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THEORY OF THE WIENER NUMBER OF GRAPHS. II. TRANSFER GRAPHS AND SOME OF THEIR METRIC PROPERTIES

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ABSTRACT

A particular class of simple connected graphs is specified. Each graph H of this class, consists of two subgraphs G_1 and G_2 which are connected either by an edge (a bridge), or by a common vertex, or by a common edge. Within this class of graphs, there always exist pairs of graphs H_1 and H_2 such that H_2 can be regarded as obtained from H_1 by the transfer of G_2 with respect to G_1. A subclass of transfer chain graphs is also specified by the presence of a transfer chain (a path subgraph) to the vertices of which any kind of other subgraphs may be attached. The interconversions of graphs H_1 and H_2 correspond to molecular rearrangements in isomeric compounds. The metric properties of these graphs and, particularly, the change in the Wiener number ΔW produced by the various fragment transfers is studied in detail making use of the formalism developed in Part I [1]. A number of properties and corollaries is proved for ΔW of transfer graphs including here the conditions for two such graphs to have the same Wiener number (isowiener graphs) or to have the same difference in the Wiener number (isodifferent graphs).

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INTRODUCTION

The metric properties of graphs have been for a long time of interest in mathematics [2-7] and in applying mathematical methods in various scientific fields such as electrical engineering [8], transport networks [9], geology [10], biology [11], psychology [12], sociology [13], etc. Due to its specific nature, chemistry is that particular branch of science in which the concept of graph distances has found the most extensive applications [14,15].

Among the various metric characteristics of a graph G [6] it is the Wiener number W(G) which is of greatest importance. Introduced empirically by H. Wiener [16] this number equals the sum of distances between any pair of graph vertices or, otherwise, it is the half-sum of all distance matrix entries [2]. Being a good numerical measure for the compactness of a system or for its element interdependence, the Wiener number proved to be of relevance to physico-chemical properties of chemical compounds [17-20], polymers [21-24], and crystals [25-27]. It has been applied to quantitative structure-property [28-33] and structure-activity [34] correlations. More general studies on molecular branching [35] and cyclicity [36-37] have also been performed on this basis. Very recently, some light was shed [38] on the problem why does this topological index work so well in structural chemistry by establishing its close relation to another graph-invariant, the number of self-returning walks, which has a direct quantum-mechanical background [39].

In Part I of this series [1] we developed a new formalism for the study of the Wiener number based on the distance numbers of graph vertices (the sum of the distances from all graph vertices to a certain vertex). The changes in the distance and Wiener numbers after some graph operations were studied, and a number of properties was proved for these quantities. In the present work we introduce some particular classes of graphs for which the changes in the Wiener number after some specified graph transformations are investigated and expressed in a number of properties and formulae including essential structural parameters. As shown in a subsequent publication {40} the new formalism provides a more general treatment of molecular branching.

2. GENERAL "TRANSFER GRAPHS"

Let the simple connected non-isomorphous graphs H_1 and H_2 be considered. Let also H_1 and H_2 be built by the same non-isomorphous graphs G_1 and G_2 which are either: (i) linked by an edge (a bridge) $\{au\}$ or $\{av\}$, $a \in G_2$, $u, v \in G_1$, or (ii) are covered upon a vertex $a \equiv u$ or $a \equiv v$, or (iii) are covered upon an edge $\{a_1 a_2\} \equiv \{u_1 u_2\}$ or $\{a_1 a_2\} \equiv \{v_1 v_2\}$; $u_1, u_2, v_1, v_2 \in G_1$; $a_1, a_2 \in G_2$. Obviously, H_1 and H_2 differ solely in the specific location of G_2 with respect to G_1. Otherwise, H_2 may be regarded as obtained from H_1 upon a transfer of G_2 from the initial vertex(es) u (u_1, u_2) to another one(s) $v(v_1, v_2)$. In the following each pair of graphs H_1 and H_2 that obeys the above conditions will be termed "transfer graphs". An important type of transfer graphs will be considered in Section 3. Depending on the kind of G_1 - G_2 linkage, the transfer graphs can be divided into three classes corresponding to cases (i), (ii), and (iii) given above. The notations H^b, H^s, and H^f will be used in these cases where the symbols b, s, and f are abbreviations for bridged-, spiro-, and fused G_1 and G_2. Each H^b or H^s graph posseses at least one cut set of vertices [41] of cardinality 1 while each H^f graph possesses at least one cut set of edges [3] of cardinality one. The vertices u and v in H^b and H^s, as well as the pair of vertices u_1, u_2 and v_1, v_2 in H^f will be termed "transfer vertices". Evidently, in the case of molecular graphs, H_1 and H_2 may be regarded as representatives of the well known case of constitutional isomers in chemistry. Hence, the $H_1 \longrightarrow H_2$ transformation corresponds to the large class of molecular rearrangements in which a molecular fragment (linked by a sole bond or spiro-linked or fused to the remaining part of the molecule) is displaced from its initial location to another one (Fig. 1a, b, c).

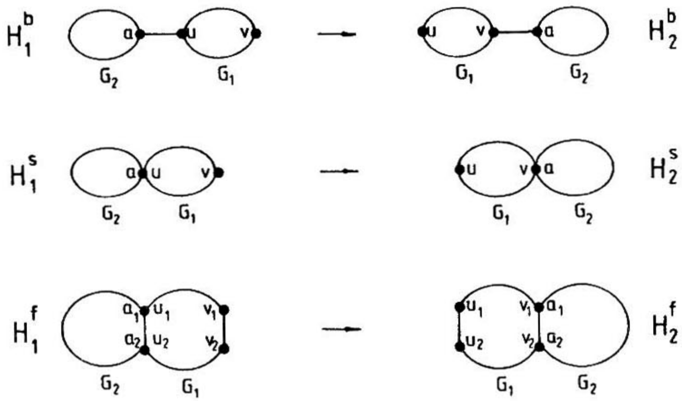


Fig. 1. A general presentation of the so-called transfer graphs H_1 and H_2 and their three classes in which the subgraph G_2 is linked with the subgraph G_1:
a) by an edge (a bridge), class H^b; b) by a common vertex (spiro-linkage), class H^s; c) by a common edge (cycle fusion), class H^f

The Wiener number of both H_1 and H_2 can be determined by making use of the equations derived in Part I of this series [1] (eqs. I.22, I.16, and I.19) for graphs obtained from their subgraphs G_1 and G_2 by the three operations described above (joined by an edge and covered upon a vertex or edge, respectively). Eq. (I.16) is given here in a simplified form:

$$W(H^b) = W(G_1) + W(G_2) + n_1 n_2 + n_2 d(u|G_1) + n_1 d(a|G_2) \quad (1)$$

$$W(H^s) = W(G_1) + W(G_2) + (n_2-1) d(u|G_1) + (n_1-1) d(a|G_2) \quad (2)$$

$$W(H^f) = W(G_1) + W(G_2) + 1 + 1/2\{|q|.|t| - n_1 n_2 + (n_2-2)[d(u_1|G_1) + d(u_2|G_1)] + (n_1-2)[d(a_1|G_2) + d(a_2|G_2)] - [d(u_1|G_1) - d(u_2|G_1)].[d(a_1|G_2) - d(a_2|G_2)] \quad (3)$$

Here n_1 and n_2 stand for the number of vertices of subgraphs G_1 and G_2, $W(H_1)$ and $W(H_2)$ are the Wiener numbers of graphs H_1 and H_2, respectively; $d(x|G_j)$ is the distance number of vertex x in the subgraph G_j, j = 1 or 2; $|q|$ and $|t|$ are the cardinalities of the subsets of those vertices in G_1 and G_2 that are equally distant from vertices u_1 u_2, and a_1, a_2, respectively.

It should be noted that eq. (1) is valid for three different classes of chemical compounds: those of acyclic, branched cyclic, and bridged cyclic compounds, since they all are represented by the same class of transfer graphs, H^b (See also Fig. 2a, b, and c).

Eqs. (1) to (3) are actually written for graph H_1. They can also be used for H_2 by replacing u, u_1, and u_2 by v, v_1, and v_2, respectively (See Fig. 1). Then, by subtracting the three pairs of equations for $W(H_1)$ and $W(H_2)$ one arrives at equations (4) to (6) for the change in the Wiener number produced by the described transformations of H_1 into H_2:

$$\Delta W^{b} = W(H^{b}_{1}) - W(H^{b}_{2}) = n_2 [d(u|G_1) - d(v|G_1)] \quad (4)$$

$$\Delta W^{s} = W(H^{s}_{1}) - W(H^{s}_{2}) = (n_2 - 1) [d(u|G_1) - d(v|G_1)] \quad (5)$$

$$\Delta W^{f} = W(H^{f}_{1}) - W(H^{f}_{2}) = 1/2\{(n_2 - 2)[d(u_1|G_1) + d(u_2|G_1) -$$
$$- d(v_1|G_1) - d(v_2|G_1)] + [d(a_1|G_2) - d(a_2|G_2].$$
$$[d(v_1|G_1) - d(u_1|G_1) + d(u_2|G_1) - d(v_2|G_1)]\} \quad (6)$$

Equations (4) to (6) provide the basis for the calculation of the change in the Wiener number produced by any kind of fragment displacement in molecular graphs. They can be applied to the study of molecular property alterations occurring during the respective molecular rearrangements. (See e.g. refs. [4-6]). The real importance of eqs. (4) to (6), however, is in the possibility they offer for a generalized treatment of the alterations in topology occuring during such molecular transformations. More specifically, relations of inequality or equality of the Wiener number of molecular graphs can be thus deduced, providing also a generalized approach towards molecular branching and cyclicity. The fragment displacements can be thus studied in chemical compounds with any possible topology: acyclic, branched cyclic, bridged cyclic, spiro-cyclic, and fused cyclic ones, as well as in compounds with a mixed type of cycle linkage (Fig. 2).

Equations (4) to (6) exhibit some properties of the change in the Wiener number ΔW occurring during the graph transformation under study: the transfer of graph G_2, which is joined to graph G_1 by an edge, or has a common vertex or a common edge with G_1, from the initial transfer vertex(es) to the final one(s).

Property 2.1. For transfer graphs of H^b and H^s classes neither the sign of ΔW nor its annulation depends on the transferred graph G_2.

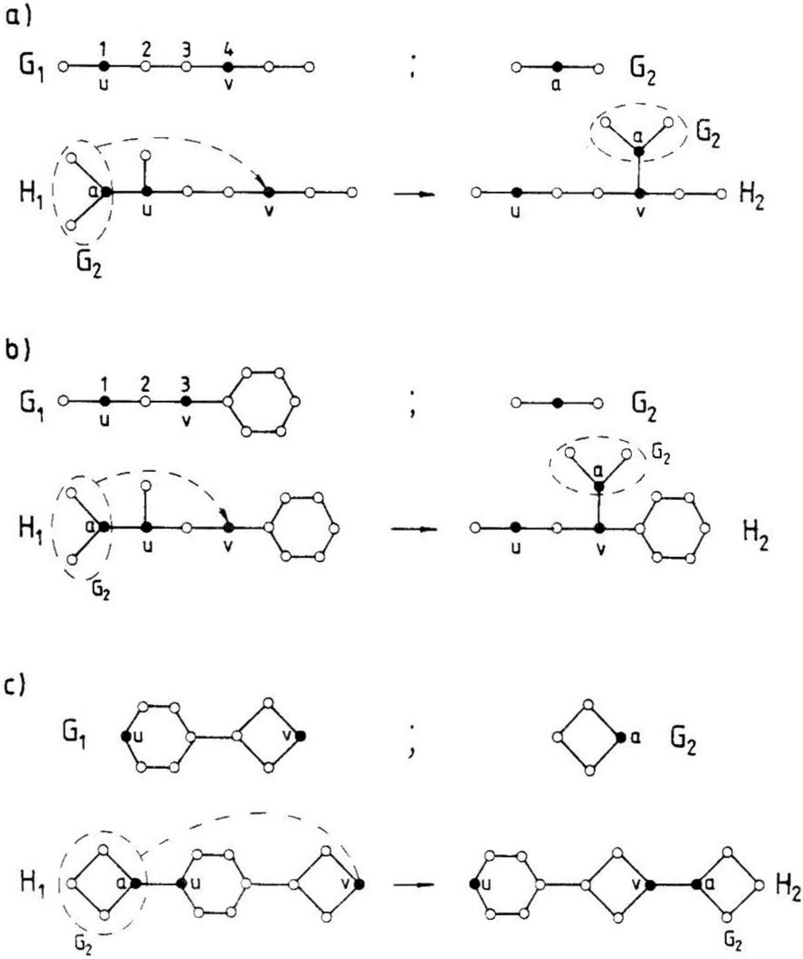
Property 2.2a. For transfer graphs of H^b and H^s classes the value of ΔW does not depend on the topology of the transferred graph G_2 but it depends on the total number of vertices of G_2.

Property 2.2b. For transfer graphs of H^f class the value of ΔW depends on the total number of vertices of the transferred graph G_2, as well as on the distance numbers of the pair of its transfer vertices.

For lack of more specific requirements in Properties 2.1, 2.2a, and 2.2b, the transferred graph G_2 can be any acyclic or cyclic graph.

Property 2.3. Both the sign and value of ΔW depend on the difference in the distance numbers of the two transfer vertices in G_1.

As a consequence of Property 2.3 the graph G_1 can also be any cyclic or acyclic graph. Two more general consequences can also be formulated.


a)
G1
1 2 3 4
u v
G2
a
H1
a
u
v
G2
H2
b)
c)

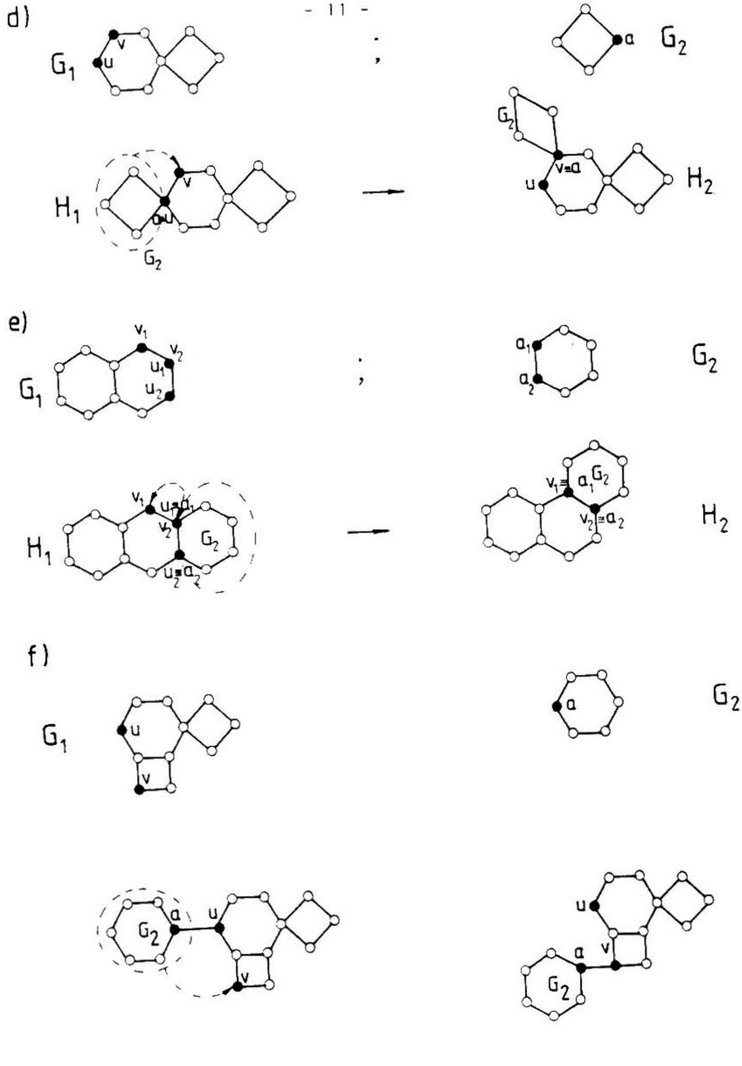


Fig. 2. Illustration of the possible application of eqs. (1) to (6) to fragment transfers in: a) a cyclic, b) branched cyclic, c) bridged cyclic, d) spiro-cyclic, e) fused cyclic, f) cyclic compounds with mixed type of cycle linkage

Corollary 1 to Property 2.3

If for graphs H^b_1 and H^b_2, as well as for H^s_1 and H^s_2

$$d(u|G_1) = d(v|G_1) \tag{7a}$$

or if for graphs H^f_1 and H^f_2

$$d(u_1|G_1) = d(v_1|G_1), \quad d(u_2|G_1) = d(v_2|G_1) \tag{7b}$$

hold, then

$$\Delta W^i = W(H^i_1) - W(H^i_2) = 0 \tag{8}$$

where $i = b, s,$ or f.

The proof of eq. (8) follows immediately from the insertion of condition (7a) into eqs. (4) and (5), as well as from the insertion of (7b) into eq. (6).

Evidently, equalities (7) represent the necessary and sufficient conditions for the transfer graphs H_1 and H_2 to have the same Wiener number. Such graphs will be termed "isowiener graphs". Some classes of isowiener transfer graphs will be specified in Section 3.

Corollary 2 to Property 2.3

If for graphs H^i_1 and H^i_2

$$n_2 = \text{const} \tag{9a},$$

and $\quad d(u|G_1) - d(v|G_1) = \text{const} = b$

or $\quad d(u_1|G_1) - d(v_1|G_1) = d(u_2|G_1) - d(v_2|G_1) = \text{const} = b \quad$ (9b)

hold at a time. Then one obtains

$$\Delta W^i = W(H^i_1) - W(H^i_2) = \text{const} = (n_2 - a)b \tag{10}$$

where $a = 0, 1$ or 2 for $i \equiv b, s,$ and f, respectively.

In addition, if for graphs H^f_1 and H^f_2

$$n_2 = \text{const} \tag{9c}$$

$$d(u_1|G_1) - d(v_1|G_1) = \text{const} \neq d(u_2|G_1) - d(v_2|G_2) = \text{const} \tag{9d}$$

$$d(a_1|G_2) - d(a_2|G_2) = \text{const} \tag{9e}$$

hold at a time. Then again one obtains

$$\Delta W^{f} = W(H^{f}_{1}) - W(H^{f}_{2}) = \text{const} \qquad (10a)$$

The proof is analogous to that of Corollary 1. Equalities (9a) to (9e) represent the necessary and sufficient conditions for the pairs of transfer graphs H_1 and H_2 to have a constant difference in their Wiener numbers for different pairs of subgraphs G_1 and G_2. The pairs of graphs H_1 and H_2 satisfying eq. (10) will be termed "isodifferent transfer graphs". Some classes of such transfer graphs will be specified in Section 3.

Properties 2.1 to 2.3 and the two corollaries to Property 2.3 formulated in the foregoing refer to such G_2 transfers which preserve the kind of their linkage with G_1 (b-, s-, or f-class). When, however, the attachment type is changed upon the G_2 transfer, ΔW loses a great deal of these properties. This is demonstrated below proceeding from two new equations (11a) and (11b) for the change in the Wiener number during the G_2 transfer at which the linkage of G_2 and G_1 alters from b- to s-class, and vice versa. These equations are derived by the subtraction of eqs. (1) and (2).

$$\Delta W^{b,s} = W(H^{b}_{1}) - W(H^{s}_{2}) = n_2 [d(u|G_1) - d(v|G_1)] + n_1 n_2 + d(v|G_1) + d(a|G_2), \qquad (11a)$$

$$\Delta W^{s,b} = W(H^{s}_{1}) - W(H^{b}_{2}) = n_2 [d(u|G_1) - d(v|G_1)] - n_1 n_2 - d(u|G_1) - d(a|G_2). \qquad (11b)$$

As seen from eqs. (11a,b) Properties 2.1 and 2.2a are no more valid while Property 2.3 and its two corollaries become more complicated, due to the three additional terms as compared with eqs. (4) and (5).

Property 2.4. When the transfer of graph G_2 occurs with a simultaneous change in the kind of $G_1 - G_2$ attachment from bridged to spiro- one, or vice versa, the sign and value of ΔW depend on the distance numbers of the transfer vertices G_1 and G_2, as well as on the total number of vertices in G_1 and G_2.

Corollary 1 to Property 2.4.

If for graphs H^b_1 and H^s_2, the first one being of b-class and the second one being of s-class

$$n_2 [d(u|G_1) - d(v|G_1)] = - n_1 n_2 - d(v|G_1) - d(a|G_2) \tag{12a}$$

or, vice versa, if for graphs H^s_1 and H^b_2

$$n_2 [d(u|G_1) - d(v|G_1)] = n_1 n_2 + d(u|G_1) + d(a|G_2) \tag{12b}$$

hold, then

$$\Delta W^{b,s} = W(H^b_1) - W(H^s_2) = \Delta W^{s,b} = W(H^s_1) - W(H^b_2) = 0 \tag{13}$$

Equalities (12) represent the necessary and sufficient condition for pairs of graphs (H^b_1, H^s_2) and (H^s_1, H^b_2) to be isowiener graphs.

Corollary 2 to Property 2.4 could be similarly formulated.

A more general property can be formulated which refers to all the three cases of G_2 transfers that change the $G_1 - G_2$ linkage type; bridging $\rightleftharpoons$ spiro, bridging $\rightleftharpoons$ fusion, and spiro $\rightleftharpoons$ fusion ($b \rightleftharpoons s$, $b \rightleftharpoons f$, and $s \rightleftharpoons f$, respectively). The notation $W^{i,j}$ will be used where $i, j = b, s$, or f, and $i = j$.

Property 2.5. Both sign and value of $\Delta W^{i,j}$ depend on the change in the type of linking graphs G_2 to G_1.

This property follows for $b \rightleftharpoons s$ transfers from inspection of eqs. (11a,b) where the three additional terms, taken with their signs plus or minus, influence strongly the value of ΔW and can alter the ΔW sign.

Inspite of the generality of our approach we do not present here the equations describing the other two cases of $H_1 \rightarrow H_2$ transformation which alter the linkage type of G_1 and G_2 (fusion $\rightleftharpoons$ bridging and fusion $\rightleftharpoons$ spiro). Albeit, as readily obtained by subtracting the pairs of eqs. (1), (3) and (2), (3) these equations will be given in a subsequent publication [42], devoted to cyclicity of molecular systems. Fragment transfers in bridged, spiro-, and fused cyclic compounds will be analyzed in detail there. In the present paper, besides the general properties of the change in the Wiener number upon the specified graph transformations, we develop the basis for treating from a general view-point [40] the problem of branching in acyclic (Fig. 2a) and branched cyclic (Fig. 2b) compounds, as well as in some bridged, spiro-, and fused cyclic compounds (Fig. 2c, d). With this aim in mind some metric properties of the important class of transfer graphs, which we term "transfer chain graphs", are studied in the next Section 3.

3. TRANSFER CHAIN GRAPHS

3.1. Basic notions and equations

Let now the transfer graphs H_1 and H_2 meet the additional condition to contain the same path-subgraph P_{uv} (called further "transfer chain") whose vertices u, u+1, ..., k, ..., v-1, v have positions j_k = 1, 2, 3, ..., n_0, respectively. Let further H_1 and H_2 contain the same subgraphs G_{jl} (l = 0, 1, 2, ..., p), the latter being any kind of simple connected graphs (either acyclic or cyclic ones) with n_{jl} vertices each one. Each of G_{jl} is connected with P_{uv} either by an edge not belonging to them (G^b_{jl} = acyclic or bridged cyclic subgraphs) or by a common vertex (G^{s}_{jl} : spiro-linked cyclic subgraph or by a common edge or edges (G^{f}_{jl} = fused cyclic subgraph). Two G_{jl} could be connected only by the vertex(es) they have common with P_{uv}. The subgraph G_2 to be transferred will be denoted by G_{1l}. By definition $G_{1l} \notin G_1$. In this study $G_{1l} \equiv G^{b}_{1l}$ or $G_{1l} \equiv G^{s}_{1l}$ are considered.

The shortest path $W_{kk'}$ ($k, k' \in P_{uv}$) between any pair of vertices k and k' belonging to the transfer chain is always a subgraph of the latter. Due to this, any transfer of G_{1l} from vertex k to vertex k' can be treated as a transfer between positions $j_u = 1$ and $j_v = n_0$, i.e. as a transfer between the two terminal vertices u and v in P_{uv}. Actually, if a fragment transfer between non-terminal vertices occurs, the two ends of the transfer chain together with all subgraphs attached to them could always be treated as two large subgraphs G_{1l} and $G_{n_0 l}$ connected with the two terminal vertices of a shorter transfer chain (Fig. 3a).

Similarly, all G_{1l} transfers between two vertices t and t' belonging to two different acyclic subgraphs (branches) of G_1 will be handled as a transfer between the two terminal vertices $u \equiv t$ and $v \equiv t'$ of the new transfer chain (Fig. 3b). The same holds for cases when G_{1l} transfers occur between two vertices r and r' belonging to one or two cyclic G_{jl} (Fig. 3c). Clearly, the shortest path $W(rr'|G_1)$ should be selected as a transfer chain in the latter case.

When one or more cyclic G_{jl} are fused to the transfer chain ($G_{jl} \equiv G^f_{jl}$ then multiple (double, triple, etc.) transfer chain or subchain could exist (Fig. 3d). Sets (pairs, triplets, etc.) of vertices k, k', k", ..., m, m', m", ..., etc., located within the multiple transfer (sub)chains are equivalent with respect to their distance to u and v.

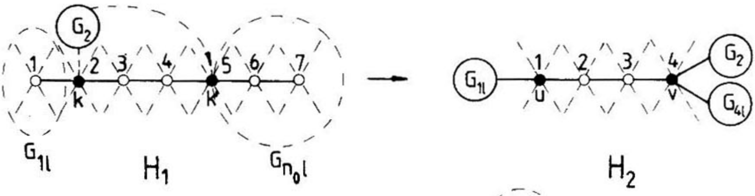
It follows from the foregoing and eqs. (4) and (5) that the basic properties of the change in the Wiener number ΔW upon the graph transformations under study are determined by the difference in the distance numbers of the two terminal vertices of the transfer chain, $d(u|G_l)$ and $d(v|G_l)$, respectively. This difference can be presented as consisting of two types of contributions: the distances from u and v to the remaining vertices in the transfer chain and those to the vertices in all subgraphs G_{jl} :

$$\begin{aligned} d(u|G_l) - d(v|G_l) = & \sum_{k \in P_{uv}} (d(uk|G_l) + \sum_{j} \sum_{l \neq l'} \sum_{t \in G_{jl}} d(ut|G_l) - \\ & - \sum_{k \in P_{uv}} d(vk|G_l) - \sum_{j} \sum_{l \neq l'} \sum_{t \in G_{jl}} d(vt|G_l) \end{aligned} \qquad (14)$$

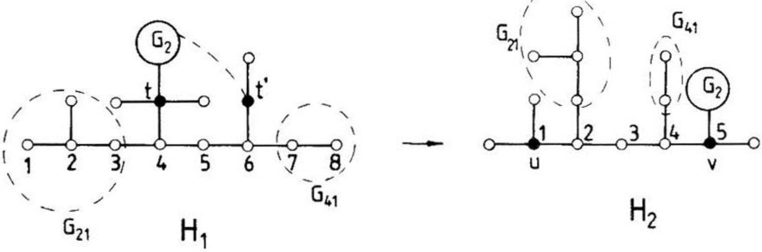
Here, k and t stand for an arbitrary vertex from the transfer chain and from the lth subgraph linked with the transfer chain in a vertex having position j, respectively.

The subgraph, $G_2 \equiv G_{11}$, having j = 1 and l = 1 is denoted for convenience with a different subscript l = 1'; it is not taken into account in the summation since by definition it does not belong to G_l.

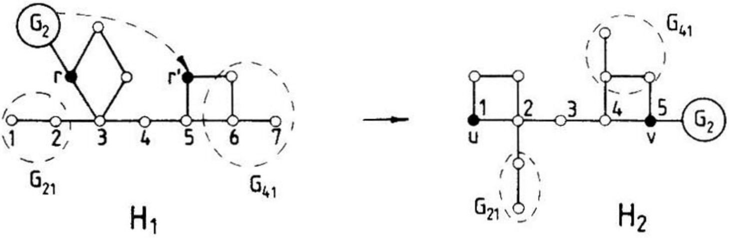
a)

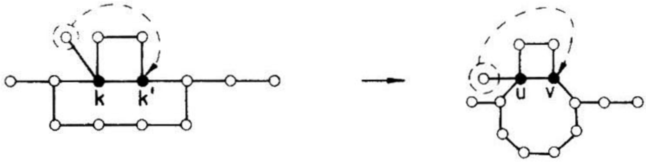

G2
1
2
3
4
5
6
7
k
k'
G1l
H1
Gn0l
G1l
u
v
G4l
H2

b)


G2
t
t'
1
2
3
4
5
6
7
8
G21
H1
G41
u
v
H2

c)


G2
r
r'
1
2
3
4
5
6
7
G21
H1
G41
u
v
H2


k
k'
u
v

d)

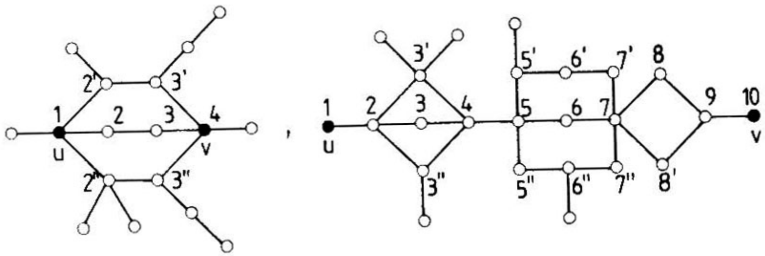


Fig. 3. Illustration of the flexibility of the transfer chain notion. New transfer chains are formed so as to have always <u>terminal</u> transfer vertices u and v in case of G_2 displacement: a) from an internal chain vertex to another internal chain vertex, b) from a branch vertex to another branch vertex, c) from a vertex belonging to one cycle to a vertex belonging to another cycle, or to the same cycle; d) triple transfer chain and a multiple transfer chain having two triple and one double transfer subchains

3.2. Transfer chain graphs with subgraphs attached to the chain by an edge (a bridge) or common vertex

The first and third term in eq. (14) mutually cancel because u and v are the two terminal vertices of P_{uv}. The sums of the distances from vertices u and v to all the vertices t in all bridged or spiro linked subgraphs G^b_{jl} and G^s_{jl} can on their turn be divided into two contributions: the distances to the respective transfer chain vertices k_j and the distances from the latter to each of the vertices within the subgraphs. The second contributions are constant and vanish upon the subtraction in eq. (14). One thus obtains for $G_{jl} \equiv G^b_{jl}$ and $G_{jl} \equiv G^s_{jl}$:

$$d(u|G_l) - d(v|G_l) = \sum_j \sum_{l \neq l'} \sum_{t \in G_{jl}} \{[(j-1).$$

$$.(n^i_{jl} + d(k_j t|G_{jl})] - [(n_0 - j)n^i_{jl} + d(k_j t|G_{jl})]\} \quad (15),$$

or simply:

$$d(u|G_l) - d(v|G_l) = \sum_j \sum_{l \neq l'} (2j - n_0 - 1)n_{jl} \quad (16)$$

Here n^i_{jl} is the number of those vertices in G^i_{jl} which are not common with P_{uv}. For $G_{jl} \equiv G^b_{jl}$ and $G_{jl} \equiv$ $\equiv G^s_{jl}$, $n^b_{jl} = n_{jl}$ and $n^s_{jl} = n_{jl} - 1$ holds, respectively, n_{jl} being the total number of vertices in G_{jl}.

Eq. (16) can be ultimately simplified by taking into account the fact that the double summation in this equation runs over the different variables j and l which can be separated:

$$d(u|G) - d(v|G) = \sum_j (2j - n_0 - 1) \sum_{l \neq l'} n^i_{jl} =$$

$$= \sum_j (2j - n_0 - 1).n'_j \quad (17)$$

where $n'_j = \sum_{l \neq l'} n^i_{jl}$ is the total number of vertices not belonging to P_{uv} in all subgraphs G_{jl} attached to vertex k_j having position j in P_{uv}. When all $n^i_{jl} \equiv n^b_{jl}$, then $n'_j = n_j$ (the total number of vertices in all subgraphs G^b_{jl}). When all $n^i_{jl} = n^s_{jl}$, then $n'_j = n_j - s_j$. s_j being the total number of G^s_{jl}. $G_2 \equiv G_{11} \equiv G_{1l'}$ which is attached to vertices u or v is excluded from the summation since $G_2 \notin G_1$ by definition.

Proceeding from eqs. (4) and (5) one arrives thus to the general equation (18) for the change in the Wiener number occurring upon the transformation of the transfer chain graph H_1 into another transfer chain graph H_2. More specifically, eq. (18) refers to transfers of the subgraph $G_2 \equiv G_{11}$ which is connected by an edge (a bridge) or by a vertex (but not by a common edge!) with the transfer chain subgraph G_1, the latter having subgraphs G^b_{jl} or G^s_{jl}, bridged or spiro- linked to the transfer chain P_{uv}, respectively:

$$\Delta W^i = W(H^i_1) - W(H^i_2) = (n_{11} - a) \sum_j (2j - n_0 - 1) . n'_j \qquad (18)$$

where a = 0 for i = b, and a = 1 for i = s. The number of vertices in the subgraph $G_2 \equiv G_{11}$ is denoted here by n_{11} instead of n_2 to avoid any confusion with the term n_j in case of j = 2.

The case i = b refers to fragment transfers in acyclic, branched cyclic, and bridged cyclic compounds whilst i = s treats the spiro-cyclic compounds which can also contain acyclic branches or/and cyclic fragments linked by bridges. For examples illustrating such applications of eq. (18) one may go back to Figs. 2a to 2d. A more general example, illustrating eq. (18) is given in Fig. 4.

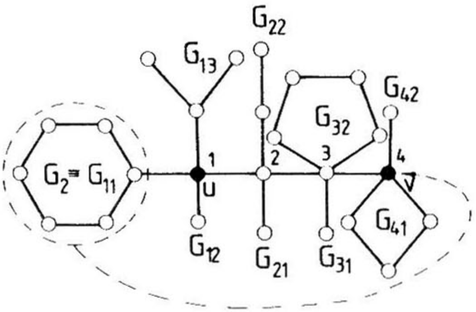


Fig. 4. Illustration to the application of eq. (18). Here $n_{11} = 6$, $a = 0$, $n_0 = 4$, $n'_1 = 4$, $n'_2 = 3$, $n'_3 = 5$ ($n_3 = 6$, $s_3 = 1$), $n'_4 = 4$ ($n_4 = 5$, $S_4 = 1$), $\Delta W^b = 12$. For comparison with eq. (4): $d(u|G_1) = 53$, $d(v|G_1) = 51$.

Eq. (18) exhibits some of the properties of eqs. (4) and (5), and more specifically, Property 2.1 and Property 2.2a. Property 2.3 is certainly no longer valid in the same formulation because the difference in the distance numbers of the transition vertices u and v was expressed by other variables. New properties, however, can be formulated or proved for the transfer chain graphs.

<u>Property 3.1</u>. The value of ΔW does not depend neither on the structure nor on the number of subgraphs G_{jl} attached to a certain vertex k_j having position j in the transfer chain P_{uv} but it depends on the position j, as well as on the total number of those vertices in the these subgraphs, n'_j, that are not common with P_{uv} .

The proof of this property was actually given by deducing eq. (17) from eq. (16), i.e. it results from the possibility for a separate summation over the two variables j and l in eq. (16).

It should be noted that unlike ΔW, the Wiener numbers $W(H_l)$

and $W(H_2)$ depend on the number of subgraphs at each vertex.

Another property considers the mutual influence on ΔW of the total number of vertices in the subgraphs attached to vertex K_j, n_j, and the position of attachment j in the transfer chain. In the general case, the subgraph transfer can result in ΔW positive or negative or zero, depending on the relative contribution of j and n_j. Some conclusions, however, can be made by using the symmetry of j with respect to the initial and final transfer vertices u and v.

Property 3.2. The transfer of a subgraph G^b_{11} or G^s_{11} between the two terminal vertices u and v of the transfer chain P_{uv} without changing the attachment type decreases (increases) the Wiener number when for each pair of vertices x, $y \in P_{uv}$, located symmetrically with respect to u and v, respectively, the total number of vertices in the subgraphs attached to y is larger (smaller) than, or equal to, the one for x. In case of equality for each pair (x, y) the Wiener number does not change.

Proof. Let the closed interval $[j_u, j_v]$ be divided into two equal parts, as shown in Fig. 5.

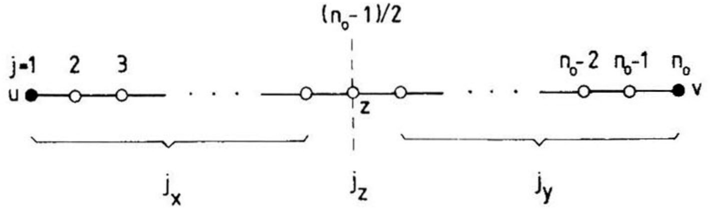


Fig. 5. Division of the closed interval $[j_u, j_v]$ into two equal parts containing pairs of symmetrically located vertices j_y and j_x (equality 19c)

The following relations hold:

$$1 = j_u \leq j_x \leq (j_u + j_v)/2 = (n_0 + 1)/2, \qquad (19a)$$

$$(n_0 + 1)/2 \leq j_y \leq j_v = n_0 , \qquad (19b)$$

$$j_y = j_u + j_v - j_x = n_0 + 1 - j_x \qquad (19c)$$

In case of $n_0 + 1 = 2m = 2j_z$, vertex z exists and it is always the center of the $[j_u, j_v]$ interval. Due to the equality $2j_z - n_0 - 1 = 0$ occurring for $j = j_z$ in eq. (18) vertex z has a zero contribution to ΔW and can be neglected in the summation in this equation. On the other hand, all summands for $j = j_x$ in eq. (18) are negative while those for $j = j_y$ are positive. One can then present eq. (18) in the form:

$$\Delta W^i = W(H^i_1) - W(H^i_2) =$$
$$= (n_{11} - a) \sum_{1 \leq j_x \leq (n_0+1)/2} (n_0 + 1 - 2j_x)(n'_{j_y} - n'_{j_x}) \qquad (20)$$

where a = 0 and 1 for i ≡ b and s, respectively.

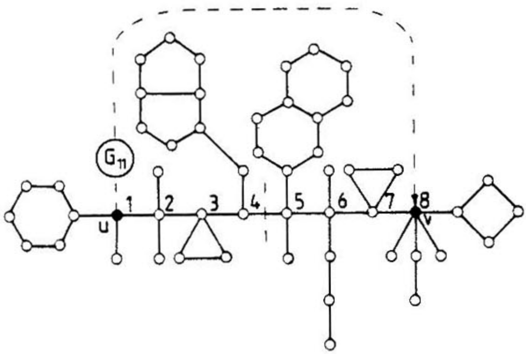


Fig. 6. Illustration of the application of eq. (20). Here $n_0 = 8$, $j_x = 1, 2, 3, 4$; $j_y = 8, 7, 6, 5$; $n'_1 = 7 < n'_8 = 8$, $n'_2 = 2 = n'_7$, $n'_3 = 2 < n'_6 = 4$, $n'_4 = 9 < n'_5 = 11$, $\Delta W = n_{11} . 15 > 0$

Clearly, if for all pairs of symmetrically located vertices (x, y), specified by (19c), $n'_{j_y} \geq n'_{j_x}$ holds, then $\Delta W \geq 0$. The equality for ΔW occurs when $n'_{j_y} = n'_{j_x}$ holds for each of the pairs (x, y) while ΔW will be positive if at least for one such pair $n'_{j_y} > n'_{j_x}$ and if, further, $n'_{j_y} = n'_{j_x}$ for the remaining pairs (x, y); (See Fig. 6 above).

<u>Corollary 1 to Property 3.2</u>. Classes of pairs of transfer chain graphs H^b_1, H^b_2 or H^s_1, H^s_2 exist with the same Wiener number.

The necessary and sufficient condition for the existence of such pairs of graphs is

$$n'_{j_y} = n'_{j_x} \tag{21}$$

It should hold for any pair of vertices x, y in H_1 and H_2 specified by (19c).

The proof follows from the insertion of (21) into (20). The class of isowiener transfer chain graphs is thus defined. Evidently, such a pair of isowiener graphs possesses a certain symmetry of the basic subgraph G_1. A non-trivial example of such pair of isowiener graphs H^b_1 and H^b_2 is shown in Fig. 7.

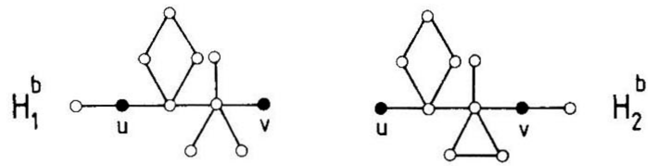


Fig. 7. A pair of transfer chain graphs having the same Wiener number (isowiener graphs)

<u>Corollary 2 to Property 3.2</u>. Classes of pairs of transfer chain graphs H^b_1, H^b_2 or H^s_1, H^s_2 exist for which W is constant.

The classes thus specified are termed isodifferent transfer chain graphs.

The proof of this corollary follows directly from eq. (20) where the difference

$$\Delta n'_j = n'_{y^j} - n'_{x^j} = (n'_{y^j} + k) - (n'_{x^j} + k), \quad k = 0, 1, 2, \ldots$$

can vary depending on k.

This corollary allows to reduce the calculation of the change in the Wiener number in complicated graphs to that of the simplest case in the class, for which $k = 0$. Thus, the graph shown in Fig. 6 can be reduced to the simplest graph in this class having $\Delta n'_1 = 1$, $\Delta n'_2 = 0$, $\Delta n'_3 = 2$, and $n'_4 = 2$ (Fig. 8).

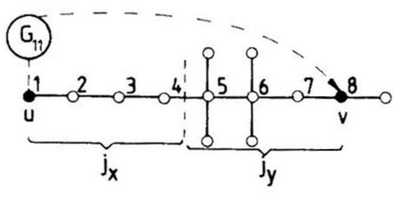


Fig. 8. The simplest graph from the class of isodifferent transfer chain graphs to which the graph from Fig. 6 also belongs

Dealing with Property 3.2 one is tempted to suppose the existence of a more general trend for $W(H_1)$ to decrease (or for $\Delta W = W(H_1) - W(H_2)$ to be positive) upon the fragment transfer when the total number of vertices in the subgraphs G_{ji} attached to the transfer chain vertices located closer to v is larger than that for the respective vertices located closer to u ($\Sigma n'_{j_{y}} > \Sigma n'_{j_{x}}$). Such a trend actually exists but it is not a general property since the position of j also influences greatly ΔW. An example is shown in Fig. 9.

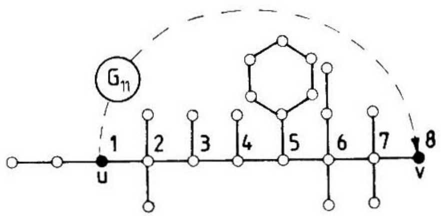


Fig. 9. Illustration of the insufficiency of the inequality $\Sigma n'_{j_{y}} > \Sigma n'_{j_{x}}$ to produce $\Delta W > 0$: $\Delta n'_1 = -2$, $\Delta n'_2 = 0$, $\Delta n'_3 = 2$, $\Delta n'_4 = 5$, $\Sigma n'_{j_{y}} = 11 > \Sigma n'_{j_{x}} = 6$, $\Delta W = -3 < 0$

3.3. Transfer chain graphs with subgraphs attached to the chain by a common edge

Consider now the more complicated case of transfer chain graphs having cycles fused to the transfer chain P_{uv}. When some $G_{jl} \equiv G^{f}_{jl}$ (a monocyclic graph, or a cyclic graph with acyclic branches, or a cyclic graph linked with other cycle(s) by a bridge, spiro-linkage or fusion) eq. (14) cannot be transformed into such a simple equation such as eq. (15). The first and third term in eq. (14) once again cancel each other Only part of the distances, however, from the transition vertices u and v to the vertices $t = t_l \in G^{f}_{jl}$ can be taken via the same fusion vertex j or j'. For the other part the vertices t will be reached from u via j while from v via j', or vice versa:

$$d(ut_i|G_i) = d(uj|G_i) + d(jt_i|G^f_{jl}) \qquad (22a),$$

$$d(vt_i|G_i) = d(vj|G_i) + d(jt_i|G^f_{jl}) \qquad (22b),$$

or

$$d(ut_i|G_i) = d(uj'|G_i) + d(j't_i|G^f_{jl}) \qquad (22c),$$

$$d(vt_i|G_i) = d(vj'|G_i) + d(j't_i|G^f_{jl}) \qquad (22d);$$

and

$$d(ut|G_i) = d(uj|G_i) + d(jt|G^f_{jl}) \qquad (23a)$$

$$d(vt|G_i) = d(vj'|G_i) + d(j't|G^f_{jl}) \qquad (23b)$$

Proceeding from eq. (14) we have

$$\Delta = d(u|G_i) - d(v|G_i) = \sum_j \sum_{l \neq l'} \sum_{t \in G_{jl}} [d(ut|G_i) - d(vt|G_i)] \qquad (24)$$

The second terms in the pairs of equations (22a)/(22b), and (22c)/(22d) mutually cancel upon the subtraction in eq. (24), as was the case of deriving eq. (15) from eq. (14). In subtracting eqs. (23a), and (23b) however, no terms are cancelled. For these reasons the quanitity Δ from eq. (24) will be determined in case of one fused subgraph G^f_{jl} as a sum of two contributions: $\Delta^f = \Delta_1 + \Delta_2$. They account for the distances from the transfer vertices u and v to the fusion vertices j and j', and from the latter to the G^f_{jl} vertices $t \notin P_{uv}$, respectively. Hence,

$$\Delta_1 = \sum_{t \in G^f_{jl}} [d(uj|G_i) + d(uj'|G_i) - d(vj|G_i) - d(vj'|G_i)] \quad (25)$$

Then by substituting $d(uj|G_i) = j-1$, $d(uj'|G_i) = j'-1$, $d(vj|G_i) = n_0 - j$, $d(vj'|G_i) = n_0 - j'$ in eq. (25), as well as by summing over all vertices t in the fused subgraph G_{jl} having the transfer chain vertex j as a fusion point we arrive at the equation:

$$\Delta_1 = \{[(j-1)n^u_j + (j'-1)n^u_{j'}] - [(n_0 - j)n^v_j + (n_0 - j')n^v_{j'}]\} \quad (26)$$

Here n^u_j and n^v_j stand for the number of vertices $t \in G^f_{jl}$,

$t \notin P_{uv}$ whose shortest paths (distances) from u and v pass via j, and similarly, $n^u_{j'}$, and $n^v_{j'}$' stand for the number of those t that are reached via j'. By summing both pairs of these quantities we obtain the total number of vertices in G^f_{jl}, n^f_j which do not belong to the transfer chain. Denoting those which belong to the latter by k = 2, 3, ..., as well as the number of all vertices in G^f_{jl} by N^f_j we have

$$n^u_j + n^u_{j'}{}' = n^v_j + n^v_{j'} = n^f_j = N^f_j - k \qquad (27)$$

Let us further divide each of these four quantities into two contributions: the respective number of vertices in the fused cycle (superscript c) and its branches (superscript b):

$$\begin{aligned} n^u_j + n^v_j &= (n^{u,c}_j + n^{v,c}_j) + (n^{u,b}_j + n^{v,b}_j) \\ n^u_{j'} + n^v_{j'} &= (n^{u,c}_{j'} + n^{v,c}_{j'}) + (n^{u,b}_{j'} + n^{v,b}_{j'}) \end{aligned} \qquad (28)$$

The sum of two first terms in eqs. (28) equals the total number of vertices in the cycle C_n which has k vertices common with the transfer chain P_{uv}:

$$n^{u,c}_j + n^{v,c}_j = n^{u,c}_{j'} + n^{v,c}_{j'} = N_c - k \qquad (29)$$

Denote also the sums of the two second terms in eq. (28) by n^b_j and $n^b_{j'}$ which thus represent the number of all branch vertices the shortest paths (distances) to which from u and v pass via j and via j', respectively:

$$n^{u,b}_j + n^{v,b}_j = n^b_j \quad ; \quad n^{u,b}_{j'} + n^{v,b}_{j'} = n^b_{j'} \qquad (30)$$

Making use of eqs. (27) to (30) we obtain from eq. (26) after some transformations:

$$\Delta_l = [(j+j')(N_c - k) + j n^b_j + j' n^b_{j'} - (n_0 + 1) n^f_j] \qquad (31)$$

Now, adding to and subtracting from eq. (31) the term (j+j'). . $(n^b_j + n^b_{j'})/2$, and taking into account that

$$n^b_j + n^b_{j'} = 2n^{f,b}_j = 2(N^f_j - N_c) \qquad (32),$$

where $n^{f,b}_j$ is the total number of all vertices in G^f_{jl}

which do not belong to C_n, we arrive at the final expression for Δ_1

$$\Delta_1 = (j + j' - n_0 - 1)n^f_j + (j'-j)(n^b_{j'} - n^b_j)/2 \qquad (33)$$

In deriving eq. (33) it was also assumed that $j'>j$ always holds.

Determine now the term Δ_2 emerging from eq. (24) for one cyclic fragment G^f_{jl} fused to the transfer chain. This term accounts for the distances from the two fusion vertices j and j' to those vertices $t \in G^f_{jl}$, $t \notin P_{uv}$, for which $d(jt|G_{jl}) \neq d(j't|G^f_{jl})$. The terms accounting for the remaining vertices $t_1 \in G^f_{jl}$, $t_1 \notin P_{uv}$, for which $d(jt|G_{jl}) = d(j't|G^f_{jl})$ mutually cancel upon subtracting the pairs of eqs. (22a, b) and (22c, d) as follows from eq. (24). Thus one has

$$\Delta_2 = \sum_{t \in G^f_{jl}} [d(jt|G^f_{jl}) - d(j't|G^f_{jl})] \qquad (34)$$

Part of vertices t belong to the fused cycle C_n. They will be denoted by t'. The remaining t vertices belong to the C_n branches which could be acyclic, as well as cyclic ones. Denote this subgraph of G^f_{jl} by $C'_n = G^f_{jl} \setminus C_n$. One obtains thus

$$\begin{aligned} d(jt|G^f_{jl}) &= d(jt'|C_n) + d(t't|C'_n) \\ d(j't|G^f_{jl}) &= d(j't'|C_n) + d(t't|C'_n) \end{aligned} \qquad (35)$$

Clearly, the second terms in eq. (35) vanish upon the subtraction of eq. (34). On the other hand, due to the bilateral symmetry existing in C_n with respect to the fusion vertices j and j', the t' vertices divide into two groups, t'_1 and t'_2 for which the following holds:

$$d(jt'_1|C_n) = d(j't'_2|C_n) \qquad (36)$$

Hence, one obtains

$$\Delta_2 = \sum_{t' \in C_n} [d(jt'|C_n) - d(j't'|C_n)]n_{t'} \qquad (37)$$

where $n_{t'}$ stands for the number of vertices $t \in C'_n$ in the branch(es) attached to vertex $t' \in C_n$. If a second cycle C''_n or more cycles are fused to C_n in vertices $t'_1, t'_2, t'_3, \ldots, t'_i$, then $n_{t'}$ equals the number of those vertices in C''_n that are closer to vertex t'_i than to the other fusion vertices.

By summing eqs. (33) and (37) one arrives thus to the equation for the difference Δ between the distance numbers of the transition vertices u and v which is due to the presence of the subgraph G^f_{ji} fused to the transfer chain P_{uv} :

$$\Delta_f = (j+j'-n_0-1)n^f_j + (j'-j)(n^b_{j'} - n^b_j)/2 + \\ + \sum_{t'\in C_n} [d(jt'|C'_n) - d(j't'|C_n)]n_{t'} \tag{38}$$

Evidently, $\Delta_f = 0$ when no G_{ji} is fused to P_{uv}, since in this case in eq. (38) $j' = j$, $n^f_j = 0$, and $n_{t'} = 0$. On the other hand, when n^f_j is replaced by n^s_j or n^j_b or generally by n'_j, and $j = j'$, the first term in eq. (38) transforms into $(2j-n_0-1)n'_j$. Thus, eq. (18) obtained in the foregoing for transfer chain graphs having only bridged or spiro-linked subgraphs G_{ji} may be regarded as a specific case of the more general equation (39) which comprises also the cases of fused subgraphs:

$$\Delta W^i = W(H^i_1) - W(H^i_2) = (n_{1i} - a)\{\sum_j \sum_{i \neq i'} (j+j'-n_0-1)n'_{ji} + \\ + (j'-j)(n^b_{j'} - n^b_j)/2 + \sum_{j=j_f} \sum_{i=i_f} \sum_{t'} [d(jt'|C_n) - \\ - d(j't'|C_n)n_{t'}]\} \tag{39}$$

where $a = 0$ and 1 for $i = b$ and s, respectively, and $j = j'$, for $G_{ji} \equiv G^b_{ji}$ or G^s_{ji} .

The third term in eq. (39) takes into account also the the possibility several G_{ji} to be fused at the same vertex j, as well as several G_{ji} to be fused at different transfer chain vertices j. The superscript i here refers to the transferred subgraph G_{11}, i.e. eq. (39) is not applicable to transfers of a fused G_{11} but it is of use for transfers of any bridged or spiro-linked G_{11} along the transfer chain P_{uv} which can be bridged or spiro-linked or fused to any simple connected subgraph G_{jl} (acyclic or cyclic one). An example of the application of eq. (39) is given in Fig. 10.

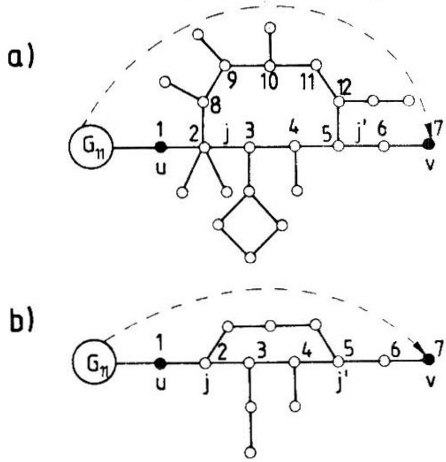


Fig. 10. Illustrations of the application of eq. (39): a) the general case: $n_0 = 7$, $j = 2$, $j' = 5$, $n_2 = 2$, $n^f_2 = 10$, $n_3 = 4$, $n_4 = 1$, $n^b_{j'} = 2+4 = 6$, $n^b_j = 1+3 = 4$, $t' = 9, 10$; $n'_9 = 1$, $n'_{10} = 1$, respectively. For $n_{11} = 1$, $\Delta W = -25$; b) the specific case when eq. (29) reduces to eq. (18): $j = 2$, $j' = 5$, $n_3 = 2$, $n_4 = 1$; for $n_{11} = 1$, $\Delta W = -7$

Eq. (39) reduces to the much simpler equation (18) not only in case the fused subgraphs are missing in the transfer chain graphs H_1 and H_2. Due to symmetry existing for multiple transfer chains, $j' = j$ always holds, though $j \equiv j_f$ (See Fig. 3d). Thus, the simple eq. (18) describes also these rather complicated graphs. Finally, in a slightly modified form eq. (18) can be deduced from eq. (39) for the case when G^f_{jl} is a fused cycle having no other branches than those attached to transfer chain vertices. $n^b_{j'} = n^b_j = n_{l'} = 0$ holds here and one thus obtains

$$\Delta W^{b(s)} = (n_{l1} - a) \sum_j \sum_{l \neq l'} (j + j' - n_0 - 1) n'_j \qquad (40)$$

In eq. (40) $j \neq j'$ is taken only for those j and l for which $G_{jl} = G^f_{jl}$.

Eq. (39) do not possess Property 3.1, specified in this Section for eq. (18), due to the second and third term in it which depends on the subgraph structure. This property is, however, attributed to graphs with multiple transfer chain, as well as for graphs having branches attached only to transfer chain vertices since in these cases the second and third term in (39) vanish. Property 3.2 in general does not hold except in some very specific cases, due to the asymmetry of j and j' with respect to u and v.

4. CONCLUDING REMARKS

In the search for a more general graph-theoretical approach to the description of molecular rearrangements the classes of transfer graphs and transfer chain graphs were introduced in this study. The change in molecular topology occurring upon such rearrangements is evaluated by the change in the metric properties of molecular graphs and, more specifically, by the change in the Wiener number, ΔW. Two types of dependences were obtained for ΔW. The first one given by eqs. (4) to (6) expresses the Wiener number change by means of the distance numbers of the two atoms in the basic molecular fragment to which the transferred fragment is attached before and after the molecular rearrangement. Alternatively, eqs. (18), (20), and the most general eq. (39) present ΔW as a finction of more specific structural parameters: the total number of atoms in the transfer chain, in the transferred fragment, and in all fragments attached to a certain transfer chain atom, the position of the latter being also of importance. In case some fragments are fused to the transfer chain, the change in the Wiener number depends also on the distances between the terminal fusion atoms and those atoms from the fused ring which are not included in the transfer chain, as well as on the total number of atoms in the side-chains attached to each of these ring atoms.

A number of properties and corollaries proved for ΔW showing e.g. that when some fragment is attached to the transfer chain of atoms by a bond (a bridge) or a common atom (a spiro-linkage) it is not its specific topology but its total number of atoms which is of importance. Of special interest are the necessary and sufficient conditions for two graphs to have the same Wiener number (isowiener graphs) or the same difference in their Wiener numbers, i.e. for two molecules to have the same total topological distance or a constant difference of these topological characteristics. Differing from the general conditions for two graphs to have the same metric properties [6], which are based on vertex neighborhood considerations, our conditions are formulated proceeding from exact equations for the Wiener number. They include some more specific parameters such as the total number of vertices in the graph fragments. The conditions for two graphs to have the same Wiener number presented in this work actually refer to cases not covered by the Skorobogatov conditions [6], i.e. they refer to pairs of graphs having different metric properties except the total distance of the graphs. Our approach also allows to simplify the calculation of the difference in the Wiener number of two chemical structures, as well as to generate readily classes of compounds having the same Wiener number or the same difference in this number.

The main importance of the equations derived for the Wiener number is, however, in the possibility they offer for comparison and ordering of chemical structures and first of all of isomeric compounds. Most of isomeric molecules may in principle be interconverted by means of one or a series of molecular rearrangements in which a fragment is transferred from one part of the molecule to another one. Typical rearrangements might be expected to be associated with a regular change in the Wiener number. One thus arrives at a classification of molecular rearrangements on topological basis making use of generalized structural rules. Some examples of such fragment transfers which change the molecular branching are shown in Fig. 11 making use of molecular hydrogen-depleted graphs. The basic part of the molecule in Figs. 11a to 11d is taken to be acyclic, branched monocyclic or fused polycyclic, spiro- and bridged polycyclic, respectively. The fragment R to be transferred might be acyclic or cyclic one.

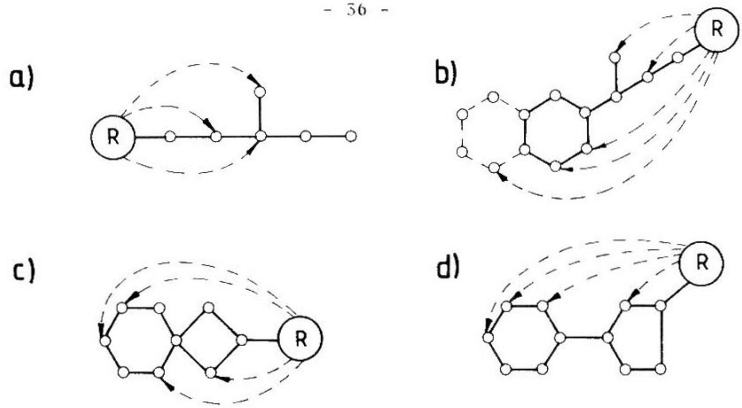


Fig. 11. Illustration of molecular branching treatment. Molecular rearrangements upon which the transfer of fragment R occurs in branched: a) acyclic, b) monocyclic or fused polycyclic, c) spiro-cyclic, and d) bridged polycyclic compounds

A detailed analysis of molecular branching based on the present study will be given in a subsequent publication [40]. Bearing in mind that the same trend is observed [35] in the typical molecular rearrangements for the change in the Wiener number and numerous thermodynamic and other physico-chemical properties one may expect such a generalized treatment of molecular branching to result in a method for finding regularities in molecular properties, as well as for property calculations. Similar treatment of molecular cyclicity will be done later [42] proceeding from eq. (6). An extension of the approach to crystal and polymer modellings is also in progress.

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