, J.R. High-nitrogen Energetic Materials Derived from Azotetrazolate. *J. Energ. Mater.* **1998**, *16*(2-3): 119-127.

[8] Hiskey, M.A.; Chavez, D.E.; Naud, D. *Preparation of 3,3'-Azobis(6-amino-1,2,4,5-tetrazine).* US Patent 6342589, **2002**.
[9] Chavez, D.E.; Hiskey, M.A. 1,2,4,5-Tetrazine Based Energetic Materials. *J. Energ. Mater.* **1999**, *17*(4): 357-377.
[10] Agrawal, J.P. *High Energy Materials: Propellants, Explosives and Pyrotechnics*. WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, **2010**.
[11] Keshavarz, M.H.; Klapötke, T.M. *Energetic Compounds: Methods for Prediction of Their Performance*. Walter de Gruyter GmbH & Co KG, **2017**.
[12] Klapötke, T.M. *Chemistry of High-Energy Materials*. Walter de Gruyter GmbH & Co KG, **2017**.
[13] Keshavarz, M.H.; Klapötke, T.M. *The Properties of Energetic Materials: Sensitivity, Physical and Thermodynamic Properties*. Walter de Gruyter GmbH & Co KG, **2017**.
[14] Keshavarz, M.H. *Liquid Fuels as Jet Fuels and Propellants*. Nova Science Publishers, New York, **2018**.
[15] Klapötke, T.M. The Synthesis Chemistry of Energetic Materials. In: *Energetics Science and Technology in Central Europe.* (Armstrong, R.W., Ed.) CALCE EPSC Press, Maryland, **2012**, pp. 57-71.
[16] Atkins, R.L. *Advanced Energetic Materials*. Washington DC, USA, **2004**, p. 51.
[17] Damse, R.; Ghosh, M.; Naik, N.; Sikder, A. Thermoanalytical Screening of Nitrogen-rich Compounds for Ballistic Requirements of Gun Propellant. *J. Propul. Power* **2009**, *25*(1): 249-256.
[18] Nair, U.R.; Asthana, S.N.; Rao, A.S.; Gandhe, B.R. Advances in High Energy Materials (Review Paper). *Def. Sci. J.* **2010**, *60*(2): 137-151.
[19] Gao, H.; Shreeve, J.M. Azole-based Energetic Salts. *Chem. Rev.* **2011**, *111*(11): 7377-7436.
[20] Rice, B.M., Hare, J.J.; Byrd E.F. Accurate Predictions of Crystal Densities using Quantum Mechanical Molecular Volumes. *J. Phys. Chem., A* **2007**, *111*(42): 10874-10879.
[21] Qiu, L.; Xiao, H.; Gong, X.; Ju, X.; Zhu, W. Crystal Density Predictions for Nitramines Based on Quantum Chemistry. *J. Energ. Mater.* **2007**, *141*(1): 280-288.
[22] Rice, B.M.; Byrd, E.F. Evaluation of Electrostatic Descriptors for Predicting Crystalline Density. *J. Comput. Chem.* **2013**, *34*(25): 2146-2151.
[23] Wang, G.; Gong, X.; Liu, Y.; Du, H.; Xiao, H. Prediction of Crystalline Densities of Polynitro Arenes for Estimation of Their Detonation Performance Based on Quantum Chemistry. *J. Mol. Struct.: THEOCHEM* **2010**, *953*(1-3): 163-169.
[24] Wang, G.; Xu, Y.; Xue, C.; Ding, Z.; Liu, Y.; Liu, H.; Gong, X. Prediction of the Crystalline Densities of Aliphatic Nitrates by Quantum Chemistry Methods. *Cent. Eur. J. Energ. Mater.* **2019**, *16*(3): 412-432.
[25] Nirwan, A.; Devi, A.; Ghule, V.D. Assessment of Density Prediction Methods Based on Molecular Surface Electrostatic Potential. *J. Mol. Model.* **2018**, *24*(7): 166.
[26] Rahimi, R.; Keshavarz, M.H.; Akbarzadeh, A.R. Prediction of the Density of

Energetic Materials on the Basis of their Molecular Structures. *Cent. Eur. J. Energ. Mater.* **2016**, *13*(1): 73-101.

[27] Fathollahi, M.; Sajady, H. Prediction of Density of Energetic Cocrystals Based on QSPR Modeling using Artificial Neural Network. *Struct. Chem.* **2018**, *29*(4): 1119-1128.

[28] Zohari, N.; Bajestani, I.R. A Novel Correlation for Predicting the Density of Tetrazole – *N*-oxide Salts as Green Energetic Materials through Their Molecular Structure. *Cent. Eur. J. Energ. Mater.* **2018**, *15*(4): 629-651.

[29] Nazari, B.; Keshavarz, M.H.; Hamadanian, M.; Mosavi, S.; Ghaedsharafi, A.R.; Pouretedal, H.R. Reliable Prediction of the Condensed (Solid or Liquid) Phase Enthalpy of Formation of Organic Energetic Materials at 298 K through Their Molecular Structures. *Fluid Phase Equilib.* **2016**, *408*: 248-258.

[30] Zohari, N.; Abrishami, F.; Zeynali, V. Using the QSPR Approach for Estimating the Density of Azole-based Energetic Compounds. *Z. Anorg. Allg. Chem.* **2017**, *643*(24): 2124-2137.

[31] Zohari, N.; Sheibani, N. Link between Density and Molecular Structures of Energetic Azido Compounds as Green Plasticizers. *Z. Anorg. Allg. Chem.* **2016**, *642*(24): 1472-1479.

[32] Keshavarz, M.H.; Pouretedal, H.R. A Reliable Simple Method to Estimate Density of Nitroaliphatics, Nitrate Esters and Nitramines. *J. Hazard. Mater.* **2009**, *169*(1-3): 158-169.

[33] Keshavarz, M.H. Novel Method for Predicting Densities of Polynitro Arene and Polynitro Heteroarene Explosives in order to Evaluate Their Detonation Performance. *J. Hazard. Mater.* **2009**, *165*(1-3): 579-588.

[34] Keshavarz, M.H. New Method for Calculating Densities of Nitroaromatic Explosive Compounds. *J. Hazard. Mater.* **2007**, *145*(1-2): 263-269.

[35] Keshavarz, M.H.; Motamedoshariati, H.; Moghayadnia, R.; Ghanbarzadeh, M.; Azarniamehraban, J. A New Computer Code for Assessment of Energetic Materials with Crystal Density, Condensed Phase Enthalpy of Formation, and Activation Energy of Thermolysis. *Propellants, Explos., Pyrotech.* **2013**, *38*(1): 95-102.

[36] Keshavarz, M.H.; Pouretedal, H.R.; Saberi, E. A Simple Method for Prediction of Density of Ionic Liquids through Their Molecular Structure. *J. Mol. Liq.* **2016**, *216*: 732-737.

[37] Keshavarz, M.H. Prediction of Densities of Acyclic and Cyclic Nitramines, Nitrate Esters and Nitroaliphatic Compounds for Evaluation of their Detonation Performance. *J. Hazard. Mater.* **2007**, *143*(1-2): 437-442.

[38] Keshavarz, M.H.; Klapötke, T.M.; Sućeska, M. Energetic Materials Designing Bench (EMDB), Version 1.0. *Propellants, Explos., Pyrotech.* **2017**, *42*(8): 854-856.

[39] Keshavarz, M.H.; Rahimi, R.; Akbarzadeh, A.R. Two Novel Correlations for Assessment of Crystal Density of Hazardous Ionic Molecular Energetic Materials using Their Molecular Structures. *Fluid Phase Equilib.* **2015**, *402*: 1-8.

[40] Qu, Y.; Zeng, Q.; Wang, J.; Ma, Q.; Li, H.; Li, H.; Yang, G. Furazans with Azo Linkages: Stable CHNO Energetic Materials with High Densities, Highly Energetic

Performance, and Low Impact and Friction Sensitivities. *Chem. – Eur. J.* **2016**, *22*(35): 12527-12532.
[41] Joo, Y.-H.: Shreeve, J.M. 1-Substituted 5-Aminotetrazoles: Syntheses from $CNN_3$ with Primary Amines. *Org. Lett.* **2008**, *10*(20): 4665-4667.
[42] Joo, Y.H.; Shreeve, J.M. Energetic Mono-, Di-, and Trisubstituted Nitroiminotetrazoles. *Angew. Chem., Int. Ed.* **2009**, *48*(3): 564-567.
[43] Tang, Y.; Shreeve, J.M. Nitroxy/Azido-Functionalized Triazoles as Potential Energetic Plasticizers. *Chem. – Eur. J.* **2015**, *21*(19): 7285-7291.
[44] Abe, T.; Joo, Y.H.; Tao, G.H.; Twamley, B.; Shreeve, J.M. Disubstituted Azidotetrazoles as Energetic Compounds. *Chem. – Eur. J.* **2009**, *15*(16): 4102-4110.
[45] Chen, D.; Yang, H.; Yi, Z.; Xiong, H.; Zhang, L.; Zhu, S.; Cheng, G. $C_8N_{26}H_4$: an Environmentally Friendly Primary Explosive with High Heat of Formation. *Angew. Chem.* **2018**, *130*(8): 2103-2106.
[46] Xu, Z.; Cheng, G.; Yang, H.; Ju, X.; Yin, P.; Zhang, J.; Shreeve, J.M. A Facile and Versatile Synthesis of Energetic Furazan-Functionalized 5-Nitroimino-1,2,4-Triazoles. *Angew. Chem., Int. Ed.* **2017**, *56*(21): 5877-5881.
[47] Allen, F.H. The Cambridge Structural Database: a Quarter of a Million Crystal Structures and Rising. *Acta Crystallogr., Sect. B: Struct. Sci.* **2002**, *58*(3): 380-388.
[48] Wang, Q.; Pang, F.; Wang, G.; Huang, J.; Nie, F.; Chen, F.-X. Pentazadiene: a High-nitrogen Linkage in Energetic Materials. *Chem. Commun.* **2017**, *53*(15): 2327-2330.
[49] Kumar, D.; He, C.; Mitchell, L.A.; Parrish, D.A.; Shreeve, J.M. Connecting Energetic Nitropyrazole and Aminotetrazole Moieties with *N*,*N*′-Ethylene Bridges: A Promising Approach for Fine Tuning Energetic Properties. *J. Mater. Chem. A* **2016**, *4*(23): 9220-9228.
[50] Kumar, D.; Mitchell, L.A.; Parrish, D.A.; Shreeve, J.M. Asymmetric N,N′-Ethylene-bridged Azole-based Compounds: Two Way Control of the Energetic Properties of Compounds. *J. Mater. Chem. A* **2016**, *4*(25): 9931-9940.
[51] Thottempudi, V.; Forohor, F.; Parrish, D.A.; Shreeve, J.M. Tris(triazolo)benzene and Its Derivatives: High-Density Energetic Materials. *Angew. Chem., Int. Ed.* **2012**, *51*(39): 9881-9885.
[52] Yin, P.; Parrish, D.A.; Shreeve, J.M. N-Diazo-Bridged Nitroazoles: Catenated Nitrogen-Atom Chains Compatible with Nitro Functionalities. *Chem. – Eur. J.* **2014**, *20*(22): 6707-6712.
[53] Zhang, J.; Yin, P.; Mitchell, L.A.; Parrish, D.A.; Shreeve, J.M. N-functionalized Nitroxy/Azido Fused-ring Azoles as High-performance Energetic Materials. *J. Mater. Chem. A* **2016**, *4*(19): 7430-7436.
[54] Joo, Y.-H.; Shreeve, J.M. 1,3-Diazido-2-(azidomethyl)-2-propylammonium Salts. *Inorg. Chem.* **2009**, *48*(17): 8431-8438.
[55] Klapötke, T.M.; Schmid, P.C.; Schnell, S.; Stierstorfer, J. Thermal Stabilization of Energetic Materials by the Aromatic Nitrogen-rich 4,4′,5,5′-Tetraamino-3,3′-bi-1,2,4-triazolium Cation. *J. Mater. Chem. A* **2015**, *3*(6): 2658-2668.
[56] Joo, Y.H.; Shreeve, J.M. High-density Energetic Mono- or Bis(oxy)-5-

nitroiminotetrazoles. *Angew. Chem., Int. Ed.* **2010**, *49*(40): 7320-7323.
[57] Klapötke, T.M.; Leroux, M.; Schmid, P.C.; Stierstorfer, J. Energetic Materials Based on 5,5′-Diamino-4,4′-dinitramino-3,3′-bi-1,2,4-triazole. *Chem. – Asian J.* **2016**, *11*(6): 844-851.
[58] Joo, Y.H.; Twamley, B.; Shreeve, J.M. Carbonyl and Oxalyl Bridged Bis(1,5-Diaminotetrazole)-based Energetic Salts. *Chem. – Eur. J.* **2009**, *15*(36): 9097-9104.
[59] Liu, W.; Li, S.-h.; Li, Y.-c.; Yang, Y.-z.; Yu, Y.; Pang, S.-p. Nitrogen-rich Salts Based on Polyamino Substituted *N*,*N*′-azo-1,2,4-triazole: a New Family of High-performance Energetic Materials. *J. Mater. Chem. A* **2014**, *2*(38): 15978-15986.
[60] Xu, Z.; Cheng, G.; Yang, H.; Zhang, J.; Shreeve, J.M. Synthesis and Characterization of 4-(1,2,4-Triazole-5-yl)furazan Derivatives as High-Performance Insensitive Energetic Materials. *Chem. – Eur. J.* **2018**, *24*(41): 10488-10497.
[61] Tang, Y.; He, C.; Gao, H.; Shreeve, J.M. Energized Nitro-substituted Azoles through Ether Bridges. *J. Mater. Chem. A* **2015**, *3*(30): 15576-15582.
[62] He, C.; Shreeve, J.M. Energetic Materials with Promising Properties: Synthesis and Characterization of 4,4′-bis(5-Nitro-1,2,3-2*H*-triazole) Derivatives. *Angew. Chem., Int. Ed.* **2015**, *54*(21): 6260-6264.
[63] Yin, P.; Shreeve, J.M. From N-Nitro to N-Nitroamino: Preparation of High-Performance Energetic Materials by Introducing Nitrogen-Containing Ions. *Angew. Chem., Int. Ed.* **2015**, *54*(48): 14513-14517.
[64] Dippold, A.A.; Klapötke, T.M.; Oswald, M. Asymmetrically Substituted 5,5′-Bistriazoles–Nitrogen-rich Materials with Various Energetic Functionalities. *Dalton Trans.* **2013**, *42*(31): 11136-11145.
[65] Fendt, T.; Fischer, N.; Klapötke, T.M.; Stierstorfer, J. *N*-Rich Salts of 2-Methyl-5-nitraminotetrazole: Secondary Explosives with Low Sensitivities. *Inorg. Chem.* **2011**, *50*(4): 1447-1458.
[66] Tao, G.-H.; Twamley, B.; Shreeve, J.M. A Thermally Stable Nitrogen-rich Energetic Material – 3,4,5-Triamino-1-tetrazolyl-1,2,4-triazole (TATT). *J. Mater. Chem.* **2009**, *19*(32): 5850-5854.
[67] Klapötke, T.M.; Schmid, P.C.; Schnell, S.; Stierstorfer, J. 3,6,7-Triamino-[1,2,4] triazolo [4,3-b][1,2,4]triazole: a Non-toxic, High-Performance Energetic Building Block with Excellent Stability. *Chem. – Eur. J.* **2015**, *21*(25): 9219-9228.
[68] Stierstorfer, J.; Tarantik, K.R.; Klapötke, T.M. New Energetic Materials: Functionalized 1-Ethyl-5-aminotetrazoles and 1-Ethyl-5-nitriminotetrazoles. *Chem. – Eur. J.* **2009**, *15*(23): 5775-5792.
[69] Chand, D.; Parrish, D.A.; Shreeve, J.M. Di(1*H*-tetrazol-5-yl)methanone Oxime and 5,5′-(Hydrazonomethylene)bis(*1*H-tetrazole) and Their Salts: a Family of Highly Useful New Tetrazoles and Energetic Materials. *J. Mater. Chem. A* **2013**, *1*(48): 15383-15389.
[70] Dachs, M.; Dippold, A.A.; Gaar, J.; Holler, M.; Klapötke, T.M. A Comparative Study on Insensitive Energetic Derivatives of 5-(1,2,4-Triazol-*C*-yl)-tetrazoles and Their 1-Hydroxy-tetrazole Analogues. *Z. Anorg. Allg. Chem.* **2013**, *639*(12-13): 2171-2180.

[71] Bian, C.; Zhang, M.; Li, C.; Zhou, Z. 3-Nitro-1-(2*H*-tetrazol-5-yl)-1*H*-1,2,4-triazol-5-amine (HANTT) and Its Energetic Salts: Highly Thermally Stable Energetic Materials with Low Sensitivity. *J. Mater. Chem. A* **2015**, *3*(1): 163-169.
[72] Yin, P.; Zhang, Q.; Zhang, J.; Parrish, D.A.; Shreeve, J.M. *N*-Trinitroethylamino Functionalization of Nitroimidazoles: a New Strategy for High Performance Energetic Materials. *J. Mater. Chem. A* **2013**, *1*(25): 7500-7510.
[73] Yin, P.; Zhang, J.; He, C.; Parrish, D.A.; Shreeve, J.M. Polynitro-substituted Pyrazoles and Triazoles as Potential Energetic Materials and Oxidizers. *J. Mater. Chem. A* **2014**, *2*(9): 3200-3208.
[74] Klapötke, T.M.; Sabaté, C.M.; Rasp, M. Synthesis and Properties of 5-Nitrotetrazole Derivatives as New Energetic Materials. *J. Mater. Chem.* **2009**, *19*(15): 2240-2252.
[75] Klapötke, T.M.; Sabaté, C.M. Bistetrazoles: Nitrogen-rich, High-performing, Insensitive Energetic Compounds. *Chem. Mater.* **2008**, *20*(11): 3629-3637.
[76] He, C.; Yin, P.; Mitchell, L.A.; Parrish, D.A.; Shreeve, J.M. Energetic Aminated-azole Assemblies from Intramolecular and Intermolecular N–H···O and N–H···N Hydrogen Bonds. *Chem Commun.* **2016**, *52*(52): 8123-8126.
[77] Klapötke, T.M.; Mayer, P.; Stierstorfer, J.; Weigand, J.J. Bistetrazolylamines – Synthesis and Characterization. *J. Mater. Chem.* **2008**, *18*(43): 5248-5258.
[78] Huang, Y.; Gao, H.; Twamley, B.; Shreeve, J.M. Nitroamino Triazoles: Nitrogen-Rich Precursors of Stable Energetic Salts. *Eur. J. Inorg. Chem.* **2008**, *2008*(16): 2560-2568.
[79] Wang, B.; Qi, X.; Zhang, W.; Wang, K.; Li, W.; Zhang, Q. Synthesis of 1-(2*H*-Tetrazol-5-yl)-5-nitraminotetrazole and Its Derivatives from 5-Aminotetrazole and Cyanogen Azide: a Promising Strategy towards the Development of C–N Linked Bistetrazolate Energetic Materials. *J. Mater. Chem. A* **2017**, *5*(39): 20867-20873.
[80] Fischer, D.; Klapötke, T.M.; Stierstorfer, J. 1,5-Di(nitramino) Tetrazole: High Sensitivity and Superior Explosive Performance. *Angew. Chem., Int. Ed.* **2015**, *54*(35): 10299-10302.
[81] Stierstorfer, J.; Klapötke, T.M.; Hammerl, A.; Chapman, R.D. 5-Azido-1*H*-tetrazole–Improved Synthesis, Crystal Structure and Sensitivity Data. *Z. Anorg. Allg. Chem.* **2008**, *634*(6-7): 1051-1057.
[82] Göbel, M.; Karaghiosoff, K.; Klapötke, T.M.; Piercey, D.G.; Stierstorfer J. Nitrotetrazolate-2*N*-oxides and the Strategy of *N*-Oxide Introduction. *J. Am. Chem. Soc.* **2010**, *132*(48): 17216-17226.
[83] Palm, W.J. *Introduction to MATLAB for Engineers. 3rd* Ed. McGraw-Hill, New York, **2011**.
[84] Keshavarz, M.H.; Gharagheizi, F.; Shokrolahi, A.; Zakinejad, S. Accurate Prediction of the Toxicity of Benzoic Acid Compounds in Mice via Oral without using any Computer Codes. *J. Hazard. Mater.* **2012**, *237*: 79-101.
[85] Billo, E.J. *Excel for Chemists: a Comprehensive Guide.* 2nd *Ed.,* Wiley, New York, **2001**.
[86] Becke, A.D. Density-functional Thermochemistry. III. The Role of Exact Exchange.

*J. Chem. Phys.* **1993**, *98*(7): 5648-5652.

[87] Frisch, M.J.; Trucks, G.W.; Schlegel, H.B.; Scuseria, G.E.; Robb, M.A.; Cheeseman, J.R.; Scalmani, G.; Barone, V.; Mennucci, B.; Petersson, G.A.; Nakatsuji, H.; Caricato, M.; Li, X.; Hratchian, H.P.; Izmaylov, A.F.; Bloino, J.; Zheng, G.; Sonnenberg, J.L.; Hada, M.; Ehara, M.; Toyota, K.; Fukuda, R.; Hasegawa, J.; Ishida, M.; Nakajima, T.; Honda, Y.; Kitao, O.; Nakai, H.; Vreven, T.; Montgomery, J.A., Jr.; Peralta, J.E.; Ogliaro, F.; Bearpark, M.; Heyd, J.J.; Brothers, E.; Kudin, K.N.; Staroverov, V.N.; Kobayashi, R.; Normand, J.; Raghavachari, K.; Rendell, A.; Burant, J.C.; Iyengar, S.S.; Tomasi, J.; Cossi, M.; Rega, N.; Millam, J.M.; Klene, M.; Knox, J.E.; Cross, J.B.; Bakken, V.; Adamo, C.; Jaramillo, J.; Gomperts, R.; Stratmann, R.E.; Yazyev, O.; Austin, A.J.; Cammi, R.; Pomelli, C.; Ochterski, J.W.; Martin, R.L.; Morokuma, K.; Zakrzewski, V.G.; Voth, G.A.; Salvador, P.; Dannenberg, J.J.; Dapprich, S.; Daniels, A.D.; Farkas, O.; Foresman, J.B.; Ortiz, J.V.; Cioslowski, J.; Fox, D.J. *Gaussian 09, Revision A.1*. Gaussian, Inc., Wallingford, CT, **2009**.

[88] Politzer, P.; Martinez, J.; Murray, J.S.; Concha, M.C.; Toro-Labbe, A. An Electrostatic Interaction Correction for Improved Crystal Density Prediction. *Mol. Phys.* **2009**, *107*(19): 2095-2101.

[89] Willmott, C.J.; Matsuura, K.; Robeson, S.M. Ambiguities Inherent in Sums-of-squares-based Error Statistics. *Atmos. Environ.* **2009**, *43*(3): 749-752.

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