Circuit Resonance Energy. On the Roots of Circuit Characteristic Polynomial

Noriyuki MIZOGUCHI
Department of Physics, Meiji College of Pharmacy, Nozawa, Setagaya-ku, Tokyo 154
(Received June 5, 1989)

The circuit characteristic polynomial plays an important role in the theory of circuit resonance energy of polycyclic conjugated molecules. Although in the use of circuit resonance energy it is essential that the circuit characteristic polynomial has real roots, the definition of circuit characteristic polynomial does not ensure that the roots of this polynomial are real numbers. It is proved that the circuit characteristic polynomial for any circuit in a polycyclic conjugated molecule has no imaginary roots if this molecule is a nonfused polycyclic conjugated molecule or a fused bicyclic system.

Re-examination of Hückel molecular orbital theory from a graph-theoretical point of view clarified that the coefficients of the characteristic polynomial of a graph G can be calculated from a set of certain subgraphs (called Sachs graph) of G.[1-3] The topological resonance energy TRE defined on the basis of this finding was found to be an excellent index for aromaticity. The coefficients of the reference polynomial in the TRE theory are constructed from acyclic Sachs graphs for G.[2] Thus the reference polynomial $R(G; X)$ does not contain the contribution of any circuit found in G while the characteristic polynomial $P(G;X)$ contains the contributions of all the circuits. The TRE of a cyclic conjugated molecule, which is given by the difference between the π-electron energy of the system and that of the reference system, represents the degree of cyclic conjugation of π-electrons due to the presence of the circuits found in the system.

Ring aromaticity or local aromaticity is a useful concept for prediction of the chemical behavior of a given ring in a polycyclic hydrocarbon.[4] The circuit resonance energy plays a significant role in the theory of local aromaticity. Although the TRE theory is very useful in predicting the whole aromaticity of a conjugated molecule, this theory does not allow one to estimate quantitatively the contribution of each circuit in a polycyclic conjugated molecule to the TRE (circuit resonance energy, CE). This difficulty was overcome by introducing two new polynomials, the circuit characteristic polynomial, $P(G/C_n; X)$,[5] and the circuit reference polynomial, $R(G/C_n; X)$.[6] The former polynomial contains the contribution of the circuit C_n only while the later polynomial contains the contributions of the other circuits except C_n. By using the two polynomials circuit resonance energy was defined in two different manners by Aihara[5] and by Gutman and Bosanac.[6]

Two problems of circuit resonance energy CE have been pointed out. One is a problem associated with the Hückel rule.[6] Since the stabilities of cyclic conjugated molecules obey the Hückel rule,[7] it is desirable that the circuit resonance energies obey rules similar to the Hückel rule. However circuit resonance energies defined by Gutman violate the Hückel rule.[6] Recently, by using a Coulson-type integral formula[8] we proved mathematically that the circuit resonance energies defined by Aihara obey the $4n+2$ rules.[9] The stabilities of Möbius polycyclic conjugated molecules obey the anti-Hückel rule.[10-12] We applied the theory of circuit resonance energy also to Möbius polycyclic conjugated molecules and proved the rules for circuit resonance energies of Möbuis-type circuits.[9] These rules are, of course, in accord with the anti-Hückel rule for the stabilities of Möbius systems.[10,11] Another problem is that the circuit resonance energies should be real values. The existence of imaginary roots of circuit characteristic polynomial or circuit reference polynomial obscures the meaning of the estimated values of ring aromaticities. The definitions of the two polynomials do not ensure that these polynomials have no imaginary roots. It has been reported that the circuit reference polynomial has imaginary roots in some cases.[13] We have calculated the roots of the circuit characteristic polynomials for a large number of polycyclic conjugated molecules and found that the roots are real numbers in all cases. Accordingly it is conjectured that the circuit characteristic polynomial for any circuit of any system has real roots. The Coulson integral expression for the circuit resonance energy enables us to show that circuit resonance energies are real values for any Hückel and Möbius polycyclic conjugated molecules with an even number of carbon atoms. However this does not mean that the circuit characteristic polynomial has no imaginary roots.[14]

The aim of this paper is to discuss the problem of whether the roots of the circuit characteristic polynomials are real or not.

Circuit Characteristic Polynomial

Let G be a graph representing the π-electron network of a polycyclic conjugated molecule without bond alternation and C_n be a circuit in G. The circuit resonance energy of the circuit C_n is given by[5]

$$\mathrm{CE}(\mathrm{C}_n) = \sum_j g_j\{X_j(P(\mathrm{G}/\mathrm{C}_n;\ X)) - X_j(R(\mathrm{G};X))\}. \tag{1}$$

In Eq. 1 $X_J(P(\mathrm{G}/\mathrm{C}_n;\ X))$ and $X_J(R(\mathrm{G};X))$ are the J-th roots of the circuit characteristic polynomial $P(\mathrm{G}/\mathrm{C}_n;X)$ and the reference polynomial $R(\mathrm{G};X)$, respectively, and g_J is the occupation number of the J-th MO.

Hückel and Möbius polycyclic conjugated molecules are represented by generalized graphs in which each edge has a weight of 1 or −1.[15] Circuits in generalized graphs are classified into Hückel-type circuits or Möbius-type circuits.[11] The former is a circuit in which the number of edges with the weight −1 is even and the latter is a circuit in which the number of edges with the weight −1 is odd. A generalized graph is said to be Hückel-type if it does not have any Möbius-type circuit and Möbius-type if it has at least one Möbius-type circuit.[11] The thermodynamic stabilities of generalized graphs obey module 4 rules, which are the unified rules for Hückel and Möbius polycyclic conjugated molecules.[11]

Let G^{G} be one of the generalized graphs for G obtained by giving each edge in G the weight 1 or −1. The explicit equations for the circuit characteristic polynomials for Hückel-type and Möbius-type circuits in the generalized graph G^{G} were obtained as follows:[9] for Hückel-type circuit C_n

$$P(\mathrm{G}^{\mathrm{G}}/\mathrm{C}_n;\ X) = R(\mathrm{G};X) - 2\,R(\mathrm{G}\ominus\mathrm{C}_n;X), \tag{2}$$

for Möbius-type circuit C_n

$$P(\mathrm{G}^{\mathrm{G}}/\mathrm{C}_n;\ X) = R(\mathrm{G};\ X) + 2\,R(\mathrm{G}\ominus\mathrm{C}_n;\ X), \tag{3}$$

where the graph $\mathrm{G}\ominus\mathrm{C}_n$ is a subgraph of G obtained by deleting the circuit C_n and all the edges incident to C_n from G.[1] In the derivation of Eqs. 2 and 3 it was used that the reference polynomial of the generalized graph G^{G} is identical with that of the original graph G.[11] Since the second term of the right-hand side of Eq. 2 (or Eq. 3) represents the contribution of the Hückel-type (or Möbius-type) circuit C_n, the circuit resonance energy of the circuit given by Eq. 1 is considered to represent the contribution of the circuit C_n to the TRE.

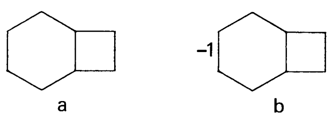

Fig. 1. Two generalized graphs for benzocyclobutene. Graph **a** is Hückel-type and graph **b** is Möbius-type.

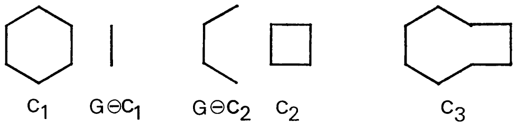

Fig. 2. Three circuits in benzocyclobutene and the subgraphs associated with them.

Let us take one example to illustrate the above terminologies. Two of the generalized graphs **1a** and **1b** for the benzocyclobutene graph are shown in Fig. 1. (In these graphs we have not shown the weight 1. This omission is used throughout this paper). Figure 2 shows three circuits contained in the benzocyclobutene graph and three subgraphs associated with these circuits. Graph **1a**, which is the benzocyclobutene graph itself, contains the Hückel-type circuits only, while **1b** is a Möbuis-type graph because this graph contains two Möbius-type circuits C_1 and C_3. So we have for the Hückel-type circuit C_1 of **1a**

$$P(\mathbf{1a}/\mathrm{C}_1;X)=R(\mathrm{G};\ X)-2R(\mathrm{G}\ominus\mathrm{C}_1;\ X),$$

and for the Möbius-type circuit C_1 of **1b**

$$P(\mathbf{1b}/\mathrm{C}_1;X)=R(\mathrm{G};X)+2R(\mathrm{G}\ominus\mathrm{C}_1;X).$$

Graph Corresponding to Circuit Characteristic Polynomial. The definition of the circuit characteristic polynomial does not give any information about structure corresponding to the polynomial. The structure is represented by a graph whose characteristic polynomial is equal to the circuit characteristic polynomial. Let such graph be denoted by C^{CP}. The characteristic polynomial for G^{CP}, $P(\mathrm{G}^{\mathrm{CP}};\ X)$, satisfies

$$P(\mathrm{G}^{\mathrm{CP}};\ X) = \det|X\boldsymbol{I}-\boldsymbol{A}(\mathrm{G}^{\mathrm{CP}})| = P(\mathrm{G}^{\mathrm{G}}/\mathrm{C}_n;\ X). \tag{4}$$

In Eq. 4 $\boldsymbol{A}$ $(\mathrm{G}^{\mathrm{CP}})$ is the adjacency matrix of G^{CP}; $\boldsymbol{I}$ is the unit matrix. The objective of this paper is to obtain G^{CP} under the condition that the adjacency matrix is Hermitian.

In order to get G^{CP} we consider a directed and edge-weighted graph G* which is obtained by replacing each (undirected) edge r—s of the original graph G with the two directed edges $r\rightarrow s$ and $s\rightarrow r$ with the weights given by

$$w_{rs} = \exp(\mathrm{i}v_{rs}) \quad \text{for } r \rightarrow s \text{ edge},$$
$$w_{sr} = \exp(\mathrm{i}v_{sr}) \quad \text{for } s \rightarrow r \text{ edge}, \tag{5}$$

where $\mathrm{i}=\sqrt{-1}$. We suppose that v_{rs} is a real number but satisfies the condition:

$$v_{sr} = -v_{rs}. \tag{6}$$

This condition ensures that the characteristic polynomial for G* has real roots. The Sachs formula[3] allows us to express the characteristic polynomial of G* as follows:[16]

$$P(\mathrm{G}^*;\ X) = R(\mathrm{G};\ X) - 2\sum_j R(\mathrm{G}\ominus\mathrm{C}_j;X)\cos(V(\mathrm{C}_j))$$
$$+ 4\sum_j\sum_k R(\mathrm{G}\ominus\mathrm{C}_j\ominus\mathrm{C}_k;X)\cos(V(\mathrm{C}_j))\cos(V(\mathrm{C}_k)) - \cdots\cdots \tag{7}$$

In Eq. 7 the graph $G\ominus C_j\ominus C_k$ is a subgraph of G obtained by deleting two disjoint circuits C_j and C_k and all the edges incident to the two circuits; the first sum runs over all the circuits found in G, and the second one over all the possible pairs of disjoint circuits; $V(C_j)$ is the sum of v_{rs} over all the edges in the circuit C_j along one direction. Equation 7 shows that $P(G^*; X)$ is dependent on the $\cos(V(C))$ values (not on the v_{rs} values for individual edges).

It is seen from Eq. 7 that if the parameters v_{rs} satisfy

$$\cos(V(C_j)) = 0 \quad \text{for any circuits except } C_n, \tag{8}$$

then we have

$$P(G^*; X) = R(G; X) - 2R(G\ominus C_n; X)\cos(V(C_n)),$$

which contains the contribution of C_n only. Further, if the parameters v_{rs} for the edges in C_n satisfy

$$\cos(V(C_n)) = \begin{cases} 1 & \text{when } C_n \text{ in } G^G \text{ is Hückel-type} \\ -1 & \text{when } C_n \text{ in } G^G \text{ is Möbius-type,} \end{cases} \tag{9}$$

then $P(G^*;X)$ is equal to $P(G/C_n;X)$ for C_n in the generalized graph G^G (see Eqs. 2 and 3). Thus it is seen that if the parameters v_{rs} in the graph G^* satisfy Eqs. 8 and 9, then the graph G^* is a desired graph G^{CP} for the circuit C_n in G^G.

One cannot always assign arbitrary values to $V(C_j)$'s for all the circuits in a polycyclic graph because the $V(C_j)$ terms for all the circuits are not independent as shown below. Let C_i and C_j be two circuits with some common edges and circuit C_k be C_i+C_j (see Fig. 3). For these circuits it follows from Eq. 6 that

$$\begin{aligned} & V(C_i) + V(C_j) \\ &= (v_{12} + v_{23} + \cdots\cdots \; v_{M-1M} + v_{Mr} + v_{rs} + \cdots\cdots + v_{z1}) \\ &+ (v_{MM+1} + \cdots\cdots + v_{N-1N} + v_{N1} + v_{1z} + \cdots\cdots + v_{sr} \\ &+ v_{rM}) \\ &= v_{12} + v_{23} + \cdots\cdot + v_{M-1M} + v_{MM+1} + \cdots\cdot \\ &+ v_{N-1N} + v_{N1} \\ &= V(C_k). \end{aligned} \tag{10}$$

In the above the two circuits C_i and C_j are not necessarily fundamental circuits. Fundamental circuits in a graph are called ring.[17] Thus Eq. 10 means that the number of the independent $V(C_j)$ quantities is equal not to the number of the circuits but to that of the rings. For example, in the case of benzocyclobutene, we have

$$V(C_1) + V(C_2) = V(C_3). \tag{11}$$

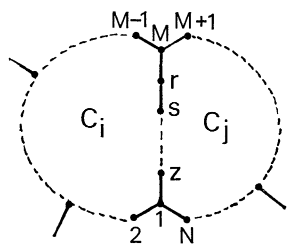

Fig. 3. Two circuits C_i and C_j.

The difference between the number of rings and the number of circuits in graph arises from condensation of rings. Accordingly we divide polycyclic graphs into three categories as shown below.

a) Polycyclic Generalized Graphs without Fused Rings. The number of the circuits in any graph of this type is equal to the number of rings. Therefore, for any circuit in the graph it is possible to choose the parameters v_{rs} so as to satisfy Eqs. 8 and 9. Thus we can obtain a graph G^{CP} for any circuit in any generalized graph of this type.

Two graphs **4a** and **4b** in Fig. 4 are two examples for the *p*-terphenyl graph G1 which contains three fundamemtal circuits (rings), C_1, C_2, and C_3. The characteristic polynomial of the directed and edge-weighted graph G1* for G1 (see Eq. 7) is

$$\begin{aligned} P(G1^*; X) &= R(G1; X) - 2\{R(G1\ominus C_1; X)\cos(V(C_1)) \\ &+ R(G1\ominus C_2; X)\cos(V(C_2)) \\ &+ R(G1\ominus C_3; X)\cos(V(C_3))\} \\ &+ 4\{R(G1\ominus C_1\ominus C_2; X)\cos(V(C_1))\cos(V(C_2)) \\ &+ R(G1\ominus C_1\ominus C_3; X)\cos(V(C_1))\cos(V(C_3)) \\ &+ R(G1\ominus C_2\ominus C_3;X)\cos(V(C_2))\cos(V(C_3))\}. \end{aligned} \tag{12}$$

In graph **4a** the circuit C_1 contains the edges with the weight 1 only while two circuits C_2 and C_3 contain a pair of directed edges with the weights i and −i besides the edges with the weight 1. Accordingly we have $V(C_1)=0$ and $V(C_2)=V(C_3)=\pm\pi/2$, which satisfy Eqs. 8 and 9. Thus Eq. 12 for **4a** reduces to

$$P(\mathbf{4a}; X) = R(G1; X) - 2R(G1\ominus C_1; X). \tag{13}$$

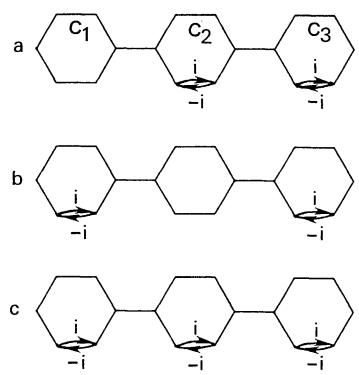

Fig. 4. Graphs (**a** and **b**) corresponding to the circuit characteristic polynomials for C_1 and for C_2 in *p*-terphenyl. Graph **c** is a graph corresponding to the reference polynomial of *p*-terphenyl.

Therefore it follows that **4a** is a graph G^{CP} for the circuit C_1 of the *p*-terphenyl graph. As seen from this example, a circuit with a pair of directed edges with the weights i and −i makes no contribution to the characteristic polynomial.[18] This effect is not dependent on the type nor the size of the circuit. Similarly it is easily understood that **4b** contains the contribution of C_2 only and thus this graph is a graph G^{CP} for C_2 in the *p*-terphenyl graph.

It should be noted that the results for **4a** and **4b** are not dependent on the sizes of the circuits in these graphs. So it is seen that if one edge in each circuit except a circuit C_n in a nonfused polycyclic generalized graph is replaced by a pair of directed edges with the weights ±i, then such a graph is a graph G^{CP} for the circuit C_n of the generalized graph.

b) Fused Bicyclic Generalized Graphs. Although for graphs of this type it is impossible to assign arbitrary values to $V(C_j)$'s for all the circuits, we can obtain a graph G^{CP} for any circuit in any graph of this type because any graph of this type contain only three circuits.

To illustrate this result let us take benzocyclobutene as an example. Since no disjoint circuit is found in the benzocyclobutene graph G2 (see Figs. 1 and 2), the characteristic polynomial of the directed and edge-weighted graph G2* for G2 (see Eq. 7) is

$$P(\text{G2*}; X) = R(\text{G2}; X) - 2\{R(\text{G2}\ominus C_1; X)\cos(V(C_1)) + R(\text{G2}\ominus C_2; X)\cos(V(C_2)) + R(\text{G2}\ominus C_3; X)\cos(V(C_3))\}. \quad (14)$$

Although the $V(C)$ values for the three circuits C_1, C_2, and C_3 satisfy Eq. 11, we can obtain graphs G^{CP} for the Hückel-type graph **1a** and for the Möbius-type graph **1b**, which are shown in Fig. 5. For **5b** we have $V(C_1)=V(C_3)=\pm\pi/2$ and $V(C_2)=0$. So, from Eq. 14 we have

$$P(\mathbf{5b}; X) = R(\text{G2}; X) - 2R(\text{G2}\ominus C_2; X),$$

which is the circuit characteristic polynomial for the Hückel-type circuit C_2 in the Möbius-type graph **1b.** Therefore it was shown that graph **5b** is a graph G^{CP} for the circuit C_2 in the Möbius-type benzocyclobutene graph **1b**. Since for graph **5c** $V(C_1)=V(C_2)=\pm\pi/2$ and $V(C_3)=\pi$, from Eq. 14 we have

$$P(\mathbf{5c}; X) = R(\text{G2}; X) + 2R(\text{G2}\ominus C_3; X).$$

Therefore it follows that graph **5c** is a graph G^{CP} for the Möbius-type circuit C_3 in the Möbius-type graph **1b**. Now it will be easily understood that graph **5a** is a graph G^{CP} for the Hückel-type circuit C_1 in the Hückel-type graph **1a**.

As seen from the above examples, it is possible to obtain a graph G^{CP} for any circuit in any fused bicyclic graph because any circuit that contains a pair of directed edges with the weights ±i makes no contribution to the characteristic polynomial and the number of the circuits in the graph is larger than the number of the rings by only one. It should be noted that the results for **5a, 5b,** and **5c** are not dependent on the sizes of the circuits in these graphs. Accordingly it follows that one can obtain a graph G^{CP} for any circuit of any fused bicyclic generalized graph.

c) Polycyclic Generalized Graphs with More than Two Fused Rings. In the case of a graph of this type the difference between the number of the circuits and that of the rings is at least larger than three. This means that it is impossible to obtain graphs corresponding to the circuit characteristic polynomials for graphs of this type.

This result will be illustrated by considering anthracene as an example. The anthracene graph contains six circuits. The three fundamental circuits are shown in Fig. 6 and the other three circuits are $C_4(=C_1+C_2)$, $C_5(=C_2+C_3)$, and $C_6(=C_1+C_2+C_3)$. Now we try to obtain a graph G^{CP} fot the circuit C_1. Graph **6b** in Fig. 6 is obtained by giving the weights i and −i to each edge in C_2 and C_3. For this graph we have $V(C_1)=0$, $V(C_2)=V(C_3)=V(C_4)=\pm\pi/2$, and $V(C_5)=V(C_6)=\pm\pi$ (see Eq. 10). So the contributions of three circuits C_2, C_3, and C_4 are eliminated but those for two circuits C_5 and C_6 are not eliminated (the $\cos(V(C))$ values for the two circuits are −1 and so the two circuits are Möbius-type). Therefore graph **6b** in Fig. 6 is not a graph G^{CP} for the circuit C_1 in the anthracene graph.

It is seen from this example that it is impossible to obtain a graph G^{CP} for any polycyclic generalized graph with more than two fused rings because of Eq. 10.

The method of giving the weights i and −i to some edges in generalized graphs is useful also in the study of graphs corresponding to the reference polynomials.

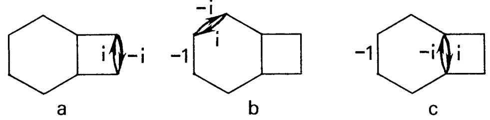

Fig. 5. Graphs corresponding to the circuit characteristic polynomials for C_1 in **1a** and for C_2 and C_3 in **1b**. Graphs **1a** and **1b** are the generalized graphs for benzocyclobutene (see Fig. 1).

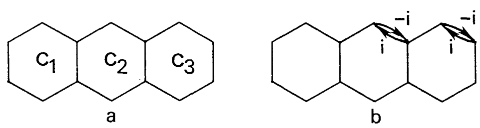

Fig. 6. Three fundamental circuits in anthracene and graph obtained by giving the weights i and −i to two edges in the two circuits C_2 and C_3.

From the above discussion, it will be evident that for a nonfused polycyclic system an appropriate choice of the v_{rs} values allows us to obtain a graph corresponding to the reference polynomial. Graph **4c**, shown in Fig. 4, is an exmple for *p*-terphenyl. Since any circuit in this graph does not contribute to the characteristic polynomial, the characteristic polynomial of this graph is equal to the reference polynomial of the *p*-terphenyl graph. Although it was proved that the roots of the reference polynomial for any graph are real,[19] Eq. 10 does not allow us to obtain graphs corresponding to the reference polynomials for fused polycyclic systems without certain symmetries.[18,20]

Concluding Remarks

It has been shown that under the condition that the adjacency matrix is Hermitian, one can obtain a graph whose characteristic polynomial is equal to the circuit characteristic polynomial for any circuit in a polycyclic conjugated molecule provided that this system is a nonfused polycyclic conjugated molecule or a fused bicylic system. Thus it has been proved that the roots of the circuit characteristic polynomial for any circuit in such a molecule are real numbers. This result holds irrespective of the type of system, namely Hückel-type or Möbius-type.

In the use of Aihara's circuit resonance energy it is essential that the circuit characteristic polynomial has real roots. Although the method used in this paper can be applied to only certain types of molecules, this does not mean that Aihara's circuit resonane energy cannot be used for other types of molecules because extensive numerical calculations show that all the circuit characteristic polynomials which have been calculated have real roots.

References

1) A. Graovac, I. Gutman, and N. Trinajstić, "Topological Approach to the Chemistry of Conjugated Molecules," Springer-Verlag, Berlin (1977); N. Trinajstić, "Chemical Graph Theory," CRC Press, Inc., Boca Raton, Florida (1983); I. Gutman and O. E. Polansky, "Mathematical Concept in Organic Chemistry," Springer-Verlag, Berlin (1986).

2) J. Aihara, *J. Am. Chem. Soc.*, **98**, 2750 (1976); I. Gutman, M. Milun and N. Trinajstić, *ibid.*, **99**, 1692 (1977).

3) H. Sachs, *Publ. Math. (Debrecen)*, **11**, 119 (1963).

4) O. E. Polansky and G. Derflinger, *Int. J. Quantum Chem.*, **1**, 379 (1976); W. C. Herndon and M. L. Ellzey, Jr., *J. Am. Chem. Soc.*, **96**, 6631 (1974); M. Randic, *Tetrahedron*, **31**, 1477 (1975); M. Aida and H. Hosoya, *ibid.*, **36**, 1317 (1980); K. Sakurai, K. Kitaura, and K. Nishimoto, *Theor. Chim. Acta*, **69**, 23 (1986); Y. Jiang and H. Zhang, *ibid.*, **75**, 279 (1989).

5) J. Aihara, *J. Am. Chem. Soc.*, **99**, 2048 (1977).

6) S. Bosanac and I. Gutman, *Z. Naturforsch.*, **32a**, 10 (1977); I. Gutman and S. Bosanac, *Tetrahedron*, **33**, 1809 (1977).

7) E. Hückel, *Z. Phys.*, **76**, 628 (1932); H. Hosoya, K. Hosoi and I. Gutman, *Theoret. Chim. Acta*, **38**, 37 (1975); I. Gutman, *Z. Naturforsh*, **33a**, 214 (1978); I. Gutman, *J. Chem. Soc., Faraday Trans. 2*, **75**, 799 (1979).

8) C. A. Coulson and H. C. Longuet-Higgins, *Proc. R. Soc. London, Ser. A*, **191**, 39 (1947); **192**, 16 (1947).

9) N. Mizoguchi, *Chem. Phys. Lett.*, **158**, 383 (1989).

10) E. Heilbronner, *Tetrahedron Lett.*, **1964**, 1923; H. E. Zimmerman, *J. Am. Chem. Soc.*, **88**, 1564 (1966); H. E. Zimmerman, "Quantum Mechanics for Organic Chemistry," Academic press, New York (1975); M. J. S. Dewar and R. C. Dougher, "The PMO Theory of Organic Chemistry," Plenum Press, New York and London (1975).

11) N. Mizoguchi, *J. Phys. Chem.*, **92**, 2754 (1988).

12) A similar opposite tendency between Hückel systems and Möbius systems was found also in magnetic susceptility. See N. Mizoguchi, *Chem. Phys. Lett.*, **106**, 451 (1984); N. Mizoguchi, *Chem. Phys. Lett.*, **134**, 371 (1987); N. Mizoguchi, *J. Mol. Struct. (THEOCHEM.)*, **181**, 245 (1988).

13) W. C. Herndon, *J. Am. Chem. Soc.*, **104**, 3541 (1982).

14) I. Gutman and W. C. Herndon, *Chem. Phys. Lett.*, **105**, 281 (1984).

15) A. Graovac and N. Trinajstić, *Croat. Chem. Acta*, **47**, 95 (1975).

16) J. Aihara, *J. Am. Chem. Soc.*, **101**, 5913 (1979).

17) R. B. Mallion, *Proc. R. Soc. London, Ser. A.*, **341**, 429 (1975).

18) N. Mizoguchi, *J. Am. Chem. Soc.*, **107**, 4419 (1985).

19) O. J. Heilman and E. H. Lieb, *Commun. Math. Phys.*, **25**, 190 (1972).

20) J. Aihara, *Bull. Chem. Soc. Jpn.*, **52**, 1529 (1979); L. J. Schaad, B. A. Hess, Jr., J. B. Nation, and N. Trinajstić, *N. Croat. Chem. Acta*, **51**, 233 (1979); A. Graovac, *Chem. Phys. Lett.*, **82**, 248 (1981); I. Gutman, A. Graovac, and B. Mohar, *Match*, **13**, 129 (1982); A. Graovac and O. E. Polansky, *ibid.*, **21**, 33 (1986); A. Graovac and O. E. Polansky, *ibid.*, **21**, 81 (1986); H. Hosoya and K. Balasubramanian, *J. Comput. Chem.*, **10**, 698 (1989).