Resonance Energy in Graphite

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According to Zhu et al. the resonance energy/electron (REPE) in infinite graphite sheets is equal to 0.17 eV. In the present work the REPE was calculated for parallelogram-shaped graphite sheets (PSGSs). The number of Kekulé structures and contributions of benzene-like and naphthalene-like conjugated circuits were taken into account. Analogously to polyacenes, it was found that REPE = 0.00 eV in PSGSs. The convergence is slow. These results indicate that PSGS is less "aromatic" than its infinite counterpart. Therefore addition reactions are expected to be less difficult to carry out in PSGSs than in infinite or rectangular graphite sheets.

INTRODUCTION

Graphite is composed of "infinite" (two-dimensional) sheets, which themselves are made up by hexagonal lattices consisting of sp^2 carbon atoms. Each carbon contributes exactly one π-electron to the system. Therefore the number of carbon atoms and the number of π-electrons is equal. The parallel sheets are held together by dispersion forces, being much weaker than the chemical bonds between the adjacent carbon atoms of the same sheet. Graphite sheets are therefore special cases of polycyclic conjugated "benzenoid" structures,[1] like coronene, polyacenes, polyphenanthrenes, etc. Each benzenoid structure is characterized by the number of ways the double bonds are assigned to the carbon atoms. Each actual allocation of the double bonds is a Kekulé structure. The number of Kekulé structures (or Kekulé-count, K) is a measure of the stability of the underlying structure.

The enumeration of Kekulé structures in honeycomb lattice strips has been carried out by using the transfer matrix approach.[2] Klein also devised a Hückel-model solution for poly-polyphenanthrene strips.[3] The resonance in elemental benzenoids was investigated by Klein and Zhu.[4] The number of resonance structures in concealed non-Kekuléan benzenoid hydrocarbons was determined by Dias and Cash.[5] Ivanciuc, Klein, and Bytautas investigated the unpaired π-spin density in defected graphite by using band-theoretic methods.[6]

The stability of benzenoid structures may most elegantly be accounted for in terms of the resonance-energy, which can be reduced to the enumeration of "conjugated circuits" (CCs) in the benzenoid under investigation.[7] The problem of enumeration of CCs is in turn closely related to the enumeration of Kekulé structures.[7] The method of CCs used in its present form was proposed by Randić[8] and was subsequently applied to a great number of benzenoid structures. It has to be mentioned that Herndon discovered[9] the method of CCs already in 1973. Nearly 30 years after this discovery, it was again Randić, who published a comprehensive review on aromaticity (a concept closely related to resonance energy) and the role of CCs in polycyclic

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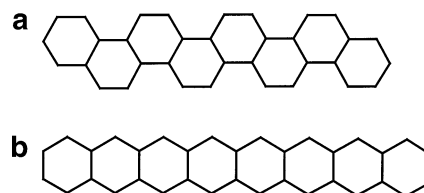
Figure 1. Polyphenanthrene (a), REPE = 0.140 eV, and polyacene (b), REPE = 0.058 eV. Both structures are composed of eight hexagons. The double bonds are not shown for the sake of clarity.

conjugated hydrocarbons.[10] How fruitful the concept of CCs is, can be illustrated by the fact, that Randić cited more than 1000 (!) literature references in his review. Recently, Randić introduced a numerical representation of the Kekulé count of aromatic structures.[11] This new concept has already been applied to the ordering of Kekulé structures.[12]

Before discussing the stability of graphite sheets, let us consider a closely related problem: It was demonstrated[13] that the value of the resonance energy/electron (REPE) is 0.141 eV in polyphenanthrenes (Figure 1) of infinite length, whereas its value is equal to 0 eV in polyacenes (Figure 1) of infinite length.[13] By applying periodic boundary conditions, and second quantization, it was found[1,14] that the REPE is approximately equal to 0.17 eV in infinite graphite sheets.

The aim of the present work was to determine the value of the REPE in parallelogram-shaped graphite sheets (PSGSs). It has to be noted that the number of Kekulé structures is equal to $h + 1$ (h denotes the number of hexagons in the polyacene molecule) in polyacenes, whereas in their cyclic analogues, (polyacene-belts) the number of Kekulé structures is equal to four, irrespectively of the number of hexagon units involved. A similar relationship exists between the number of Kekulé structures in PSGSs and their cyclic analogues.[15] The value of K is nearly identical in polyphenanthrenes and the cyclic analogue (polyphenanthrene belt), provided that h is large enough.[16] In this work it will be demonstrated that in PSGSs the value of index REPE is equal to 0 eV. It seems that PSGSs have been less often investigated than their infinite counterpart, although the Wiener index[17] and a formulas for K in various kinds of graphite sheets (including PSGSs) have been derived.[18,19] The
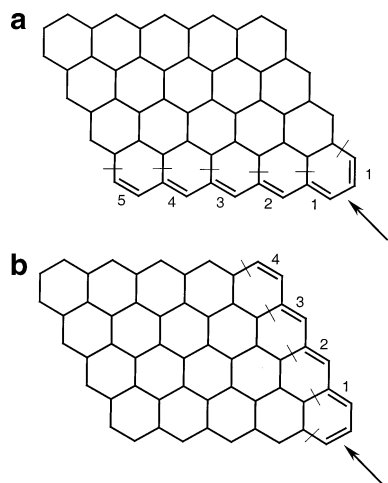
Figure 2. Hydrogen suppressed scheme of (4,5) graphite sheet with a single bond (a) and a double bond (b) at the specified site. The numbers denote the bonds which are double bonds as a consequence of the definition of the bond at the specified edge.

Table 1. Number of Kekulé Structures, $K_{m,n}$, in (m,n) Parallelogram-Shaped Graphite Sheets

row/column	1	2	3	4	5	6
1	2	3	4	5	6	7
2	3	6	10	15	21	28
3	4	10	20	35	56	84
4	5	15	35	70	126	210
5	6	21	56	126	252	462
6	7	28	84	210	462	924

reader might want to study a review[20] on Kekulé structures and benzenoids which has already been published in 1988.

ENUMERATION OF KEKULE STRUCTURES AND CONJUGATED CIRCUITS IN GRAPHITE SHEETS

Figure 2 displays a (4,5) graphite(like) sheet possessing four "rows" and five "columns" of hexagons with an arrow at the lower rhs pointing to a bond which is either a single bond (Figure 2a) or a double bond (Figure 2b). Once this bond is defined (fixed), the bonds denoted by index 1 must be double bonds, and if the bonds denoted by 1 are double bonds, the bonds denoted by index 2 must also be double bonds. In a similar manner it can be shown that bonds denoted by indices 3, 4, and 5 must also be double bonds. The thin lines crossing one of the single bonds indicate that the respective bond *must* be a single bond. In this way we have shown that the number of Kekulé structures of the (4,5) sheet, $K_{4,5}$ is equal to the sum of $K_{3,5}$ and $K_{4,4}$; i.e., $K_{4,5} = 56 + 70 = 126$ (Table 1). In general for any (m,n) sheet we obtain

$$K_{m,n} = K_{m-1,n} + K_{m,n-1} \tag{1}$$

Because $K_{1,n} = K_{n,1} = n + 1$,[20] eq 1 allows to derive the value of $K_{m,n}$ for any PSGS, composed of m rows and n columns. Table 1 lists the values of $K_{m,n}$ for several values of m and n ($m, n \leq 6$). Equation 1 remains valid if $K_{m-1,n}$ and $K_{m,n-1}$ are equal to zero. ($K_{m,n}$ is zero if the actual structure cannot be decomposed into 1-factors, i.e., it is impossible to assign the double bonds in such a way that each carbon atom has one incident double bond.)

To derive an analytical formula for $K_{m,n}$, we proceed in an identical manner as in the derivation of the number of

Table 2. Number of "Benzene-like" Conjugated Circuits ($6_{m,n}$) in (m,n) Parallelogram-Shaped Graphite Sheets

row/column	1	2	3	4	5	6
1	2	4	6	8	10	12
2	4	12	24	40	60	84
3	6	24	60	120	210	336
4	8	40	120	280	560	1008
5	10	60	210	560	1260	2520
6	12	84	336	1008	2520	5544

Morgan-trees.[21] Observe (Table 1) that $K_{1,n}$ is equal to

$$K_{1,n} = (n + 1)/1! \tag{2}$$

whereas $K_{2,n}$ is equal to

$$K_{2,n} = (n + 1)(n + 2)/2! \tag{3}$$

and

$$K_{3,n} = (n + 1)(n + 2)(n + 3)/3! \tag{4}$$

In general we obtain

$$K_{m,n} = (m + n)!/(m!n!) \tag{5}$$

Eq 5 is obviously valid for entries $n, m \leq 6$ (Table 1). Let us suppose that eq 5 remains valid for a $(m-1,n)$ and a $(m,n-1)$ sheet. Then $K_{m-1,n} = (m + n - 1)!/[(m - 1)!n!]$, and $K_{m,n-1} = (m + n - 1)!/[m!(n - 1)!]$. Therefore

$$K_{m-1,n} + K_{m,n-1} = \{(m + n - 1)!/[(m - 1)!n!]\} + \{(m + n - 1)!/[m!(n - 1)!]\} = (m + n)!/(m!n!) = K_{m,n} \tag{6}$$

and we have proved eq 5, by induction. It has to be noted that eq 5 has been derived earlier by using a different argumentation.[18]

The derivation of the number of CCs composed of benzene rings ($6_{m,n}$) and naphthalene rings ($10_{m,n}$) in (m,n) PSGSs is a more complicated task. In any graph G, the contribution of (benzene- or naphthalene-like) CCs to the overall count is equal to $\Sigma[2 \times$ (number of Kekulé structures in G$')]$, where G$'$ denotes a subgraph obtained from G by deleting the actual CC—the contribution of which is sought—and all incident edges. (The number of Kekulé structures in G$'$ will be denoted by $K_G{'}$, hereafter.) The summation has to be performed for all benzene-like and naphthalene-like CCs in G. The first factor equal to two indicates that the actual CC has two Kekulé structures, and this factor has to be multiplied by the number of the Kekulé structures in G$'$.

Table 2 lists the number of "benzene-like" CCs ($6_{m,n}$) for several values of m and n ($m, n \leq 6$). Computational details of this (simple) procedure have been omitted. Analogously to eqs 2—4 we obtain
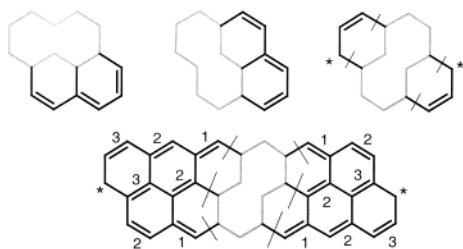
$$6_{1,n} = 2n/0! \tag{7}$$

$$6_{2,n} = 2n(n + 1)/1! \tag{8}$$

$$6_{3,n} = 2n(n + 1)(n + 2)/2! \tag{9}$$

and in general

$$6_{m,n} = 2(m + n - 1)!/[(m - 1)!(n - 1)!] \tag{10}$$

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Figure 3. Upper row, left to right: the definition of the horizontal, vertical, and skew naphthalene-like conjugated circuits in pyrene. Lower row: a (2,6) PSGS containing a skew naphthalene-like conjugated circuit. The conjugated circuit (in gray) has been indicated by its contours.
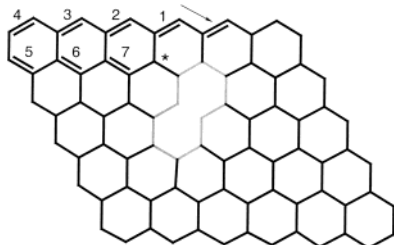
Figure 4. (5,6) PSGS with a skew naphthalene like conjugated circuit in the second and third rows. The conjugated circuit (in gray) has been indicated by its contours. The fixed bond has been indicated by an arrow,

Observe (Table 2) that

$$6_{m,n} = 6_{m,n-1} + 6_{m-1,n} + 6_{m-1,n}/(m-1) \qquad (11)$$

In complete analogy with the proof used to derive eq 5, it can be shown from eqs 6–9 and 11 that eq 10 is valid for any (m,n) PSGS.

The number of naphthalene-like CCs in (m,n) PSGSs will be denoted by $10_{m,n}$. Figure 3 defines the "horizontal", "vertical", and "skew" alignments of the naphthalene-like CCs (gray contours) in pyrene, respectively (upper row), and a more extended structure containing a "skew" naphthalene-like CC (lower row). The bonds indicated by number one (Figure 3, lower row) must be double bonds. The bonds denoted by number two must be double bonds if bonds denoted by number one are double bonds, etc. The "skew" orientation will lead to two unpaired electrons. The asterisk denotes that the actual atom contains unpaired electron and therefore the structure is non-Kekuléan. This observation is valid for any PSGS: Figures 4 and 5 demonstrate a more general example: a (5,6) PSGS containing a "skew" naphthalene-like CC in the second and third rows. The arrow no. 1 indicates the bond which is once a double bond (Figure 4) and once a single bond (Figure 5). The numerals define the logical order of inference, as a result of which the actual bond i is (provided that preceding bonds 1, 2, ..., i-1, are double bonds) a double bond. Therefore the structure is non-Kekuléan. Obviously, the Kekulé-count for such a structure is equal to zero.

Figure 5 contains a second (numbered) arrow defining the position of the vertical double bond (VDB). The first row of any PSGS can contain just a single VDB. Let us assume that there is a structure G″ containing *two* VDBs, $d_1 = (x_1, y_1)$ and $d_2 = (x_2, y_2)$ in the same row, where x denotes the peripheral vertex, and y denotes the internal vertex. Evidently, the number of peripheral C–C bonds separating x_1 and x_2 is
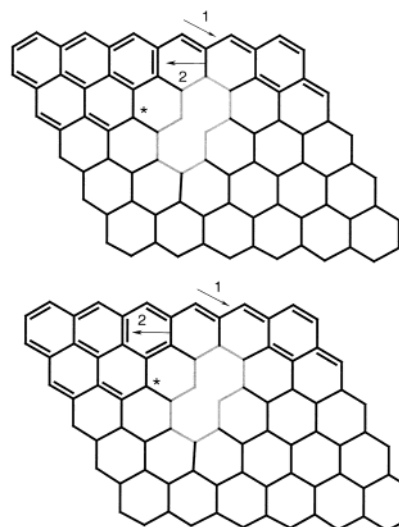
Figure 5. (5,6) PSGS with a skew naphthalene like conjugated circuit in the second and third rows. The conjugated circuit (in gray) has been indicated by its contours. The fixed bonds have been indicated by numbered arrows (see text).
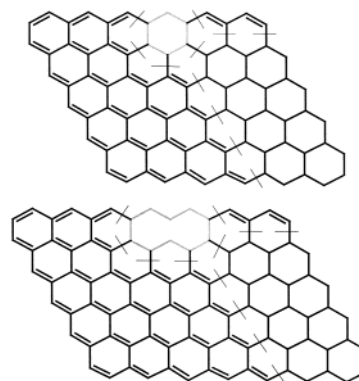
Figure 6. An (5,6) PSGS containing a benzene-like conjugated circuit and a (5,7) PSGS containing a naphthalene-like conjugated circuit in the first row.

even, containing a single bond at the beginning and at the end of the path. However, the path must contain an equal number of single and double bonds. The last two propositions cannot be satisfied simultaneously; therefore, we have proved that G″ cannot exist and therefore there cannot be two VDBs in the first row of PSGSs. The same reasoning applies to the last row of any PSGS.

The structures shown in Figure 5 display different positions of the VDBs, and in both cases the resulting structure will contain unpaired electrons, irrespectively of the position of the VDB. Similarly, at any position of the VDB a non-Kekuléan structure will result. It can be shown easily, that if the naphthalene-like CC is placed in the first row of any PSGS, then always an open-shell structure results.

Now the Kekulé count of any PSGS can be obtained by using eq 1. Starting with a structure containing a "skew" naphthalene-like CC in the first and second row, the Kekulé count of any (m,n) PSGS is the sum of $K_{m-1,n}′$ and $K_{m,n-1}′$, and it will always be equal to zero. Therefore we have shown that PSGSs do not contain "skew" naphthalene-like CCs.

Figure 6 displays one (5,6) PSGS containing a "benzene-like" CC and a (5,7) PSGS containing a "horizontal" naphthalene-like PSGS in the first row. Depending on the position of the respective CC, the left-hand side of each

Table 3. Enumeration of Kekulé Structures in a PSGS with a "Benzene-like" CC in the Fourth Position of the First Row (Figure 6)[a]

row/column	1	2	3	4	5	6
1	0	0	0	0	1	1
2	0	0	0	1	2	3
3	0	0	0	1	3	6
4	0	0	0	1	4	10
5	0	0	0	1	5	15
6	0	0	0	1	6	21

[a] The total count is $K = 21$.
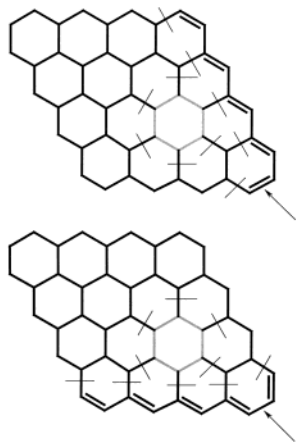
Figure 7. Decomposition of a (4,4) sheet containing a benzene-like CC (in position 3,3) into (3,4) and (4,3) sheets.

structure is fixed, and there are only $m - 1$ rows and $n - j$ or $n - j - 1$ columns, respectively, with nonfixed double bonds, where j denotes the position of the benzene (or of the left-hand side of the naphthalene) ring. Observe that the sector containing a nonfixed double bond is identical in both structures. Both structures can be extended in all directions, and an $(m, n-1)$ PSGS enclosing a benzene-like CC will contain the same number of Kekulé structures as an (m, n) PSGS enclosing a horizontal naphthalene-like CC. The Kekulé count of any (m, n) PSGS containing a benzene-like CC in position j of the first row is equal to $K_{m-1, n-j}'$. Similarly, the Kekulé count of any (m, n) PSGS containing a naphthalene-like CC in positions j and $j+1$ of the first row is equal to $K_{m-1, n-j-1}'$. The numerical procedure (eq 1) is illustrated in Table 3.

Figure 7 is an illustration of the evaluation of a (4,4) PSGS containing a CC in position (3,3). The number of Kekulé structures is equal to the sum of $K_{3,4}' + K_{4,3}' = 12$. It can be seen that the same decomposition procedure used to derive eq 1 remains valid, but each entry in the first row and column of the matrix is equal to the number of Kekulé structures in the upper left-hand corner (i.e. in this case; six). As an illustration let us enumerate the Kekulé structures in a (6,7) PSGS sheet, containing a "benzene-like" CC in position 3,3. From Table 4 we obtain that entry 4,5 is equal to 210; therefore, $K_{6,7}' = 210$. The total contribution of this CC in position 3,3 is therefore equal to $2 \times 210 = 420$.

All these arguments remain valid if the words "row" and "column" are interchanged. Therefore the sum of horizontal and vertical contributions of naphthalene-like CCs ($10_{m,n}$) is equal to

$$10_{m,n} = 6_{m-1,n} + 6_{m,n-1} \quad (12)$$

Table 4. Enumeration of Kekulé Structures in the Lower Right-Hand Side of Any $(m+2, n+2)$ PSGSs with a Pyrene-like Structure in the Upper Left-Hand Corner and a Conjugated "Benzene-like" Circuit in Position 3,3[a]

row/column	2+1	2+2	2+3	2+4	2+5	2+6
2+1	0	6	6	6	6	6
2+2	6	12	18	24	30	36
2+3	6	18	36	60	90	126
2+4	6	24	60	120	210	336
2+5	6	30	90	210	420	756
2+6	6	36	126	336	756	1512

[a] The total Kekulé count of the structure depicted in Figure 7 may be found in the entry 2+2, 2+2 (i.e. $K' = 12$).

Table 5 lists the number of naphthalene-like CCs in (m,n) PSGSs. From eqs 11 and 12 it follows that if m and n are sufficiently large, then $6_{m,n} = 10_{m,n}$ in PSGSs. Setting m equal to n, and from eqs 11 and 12 we obtain after simple manipulations that

$$10_{n,n} = 2\{(2n - 2)!(2n - 2)/[(n - 1)!]^2\} \quad (13)$$

In any aromatic system G the REPE is equal to[7,22]

$$\text{REPE}_G = (6_G R_6 + 10_G R_{10})/(K_G \#_G) \quad (14)$$

where R_6 and R_{10} denote the energy contribution of a single benzene-like or naphthalene-like CC, respectively, and $\#_G$ denotes the number of carbon atoms in the actual structure G. For (n,n) PSGSs $\#_{n,n}$ is equal to

$$\#_{n,n} = 2(n^2 + 2n) \quad (15)$$

From eqs 5, 10, 13, 14, and 15 we obtain after simplification that

$$\text{REPE}_{n,n} = [n R_6/(2n^2 + 4n)] + \{(n^2 - n)R_{10}/ \\ [(2n - 1)(n^2 - 2n)]\} \quad (16)$$

where two lower indices indicate the number of rows and columns in the PSGSs. The values of the parameters R_6 and R_{10} were taken from the literature[22] and are as follows: $R_6 = 0.8220$ and $R_{10} = 0.3355$ (eV).

The value of the third parameter (i.e. the contribution of an anthracene-like CC) in this series is rather small[22] ($R_{14} = 0.058$ eV). Whether it can be neglected or not, will depend of course of the number of anthracene-like CCs ($14_{m,n}$) in (m,n) PSGSs. Argumentation leading to eq 12 can be extended to anthracene-like CCs, and we obtain

$$14_{m,n} = 6_{m-2,n} + 6_{m,n-2} \quad (17)$$

Therefore $14_{m,n}$ is less than $10_{m,n}$, and its contribution to the REPE can be safely neglected.

RESULTS AND DISCUSSION

Figure 8 shows the dependence of the values of the $\text{REPE}_{n,n}$ in terms of n (the number of hexagon "rows" and "columns") in PSGSs. In addition to Figure 8 we find that $\text{REPE}_{1000,1000} = 0.00058$ eV, $\text{REPE}_{10000,10000} = 0.00006$ eV. At sufficiently large values of n, $\text{REPE}_{n,n} = 0$. The

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Table 5. Number of "Naphthalene-like" Conjugated Circuits ($10_{m,n}$) in (m,n) Parallelogram-Shaped Graphite Sheets

row/column	1	2	3	4	5	6
1	0	2	4	6	8	10
2	2	8	18	32	50	72
3	4	18	48	100	180	294
4	6	32	100	240	490	896
5	8	50	180	490	1120	2268
6	10	72	294	896	2268	5040
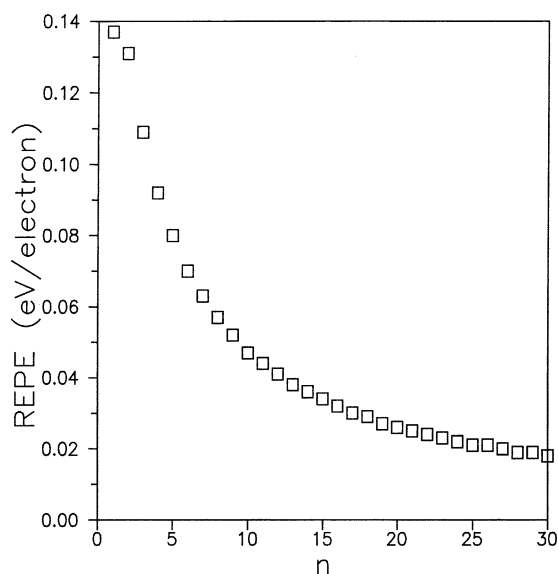
Figure 8. The value of the $REPE_{n,n}$ (eV) in terms of n in (n,n) graphite sheets.

convergence is slow. Observe that the left-hand side of eq 15 will always be equal to zero if m and n are sufficiently large, *irrespectively* of the actual value of the parameters R_6 and R_{10} (or R_{14}). This result indicates that the REPE in PSGSs is less than the corresponding value (0.17 eV) in infinite graphite sheets.[1] Therefore PSGSs are less "aromatic" than their infinite counterparts. In terms of the resonance energy, rectangular graphite is an extended version of polyphenanthrene, whereas PSGS is an extended version of polyacene.

The expression being less aromatic means that addition reactions[23] are most probably easier to accomplish in PSGSs than in their infinite counterparts. This means that starting from PSGSs, functionalized graphite sheets could be manufactured.

Concerning the aromatic character, PSGSs are less aromatic than buckminsterfullerene[24] ($REPE_{buckminsterfullerene}$ = 0.120 eV), naphthalene[22] ($REPE_{naphthalene}$ = 0.132 eV), pyrene[22] ($REPE_{pyrene}$ = 0.133 eV), benzene[7] ($REPE_{benzene}$ = 0.137 eV), phenanthrene[22] ($REPE_{phenanthrene}$ = 0.138 eV), benzphenanthrene[22] ($REPE_{benzphenanthrene}$ = 0.139 eV), triphenylene[22] ($REPE_{triphenylene}$ = 0.147 eV), and armchair carbon nanotubes (ACNT)[1,25] (0.160 eV ≤ $REPE_{ACNT}$ ≤ 0.170 eV).

Reactivity of graphite sheets will also depend on the edge types.[6,26] Seitz et al.[27] suggested that zigzag edges will be more reactive than the armchair counterparts. Therefore PSGSs will be more reactive, because aromaticity in PSGSs is less, and the reactivity of the edges are greater than in rectangular graphite.

The numerical procedure (eq 1) used to determine the number of Kekulé structures in PSGSs is identical to the procedure used to obtain the Catalan numbers (i.e. the number of Morgan trees).[28] However, the analogy is superficial; the starting values are different in the two cases. In addition the diagonal values of the Catalan numbers are zero, whereas they are not zero in the case of Kekulé structures.

To calculate the Kekulé count exactly, Živković et al.[29] devised a method which makes it possible to manipulate very large (up to 500 digits) numbers and applied this approach to benzenoid polymers. At the same time Živković et al.[29] derived analytical formulas for polymers. Both Živković's and the present results indicate that the formulas can be easily handled. The actual values of the REPE will nevertheless be small (i.e. ≤ 1.00) numerals, where the first three digits are important, only.

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