

NEW VERTEX INVARIANTS AND TOPOLOGICAL INDICES OF CHEMICAL GRAPHS BASED ON INFORMATION ON DISTANCES

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Abstract

By applying information theory to the set of topological distances from one vertex to all other graph vertices, one obtains four new types of vertex invariants (u_i, v_i, x_i, y_i) which are real numbers (as opposed to integers). They may be combined in many ways to afford new topological indices. One such type leads to indices U, V, X and Y which show no degeneracy for alkanes with up to 15 vertices.

1. Introduction

Topological indices (TIs) continue to be a topic attracting interest, as attested by the publication of numerous reviews, some of which appeared recently [1 – 10].

TIs are numbers associated with graphs, which in turn are models of chemical structures. In order to obtain TIs, one has to start from various graph invariants (e.g. local vertex invariants), and then one has to combine these invariants by means of various operations into a TI for the corresponding graph. Because physico-chemical or biological properties of chemical compounds depend on their structure in various ways, so far no single TI can serve for modeling all properties, and many different TIs have been proposed.

Important drawbacks of TIs are their degeneracy and the derived structure irretrievability: when for more than one structure one obtains the same TI, one calls these TIs degenerate. The search led to new TIs which may be practically non-degenerate and therefore may serve as graph codes [11, 12] or molecular ID numbers [13].

Our efforts were aimed at devising TIs with lower degeneracy than the previously existing ones, and we thus proposed the index J (average distance sum con-

nectivity [14] and, in collaboration with Bulgarian colleagues, two TIs based upon the HOC algorithm (hierarchically ordered extended connectivities) [15]. More recently, we devised two new types of vertex invariants: one type is based on converting the adjacency or distance matrix of a graph into a system of linear equations, from which we devise these new vertex invariants [16] using techniques related to those previously employed by Gollender et al. [17]; the other type is based on the various external fragment topological indices (EFTI) where the fragment is, in turn, each single vertex of the molecular graph [18]. Then we may combine these vertex invariants in a manifold of TIs.

2. Local graph invariants

By removing all hydrogen atoms from the formula of a chemical compound containing covalent bonds, we obtain the hydrogen-depleted graph (or molecular graph) of that compound, whose vertices correspond to non-hydrogen atoms. In the particular case of hydrocarbons, the vertices of the molecular graph denote carbon atoms.

Several simple graph invariants have been known:

(i) *Vertex degrees* are the number of lines (covalent bonds to non-hydrogen atom) meeting at each vertex. In hydrocarbon graphs, vertices can have only degrees 1, 2, 3, or 4. Vertex degrees are sums over rows or columns in the adjacency matrix of the graph.

(ii) *Distance sums* are the sums of topological distances from a given vertex to all other graph vertices. Distances (i.e. topological distances) are the number of lines along the shortest path between two vertices. Distance sums also result as sums over rows and columns in the distance matrix of the graph. Unlike the previous invariant, there is no restriction on the distance sums. Vertices with lowest distance sums form the centroid of the graph.

(iii) *Eccentricities* are the longest topological distances from a given vertex to any other graph vertex. The vertices with largest eccentricities are peripheral ones, whereas those with lowest eccentricities form the graph center.

In acyclic graphs (trees), the center as well as the centroid is a vertex or a pair of adjacent vertices. The graph center and centroid may not coincide.

Figure 1 presents as examples of the above graph invariants the unique identity 4-trees with 7 and 8 vertices. Hydrogen-depleted graphs of acyclic hydrocarbons are called 4-trees because the degree of any point (vertex) is at most 4. Among the alkane molecular graphs, in a single isomer of heptane and a single isomer of octane there are no equivalent carbon atoms; these isomers correspond to identity trees (thus called because they have no symmetry except for the identity operation).

It may be seen that vertex degrees and eccentricities have a high degree of degeneracy: although the above identity trees have no equivalent vertices, there are

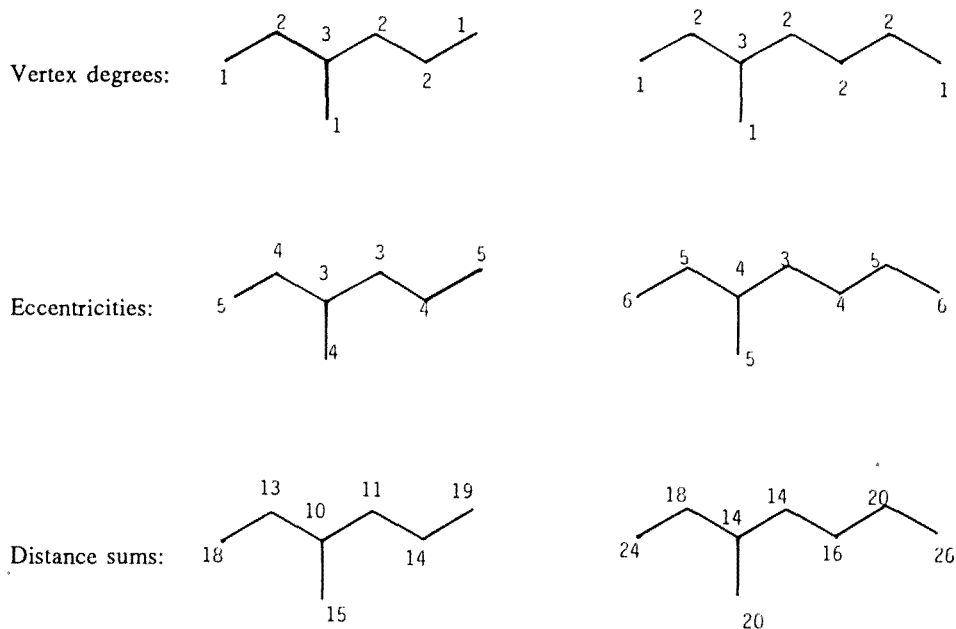


Fig. 1. Identity trees with $n = 7$ and 8 vertices and three of their vertex invariants which are integers.

in the same graph many vertices with the same vertex degree or eccentricities. It is normal that TIs based upon such invariants will also have high degeneracies, even with sophisticated operations for combining vertex invariants into TIs.

On the other hand, the degeneracy of distance sums is lower: none for the vertices of the identity tree with 7 vertices, and only two pairs for the identity tree with 8 vertices. The index J , based on distance sums, has low degeneracy: the smallest 4-trees for which J is degenerate are six pairs of dodecane isomers. Interestingly, there are only three such pairs for 4-trees with $n = 13$; fifty pairs with degenerate J for $n = 14$, but only forty-two for $n = 15$.

(iv) *Weights* in acyclic graphs are the number of lines in the longest branch starting from each vertex. Vertices with the lowest weights constitute the centroid. Weights have higher degeneracy than eccentricities: all endpoints of a given tree have the same weights, equal to q (the number of graph edges).

(v) *External fragment TI invariants*. As already mentioned, one may devise TIs for molecular fragments. If the fragment in question is reduced to a vertex, by this procedure one obtains sets of vertex invariants. These may then be used for devising new TIs [15].

(vi) *Eigenvectors*. As known from quantum chemistry, for any given eigenvalue, the secular equation leads to a set of eigenvectors which are actually graph invariants. However, they have been little used for TIs.

(vii) *Solutions of linear equations obtained from the adjacency or distance matrices.* It was mentioned earlier that on inserting on the main diagonal of the above matrices a set of parameters and by equating each line of these matrices with another set of parameters (which may be identical or different from the previous set), one obtains a set of linear equations whose solutions are new vertex invariants. The parameters may be chemical (e.g. Z , the atomic number of the atom symbolized by the vertex), topological (e.g. vertex degree V , distance sum S), or constants. Three letters indicate the type of invariant: matrix, parameter for main diagonal, and parameter for variable-free term. Thus, invariants AZV lead to a TI which gives an excellent correlation with boiling points of alkanes [16].

These are the main local graph invariants known so far. All but the last two are integers.

Historically, the first TI, which is denoted by w , was proposed by Wiener [19] in 1947, and consisted of the half-sum of all entries in the distance matrix, i.e. of all topological distances in the graph; since the adjacency matrix is symmetrical relative to its main diagonal, each entry is present twice because $d_{ij} = d_{ji}$. Another way of expressing w is by summing the distance vectors of each vertex. As an example, the single vertex of degree three in the unique identity trees with $n = 7$ and 8 (fig. 1) has distance vectors equal to 1, 1, 1, 2, 2, 3 and 1, 1, 1, 2, 2, 3, 4, respectively; the same vectors are expressed more compactly as 1^32^23 and 1^32^234 , respectively, or in general by expressing all distances j with an exponent g_j indicating how many vertices there are at distance j from the given vertex i . Therefore, we have the primitive local vertex invariant s (distance sum) and derived TI:

$$s_i = \sum_j d_{ij} = \sum_j j g_j,$$

$$w = 1/2 \sum_i \sum_j d_{ij} = 1/2 \sum_i s_i.$$

3. New local graph invariants: information on distances

It was seen that distance sums in alkanes may give rise to degeneracy of indices which, like J , are based on invariants of endpoints of each edge (line) in the graph. Taking into account the rapid increase in the number of isomeric alkanes with increasing numbers of carbon atoms, a lumping together of distances by such a crude method as summation, loses information contained in the sequence of distances from a given vertex to all other graph vertices.

We therefore propose new vertex invariants based upon the sequence of topological distances from a given vertex to all other vertices in the graph. We make use of Shannon's formula for the information contained in a sequence of numbers,

which has been extensively used for TIs by Bonchev and Trinajstić [20,21]. The use of information theoretical TIs was reviewed in a book by Bonchev [22].

So far, information theoretic approaches have been extensively used for reduction of degeneracy in TIs at the global level and only very little at the local invariant level. Only Basak, Raychaudhury, Klopman and coworkers [23] have used information theory at the local (vertex) level. Among the TIs defined with the local invariants, the graph distance complexity H^D was found to be least degenerate [23].

In order to replace the crude operation of global distance summation by a more refined approach, we first convert the distance vector into new local (vertex) graph invariants by means of information theory. On applying Shannon's formula to the information content in distance vectors, we obtain (in bits) for each vertex i the mean local information on the magnitude of distances u_i :

$$u_i = - \sum_j \frac{j g_j}{s_i} \log_2 \frac{j}{s_i}$$

and the local information on the magnitude of distances v_i :

$$v_i = s_i \log_2 s_i - u_i.$$

For obtaining more convenient numbers, albeit not derived from rigorous formulas, we also propose related vertex invariants which may be called, by analogy with the previous ones, the extended local information on distance magnitude (x_i), and the mean extended local information on distance magnitude (y_i):

$$x_i = s_i \log_2 s_i - y_i,$$

$$y_i = \sum_j g_j j \log_2 j.$$

We denote by $\log_2 x$ binary logarithms, and by $\log x$ decimal logarithms. Evidently, we have:

$$\log_2 x = \log x \log_2 10 = \log x / \log 2 = 3.322 \log x.$$

In fig. 2, these four local vertex invariants are presented for the two identity trees with 7 and 8 vertices, together with the compact distance vectors from which these invariants were derived. In fig. 3, the vertex invariants are shown for the three identity trees with $n = 9$.

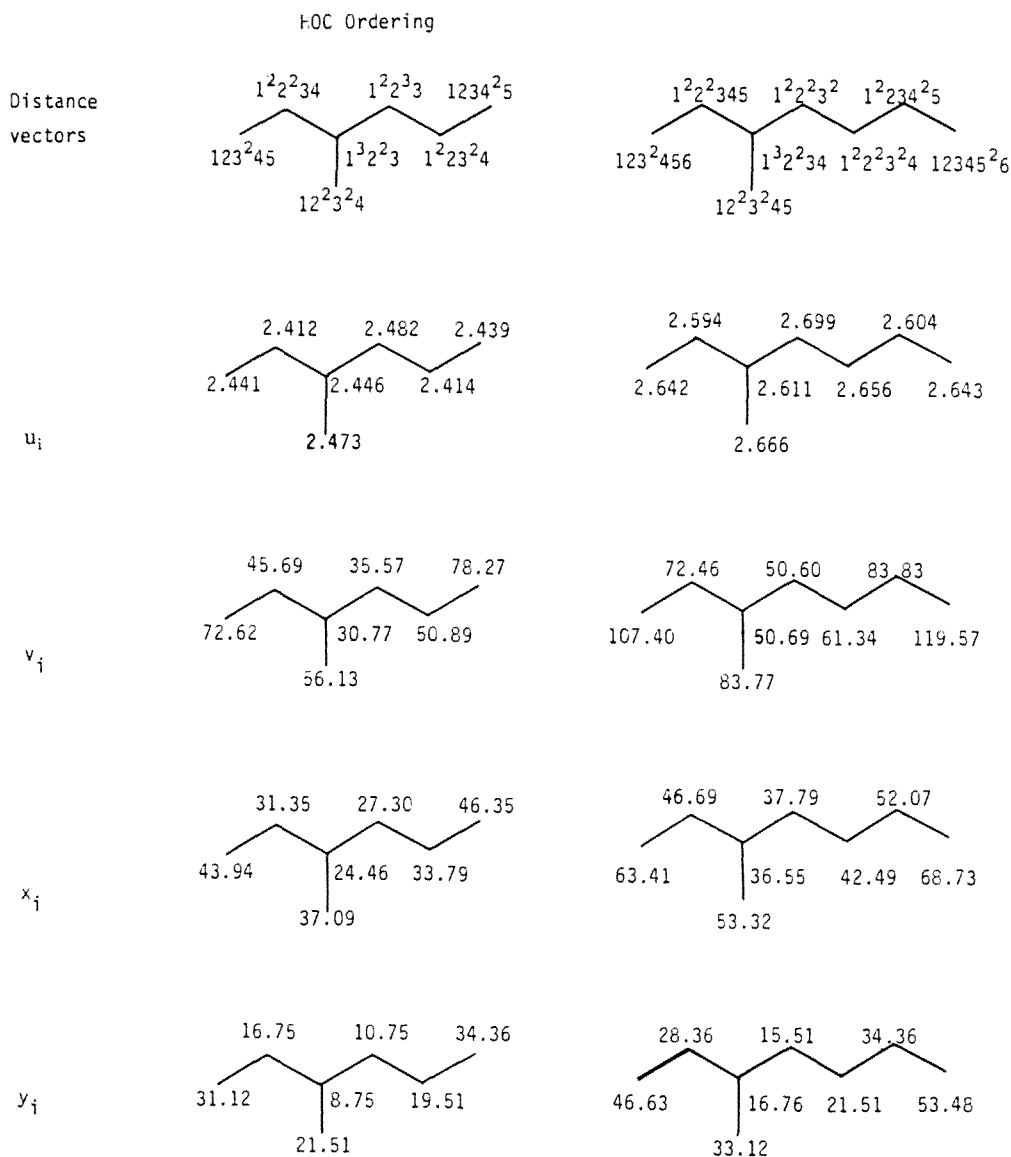


Fig. 2. Identity trees with $n = 7$ and 8 vertices with the distance vectors and the new vertex invariants.

For any 4-tree wherein the distance vectors of the vertices i, j are different, these two vertices will differ in their local vertex invariants u, v, x , and y .

Of course, when the distance vectors are equal by accidental degeneracy, the new vertex invariants will also be equal. Thus, fig. 4 shows that among the cyclic

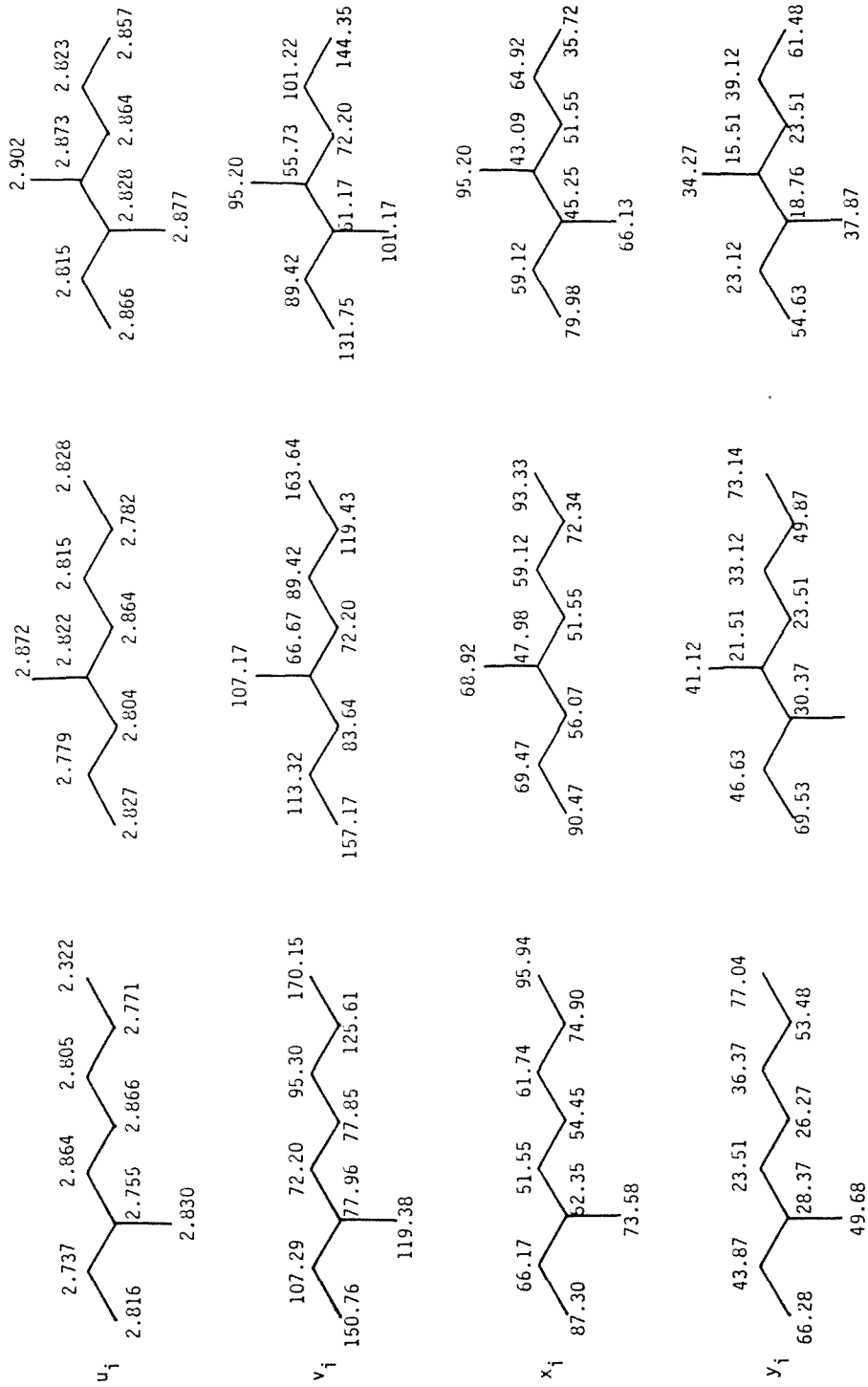
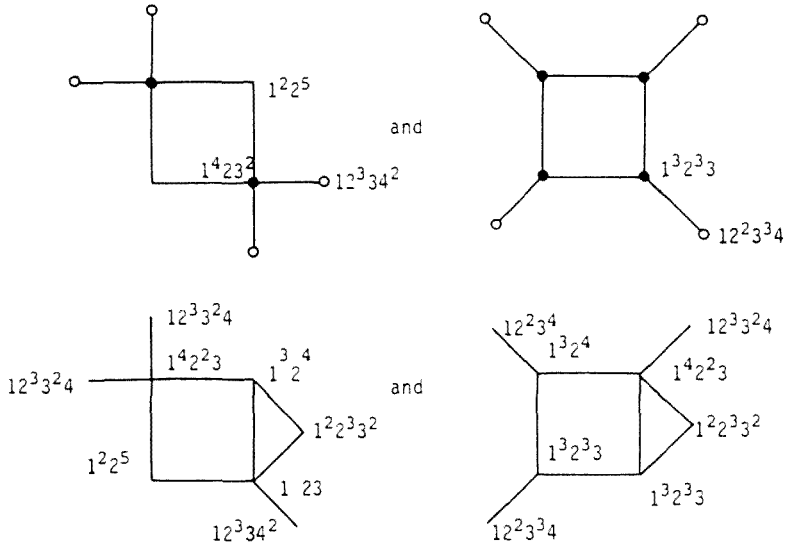


Fig. 3. Local (vertex) invariants for the identity trees with $n = 9$ vertices.

J Degenerate, U, V, X, Y Non-degenerate



J, U, V, X, Y Degenerate :

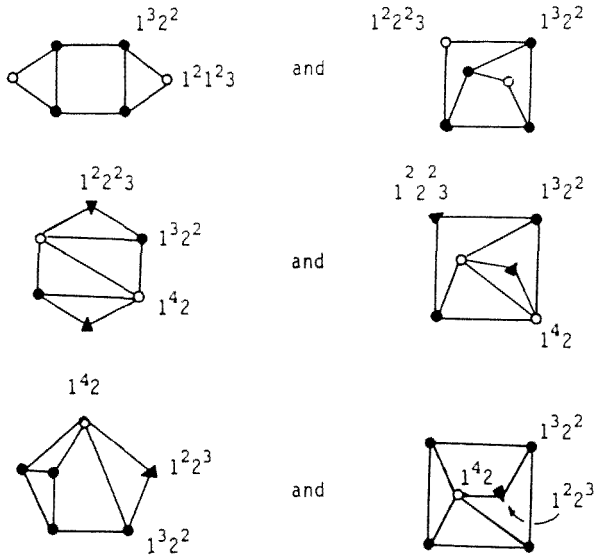


Fig. 4. Examples of cyclic graphs with degenerate J values and non-degenerate U, X, Y, Z values (upper part) and of poly-cyclic graphs with degenerate distance vectors (lower part).

graphs which give degenerate J -values, the monocyclic and bicyclic pair with $n = 8$ have distinct distance vectors, but the tri- and tetracyclic pairs with $n = 7, 6, 5$, respectively, have the same distance vectors, therefore they will give rise to degeneracy.

Alkanes with degenerate distance vectors will be presented in section 5.

4. New topological indices with low degeneracy

From local graph invariants, one may form TIs either by means of simple operations confined to one vertex (e.g. summation of invariants, of their positive or negative powers, etc.), or by more sophisticated operations involving more than one vertex at a time. Such operations are: solving the determinant corresponding to the adjacency matrix, Randić's formula [24] involving products of invariants corresponding to endpoints of each edge, etc.

Summation is likely to lead to high degeneracy; therefore, by analogy with the Randić formula and with the J index, we propose for all new local graph invariants one and the same operation to convert them into new TIs:

$$U = \frac{q}{\mu + 1} \sum (u_i u_j)^{-1/2},$$

$$V = \frac{q}{\mu + 1} \sum (v_i v_j)^{-1/2},$$

$$X = \frac{q}{\mu + 1} \sum (x_i x_j)^{-1/2},$$

$$Y = \frac{q}{\mu + 1} \sum (y_i y_j)^{-1/2}.$$

In all these formulas, summations are over all edges ij ; q and μ denote the number of edges and cycles in the graph, respectively; μ is also called the cyclomatic number of the graph, i.e. $\mu = q + 1 - n$. For trees, $\mu = 0$. Table 1 presents the new indices for all alkanes (4-trees) with $n = 4$ through 8.

Alkane structures are indicated in table 1 not only by the abbreviated name, but also by a code devised recently [11] which gives vertex numberings for vertices adjacent to vertices numbered 3, 4, etc. in increasing order. Table 1 presents alkanes in order of their increasing codes.

Table 1

Topological indices U, V, X and Y for alkane isomers with 4 to 8 carbon atoms

Alkane	Code	Index U	Index V	Index X	Index Y
2-Me-C ₃	11	5.79	–	1.4962	–
C ₄	12	6.06	1.0894	1.3278	3.1324
2, 2-Me ₂ -C ₃	111	8.10	–	1.5310	–
2-Me-C ₄	112	8.36	1.1056	1.3271	3.9156
C ₅	123	8.59	0.8527	1.1751	2.3404
2, 2-Me ₂ -C ₄	1112	11.12	1.2025	1.4095	5.1838
2, 3-Me ₂ -C ₄	1122	11.19	1.0878	1.3399	3.9707
3-Me-C ₅	1123	11.30	0.9548	1.2441	3.1352
2-Me-C ₅	1125	11.43	0.8821	1.2008	2.6360
C ₆	1234	11.60	0.7370	1.0836	1.9580
2, 2, 3-Me ₃ -C ₄	11122	14.38	1.1586	1.4147	4.6563
3, 3-Me ₂ -C ₅	11123	14.44	1.0737	1.3479	4.0959
2, 2-Me ₂ -C ₅	11126	14.65	0.9705	1.2838	3.2097
2, 3-Me ₂ -C ₅	11223	14.55	0.9655	1.2706	3.2531
3-Et-C ₅	11234	14.56	0.8992	1.2095	2.9330
3-Me-C ₆	11235	14.74	0.8260	1.1588	2.4597
2, 4-Me ₂ -C ₅	11255	14.73	0.8752	1.2074	2.6724
2-Me-C ₆	11256	14.91	0.7585	1.1078	2.1070
C ₇	12345	15.01	0.6661	1.1095	1.7297
2, 2, 3, 3-Me ₄ -C ₄	111222	17.97	1.2012	1.4745	4.9756
2, 3, 3-Me ₃ -C ₅	111223	18.08	1.0684	1.3678	4.0126
2, 2, 3-Me ₃ -C ₅	111226	18.16	1.0306	1.3428	3.6700
3-Et-3-Me-C ₅	111234	18.08	1.0191	1.3217	3.7505
3, 3-Me ₂ -C ₆	111236	18.29	0.9310	1.2591	3.0706
2, 2, 4-Me ₃ -C ₅	111266	18.36	0.9319	1.2694	3.0046
2, 2-Me ₂ -C ₆	111267	18.57	0.8255	1.1791	2.4356
2, 3, 4-Me ₃ -C ₅	112233	18.21	0.9638	1.2868	3.2648
3-Et-2-Me-C ₅	112234	18.18	0.9227	1.2445	3.0824
3, 4-Me ₂ -C ₆	112235	18.28	0.8972	1.2276	2.8870
2, 3-Me ₂ -C ₆	112237	18.40	0.8492	1.1898	2.6100
3-Et-C ₆	112345	18.34	0.8144	1.1502	2.4808
2, 4-Me ₂ -C ₆	112355	18.45	0.8202	1.1659	2.4512
4-Me-C ₇	112356	18.52	0.7557	1.1026	2.1650
3-Me-C ₇	112357	18.60	0.7336	1.0855	2.0490
2, 4-Me ₂ -C ₆	112566	18.63	0.7551	1.1116	2.1249
2-Me-C ₇	112567	18.75	0.6803	1.0380	1.8121
C ₈	123456	18.80	0.6170	0.9707	1.5743

5. Discussion of the results

Consideration of table 1 and figs. 3 and 4 leads to the following conclusions:

(1) Local invariants u_i are the smallest and fall within a narrow range. This is evident from the data of the identity trees. Therefore, it is normal that U for a given series of isomeric alkanes also falls within a narrow range. On the other hand, it can be seen that U increases regularly in the alkane series with increasing n . There is no overlap between values of U for alkanes with different n values. Index U , unlike indices J , V , X , and Y , decreases with increasing branching; it is the largest among all these indices.

(2) Local invariants v_i are the largest and fall within a wide range; in a given alkane, the ratio of largest-to-smallest v_i values can be higher than 2. Values of v_i are lower for vertices of higher degree and lower eccentricity. The topological index V is the smallest among all indices discussed here and decreases slightly with increasing n for alkanes. All values of V for the alkanes with $n = 4$ to 10 overlap, but V is a good measure for branching.

(3) Local invariants x_i and y_i behave similarly to v_i , and the TIs denoted by X and Y behave similarly to V . The range of values is largest for y within a given isomer. Among v , x , and y , invariants y are the smallest; among indices V , X , and Y , the largest values appear for Y . The range of values is largest for Y on comparing alkanes with various n and for series of isomeric alkanes. Parameters x and index X are intermediate.

(4) Alkane ordering within isomeric series is an important diagnostic for the value of topological indices. There is complete parallelism in the ordering of alkanes by J , U , V , X , and Y for alkanes with $n = 4, 5$, and 6. For higher n values (we checked up to $n = 15$), there are slight differences between the ordering induced by J and that caused by X , Y , or V : one pair of alkanes is ordered differently by Y for $n = 7$ (2, 2- and 2, 3-dimethylpentane), one pair is ordered differently for $n = 8$ by V (4-methylheptane and 2, 5-dimethylhexane), and three pairs by Y (same as above, and in addition: 2, 2-dimethyl- and 3-ethylhexane; 2, 2, 4-trimethyl- and 3-ethyl-2-methylhexane); for $n = 9$, one pair by Y (2, 2- and 2,5-dimethylheptane), and four pairs by X (2, 2-dimethylheptane and 3-ethylheptane; 2, 3-dimethylheptane and 4-ethylheptane; 2, 2, 5-trimethylhexane and 4-ethyl-2-methylhexane; 2, 2, 4-trimethylhexane and 3-ethyl-4-methylhexane). These are actually minor differences. Bertz [10] has presented a thorough discussion of alkane ordering via topological indices, and it is gratifying to find that indices V , X , and Y give an ordering which is quite close to that determined by J ; this, in turn, is very similar to that induced by Bertz's procedure based on iterating line graphs.

On the other hand, U induces (in reverse order) a markedly different ordering of the higher alkanes, considering e.g. 2, 2-dimethylheptane as less branched than 2, 3-dimethyl- and 3-ethylpentane. In fig. 5, we present the alkanes with $n = 7$ to

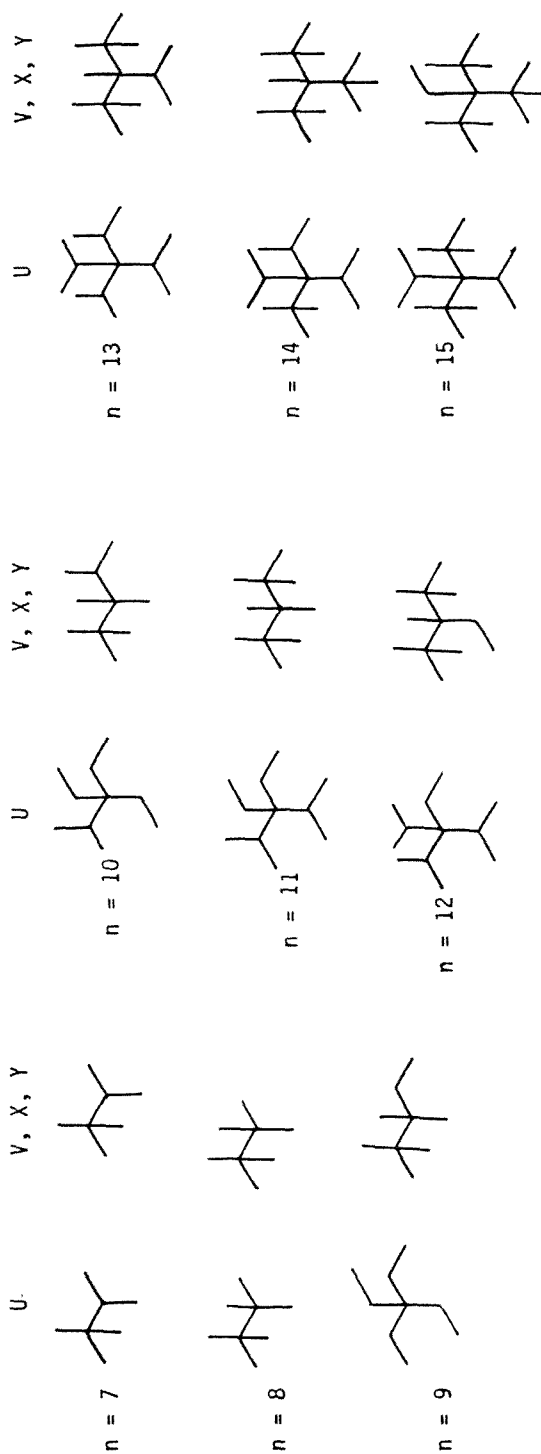


Fig. 5. 4-trees with the lowest value for index U and highest value for indices V , X , and Y among all 4-trees of order n .

15 vertices, for which the lowest value was obtained for index U and the highest values were obtained for indices V , X , and Y within a class of isomeric alkanes. These graphs represent the "most branched" 4-trees with a given n according to the new indices.

Unlike index J , which is degenerate for 4-trees with $n \geq 12$, we found no degeneracy for 4-trees with $n \leq 15$ for any of the indices U , V , X , and Y .

By observing the values of these TIs for linear alkanes with increasing number of vertices (table 2), it is evident that U increases fast towards infinity, whereas V , X , and Y decrease slowly. Even if in calculating U we would omit the factor q (i.e. for U/q), the resulting values increase monotonously for linear alkanes with increasing n .

Table 2

Values of various TIs for n -alkanes with increasing number n of carbon atoms

n \ TI	Index J	Index U	Index V	Index X	Index Y
5	2.1906	8.59	0.8526	1.1751	2.3404
7	2.4475	15.02	0.6661	1.0195	1.7297
9	2.5951	23.01	0.5804	0.9315	1.4601
21	2.9014	99.11	0.4121	0.7181	0.9636
101	3.0909	—	—	0.4894	0.5837
201	3.1160	5475.49	0.2289	0.4263	0.4952
501	3.1313	—	—	0.3630	0.4113
1001	3.1365	103929.69	0.1719	0.3258	0.3641
2001	3.1390	—	—	0.2954	0.3265

Local invariants of complete graphs (regular graphs of degree $n - 1$, in which each vertex is connected to every other vertex) all of whose distances are equal to one are, of course, zero; hence, their TIs U , V , X , and Y are meaningless. For two 4-trees, there exist vertices whose distance vectors are composed of distances 1: the central vertex in isobutane and neopentane; for these alkanes, V and Y are equal to zero.

It should be noted that for trees, the distance vectors are synonymous with the path counts used extensively by Randić [25].

Degenerate local graph invariants for 4-trees modeling alkanes are encountered starting with one isomer of decane, namely 4-ethyloctane (an identity tree), in which the two indicated vertices in fig. 6 have identical distance vectors.

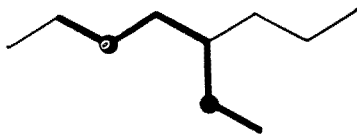


Fig. 6. Graph of 4-ethyloctane which has for the marked vertices degenerate distance vectors $1^22^23^24^25$.

By attaching to each of these indicated vertices one or two identical side chains, or by attaching to symmetrically placed vertices on the thick line such side chains, one obtains 4-trees with 12, 14, 16, . . . vertices, having pairwise identical distance vectors for non-equivalent vertices in the same graph. In fig. 7, we present the three resulting 4-trees with 12 vertices (the last one is an identity tree), and the nine derived 4-trees with 14 vertices (two of the last three are identity trees); indicated vertices have degenerate distance vectors.

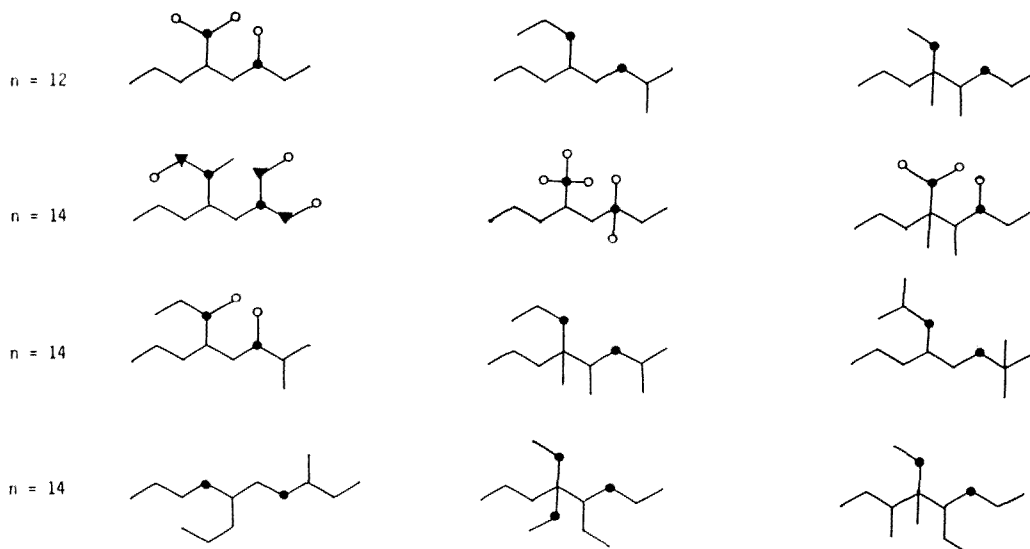


Fig. 7. 4-trees with degenerate distance vectors with 14 and 15 vertices. Indicated nonequivalent vertices have the same distance vector.

Using the mathematical formulas outlined on the basis of Shannon's information theory, these nonequivalent vertices with identical distance vectors will be assigned identical (degenerate) local invariants. Nevertheless, on combining local vertex invariants with topological indices U , V , X , and Y , these indices are found to have no degeneracy up to, and including, $C_{15}H_{32}$. It will be interesting to find the smallest 4-trees with degenerate indices U , V , X , or Y .

6. Conclusions

So far, information theory has been applied mainly for reducing degeneracy of TIs at the global level and very little at the local level. Here, we have proposed an application at the local (vertex) level, and we use the local invariants for new TIs which are less degenerate than the index J . One of these TIs, namely U , has a small range of variation for isomeric families of alkanes, and increases rapidly with increasing number of carbon atoms in the alkane; therefore, it is different from

the other three new TIs, namely V , X , and Y , which behave fairly similarly to the index J , but with more pronounced discriminating ability than J among isomeric alkanes: no degeneracy was found for any of the tested alkanes with 4 to 15 carbon atoms. The closest similarity in alkane ordering is between J and V , as proved by the fact that from 4347 isomeric alkanes $C_{15}H_{32}$, ordered according to J or V values, very few inversions exist (less than 1%).

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- [26] *Note added in proof*: 4-Trees with 18 vertices may be U , V , X and Y degenerate, as will be shown: O. Ivanciuc, T.-S. Balaban and A.T. Balaban, *J. Math. Chem.*, submitted.