

## Benzenoid catafusenes: Perfect matchings, isomerization, automerization

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**Abstract.** It is shown that for a given number  $h$  of benzenoid rings, the polycyclic aromatic hydrocarbons with the maximum number  $K(\max)$  of Kekulé structures (or of perfect matchings) are cata-condensed. The structures of such catafusenes with  $K(\max)$  present interesting 4- and 3-periodicities in terms of  $h$ . By analogy with known catalytic isomerizations of benz[a]anthracene into chrysene, i.e. of a catafusene with a smaller number of perfect matchings into one with a higher number, it was inferred in 1965 that degenerate rearrangements (automerizations) of catafusenes may be possible, and this was demonstrated thermally in 1977 by Scott. Data for the  $AlHal_3$ -catalyzed automerization of phenanthrene labeled with carbon-13 are reviewed, and the reaction mechanism is discussed. Prospects for new thermal or catalytic isomerizations or automerizations of catafusenes are considered.

### 1. THEORY

**1.1 Introduction.** It is one of the tenets of valence-bond theory (ref.1) that among isomeric benzenoids, the systems with larger numbers of Kekulé structures are more stable. Later developments in this field include Clar's all-benzenoid structures (ref.2), Randić's conjugated circuits model (ref.3), Gordon and Davison's algorithm for counting Kekulé structures (ref.4), and the representation of benzenoids via their dualists (ref.5). Many of these advances have benefitted from the interplay between organic chemistry and graph theory; chemical graph theory, or graph-theoretical chemistry, is now a rapidly expanding research area with its own journals, symposia, and monographs (refs.6-10).

**1.2 Definitions.** We shall call benzenoid or polyhex any polycyclic aromatic hydrocarbon with fused benzene rings, irrespective whether it is a portion of the graphite lattice in its hydrogen-depleted representation ("in-plane benzenoid") or it has helicenic portions ("out-of-plane benzenoid", with two or more carbons projected together on the hexagonal lattice as in hexahelicene or higher helicenes); even in pentahelicene the hydrogen atoms would overlap.

The dualist graph (inner dual, or simply dualist) of a benzenoid consists of vertices which are centers of hexagons, and of edges which connect vertices corresponding to condensed hexagons, i.e. hexagons sharing an edge; unlike most graphs, in dualists the angles between edges do matter. Benzenoids are of three kinds: cata-condensed (catafusenes) whose dualist graphs are acyclic; peri-condensed (perifusenes) whose dualists contain 3-membered rings; and coronoids whose dualists contain larger rings which are not the periphery of 3-membered ring aggregates.

If one views dualists of catafusenes similarly to alkane carbon chains, then any rotation by  $180^\circ$  around an edge which leads to non-isomorphic benzenoid structures leaves unchanged the number of Kekulé structures (called also the number of perfect matchings) ; in other words, benzenoids which differ by reversing "left-hand kinks" of their dualists into "right-hand kinks" have the same number  $K$  of perfect matchings, and are called isoarithmic (ref.11).

### 1.3 Benzenoids with maximum numbers of Kekulé structures.

In this section we shall solve the following problem : what are the structures of benzenoids with the maximum number of Kekulé structures for given numbers  $h$  of hexagons ?

Gutman (ref.12) demonstrated that for certain numbers of hexagons :

$$h(X_k) = 3 \cdot 2^{k-1} - 2 \quad (1)$$

i.e.  $h = 4, 10, 22, 46$  for  $k = 2, 3, 4, 5$  and so on, the structures of benzenoids with maximum numbers of Kekulé structures are highly branched catafusenes with trigonal symmetry, whose dualists have vertices of degree 3 (i.e. with three lines or edges emerging from them) and 1 (endpoints). The corresponding numbers of perfect matchings or of Kekulé structures are :

$$K(X_k) = K(Y_{k-1})^3 + K(Y_{k-2})^6 \quad (2)$$

The auxiliary series  $Y_k$  has bilateral symmetry with one central point of degree 2 in the dualist, i.e. series  $X_k$  lacking one of the three main branches. The corresponding numbers of benzenoid rings and Kekulé structures are :

$$h(Y_k) = 2^k - 1 \quad (3)$$

$$K(Y_k) = K(Y_{k-1})^2 + K(Y_{k-2})^4 \quad (4)$$

Recurrences (2) and (4) allow the easy determination of  $K$  values. One can see, however, that the  $h$  values in eq. (1) are very sparse.

In a joint effort with X. Liu, D. J. Klein and S. J. Cyvin, we solved the problem stated at the beginning of this section for out-of-plane benzenoids (ref.13). For systems which exclude heli-cenic systems, the problem is still unsolved ; the general problem was proposed as a challenge at the Symposium "Quo Vadis Graph Theory ?" (ref.14).

It is easy to prove that catafusenes with at least three linearly condensed hexagons have smaller numbers of Kekulé structures than isomeric catafusenes devoid of such a type of condensation. As it is well-known, acenes have smaller numbers of perfect matchings than their zig-zag condensed isomers which were called fibonacenes because their  $K$  values form the Fibonacci sequence (refs.4, 11). Therefore for catafusenes with maximum number of perfect matchings for a given number of hexagons, no fragment with three or more linearly condensed benzenoid rings should be present. Such systems will be henceforth be called  $K(\max)$  benzenoids.

TABLE 1.  $K(\max)$  values for polyhexes with given number  $h$  of hexagons

$h$	$K(\max)$	$h$	$K(\max)$	$h$	$K(\max)$	$h$	$K(\max)$	$h$	$K(\max)$
1	2	6	24	11	305	16	3987	21	49913
2	3	7	41	12	510	17	6515	22	84546
3	5	8	66	13	863	18	10905	23	138170
4	9	9	110	14	1425	19	18254	24	231117
5	14	10	189	15	2345	20	30135	25	386222

Table 2. Dualist graphs of benzenoids with maximum number of Kekulé structures for a given number  $b$  of hexagons (asterisks indicate systems which have two or more isoarithmic isomers : their dualist graphs differ by rotation around edges with angles of  $180^\circ$ )

$b = 3(\text{mod } 4)$		$b = 0(\text{mod } 4)$		$b = 1(\text{mod } 4)$		$b = 2(\text{mod } 4)$	
$b$		$b$		$b = 1$	$\bullet$	$b = 2$	$\bullet$
3		4		5		6	
7		8		9*		10	
11		12*		13*		14	
15*		16		17*		18	
19		20*		21*		22	

A computer program was developed by D. J. Klein and X. Liu to generate  $K(\text{max})$  benzenoids (ref.13) and calculations were performed for  $h \leq 60$ . Results for  $K(\text{max})$  are presented in Table 1, and structures of  $K(\text{max})$  benzenoids are shown in Table 2 only for the first 22, but all structures may be found in ref.13. It must be stressed that in most cases, indicated by asterisks in Table 2, two or more isoarithmic benzenoids result as the solution for certain  $h$  values. In such cases, the least sterically hindered structure is displayed for convenience.

Although calculations were performed only for  $h \leq 60$ , the 4-periodicity in terms of  $h$  which is observable from Table 2 can safely be assumed to hold for all  $h$  values. By using abbreviations P, X, Y, Z for subgraphs with 3, 6, 9, and 12 vertices, respectively, as indicated in the heading of Table 3, one may convert Table 2 into a simpler format : the resulting Table 3 shows the same remarkable 4-periodicity and, in addition, a 3-periodicity in terms of  $h$  if one of the letters P is replaced by X, Y, or Z. One may see from Table 3 that the same core graph is repeated vertically in each of the four columns starting from the upper right-hand corner and stepping

Table 3. Dualist graphs of  $K_{\max}$  catafusenes with their core graphs surrounded by P,X,Y,Z subgraphs

$h \equiv 3 \pmod{4}$ ; $Z = \begin{array}{c} P \\ \diagdown \quad \diagup \\ P \end{array}$	$h \equiv 0 \pmod{4}$ ; $Y = \begin{array}{c} P \\ \diagdown \quad \diagup \\ P \end{array}$	$h \equiv 1 \pmod{4}$ ; $X = \begin{array}{c} P \\ \diagdown \quad \diagup \\ P \end{array}$	$h \equiv 2 \pmod{4}$ ; $P = \begin{array}{c} P \\ \diagdown \quad \diagup \\ P \end{array}$
			10 $\bullet P_3$
		13 $\bullet P_2 X$	14 $P_2 \longleftrightarrow P_2$
	16 $\bullet P_2 Y$	17 $P_2 \longleftrightarrow P X$	18 $P_2 \longleftrightarrow \begin{array}{c} P \\ \diagdown \quad \diagup \\ P_2 \end{array}$
19 $\bullet P_2 Z$	20 $P_2 \longleftrightarrow P Y$	21 $P_2 \longleftrightarrow \begin{array}{c} X \\ \diagdown \quad \diagup \\ P_2 \end{array}$	22 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$
23 $P_2 \longleftrightarrow P Z$	24 $P_2 \longleftrightarrow \begin{array}{c} Y \\ \diagdown \quad \diagup \\ P_2 \end{array}$	25 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P X \end{array}$	26 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$
27 $P_2 \longleftrightarrow \begin{array}{c} Z \\ \diagdown \quad \diagup \\ P_2 \end{array}$	28 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P Y \end{array}$	29 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	30 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$
31 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P Z \end{array}$	32 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	33 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	34 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$
35 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	36 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	37 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	38 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$
39 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	40 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	41 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	42 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$
43 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	44 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	45 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$	46 $P_2 \longleftrightarrow \begin{array}{c} P_2 \\ \diagdown \quad \diagup \\ P_2 \end{array}$

down diagonally for each row. Thus, starting with the right-most column with  $h \equiv 2 \pmod{4}$ , which in Table 2 had dualists whose vertices had only degrees 3 and 1, the core graph is completely surrounded by P subgraphs and has  $h'$  vertices, where  $h' = (h-6)/4$ . The core graph has in this column vertices of degree 3 and 1, and for  $h \equiv 2 \pmod{8}$  one vertex of degree 2; this is the vertex where in the other columns the letter P is replaced by X, Y, or Z for  $h \equiv 5 \pmod{8}$ ,  $h \equiv 0 \pmod{8}$ , and  $h \equiv 3 \pmod{8}$ , respectively. For  $h \equiv 1 \pmod{8}$ ,  $h \equiv 4 \pmod{8}$ , and  $h \equiv 7 \pmod{8}$ , the replacement of P by X, Y, or Z, respectively, occurs at an endpoint which is closest to the center of the core graph.

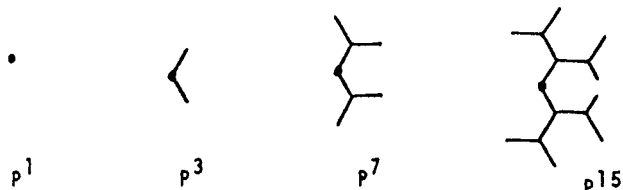
Thus, given the core graphs of the right-most column in Table 3, the remaining columns are perfectly determined. For the right-most column of Table 3 with  $h \equiv 2 \pmod{4}$  the following rules apply: whenever possible, trigonal symmetry around the center of the dualist is achieved ( $h = 10, 22, 46, 94, \text{etc.}$ ). Bilateral symmetry is more frequent ( $h = 2, 6, 14, 18, 30, 34, 38, \text{etc.}$ ).

A recursive method to obtain the last vertical column for the core graphs in Table 3 with  $h \equiv 2 \pmod{4}$  is to take sequentially (row after row) the dualists from Table 2, then surround them by letters P so that all vertices obtain degree 3. Thus, starting from a dualist with  $h-1$  vertices in Table 2, one obtains a dualist with  $4h+2$  vertices in the last column of Table 3, and from this by appropriate substitution of one letter P by X, Y, or Z, one obtains all other dualists from Table 3.

Yet another method for obtaining all structures from Tables 2 or 3 is shown in Table 4. If  $P^3$  (previously denoted by P) is a subgraph with 3 vertices connected via its mid-point, one may form the series mentioned earlier as  $Y_k$  (in equations 2-4), which will be denoted by :

$p^1, p^3, p^7, p^{15}, p^{31}, \dots, p^h$  with  $h = 2^k - 1$  hexagons, whose dualists have two identical branches joined to a mid-point; each branch has only vertices of degrees 3 and 1.

There is no ambiguity in joining together  $k = 2$  such subgraphs by one edge, or  $k = 3$  such subgraphs to one extra vertex; however, in joining  $k = 4$  such subgraphs



to a pair of extra adjacent vertices, several non-isomorphic dualists are formed: from them only that one corresponds to  $K(\max)$  which has two identical subgraphs connected geminally, or when all four subgraphs are distinct, the one with the extremal subgraphs attached vicinally.

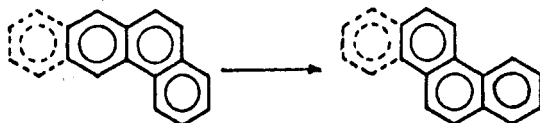
TABLE 4. Structures of dualists for  $K(\max)$  benzenoids formed by joining together  $k$  subgraphs of  $P^h$  series

h	$p^3$	$p^7$	$p^{15}$	$p^{31}$	k	h	$p^3$	$p^7$	$p^{15}$	$p^{31}$	k	h	$p^3$	$p^7$	$p^{15}$	$p^{31}$	k
6	2	-	-	-	2	26	1	1	1	-	3	42	1	1	-	1	3
10	1	1	-	-	2	1	3	-	-	4	1	1	2	-	-	4	
	3	-	-	-	3	3	-	1	-	4	3	-	-	-	1	4	
14	-	2	-	-	2	30	-	-	2	-	2	46	-	-	1	1	2
	2	1	-	-	3	-	2	1	-	3	-	2	-	1	-	3	
	4	-	-	-	4	-	4	-	-	4	-	-	-	3	-	3	
18	1	-	1	-	2	2	1	1	-	4	-	2	2	-	-	4	
	1	2	-	-	3	34	1	-	-	1	2	50	1	-	1	1	3
	3	1	-	-	4	1	-	2	-	3	1	1	-	3	-	4	
22	-	1	1	-	2	1	2	1	-	4	1	54	-	1	1	1	3
	2	-	1	-	3	36	-	1	-	1	2	2	2	-	1	1	4
	-	3	-	-	3	2	-	-	-	1	3	-	3	-	-	1	4
	2	2	-	-	4	-	1	2	-	3	-	-	-	-	-	-	4
						2	-	2	-	4	-						

## 2. EXPERIMENT

### 2.1 Isomerizations of benzenoids.

It had been reported (ref.15) that anhydrous aluminum chloride isomerized benz[a]anthracene into chrysene on refluxing in benzene. This reaction involves the conversion of a catafusene with fewer Kekulé structures into an isomer with higher K value. No mechanistic indications or speculations were presented.



(5)

**2.2 Automerizations of benzenoids.** In a paper published in 1965 (ref.16), speculating about the mechanism of the above reaction, I made the supposition that on deleting the anthracenic marginal ring indicated above by broken lines, one would arrive at the degenerate rearrangement of phenanthrene; by chopping one more benzenoid ring, it might even be possible to isomerize naphthalene into itself:

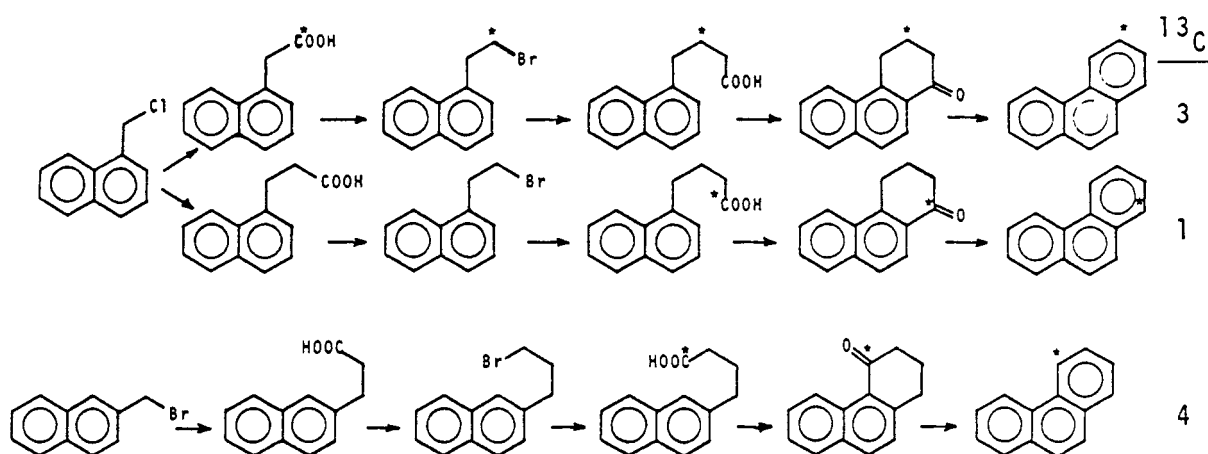


At that time, it was not possible to embark on a study of carbon-labeled phenanthrene, but for naphthalene it was easier. As indicated by the above formulas, such reactions involving bond-breaking and new bond formation, but conserving the chemical structure ("automerizations") would involve scrambling of isotopic labels. By synthesizing naphthalene labeled with carbon-14 in position 1, treating it with aluminum chloride, followed by stepwise degradation, conversion of the fragments into carbon dioxide and measuring the gas radioactivity, it appeared that a scrambling of the label did take place (ref.17). However, there were so many pitfalls in working with carbon-14 that we decided to check the results with carbon-13 and replace chemical degradations by NMR spectrometry. At that time this was impossible in Roumania, but in a cooperation with the Institute of Organic Chemistry in Novosibirsk we did check up the results. To our disappointment, we did not find any automerization under the above conditions so that in the previous experiments some unknown radiochemical impurity formed during the oxidative degradations must have led to erroneous data. Independently, Staab had also found that there was no catalytic automerization of naphthalene. In a letter exchange, I proposed to publish these findings in the same journal issue, but Staab refused, so that we published separately (refs. 18,19).

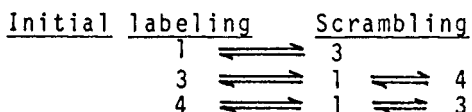
Then the problem lay dormant for ten years, till 1978, when Scott and his coworkers discovered (ref.20) that thermolysis of naphthalene (labeled with carbon-13) at 1000-1050° did lead to automerization. The mechanism of this reaction is still under investigation. No isomerization of anthracene into phenanthrene was observed under comparable conditions. An extensive and thorough study of many such thermolytic processes was made (ref.21).

In the last few years, in a cooperation with Mircea Gheorghiu, Ana Schiketanz (Racoveanu), Atena Necula, and Iuliu Pogany (refs.22-25), we returned to the initial speculation and reasoned that if the automerization (7) proceeds only thermally, perhaps the automerization (6) might take place catalytically, like the isomerization (5). Therefore, in a considerable synthetic effort outlined in the following formulas, phenanthrene labeled with carbon-13 in positions 1 or 3 was prepared and submitted to catalytic conditions (a melt of  $AlCl_3 + NaCl$  at 180-220°). The  $^{13}C$ -NMR assignments in deuterated acetone allow a clear distinction between all carbon atoms. Catalytic automerization does take place; in parallel, a dehydrogenating condensation also occurs (Scholl reaction, ref. 26), leading to higher condensed aromatics, therefore thermodynamic equilibration for the isotope scrambling cannot be attained.

Newer synthetic efforts by M.D.Gheorghiu, A.Racoveanu-Schiketanz and A. Necula led to the synthesis of phenanthrene labeled with carbon-13 in position 4. Its catalytic automerization under milder conditions ( $AlBr_3 + H_2O$ , 1:1 moles, in refluxing benzene, with 2 moles labeled phenanthrene) led to automerization with considerably less side-reactions.

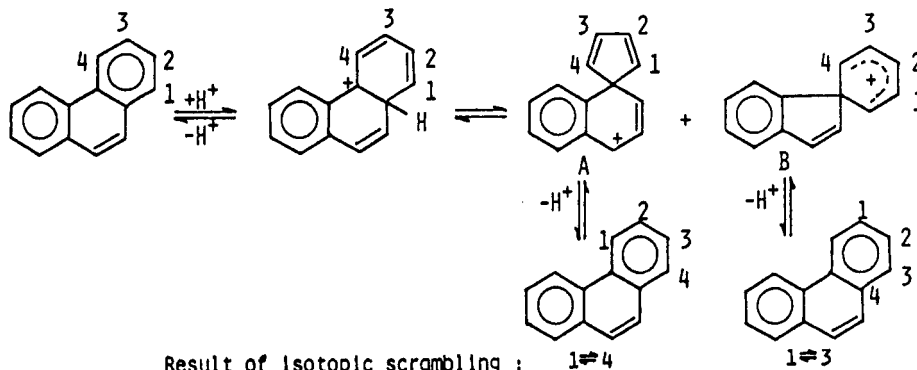


The experimental results on the catalytic automerization of phenanthrene may be summarized as follows :



Isotopic scrambling to quaternary sites is more difficult to observe because of low NMR peak intensities, therefore a special search must be undertaken for this purpose in the future.

The results obtained so far can be explained satisfactorily by the following mechanism. It is known that strong Lewis acids (aluminum or antimony halides) form with water Brönsted superacids which are able to protonate even sigma C-H bonds, leading to carbonium ions which can decompose into carbenium ions and hydrogen which is evolved. When such acids react with phenanthrene they will certainly protonate the 9,10 C=C bond, but this process does not lead to any reaction. However, protonation of other bonds does lead to rearrangements ; the simplest and most plausible mechanism (which by virtue of Occam's Razor can be considered as satisfactory for explaining the known facts till now) is outlined below :

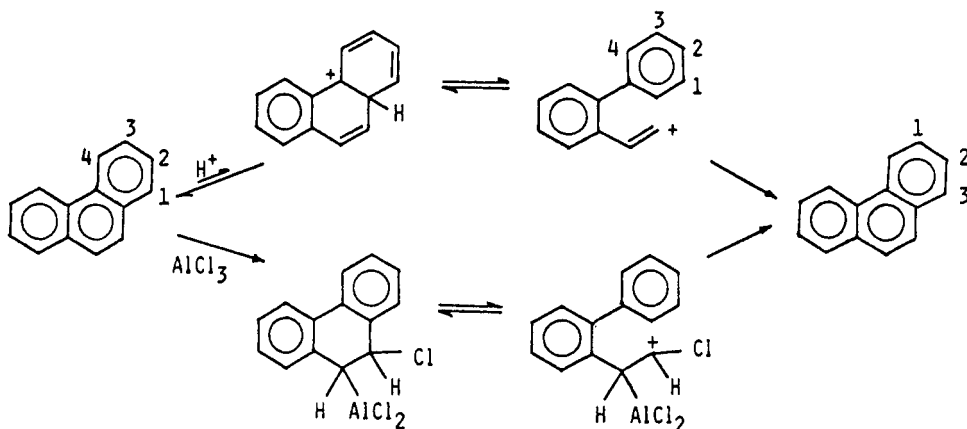


In all above formulas, we conserved for clarity the original numbering of carbon atoms as in the starting material.

Ab initio calculations at the STO-3G level performed by R. D. Topsom (La Trobe University, Bundoora, Australia) indicate that the intermediate A may be more stable than B by 2.6 kcal/mol.

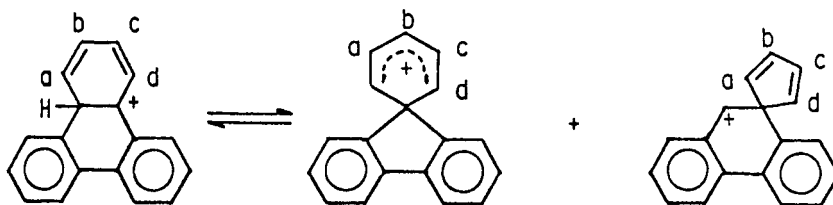
The above mechanism also explains why there is no catalytic automerization of naphthalene : no benzylic-type stabilization of cationic intermediates analogous to A is possible in that case (evidently, for naphthalene, A and B intermediates are identical).

Other mechanisms may be taken into consideration also, as shown below :



Such mechanisms may account for isotopic scrambling between positions 1 and 3, but not 4. Other conceivable mechanisms involve dications.

2.3 Outlook. It is possible that under similar catalytic conditions, triphenylene may undergo automerization :



The cyclohexadienylic cationic intermediate would lead to isotopic scrambling  $a \rightleftharpoons c$  between non-equivalent carbons in the starting material, while the benzylic cationic intermediate would undergo a label scrambling  $a \rightleftharpoons d$  between two equivalent carbons in triphenylene. A synthetic approach to the cyclohexadienylic cation might start from 9-cyclopentadienyl-9-fluorenyl.

Future work in the phenanthrene series will involve the search for detecting any scrambling involving quaternary carbon atoms, as well as the investigation of phenanthrene labeled with carbon-13 in position 2 : cation B conserves the labeling in position 2, but cation A leads to scrambling between positions 2 and 3, yet phenanthrene labeled in position 3 did not lead to detectable scrambling to position 2.

As far as catalytic isomerizations of benzenoids are concerned, the following two seem possible : dibenz[a,c]anthracene into the K(max) benzenoid with  $h = 5$ , and tetrabenz[a,c,h,j]anthracene into the K(max) benzenoid with  $h = 7$  ; however, in the latter case, a Scholl-type intramolecular ring closure to a perifusene might also occur.

### 3. CONCLUSIONS

Benzenoids with maximum number of Kekulé structures for given numbers of hexagons are branched catafusenes having no anthracenic portion ; the structures of these K(max) benzenoids present remarkable 4- and 3-periodicities in terms of the number  $h$  of benzenoid rings. These periodicities allow the prediction of such K(max) benzenoid structures, which may correspond to several isoarithmic structures differing only in the directions of kinks for their dualist graphs (refs.28,29).

Gas-phase thermolytic isomerizations and automerizations of benzenoids at temperatures around  $1000^\circ$  are likely to involve different mechanisms and intermediates than catalytic reactions taking place in liquid state at much lower temperatures, in conditions which are closer to organic chemical experiments.



It is interesting to note that most of the carcinogenic benzenoids have anthracenic subgraphs, whereas none of the K(max) benzenoids are among the known carcinogens. Since the former are less stable thermodynamically than the latter, it would be highly desirable to find an easy catalytic way to convert the carcinogenic catafusenes into the K(max) non-carcinogenic isomers ; so far, no feasible pathway for converting a non-branched catafusene into a branched one is known. Unfortunately, for carcinogenic perifusenes such as benzo[a]pyrene, no such hope exists.

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#### REFERENCES

1. L.Pauling and G.W.Wheland, J.Chem.Phys., 1933, 1, 362.
2. E.Clar, The Aromatic Sextet, Wiley, New York, 1972.
3. M.Randić, J.Am.Chem.Soc., 1977, 99, 444 ; Tetrahedron, 1977, 33, 1905.
4. M.Gordon and W.H.T.Davison, J.Chem.Phys., 1952, 20, 428.
5. A.T.Balaban and F.Harary, Tetrahedron, 1968, 24, 2505.
6. A.T.Balaban (ed.), Chemical Applications of Graph Theory, Academic Press, London - New York, 1976.
7. N.Trinajstić, Chemical Graph Theory, 2nd Ed., CRC Press, Boca Raton, FL 1992.
8. S.J.Cyvin and I.Gutman, Kekulé Structures in Benzenoid Hydrocarbons, Lecture Notes in Chemistry No.16, Springer, Berlin, 1988.
9. I.Gutman and S.J.Cyvin, Introduction to the Theory of Benzenoid Hydrocarbons, Springer, Berlin, 1989.
10. A.T.Balaban, Pure Appl.Chem., 1982, 54, 1075 ; J.Chem.Inf.Comput.Sci., 1985, 25, 334.
11. A.T.Balaban, Math.Chem.(Mulheim/Ruhr), 1989, 24, 29.
12. I.Gutman, J.Serbian Chem.Soc., 1988, 53, 607.
13. A.T.Balaban, X.Liu, S.V.Cyvin and D.J.Klein, submitted for publication.
14. A.T.Balaban, Annals of Discrete Math. 1992 (to appear).
15. A.Dansi and E.Salvioni, Gazz.Chim.Ital., 1949, 71, 549 ; N.P.Buu-Hoi and D.Lavit-Lamy, Proc.Chem.Soc., 1960, 120 ; Bull.Soc.Chim.France, 1961, 1657 ; 1962, 1398 ; 1963, 341 ; D.Lavit-Lamy and N.P.Buu-Hoi, Chem.Commun., 1966, 92 ; Bull.Soc.Chim.France, 1966, 2613, 2619.
16. A.T.Balaban and Z.Simon, Rev.Roum.Chim., 1965, 10, 1059.
17. A.T.Balaban and D.Farcasiu, J.Am.Chem.Soc., 1967, 89, 1958.
18. A.T.Balaban, D.Farcasiu, V.A.Koptyug, I.S.Isaev, M.I.Gorfinkel and A.T.Rezvukhin, Tetrahedron Lett., 1968, 4757.
19. H.A.Staab and M.Haenel, Angew.Chem.Int.Ed.Engl., 1968, 7, 52 ; Chem.Ber., 1970, 103, 1095.
20. L.T.Scott and G.K.Agopian, J.Am.Chem.Soc., 1977, 99, 4506.
21. L.T.Scott, Acc.Chem.Res., 1982, 15, 52.
22. A.T.Balaban, M.D.Gheorghiu, A.Schiketanz and A.Necula, J.Am.Chem.Soc., 1989, 111, 735.
23. A.Schiketanz, I.Pogany, M.D.Gheorghiu, A.Necula and A.T.Balaban, J.Labelled Comp.Radiopharm., 1989, 27, 971.
24. M.D.Gheorghiu, A.Schiketanz, A.Necula, I.Pogany and A.T.Balaban, in Synthesis and Applications of Isotopically Labelled Compounds (eds. T.A.Baillie and J.R.Jones), Elsevier, Amsterdam, 1989, p. 689.
25. M.D.Gheorghiu, A.Schiketanz, A.Necula and A.T.Balaban, Rev.Roum.Chim., 1989, 34, 1005.
26. A.T.Balaban and C.D.Nenitzescu, in Friedel-Crafts and Related Reactions, ed. G.A.Olah, Wiley-Interscience, New York, 1964, vol.2, p.979.
27. M.D.Gheorghiu, A.Racoveanu-Schiketanz and A.Necula, to be published.
28. A.T.Balaban, Rev.Roum.Chim., 1977, 22, 45.
29. I.Gutman, Theoret.Chim.Acta (Berlin), 1977, 45, 309.