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## DISTANCE BASED TOPOLOGICAL INDICES ON MULTIWALL CARBON NANOTUBES SAMPLES OBTAINED BY ELECTROLYSIS IN MOLTEN SALTS

BETI ANDONOVIC, VESNA ANDOVA, TATJANA ATANASOVA PACEMSKA, PERICA PAUNOVIC, VIKTOR ANDONOVIC, JASMINA DJORDJEVIC AND ALEKSANDAR T. DIMITROV

**Abstract.** The interest for the intensive studies and methods of structural characterization of multiwall carbon nanotubes (MWCNTs) to date has resulted in many valuable contributions and an amazingly wide application area. This paper offers an approach that combines several techniques. It is the first direct application of the graph theory upon nanotubical structures obtained by electrolysis in molten salts using non-stationary current regimes. The spectroscopic data enables studying the diameters and performing an  $(n,m)$  assignment of nanotube samples. Using the graph representation and the chirality of the studied samples, different distance based topological indices (Wiener, Balaban, Sum-Balaban, and Gutman indices) have been evaluated in order to enable further prediction of index-related properties of the molecules.

### 1. Introduction

The electrochemical method is a proven low-cost method for a high-yield production of carbon based nanostructures [1-3]. Obtained carbon nanotubes' (CNTs) properties largely depend on their atomic structure, hence the determination of the CNT's diameter and/or of the chiral angle is essential. Accurate determination of chiral indices  $m$ ,  $n$  enables the calculations of any CNT's structural parameter. There are studies that suggest some methods of  $(m,n)$  assignment for SWCNTs, as well as for MWCNTs. However, precise determination becomes extremely complicated for more than three walls (layers) [4]. The obtained CNT samples that are the subject of this study were produced by electrolysis in molten salts using non stationary current regimes, and are multi-wall tubes having highly ordered structures and undetermined diameter and chirality. Therefore, this research is strictly focused on determining the outermost wall chiral indices and corresponding distance based topological indices of these graphitic nanostructures. In the chemical graph theory Quantitative Structure Activity Relations (QSAR) and Quantitative Structure Property Relations (QSPR) are showing the tendency of contemporary theoretical and mathematical chemistry to predict the properties of a compound based on its molecule structure. The research in this field is mainly done by combining graph molecular descriptors and experimental results by using a method of representing molecules as graphs. Each atom is presented as a vertex and the bonds between the atoms are the edges in the graph [5-10].

It is important to notice that for this study only the distance based topological indices such as Wiener index  $W(G)$ , Gutman index  $Gut(G)$ , Balaban index  $J(G)$ , and Sum-Balaban index  $SJ(G)$  are considered, as it is known that the graph representations of nanotubes are cubic or sub – cubic (all the vertices are of degree three except the vertices on the outer face). The results in this paper are motivated by [11,12], where some distance



based topological indices are evaluated for open and closed nanotubes of a given type. The number of vertices at a given distance from a vertex  $u$  on an infinite hexagonal tube was determined and hence used [12].

## 2. Materials and techniques

The carbonaceous phases extracted from the solidified electrolyte were observed by scanning with electron microscopy, using JEOL 6340F (SEM, 10 kV). The structural characteristics of the carbon nanostructures were studied by means of Raman spectroscopy. Non-polarized Raman spectra were recorded by a confocal Raman spectrometer (Lab Ram ARAMIS, Horiba Jobin Yvon) operating with a laser excitation source emitting at 532 nm. The Software Mathematica is applied to determine possible chiral indices assignment as well as the distance based topological indices.

## 3. Analysis and methods

Let  $G$  be a connected graph. By  $V(G)$  and  $E(G)$  the vertex and edge set of  $G$  are respectively denoted. Let  $k = |V(G)|$  and  $e = |E(G)|$ . Two vertices  $u, v \in V(G)$  being given,  $dist(u, v)$  is the distance from  $u$  to  $v$  in  $G$ . The degree (valence) of the vertex  $v$  is denoted by  $d(v)$ . The corresponding formulas are:

$$W(G) = \sum_{u,v \in V(G)} dist(u, v) \quad (3.1)$$

$$J(G) = \frac{e}{e-k+2} \sum_{uv \in E(G)} \frac{1}{\sqrt{w(u)w(v)}}, \quad \text{where} \quad (3.2)$$

$$w(x) = \sum_{y \in V(G)} dist(x, y)$$

$$SJ(G) = \frac{e}{e-k+2} \sum_{uv \in E(G)} \frac{1}{\sqrt{w(u)+w(v)}} \quad (3.3)$$

$$Gut(G) = \sum_{u,v \in V(G)} d(u)d(v)dist(u, v) \quad (3.4)$$

The CNT sample considered for this study, having a nomenclature CNT – 3, is a multiwall tube, having an outermost diameter estimated to  $\sim 30$  nm and a length  $\sim 5$   $\mu$ m. Part of its Raman spectrum is displayed on Fig 4.1B, showing its D and G modes. The ratio of their intensities  $I_D/I_G = 0.296$  is very low, which points to its highly ordered structure. Furthermore, the G-mode having two components (G- and G+) being both narrow and symmetric, identify the studied CNT as a semiconducting chiral one. The latter is used to narrow down the choice of the most probable chiral indices  $(m, n)$  assignment, as it must be satisfied that  $MOD(2m + n, 3) \in \{1, 2\}$  [13].

#### 4. Results and discussion

The obtained results are summarized with regard to the closest values of  $\tilde{A} = m^2 + n^2 + mn$  to  $A = \left(\frac{d}{78.3}\right)^2 (\pm 2 pm^2)$  [13] and presented in Table 4.1.

Table 4.1. Possible  $(m, n)$  assignment for CNT - 3 with estimated diameter (in nm)

d	A	(m, n)	$\tilde{A}$	MOD	Type	d	A	(m, n)	$\tilde{A}$	MOD	Type
25	101943	(44,295)	101941	2	ch. sc	30	146798	(12,377)	146797	2	ch. sc
		(95,261)	101941	1	ch. sc			(117,311)	146797	2	ch. sc
		(37,312)	110257	2	ch. sc			(172,267)	146797	2	ch. sc
26	110261	(123,253)	110257	1	ch. sc			(153,283)	146797	1	ch. sc
		(128,249)	110257	1	ch. sc			(165,273)	146799	0	ch. m.
		(176,207)	110257	1	ch. sc			(180,260)	146800	2	ch. sc
27	118906	(162,234)	118908	0	ch. m.			31	156747	(218,239)	156747*
28	127877	(66,320)	127876	2	ch. sc	32	167023	(54,379)	167023*	1	ch. sc
		(90,304)	127876	1	ch. sc	33	177625	(122,347)	177627	0	ch. m.
29	137174	(43,347)	137179	1	ch. sc	34	188553	(29,419)	188553*	0	ch. m.
		(118,297)	137179	2	ch. sc	35	199808	(143,358)	199807	2	ch. sc
By * are denoted the exact matches of $\tilde{A}$ to A.								(0,447)	199809	0	zz. m.

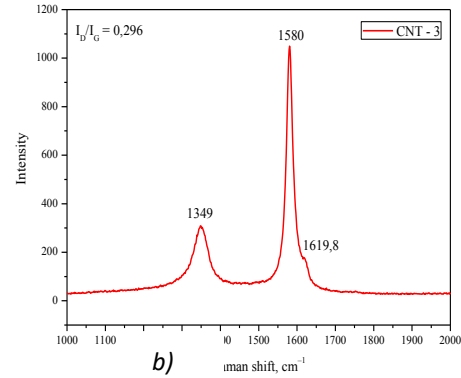
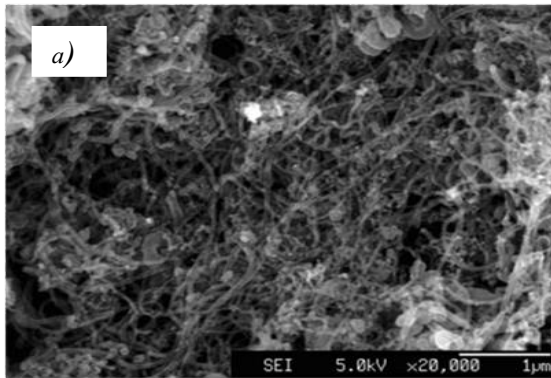


Figure 4.1. SEM image of CNTs obtained by electrolysis in molten salts (Fig. 4.1a); Raman spectrum D and G-modes display of CNT - 3 (Fig. 4.1b).

Possible assignments are considered with regard to  $|\tilde{A} - A| < 2$ , to non-metallic, and to diameters closest to 30 nm. Based on these results, there are seven possible chiral indices assignment out of 23 satisfying every condition. However, it should be stressed that (54, 379) is the most probable assignment, as the only chiral semiconducting CNT precisely satisfying  $|\tilde{A} - A| = 0$ .

In order to evaluate the distance based topological indices, the number of vertices/atoms in the nanotube is highly important. The vector  $(m, n)$  determines the

diameter of the tube, but the distance between any two vertices depends on the length of the nanotube as well, or on the length of its unit cell, which is a rectangular lattice determined by the vector  $(m, n)$  and the translation vector  $\vec{T}$ ,

$$\vec{T} = \frac{\sqrt{3}d}{d_R} \quad (4.1)$$

where  $d_R = GCD(2m + n, 2n + m)$ ,  $GCD$  standing for the greatest common divisor [13]. Using the length and the type of the nanotube, the number of vertices is asymptotically determined. The length of the translation vector and the length of the nanotube determine the number of (repeating) unit cells and hence the total number of vertices  $N$ . It is important to notice that it is possible to obtain the topological indices for the unit cell itself only and use them for further research, whenever the length of the nanotube is unknown for any reason. The obtained results are presented in Table 4.2.

Table 4.2. Summarized results for CNT - 3

<b>d</b> (in nm)	<b>(m,n)</b> assignment	<b>T</b>	<b>N<sup>uc</sup></b>	<b>N</b>	<b>W(G) *</b> <b>10<sup>19</sup></b>	<b>J(G)</b>	<b>SJ(G)</b> <b>* 10<sup>11</sup></b>	<b>Gut(G)</b> <b>* 10<sup>20</sup></b>
<b>28</b>	(66,320)	24249	255752	52684912	6.31423	110.2	1.57031	3.78854
<b>28</b>	(90,304)	24248	255752	52684912	6.18602	111.3361	1.60286	3.71161
<b>30</b>	(12,377)	51961	587188	56370048	7.6744	110.6274	1.69965	4.60464
<b>30</b>	(117,311)	51961	587188	56370048	6.9751	116.0405	1.87005	4.18506
<b>30</b>	(172,267)	51961	587188	56370048	6.80032	117.5222	1.91811	4.08019
<b>30</b>	(153,283)	51961	587188	56370048	6.84711	117.12	1.905	4.10827
<b>32</b>	(54,379)	43323	408172	46939780	5.02548	103.8807	1.23514	3.01529

## 5. Conclusion

Several methods and techniques have been combined to enable and propose this approach presented on the sample CNT – 3 for determination of its chiral indices assignment as well as of its distance based topological indices. As the latter was the most important aim, it is useful to notice that evaluating the topological indices for any CNT unit cell alone and regardless of the nanotube's length is possible using the same approach. However, the calculations of the indices herein are given considering the previously estimated value of the nanotube's length of 5000 nm. It is evident by the results that the determined indices distinguish the nanotubes with regard to their chirality and hence open a wide research area for finding which properties of the nanotubes could be described with evaluating the topological indices.

## 6. Acknowledgements

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