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A Topological Index for the Total π-Electron Energy

Proof of a Generalised Hückel Rule for an Arbitrary Network

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A modified topological index $\hat{Z}_G$ is proposed to be defined as

$$\hat{Z}_G = \sum_{k=0}^{[N/2]} (-1)^k a_{2k}$$

for characterising the π-electronic system of a conjugated hydrocarbon G with N carbon atoms, where a_{2k} is the coefficient of the characteristic polynomial of G defined as

$$P_G(X) = (-1)^N \det |A - XE| = \sum_{k=0}^{N} a_k X^{N-k}$$

with an adjacency matrix A and the unit matrix E.

$\hat{Z}_G$ is identical to Z_G for a tree graph, or a chain hydrocarbon. Z_G increases with a $(4n+2)$-membered ring formation and decreases with a $4n$-membered ring formation. The total π-electron energy E_π of the Hückel molecular orbital is shown to be related with $\hat{Z}_G$ as $E_\pi = C \ln \hat{Z}_G$. With this relation generalised and extended Hückel rules for predicting the stability of an arbitrary network are proved.

Key words: Graph theory – Topological studies in organic chemistry

1. Introduction

In the early stage of the development of chemistry, the structural formula has played a significant role as a chemical language and sometimes as the basis of the chemical logic. This topological aspect of chemistry has gradually been overwhelmed by more quantitative measurements and calculations. However, a number of the graph-theoretical techniques have recently been shown to be applied to chemical problems and have revived the topological studies, especially in organic chemistry [1–5]. In despite of the scepticism of many rigorous theoreticians the topological methods have exhibited a strong descriptive and predictive power in many areas of organic chemistry. Particularly, it is possible to predict a number of physical and chemical properties both of saturated [6–10] and conjugated hydrocarbons [4, 11, 12] through several graph-theoretical countings, e.g., the number of the Kekulé structures [13–17], the values of topological indices [6–8], and the coefficients of the secular polynomial of molecular orbitals [16, 18–22]. Many empirical rules which have never been proved by rigorous treatments were shown to be true by the topological studies with as few assumptions as possible [7].

In this paper a quantity $\tilde{Z}$, modified topological index, is proposed for estimating the total π-electron energy E_π of the Hückel molecular orbitals of unsaturated hydrocarbons and with this index a generalised and extended Hückel rule, or a stability criterion for an arbitrary network, is proved. Before presenting the theory proposed, let us give a short summary of the graph-theoretical definitions and theorems relevant to this problem [23].

2. Premises

Graph $\boldsymbol{G}$, non-adjacent number $p(\boldsymbol{G}, k)$, topological index Z_G, and characteristic polynomial $P_G(X)$ have been defined [6, 18] and tabulated [24, 25] elsewhere. However, $P_G(X)$ is chosen here as

$$P_G(X) = (-1)^N \det|\boldsymbol{A} - X\boldsymbol{E}|, \tag{1}$$

which is related to the Hückel molecular orbitals through the relation

$$X = (\varepsilon - \alpha)/\beta. \tag{2}$$

For the later discussion let us divide $P_G(X)$ into two parts as

$$\begin{aligned} P_G(X) &= S_G(X) + A_G(X) \\ &= \sum_{k=0}^{m} a_{2k} X^{N-2k} + \sum_{k=0}^{m-1} a_{2k+1} X^{N-2k-1}\,{}^{[1]}. \end{aligned} \tag{3}$$

For an alternant hydrocarbon [26, 27] we have

$$a_{2k+1} = 0 \qquad \text{for all } k \tag{4}$$

or

$$A_G(X) \equiv 0, \tag{4'}$$

and the sign of the coefficient a_{2k} alternates

$$(-1)^k a_{2k} \geqq 0. \tag{5}$$

There have been a number of attempts to relate the coefficients of the characteristic polynomial with the topology of a graph [6, 16, 18–22, 28–31]. In this paper the approach by the topological index [6, 18] will be developed. For a tree graph the coefficients of the characteristic polynomial are directly related with the $p(\boldsymbol{G}, k)$ numbers as [6]

$$P_G(X) = \sum_{k=0}^{m} (-1)^k p(\boldsymbol{G}, k) X^{N-2k}. \tag{6}$$

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[1] $m = [N/2]$, the largest integer not exceeding $N/2$.

Correction terms are necessary for a non-tree graph [18][2]

$$\begin{aligned} P_G(X) = & \sum_{k=0}^{m} (-1)^k p(G, k) X^{N-2k} \\ & + \sum_{R_i \in G} \sum_{k=0}^{m_i} (-2)^{r_i} (-1)^k p(G \ominus R_i, k) X^{N-n_i-2k} . \end{aligned} \tag{7}$$

Let R_i be a ring or a set of disjoint rings in G. The numbers of the points and rings in R_i are, respectively, n_i and r_i. Graph $G \ominus R_i$ is the subgraph of G obtained from G by deleting R_i together with all the lines incident to R_i. The first summation in the second terms runs over all the rings and all the possible combinations of disjoint rings in G. Examples for using Eq. (7) will be given later.

Through Eqs. (6) and (7) the topological index is potentially related to the Hückel molecular orbitals. This relationship is more compactly but implicitly stated in the Sachs' theorem [19, 21]. which, however, will not be treated here.

3. Relation between the Total π-Electron Energy and the Characteristic Polynomial

In what follows let us confine ourselves to unsaturated hydrocarbons (alternant and non-alternant) with even number ($N = 2m$) of carbon atoms, and study their π-electronic structures of the ground state by the Hückel molecular orbital method. According to Coulson and Longuet-Higgins [32, 33] the total π-electron energy

$$E_\pi = 2 \sum_{k=1}^{m} X_k \text{[3]} \tag{8}$$

is expressed as an integral form on the complex plane,

$$E_\pi = \frac{1}{\pi i} \oint_\gamma \left\{ \frac{z P'_G(z)}{P_G(z)} - N \right\} dz , \tag{9}$$

where the path of integration γ is the imaginary axis from ∞i to $-\infty i$ and the infinite semi-circle to the right of the y-axis. For a molecule with an NBMO path γ should be changed so as to detour the origin [33].

Since the integration over the infinite semi-circle is vanished we have

$$E_\pi = \frac{1}{\pi} \int_{-\infty}^{\infty} \left\{ N - \frac{iy \cdot P'_G(iy)}{P_G(iy)} \right\} dy . \tag{10}$$

It is clear from the above discussion that we had better exclude the following three classes of molecules:

Class 1) molecule like I whose highest occupied molecular orbital (HOMO) is anti-bonding.

Class 2) molecule like II whose lowest unoccupied molecular orbital (LUMO) is bonding.

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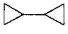
[2] Note the difference in the definition of $P_G(X)$ adopted in the original paper [18].

[3] X_k is to be numbered in the decreasing order.

Class 3) molecule with an NBMO or NBMO's.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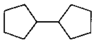

II

If one divides $P_G(X)$ into two parts as in Eq. (3), $S_G(X)$ and $A_G(X)$ will respectively be even and odd functions with respect to X, namely

$$\begin{aligned} S_G(-X) &= S_G(X) \\ A_G(-X) &= -A_G(X). \end{aligned} \tag{11}$$

The opposite is true for their first derivatives,

$$\begin{aligned} S'_G(-X) &= -S'_G(X) \\ A'_G(-X) &= A'_G(X). \end{aligned} \tag{12}$$

With the relations (11) and (12) the integral (10) is expressed as

$$E_\pi = \frac{2}{\pi} \int_0^\infty \left[N - \frac{iy\{S_G(iy)S'_G(iy) - A_G(iy)A'_G(iy)\}}{\{S_G(iy)\}^2 - \{A_G(iy)\}^2} \right] dy. \tag{13}$$

For an alternant hydrocarbon the term $A_G(iy)$ vanishes [see Eq. (4′)] and we get

$$E_\pi = \frac{2}{\pi} \int_0^\infty \left\{ N - \frac{iy \cdot S'_G(iy)}{S_G(iy)} \right\} dy. \tag{14}$$

Let us show that even for a non-alternant hydrocarbon Eq. (14) is approximately true, unless it has an NBMO. For smaller values of y the term a_{2m} predominates in $S_G(iy)$, making the value $|S_G(iy)|$ larger than $|A_G(iy)|$. For larger values of y the highest term, y^N, of $S_G(iy)$ is larger than that, y^{N-1}, of $A_G(iy)$. Then the following inequality,

$$|S_G(iy)| > |A_G(iy)|,$$

is expected for all y to give approximately Eq. (14). This implies that the contribution by the odd function $A_G(X)$ to the integration in Eq. (10) is cancelled between the two regions $(-\infty, 0)$ and $(0, \infty)$. In other words the total π-electron energy of a hydrocarbon seems to be mostly determined by the coefficients of the symmetric part of the characteristic polynomial. This conjecture will be verified by the discussion below.

4. Definition of the Modified Topological Index

Define the function $q_G(X)$ for graph $\boldsymbol{G}$ as

$$q_G(X) = \sum_{k=0}^{m} (-1)^k a_{2k} X^k, \tag{15}$$

where a_{2k} is related with the characteristic polynomial through Eq. (3). For a tree graph $q_G(X)$ is identical to $Q_G(X)$ defined in Ref. [6]. The function $S_G(X)$ is related with $q_G(X)$ as

$$S_G(X) = X^N q_G(-X^{-2}). \tag{16}$$

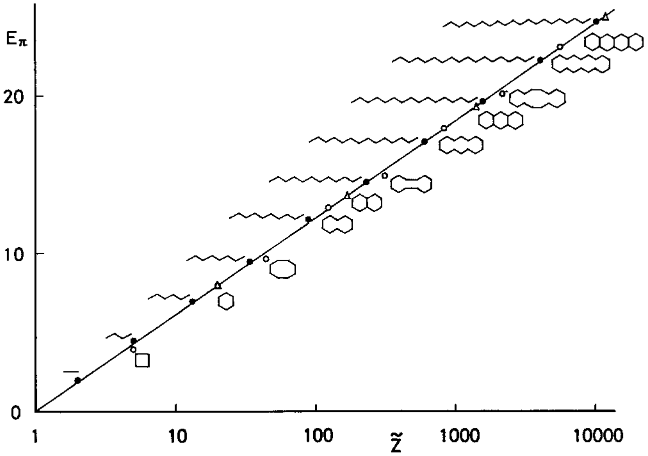


Fig. 1. Plots of E_π vs. $\log \tilde{Z}$ for linear polyenes (●), annulenes (○), and polyacenes (△)

The modified topological index $\tilde{Z}_G$ is defined as

$$\tilde{Z}_G = q_G(1) \tag{17}$$

$$= (-i)^N S_G(i) \tag{18}$$

$$= \sum_{k=0}^{m} (-1)^k a_{2k}\,. \tag{19}$$

These definitions and relations (15)–(19) can also be applied to molecules with odd number of carbon atoms.

For an alternant hydrocarbon one gets a simpler expression as

$$\tilde{Z}_G = \sum_{k=0}^{m} |a_{2k}|\,, \quad \text{(alternant hydrocarbon)} \tag{20}$$

by noticing the relation (5).

For a tree, or a chain hydrocarbon, $\tilde{Z}_G$ is identical to Z_G, i.e.,

$$\tilde{Z}_G = Z_G \quad \text{(tree)}\,. \tag{21}$$

General expressions of the $\tilde{Z}_G$ values for certain series of compounds can be obtained by using recursion formulae [6, 8, 18, 34]. Analysis shows that asymptotically the E_π values are linearly related to the logarithms of $\tilde{Z}_G$ for the following series [17]:

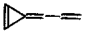
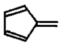
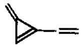
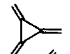
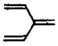
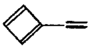
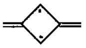
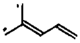
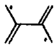
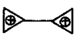
$$E_\pi \quad 6.092 \log \tilde{Z}_{\bar{N}} + 0.129 \quad \text{(linear polyene)}, \tag{22}$$

$$E_\pi = 6.092 \log \tilde{Z}_{N^0} \quad \text{(annulene)}, \tag{23}$$

$$E_\pi = 6.041 \log \tilde{Z}_{n^*} + 0.242 \quad \text{(polyacene)}, \tag{24}$$

where $\bar{N}$, N^0, and n^*, respectively, denote a linear polyene with N carbon atoms, an annulene with N carbon atoms, and a polyacene with n hexagons linearly connected. Actually in Fig. 1 all the plots of E_π against $\log \tilde{Z}_G$ for these conjugated

Table 1. Characteristic quantities of conjugated hydrocarbons with six carbon atoms

No.[a]	*G*	a_k[b]					$\tilde{Z}$[c]	Z[d]	E_π[e]	ΔZ[f]
		$k=2$	3	4	5	6				
1		-7	-2	11	2	-4	23	21	8.390	2
2		-6		9		-4	20	18	8.000	2
3		-7		7		-1	16	22	7.657	-6
4		-6	-2	8	4	-1	16	16	7.550	0
5		-6		8	-2	-1	16	16	7.466	0
6		-6	-2	7	2	-1	15	15	7.418	0
7		-6	-2	6		-1	14	14	7.301	0
8		-6		5		-1	13	15	7.208	-2
9		-5		6		-1	13	13	6.988	0
10		-5		5		-1	12	12	6.899	0
11		-8	-4	12	8		21	25	8.293	-4
12		-7		9	-4		17	21	7.565	-4
13		-6		6			13	17	6.603	-4
14		-6		5			12	14	6.472	-2
15		-5		5			11	11	6.155	0
16		-5		4			10	10	6.000	0
17		-7	-4	11	12	3	16	20	8.293	-4

[a] Compounds 11–16 belong to Class 3 and 17 (= I) belongs to Class 1.

[b] $P_G(X) = \sum_{k=0}^{6} a_k X^{6-k}$, with $a_0 = 1$ and $a_1 = 0$ for all the graphs.

[c] $\tilde{Z} = \sum_{k=0}^{3} (-1)^k a_{2k}$, Eq. (19).

[d] Tabulated in Refs. [24] and [25], where the signs of the odd terms, a_{2k+1}, are inverted, due to the different definition of $P_G(X) = \det|A + XE|$.

[e] $E_\pi = \sum_{k=1}^{6} g_k X_k$. For 1–12, $g_1 = g_2 = g_3 = 2$ and $g_4 = g_5 = g_6 = 0$. For 16 $g_1 = g_2 = 2$, $g_3 = g_4 = 1$, and $g_5 = g_6 = 0$. For 17 $g_1 = g_2 = 2$ and $g_3 = g_4 = g_5 = g_6 = 0$.

[f] Aromaticity index, Eq. (27).

hydrocarbons lie on a single straight line. It was observed that for almost typical alternant hydrocarbons with moderate number of carbon atoms the relation

$$E_\pi = C \ln \tilde{Z}_G \tag{25}$$

holds fairly well.

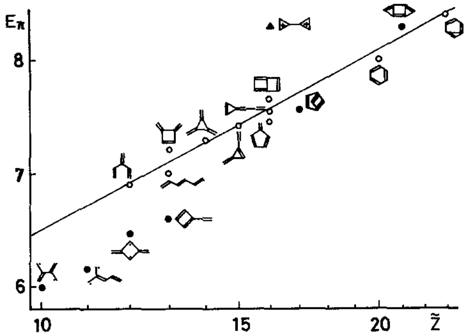


Fig. 2. Plots of E_π vs. log $\tilde{Z}$ for the conjugated hydrocarbons with six π-electrons. ▲ : Class 1, ● : Class 3

Figure 2 gives the plots of E_π vs. log $\tilde{Z}_G$ for almost all the isomers of alternant and non-alternant hydrocarbons with six carbon atoms including molecules of Classes 1–3[4], without which all the points lie on a straight line. In order to keep consistency between the energy expression (8) and the contour γ in the integral (9) E_π value of the dication instead of the neutral molecule was plotted for molecule 17 (in Table 1), or I, which has two bonding and four anti-bonding orbitals (Class 1). A number of molecules have been tested similarly and we can safely conclude that the π-electron stability of a conjugated hydrocarbon is determined by the topology of the molecule through Eq. (25); the constant being different for molecules with and without NBMO's. This good correlation proves that the coefficients of $S_G(X)$ determines the total energy E_π through Eq. (14) or its transformed form,

$$E_\pi = \frac{4}{\pi}\int_0^\infty \frac{q'_G(X^2)}{q_G(X^2)}\,dX\,. \tag{26}$$

Numerical analysis shows that Eq. (25) is a good approximation to Eq. (26) [17].

5. Aromaticity Index

In order to analyse the relation (25) the coefficients of the characteristic polynomial and the values of E_π, $\tilde{Z}_G$, and Z_G for the molecules in Fig. 2 are compared in Table 1. Correlation between E_π and Z_G is not so good but improved by modifying Z_G into $\tilde{Z}_G$. Note that molecules without a ring and those with only one odd-membered ring have common values for $\tilde{Z}_G$ and Z_G, or in other words, the increment

$$\Delta Z_G = \tilde{Z}_G - Z_G \tag{27}$$

is zero[5]. On the other hand, a $(4n+2)$-membered ring gives a positive increment and a $4n$-membered ring a negative one.

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[4] In this group there is no entry in Class 2.

[5] Molecule 1 has one triangle and one pentagon. Molecule 11 has two triangles and two pentagons. Molecule 12 has two pentagons. They all have non-zero ΔZ_G. On the other hand, molecule 4–7 with one odd-membered ring have zero ΔZ_G.

Table 2. Enumeration of $\tilde{Z}_G$ by using the composition principle

i	R_i	$G \ominus R_i$	$Z_{G \ominus R_i}$ (A)	$(-2)^{r_i}(-1)^{n_i/2}$ (B)	$A \times B$
0	0[a]	 [b]	25	1	25
1			1	-2	-2
2		0	1	2	2
3		0	1	-4	-4

[a] Vacant graph.
[b] The original graph G.

Let us call ΔZ_G as an aromaticity index, as it accounts for the stability of aromatic rings. Since the topological index $\tilde{Z}_G$ can be enumerated quite easily by the use of the composition principles (recursion formulae), the value of $\tilde{Z}_G$ and the prediction of aromatic stability of a condensed ring system can also be obtained by hand calculation if one knows a simple algorithm for getting ΔZ_G.

6. Generalised and Extended Hückel Rules

By the use of Eq. (7) the even function $S_G(X)$ can be expressed in terms of non-adjacent numbers as

$$\begin{aligned} S_G(X) = & \sum_{k=0}^{m} (-1)^k p(G,k) X^{N-2k} \\ & + \sum_{R_i \in G}^{\text{even } n_i} \left\{ (-2)^{r_i} \sum_{k=0}^{m_i} (-1)^k p(G \ominus R_i, k) X^{N-n_i-2k} \right\}, \end{aligned} \tag{28}$$

where the first summation in the second term runs over such R_i that the total number, n_i, of carbon atoms of an independent ring or a set of independent rings is even. By combining Eqs. (18) and (28) we get

$$\begin{aligned} \tilde{Z}_G = & \sum_{k=0}^{m} p(G,k) \\ & + \sum_{R_i \in G}^{\text{even } n_i} (-1)^{n_i/2} (-2)^{r_i} \sum_{k=0}^{m_i} p(G \ominus R_i, k) \\ = & Z_G + \sum_{R_i \in G}^{\text{even } n_i} (-1)^{n_i/2} (-2)^{r_i} Z_{G \ominus R_i}. \end{aligned} \tag{29}$$

Let us enumerate $\tilde{Z}_G$ for molecule 11 by using Eq. (29). Call graph 11 as G and write down all the possible independent even-membered rings (R_1 and R_2) and the possible combination (R_3) of disjoint rings with even number of carbon atoms in total (see Table 2). For convenience's sake let us call G as $G \ominus R_0$, where R_0 is

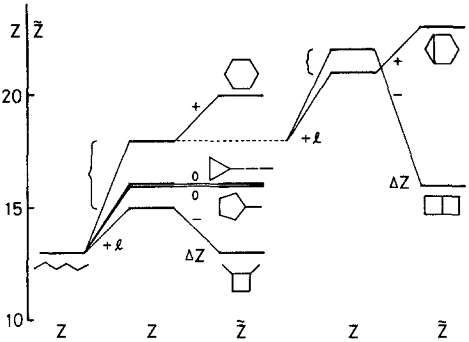
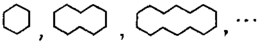


Fig. 3. Relations between the stability of molecules and topological quantities, Z, $\tilde{Z}$, and $\Delta\tilde{Z}$, for the systems with six π-electrons. Z creases with a ring closure due to a formation of a new bond $(+l)$. Difference in Z ({) among the isomers is due to the difference in the combinatorial factors. A $(4n+2)$-membered ring gives positive ΔZ, a $4n$-membered ring negative ΔZ, and odd-membered ring zero ΔZ

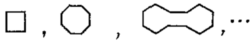
a vacant graph ($r_0 = n_0 = 0$). Calculate the Z values for all the $\boldsymbol{G} \ominus \boldsymbol{R}_i$ and add them up with the weight $(-2)^{r_i}(-1)^{n_i/2}$.

The summation of the second term to get $\tilde{Z}_G$ of Eq. (29) can be rewritten symbolically as

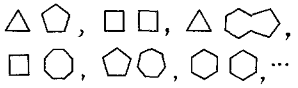
$$\Delta Z_G = 2 \sum_{\substack{r=1 \\ n=4k+2}}^{\boldsymbol{R}} Z_{\boldsymbol{G}\ominus\boldsymbol{R}}$$



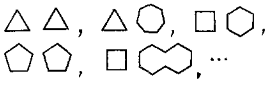
$$-2 \sum_{\substack{r=1 \\ n=4k}}^{\boldsymbol{R}} Z_{\boldsymbol{G}\ominus\boldsymbol{R}}$$



$$+4 \sum_{\substack{r=2 \\ n=4k}}^{\boldsymbol{R}} Z_{\boldsymbol{G}\ominus\boldsymbol{R}}$$



$$-4 \sum_{\substack{r=2 \\ n=4k+2}}^{\boldsymbol{R}} Z_{\boldsymbol{G}\ominus\boldsymbol{R}}$$



$$+8 \sum_{\substack{r=3 \\ n=4k+2}}^{\boldsymbol{R}} Z_{\boldsymbol{G}\ominus\boldsymbol{R}}$$

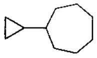
$$- \cdots \tag{30}$$

This equation shows two important factors governing the π-electron stability of a ring network, namely, the value of the topological index of subgraph $\boldsymbol{G} \ominus \boldsymbol{R}$ and the sign given to each term.

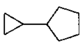
Figure 3 illustrates how the difference in the phase of ring closure, for example, of hexatriene, affects the stability of a π-electron network. Since the number of bonds is increased by the ring closure, all the topological indices of molecules 2, 4, 5, and 8 are larger than that of hexatriene (molecule 9). The differences among them are caused by the partitioning of double bonds in the molecular network. The aromaticity index of hexagon (molecule 2) is positive, while that of tetragon (molecule 8) negative, causing large difference in the π-electron stability of these two molecules. Ring closure of odd-membered ring (see molecules 4 and 5) does not make Z_G and $\tilde{Z}_G$ differ. The difference in the stability of molecules 1 and 3 is largely due to the destabilisation by the two tetragon formation in 3.

Thus the first two terms of Eq. (30) give the proof for the generalised Hückel rule, namely, a π-electron network is stabilised and destabilised additively by a closure of a $(4n+2)$- and $4n$-membered ring in an arbitrary network [36, 37].

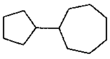
The third and fourth terms present an interesting feature of the Hückel rule extended to the system with two disjoint rings. As evident from Eq. (30) one can predict that molecules with ring skeleton I (= 17), II, or III are unstable but those with IV or V are stable.



III



IV



V

The same conclusion on the generalised and extended Hückel rules can be drawn by using the Sachs' theorem [19].

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