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RING-TRANSFORMATION GRAPHS AND THEIR APPLICATION TO DEGENERATE HETEROCYCLIC REARRANGEMENTS

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Abstract

A special type of reaction graphs – the ring-bond-redistribution graphs – is used as a new model for description, classification and enumeration of ring transformation reactions of heterocycles, particularly of the class of degenerated heterocyclic rearrangements. Combinatorial formulae for enumeration of all the possible degenerated rearrangements for heterocycles with only one heteroatom are proposed.

1. Introduction

Heterocycles form one of the most important and well-investigated classes of organic molecules due to their occurrence in the human body and wide spectra of biological activity. One characteristic of the chemistry of heterocycles consists in the reactions of ring transformation or recyclization, i.e. easy opening of the starting heterocyclic ring and easy closure into a new (or the same) one. This ability of heterocyclic compounds was discovered more than a century ago and caused the now highly developed and extensively reviewed branch of organic chemistry [1–10]. Because this type of reaction is independent of the nature and size of the heterocyclic ring, it is a useful tool in planning synthesis of target organic compounds. Nevertheless, the general picture of heterocyclic ring transformations is rather complicated: they are poorly classified, and there is still not any appropriate mathematical model capable of rationalizing this interesting and beautiful class of organic reactions. It seems that the first (and unique) attempt to apply the methodology of graph theory to this subject was an approach by Balaban [5,7], where labelled subgraphs of the pyrillium ring (incorporated into a new heterocyclic nucleus) have been used to classify pyrillium ring transformations.

Different types of mathematical models have been suggested to describe and classify chemical reactions; some of them use ideas of graph theory, for instance, to analyze the redistribution of bonds via chemical reaction [11–15]. However, tautomerism of heterocycles (especially typical for substituted heteroaromatics) makes difficult the general use of such formal models (e.g. graphs of bond redistribution) for the general description of heterocyclic rearrangements and ring transformations (see the discussion in ref. [16]). In our early review on ring transformations of azoles, we had an idea for a new type of reaction graphs for the needs of heterocyclic chemistry [16,17]. This type of graphs, which neglect tautomerism – *the ring-bond-redistribution graphs* – has been proposed to formalize and classify the known recyclizations of 5-membered heterocycles and to predict some unknown sorts of azole rearrangements. The present article is devoted to the further development of this approach: a brief analysis of some combinatorial properties and relations for the ring-bond-redistribution graphs and an application of the approach to enumeration of degenerated heterocyclic rearrangements.

2. Definitions

We shall use the term *heterocyclic ring transformation* (HRT) for any reaction containing the steps of heterocyclic ring opening and ring closure in any sequence; the only requirement is that at least one (or more) atoms of the starting heterocyclic ring should be incorporated as a fragment in the final ring. HRT can contain any number of starting reagents and any number of final products.

We call the HRT to be *simple* (SHRT) if

(a) only one ring of the starting heterocycle is transformed into only one ring of the final molecule,
(b) there is no transient formation of cycles except the formation of the final cycle, and
(c) there are no transient permutations of the atoms belonging either to the starting or to the final heterocyclic rings.

A lot of known molecular HRTs are SHRTs; this is also true for most of the known heterocyclic rearrangements (HCRs), particularly for such well-investigated "named" reactions as the Dimroth, Cornforth, and Boulton–Katritzky HCRs for azoles and azines [1,2,9,10]; the reactions in figs. 1 and 2 are also SHRTs*. To avoid confusion with the fused heterocycles, let us consider their HRTs to be SHRTs if fused rings have only one pair of common atoms (e.g. as is usual for benzannulation in indole, quinoline, etc.), and if the presence of fused rings does not violate conditions (a)–(c).

Let us now consider any SHRT. The atoms of the final structure should be numbered in accordance with the numbers of the starting structure and the mechanism of HRT. Two types of graphs – *molecular graph* and *ring-bond-redistribution graph of SHRT* – are determined.

A molecular graph (M graph) of SHRT is determined for starting reagents (symbol s) and final products (symbol f) as the pairs $M_s(V, R_s)$ and $M_f(V, R_f)$, where the vertices V and the edges R of the M graph correspond only to the atoms and bonds which are present *either in the starting or in the final cycle.* The symbols

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*Reactions (1), (3)–(7) are taken from reviews [1,3], reaction (2) from ref. [19].

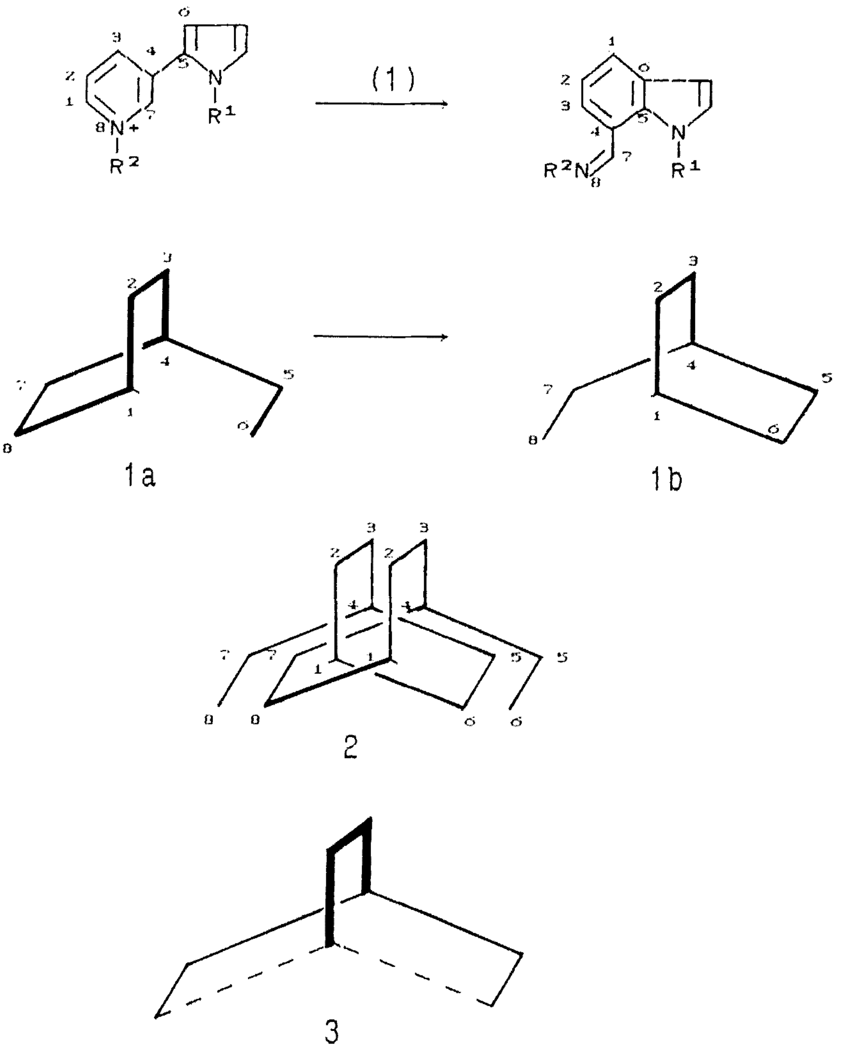


Fig. 1.

of heteroatoms, as well as hydrogen atoms, multiple bonds, substituents and any fused rings should be omitted in the M-graph structure. Examples of M graphs 1a and 1b for SHRT (1) are shown in fig. 1. M graphs are obviously monocyclic graphs; both M_s and M_f graphs contain an equal number of vertices but differ by the number and/or distribution of edges. For SHRTs that are usual intramolecular HCRs, M graphs are connected. Non-connected M graphs correspond to intermolecular SHRT only if the part of the starting (final) cycle is eliminated (entered) via reaction.

This pair of molecular M_s and M_f graphs is used to construct the graph of the SHRT-reaction – *the ring-bond-redistribution graph* (*RBR-graph*) or G_1 graph: M_s

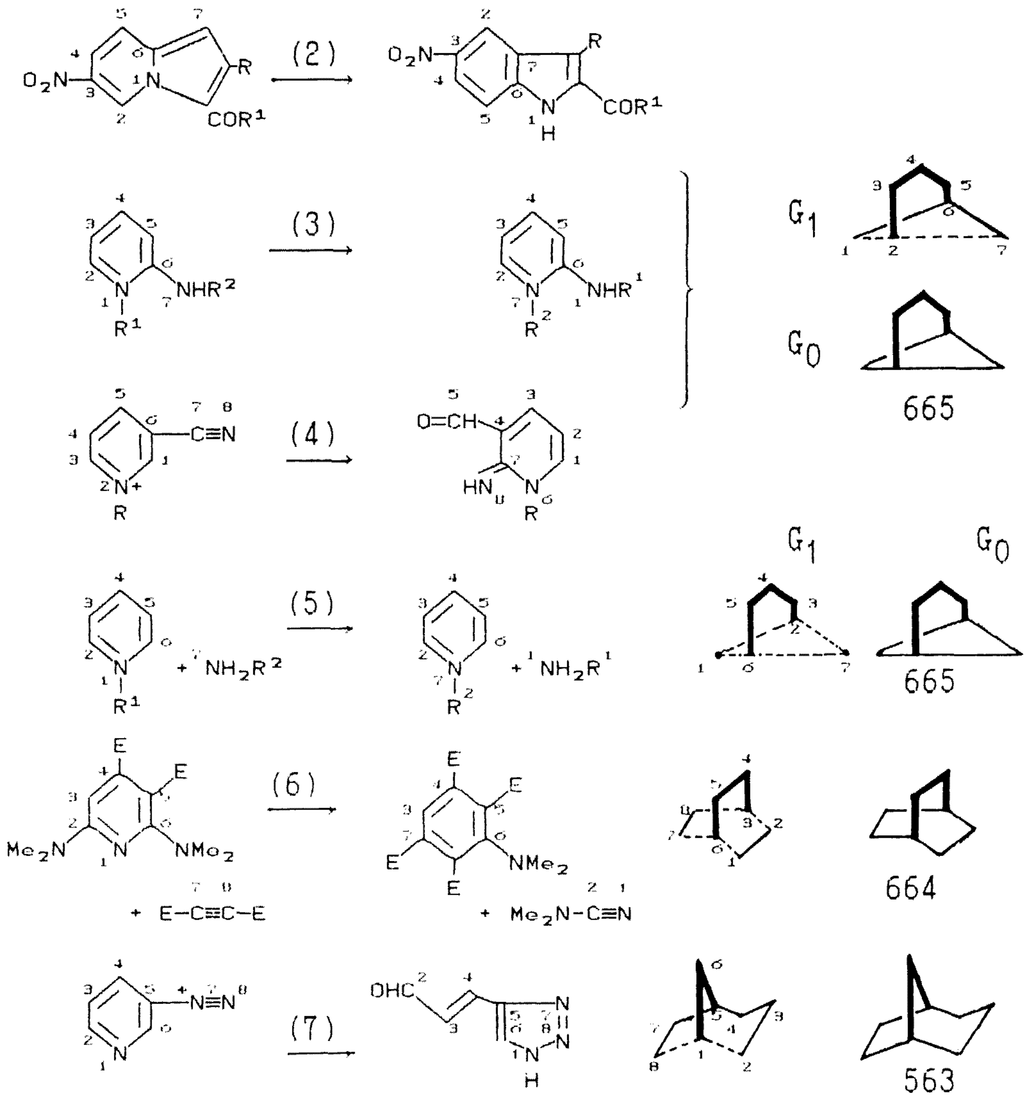


Fig. 2.

and M_f graphs of the starting and final structures are "superposed" according to the matching vertices with identical numbers. The resulting G_1 graph of reaction contains the same number of vertices (as in M graphs) and edges of different types, designated by

- *solid* lines, if the edge is present in both M_s and M_f graphs,
- *dashed* lines, if the edge is present only in one M graph, and
- *boldface* lines, if the edge belongs to both cycles of M graphs.

The necessity of each type of labels in the G_1 graph is evoked by a reasonable requirement to present maximal information on the appearance (disappearance) or conservation of any skeletal ring bond of the final (starting) cycle at the same structure of a reaction graph. An example of the superposition 2 of M graphs 1a and 1b and the resulting G_1 graph 3 for rearrangement (1) are shown in fig. 1. Followig the definition, RBR graphs of direct and reverse reactions coincide.

3. General properties and utilization of RBR graphs

The first important property can be formulated as:

THEOREM

An RBR graph G_1 for any SHRT is bicyclic; its structure corresponds to a pair of cycles with at least one common vertex.

The proof of this theorem simply follows our definitions of the *simple* HRT, M and RBR graphs. In fact, both M_s and M_f graphs contain only one cycle; any vertex and any edge of M_s, M_f or G_1 graphs belongs either to the starting cycle or to the final cycle. All these vertices and edges are present in the structure of the RBR graph, so it cannot contain more or less than two cycles. There are also no vertices or edges which do not belong to one of the two cycles, so the structure of the RBR graph can be only the pair of cycles with at least one common vertex.

In general, one can find three cycles in the structure of RBR graphs (e.g. the consequences 1234781, 1234561, and 1874561 for the G_1 graph 3 in fig. 1); however, only two cycles are linearly independent for the graphs with cyclomatic number 2 [20]. To avoid confusion, we select the cycle to be independent only *if it contains boldface-labelled edges*, and below use the term *cycle* of RBR graphs only in this sense.

Because each *cycle* of the bicyclic RBR graph has its ancestor, the cycle of the M_s or M_f graph, the pair of numbered molecular M graphs is in one-to-one correspondence with the G_1 graph. It is easy to restore the pair of M graphs from the given graph G_1 (operation reverse to superposition in fig. 1): let us choose any of the *cycles* from the G_1 graph; the dashed line(s) of only this *cycle* should be changed back to solid one(s), while the dashed line(s) of another *cycle* should be removed. This operation restores one of the M graphs. Now the same procedure should be repeated starting from the second *cycle* of the G_1 graph to restore another M graph. The retaining boldface labels should also be removed. In this manner, the G_1 graph gives rise only to the pair of numbered M graphs, but not to the direction of reaction presented by these graphs.

Let us define two different types of reaction RBR graphs [16]: the first one, G_1 graph; another type of RBR graph, G_0 graph. It can be constructed from the G_1 graph by changing all the dashed-marked edges to solid ones, i.e. it is a bicyclic graph with only two sorts of edges (solid and boldface). Corresponding examples of the G_0 and G_1 graphs for some known pyridine SHRTs (2)–(7) are shown in fig. 2.

These two constructions of G_0 and G_1 graphs permit us to pose and solve the following series of problems, which seem interesting both for mathematical and heterocyclic chemistry:

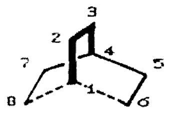
(1) *general classification and codification of SHRTs,*
(2) *enumeration of the possible types and classes of SHRTs,*
(3) *problem of prediction of the unknown SHRTs.*

All these problems are in correlation with each other; some concrete examples of how to solve them for particular cases of azole monocyclic HCRs have been discussed earlier in a review [16]. Let us briefly illustrate the use of our methodology, its generalization, and some important combinatorial results on the examples of pyridine SHRTs.

Types and classes of SHRTs. The construction of the G_0 graph can be used to reveal the degree of topological similarity of different SHRTs. Let us say the reactions belong to the same *type* if their G_0 graphs coincide, and to the same *class* when their G_1 graphs are identical. As one can see, there are three different types of pyridine SHRTs, namely, eqs. (2)–(5), eqs. (1) and (6), and eq. (7). Inside the type, reactions can differ by class: HCRs (2) (examined earlier by one of us [19]) and (4) are of the same class as the Dimroth rearrangement (3) but differ from the class of SHRT (4); classes of SHRTs (1) and (6) are also different. This clear structural classification of SHRTs is in contrast with traditional (and some vague) utilization of the "named" reactions; for instance, SHRTs (1) and (4), which differ either by G_0 or by G_1 graphs, were earlier related to the same reaction type [3].

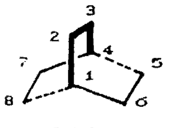
Codes for SHRT types. The simple bicyclic structure of G_0 graphs permits us to define a simple code for any *type* of SHRT. Let K and L be the size of cycles for a pair of M graphs, and N be the size of their common ring fragment ($1 \leq N \leq K$, $K \leq L$). Then the structure of the G_0 graph can be considered as a pair of annulated K- and L-membered *cycles* with an N-membered boldface-labelled bridge. The vector (K, L, N) is in one-to-one correspondence with the certain G_0 graph and can be used to codify the *type* of SHRT. The codes KLN for G_0 graphs of reactions (2)–(7) are shown in fig. 2.

Enumeration of SHRT classes. Comparing the structures of G_1 and G_0 graphs, one can conclude that the G_1 graph is the G_0 graph with labelled edges; one can easily obtain the possible G_1 graphs from an ancestor G_0 graph by changing some of its edges (designated as solid lines) to dashed ones. Since dashed edges correspond to the formed and broken ring bonds, in the particular case of HCRs only two edges of different *cycles* of the G_0 graph should be labelled. (The boldface-marked edges of the bridge should not be labelled, since they correspond to the unchanged ring bonds in SHRT.) All inequivalent manners of this labelling enumerate the number of different *classes* of rearrangements in the given *type*; an example is shown in fig. 3 for the particular G_0 graph with the code 664. The idea of the described enumeration of G_1 graphs is realized in our computer program [18] for any G_0 graphs with the number of dashed labels from 2 to 4; simple combinatorial equations for enumeration of G_1 graphs have also been suggested for rearrangements [16].



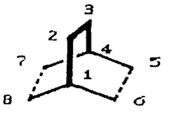
664-aa

Γ_1: (ε,α) $\varepsilon = (1)(2)(3)(4)(5)(6)(7)(8)$;
$\alpha = (1)(2)(3)(4)(57)(68)$



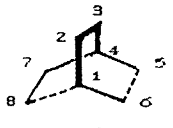
664-ac

Γ_2: (ε,β); $\beta = (14)(23)(67)(58)$



664-bb

Γ_3: $(\varepsilon,\alpha,\beta,\gamma)$; $\gamma = (14)(23)(56)(78)$



664-ab

Γ: (ε)

Fig. 3.

Codes for SHRT classes. A simple convention can be used to codify any G_1 graph on the basis of the dashed label distribution. Since the G_0 graph of the code *KLN* contains two annulated *cycles*, let us choose any node of degree 3 in the G_0 graph and denote non-boldface edges in both *cycles* by the letters *a, b, c*, . . . (starting from the *a*-edges adjacent to the chosen node). As a consequence, an expression *KLN* $(i_K j_K \ldots)$ $(i_L j_L \ldots)$, showing the position of dashed labels codifies a certain G_1 graph or a certain class of SHRT (the first is the smallest cycle; for cycles of an equal size, the letters should be of lexicographic order).

Previously, the idea of such codification was applied to HCRs of azoles [16]; it is now clear how to obtain the corresponding G_1 graphs and codify any HCRs. In particular, all the possible codes of pyridine HCRs should have the notation 66*N*-$(i)(j)$ (or simply 66*N*-*ij*), see the examples in fig. 3. Moreover, it is easy to codify SHRTs that are not HCRs: the only difference lies in the number of dashed edges used as labels of the G_0 graph and, evidently, in the corresponding number of the letters in the code. In practice, the largest number of formed and broken bonds is not more than 4, as it is for SHRTs (5) and (6) in fig. 2 (two pairs of ring bonds to be formed and to be broken). The resulting codes should have the notation KLN-$(i_K j_K)(i_L j_L)$, as for instance, 665-$(ab)(ab)$ and 664-$(ac)(ac)$ for reactions (5) and (6).

Hierarchic classification of SHRTs. The properties of RBR graphs discussed above permit us to suggest the first classification of simple heterocyclic ring transformation, which is the hierarchic one. In fact, on the first level only the sizes of K and L of the starting and final heterocycles are known. The pair (K, L) determines all the possible vectors (K, L, N): $N = 1, 2, \ldots, K$, $K \leq L$. These vectors form the second level of classification, level of G_0 graphs (or of the SHRT types) with the codes KLN. Each G_0 graph, due to its structure and symmetry, determines a new level, the level of G_1 graphs (or of SHRT classes) with the definite codes, e.g. KLN-$(i)(j)$ for HCRs. Each G_1 graph determines the corresponding pair of numbered M graphs and can be either directly compared with the real SHRT or used for the deeper levels of classification (involving, for instance, such labels as heteroatoms, substituents, fused rings, etc.).

The suggested classification of SHRTs is the full one (for not-*simple* HRT, see ref. [16]). This means that an expert chemist can easily use this classification tree not only to analyze the similarity of different ring transformations and distributions of known SHRT between the different classes but, more importantly, to predict unknown and chemically reasonable examples of simple HCRs and HRTs. The practical realization of the suggested classification (involving also deeper levels) is our computer program "GREH" (*G*raphs of *RE*cyclizations of *H*eterocycles, announced in ref. [18]), which can be used both as a database of known ring transformation and as a tool for prediction of unknown types of HCRs (see examples of some predictions in ref. [16]).

4. Degenerated ring-transformations of heterocycles

One specific sort of HRTs are *degenerated heterocyclic rearrangements* (DHCR). Elegant experimental examples of these ring transformations (often proved by the labelled atoms method or by temperature controlled NMR studies) are known for many monocyclic hetarenes, especially for 5- and 6-membered rings [1–10]. As in the case of any other HRTs, a lot of DHCR are *simple*. An example of DHCR is the reaction (3) if $R^1 = R^2$, see fig. 2.

More extensive is the bibliography on the *quasi-degenerated rearrangements*, different from the DHCR only by substituent presence and/or by tautomerism (e.g. the same reaction (3) if R^1 differs from R^2 can be considered as quasi-DHCR). An attractive peculiarity of quasi-DHCR is an illusive effect of the substituent(s) migration or exchange and a unique possibility to control the position of substituents (and/or multiple bonds) only by the temperature or pH variation; on the other hand, the existence of quasi-DHCR is often a reliable signal to search its degenerated prototype. Some reviews on HRTs include DHCR and quasi-DHCR [1–3,8,9]; nevertheless, it appears that these reactions have never been analyzed from a combinatorial point of view.

It seems we are the first to pose the question: how many degenerated rearrangements (which are SHRTs) are theoretically possible for the given heterocyclic nucleus? Although the general graph-theoretical constructions for different types of degenerated rearrangements have been discussed in the literature [21,22] with the following serious group-theoretical analysis of their reaction graphs [23,24], we did not find an answer to our question. The reason seems clear: before enumeration of DHCR for the given heterocycle, one should define the type (class) of this reaction (or a corresponding reaction graph). Because the general classification of possible types (classes) for SHRTs and HCRs now exists, our approach permits us to solve the enumeration problem of the simple DHCR.

Let us consider any SHRT of a K-membered heterocycle, which is DHCR; following our definitions, one can easily construct the corresponding M and RBR graphs. For degenerated SHRTs, the M_s and M_f graphs are obviously isomorphic (the only numeration of vertices in these two graphs is different). Let N be the number of vertices in the ring fragment common to both isomorphic M graphs ($1 < N \leq K$, we do not consider rare examples of $N = 1$); then the corresponding bicyclic G_0 graph with the code KKN can be defined immediately. Let A, B ($0 \leq A \leq B$) be the number of vertices in two side chains of the M graph (let us call them A and B fragments or chains); then, the K-membered cycle of one M graph should be constructed from these A and B chains and from the N-membered ring fragment, i.e. $A + B + N = K$.

Recall the isomorphism of M_s and M_f graphs; let us carry out the usual superposition (as in fig. 1) of these M graphs to obtain the G_1 graph; this includes the correspondence between the matching N-membered fragments of M graphs, while the chains A and B of one M graph should be placed at the $(K-N)$-membered fragment of the cycle of another M graph. It is possible in a unique manner if $A = B$, and there are two inequivalent possibilities to superpose if $A < B$. The resulting structures of G_1 graphs with the different relative disposition of the A and B fragments are symbolically shown as diagrams 4–6 in fig. 4, where the dashed edges of the

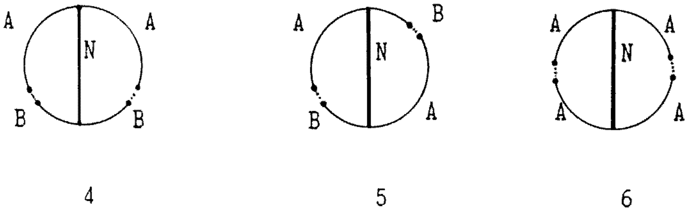


Fig. 4.

G_1 graph are located between the terminal vertices of the A and B fragments. Since there are no other variants to superpose an isomorphic M graph, diagrams 4–6 exhaust all types of G_1 graphs possible for DHCR.

Clearly, diagrams 4–6 of fig. 4 present three different types of symmetry of G_1 graphs. The symmetry of a G_1 graph (as of any other graph) can be characterized by the group of automorphisms Aut(Γ); this group is isomorphic to a certain permutation group determined, for instance, on the vertices of a graph [20]. Since the G_1 graph contains a set of labelled edges (solid, dashed and boldface), the only condition for permutations is to conserve both the incidence and the "color" of an edge between a pair of vertices.

Diagrams 4–6 present G_1 graphs with three different groups of automorphisms; let is call these groups Aut(Γ_1) = (ε, α), Aut(Γ_2) = (ε, β) and Aut(Γ_3) = (ε, α, β, γ), where ε is identical and α, β, γ are non-identical permutations of vertices. One can conclude that in the manner of permutation of the G_1 graph vertices (e.g. of the vertices from the bridge or from A and B fragments of diagrams 4–6), these groups Γ_1, Γ_2 and Γ_3 are isomorphic corresponding to the point groups C_{2v}, C_2 and D_{2h}. Evidently, the G_1 graph can be the RBR graph of the degenerated rearrangement only if its structure corresponds to any of diagrams 4–6 in fig. 4, i.e. if its group is of type Aut(Γ_i), $i = 1, 2, 3$. Examples of these groups for the concrete G_1 graphs are illustrated in fig. 3. (The last G_1 graph in this figure with the code 664-(*ab*) can never be the graph of any DHCR due to its group Aut(Γ) = (ε): there is no ancestor pair of isomorphic M graphs with such a G_1 graph.)

Now it is possible to analyze our question of DHCR enumeration for the given heterocyclic ring. Let us consider the simplest case of heterocyclic systems with only one heteroatom, e.g. such heteroaromatics as 5-membered pyrrole, furan and thiophene, or 6-membered such as pyridine, pyrillium or thiapyrillium nucleus. Some known examples of DHCR for these hetarenes can be found elsewhere [1–10].

Any heteroatom can be considered as the label for a vertex of both M graphs, and (after superposition) also for vertices of the G_1 graph. Due to only three possible (for DHCR) groups Aut(Γ_i) of G_1 graphs, the following requirements should be used for labelling the vertices of the G_1 graph:

(1) for the vertices v_i and v_j of the G_1 graph, which are permuted by permutation π of the groups Aut(Γ_i), $i = 1, 2, 3$, labelling of v_i requires labelling of v_j, and

(2) v_i and v_j should belong to the different *cycles* (in the above-mentioned sense) of the G_1 graph. (This requirement follows our consideration of a heterocycle with only one heteroatom, i.e. one label in the M graph.)

Let us consider a K-membered heterocycle that undergoes all possible DHCR to be enumerated. For the given value N (fixed G_0 graph), the possible G_1 graphs of DHCR are differing by their groups Aut(Γ_i) and the sizes of the A and B fragments. For the chosen G_1 graph, all the inequivalent possibilities of its labelling by one heteroatom (following the above conditions (1) and (2)) correpond to the required number of DHCR and can be calculated in the following manner:

(a) for group Γ_1 (diagram 4): $A + B + N$ variants. It is possible to label *any pair* of vertices that belongs to A (or B) fragments of the G_1 graph and are permuted by permutation α (i.e. $A + B$ variants) and *any vertex* from the bridge (i.e. N variants); the resulting value is equal to the size K of the heterocycle;

(b) for group Γ_2 (diagram 5): $A + B + (N \bmod 2)$ variants. It is possible to label *any pair* of vertices from fragments A (or B), which are permuted by permutation β (i.e. $A + B$ variants), and *only* central vertex of a bridge if its size is odd (i.e. $(N \bmod 2)$ variants). Labelling of any other vertex from the odd bridge (or any vertex from the even bridge) violates condition (2);

(c) for group Γ_3 (diagram 6): $2A + (N + 1)/2$ variants. Because $A = B$, *any pair* of vertices from fragments A (or B) which are permuted by permutations α *and* β can be labelled (i.e. $2A$ variants), and *only those* vertices from a bridge that are non-equivalent to the action of permutations β and γ and remain immovable to the action of permutation α (i.e. $(N + 1)/2$ variants).

Now the proposed formulae should be consequently applied for all other G_0 graphs as ancestors of G_1 graphs. Due to the hierarchic classification, the size of cycle K determined possible G_0 graphs with different $N > 1$, and the symmetry requirements select appropriate G_1 graphs to be labelled with possible A and B fragments. The resulting sum of all labelled G_1 graphs should give the number of all possible degenerated SHRTs for the given heterocycles with only one heteroatom. Let us illustrate the use of our formulae for enumeration of DHCR for ring systems of pyrrole, furan and thiophene.

There are only three possible G_0 graphs with $N > 1$ corresponding to SHRTs of 5-membered heterocycles into othe 5-membered rings; their codes are 552, 553, and 554. Among all possible G_1 graphs with only a pair of dashed-marked edges (which is necessary for any HCRs) there are only nine graphs, whose groups are Γ_1, Γ_2 or Γ_3 with the following codes:

group Γ_1 : 552-$(a)(a)$, 552-$(b)(b)$, 553-$(a)(a)$, 554-$(a)(a)$;

group Γ_2 : 552-$(a)(d)$, 552-$(b)(c)$, 553-$(a)(c)$, 554-$(a)(b)$;

group Γ_3 : 553-$(b)(b)$.

The size of fragments A, B and N for these G_1 graphs and the possible number of labellings is shown in table 1.

In the case of DHCR of 6-membered rings with one heteroatom (e.g. pyridine, pyrillium or thiapyrillium), the possible G_0 graphs have the codes 662, 663, 664 and 665. There are only fourteen G_1 graphs with groups Γ_1, Γ_2 or Γ_3 whose codes, A, B and N values, and the possible number of rearrangements are also shown in table 1.

These data exhaust all the number of DHCR (evidently, which are SHRTs) for 5- and 6-membered heterocycles with only one heteroatom. As an example, all

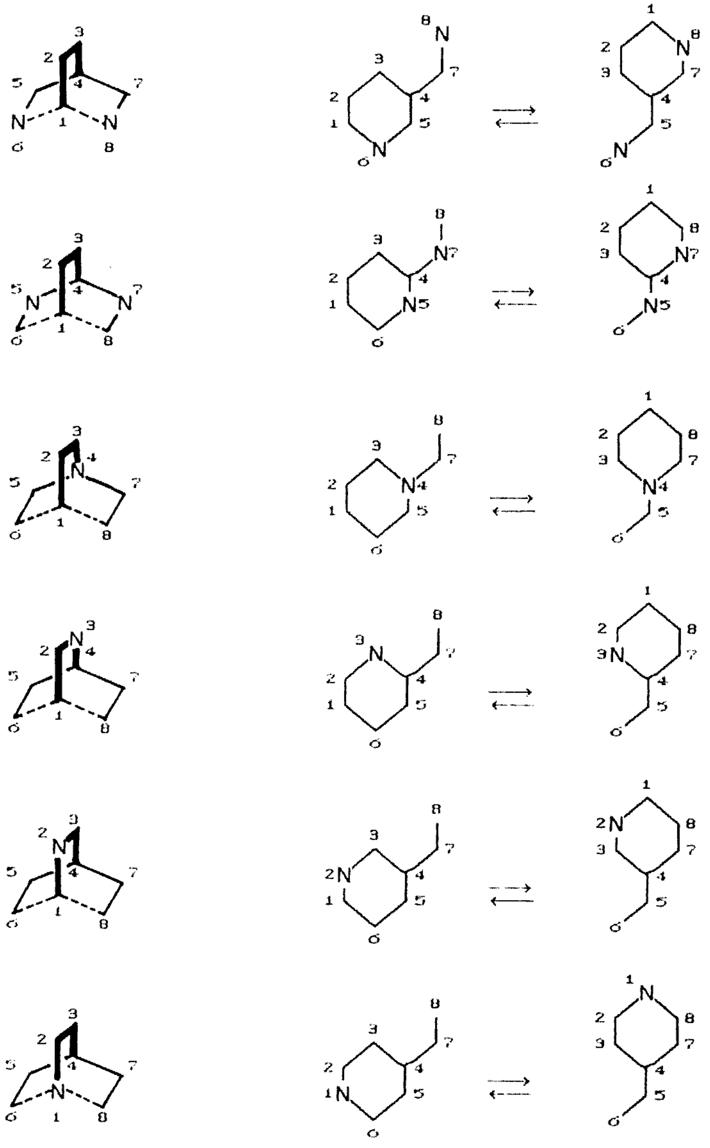


Fig. 5.

Table 1

5-membered rings					6-membered rings				
group and code of G_0 graph	N	A	B	number of DHCR	code of G_1 graph	N	A	B	number of DHCR
Γ_1: 552-*aa*	2	0	3	5	662-*aa*	2	0	4	6
552-*bb*	2	1	2	5	662-*bb*	2	1	3	6
553-*aa*	3	0	2	5	663-*aa*	3	0	3	6
554-*aa*	4	0	1	5	663-*bb*	3	1	2	6
					664-*aa*	4	0	2	6
					665-*aa*	5	0	1	6
Γ_2: 552-*ad*	2	0	3	3	662-*ae*	2	0	4	4
552-*bc*	2	1	2	3	662-*bd*	2	3	1	4
553-*ac*	3	0	2	3	663-*ad*	3	0	3	4
554-*ab*	4	0	1	1	663-*bc*	3	1	2	4
					664-*ac*	4	0	2	2
					665-*ab*	5	0	1	2
Γ_3: 553-*bb*	3	1	1	4	662-*cc*	2	2	2	5
					664-*bb*	4	1	1	4

the labelled G_1 graphs with the code 664-$(a)(a)$ and the corresponding labelled pairs of M graphs are shown in fig. 5 for the sekeleton of the pyridine nucleus. We would like to mention that the only known example of degenerated SHRT of pyridine seems to be the above discussed Dimroth rearrangement (3) in fig. 2. The results would also be applied in the search for new quasi-DHCR.

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