Enumeration of Conjugated Circuits in Nanotubes[†]

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The resonance energy of $(1,1)_n$ "armchair" carbon nanotubes and $(n,n)_1$ nanoribbons was determined by enumerating the conjugated circuits (CC) and the Kekulé structures. The lower indices denote the number of hexagon layers. It was found that the resonance energy per carbon atom is equal to 0.160 eV in $(1,1)_n$ tubes and 0.142 eV in $(n,n)_1$ tubes.

INTRODUCTION

Carbon nanotubes (CNTs) were discovered by Iijima[1] in 1991. CNTs are composed of cylindrical graphite sheets made up by sp^2 carbon atoms. This fact implies that with respect to the π-electron-structure, CNTs are similar to graphite and aromatic benzenoid molecules. Therefore the conjugated circuit model,[2] which was used successfully in the case of benzenoids, might be of use in estimating the resonance energy—and stability—of various CNTs.

Because of its importance, various approaches have been used to account for stability and other physical properties of CNTs. The most obvious method seems to be quantum chemistry, but the usual size of the objects to be investigated renders such research impossible. However, small, "CNT-like" molecules have been investigated by using quantum-chemical approaches. Cioslowski, Rao, and Moncrieff[3] found by using the density-functional approach, that the metallic character of CNTs is manifested by oscillatory dependence of the properties in terms of their size. A similar result has been reported by Li, Zhang, and Zhang.[4] Semiempirical approaches have been used by Erkoç,[5] who found that the energy of the CNTs increases linearly in terms of the tube length. The mechanism of formation of CNTs from diamond was studied by Kuznetsov et al.[6] by using the MNDO semiempirical approach. By using a local-density functional method Liu and Chan[7] found that (5,0) and (3,3) tubes are metallic, while the (4,2) tube is semiconducting. The conductivity of single-walled CNTs was computed by using the transfer matrix technique.[8] Klein et al.[9] were able to prove analytically that in (n,m) tubes, if $n-m$ is divisible by three (i.e. $n - m = 0$ mod 3), then the band gap will be zero. If $n - m = 0$, then the CNTs will be a metallic conductor. In the case of (n,n) CNTs this rule does not hold strictly; curvature effects may lead to nonzero band gaps.[10] The asymptotic band gap behavior depends on the type of the CNT: it may be different in tubes with various caps and in tori and open tubes.[11] (In the present paper open tubes were considered.)

Note, that the leapfrog operation, a graphical tool used to generate the graphs of fullerenes, tubulenes, and toroids, does not change the character of the resulting object: if a tubule is a leapfrog of another, then it is metallic and has four non-bonding orbitals, if it is not a leapfrog, then it has a properly closed shell with a nonzero HOMO−LUMO gap.[12] It was found that the magnetic field influences the electronic structure of CNTs.[13]

Molecular mechanics has also been used to model the strain in CNTs containing nonhexagonal rings.[14] The same approach was used to investigate the effect of substitution of dicoordinated carbon atoms by heteroatoms in tubulenes.[15] The fracture of CNTs was also studied by molecular mechanics.[16] The energetic stability of tori was investigated by Diudea and Kirby, who found that untwisted thin tori are more stable than analogues which are twisted and thick.[17] The effect of chirality was studied by Erkoç and Malcioğlu,[18] who found that chiral CNTs are more stable than zigzag or armchair CNTs. Thin (diameter is less than 10 Å) single-walled CNTs are more stable if they are circular, while with increasing diameter the collapsed form tends to be more stable.[19] The reader may wish to consult the general review by Saito, Dresselhaus, and Dresselhaus.[20] Recently Randić has published a comprehensive review on aromaticity of polycyclic conjugated hydrocarbons.[21]

Besides quantum chemistry and molecular mechanics the third approach used to estimate the stability of CNTs is the estimation of resonance energy. This task can be achieved by using two methods: (1) by enumerating the Kekulé structures (K) and (2) by determining K and the number of conjugated circuits (CCs) in CNTs. Determination of K in CNTs (tubulenes) was first accomplished by Sachs, Hansen, and Zheng[22] and then by Klein and Zhu,[23] John,[24] and Lukovits et al.[25]

It has been discovered that the Cartesian coordinates of the carbon atoms in tubes can easily be computed by computing the eigenvalues of the adjacency matrix.[26,27]

The aim of this work was to estimate and compare the stability of $(1,1)_n$ (armchair) CNTs and $(n,n)_1$ CNTs by using the CC approach. The lower index denotes the number of hexagon layers. Therefore $(n,n)_1$ CNTs, which contain a single hexagon layer, will also be referred to as "nanorib-

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ENUMERATION OF CONJUGATED CIRCUITS IN NANOTUBES

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Figure 1. Enumeration of 6-cycles in a $(2,2)_1$-nanotube. A thin line crossing one of the bonds, indicates that the respective bond must be a single bond independently of the configuration of the CC (shaded area). The single bonds at the right and left-hand sides refer to hexagons already depicted in the drawing.

bons". Our results indicate that resonance energy per electron is greater in nanotubes than in nanoribbons.

ENUMERATION OF CONJUGATED CIRCUITS IN NANOTUBES

A conjugated circuit (CC) for a Kekulé structure on a graph G is a self-returning path, composed of a sequence of alternating single and double bonds. In systems consisting of hexagons, the simplest CC is a path composed of six vertices three single and three double bonds, and the number of such paths in G (following the notation used by Klein[28]) will be denoted by 6_G. Similarly the number of naphthalene-like CCs will be denoted by 10_G, and the corresponding number related to anthracene is 14_G. The enumeration of CCs was discussed in detail by Nikolić et al.[2] Our main task was to enumerate CCs in long and short CNTs and to obtain from these values the resonance energy/electron (REPE).

Two special cases have been considered: (1) the $(1,1)_n$ CNTs, composed of 1, 2,...,n layers (i.e. naphthalene units) and (2) cyclic $(n,n)_1$ nanoribbons composed of n naphthalene units forming a single hexagon layer. It has to be noted that $(1,1)_n$ nanotubes may not exist, because of the high strain of the distorted structures, which the resonance energy does not account for.

The value of the REPE of structure G was computed by using the following equation[2]

$$\text{REPE}_G = (6_G R_6 + 10_G R_{10})/(K_G \#_G) \qquad (1)$$

where R_6 and R_{10} denote the contributions of circuits containing six vertices (benzene-like circuits) and 10 vertices (naphthalene-like circuits), respectively, and K_G denotes the number of Kekulé structures in G. $\#_G$ is the number of carbons (or π electrons) in G. For our calculations the values $R_6 = 0.8220$ (eV) and $R_{10} = 0.3355$ (eV) were used.[29] In this parametrization the next term, accounting for the contribution of anthracene-like circuits, R_{14}, is so small ($R_{14} = 0.058$ eV), that it can be neglected. In this section the number of 6-circuits and 10-circuits will be determined in $(1,1)_n$, as well as in $(n,n)_1$ CNTs.

Figure 1 shows a $(2,2)_1$ tube and illustrates the method used to obtain a formula for $6^1_{(n,n)}$, where the upper index indicates that there is only a single layer. The single bonds at the left-hand side denote the hexagon at the right-hand side. The single bonds at the right-hand side denote the hexagon at the left-hand side. The "gray" hexagon denotes the CC under consideration, and all adjoining edges must be single bonds. These have been indicated using thin, intersecting lines. In the case $n = 2$ (i.e. there are only two naphthalene units per tube) we obtain

$$6^1_{(2,2)} = 2 \times 2nK^1_{\text{phen}} = 2 \times 4 \times 2 = 16 \qquad (2)$$

where the first factor of 2 indicates that the "gray" hexagon

Table 1. Number of Kekulé Structures in Polyphenanthrenes in Terms of the Number of Hexagons (h) and Number of Kekulé Structures in $(1,1)_n$ Tubes and Extended $(1,1)_n$ Tubes in Terms of Naphthalene Units $(n)^a$

h (n)	$K^h_{\text{polyphenanthrene}}$	$K^1_{(n,n)}$	$K^n_{(1,1)}$	$\underline{K}^n_{(1,1)}$
-2 (-1)	0		1	1
-1	1			
0 (0)	1		2	3
1	2			
2 (1)	3	5	5	8
3	5			
4 (2)	8	9	13	21
5	13			
6 (3)	21	20	34	55
7	34			
8 (4)	55	49	89	144
9	89			
10 (5)	144	125	233	377
11	233			
12 (6)	377	324	610	987
13	610			
14 (7)	987	845	1597	2584
15	1597			
16 (8)	2584	2209	4181	6765
17	4181			
18 (9)	6765	5780	10946	17711
19	10946			
20 (10)	17711	15129	28657	46368
21	28657			
22 (11)	46368	39605	75025	121393
23	75025			
24 (12)	121393	103684	196418	317811

^{a} $h = 2n$.

contributes two CCs itself, the second factor of $2n$ indicates the number of (in our case four) equivalent hexagons, and K^1_{phen} denotes the number of Kekulé structures of the "remaining part" (i.e. benzene) of the ribbon (in Figure 1 this is the "white" hexagon). Note that in general the number of Kekulé structures in the "remaining part" is equal to K^{2n-3}_{phen} in polyphenanthrene consisting of $2n-3$ hexagons, and the value of K^{2n-3}_{phen} can easily be obtained (Table 1) by using well-known recursion formulas derived for polyphenanthrenes.[30] The double bonds appearing in Figure 1 are the consequence of the fact that bonds incident with the CC *must* be single bonds, while the double bonds are the consequence of the fact that the system is conjugated. Therefore the generalization of eq 2 is straightforward:

$$6^1_{(n,n)} = 4nK^{2n-3}_{\text{phen}} \qquad (3)$$

The number of 10-vertex CCs is obtained analogously:

$$10^1_{(n,n)} = 4nK^{2n-4}_{\text{phen}} \qquad (4)$$

that is in the case of $(2,2)_1$ nanoribbon; $10^1_{(n,n)} = 4 \times 2 \times 1 = 8$ (since $K^0_{\text{phen}} = 1$). The actual values of $6^1_{(n,n)}$ and $10^1_{(n,n)}$ are displayed in Table 2.

Figure 2 illustrates the method used to obtain a formula for $6^4_{(1,1)}$. There are four nonequivalent positions of the CC. Observe that the gray hexagon in fact "dissects" the tube: in the case of the $(1,1)_4$ tube, $6^4_{(1,1)}$ is the sum of factors $2 \times$ (number of equivalent hexagons) $\times$ (number of Kekulé structures above the hexagon) $\times$ (number of Kekulé structures below the hexagon) for all four nonequivalent hexagons

Table 2. Number of Conjugated Cycles Composed of 6 or 10 Carbons, Respectively, in $(1,1)_n$ and $(n,n)_1$-Nanotubes in Terms of the Number of Naphthalene Units (n)

| n	$6^1_{(n,n)}$	$10^1_{(n,n)}$	$6^n_{(1,1)}$	$10^n_{(1,1)}$
1	4	0	4	
2	16	8	20	4
3	60	36	76	20
4	208	128	260	76
5	680	420	840	260
6	2136	1320	2616	840
7	6524	4032	7940	2616
8	19520	12064	23644	7940
9	57492	35532	69380	23644
10	167240	103360	201220	69380
11	481624	297660	578064	201220
12	1375536	850128	1647600	578064
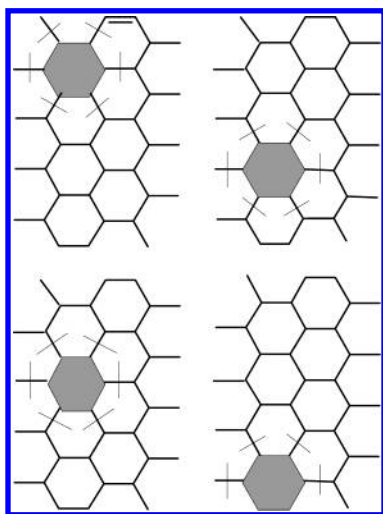
Figure 2. Enumeration of 6-cycles in a $(1,1)_4$-nanotube. A thin line crossing one of the bonds, indicates that the respective bond must be a single bond independently of the configuration of the CC (shaded area). The single bonds at the right and left-hand sides refer to hexagons already depicted in the drawing.

$$6^4_{(1,1)} = 4(\underline{K}^{-1}_{(1,1)} K^2_{(1,1)} + \underline{K}^0_{(1,1)} K^1_{(1,1)} + \underline{K}^1_{(1,1)} K^0_{(1,1)} + \underline{K}^2_{(1,1)} K^{-1}_{(1,1)}) \quad (5)$$

where $K^n_{(1,1)}$ denotes the number of Kekulé structures in $(1,1)_n$ CNTs. This is the number of Kekulé structures of the dissected tube "below" the gray hexagon. $\underline{K}^n_{(1,1)}$ denotes the number of Kekulé structures in the "extended" nanotubes (Figure 3). This is the number of Kekulé structures of the dissected tube "above" the gray hexagon. Extended $(1,1)_n$ tubes are composed of n naphthalene units and in addition contain an "extra" hexagon. Both $K^n_{(1,1)}$ and $\underline{K}^n_{(1,1)}$ can be calculated by using a recursion formula. (The computation of the actual values of $K^n_{(1,1)}$ will be explained at the end of this section.) The factor of 4 (eq 5) is the product of times (two) the number the "shaded" CC contributes to $6_3{}^{(1,1)}$ and the number of equivalent "gray" CCs (two). Application of eq 5 by inserting the values of the actual parameters yields

$$6^4_{(1,1)} = 4 \times (1 \times 13 + 3 \times 5 + 8 \times 2 + 21 \times 1) =$$
$$4 \times (13 + 15 + 16 + 21) = 260$$

The recursion formula for $(1,1)_n$ tubes has been derived earlier:[25]
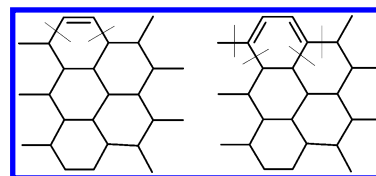
Figure 3. Enumeration of Kekulé structures in an "extended" $(1,1)_2$ nanotube. A thin line crossing one of the bonds indicates that the respective bond must be a single bond. The single bonds at the right and left-hand sides refer to hexagons already depicted in the drawing.

$$K^n_{(1,1)} = 3K^{n-1}_{(1,1)} - K^{n-2}_{(1,1)} \quad (6)$$

With $K^{-1}_{(1,1)} = 1$, $K^0_{(1,1)} = 2$.
In analogy with eq 5, we obtain for any $(1,1)_n$ tube

$$6^n_{(1,1)} = 4(\underline{K}^{n-2}_{(1,1)}K^{-1}_{(1,1)} + \underline{K}^{n-3}_{(1,1)} K^0_{(1,1)} +$$
$$\underline{K}^{n-4}_{(1,1)} K^1_{(1,1)} + ... + \underline{K}^{-1}_{(1,1)}K^{n-2}_{(1,1)}) \quad (7)$$

i.e. in each term of the right-hand side the sum of the subscripts is equal to $n-3$.

Inspection of Figure 2 makes clear that there are only six options to insert a naphthalenic CC into the $(1,1)_4$ nanotube. "Skew" naphthalenic CCs are not possible. The naphthalenic CC also dissects the tube, and an equation similar to eq 7 can be derived with the sum of the subscripts being equal to $n-4$. Therefore the value of $10^n_{(1,1)}$ can be obtained for the value of $6^{n-1}_{(1,1)}$:

$$10^n_{(1,1)} = 6^{n-1}_{(1,1)} \quad (8)$$

The actual values of $6^n_{(1,1)}$ and $10^n_{(1,1)}$ are displayed in Table 2.

A recursion formula has been derived to obtain the value of $\underline{K}^n_{(1,1)}$ in $(1,1)_n$ tubes. Figure 3 displays an "extended" $(1,1)_2$ tube; one column of this tube consists of three, and the second column consists of two hexagons. As it can be seen from Figure 3, the value of $\underline{K}^2_{(1,1)}$ can also be obtained in the following way: there are two possible Kekulé structures in the "extra" hexagon. The number of Kekulé structures related to the scheme on the left-hand side is equal to $K^2_{(1,1)}$, while the number of Kekulé structures related to the scheme on the right-hand side is equal to $\underline{K}^1_{(1,1)}$. Therefore $\underline{K}^2_{(1,1)} = 13 + 8 = 21$. This decomposition scheme applies for any positive value of n, and therefore we may write

$$\underline{K}^n_{(1,1)} = K^n_{(1,1)} + \underline{K}^{n-1}_{(1,1)} \quad (9)$$

with $\underline{K}^{-1} = 1$, $\underline{K}^0_{(1,1)} = 3$. Table 1 contains the respective values of the Kekulé counts.

The number of carbons in $(1,1)_n$, $\#^n_{(1,1)}$, and in $(n,n)_1$ tubes, $\#^1_{(n,n)}$, were obtained by using the following formulas:

$$\#^1_{(n,n)} = 8n \quad (10)$$

$$\#^n_{(1,1)} = 4n + 4 \quad (11)$$

Finally a numerical example will be given. From Tables 1 and 2 the resonance energy per electron in tube $(1,1)_5$ is equal to (eq 1): REPE = $[(840 \times 0.8220 + 260 \times 0.3355)/$
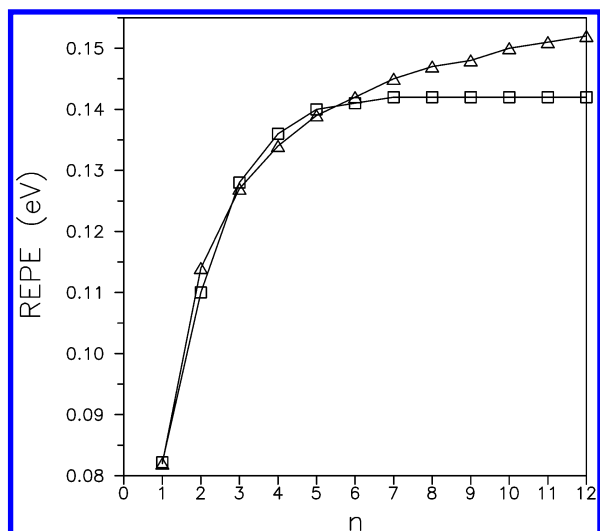
Figure 4. Variation of the resonance energy per electron (eV) in $(1,1)_n$ nanotubes ($\triangle$) and $(n,n)_1$-nanoribbons ($\square$) in terms of the naphthalene units.

233]/24 = 0.139075 eV. All calculations reported in this paper were done by using a programmable calculator.

RESULTS AND DISCUSSION

The values of REPE of $(1,1)_n$ CNTs and $(n,n)_1$ nanoribbons are depicted in terms of the naphthalene units n in Figure 4. As it can be seen up to $n = 6$, REPE is practically equal in the two systems, but starting with $n = 7$, REPE is greater in $(1,1)_n$ CNTs than in the $(n,n)_1$ counterparts. In fact, it seems that the value of the REPE converges to 0.160 eV in $(1,1)_n$ tubes. REPE is equal to 0.155 eV in a $(1,1)_{18}$ tube with $K^{18}_{(1,1)}$ = 63245986, $6^{18}_{(1,1)}$ = 789810984, $10^{18}_{(1,1)}$ = 285174360, and $\#^{18}_{(1,1)}$ = 76. (For the same system REPE = 0.158 eV, if the parameters by Hite et al.[31] are used.) REPE = 0.142 eV in a $(18,18)_1$ tube. In accordance with our earlier investigations,[25] the long (and thin) tube is therefore more stable than the thick (but short) nanoribbon. In $(1,1)_n$ tubes the value of REPE ($n \geq 20$) is about 0.10 eV less than in graphite[31,32] ($\cong 0.170$ eV). Therefore, it can be expected that $0.158 \leq$ REPE ≤ 0.170 in any $(m,m)_n$ tube, with $m \ll n$. Because of the high resonance energy, tubes with relative positive strain energy may also exist. The existence of extremely thin nanotubes of 0.75 nm diameter CNTs has already been reported.[33] This diameter may correspond to a (5,5) "armchair" CNT.

It is worthwhile to compare the values of REPEs obtained for CNTs with values obtained for other aromatic systems. REPE is greater both in $(1,1)_n$ and $(n,n)_1$ tubes than in benzene (0.137 eV), naphthalene (0.132 eV), phenanthrene[29] (0.138 eV), pyrene[29] (0.133 eV), and benzphenanthrene[29] (0.139 eV), However, the value of the REPE is greater in triphenylene[29] (0.147 eV) than in any $(n,n)_1$ tube, but it is less than the value of REPE in $(1,1)_n$ tubes with $n \geq 8$.

It was found that in polyacenes, or in aromatic hydrocarbon molecules to which polyacenees are attached, the REPE decreases with the increasing size of the polyacene chain.[29] Denoting the number of hexagons by h, it can be shown (details have been omitted) that the values of parameters $\#^h_{\text{polyacene}}$, $K^h_{\text{polyacene}}$, $6^h_{\text{polyacene}}$, and $10^h_{\text{polyacene}}$ are equal to

$$\#^h_{\text{polyacene}} = 4h + 2 \tag{12}$$

$$K^h_{\text{polyacene}} = h + 1 \tag{13}$$

$$6^h_{\text{polyacene}} = 2h \tag{14}$$

$$10^h_{\text{polyacene}} = 2h - 2 \tag{15}$$

From eqs 1 and 12−15 it follows that REPE$_{\text{polyacene}}$ = 0 in any parametrization, in accordance with Zhu and Klein's results.[34] Nanotubes of any kind are therefore more "aromatic" than polyacenes.

With polyphenanthrenes the situation is different. Numerical calculations indicated[34] that the REPE$_{\text{polyphenanthrene}} \cong$ 0.141 eV; therefore, polyphenanthrenes are about as aromatic as their cyclic analogues, the $(n,n)_1$ nanoribbons, but are less aromatic than nanotubes.

It has to be noted that the value of REPE in buckminsterfullerene is equal to[35] 0.12 eV, and, therefore, it is also less than REPE in nanotubes. In conclusion, carbon nanotubes belong to the most aromatic systems.

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