



VOLUME 2 ISSUE 2 MAY 2013

Manuscript ID : ISSN23197501-V2I2M2-0502013

SILICA NANOTUBE MUTITERMINAL JUNCTION AS A COATING FOR CARBON NANOTUBE JUNCTION

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Abstract

Silicon is an important material due to its role as the fundamental component in integrated circuits, microelectronics, and in lithium ion batteries. Continuous efforts have been focused on investigating the nanostructures of silicon for unveiling the properties and miniaturizing the microelectronics devices, including the goal of integrating the electronics and photonics on the same Si chip. The possibility of coating carbon nanotube junctions with single-walled, square-lattice silica nanotubes is considered. We describe the topology of junctions of such tubes, based on the generalized Euler formula for a tetragonal lattice with trigonal, pentagonal, and hexagonal topological defects. The types of silica nanotubes best fitting the considered carbon nanotube types are determined using a classical inter atomic force field. The influence of the silica coating on the electronic properties of the carbon nanotube is evaluated using semi-empirical Paramagnetic Model 3PM3 method calculations. It is found that the electronic properties of the carbon nanotube near the Fermi level are not changed considerably. Finally, we propose some examples of SiO₂-C composite T-, Y-, and X-junction structures.

Keywords: composite structure, C₆₀, multiwalled carbon nanotubes, silica nanotube



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Manuscript ID : ISSN23197501-V2I2M2-0502013

1. Introduction:

Silicon is an important material due to its role as the fundamental component in integrated circuits, microelectronics, and in lithium ion batteries. Continuous efforts have been focused on investigating the nanostructures of silicon for unveiling the properties and miniaturizing the microelectronics devices, including the goal of integrating the electronics and photonics on the same Si chip.

Among various carbon nanotube structures, three-terminal junctions proposed in 1992^{1,2} are potentially important not only for electronics but also for mechanical devices.³ Lately, considerable interest has been given to the preparation of carbon nanotube junctions, which are envisaged as components of nano devices, most likely functioning as a transistor or a rectification element,⁴⁻⁶ or as units in logic networks.⁷ Single-walled carbon nanotube Y-junctions grown by thermal decomposition of C₆₀ fullerene in the presence of transition metals were detected using scanning tunneling microscopy in 2000.⁸ Lately,⁹ transmission electron microscopy and Raman spectroscopy confirmed the formation of single-walled Y-junction structures with 2–5 nm tube diameters grown by chemical vapor deposition using Mo-doped Fenano particles supported on alumina substrate.

2. Silica-Coated Carbon Nanotube Structures:

It is well known that the influence of the environment on the properties of carbon nanotubes is very strong.¹⁰ For example, nanotubes can become chemically functionalized, or their electronic properties may be altered by charge doping or by pH variations in solution, both of which may be undesirable. That is why the protection of carbon nano tubes against the environment is an important problem. Recently, a single-



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walled carbon nanotube coating with thick-walled tens of nanometers amorphous silica nanotubes has been successfully implemented¹¹. However, it is desirable that the protective coatings were thin enough, so that they do not dramatically increase the size of the system or hamper its flexibility. Density functional calculations for a metallic carbon nanotube on the silicon (001) surface¹² show that Si-C bonds are formed between the substrate and the nanotube. These *sp*³-like bonds form an electronic channel along the nanotube substrate contact. Such an interaction enhances the metallic properties of the carbon nanotube, increasing the density of states around Fermi energy, and may be beneficial for devices based on metallic carbon nanotubes. On the other hand, this could be highly undesirable for devices with semi-conducting nanotubes, as it might completely change their electronic properties from semiconductor to metallic in an uncontrollable manner. In these circumstances, a carbon nanotube coating with inert silica tubes would correct this problem. In the recent work,¹³ current-voltage characteristics were measured for a single-walled carbon nanotube junction. The rectification behavior of the junction was controllably changed from along the stem nanotube to across the junction. The behavior was switched by applying voltage either to the branch nanotube or to the back-gate electrode. The authors used a doped silicon substrate as the back-gate electrode. It was covered by a 500 nm silica layer to insulate the nanotube, which is common practice in microelectronics. For making molecular electronic devices, this oxide layer should be as thin as possible. Yet electron energy-loss spectroscopy measurements¹⁴ show that there is a fundamental limit of about 0.7 nm four Si atoms across to the thickness of the silica layer in such devices associated with the spillover of silicon conduction band wave functions into the oxide layer and the formation of interfacial states. The use of coated carbon nanotubes and nanotube junctions might eliminate this problem, as they could be



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Manuscript ID : ISSN23197501-V2I2M2-0502013

placed directly on the electrode without the need for an insulating oxide layer, because the physical separation of the insulator silica nanotube wall from the electrode would prohibit the overlap of their wave functions.

Another interesting application of silica-coated carbon nanotube structures lies in the field of composite materials. It has been recently demonstrated using the nano indentation technique¹⁵, that silica-coated multiwalled carbon nanotubes can reinforce poly(methyl methacrylate) (PMMA) composites, giving hardness and Young's modulus two and three times higher than those of polymer matrix, respectively. The addition of uncoated carbon nanotubes, in contrast, did not affect the nano mechanical properties of the composites.

3. Use of Silica and silica-coated nanoparticles:

Silica and silica-coated nanoparticles are also widely used in biology. For example, dye molecules encapsulated in silica coatings show great promise in the field of fluorescent microscopy for a review. While dye molecules in solution tend to react with the solvent or with other molecules when excited by light, and consequently, to bleach out, no such bleaching is observed when dye complexes are coated by silica shells¹¹. This clearly indicates the protective effect of the latter. Moreover, silica nanoparticles exhibit little or no cytotoxicity at all, which allows their use for *in vivo* techniques. These facts demonstrate the importance of silica as a coating material for bio nanotechnology as well. Here we describe some possible multi-terminal SiO₂ nanotube structures covering multi-terminal carbon single-walled structures. We use a model of an achiral square-lattice, single-walled silica nanotube composed of parallel (-Si-O-)m rings linked by -O- bridges, i.e., of type (m,0)¹³. All the carbon nanotubes considered belong to zigzag types (n,0). While same-diameter carbon nanotubes of different



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types may have totally different electronic properties, only the diameters of tubes are important for the present study.

4. Topological Defects In Single-Walled Nanotube Junctions

Although pristine carbon single-walled nanotubes only contain carbon atoms in a hexagonal arrangement, the formation of a multi-terminal nanotube junction requires the presence of topological defects in the form of pentagons, heptagons, and octagons. This is essential for maintaining the sp_2 configuration for all carbon atoms in order to maximize the stability of the system. The number and kind of topological defects in single-walled nanotube junctions obey the generalization of well-known Euler's formula for polyhedra,

$$F + V = E + 2 - 2G \quad \dots\dots\dots (1)$$

Where F is the number of faces, V is the number of vertices, E is the number of edges, and G is the genus of the polyhedron (0 for a fullerene, 1 for a nanotube, 2 for a Y- or a T-junction, 3 for an X-junction¹². The formula (1) can be easily transformed into a modified equation,⁶

$$N_7 + 2 N_8 - N_5 = 6 (N - 2), \quad \dots\dots\dots (2)$$

Where N_5, N_7 , and N_8 are the numbers of pentagons, heptagons, and octagons, and N is the number of branches (open nanotube ends) in the N terminal junction. This formula can be extended to include even larger polygons.

In square-lattice silica nanotubes, there are two edges and one face per vertex instead of 1.5 edges and 0.5 faces per vertex in sp_2 carbon nanotubes. This leads to a different



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relation between the numbers of various polygons in silica nanotube junctions, which can also be extended to polygons of any size,

$$N_5 + 2N_6 - N_3 = 4(N - 2) \dots\dots\dots(3)$$

In this case, the positive surface curvature is formed by triangles, and the negative curvature results from pentagons, hexagons, and larger polygons. Planar T- and Y-junctions contain either two hexagons or four pentagons (configurations with one hexagon and two pentagons or with larger polyhedra would not be planar, and the inclusion of triangles in the junction would cause additional geometric strain). Planar symmetric X-junctions can be built using two octagons, four hexagons, or eight pentagons.

Equation(3) applies to, in principle, all nanostructures based on the square-lattice structural motif. Examples of such tubular structures include nanotubes of aluminium,¹³ titanium,¹⁴ and magnesium oxide¹⁰.

TABLE I. Types of silica nanotubes best corresponding to various carbon nanotube types.

| Carbon nano tube | Silica nanotube | Composite structure | | | | |
|------------------|-----------------|----------------------------|----------------|------------------------------|------------------------------------|---------------------------------|
| Type (N Atom) | Diameter (Å) | Type {Natoms} | Inner diameter | Difference of tube radii (Å) | Tube interaction energy (kcal/mol) | Distance between tube walls (Å) |
| (6,0) {144} | 4.71 | (12,0){216} (13,0){234} | 10.32 11.19 | 2.81 3.24 | -140.1 -148.3 | 2.87 3.22 |
| (7,0) {168} | 5.45 | (13,0){234} 14,0){252} | 11.19 12.06 | 2.87 3.31 | -158.4 -157.4 | 2.92 3.27 |
| (8,0) {192} | 6.21 | (14,0){252} (15,0){270} | 12.06 12.92 | 2.93 3.36 | -180.1 -169.8 | 2.97 3.31 |



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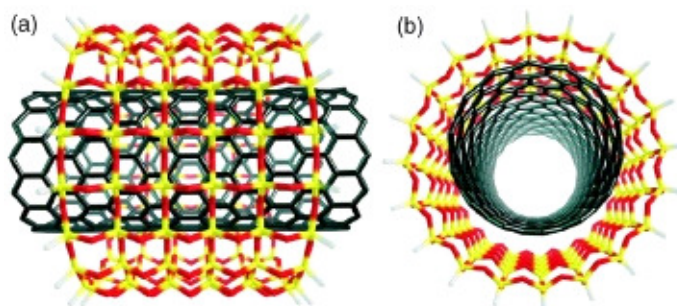
Manuscript ID : ISSN23197501-V2I2M2-0502013

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|-----------------|-------|----------------------------|----------------|--------------|--------|--------------|
| (12,0) {288} | 9.26 | (18,0){324} {324} | 15.52 16.38 | 3.13 2.80 | -239.2 | 3.11 2.92 |
| (14,0) {336} | 10.79 | (19,0){342} (20,0){360} | 17.24 | 3.23 | -261.0 | 3.17 |

*The composite system represents roughly 14.7 Å contact length of nanotube

To build geometrical models of silica-coated nanotube junctions, we had to establish the optimal relation between inner carbon nanotube and outer silica tube diameters. We used a classical inter atomic potential (the molecular mechanics (MM+) force field¹⁴) geometry optimizations because of the large size (the order of thousand atoms) of the final junction structures, and because adequate quantum mechanical modeling of van der Waals interactions, which are crucial in these structures, is unavailable for such large structures. The molecular mechanics methods have been successfully used in numerous studies of both carbon^{2,7} and silica^{8,2} nanosystems.

The best-fitting pairs of nanotubes were determined as the ones having strongest van der Waals interaction per fixed length. The calculations were carried out for double-walled composite clusters of equal length like the one shown in Fig.1 (terminal hydrogen atoms are not shown).





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FIG. 1.(Color online)The cluster structure that was used to determine the types of silica nanotubes best fitting various carbon nanotubes. The structure shown is a(14, 0) carbon nanotube inside a (20, 0) silica nanotube,(a) side view,(b) axial view.

Carbon nano-tube segments consisted of six nanotube unit cells, which corresponded to $24n$ atoms in the $(n,0)$ type cluster. Silica clusters also consisted of six nanotube unit cells, the number of atoms amounting to $18m$ ($6m$ SiO_2 molecules) for the $(m,0)$ type, neglecting the terminal atoms (this corresponds to roughly 14.7 \AA contact length). We optimized the composite structures and calculated the tube interaction energies, i.e., the differences between the energies of the composite structures and the energies of the corresponding single-walled tubes. The interaction energies as functions of the difference of tube radii are plotted in Fig.2. As the nuclei in silica tubes lie on three cylindrical surfaces (inner and outer oxygen cylinders and a silicon cylinder between them), we used the inner cylinder radius, i.e., the distance from the tube axis to the nearest oxygen nucleus. As seen from Fig.2, the best fitting pairs of carbon and silica tubes lie in the $2.8\text{--}3.4 \text{ \AA}$ range of the tube radius difference with the optimal value of about 3.0 \AA . For larger values from this interval, the carbon nanotube is somewhat stretched and the silica tube is compressed towards the axis of the structure, while for lower values the carbon tube is compressed and the silica tube is stretched. The tube radii variation in these distortions reaches nearly 0.01 \AA for carbon tubes and nearly 0.05 \AA for silica tubes. The nanotube interaction energies and geometrical parameters of isolated tubes and composite structures are presented in Table1. We considered zigzag carbon nanotubes of types $(6, 0)$, $(7, 0)$, $(8, 0)$, $(12, 0)$, and $(14, 0)$. For each carbon nanotube type, except $(12, 0)$, two types of silica



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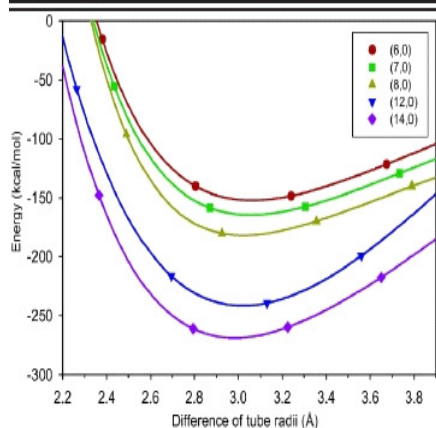


FIG. 2. The energies of interaction between carbon and silica nanotubes with respect to the difference of their radii

nanotubes were found to give interaction energies equal within a 10 kcal/mol tolerance. These belong to the higher and lower regions of the 2.8–3.4 Å interval of the tube radii difference, and we consider them to be equally suitable for corresponding carbon nanotube types. When constructing final junction structures for these carbon tube types, we chose silica nanotubes with even n among the possible silica nanotube types, as they match the symmetry of the final junctions. For the (12, 0) carbon nanotube, the only suitable (18, 0) silica tube lies in the middle of this interval (3.1 Å), which is very close to the optimal value of approximately 3.0 Å.

(Note also how the interaction of the tubes becomes stronger with increasing diameters. This is because we calculated these values for systems of fixed length, and the energy of interaction in such systems is roughly proportional to the surface area of the systems, which scales proportionally to both length and diameter.)



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When tube radii differ by less than 2.8 Å, the silica coating is too tight: the overlap repulsion leads to noticeably greater distortions of tubes, and to considerably higher interaction energies; the latter eventually reach values above zero, which means that coating the given carbon nanotube with silica nanotube of particular type is energetically unfavorable. When the tube radius difference exceeds 3.4–3.5 Å, the coating is too loose, and it is energetically favorable for the carbon tube to be positioned off the silica tube axis, causing ovoid shape of both nanotubes. The composite system then loses its axial symmetry, and the interaction energy becomes higher for greater radius differences due to the geometric strain of the silica nanotube (in contrast to carbon nanotubes, $(n,0)$ silica nanotube bond energies increase with diameter starting from a certain value^{9,10})

To examine the influence of silica coating on the electronic properties of carbon nanotubes, we have calculated local density of electronic states for the model cluster representing a (6, 0) carbon tube inside a (12, 0) silica tube. We employed the Parametric Model (PM3) semi empirical quantum chemistry method, which has successfully been used for calculations on large-scale carbon nanostructures^{10–12} as well as silica nanosystems^{3,14}. The local densities of states were calculated for the inner carbon nanotube fragment, the outer silica nanotube fragment, and the composite structure. They are presented in Fig.3, where the densities of states are shown by solid lines for carbon and silica clusters and by a dashed line for the composite structure. The comparison of the results shows that density of states of the composite structure is a superposition of densities corresponding to constituent tubes with no significant mutual influence of carbon and silica nanotubes on the electronic properties of each other. This indicates that the electronic properties of inner carbon tube, at least in the region around the Fermi energy (which is the most important for such fields as molecular electronics), are not altered significantly. The orbital localization analysis shows that the highest



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occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) are localized on the inner carbon tube. The chemical stability of the composite structure is therefore enhanced and its electronic properties are protected from the influence of the environment, compared to an isolated carbon nanotube. Another group employing density-functional theory to investigate the interaction of single-walled carbon nanotubes with walls of microporous silica channels reached similar conclusions in their study.¹⁵ They also found that the compression of carbon nanotubes due to extremely tight coatings could cause a significant change of their electronic properties, e.g., opening of band gap in metallic tubes that converted them to semiconductor.

Nanotube junctions of the simplest type^{11,12} (two terminal) are termed intra molecular or L-junctions. An example of a composite L-junction is shown in Fig.3.

The types of carbon and silica nanotube junctions are $(6, 0)-(8, 0)P1 H1$ and $(12, 0)-(14, 0) T1P1$, correspondingly. The number of branches N of an L-junction is two, and it follows from Eqs.(2) and (3) that both carbon and silica junctions must contain small polygons (a pentagon in the carbon tube and a three-member ring in the silica tube; the topological defects are highlighted in Fig.3), apart from the larger ones. L-junctions are important because they allow single-molecule metallic-to-semiconductor and $p-n$ transitions such as the $(6, 0)-(8,0)$ carbon nanotube junction presented in Fig.3, where the $(6, 0)$ tube is metallic, and the $(8, 0)$ tube is semiconductor, or tubes with varying diameter as well as permanent knee-like bends, to be constructed.

5. Silica Nanotube Junctions Corresponding To The Selected Carbon Nanotube Multi-Terminal Junctions:



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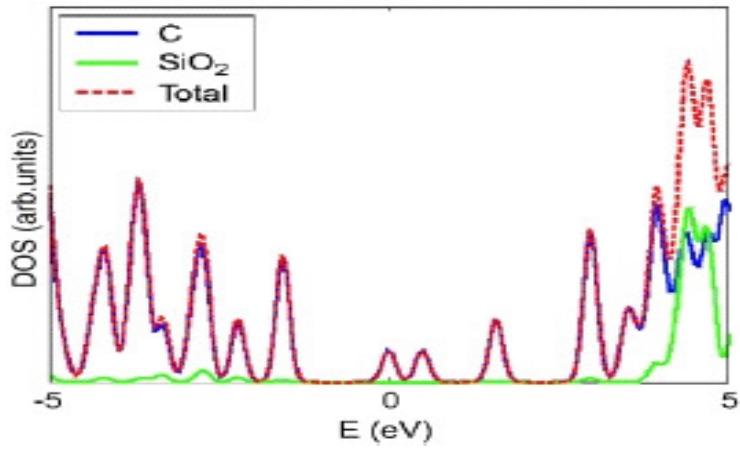
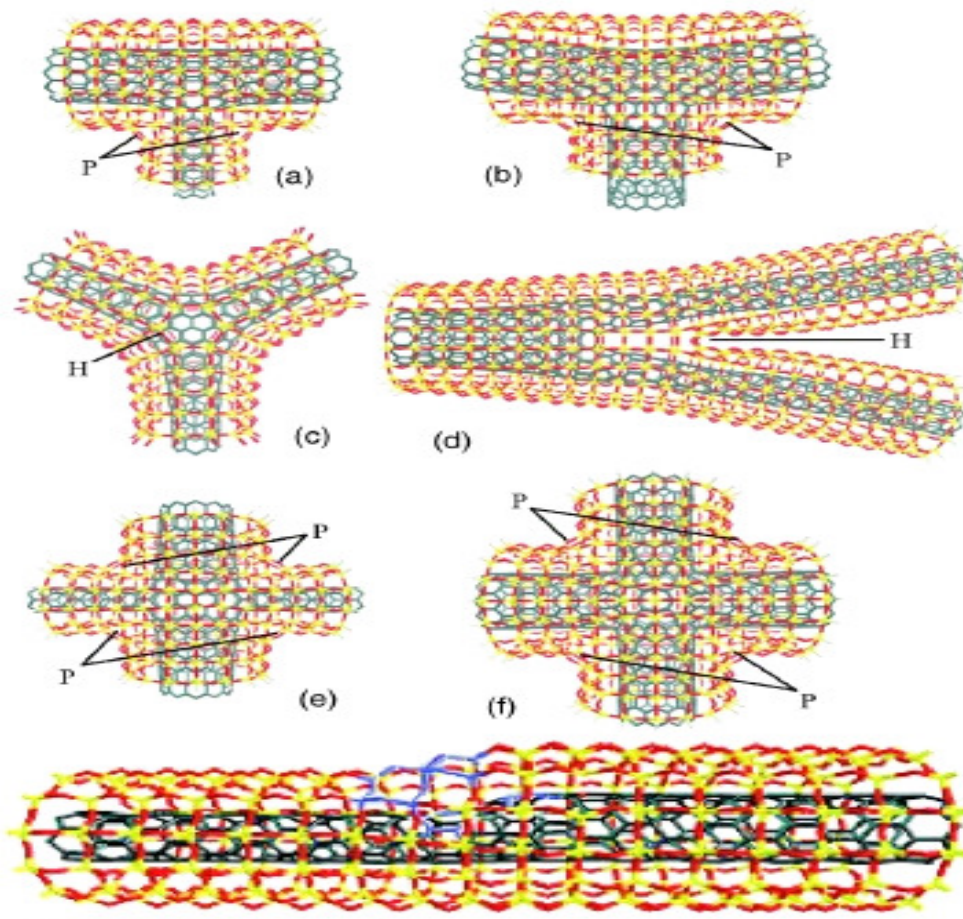
Manuscript ID : ISSN23197501-V2I2M2-0502013

Next, we have constructed silica nanotube junctions corresponding to the selected carbon nanotube multi-terminal junctions. We examined following carbon nanotube junctions: $(6, 0)-(12, 0)H2 O2$ and $(12, 0)-(12, 0)H2 O2$ T-junctions, $(8, 0)-(8, 0)-(8, 0)H6$ and $(14, 0)-(7, 0)-(7, 0)H6$ Y-junctions, and $(6, 0)-(12, 0)H4 O4$ and $(12, 0)-(12, 0)O6$ X-junctions (the numbers after H and O indicate the count of heptagons and octagons, respectively; in the case of silica junctions, the letters P and H stand for pentagons and hexagons).



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Fig.3. Multi-terminal carbon nanotube junctions coated with silica nanotube junctions; carbon nanotube junction coated with a (12, 0)–(14, 0) silica nanotube junction; The PM3 density of states for a (6, 0) carbon nanotube inside a (10, 0) silica nanotube: blue/dark line, isolated carbon cluster; green/light line, isolated silica cluster; and red/dashed line, the composite system. The Fermi level energy is taken as zero.

As the number of branches of a multi-terminal junction is three or greater, structures need not contain small polygons that cause additional geometrical strain. For each carbon nanotube junction, several coating structures with different arrangements of topological defects were examined, and from these, the ones with optimal topological defect arrangement and lowest energies were selected. The final composite junction structures are shown in Fig.3. Letters *P* and *H* indicate the topological defects in silica nanotubes.

Figures 3(a) and 3(b) show T-junction structures. The first is (6, 0)–(12, 0)*H* 2O₂ carbon nanotube junction coated with (12, 0)–(18, 0)*P*₄ silica nanotube junction. The second is (12, 0)–(12, 0)*H* 2O₂ carbon nanotube T-junction coated with (18, 0)–(18, 0)*P*₄ silica nanotube junction. The pentagons in both silica structures are located in the angles between stem and branch nanotubes. Note how the branch carbon tube thickness in the second structure causes a stem nanotube bend in the junction region, which is not the case with the silica nanotube junction (this can be attributed to the different symmetries of the corresponding layered phases—hexagonal graphite and square silica sheet¹³), although its wall is bulged in near the bend. This fact would require some additional bending of tubes in regions some distance away from the junction, but it would not change the topology and the structure of the junction itself, which is of primary interest here. Figures 3(c) and 3(d) show Y-junction structures. The first is a three-branch (8, 0)–(8, 0)–(8, 0)*H* 6 carbon nanotube junction coated with (14, 0)–(14, 0)–(14,



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0) H_2 silica nano-tube junction. The branches are geometrically equivalent and form a structure with a threefold symmetry axis. The two hexagons are located in the central part on the opposite sides of the silica nanotube junction. The second is a one stem-two branches $(14, 0)-(7, 0)-(7, 0)H_6$ coated by $(20, 0)-(14, 0)-(14, 0)H_2$ silica nanotube Y-junction. The hexagons are located in the inner part of the sharp angle formed by the branches. The equilibrium angles between the inner and outer junction branches differ somewhat, which results in geometrical strain: notice a slight bending of the carbon nanotube junction branches in Fig.3(d) respectively.

Finally, Figs.3(e) and 3(f) present X-junction structures. They are related to T-junctions in the sense that they can be thought of as double T-junctions with oppositely directed branches. The first is $(6, 0)-(12, 0)H_4O_4$ carbon nanotube junction coated with $(12, 0)-(18, 0)P_8$ silica nanotube junction, and the second is $(12, 0)-(12, 0)H_4O_4$ carbon nanotube X-junction coated with $(18, 0)-(18, 0)P_8$ silica nanotube junction. The pentagons of silica junctions are located similarly to those of T-junctions in Figs.3(a) and 3(b). We should note that while the carbon nanotube junction in Fig.3(f) has a fourfold symmetry axis perpendicular to the junction plane, the three-layer geometry of silica nanotubes decreases the symmetry of the coating junction, which has only two fold symmetry axes.

In addition to what was mentioned above, some geometrical strain is present in all structures, which results from the fact that carbon nanotubes tend to flatten towards the junction plane in the region near the junction, while in silica junctions, nanotubes tend to flatten in the perpendicular direction. These two tendencies confront each other, which raises the composite structure energy. This might possibly be overcome by building the coating junctions from tubes of diameters larger than those most suitable for coating single carbon nanotubes. The branches of the silica



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junctions could then be adjusted to fit the corresponding carbon nanotubes by introducing triangle-pentagon defect pairs forming L-junctions such as the one shown in Fig.3 at some distance from the multi-terminal junction. Yet despite all these sources of geometric strain, the energy of nanotube interaction is negative even in simplest structures considered here, which means that van der Waals attraction of tubes is stronger than overlap repulsion and geometric strain in the deformed regions (for example, the tube interaction energy in the X-junction shown in Fig.3(f) is -620.5 kcal/mol, which compares well with those of tube interaction energies as shown in Table; the number of atoms in the junction is roughly 2.9 times that of the double-wall nanotube segment used to calculate the energies in the table).

6. Conclusion:

In summary, we have considered the possibility of coating carbon nanotubes and multi-terminal nanotube junctions with thin-walled silica nanotubes, and examined some hypothetical structures of double-walled composite carbon-silica multi-terminal nanotube junctions. We have established the relation between the numbers of topological defects (here, three-, five-, and six-member rings) basing on the general-ized Euler rule for square-lattice nanotubes. Electronic structure calculations show that silica coating does not considerably change the electronic properties of the carbon nanotube around the Fermi level and that such an arrangement of the tubes does not decrease the chemical stability of the silica tube. This means that the proposed composite structures offer protection of carbon nanotubes from the influence of the environment. We should note that, although we have found that the electronic properties of carbon nanotubes remain unaltered in composite carbon-inside-silica nanotube structures, the situation might be not so perfect for nanotube junctions. This is because the latter lack the symmetry of the tubes, and pos-



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Manuscript ID : ISSN23197501-V2I2M2-0502013

sess topological defects and strained sites. Due to these facts, the I - V curves of the coated junctions may become some-what altered. Both the influence of strained geometry on the properties of carbon nanotube junctions and the direct impact of the electron density of the silica nanotubes on them remain undetermined.

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