Qualitative estimation of the dependence of the number of energetic compounds on their oxygen coefficient

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The ratio between the numbers of structural formulas of C,H,N,O-containing energetic compounds belonging to the classes of fuels (low values of the oxygen coefficient A), explosives (medium A), and oxidants (high A values) was studied by a computer generation procedure. The number of the theoretically possible structural formulas was found to decrease rapidly on going from fuels to explosives and then to oxidants; this observation agrees with the data on the numbers of various energetic compounds currently used and proposed. The strategy of the search for new compounds with the specified properties is described in brief, and its applicability to the search for explosives and oxidants with a small (up to 12) number of atoms in a molecule is evaluated.

Key words: energetic compounds, oxygen coefficient, molecular formula, computer generation, structural constraints.

One of the key directions in theoretical organic chemistry is a target-oriented search for new compounds that possess desirable physicochemical properties.[1] During the last few decades, development of this direction was greatly promoted by improvement of computer equipment. New search schemes, computational methods, and software based on them are constantly being developed. In particular, this interest concerns a search for energetic compounds, substances with a high enthalpy of formation (combustion).

As to the field of their application, energetic compounds are traditionally divided to three groups: fuels, explosives, and oxidants. The search for new energetic compounds is going on within all the three groups. However, the efficiency of this search varies: new fuels, including even new classes of fuels, are proposed regularly, new explosives notably less frequently,[2,3] and new oxidants very rarely.[4] The same is true for the numbers of energetic compounds that are known and used in practice. The most evident explanation of this can be formulated as follows: perhaps the numbers of proposed new compounds correspond to the numbers of *theoretically possible* structures that belong to these three groups. However, to our knowledge of the theoretically possible numbers of fuels, explosives, and oxidants has never been estimated previously, although such an estimation can play an important role in predicting the comparative efficiency of the search for these energetic compounds.

The oxygen coefficient as a characteristic of energetic compounds

Although the specific properties of a compound (its enthalpy of formation, density, *etc.*) depend on its structural formula, molecular geometry, crystal packing, and other characteristics, in fact, an energetic compound can be classified with reasonable confidence as a fuel, explosive, or oxidant based on its molecular formula.[5]

Let us consider the molecular formula $C_aH_bN_cO_d$ (a, b, c, $d \geq 1$). It can be characterized by its oxygen coefficient (oxygen balance) A, which is defined[6] as

$$A = d/(2a + b/2). \quad (1)$$

It is known[7] that free carbon can be present in the products of thermal decomposition of a substance $C_aH_bN_cO_d$ at $d < a + b/2$; at $d < a$, free carbon is always present. To find the A values corresponding to the latter condition, let us assume $d = a$ and transform formula (1) as

$$A = d/(2a + b/2) = a/(2a + b/2) = 1/(2 + b/2a). \quad (2)$$

The majority of known energetic substances satisfy the condition $0 < b < 2a$. The "average" value $b = a$ corresponds to $A = 0.4$; for $b \ll a$, one obtain $A = 0.5$. Thus, the compound in question ($C_aH_bN_cO_d$) forms free carbon during its thermal decomposition if it belongs to

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the $0 \le A < 0.4$—0.5 range; so, this A range can be referred to as the range of fuels. For simplicity of discussion, hereafter let us set $A = 0.4$ as the upper limit of this range.

At $d > 2a + b/2$ (that is, at $A > 1.0$), the amount of oxygen in a C, H, N, O-containing compound exceeds that necessary for complete oxidation of carbon and hydrogen to CO_2 and H_2O, respectively. Therefore, the $A > 1.0$ range can be referred to as the range of oxidants.

Finally, it is known[8] that the majority of widely used high explosives, as well as many compounds that have ever been used or proposed in this field, belong to the $A = 0.4$—1.0 range. Therefore, this range can be referred to as the range of explosives. (These boundaries of the range of explosives should not be regarded as exact: in fact, this range partially overlaps the ranges of fuels and oxidants. For example, $A = 0.39$ for trinitrotoluene and 1.06 for glycerol trinitrate. Hereafter, we will not take this slight inaccuracy into account.)

Thus, the value of the oxygen coefficient is a characteristic of an energetic compound that may be used for preliminary determination of its field of application as a fuel (A up to 0.4), high explosive (A from 0.4 to 1.0), or oxidant (A above 1.0). In the search for new energetic compounds with a specified field of application, it is enough to consider only compounds whose molecular formulas belong to the relevant range of oxygen coefficients.[9]

The numbers of fuels, explosives, and oxidants are compared in this work on the same basis. Using computer generation procedures, we made an attempt to estimate the ratio between the theoretically possible numbers of C, H, N, O-containing molecules with $A < 0.4$, $0.4 \le A \le 1.0$, and $A > 1.0$. We confined ourselves to small molecules (from 8 to 12 atoms, including hydrogens), because the number of resultant structures exponentially increases with increasing number of atoms in molecules and quickly becomes too large for exhaustive generation and analysis.

The use of generation in search for compounds with desired properties

The principal stages of the computer search for compounds with specified properties can be described in the following way. Firstly, on the basis of some objective and subjective considerations, a researcher determines the specific features that characterize the composition and structure of the target molecule; that is, a list of structural constraints is formed. The next stage is the generation of structural formulas that satisfy the constraints previously formulated. Finally, the required properties are theoretically estimated for substances that correspond to the generated structures; according to this estimate, the most promising compounds are selected.

The molecular formula of the target substance is often used as the main constraint during generation.[9] Its frequent use is due to the fact that a number of physicochemical properties of organic compounds are determined to some extent by the molecular formula. For example, many methods for theoretical calculation of density,[10] enthalpy of formation,[11] or sublimation enthalpy[12] of organic substances are based on explicit considerations of the molecular formula (of course, along with other molecular characteristics). Another important fact is that generation techniques based on the molecular formula are the most numerous and efficient among methods used at present. Therefore, if some other characteristics of the composition (the number of atoms and the oxygen coefficient in our case) are used as the starting information for search, all the molecular formulas corresponding to them are enumerated at first, and then generation is conducted separately for each of these formulas.

However, the use of the molecular formula alone as the initial information for generation is not sufficient in the majority of cases of practical significance. As mentioned above, the number of structural formulas that correspond to one molecular formula exponentially increases as the number of non-hydrogen atoms in the target molecules increases (for example, see Polya's formula[13]). Detailed analysis of all these molecules is impossible, but it is unnecessary because the overwhelming majority of these structural formulas either contain some fragments whose existence is improbable from the chemical standpoint (highly stressed structures, unstable tautomers, *etc.*) or are of no interest in the search for promising compounds with the specified properties for some other reason.

Thus, the problem is resolved by the use in the generation procedure of additional constraints and filters along with the molecular formula. Thus, on the basis of some criteria, the majority of molecules can be automatically excluded from consideration, and only the most promising ones will remain.

One of the additional constraints frequently used in the generation procedure is the list of undesired structural fragments in the target molecules (the list of forbidden fragments, BADLIST).[14] This list usually includes fragments that reduce the stability of the molecules or adversely affect the target properties of the corresponding substances in another way. Another additional constraint that can be used is the list of fragments that, on the contrary, should be present in target structures (the list of required fragments, GOODLIST).[14] All molecules that do not contain fragments from GOODLIST or contain at least one fragment from BADLIST must be automatically excluded from consideration. Varying GOODLIST and BADLIST, the user can adapt the structure-generating program to solve specific problems.

Calculation procedure

We studied molecular formulas of the general type $C_aH_bN_cO_d$ (a, b, c, $d \ge 1$) with the total number of

Table 1. Dependence of the number of all possible and nonempty compositions and PMFs on the number of atoms

Number of atoms	Number of all possible compositions (PMFs)	Number of nonempty compositions (PMFs)
1	35 (81)	19 (43)
9	56 (136)	28 (68)
10	84 (214)	43 (108)
11	120 (320)	59 (158)
12	165 (460)	82 (228)

atoms in a molecule $n = a + b + c + d$ varying from 8 to 12. The number of such molecular formulas for a specified n is calculated[15] as follows

$$K(n) = C^3_{n-1} = (n-1)!/3!(n-4)! \quad (3)$$

Structural generation without additional constraints was conducted for each molecular formula; as a result, all structural formulas of electrically neutral molecules corresponding to the original molecular formula were determined and their total number was found. The oxygen coefficient A for each molecular formula was determined according to Eq. (1) and rounded with an accuracy up to 0.01. The total number of structures for each rounded A value was found by summation over all the corresponding molecular formulas.

The generation was carried out with the use of a computer program that implements Molodtsov's modification[16] of Faradjev's graph-theoretical algorithm.[17] This program finds all structural isomers that correspond to a specific molecular formula if the valences of the chemical elements that are present in this molecular formula are known and constant. It is reasonable to consider $V(C) = 4$, $V(H) = 1$, $V(N) = 3$, and $V(O) = 2$ as such constant valences. However, this way we exclude a very large number of energetic compounds from consideration. Indeed, structural formulas of molecules containing, for example, a nitro group cannot be constructed with the use of strictly trivalent nitrogen. So, to consider nitro and azoxy compounds, amine oxides, *etc.*, we introduced an additional trivalent "macroatom" Z, which corresponded to the $N^+—O^-$ group (with a semipolar bond).* For example, generation for the molecular formula $C_2H_2N_2O_2$ included three steps: the first step corresponded to the "partial" molecular formula (PMF) $C_2H_2Z_2$ (each of the generated molecules contains two $N^+—O^-$ groups, that is, two macroatoms of the Z type); the second step corresponds to the PMF $C_2H_2Z_1N_1O_1$ (the generated molecules contain one $N^+—O^-$ group, one trivalent nitrogen atom, and one divalent oxygen atom); the third step corresponds to the formula $C_2H_2N_2O_2$ containing trivalent nitrogen and divalent oxygen (the PMF is identical to the molecular formula). The number of structures corresponding to each molecular formula was calculated by summation over the numbers of isomers generated for each of the corresponding PMFs.

The dependence of the total number of possible molecular formulas and PMFs on the n value is shown in Table 1. Those molecular formulas for which the generation of any covalently bound structures with correct atomic valences is *a priori* impossible (for example, $C_3H_3N_2O_2$ or $C_1H_9N_1O_1$) were excluded from further consideration. Hereafter, such molecular formulas are referred to as *empty compositions*, and, accordingly, molecular formulas for which at least one isomer was generated are *nonempty compositions*. Empty and nonempty PMFs can be defined in a similar way. The dependences of the numbers of nonempty compositions and PMFs on the total number of atoms in a molecule are also shown in Table 1.

At the second stage, the generation with additional constraints was conducted out for all nonempty compositions. For this purpose, we used the SMOG program developed by one of the authors.[18] This program (which also uses elements of Faradjev's aforementioned algorithm[17]) is specially adapted to solution of chemical problems and can efficiently make allowance for a wide diversity of structural constraints during generation (including required, forbidden, and allowed fragments; local environments of atoms; their hybridization and formal charges; aromaticity and nonaromaticity of cycles; molecular topology; *etc.*).

To avoid generation of most molecules that are of no interest as energetic compounds, we included the following substructures into BADLIST:

— stressed or highly stressed fragments: cycles with triple bonds, anti-Bredt structures (with a double bond at the bridgehead), polycyclic structures with the pyramidal geometry of a carbon atom (some propellanes and paddlanes), small (three- or four-membered) cycles with multiple bonds, fused small cycles, spiroconnected three-atom cycles, non-planar (from the graph-theoretical standpoint[19,20]) fragments,** *etc.*;

— unstable tautomers (enols, inols, and their nitrogen-containing analogs; primary nitrosoamines, *etc.*), as well as some compounds that are easily decomposed (gem-diols, hemiacetals/hemiketals, *etc.*);

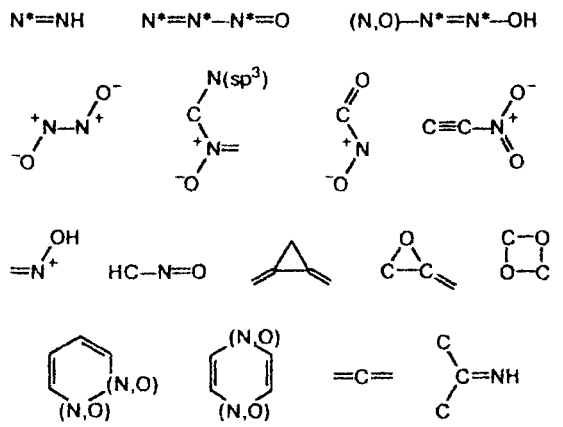
— peroxides and hydroperoxides (because of their high sensitivity to mechanical impact, they are normally used only as initiators rather than fuels or high explosives; azides were earlier excluded from consideration for the same reason);

* A similar representation of structures with semipolar bonds can be also used for generation of other classes of energetic substances, for example, azides. However, the latter compounds were not considered in this study, because they have a limited field of application due to their low stability.

** A molecular structure is regarded as a nonplanar one from the graph-theoretical standpoint if it is impossible to draw this structure on a plane, without breaking any bonds between atoms, so that bonds would not intersect and atoms would not be superimposed. Many nonplanar caged molecules have very high stress energy.

— unstable heterocycles (cycles consisting only of heteroatoms; three- or four-membered cycles containing only one carbon atom) and some chains of heteroatoms (more than four heteroatoms in succession or more than two heteroatoms in succession without conjugation or charge alternation);

— other groups (some of them are shown in Fig. 1) that reduce the stability of molecules or are very rarely encountered in known energetic compounds*.



Here (N,O) is the N atom or the O atom; N* is the N atom or the N^+—O^- group; (sp^3) is the hybridization state of the atom. If the atomic type is not explicitly specified for some atoms in a fragment, then these are arbitrary non-hydrogen atoms.

Among compounds with the N^+—O^- group, we considered only those where the nitrogen atom in this group is in the state of sp^2-hybridization (nitro compounds, azoxy compounds, oxides of aromatic nitrogen bases, *etc.*) or in the state of sp^3-hybridization and bound to three carbon atoms (oxides of tertiary amines).

The generation with the use of this set of additional constraints was carried out for all molecular formulas with $n = 8—11$.

* Strictly speaking, the choice of fragments that are to be included into BADLIST is arbitrary and subjective to a large extent. For example, although allenes are among the forbidden fragments in Fig. 1, we cannot fully exclude the possibility of the synthesis of novel energetic compounds with the allene-type carbon atoms: the decision to include allenes in BADLIST was based only on our analysis of energetic compounds currently used, because allenes are extremely rare among them. Another example is the inclusion of the enol and hemiacetal groups in BADLIST: this can be regarded as debatable, because compounds with a stable enol form (acetylacetone, *etc.*) may actually exist, as well as stable hemiacetals. However, such a subjectivity of choice is permissible in the solution of our current problem, because we are presently attempting to establish qualitative relationships rather than exact quantitative results.

Results and Discussion

The dependence of the total number of structural formulas (that is, the number of structures generated without additional constraints) on the oxygen coefficient A at specified n values ($n = 8—12$) is shown in Fig. 2. Let us compare these bar charts with the above division into three ranges: the range of fuels ($A < 0.4$), which includes the majority of known energetic compounds; the range of explosives ($0.4 \le A \le 1.0$) with a smaller number of compounds; and the range of oxidants ($A > 1.0$), where only a few substances are known. Apparently, these three ranges in Fig. 2 indeed differ greatly as to the number of structures in them: 70—80% of all generated molecules lie in the range of fuels (and this fraction increases with increasing n); 10—20% are in the range of explosives; and only 1—10% are oxidants. Thus, our data concerning the numbers of theoretically possible structures match the actual data concerning the relative diversity of energetic compounds (although, as was noted above, usually only a small fraction of structures that are generated without additional constraints may be actually synthesized).

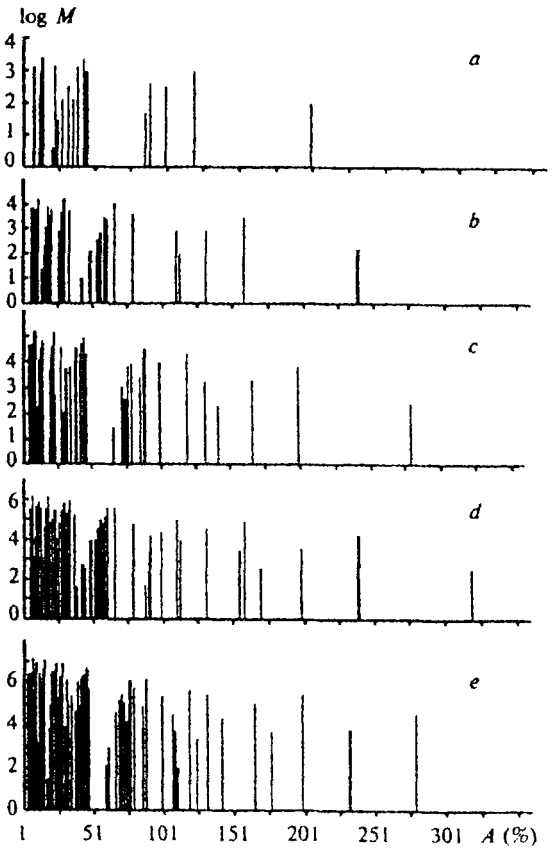


Fig. 1. Dependences of the total number of structural formulas (M) on the oxygen coefficient (A) for a constant number of atoms in a molecule: $n = 8$ (*a*), 9 (*b*), 10 (*c*), 11 (*d*), and 12 (*e*).

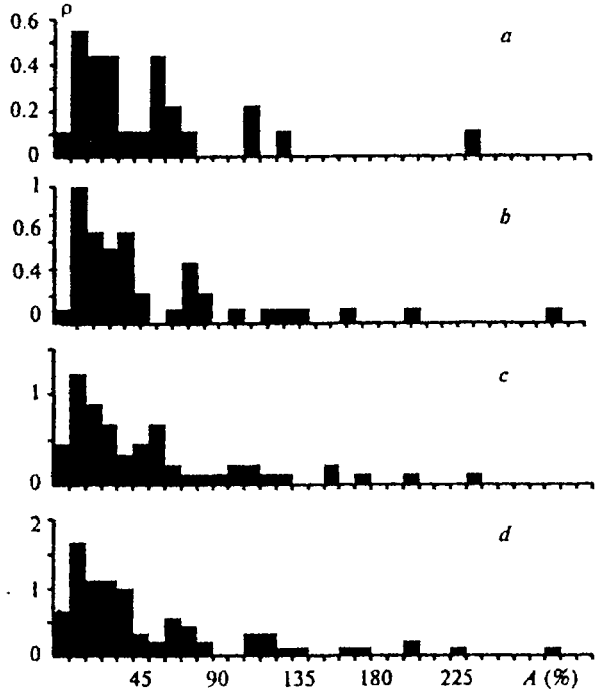


Fig. 2. Dependences of the density of nonempty compositions (ρ) on the oxygen coefficient (A) for a constant number of atoms in a molecule: n = 9 (*a*), 10 (*b*), 11 (*c*), and 12 (*d*).

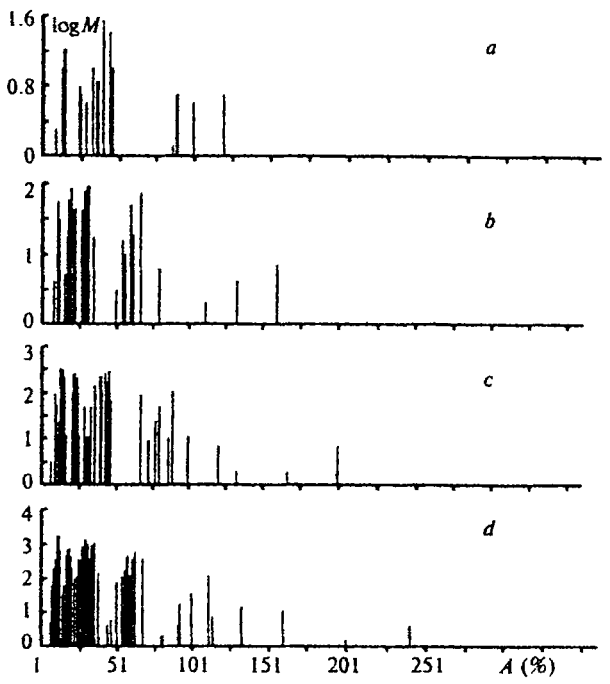


Fig. 3. Dependences of the number of structures (M) obtained by generation with additional constraints on the oxygen coefficient (A) for a constant number of atoms in a molecule: n = 8 (*a*), 9 (*b*), 10 (*c*), and 11 (*d*).

From the qualitative standpoint, such a decrease in the number of theoretically possible structures as a result of an increase in the A value could be predicted from simple mathematical considerations. Indeed, $V(C) = 4 > V(N) = 3 > V(O) = 2$. But, as a rule, the higher the valences of atoms in a molecular formula (that is, the larger the maximum number of chemical bonds that may connect these atoms), the larger the number of different structures that can be formed from them, because of the possible existence of branched chains, various polycyclic systems, *etc.* Therefore, molecules that are "rich in carbon" must be more numerous than molecules that are "rich in oxygen" but have the same total number of atoms. For example, our calculations (Fig. 2) show that the average number of isomers per one nonempty composition with $n = 10$ approximately equals $2.5 \cdot 10^4$ in the range of fuels, $1.4 \cdot 10^4$ in the range of explosives, and $5 \cdot 10^3$ in the range of oxidants. Apparently, the ratio between the corresponding numbers will not change significantly for other n values including $n > 12$.

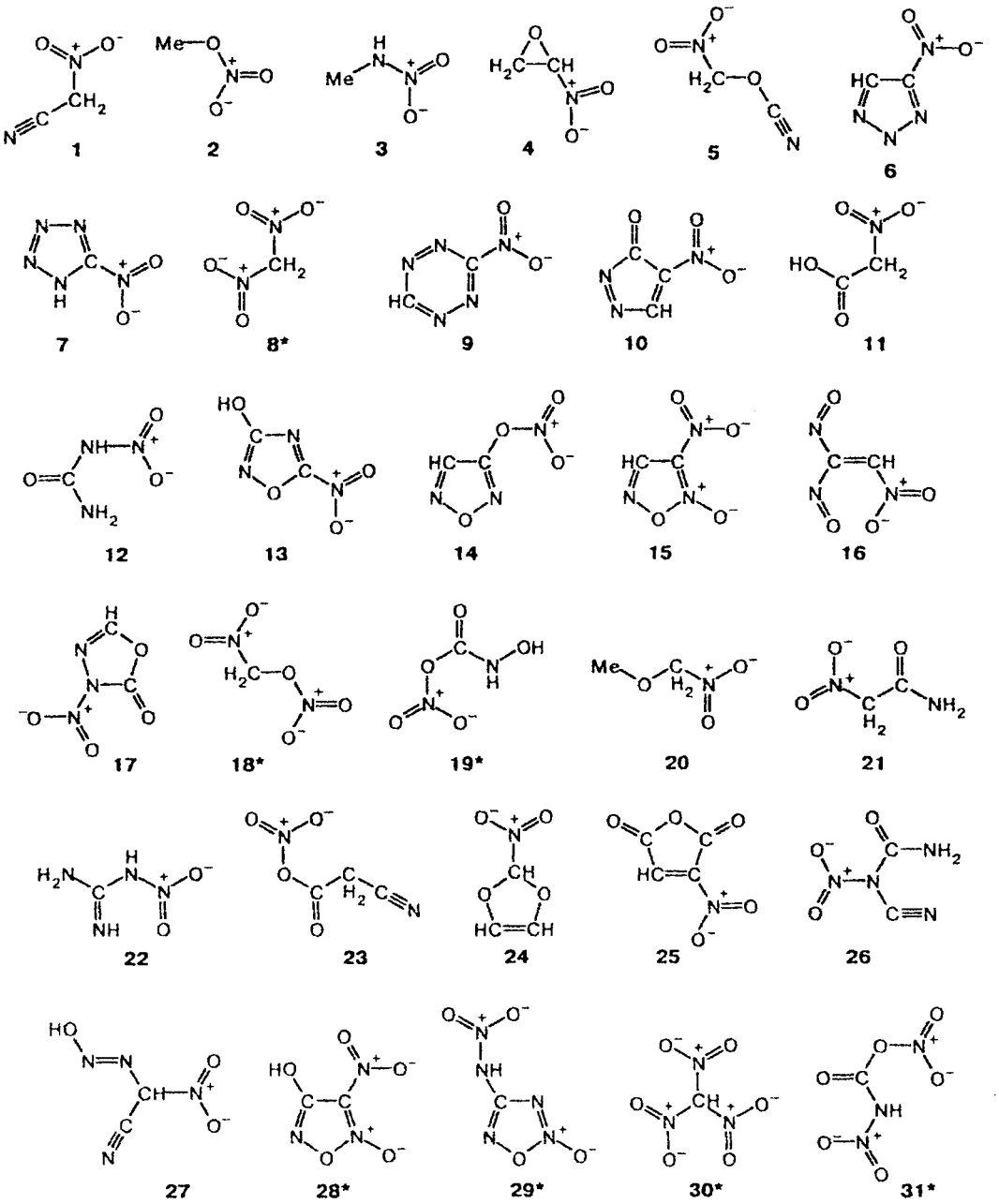
However, the fact that structures in the range of fuels are much more numerous than those in the ranges of explosives and oxidants is explained not only by the number of isomers per one nonempty composition but primarily by the number of these nonempty compositions themselves in the specific range of A values. In other words, not only the average height but also the average "density" of columns in the bar charts in Fig. 2 decrease with increasing A.

The number of nonempty compositions $N(n, A, \Delta A)$ within some interval $[A, A + \Delta A]$ is defined as the number of n-atomic molecular formulas (in this interval) for which at least one structure was obtained as a result of generation without additional constraints. The number of nonempty compositions within some interval divided by the width of this interval is hereafter referred to as the density of nonempty compositions at this interval:

$$\rho(n, A, \Delta A) = N(n, A, \Delta A)/\Delta A. \quad (4)$$

In fact, this value characterizes the density of vertical columns in Fig. 2. The dependence of the density of nonempty compositions on A (n = 9—12, $\Delta A = 0.09$) in Fig. 3 displays an obvious tendency to a decrease in the ρ value with increasing A: in the range of fuels, at least one structural formula lies within each $[A, A + \Delta A]$ interval with the selected width $\Delta A = 0.09$; in the range of explosives, this is not true for some intervals; finally, in the range of oxidants, there are but few intervals that contain any structural formulas. Moreover, the density of nonempty compositions tends to increase with increasing n, but this tendency almost totally disappears in the range of oxidants. (Strictly speaking, there are no exact boundaries between the ranges that differ by the density of nonempty compositions in Figs. 2 and 3. It

Scheme 1



1, **2** (n = 8); **3**—**8** (n = 9); **9**—**19** (n = 10); **20**—**31** (n = 11). Oxidants (A > 1.0) are marked with asterisks.

would be more accurate to say that there are transition intervals: for example, A = 0.35—0.5 between the ranges with high and medium density and A = 0.8—1.1 between the ranges with medium and low density of nonempty compositions.)

At the second stage of our work, when we performed generation using additional constraints (that is, using the above-mentioned list of forbidden fragments), the number of structures obtained for each nonempty composition decreased, on average, by two or three orders of

magnitude compared to the generation on the basis of only molecular formula (*cf.* Figs. 4 and 2). However, the ratio between the numbers of fuels, explosives, and oxidants remained the same from the qualitative standpoint: slightly less than 80% of all the molecules generated for the n values in question lie in the range of fuels; approximately 20% belong to the range of explosives; and only 1.5% are oxidants.

Let us discuss the last two ranges in more detail. Generation with the above-mentioned additional constraints yields a total of more than 3500 compounds in the range of explosives and approximately 200 oxidants for all molecular formulas with $n = 8$—11. However, these numbers can be reduced if other fragments that are not typical of known explosives and oxidants are added to BADLIST: C≡C bonds, aldehyde groups, α-lactam cycles, *etc.* Moreover, it is known that a compound, to be applicable as an explosive, must contain so-called explosophoric groups in its molecule. For high explosives, the most common among such groups is a nitro group bound to a C, N, or O atom (other explosophoric fragments are also known, but they are notably rarer). Nitro groups are also present in almost all known C, H, N, O-containing organic oxidants. Therefore, we can include the nitro group into GOODLIST for subsequent generation of explosives and oxidants.

As a result of such generation for all molecular formulas with $n = 8$—11 and $A \geq 0.4$, we obtained about 500 molecules of explosives and oxidants; the fraction of the latter was less than one-quarter. For such a number of compounds, sufficiently detailed expert analysis of each separate structure (as to the chemical stability, synthetic feasibility, and hypothetical practical applicability of the corresponding substance) becomes possible. Some of the molecules obtained as a result of this generation are shown in Fig. 5. Apparently, there are some well-known energetic compounds among them: methyl nitrate **2**, methylnitramine **3**, 5-nitro-1*H*-tetrazole **7** (an initiator), nitrourea **12**, nitroguanidine **22**, trinitromethane **30**, *etc.* At the same time, based on the data available, some structures in this figure have never been proposed as energetic compounds before (molecules **5**, **9**, **19**, **24**, **26**, **29**, *etc.*). A quantum chemical calculation of their geometry and enthalpy of formation is the subject of a separate study.

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Let us sum up the results of this study. Using a generation method without additional constraints, we showed that the number of theoretically possible compounds in the range of fuels is much greater than that of the structures in the range of explosives; the latter, in turn, exceeds the number of oxidants (see Fig. 2). This result is due both to the decreasing density of nonempty compositions in the fuels—explosives—oxidants sequence (see Fig. 3) and the decreasing average number of isomers that can be constructed for one molecular formula. The above-mentioned qualitative relationship between the numbers of generated fuels, explosives, and oxidants remains the same in the case of generation with the use of the above-mentioned additional constraints (see Fig. 4) and agrees with the data on the ratio between the numbers of known or virtually proposed energetic compounds (see the Introduction).

Our method for the generation of compounds that conform to the specified molecular formula and satisfy a certain set of additional structural constraints can be used almost without changes (probably only with some variations of GOODLIST and BADLIST, as well as with the addition of detailed criteria for estimation of molecular stability) for finding new oxidants, especially with a small number of atoms in the molecule. Since the compounds thus generated are not very numerous, fairly detailed expert analysis of each structure is possible. The search for explosives, whose number is several times greater than that of oxidants, should better be conducted with the use of stricter selection criteria,[9] especially for $n > 12$. Finally, when we search for new fuels, the number of generated structural formulas increases very significantly. Therefore, to obtain a moderate number of structures, we should either use much stricter constraints (for example, confine our search to a given class of compounds) or change the very approach to the solution of the problem — for example, to use stochastic methods instead of exhaustive enumeration of all structures that satisfy the given constraints.[21]

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