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A RE-VISIT TO THE RELATIVE IMPORTANCE OF KEKULÉ STRUCTURES

On a novel non-numerical ordering scheme

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ABSTRACT

A purely graph-theoretical (non-numerical) method is described to order a set of Kekulé structures. The method is based on transforming the VB structures into the subspace of their double bonds to form the corresponding factor-graphs. Two factor graphs are connected only if they are related by a degenerate transformation (refs 5, 11). The resulting hierarchies lead to ordering MO-VB quantities such as Kekulé indices as well as many graph-theoretical properties including connectivities, non-adjacent numbers, spectral moments and conjugated circuits. A topological analysis of the results is given.

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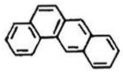
INTRODUCTION AND STATEMENT OF THE PROBLEM

Kekulé's theorem of a hexagonal structure for benzene laid the basis of a great deal of theoretical organic chemistry. The results of VB calculations would be of considerable interest as they would indicate the relative weights of different Kekulé-type structures. It has often been assumed that all Kekulé structures have identical weights. For example Pauling bond order concept[1, 2] implies the same weight for all Kekulé structures belonging to a given hydrocarbon. However, the idea that some valence structures are more important than others emerged as early as 1927 in the form of an empirical rule suggested by Fries[3]. Nearly a decade ago an index, called Kekulé index, (K(L), was introduced[4] and is given by eqn. 1, viz.,

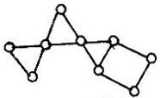
$$K(L) = \frac{1}{2N} \sum_{(u,\nu)\in L} (q_u + q_\nu + 2\, p_{u\nu})^{\frac{1}{2}} \tag{1}$$

where the symbols have their conventional meanings. Recently one of the authors studied the graph-theoretical properties of graphs resulting when the Kekulé VB structures are transformed into the subspace of their double bonds[5-16]. Such graphs are now known in literature as factor graphs[17]. As an illustration we consider some Kekulé structures and their corresponding factor graphs, F(k)'s:

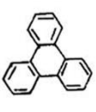


k_1

→

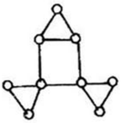


F(k_1)



k_2

→



F(k_2)

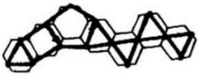
(See later for definitions)

A relation appeared to exist[5-8] between certain graph-theoretical properties of the F(k)'s and the Kekulé indices of the corresponding Kekulé structures. Of these properties, the two most heavily studies are the connectivity index[18] of F(k), χ and Hosoya's Z function[19] given by the sum of nonadjacent numbers of an F(k). As a measure of connectivity of an F(k) we took the reciprocal of the connectivity index of Randić[18], i.e.

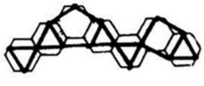
$$\chi = \Big[\sum_{i,j} (v_i v_j)^{-\frac{1}{2}} \Big]^{-1} \qquad (2)$$

where v_i is degree (valency) of ith vertex and the summation is taken over all edge-types[18] in the factor graph. The concept of F(k) has also led to the discovery of important topological properties[12-15] of benzenoid hydrocarbons. An account of the mathematical properties of these graphs is available[9]. Although the general conclusion reached from the above studies (particularly refs. 5-8, 16) is that the graph-theoretical properties of the F(k)'s almost preserve the ordering of the individual k's based on numerical values of their Kekulé indices, (and thus offered a novel insight into the problem of assigning weights to the VB structures), yet on studying more involved situations, violations appeared which seem to be more than just discrepancies. The following examples illustrate the problem (the numbers are, respectively from left to right, conjugation circuits[20], connectivity indices, eqn. 2, and Kekulé indices, eqn. 1): Throughout the paper F(k)'s are heavily outlined into their benzenoid graphs.

1)

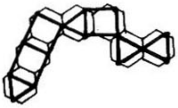


$5R_1+R_2+R_3$, 0.8755, 0.1364

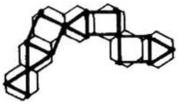


$5R_1+R_2+R_3$, 0.8812, 0.1363

2)

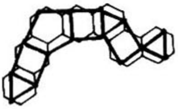


$4R_1+2R_2+R_3$, 0.8712, 0.1365

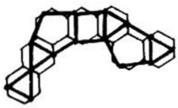


$4R_1+3R_2$, 0.8769, 0.1364

3)



$3R_1+3R_2+R_3+R_5+R_6$,
0.8564, 0.1357



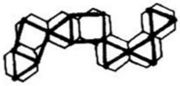
$4R_1+R_2+R_3+2R_4+R_6$
0.8622, 0.1355

4)

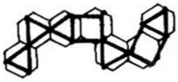
$6R_1+R_3$, 0.8794
0.1372

$5R_1+2R_2$, 0.8769,
0.1373

5)



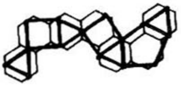
$5R_1+2R_2$, 0.8781,
0.1373



$5R_1+2R_2$, 0.8757,
0.1373

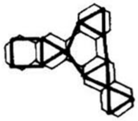
6)

$4R_1+2R_2+2R_3$,
0.8667, 0.1363



$4R_1+2R_2+R_3+R_4$,
0.8643, 0.1364

7)



$4R_1+R_2+R_3$,
0.8714, 0.1561

$3R_1+2R_2+R_3$,
0.8573, 0.1559

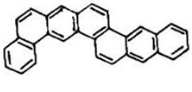
8)

$4R_1+R_2+R_5$,
0.8667, 0.1558

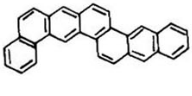
$3R_1+2R_2+R_5$,
0.8526, 0.1558

The above pairs are symptomatic of some five hundred Kekulé structures studied. They all represent inconsistent orderings of the Kekulé structures: thus our previous studies[6-9] indicate that the value of $\varkappa$ (eqn. 2) of an F(k) goes up as K(L) of the corresponding k's (eqn. 1) goes up. The above examples, thus, seem to illustrate counterexamples of this conclusion. In a recent paper, Randić[21], in an attempt to develop some adequate and sufficiently general structural vocabulary to order a tuplet of Kekulé VB structures suggested the use of his conjugated circuits[22]. Although such a proposition seems simple and fundamental, it does not seem to be discriminative enough to the ordering purpose as we demonstrate from the following quartet of Kekulé structures: they all have a conjugation content given by $\underline{5R_1+2R_2}$, and also the same branching index, $\varkappa$ (eqn. 2) but their K(L)'s are not all identical:

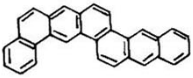
9)



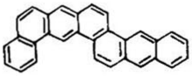
K(L) = 0.8859



0.8859



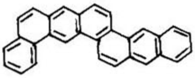
0.8859



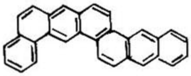
0.8830

Another illustration is provided by the following pair:

10)



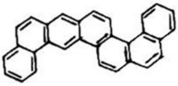
$5R_1+R_2+R_3$
0.8829, 0.1363



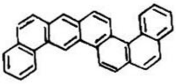
$5R_1+R_2+R_3$
0.8800. 0.1360

In the above pair K(L) and χ values lead to identical ordering i.e. both predict that the k to the left is "more important" than the one to the right but the conjugated circuits are the same for both. Some other situations are desirable to demonstrate that neither Randić's circuits of the k's nor connectivity of the F(k), χ, are adequate ordering parameters is illustrated below:

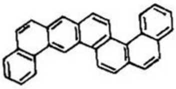
11)



$5R_1+2R_2$, $\underline{0.8781}$, 0.1373



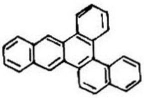
$5R_1+2R_2$, $\underline{0.8757}$, 0.1373



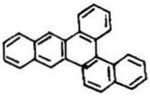
$5R_1+2R_2$, $\underline{0.8769}$, 0.1373

For the above triplet only the Kekulé index, the middle number, succeeds in distinguishing the individual double-bond permutations.

Examples from branched hydrocarbons are given below

12) 

$5R_1+R_2$, 0.8850, 0.1583



$5R_1+R_2$, 0.8762, 0.1575

The above and such numerous examples indicate that numerical ordering of Kekulé structures using Kekulé indices may lead to inconsistencies when compared with orderings based on graph-theoretical properties of the factor graphs. Furthermore neither connectivity nor conjugated circuits are sufficient discriminators of two or more double bond permutations representing the Kekulé structures. Therefore the search for more exact non-numerical method of ordering a particular set of valence structures seems desirable. The method we propose here introduces a definition for a greater relation[22] among a tuplet of Kekulé structures. Furthermore, we propose a definition of comparability[23] of a set of Kekulé structures based on purely non-numerical conditions. Such a definition allows the prediction of "inconsistencies" as exemplified by situations 1-12 given above. Thus this paper deals mainly with the problem of ordering[24] which became of interest recently in the chemical literature, but using Kekulé structures of benzenoid hydrocarbons as our substrates. The important role played by Kekulé structures in organic chemistry[25] together with the relatively novel concept of ordering in chemistry justify this work.

DEFINITIONS AND PRELIMINARIES

We let $\hat{F}$ be defined as an operator which transforms an ith structure, k_i, into the subspace of its double bonds to give the corresponding factor graph, $F(k_i)$. An inverse operator, $\hat{F}^{-1}$ is defined, thus

$$\hat{F}k_i = F(k_i) \;; \tag{3}$$

$$\hat{F}^{-1}F(k_i) = k_i \tag{4}$$

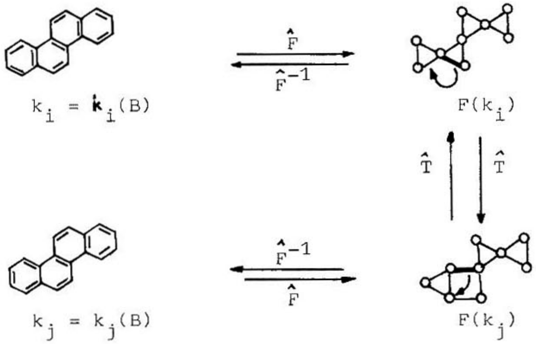
A quantity, σ_i to be associated with a particular $F(k_i)$ defined as follows: let the size of a cycle in some $F(k)$ be m, then a number is given to this cycle = m-2. The sum of all numbers given to all cycles in an $F(k_i)$ will be called $\sigma_{F(k_i)}$ or simply σ_i. E.g. σ_1 corresponding to $F(k_1) = 1+1+1+2 = 5$.

We define a transformation operator, $\hat{T}$, between two $F(k)$'s, as an operator which rearranges one edge of an $F(k_i)$ to an adjacent vertex to generate another factor graph, $F(k_j)$, thus

$$\hat{T}F(k_i) = F(k_j) \tag{5}$$

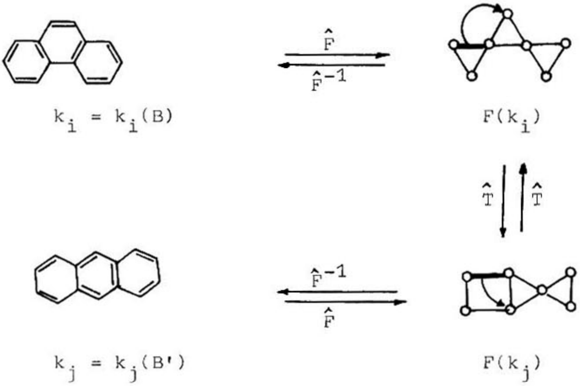
Eqn 5 represents three possibilities:

i) k_i and k_j both belong to the same benzenoid hydrocarbon, B i.e. K_i, $k_j \in B$. E.g. when B = chrysene we might have:



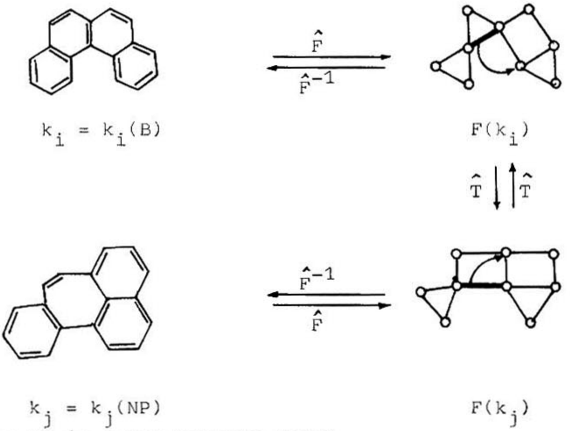
The above situation defines what has been called before[10], a degenerate transformation.

ii) $k_i \in B$; $k_j \in B'$ but both B and B' are polyhex graphs[26]. E.g. B = phenanthrene, B' = anthracene



Case ii) represents a <u>non-degenerate transformations</u>.

iii) A transformation leading to a non-polyhex graph, E.g.



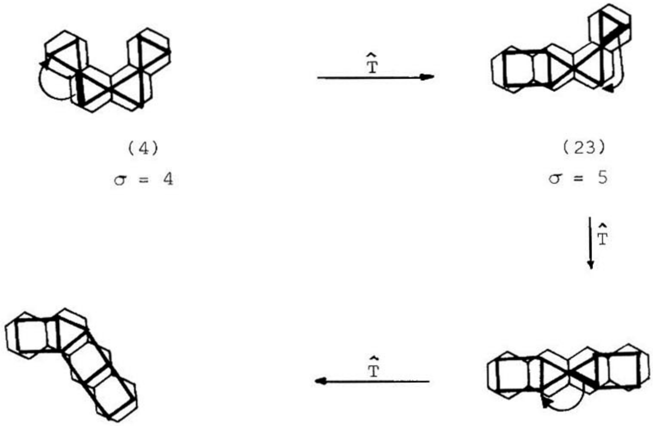
where NP is a non-polyhex graph.

In this paper only degenerate transformations (type i) involving catacondensed benzenoid hydrocarbons will be considered.

A detailed account of the mathematical properties of the above transformations is under consideration.

Transformation hierarchy

Consider an $F(k)$ containing R cycles. Let $\hat{T}$ successively operates on it (eqn. 5) to generate R transformates: $F(k_i)$, $F(k_j)$, ... , $F(k_R)$ ordered such that $\sigma_i < \sigma_j < \ldots < \sigma_R$. The above set of $F(k)$'s is said to define a transformation hierarchy. As an illustration we consider a series of $\hat{T}$ operations on benzo[c]phenanthrene: (Factor graphs are properly implanted into molecular graphs of the corresponding hydrocarbons)



(4) $\sigma = 4$

(23) $\sigma = 5$

(1) $\sigma = 6$

(28) $\sigma = 7$

(Numbers refer to Fig 1)

When the transformation hierarchy involves only degenerate transformations, it will be called a degenerate transformation hierarchy. This latter type is the subject of this paper (see Results Section).

THE MAIN RESULT: A non-numerical definition of a greater relation among a set of Kekulé structures:
In a set of F(k)'s defining a transformation hierarchy, $F(k_i)$ is defined here to be greater than $F(k_j)$, written $F(k_i) \supset F(k_j)$ if $\sigma_i < \sigma_j$.

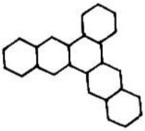
Conjecture 1
When $F(k_i) \supset F(k_j)$, then:
(a) Graph-theoretical properties (connectivity, χ, eqn 2, sum of nonadjacent numbers of Hosoya, as well as other parameters[27]) will be greater for $F(k_i)$, (b) k_i will be more important Kekulé structure than k_j as measured from its K(L), eqn. 1, and conjugated circuits.

A non-comparability criterion
$F(k_i)$ is non-comparable with $F(k_j)$ if (a) $\sigma_i = \sigma_j$ and (b) $\hat{T}\, F(k_i) = F(k_j)$ is not defined. Discrepancies (inconsistent orders) are expected to occur among such pairs. Examples being cases 1-8 above.

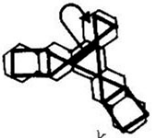
The definitions of greater relation and non-comparability allow a consistent non-numerical ordering of a set of Kekulé structures of the factor graphs.

RESULTS

Throughout this work Pauling bond orders[1, 2] are used in place of MO bond orders. (The latter are sometimes not available). The two parameters are known to be proportional[1, 2]. Eqn. 1 was used to calculate K(L) which simplifies more because $q_u = q_\nu = 1$ for alternant hydrocarbons. We studied degenerate hierarchy transformations for the Kekulé structures of all cata-condensed benzenoid hydrocarbons containing 2, 3, 4, 5, 6 and 7 rings. The results support our conjecture and prediction for inconsistent orders. It is not practical, however, to list all the individual cases studied. Therefore we will mention only a few representative examples but other data are available on the request.

I.  $K = 19$

We will consider all its Kekule structures. The following degenerate transformations are identified (Numbers from left to right are conjugated circuits, K(L) and $\varkappa$).

1)  $4R_1+2R_2$, 0.8620, 0.15596

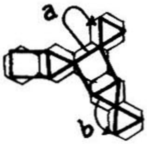
k_{11}

↓

 $3R_1+3R_2$, 0.8597, 0.15547

k_{12}

2)

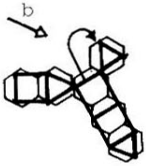


$4R_1+2R_2$, 0.8737, 0.1567

k_{13}

$4R_1+R_2+R_3$,
0.8714, 0.1561

k_{14}



$3R_1+2R_2+R_3$,
0.8573, 0.15596

k_{15}

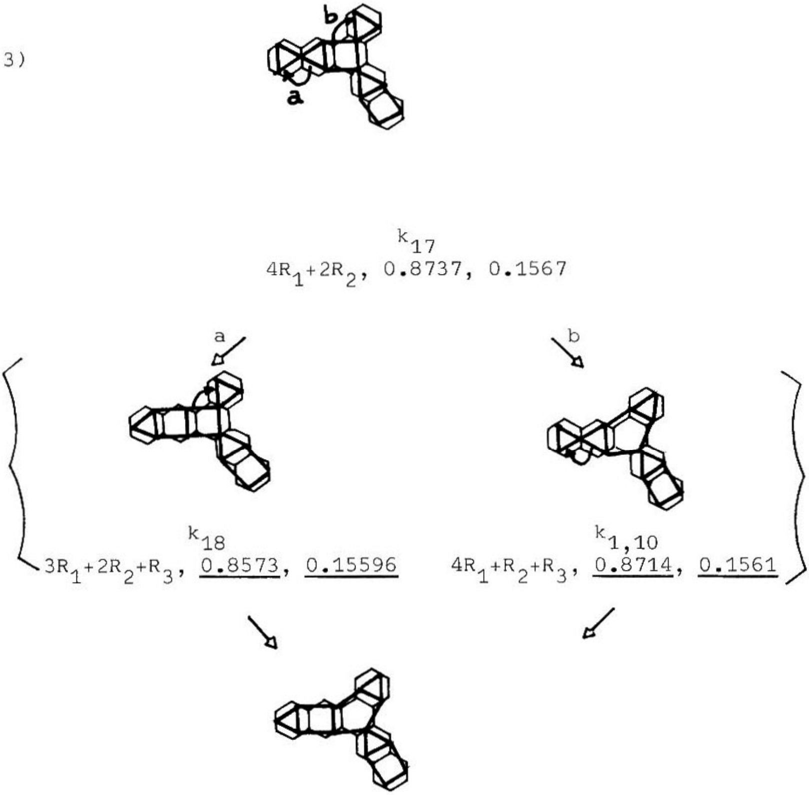
k_{16}
$3R_1+2R_2+R_4$,
0.8550, 0.1553

We define the following greater relations:

$$k_{11} \supset k_{12}, \quad (\sigma_{11} = 8,\ \sigma_{12} = 9)$$
$$k_{13} \supset k_{14} \supset k_{16} \quad (\sigma_{13} = 8,\ \sigma_{14} = 9,\ \sigma_{16} = 10)$$
$$k_{13} \supset k_{15} \supset k_{16} \quad (\sigma_{13} = 8,\ \sigma_{15} = 9,\ \sigma_{16} = 10)$$

An example of a non-comparable pair is $\{k_{14}, k_{15}\}$. As expected values of χ and K(L) lead to inconsistent orders. Such pairs (or higher tuplets) of non-comparable VB structures will be placed in braces.

3)



k_{17}
$4R_1+2R_2$, 0.8737, 0.1567

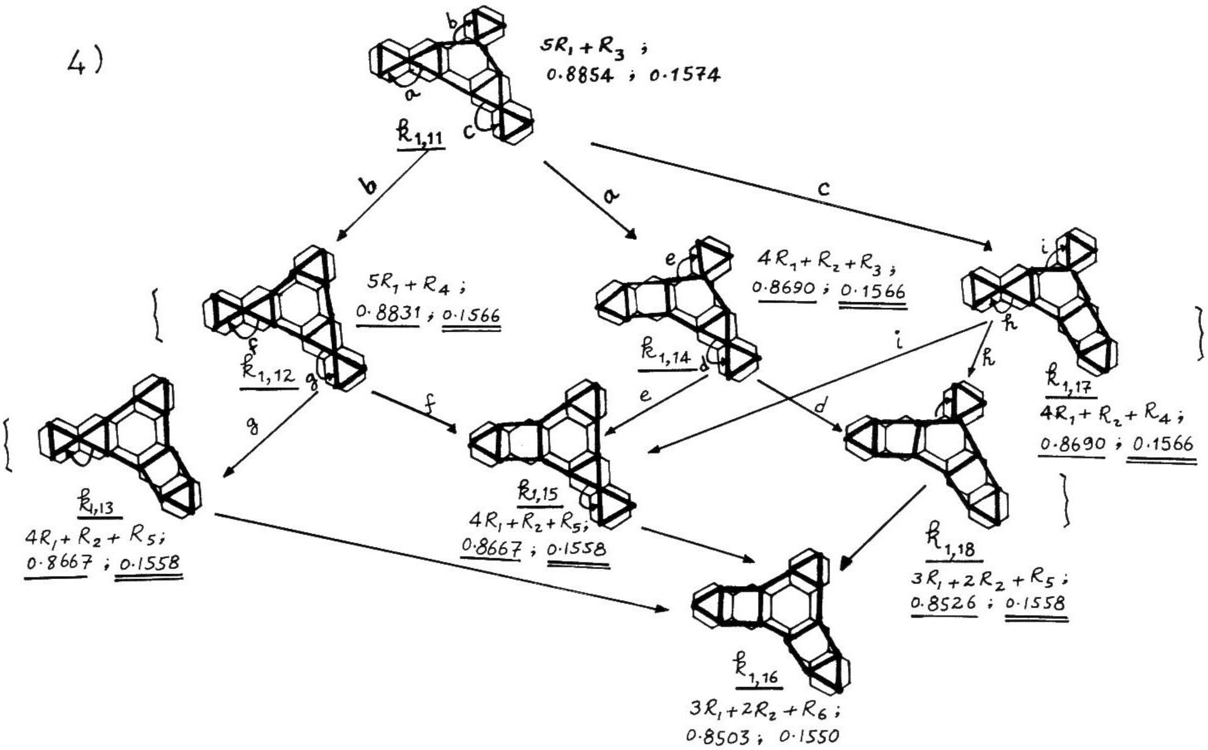
k_{18}
$3R_1+2R_2+R_3$, $\underline{0.8573}$, $\underline{0.15596}$

$k_{1,10}$
$4R_1+R_2+R_3$, $\underline{0.8714}$, $\underline{0.1561}$

k_{19}
$3R_1+2R_2+R_4$, 0.8550, 0.1553

Again the pair $\{k_{18}, k_{1,10}\}$ is non-comparable. Analogous greater relations might be defined.

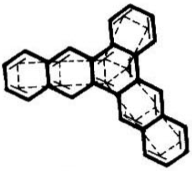
4)


$\mathcal{R}_{1,11}$
$5R_1 + R_3$;
0.8854 ; 0.1574
a
b
c
$\mathcal{R}_{1,12}$
$5R_1 + R_4$;
0.8831 ; 0.1566
f
g
$\mathcal{R}_{1,14}$
$4R_1 + R_2 + R_3$;
0.8690 ; 0.1566
e
d
$\mathcal{R}_{1,17}$
$4R_1 + R_2 + R_4$;
0.8690 ; 0.1566
i
h
$\mathcal{R}_{1,13}$
$4R_1 + R_2 + R_5$;
0.8667 ; 0.1558
$\mathcal{R}_{1,15}$
$4R_1 + R_2 + R_5$;
0.8667 ; 0.1558
$\mathcal{R}_{1,18}$
$3R_1 + 2R_2 + R_5$;
0.8526 ; 0.1558
$\mathcal{R}_{1,16}$
$3R_1 + 2R_2 + R_6$;
0.8503 ; 0.1550

Several greater relations might be identified, e.g. $k_{1,11} \supset k_{1,12} \supset k_{1,13} \supset k_{1,16}$. Non-comparable triplets are placed in braces. One observes that some non-comparable pairs might be symmetry-equivalent, e.g. $\{k_{1,14}, k_{1,17}\}$.

An odd Kekulé structure

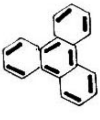
If one of the Kekulé structures is not transformable into another member of the set, it will be called an odd member. For the case at hand such a member is shown below.



$R_1+3R_2+3R_3+2R_4+R_5$
0.8386, 0.15596

$k_{1,19}$

It has been suggested[10, 28] that such Kekulé structures have no contribution to the total VB wave function of the benzenoid hydrocarbon. Such a view has been previously conjectured by Clar[29] for analogous structures. E.g. a Kekule structure of triphenylene in which only the central ring has an aromatic sextet does not represent the actual state of the molecule[29].

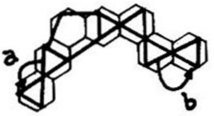


k_3

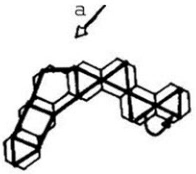
In fact such structures lead to inconsistent orderings (as predicted, being non-transformable to any other structure by $\hat{T}$). E.g. $\chi(k_{1,19})$ predicts $k_{1,19}$ to be one of the most important states, but its K(L) predicts it to be the least important. The same observation holds for k_3[5, 11].

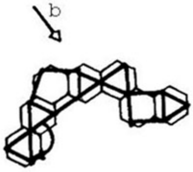
Instead of going through all the transformations in other hydrocarbons studied, we will mention only some selected cases to illustrate our concepts of greater relation and of non-comparability.

II.

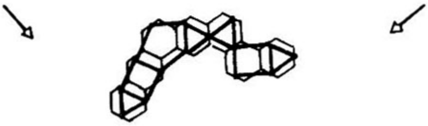


$6R_1+R_3$, 0.8912, 0.1370



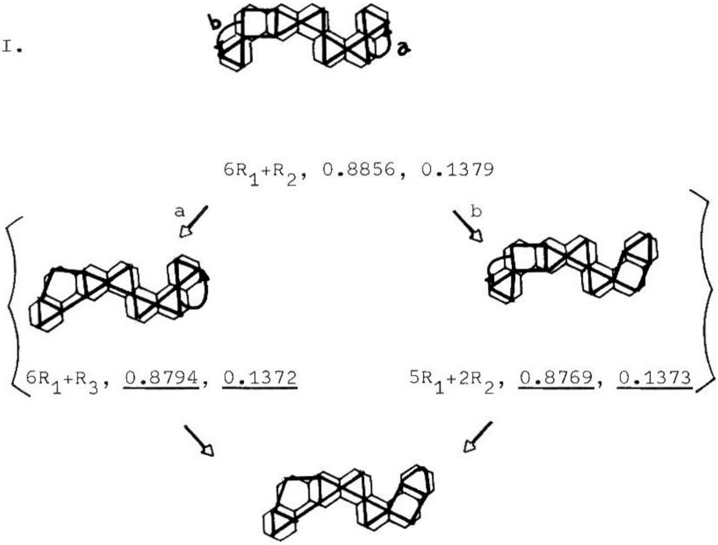


$5R_1+R_2+R_3$, $\underline{0.8755}$, $\underline{0.1364}$ $5R_1+R_2+R_3$, $\underline{0.8812}$, $\underline{0.1363}$



$4R_1+2R_2+R_4$, 0.8655, 0.1357

III.



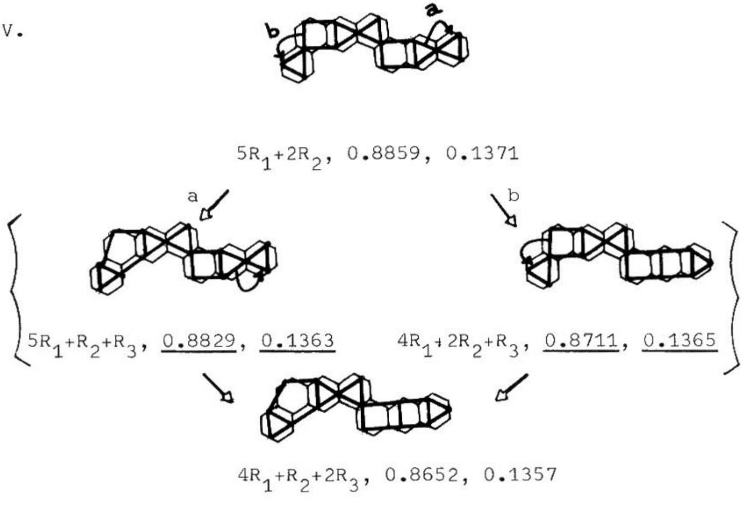
$6R_1+R_2$, 0.8856, 0.1379

$6R_1+R_3$, $\underline{0.8794}$, $\underline{0.1372}$

$5R_1+2R_2$, $\underline{0.8769}$, $\underline{0.1373}$

$5R_1+R_2+R_3$, 0.8706, 0.1365

IV.



$5R_1+2R_2$, 0.8859, 0.1371

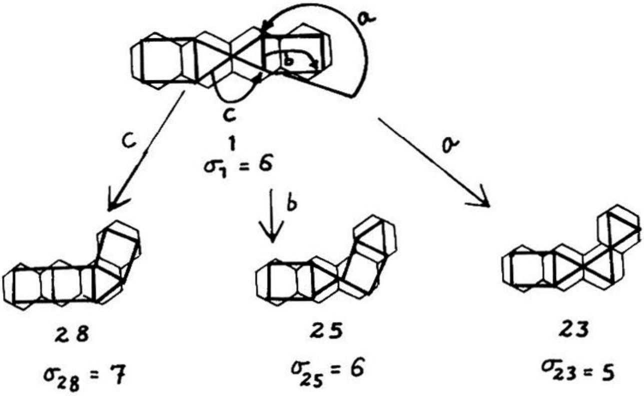
$5R_1+R_2+R_3$, $\underline{0.8829}$, $\underline{0.1363}$

$4R_1+2R_2+R_3$, $\underline{0.8711}$, $\underline{0.1365}$

$4R_1+R_2+2R_3$, 0.8652, 0.1357

We conclude our results by exposing all transformation hierarchies involved in all F(k)'s belonging to nonbranched cata-condensed benzenoid hydrocarbons containing four rings. Such F(k)'s are shown in Fig 1. Two members which are related by eqn. 5 define a greater relation between them. Some graph-theoretical properties of these F(k)'s as well as the Kekulé indices of the corresponding Kekulé structures are listed in Table 1.

We consider, e.g. graph (1) and all possible $\hat{T}$ transformates: (Numbers refer to Fig. 1).



The above hierarchy may be expressed by writing

$$\hat{T}: 1 \longrightarrow \{23, 25, 28\}$$

From the values of the σ's, the above mapping leads to the greater relation : $1 \supset 28$

Other $\hat{T}$ mappings are as follows

$$\hat{T}: \; 2 \longrightarrow \{4, 18, 22, 23, 24, 28\}$$
$$\hat{T}: \; 3 \longrightarrow \{5, 17, 22, 23, 24, 28\}$$
$$\hat{T}: \; 4 \longrightarrow \{2, 21, 25, 26, 27, 28\}$$

$\hat{T}: 5 \longrightarrow \{3, 20, 25, 26, 27, 28\}$
$\hat{T}: 6 \longrightarrow \{7, 8, 22, 23\}$
$\hat{T}: 7 \longrightarrow \{6, 11, 23, 24, 25, 26\}$
$\hat{T}: 8 \longrightarrow \{6, 11, 23, 24, 25, 26\}$
$\hat{T}: 9 \longrightarrow \{13, 22, 23, 26, 28\}$
$\hat{T}: 10 \longrightarrow \{12, 22, 23, 26, 28\}$
$\hat{T}: 11 \longrightarrow \{7, 8, 25, 27\}$
$\hat{T}: 12 \longrightarrow \{10, 20, 24, 25, 27, 28\}$
$\hat{T}: 13 \longrightarrow \{9, 21, 24, 25, 27, 28\}$
$\hat{T}: 14 \longrightarrow \{15, 16, 22, 23\}$
$\hat{T}: 15 \longrightarrow \{14, 19, 23, 24, 25, 26\}$
$\hat{T}: 16 \longrightarrow \{14, 19, 23, 24, 25, 26\}$
$\hat{T}: 17 \longrightarrow \{3, 20, 22, 23, 26, 28\}$
$\hat{T}: 18 \longrightarrow \{2, 21, 22, 23, 26, 28\}$
$\hat{T}: 19 \longrightarrow \{15, 16, 25, 27\}$
$\hat{T}: 20 \longrightarrow \{5, 12, 17, 24, 25, 27, 28\}$
$\hat{T}: 21 \longrightarrow \{4, 13, 18, 24, 25, 27, 28\}$
$\hat{T}: 22 \longrightarrow \{2, 3, 6, 9, 10, 14, 17, 18, 24, 26\}$
$\hat{T}: 23 \longrightarrow \{1, 2, 3, 6, 7, 8, 9, 10, 14, 15, 16, 17, 18, 25\}$
$\hat{T}: 24 \longrightarrow \{2, 3, 7, 8, 12, 13, 15, 16, 20, 21, 22, 26, 27\}$
$\hat{T}: 25 \longrightarrow \{1, 4, 5, 7, 8, 11, 12, 13, 15, 16, 19, 20, 21, 23\}$
$\hat{T}: 26 \longrightarrow \{4, 5, 7, 8, 9, 10, 15, 16, 17, 18, 22, 24, 27\}$
$\hat{T}: 27 \longrightarrow \{4, 5, 11, 12, 13, 19, 20, 21, 24, 26\}$
$\hat{T}: 28 \longrightarrow \{1, 2, 3, 4, 5, 9, 10, 12, 13, 17, 18, 20, 21\}$

Greater relations might easily be derived from the values of the σ's. These are graphically represented by connecting members defining greater relations in Fig 1. In no case our conjecture 1 was violated. This might be inferred from Fig 1 and Table 1. The situation exemplified here is symptomatic of other transformation hierarchies involving hydrocarbons containing higher number of rings.

Fig. Legend

Fig. 1

Factor graphs of nonbranched catacondensed benzenoid hydrocarbons containing four rings (They are boldly outlined and properly implanted into their corresponding polyhex graphs). Four levels of importance are defined, viz., $\sigma = 4, 5, 6, 7$. Members defining greater relations are connected.

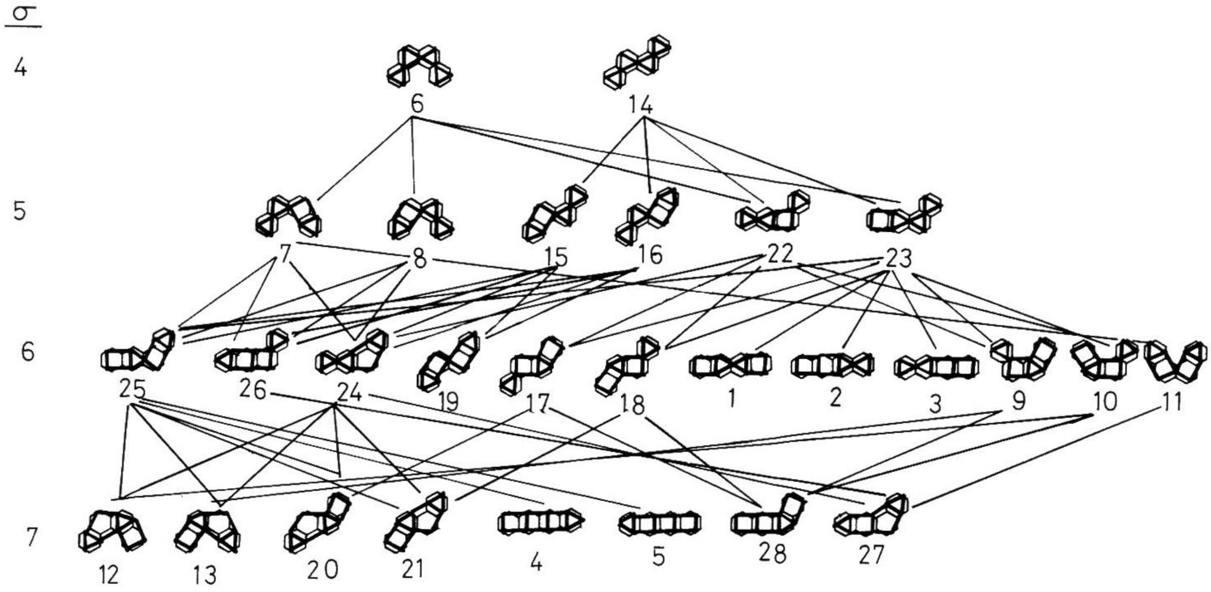

σ
4
5
6
7
6
14
7
8
15
16
22
23
25
26
24
19
17
18
1
2
3
9
10
11
12
13
20
21
4
5
28
27

FIG 1

Topological description of degenerate transformations

A set of Kekulé structures belonging to a particular hydrocarbon are divided into subsets according to $\hat{T}$ transformations defined in the set. We exemplify the method by considering the Kekulé structures of the branched hydrocarbon, I. Five subsets are identified and shown in Fig 2. We define the following relations: T_1 being a relation from A to B, T_2 as a relation from B to C and T_3 from C to D. Then we may write

$$T_1 \subset A \times B,$$

$$T_2 \subset B \times C,$$

$$T_3 \subset C \times D.$$

The following graphs are defined for the T relations:

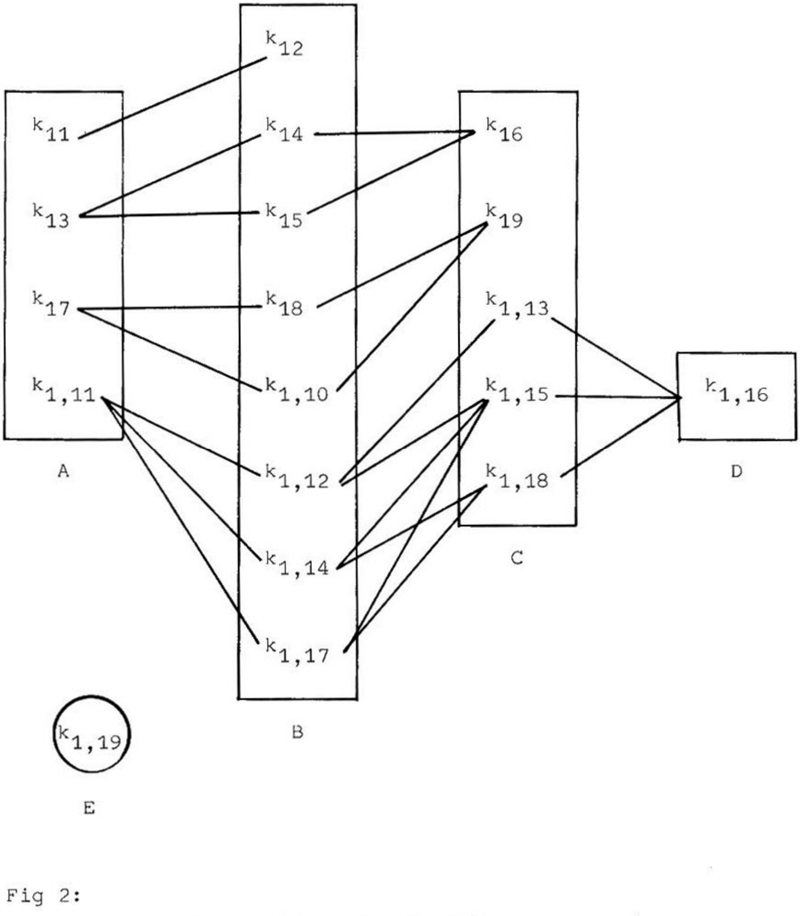


Fig 2:
Degenerate transformations involved in the Kekulé structures of hydrocarbon I. Pairs or triplets of connected graphs define greater relations. Set E is an "odd member".

$$T_1 = \{ < k_{11}, k_{12} > , < k_{13}, k_{14} > , < k_{13}, k_{15} > ,$$

$$< k_{17}, k_{18} > , < k_{17}, k_{1,10} > , < k_{1,11}, k_{1,12} > ,$$

$$< k_{1,11}, k_{1,14} > , < k_{1,11}, k_{1,17} > \} ;$$

$$T_2 = \{ < k_{14}, k_{16} > , < k_{15}, k_{16} > , < k_{18}, k_{19} > ,$$

$$< k_{1,10}, k_{19} > , < k_{1,12}, k_{1,13} > , < k_{1,12}, k_{1,15} > ,$$

$$< k_{1,14}, k_{1,15} > , < k_{1,14}, k_{1,18} > , < k_{1,17}, k_{1,15} > ,$$

$$< k_{1,17}, k_{1,18} > \} ;$$

$$T_3 = \{ < k_{1,13}, k_{1,16} > , < k_{1,15}, k_{1,16} > , < k_{1,18}, k_{1,16} > \}$$

One might identify pairs such as:

$$< k_{13}, k_{16} > \in T_2 \circ T_1 ,$$

$$< k_{1,16}, k_{1,12} > \in T_3 \circ T_2 \quad \text{etc.}$$

It must be observed that the T_i relations ($i = 1, 2, 3$) are not one-to-one functions because T transforms a particular element of a subset into two or more elements of another set. The topological spaces, A, B, C, D are thus not hemeomorphic

Conclusion

We developed a purely graph-theoretical (non-numerical) method for consistently ordering a set of Kekulé structures, not necessarily, all belonging to one hydrocarbon. The resulting hierarchies reproduces relative orders of Kekulé indices and conjugated circuits of the VB structures as well as several graph-theoretical parameters of the associated factor graphs. Such graph-theoretical parameters include connectivity, non-adjacent numbers and spectral moments of the factor graphs. The method induces topological descriptions of the individual hydrocarbons.

Table 1
MO-VB and Graph-Theoretical properties of Factor-graphs corresponding to all Kekulé structures of nonbranched cata-condensed benzenoid hydrocarbons containing four rings. Numbers are ordered from left to right correspondingly with Fig 1.

Level: σ =	Property
	K(L): (Quoted from Ref. 4)
4	0.912, 0.912
5	0.9069, 0.9069, 0.9069, 0.9069, 0.9112, 0.9083
6	0.9042, 0.9028, 0.9071, 0.901, 0.902, 0.902, 0.909, 0.908, 0.908, 0.9022, 0.9022, 0.9012
7	0.8964, 0.8964, 0.8964, 0.8964, 0.898, 0.898, 0.896, 0.8986.
	Conjugated Circuits[20]
4	$(4R_1)$, $(4R_1)$
5	$(3R_1+R_2)$, $(3R_1+R_2)$, $(3R_1+R_2)$, $(3R_1+R_2)$, $(3R_1+R_2)$, $(3R_1+R_2)$
6	$(2R_1+2R_2)$, $(2R_1+R_2+R_3)$, $(3R_1+R_3)$, $(2R_1+2R_2)$, $(2R_1+2R_2+R_3)$, $(2R_1,2R_2+R_3)$, $(2R_1+2R_2)$, $(2R_1+R_2+R_3)$, $(2R_1+2R_2+R_3)$, $(2R_1+2R_2+R_3)$, $(2R_1+2R_2+R_3)$, $(2R_1+2R_2)$
7	$(2R_1+R_2+R_3+R_4)$, $(2R_1+R_2+R_3+R_4)$, $(2R_1+R_2+R_3+R_4)$, $(2R_1+R_2+R_3+R_4)$, $(R_1+R_2+R_3+R_4)$, $(R_1+R_2+R_3+R_4)$, $(R_1+2R_2+2R_3+R_4)$, $(2R_1+R_2+R_4)$
	Nonadjacent number sums[19] defined as $\sum_{i=2}^{m} p(i)$
4	95, 95
5	101, 101, 100, 100, 107, 107
6	114, 110, 109, 107, 105, 105, 115, 120, 120, 108, 108, 106

Table 1 (Cont.)

7	114, 114, 111, 111, 122, 122, 120, 119
	Sum of first five spectral moments[27]
4	408, 408
5	346, 346, 346, 346, 346, 366
6	304, 274, 334, 284, 308, 308, 324, 294, 294, 308, 308, 284
7	296, 296, 296, 296, 222, 222, 256, 272

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