CROATICA CHEMICA ACTA CCACAA **69** (3) 1117–1148 (1996)

ISSN-0011-1643
CCA–2364
Original Scientific Paper

Combinatorial Self-Similarity*

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Received May 11, 1995, revised September 26, 1995, accepted October 3, 1995

Combinatorial (or numerical) self-similarity is an apparently new concept, introduced here in an attempt to explain the similarity of properties of the members of a homologous series that are not (geometrically) self-similar and whence are not (deterministic) fractals. The term is defined in the following steps:

a) Select a numerical invariant, φ, characteristic of the member of the series

b) Partition this property, φ, into a finite number of parts through a prescribed algorithm

c) Members are described so as to be combinatorially self-similar (or to represent a »numerical« fractal) if the limits of the ratios of φ of two successive members at infinite stages of homologation are equal for all parts, and equal the corresponding limit for the total property. In the present work, φ is taken to be the Kekulé count, K, when dealing with benzenoid systems and the topological index, Z, (H. Hosoya, *Bull. Chem. Soc. Japan* **44** (1971) 2332) when dealing with saturated hydrocarbons. The previously described equivalence relation, l, [S. El-Basil, *J. Chem. Soc. Faraday Trans.* **89** (1993) 909; *J. Mol. Struct. (Theochem)* **288** (1993) 67; *J. Math. Chem.* **14** (1993) 305; *J. Mol. Struct. (Theochem)* **313** (1994) 237; *J. Chem. Soc. Faraday Trans.* **90** (1994) 2201], is used to partition K when the number of *terminal* hexagons remains constant throughout the series; otherwise the method of Klein and Seitz [D. J. Klein and W. A. Seitz, *J. Mol. Struct. (Theochem)* **169** (1988) 167] is used. For alkanes, an appropriate recurrence relation is used to partition the Z values. It was found that φ for any homologous series of *unbranched* benzenoids, alkanes, Clar graphs, rook broads and King polyominos are all scaled by the golden mean, τ = 1,618033989, while homologous

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* Dedicated to Professor Sven J. Cyvin for his elegant applications of combinatorics in chemistry proper.

series of other types of benzenoids also represent »numerical« fractals, but the characteristic scaling factors depend on the closed form expressions of their K values. In all cases, self-similarities were manifested by expressing the ratios of adjacent φ's in the form of continued fractions, which in some cases led to *exact* self-similarity but in most cases self-similarity was only approximate.

»There is no escape from these foxy rabbits!«
Manfred Shroeder

1. INTRODUCTION

While Euclidean geometry classically considers lines, circles, spheres, etc., geomtry of fractals deals with *algorithms* which can generate shapes and structures that may be used to model and analyze complex forms found in nature.[1] It is interesting to observe that the classical fractals,[2] such as the Cantor dust,[3] the Sierpinski gasket[3] and the Koch curve,[3] have been known for about a century; however, the term *self-similarity* is only some twenty years old![4] When a larger lattice shows exactly the same cluster distribution as a smaller one, it is described as being self similar or »free of scale«. It is not true, however, that if a structure is self similar, then it is also a fractal. For example, a line segment, or a square or a cube can be broken into smaller copies which are obtained by similarity transformations, yet these structures are not fractals.[5] The latter must have a *characteristic* reduction (scaling) factor which is related to the number of scaled down self-similar pieces into which the object is divided.[5] This latter parameter enters into the definition of the *fractal dimension*,[6] D, as given by Eq. (1).

$$D = \frac{\text{Log(number of scaled down pieces)}}{\text{Log(magnification factor)}} \tag{1}$$

As an illustration, we consider a fractal object in Figure 1 whose initial stage is an equilateral triangle to be divided into five triangles, each being scaled down by a factor of 2.5. The fractal dimension between any two successive stages of the construction is given by:

$$D = \frac{\text{Log } 5}{\text{Log } 2.5} = \frac{\text{Log } (5)^2}{\text{Log } (2.5)^2} = \frac{\text{Log } (5)^3}{\text{Log } (2.5)^3} = \ldots = 1.756470797 \tag{2}$$

The above value (which happens to be less than the dimension of a square but more than that of a line)[7] is certainly *not* arbitrary. That is why the various stages of the object shown in Figure 1 form a fractal, while the corre-

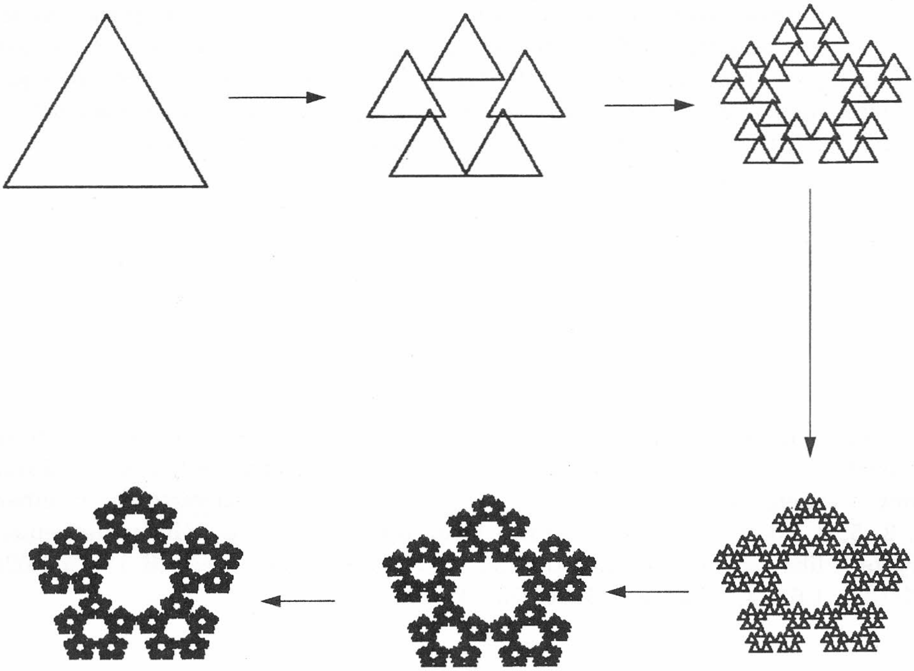


Figure 1. A fractal[3] whose intial stage is an equilateral triangle. At each stage, each triangle is divided into five ones, each being scaled down by a factor of 2.5.

sponding ones of a cube, say, although self-similar, do not define a fractal because its reduction factor is arbitrary.

Naturally, fractals have added a new dimension to the *Chemical Graph Theory.*[8–14] In the present work, homologous series of chemical structures (such as benzenoid hydrocarbons, alkanes and cycloalkanes) are further reconsidered. The question to be posed is the following: An arbitrary homologous sequence of (chemical) structures is *not* self-similar from the viewpoint of the fractal geometry (unless the series is deterministic, such as those of Klein *et al.*),[9] yet the physico chemical properties of the individual members are indeed very similar. Take for instance the class of unbranched benzenoid hydrocarbons of the polyphenanthrenoid polyphenes[15] (commonly called zig-zag polyacenes)[16] which represents the simplest form of quasicrystal – like benzenoid systems: Keeping the number of hexagons constant, the members of this particular series have the highest Kekulé counts, shortest wavelength for their UV spectra, largest sum of non-adjacent numbers and highest resonance energy.[12] This similarity in properties is *not* a result of geometrical (self-) similarity of their skeleton (molecular) graphs. The goal here it then to look for other types of self-similarities, such as numerical or combinatorial ones, which are likely to be generated from a given graph invariant of these structures. Consider for example the golden mean,[12] τ, (a ratio of cosmopolitan occurrence!): in its decimal notation, 1.618033989... it is just an irrational number like many others but if presented as a *continued fraction*,[17] its interesting number-theoretic properties get revealed, *viz.*

$$\tau = 1 + \cfrac{1}{1 + \cfrac{1}{1 + \cfrac{1}{1 + \dots}}}$$

$$= [1;\ 1,\ 1,\ 1,\ 1,\ .....]$$

In fact, it is said that τ is a fixed point in the hyperbolic (Gaussian) map. Indeed, its continued – fraction form Eq. (3) *is* geometrically self-similar! Now, a »chemical graph« theoretician will immediately recognize the numbers 2, 3, 5, 8, 13, 21, 34, 55, ... as the Kekulé counts of the unbranched.quasicrystal – like benzenoids,[12] and that 3/2 = 1.5; 5/3 = 1.6667; 8/5 = 1.6; 13/8 = 1.625; 21/13 = 1.6154; 34/21 = 1.6190; 55/34 = 1.6176; ......

approach

$$\tau = \lim_{n - \infty} \frac{K(B_o(n+1))}{K(B_o(n))} \tag{4}$$

where $K(B_o(n))$ = Kekulé count of an unbranched quasicrystal – like benzenoid which contains n hexagons.

Now, one might say that the (*hidden*) geometrical self-similarity of the series: benzene, naphthalene, phenanthrene, chrysene, picene,[16] .... *etc.* »*hides*« in their Kekulé counts and will be clear when the latter are expressed as *continued fractions*:

$$1 + \cfrac{1}{1 + \cfrac{1}{1 + \cfrac{1}{1 + \dots}}}$$

2. NUMERICAL (COMBINATORIAL) SELF-SIMILARITY POSTULATE

Since arbitrary homologous sequences of (chemical) structures, unless deteministic,[9] are *not*, generally, geometrically self-similar, an attempt is made here to look for other types of self-similarities, such as numerical or combinatorial ones. Namely, let the members of a homologous sequence possess a set of graph invariants, and suppose one selects a combinatorial property φ. Then, for the individual structures of a homologous series, one has the sequence $\phi\{S_1\}$, $\varphi\{S_2\}$, $\varphi\{S_3\}$, ... where S_i is the i^{th} structure in the series. Now, a function $\mathscr{F}$ is selected, which *partitions* $\varphi\{S_i\}$, i = 1, 2, ... into j parts, *viz.*

$$\begin{aligned}\mathscr{F}\varphi\{\mathrm{S}_1\} &= \mathrm{P}_{1,1} + \mathrm{P}_{1,2} + \mathrm{P}_{1,3} + \ldots + \mathrm{P}_{1j}\\ \mathscr{F}\varphi\{\mathrm{S}_2\} &= \mathrm{P}_{2,1} + \mathrm{P}_{2,2} + \mathrm{P}_{2,3} + \ldots + \mathrm{P}_{2j}\\ &\vdots\\ \mathscr{F}\varphi\{\mathrm{S}_i\} &= \mathrm{P}_{i,1} + \mathrm{P}_{i,2} + \mathrm{P}_{i,3} + \ldots + \mathrm{P}_{i,j}\end{aligned} \tag{5}$$

where, in general, the sum of the individual partitions, $\sum_{k=1}^{j} \mathrm{P}_{ik} = \varphi\{\mathrm{S}_i\}$ for the i^{th} member.

Then, the homologous series S_1, S_2, ... is said (*here*) to have a combinatorial self-similarity if

$$\lim_{i\to\infty} \frac{\varphi\{\mathrm{S}_{i+1}\}}{\varphi\{\mathrm{S}_i\}} = \lim_{i\to\infty} \frac{\varphi\{\mathrm{P}_{i+1,j}\}}{\varphi\{\mathrm{P}_{i,j}\}} \tag{6}$$

for all values of j, that is, if the individual limits of *all* partitions are mutually identical *and* equal the limit of the total property.

It is natural when dealing with a homologous series of benzenoid hydrocarbons to think of K, the number of Kekulé structures,[18] as the »most promising« candidate of such a combinatorial quantity, namely in Eq. (5) one has:

$$\varphi\{\mathrm{S}_i\} \equiv K\{\mathrm{B}_i\} \equiv K_i \tag{7}$$

where B_i is an »i^{th}« benzenoid hydrocarbon in a given homologous series. Function $\mathscr{F}$ will then be our (previously introduced)[13] equivalence relation l, which is based on the conjugated circuit model of Randić.[19] (section 3.1).

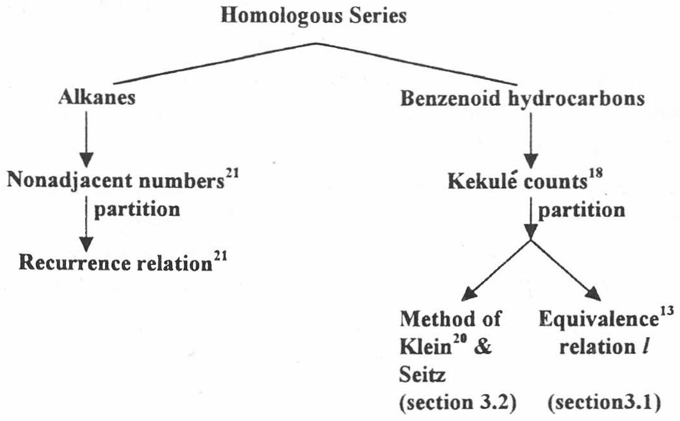


Chart 1.

Sometimes the function l is not convenient to work with, in which cases we use the method of Klein and Seitz[20] to partition the individual K values (section 3.2). When dealing with alkanes (and possibly cycloalkanes), the other property $\varphi\{S_i\}$ (of Eq. (5)) will be taken as the sum of Hosoya's nonadjacent numbers[21] and our (partition) function is the appropriate recurrence relation. (Section 3.4). The following chart 1 outlines the main themes of this research.

3. RESULTS

3.1. Homologous Series of Benzenoid Hydrocarbons with a Constant Number of Terminal Hexagons

Figure 2. shows the representative members of this type of homologous series and some auxilliary systems.

Appendix 1 lists explicit formulas for the K values[18] of these series. For the six types of these hydrocarbons, $P(n)$, $P_1(n)$, $P_2(n)$, $Q(n)$, $Q_1(n)$, $Q_2(n)$, the equivalence relation l, previously introduced, is used as the partition function of the Kekulé counts.

The algorithm of the l equivalence relation[13] is outlined below:

a) Label the terminal hexagons of B by t_1, t_2, ..., t_T. If $T = 0$, find some other method to partition K.

b) Place circles in all terminal hexagons, each circle indicates a resonant sextet, *i.e.*, an R_1 conjugated circuit[19] which contains 6 pi-electrons, and prune out all such terminal hexagons (with their single bonds) from the rest of the benzenoid skeleton.

c) Find the K value of the remaining (resonating) part. If such $K = 0$, the above assignment of terminal R_1's are not allowed and one may look for other distributions.

d) The above procedure is repeated for all terminal hexagons less 1 and so on until all terminal hexagons are exhausted.
Each assignment generates a *subset* of Kekulé structures which share the property of having the same number of terminal R_1's.

e) The individual subsets are nothing else but *equivalence* classes[22] of the original *Kekulé space*, $\kappa(\mathrm{B}) \equiv \kappa$, *i.e.*.

$$l\kappa = \sum_{k=1}^{i} \alpha_k K_k \qquad (8)$$

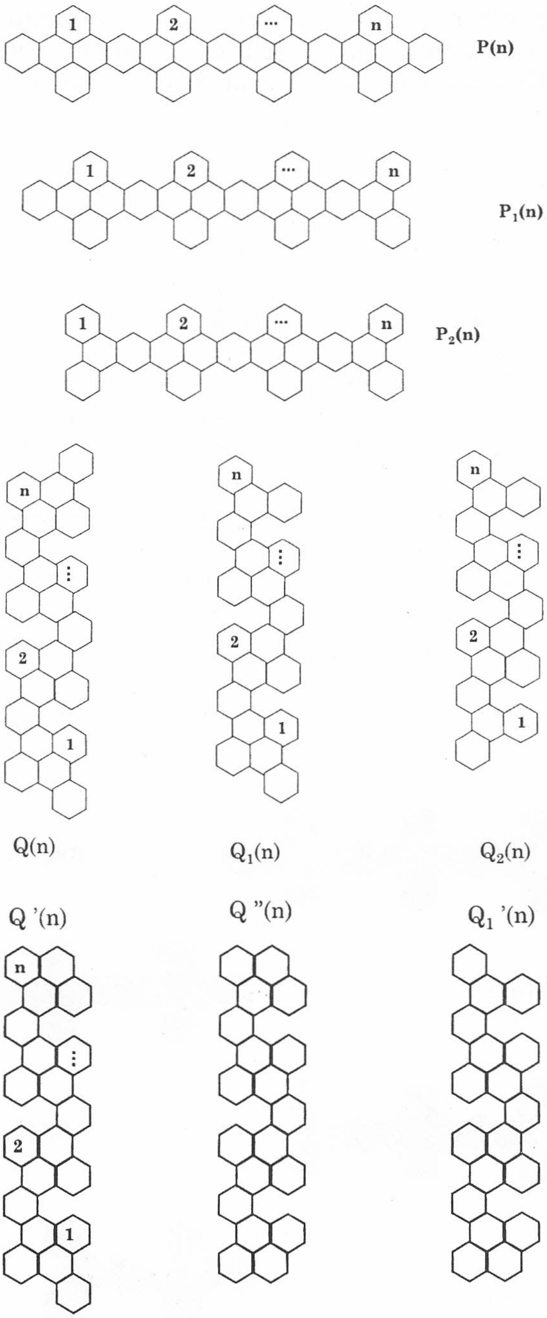


Figure 2. Homologous series of benzenoid hydrocarbons which possess a constant number of terminal hexagons throughout the series. Boldly drawn systems are the auxilliary ones.

where K_k is the number of Kekulé structures of the resonating part of B generated by a given assignment of R_1's to the terminal hexagons, and α_k is the multiplicity of this equivalence class of Kekulé structures. It has been shown[12,13] that

$$\alpha_k = 2^r \tag{9}$$

wherte r = # terminal R_1's in the k^{th} equivalence class.

A convenient (pictorial) way to invisage Eq. (8) is to imagine the individual Kekulé structures in a given equivalence class as »vertices«, and make use of the »spin – like« property of the resonant sextet types[23] (proper ≡ + 1/2 and improper ≡ –1/2)[23] by connecting any two »vertices« that possess »opposite spins«! In this way, a t-dimensional cluster (cube) results *at each* assignment of terminal R_1's. Naturally, when $r = 0$, the corresponding equivalence class contains only vertices (*o*-dimensional cube) while $r = 1$ leads to a collection of »edges« (1-cube), $r = 2$ generates squares (2-cubes), $r = 3$ yields (3-cubes), $r = 4$ forms tesseracts (4-cubes) and so on.[12,13] Interestingly, these hypercubes have found chemical, biochemical and computational applications.[12,13] One is then »tempted« to describe our l relation as a form of »*cluster expansion of Kekulé space*«.[24]

Figure 3 pictorially demonstrates this type of *cluster expasion of Kekulé spaces* through application of the equivalence relation l by showing all al-

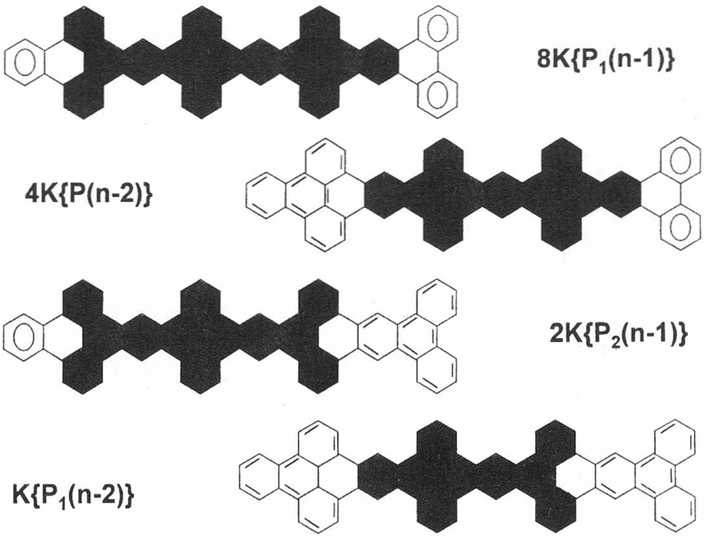
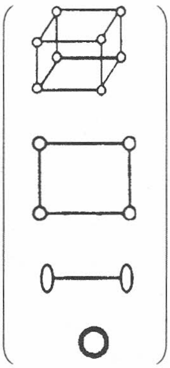


Figure 3. The four types of Kekulé structures of the $P_1(n)$ series (C. F. Figure 2), generated by the equivalence relation[13] l on the Kekulé space $\kappa(P_1(n))$.

lowed assignments of terminal R_1's. Where the shaded graphs are the resonating parts, in all cases the multiplicities of all these subsets will always be integral powers of 2 (possibly times a symmetry factor). No other multiplicity is possible. This is a direct result of the »spin-like« property[23] of the aromatic sextet (*c.f.* Eq. (9)).

It turns out that all benzenoids shown in Figure 2 are combinatorially self-similar (from the point of view of our defining Eq. (6) where the partition function $\mathscr{F}$ is the equivalence relation l). Furthermore, this type of numerical self-similarity is associated in each case with a *characteristic* scaling factor (given by the limit defined in Eq. 6) and whence the Kekulé *counts* possess the structure of a *fractal*. To demonstrate this form of self-similarity, we compute K values of the $P_1(n)$ series shown in Figure 2 as portrayed in the following (convenient)[25] matrix form:

$$\begin{pmatrix} 89 & 20 & 40 & 9 \\ 881 & 198 & 396 & 89 \\ 8721 & 1960 & 3920 & 881 \\ 86329 & 19402 & 38804 & 8721 \\ \vdots & \vdots & \vdots & \vdots \\ P_1(n-1) & P(n-2) & P_2(n-1) & P_1(n-2) \end{pmatrix}$$



$$= \begin{pmatrix} 881 \\ 8721 \\ 86329 \\ 854569 \\ \vdots \\ P_1(n) \end{pmatrix} \qquad (10)$$

where $P_1(n) \equiv K\{P_1(n)\}$ and so on and a t-cube $\equiv 2^t$. For example, the leading member has a value of $K = 881$ which is partitioned as $89 \times 8 + 20 \times 4 + 40 \times 2 + 9 \times 1$. Now, we find the limits (of Eq. 6), we form a matrix; the i^{th} element of its j^{th} column is given by

$$\frac{a_{i+1},i}{a_{i},j}$$

i.e. we simply list the ratio of each two successive elements in each column. One obtains the following result:

$$\begin{pmatrix} 9.8989 & 9.9000 & 9.9000 & 9.8989 \\ 9.8989 & 9.8989 & 9.8989 & 9.8989 \\ 9.8989 & 9.8989 & 9.8989 & 9.8989 \end{pmatrix} = \begin{pmatrix} 9.8989 \\ 9.8989 \\ 9.8989 \end{pmatrix} \qquad (11)$$

This result is general for all benzenoid homologous series of Figure 2. In Appendix 1, the closed form expressions for K values lead to the following approximate formulas for very large values of n:

$$\mathrm{P}(n) \approx 0.204\ (9.8989)^{n+1} \tag{12}$$

$$\mathrm{P}_1(n) \approx 0.908\ (9.8989)^{n+1} \tag{13}$$

$$\mathrm{P}_2(n) \approx 0.408\ (9.8989)^{n+1} \tag{14}$$

$$\mathrm{Q}(n) \approx 0.196\ (10.0990)^{n+1} \tag{15}$$

$$\mathrm{Q}_1(n) \approx 0.892\ (10.0990)^{n} \tag{16}$$

$$\mathrm{Q}_2(n) \approx 0.402\ (10.0990)^{n} \tag{17}$$

where $\mathrm{P}(n) \equiv K\{\mathrm{P}(n)\}$ and so on.

Table I lists the multiplicities of the various clusters (t-cubes) generated upon the effect of the l operation on the Kekulé space of the homologous series of Figure 2. These values are the Kekulé counts of the »resonating portion« of the respective molecular graphs upon a given assignment of terminal R_1 circuits. For both $\mathrm{Q}_1(n)$ and $\mathrm{Q}_2(n)$ systems Kekulé counts of the auxilliary systems $\mathrm{Q}'(n)$, $\mathrm{Q}''(n)$ and $\mathrm{Q}_1'(n)$, (Figure 2, Appendix 2) are used.

Now, it is easy to see the combinatorial self-similarity as defined by Eq. (6) using Table I. For example, for the $\mathrm{P}_1(n)$ series, one may write the following limits, using Eqs. (12)–(14):

$$\lim_{n-\infty}\frac{K\{\mathrm{P}_1(n+1)\}}{K\{\mathrm{P}_1(\mathrm{n})\}} = \lim_{\mathrm{n}-\infty}\frac{K\{\mathrm{P}_2(n+1)\}}{K\{\mathrm{P}_2(\mathrm{n})\}} = \lim_{n-\infty}\frac{K\{\mathrm{P}(n+1)\}}{\mathrm{K}\{\mathrm{P}(n)\}} = 9.8989 \tag{18}$$

where one asumes that when

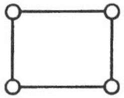
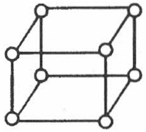
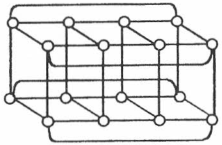
$$n \to \infty\,;\quad K\{\mathrm{P}_1(n-j)\} \to K\{\mathrm{P}(n)\}\ etc... \tag{19}$$

(j = 1, 2, *c.f.* Table I).

A similar arithmetic leads to this type of self-similarity for the $\mathrm{Q}(n)$, $\mathrm{Q}_1(n)$ and $\mathrm{Q}_2(n)$ series. In thise cases, the corresponding limit = 10.0990 (*c.f.* Eqs. (15)–(17)).

TABLE I

Multiplicities of the t-cube upon the »cluster-expansion« of Kekulé spaces of benzenoid systems shown in Figure 2. The symbol of a given homologous series stands for its Kekulé count.

					
$P(n)$	$P(n-2)$	$2^*P_1(n-1)$	$P_2(n)$		
$P_1(n)$	$P_1(n-2)$	$P_2(n-1)$	$P(n-2)$	$P_1(n-1)$	
$P_2(n)$	$P_2(n-2)$		$2^*P_1(n-2)$		$P(n-2)$
$Q(n)$	$Q(n-2)$	$2^*Q_1(n-1)$	$Q_2(n)$		
$Q_1(n)$	$1/2\ [Q(n-2)+Q(n-3)]$	$1/4\ [Q(n-1)-Q(n-3)]$	$Q(n-2)$	$Q_1(n-1)$	
$Q_2(n)$	$1/4\ [Q(n-2)+Q(n-3)+Q(n-4)]$		$Q(n-2)+Q(n-3)$		$Q(n-2)$

* Symmetry factor

3.2. Homologous Series of Benzenoid Hydrocarbons with a Varying Number of Terminal Hexagons

Figure 4 shows seven such benzenoid families. The notations are those adopted in Ref. 18. It is not convenient to use the equivalence relation l in these cases to study the combinatorical properties and self-similarities in their Kekulé counts. This is because the number of terminal hexagons increases as one moves up in the homologous series. A suitable and illustrative method for investigating fractal properties of their Kekulé counts is due to Klein and Seitz.[20] These authors use a pictorial recursion by writing a sequence of numbers inside the hexagons (starting at either end of the chain) where a 2 is assigned to terminal rings and a 1 outside each such ring so that the number at a given position is the sum of the two *preceding ones*. The desired Kekulé count is the sum of two products: the product of numbers around a given branched hexagon plus the product of numbers immediately following another phenanthrenoid ring.[15] (Terminal rings are taken to be phenanthrenoid). Thence, these two products represent a given *partition* of K. Appendix 3 lists closed forms for the Kekulé counts of the seven series of Figure 4. The method of Klein and Seitz is best illustrated on the

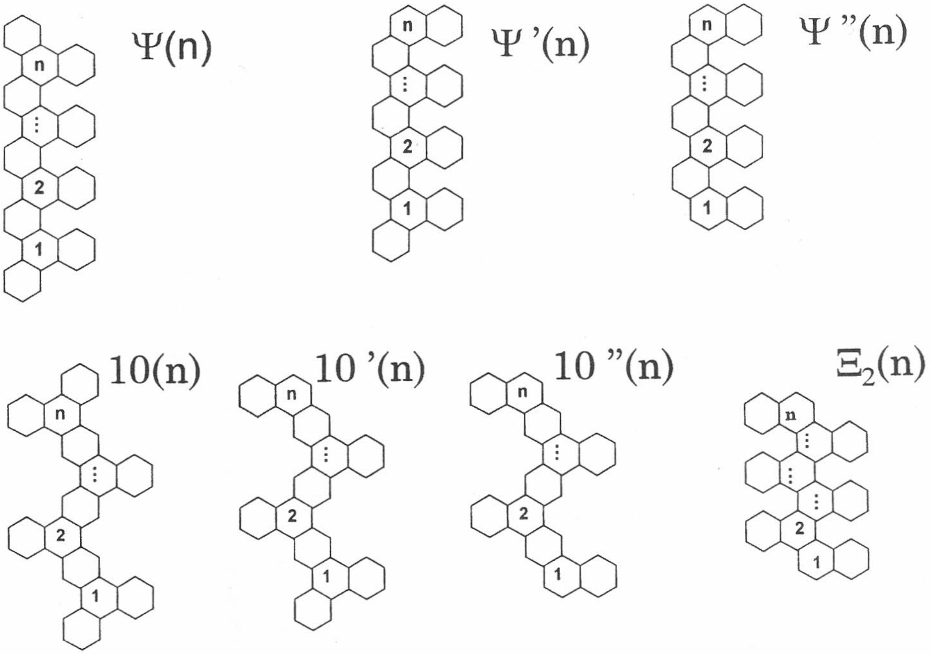


Figure 4. Homologous series of benzenoid hydrocarbons whose number of terminal hexagons increases with homologation.

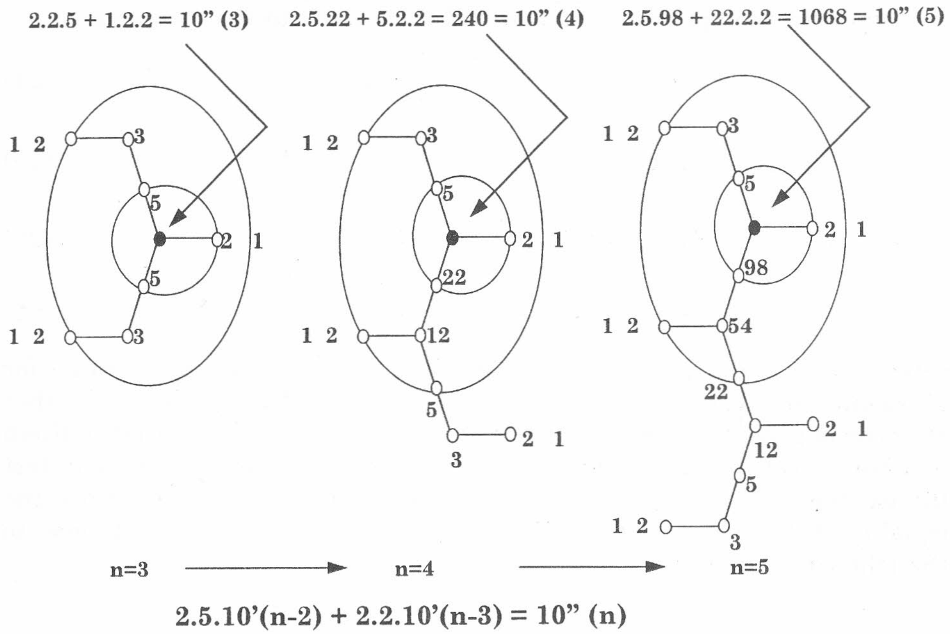


Figure 5. Recursive relation of Kekulé counts of the 10"(n) series derived through the application of the method of Klein and Seitz[20] (*c.f.* Figure 4. Members are represented in dualist[18] forms).

10"(n) systems in Figure 5. The relevant sums of the two products are shown for n = 3, 4, and 5, and lead to the following recursive relations:

$$\begin{aligned} n = 3 &\rightarrow 2.5.5 + 1.2.2 = 5 = K\{10"(3)\} \\ n = 4 &\rightarrow 2.5.22 + 5.2.2 = 240 = K\{10"(4)\} \\ n = 5 &\rightarrow 2.5.98 + 22.2.2 = 1068 = K\{10"(5)\} \end{aligned} \tag{20}$$

and by recalling the closed form K expressions of Appendix 3, one may generalize Eqs. (20) as follows:

$$2.5.10'(n - 2) + 2.2.10'(n - 3) = 10"(n) \tag{21}$$

Analogously, one might write the following two recursions:

$$2.2.\Psi(n - 1) + \Psi'(n - 1) = \psi(n) \tag{22}$$

$$2.3.\Xi_2(n - 2) + 2.2\Xi_2(n - 3) = \Xi_2(n) \tag{23}$$

Similarly, the approach of Klein and Seitz leads to the recursions:

$$2.\Psi'(n-1) + \Psi''(n-1) = \Psi''(n) \tag{24}$$

$$2.\Psi(n-1) + \Psi'(n-1) = \Psi'(n) \tag{25}$$

$$10'(n) + 2.10(n-1) = 10(n) \tag{26}$$

$$10(n-1) + 2.10(n-1) = 10'(n) \tag{27}$$

where, in the above equations, the symbol of a homologous series stands for its Kekulé counts, *e.g.* $\Psi(n) \equiv K\{\Psi(n)\}$ and so on. Naturally, there are other methods of generating the above recursive relations but the method of Klein and Seitz[20] can be easily applied in a consistent way. Now, in order to test the existence of combinatorial self-similarity (Eq. (6)), one must prove the equality of the relevant limits. For instance, Eq. (22) is expected to lead to the following equality

$$\lim_{n\to\infty}\frac{K\{\Psi(n+1)\}}{K\{\Psi(n)\}} = \lim_{n\to\infty}\frac{K\{\Psi'(n+1)\}}{K\{\Psi'(n)\}} \tag{28}$$

and similarity for other homologous series.

Indeed, all the relevant limits that appear in Eqs. (21)–(27) equal, as it can be easily envisaged by inspection of the following approximate K formulas for very large n values (*c.f.* Appendix 3)

3.3. *Characteristic Scaling Factors*

The Kekulé counts of a homologous series of benzenoid hydrocarbons, B(n), form a fractal whose characteristic scaling factor, $\mathfrak{S}$, is defined (here) by the following equation:

$$\mathfrak{S} = \lim_{n \to \infty} \frac{K\{\mathrm{B}(n+1)\}}{K\{\mathrm{B}(n)\}} \tag{36}$$

For example, the polyphenanthrenoid polyphenes[15] (benzene, naphthalene, phenanthrene, chrysene, picene, fulminene, ...)[16], the recently called unbranched quasicrystal – like benzenoids[12,13] $\equiv$ $\mathrm{B}_0(n)$ genrate the famous *»prototype«, self-similar* Fibonacci's sequence

2, 3, 5, 8, 13, 21, 34, ...

Self-similarity in this case might be envisaged by multiplying each term by 1.6 and rounding up to the nearest integer to obtain

3, 5, 8, 13, 21, ...

i.e. the sequence is generated again. The (characteristic) scaling factor is the (popular) golden mean,[12, 13] τ. obtained as the limit of two successive numbers (the larger one being the numerator). The limit is approached gradually:

$$3/2 = 1.5;\ 5/3 = 1.6;\ 8/5 = 1.6;\ 13/8 = 1.625;\ 21/13 = 1.615384615,\ ...,$$
$$\lim_{n\,-\,\infty} \mathrm{B}_0(n+1)/\mathrm{Bo}(n) = 1.6180330/989 = (\sqrt{5} - 1)/2\ .$$

The above limits has the following peculiar property:

$$\tau = 1 + \tau^{-1} \tag{37}$$

where $\tau^{-1} - 0.618033989$. Indeed, this irrational limit can be approached by the following rule:

Add one and take the reciprocal : viz:

$$1 \to (1 + 1)^{-1} = 1/2 \to (1/2 + 1)^{-1} = 2/3 \to (2/3 + 1)^{-1} =$$
$$= 3/5 \to 5/8 \to 8/13\ ...$$

Self-similarity in the above fraction is best revealed if expressed in the form of *continued fractions*:[17]

$$\frac{1}{2} = \cfrac{1}{1 + \cfrac{1}{1}} \equiv [1,1]$$

$$\frac{2}{3} = \cfrac{1}{\cfrac{3}{2}} = \cfrac{1}{1 + \cfrac{1}{2}} \equiv [1,2] \equiv [1,1,1]$$

$$\frac{3}{5} = \cfrac{1}{\cfrac{5}{3}} = \cfrac{1}{1 + \cfrac{1}{\cfrac{3}{2}}} = \cfrac{1}{1 + \cfrac{1}{1 + \cfrac{1}{2}}} \equiv [1,1,1,1] \tag{38}$$

$$\frac{5}{8} = \cfrac{1}{\cfrac{8}{5}} = \ldots \qquad\qquad [1,1,1,1,1]$$

and so on. Indeed, $\tau^{-1} \equiv [1,1,.....1] \equiv [1]$

Naturally, it is possible to compute analogous scaling factors which characterize the Kekulé counts of the homologous series of benzenoid systems studied in this paper. These are collected in Table II. The self-similar scaling properties of the $\Xi_2(n)$ homologous series are outlined in Table III as a representative example of other series. While using K_o underestimates K_n values (second column), K_1 seems to exaggerate these values (third column). However, the use of chrysene ($K_2 = 8$) leads to reasonable values (e.g. error in K_{10} is about 0.5%). It is perhaps safe to asume that *the higher members of the series are better reprentatives than the very early ones.*

TABLE II

Characteristic scaling factors, $\mathscr{S}$, of the Kekulé counts of the homologous series of benzenoids studied in this work.

Series	$\mathscr{S}$
Ξ_2	2.732050808
Ψ, Ψ', Ψ"	4.561552813
10, 10', 10"	4.449489743
P, P_1, P_2	9.898979486
Q, Q_1, Q_2	10.09901951

TABLE III

Self-similarity of the Kekulé counts of $\Xi_2(n)$ series with ϑ = 2.732050808 (*c.f.* Table II). Computed values are rounded to the nearest integer.

n	K_n	$\mathrm{K}_0\vartheta^n$	$K_1\vartheta^{n-1}$	$K_2\vartheta^{n-2}$
0	1	1		
1	3	3	3	
2	8	7	8	8
3	22	20	22	22
4	60	56	61	60
5	164	152	167	163
6	448	416	457	446
7	1224	1136	1248	1218
8	3344	3104	3408	3327
9	9136	8480	9312	9089
10	24960	23168	25440	24831

3.4. Homologous Series of Alkanes

In the early 1970's, Hosoya[21,26] defined the quantity $p(\mathrm{G}, k)$ as the number of selections of k edges in graph G so that no two of them are adjacent, sometimes called the number of k – matchings. These numbers are the coefficients of the various powers of a variable x in Hosoya's[21,26] counting polynomial, $H(\mathrm{G}\ x)$, *viz.*

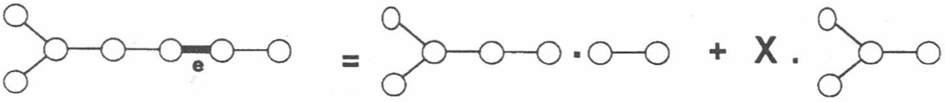
$$H(\mathrm{G};x) = \sum_{k=0}^{M} p(\mathrm{G},k)\, x^k \tag{39}$$

where M is the value for a maximal matching in G. The sum of these numbers, *i.e.* $H(\mathrm{G};\ 1)$, is the topological index, $Z(\mathrm{G})$ of graph G. Here, we take this sum to correspond to the property φ of eqns 5 and 6. It is possible to partition $Z(\mathrm{G})$ by application of the standard recursive relation:[21]

$$H(\mathrm{G}\ x) = H(\mathrm{G} - e;\ x) + xH(\mathrm{G} - e;\ x) \tag{40}$$

where e ($\in$G) is an edge in G; G – e is a (disconnected) graph obtained when e is pruned out of G and G – e is obtained from G when e plus its two vertices are removed from G. In order to make sense (of our postulated definition of combinatorial self-similarity), edge e should be the same throughout the members of the homologous series. We illustrate the procedure on 2-

methyl alkanes where (arbitrarily) the third edge after the branched vertex is selected. This limits the »first« member of this series at 2 – methylhexane. Application of Eq. (40) leads to 18 = 14 + 4, as illustrated below:



$$
\begin{aligned}
1 + 6X + 9X^2 + 2X^3 &= (1 + 4X + 2X^2)(1 + X) &&+ X(1 + 3X) \\
&= 1 + 5X + 6X^2 + 2X^3 &&+ X(1 + 3X)
\end{aligned}
$$

The sequences of $p(G,k)$ for G = 2-methyl hexane are:

$$\{1, 6, 9, 2\} = \{1, 5, 6, 2\} \ \& \ \{1, 3\} \tag{41}$$

$$\text{or} \qquad 18 = 14 + 4 \tag{42}$$

where the left-hand side of Eq. (42) is obtained as the sum of the integers in brakets to the right of the equality sign in Eq. (41). The righthand side of Eq. (41) are the corresponding values of the disconnected graphs. For example, the 2 represents the case where $k = 3$, *i.e.*

Now, because of the factor x in the recursive relation, the *second* number in the first brakets is added to the first one after the union sign and so on. Doing this leads to replacement of the & sign (in 41) by the + sign (in Eq. (42)).

Table IV lists the topological indices of 2-methyalkanes and their partitions as defined above, while Table V shows the sum and partial sum ratios for adjacent members in the homologous series. In all cases, these ratios approach the golden mean, $\tau = 1/2(1 + \sqrt{5}) = 1.618033989$, for large numbers of carbon atoms, thus demonstrating the combinatorial self-similarity of the homologous series of alkanes in the light of our postulated definition (Eq. (6)). The same pattern is obtained when studying cycloalkanes using a similar approach.

TABLE IV

Topological indices, $Z(\mathrm{G}_i)$ od 2-methylalkanes and a possible partition through application of Eq. (40) (G_6 = 2-methylhexane, G_7 = 2-methylheptane *etc*...)

i	$Z(\mathrm{G}_i)$	=	$Z'(\mathrm{G}_i)$	+	$Z''(\mathrm{G}_i)$
6	18		14		4
7	29		21		8
8	47		35		12
9	76		56		20
10	123		91		32
11	199		147		52
12	322		238		84
13	521		385		136
14	843		623		220
15	1264		1008		356
16	2207		1631		576
17	3571		2639		932
18	5778		4270		1508
19	9349		6909		2440
20	15127		11179		3948
21	24476		18088		6388
22	39603		29267		10336
23	64079		47355		16724
24	103682		76622		27060
25	167761		123977		43784
26	271443		200599		70844
27	439204		324576		114628
28	710647		525175		185472
29	1149851		849751		300100
30	1860498		1374926		485572
31	3010349		2224677		785672
32	4870847		3599603		1271244
33	7881196		5824280		2056916
34	12752043		9423883		3328160
35	20633239		15248163		5385076
36	33385282		24672046		8713236
37	54018521		39920209		14098312
38	87403803		64592255		22811548
39	141422324		104512464		36909860
40	228826127		169104719		59721408
41	370248451		273617183		96631268
42	599074578		442721902		156352676
43	969323029		716339085		252983944
44	1568397607		1159060987		409336620

TABLE V

$Z(\mathrm{G}_{i+1}) / Z(\mathrm{G}_i)$ values and the corresponding partial sums listed in Table IV. In all cases, the ratios approach the golden-mean, $\tau = 1.618033989$, at large values of i. The table is intended to demonstrate combinatorical self-similarity (*c.f.* Eq. (6))

n	$Z(\mathrm{G}_{i+1}) / Z(\mathrm{G}_i)$	$Z'(\mathrm{G}_{i+1}) / Z'(\mathrm{G}_i)$	$Z''(\mathrm{G}_{i+1}) / Z''(\mathrm{G}_i)$
6	1.611111111111111	1.5	2
7	1.620689655172414	1.666666666666667	1.5
8	1.617021276595745	1.6	1.666666666666667
9	1.618421052631579	1.625	1.6
10	1.617886178861789	1.615384615384615	1.625
11	1.618090452261306	1.619047619047619	1.615384615384615
12	1.618012422360248	1.617647058823529	1.619047619047619
13	1.618042226487524	1.618181818181818	1.617647058823529
14	1.61803084223013	1.617977528089888	1.618181818181818
15	1.618035190615836	1.618055555555556	1.617977528089888
16	1.618033529678296	1.618025751072961	1.618055555555556
17	1.618034164099692	1.618037135278515	1.618025751072961
18	1.618033921772239	1.618032786885246	1.618037135278515
19	1.618034014333084	1.618034447821682	1.618032786885246
20	1.618033978977986	1.618033813400125	1.618034447821682
21	1.618033992482432	1.618034055727554	1.618033813400125
22	1.618033987324193	1.618033963166706	1.618034055727554
23	1.618033989294465	1.618033998521803	1.618033963166706
24	1.618033988541888	1.618033985017358	1.618033998521803
25	1.618033988829346	1.618033990175597	1.618033985017358
26	1.618033988719547	1.618033988205325	1.618033990175597
27	1.618033988761487	1.618033988957902	1.618033988205325
28	1.618033988745467	1.618033988670443	1.618033988957902
29	1.618033988751586	1.618033988780243	1.618033988670443
30	1.618033988749249	1.618033988738303	1.618033988780243
31	1.618033988750142	1.618033988754322	1.618033988738303
32	1.618033988'749801	1.618033988748204	1.618033988754322
33	1.618033988749931	1.618033988750541	1.618033988748204
34	1.618033988749881	1.618033988749648	1.618033988750541
35	1.6180339887499	1.618033988749989	1.618033988749648
36	1.618033988749893	1.618033988749859	1.618033988749989
37	1.618033988749896	1.618033988749909	1.618033988749859
38	1.618033988749894	1.61803398874989	1.618033988749909
39	1.618033988749895	1.618033988749897	1.61803398874989
40	1.618033988749895	1.618033988749894	1.618033988749897
41	1.618033988749895	1.618033988749895	1.618033988749894
42	1.618033988749895	1.618033988749895	1.618033988749895
43	1.618033988749895	1.618033988749895	1.618033988749895
44	1.618033988749895	1.618033988749895	1.618033988749895

3.5. Manifestations of Self-Similarity

Self-similarity of the golden mean becomes apparent if expressed in the form of an infinite continued fraction (*c.f.* Eq. 38).

In order to investigate the fractal (scaling) self-similarity properties of combinatorial characteristics of homologous series of (chemical) structures, the ratios of relevant values of adjacent members are represented as their continued fractions. Figures 6–9 show these representations for the classes of benzenoids studied, where K values are used to generate the required ra-

P(n)

20	[10]
198	[9, 1, 9]
1960	[9, 1, 8, 1, 9]
19402	[9, 1, 8, 1, 8, 1, 9]
192060	[9, 1, 8, 1, 8, 1, 8, 1, 9]
1901198	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]
18819920	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]
186298002	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]
1844160100	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]
18255302998	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]

P_1(n)

9	[9]
89	[9, 1, 8]
881	[9, 1, 8, 1, 8]
8721	[9, 1, 8, 1, 8, 1, 8]
86329	[9, 1, 8, 1, 8, 1, 8, 1, 8]
854569	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8]
8459361	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8]
83739041	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8]
828931049	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8]
8205571449	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8]

P_2(n)

4	[8]
40	[10]
396	[9, 1, 9]
3920	[9, 1, 8, 1, 9]
38804	[9, 1, 8, 1, 8, 1, 9]
384120	[9, 1, 8, 1, 8, 1, 8, 1, 9]
3802396	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]
37639840	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]
372596004	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]
3688320200	[9, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 8, 1, 9]

Figure 6. Self-similarity of ratios of Kekulé counts, of successeive members of P(n), $P_1(n)$ and $P_2(n)$ series shown in Figure 2 Boldly printed numbers are Kekulé counts while sequences in square brackets are ratios of K's represented in the form of continued fraction.[17]

	20	[10]
	202	[10, 10]
Q(n)	**2040**	[10, 10, 10]
	20602	[10, 10, 10, 10]
	208060	[10, 10, 10, 10, 10]
	2101202	[10, 10, 10, 10, 10, 10]
	21220080	[10, 10, 10, 10, 10, 10, 10]
	214302002	[10, 10, 10, 10, 10, 10, 10, 10]
	2164240100	[10, 10, 10, 10, 10, 10, 10, 10, 10]
	21856703002	[10, 10, 10, 10, 10, 10, 10, 10, 10, 10]

	9	[9]
	91	[10, 9]
Q_1(n)	**919**	[10, 10, 9]
	9281	[10, 10, 10, 9]
	93729	[10, 10, 10, 10, 9]
	946571	[10, 10, 10, 10, 10, 9]
	9559439	[10, 10, 10, 10, 10, 10, 9]
	96540961	[10, 10, 10, 10, 10, 10, 10, 9]
	974969049	[10, 10, 10, 10, 10, 10, 10, 10, 9]
	9846231451	[10, 10, 10, 10, 10, 10, 10, 10, 10, 9]

	4	[4]
	41	[10, 4]
Q_2(n)	**414**	[10, 10, 4]
	4181	[10, 10, 10, 4]
	42224	[10, 10, 10, 10, 4]
	426421	[10, 10, 10, 10, 10, 4]
	4306434	[10, 10, 10, 10, 10, 10, 4]
	43490761	[10, 10, 10, 10, 10, 10, 10, 4]
	439214044	[10, 10, 10, 10, 10, 10, 10, 10, 4]
	4435631201	[10, 10, 10, 10, 10, 10, 10, 10, 10, 4]

Figure 7. The same as in Figure 6 for the indicated series of Figure 2. The Q(n) system is *exactly* self-similar!

tios. Figure 10 is the corresponding representation for topological indices of 2-methyl alkanes and their partial sums. In all cases, the resulting sequences are either exactly self-similar (*e.g.* Q(n) system, Figure 7) or approximately so (*e.g.* Ψ" series, Figure 4). Scaling factors can be expressed in closed forms, *e.g.* K values of the 10 n, 10 n', 10 n'' benzenoids generate the self-similar fractal

$$4\ 2\ 4\ 2\ 4\ 2\ 4\ 2\ \ldots \tag{43}$$

whose characteristic scaling factor is

Ψ_n

9	[4, 2]
41	[4, 1, 1, 4]
187	[4, 1, 1, 3, 1, 1, 2]
853	[4, 1, 1, 3, 1, 1, 3, 3]
3891	[4, 1, 1, 3, 1, 1, 3, 1, 1, 6]
17749	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 2, 2]
80963	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 2, 1, 4]
369317	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 2, 9]
1684659	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 4, 4, 2]
7684661	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 1, 1, 1, 1, 3]

Ψ_n'

5	[5]
23	[4, 1, 1, 2]
105	[4, 1, 1, 3, 3]
479	[4, 1, 1, 3, 1, 1, 6]
2185	[4, 1, 1, 3, 1, 1, 3, 1, 2, 2]
9967	[4, 1, 1, 3, 1, 1, 3, 1, 1, 2, 1, 4]
45465	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 2, 9]
207391	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 4, 4, 2]
946025	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 1, 1, 1, 1, 3]
4315343	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 4, 7]

Ψ_n''

3	[3]
13	[4, 3]
59	[4, 1, 1, 6]
269	[4, 1, 1, 3, 1, 2, 2]
1227	[4, 1, 1, 3, 1, 1, 2, 1, 4]
5597	[4, 1, 1, 3, 1, 1, 3, 2, 9]
25531	[4, 1, 1, 3, 1, 1, 3, 1, 1, 4, 4, 2]
116461	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 1, 1, 1, 1, 3]
531243	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 4, 7]
2423293	[4, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 3, 1, 1, 8, 3, 2]

Figure 8. The same as in Figure 6 for the indicated series of Figure 4.

where $j = 0, 1, 2, ...$ generating the values:

$$2, 1/2, 2, 1/2, ...$$

The above sequence can be obtained from the sequence of Eq. (43):

$$4/2, 2/4, 4/2, 2/4, ...$$

Analogously, for the Ψ_n, Ψ'_n and Ψ''_n series, one might see the sequence

$$1\ 1\ 3\ 1\ 1\ 3\ 1\ 1\ 3\ 1\ 1\ 3\ ... \tag{45}$$

after neglecting the initial term and a few last terms. The characteristic scaling factor in this case is

$$(3)^{(-1)^j} \tag{46}$$

10 n	9	[4, 2]
	40	[4, 2, 4]
	178	[4, 2, 4, 2]
	792	[4, 2, 4, 2, 4]
	3524	[4, 2, 4, 2, 4, 2]
	15680	[4, 2, 4, 2, 4, 2, 4]
	69768	[4, 2, 4, 2, 4, 2, 4, 2]
	310432	[4, 2, 4, 2, 4, 2, 4, 2, 4]
	1381264	[4, 2, 4, 2, 4, 2, 4, 2, 4, 2]
	6145920	[4, 2, 4, 2, 4, 2, 4, 2, 4, 2, 4]

10 n'	5	[5]
	22	[4, 2, 2]
	98	[4, 2, 5]
	436	[4, 2, 4, 2, 2]
	1940	[4, 2, 4, 2, 5]
	8632	[4, 2, 4, 2, 4, 2, 2]
	38408	[4, 2, 4, 2, 4, 2, 5]
	170896	[4, 2, 4, 2, 4, 2, 4, 2, 2]
	760400	[4, 2, 4, 2, 4, 2, 4, 2, 5]
	3383392	[4, 2, 4, 2, 4, 2, 4, 2, 4, 2, 2]

10 n''	3	[2]
	12	[4]
	54	[4, 2]
	240	[4, 2, 4]
	1068	[4, 2, 4, 2]
	4752	[4, 2, 4, 2, 4]
	21144	[4, 2, 4, 2, 4, 2]
	94080	[4, 2, 4, 2, 4, 2, 4]
	418608	[4, 2, 4, 2, 4, 2, 4, 2]
	1862592	[4, 2, 4, 2, 4, 2, 4, 2, 4]

$\Xi_2(n)$	3	[3]
	8	[2, 1, 2]
	22	[2, 1, 3]
	60	[2, 1, 2, 1, 2]
	164	[2, 1, 2, 1, 3]
	448	[2, 1, 2, 1, 2, 1, 2]
	1224	[2, 1, 2, 1, 2, 1, 3]
	3344	[2, 1, 2, 1, 2, 1, 2, 1, 2]
	9136	[2, 1, 2, 1, 2, 1, 2, 1, 3]
	24960	[2, 1, 2, 1, 2, 1, 2, 1, 2, 1, 2]

Figure 9. The same as in Figure 6 (*c.f.* Figure 4).

where

$$j = 0,\ 1,\ 2,\ 0,\ 1,\ 2,\ 0,\ 1,\ 2,\ \ldots$$

which generates

$$1,\ 1/3,\ 3,\ 1,\ 1/3,\ 3,1,\ 1/3,\ 3,\ \ldots \tag{47}$$

The above values can be obtained as ratios of successive values of the fractal of Eq. (45). A similar approach may be used to investigate cycloalkanes.

29	[1. 1, 1, 1, 1, 3]
47	[1, 1, 1, 1, 1, 1, 3]
76	[1, 1, 1, 1, 1, 1, 1, 3]
123	[1, 1, 1, 1, 1, 1, 1, 1, 3]
199	[1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
322	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
521	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
843	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
1364	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
2207	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
3571	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
5778	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
9349	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]
15127	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 3]

21	[1, 2]
35	[1, 1, 2]
56	[1, 1, 1, 2]
91	[1, 1, 1, 1, 2]
147	[1, 1, 1, 1, 1, 2]
238	[1, 1, 1, 1, 1, 1, 2]
385	[1, 1, 1, 1, 1, 1, 1, 2]
623	[1, 1, 1, 1, 1, 1, 1, 1, 2]
1008	[1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
1631	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
2639	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
4270	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
6909	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
11179	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]

8	[2]
12	[1, 2]
20	[1, 1, 2]
32	[1, 1, 1, 2]
52	[1, 1, 1, 1, 2]
84	[1, 1, 1, 1, 1, 2]
136	[1, 1, 1, 1, 1, 1, 2]
220	[1, 1, 1, 1, 1, 1, 1, 2]
356	[1, 1, 1, 1, 1, 1, 1, 1, 2]
576	[1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
932	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
1508	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
2440	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]
3948	[1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 2]

Figure 10. Continued fraction[17] representation of the ratios of topological indices[21] of two successive members of 2-methyl alkanes (top third) and the corresponding partial sums (lower two-thirds). *c.f.* Table IV (*e.g.* 29 = 21 + 8, 47 = 35 + 12, and so on).

4. DISCUSSION AND CONCLUSIONS

A basic fact of organic chemistry is that members of a homologous series of structures exhibit similar properties. For example, the linear acenes are known[2] to loose stability beyound six rings: Heptacene is green-black and cannot be obtained in a pure state. On the other hand, the polyphenan-

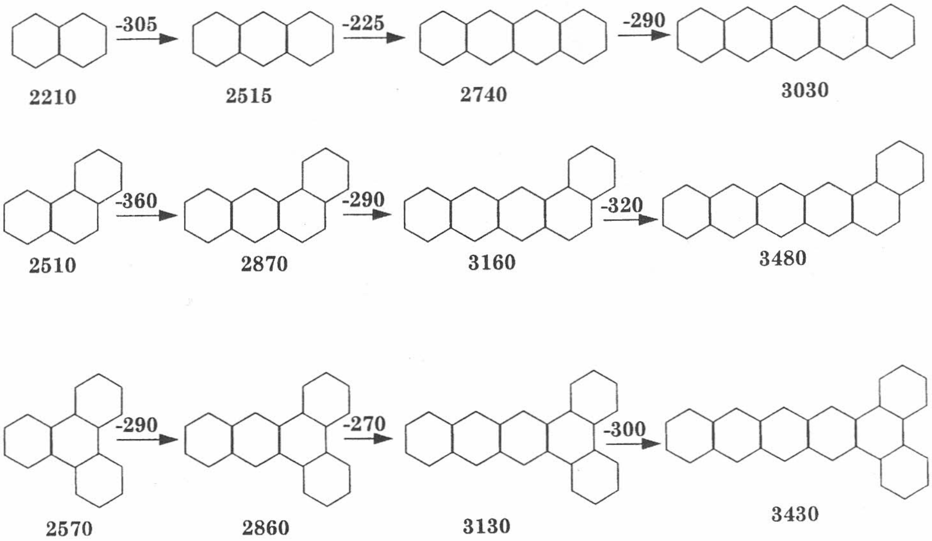


Figure 11. Three homologous series of benzenoid hydrocarbons. Numbers below molecular graphs are the UV wave-lengths of their β band.[16]

threnoids[12,15,16] (polyphenes in which the benzene rings are annellated in a zig-zag way) show much higher stabilities (highest K values and shortest wavelengths). In Figure 11, three homologous series of benzenoid hydrocarbons are shown with their wavelengths of the β bands[27]. It seems that, initially, in each series each additional benzene ring leads to an »average« decrease of approximately 300 nm. However, members of the series themselves are *not* geometrically self-similar, but they generate combinatorial parameters which form self-similar sequences with *characteristic* scaling factors and, hence, might be viewed as fractals. Homologous series of alkanes (and cycloalkanes) are Fibonacci graphs[12,28], for which a given combinatorial property recurs as

$$\varphi(n+2) = \varphi(n+1) + \varphi(n) \tag{48}$$

and hence is associated with the auxilliary Eq.[29]

$$r^2 - r - 1 = 0 \tag{49}$$

The general solution, then, would be

$$\varphi(n) = C_1\tau^n - C_2\tau^{-n} \tag{50}$$

where C_1 and C_2 are constants which depend on the initial conditions. At very large values of n, only the first term (Eq. (50)) dominates and the limit of two successive values approaches τ, the golden mean. In fact, all homologous series of compounds which are Fibonacci graphs (with either internal or external homologation)[28] comply with Eq. (6) and, hence, exhibit combinatorial self-similarity with the golden mean as their characteristic scaling factor (regardless, of course, of their initial conditions). This fundamental fact has computational significance for infinitely large graphs; as an illustration, the numbers of Kekulé structures of the benzenoid hydrocarbons whose hexagons are annelated[30] (respectively) as $L^3A^{31}L^3$ can be calculated from a much earlier member, say $L^3A^4L^3$ ($K = 65$) by a direct application of the *scaling* properties of the Kekulé counts of a homologous series of hydrocarbons. The required value = $(65)(1.618033989)^{27} \cong 28548260$; the exact value = 28710853 ; the percentage error is just a little 0.5%. Here, we emphasize that the fractal properties of Fibonacci graphs do not offer a solution to an unsolved problem (there are many methods for obtaining K), but it is always illuminating to have options and alternatives. One recalls here that the calculated value of K is also the topological index (of Hosoya)[26] of an alkane containing 38 carbon atoms! Since this topological index orders the physical properties of hydrocarbons, the scaling (fractal $\equiv$ self-similar) properties of homologous series have both computational *and* chemical importance. Further, if one recalls the concept of equivalence of objects of physicochemical interest,[31,32] one immediately recognizes the computational K values as a combinatorial property of certain Clar graphs, king polyomino, rook board, Young diagram and a caterpillar tree!

Construction of a homologous series of structures (be it chemical or not) is a »self-same« operation (*i.e.* repeated addition of carbon atoms, annelation of hexagons, ring expansion, ...) and this sort of construction is more like iteration, which is one of the richest sources of self-similarity, and because of the mathematical equivalence of benzenoid systems with almost all objects of physical interest (trees, diagrams, boards, *etc*.....), interest will continue in the study of benzenoid hydrocarbons[33] in addition to the theory of Kekulé[18] and the model of Clar, their mathematical equivalence to caterpillar trees (which is a case of data reduction) make a homologous series of unbranched benzenoids scalable by the golden mean! Indeed, combinatorial properties of homologous series of other objects (Young diagrams, rook boards *etc*....)[32] will also be scalable by τ, which seems, with the discovery of fractals, to assume the importance of fundamental quantities such as π and Planck's constant and will always remind us of the »Fibonacci rabbits«!

Acknowledgment – The Author's son Islam El-Basil computed the continued fractions, did the art work and typed the manuscript.

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$$X(\Gamma) = \sum_{\mathrm{G} \in \mathrm{C}(\Gamma)} x(\mathrm{G}) \qquad (R-1)$$

where C (Γ) is a set of connected subgraphs of Γ, Eq. $(R-1)$ is analogous to Eq. (8) and $\mathrm{C}(\Gamma)$ may be thought to correspond to an equivalence class of Kekulé structures $\in$ B. The same approach is applied by : T. G. Schmalz, T. Živković, and D. J. Klein, in R. C. Lacher (Ed.) MATH/CHEM/COMP 1987; Studies in Physical and Theoretical Chemistry **54**, Elsevier, Amsterdam, 1988, p. 173. More recently, it was used to »re-visit« the concept of resonance energy of conjugated hydrocarbons: D. Babić and N. Trinajstić, *Croat. Chem. Acta* **65** (1992) 881.

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Appendix 1

Formulas of Kekulé counts,[18] K's of the benzenoid systems shown in Figure 2

$$K\{\mathrm{P}(n)\} = \frac{1}{2\sqrt{6}}\left[(5+2\sqrt{6})^{n+1} - (5-2\sqrt{6})^{n+1}\right]$$

$$K\{\mathrm{P}_1(n)\} = \frac{1}{2\sqrt{6}}\left[(\sqrt{6}+2)(5+2\sqrt{6})^{n} + (\sqrt{6}-2)(5-2\sqrt{6})^{n}\right]$$

$$K\{\mathrm{P}_2(n)\} = \frac{1}{\sqrt{6}}\left[(5+2\sqrt{6})^{n} - (5-2\sqrt{6})^{n}\right]$$

$$K\{\mathrm{Q}(n)\} = \frac{1}{\sqrt{26}}\left[(5+\sqrt{26})^{n+1} - (5-\sqrt{26})^{n+1}\right]$$

$$K\{\mathrm{Q}_1(\mathrm{n})\} = \frac{1}{2\sqrt{26}}\left[(\sqrt{26}+4)(5+\sqrt{26})^{n} + (\sqrt{26}-4)(5-\sqrt{26})^{n}\right]$$

$$K\{\mathrm{Q}_2(n)\} = \frac{1}{2\sqrt{26}}\left[(\sqrt{26}-1)(5+\sqrt{26})^{n} + (\sqrt{26}+1)(5-\sqrt{26})^{n}\right]$$

Appendix 2

Expressions of the Kekulé counts[18], K's of auxilliary benzenoid systems shown (in bold) in Figure 2

$$K\{\mathrm{Q}'(n)\} = \frac{1}{2}\left[K\{\mathrm{Q}(n)\} + K\{\mathrm{Q}(n-1)\}\right]; \quad n \geq 1$$

$$K\{\mathrm{Q}''(n)\} = \frac{1}{4}\left[K\{\mathrm{Q}(\mathrm{n})\} + 2K\{\mathrm{Q}(n-1)\} + \mathrm{K}\{\mathrm{Q}(n-2)\}\right]; \quad n \geq 2$$

Appendix 3

$$K\{\Psi(n)\} = \frac{1}{\sqrt{17}}\left[(\sqrt{17}+4)\,\frac{(5+\sqrt{17})^n}{2} + (\sqrt{17}-4)\,\frac{(5-\sqrt{17})^n}{2}\right]$$

$$K\{\Psi'(n)\} = \frac{1}{\sqrt{17}}\left[\frac{(5+\sqrt{17})^{n+1}}{2} - \frac{(5-\sqrt{17})^{n+1}}{2}\right]$$

$$K\{\Psi''(n)\} = \frac{1}{2\sqrt{17}}\left[(\sqrt{17}+1)\,\frac{(5+\sqrt{17})^n}{2} + (\sqrt{17}-1)\,\frac{(5-\sqrt{17})^n}{2}\right]$$

$$K\{10(n)\} = \frac{1}{4\sqrt{6}}\left[(2+\sqrt{6})^{n+2} - (2-\sqrt{6})\right]^{\mathrm{n+2}}$$

$$K\{10'(n)\} = \frac{1}{4}\left[(2+\sqrt{6})^{n+1} - (2-\sqrt{6})^{n+1}\right]$$

$$K\{10''(n)\} = \frac{1}{2\sqrt{6}}\left[(2+\sqrt{6})^{n} - (2-\sqrt{6})^{n}\right]$$

Appendix 4

Calculation of continued fractions of Figure 6

$$\frac{20}{202} = \frac{1}{\frac{202}{20}} = \frac{1}{10+\frac{2}{20}} = \frac{1}{10+\frac{1}{\frac{20}{2}}} = \frac{1}{10+\frac{1}{10}} \equiv [10,10]$$

$$\frac{9}{91} = \frac{1}{\frac{91}{9}} = \frac{1}{10+\frac{1}{9}} \equiv [10,9]$$

$$\frac{91}{919} = \frac{1}{\frac{919}{91}} = \frac{1}{10+\frac{9}{91}} = \frac{1}{10+\frac{1}{\frac{91}{9}}} = \frac{1}{10+\frac{1}{10+\frac{1}{9}}} \equiv [10,10,9]$$

$$\frac{414}{4181}=\frac{1}{\dfrac{4181}{414}}=\frac{1}{10+\dfrac{41}{414}}=\frac{1}{10+\dfrac{1}{\dfrac{414}{41}}}=\frac{1}{10+\dfrac{1}{10+\dfrac{4}{41}}}=\frac{1}{10+\dfrac{1}{10+\dfrac{1}{\dfrac{41}{4}}}}=$$

$$=\frac{1}{10+\dfrac{1}{10+\dfrac{1}{10+\dfrac{1}{4}}}}\equiv[10,10,10,4]$$

SAŽETAK

Kombinatorna samosličnost

Sherif El-Basil

Kombinatorna samosličnost nov je koncept koji se uvodi da bi se objasnila sličnost svojstava članova homolognih nizova koji nisu geometrijski samoslični i stoga nisu deterministički fraktali. Pri definiranju koncepta treba:

a) odabrati neku numeričku invarijantu φ koja karakterizira članove niza,

b) razdijeliti svojstvo φ u konačan broj dijelova po nekom propisanom algoritmu,

c) za članove niza kažemo da su samoslični (ili da predstavljaju »numerički« fraktal) ako omjer svojstva φ za sukcesivne, sve veće članove niza teži nekoj graničnoj vrijednosti, a isto svojstvo moraju pokazati i u b) uvedene particije tog svojstva.

U ovom radu smo promatrali benzenoidne sustave (svojstvo φ bio je pripadni broj Kekuléovih struktura), te zasićene ugljikovodike (svojstvo φ bili su Hosoyini indeksi, Z). Ako je broj terminalnih heksagona u benzenoidima konstantan, rabi se ranije uvedena relacija ekvivalencije za particiju broja K, a kada to nije slučaj, onda postupak Kleina i Seitza. Kod alkana je particija broj Z izvedena upotrebom prikladne rekurzivne relacije, pa je za njihove homologne nizove, kao i za homologne nizove nerazgrananih benzenoida i neke druge sustave od interesa u matematičkoj kemiji, nađeno da je svojstvo Z skalirana zlatnim rezom. Nađeni su skalirajući faktori i za neke druge sustave, kao npr. za razgranane benzenoide. U svima tim slučajevima bilo je moguće prikazati omjere svojstava φ za sukcesivne članove niza u obliku neprekinutog razlomka, što u posebnim slučajevima vodi do točne, a u općem slučaju samo do približne samosličnosti.