Quantum Modeling of Doped Carbon Nanotubes for High Ampacity Conductor Design

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ABSTRACT

Carbon based electrical conductors have attracted considerable attention as potential replacements for copper, since they may offer improved specific conductance or higher ampacity. A series of calculations has been performed to estimate the ballistic conductance properties of iodine doped carbon nanotubes and iodine doped carbon nanotube junctions. The results suggest that doped carbon nanotube conductors are viable research and development candidates for electrical conductors in ship and aircraft applications, where mass specific conductivity is of central interest.

Keywords: nanotubes, conductance, doping

1 INTRODUCTION

The widespread use of copper in power and data cabling for aircraft, ships, and ground vehicles imposes significant mass penalties and can limit system performance, due to thermal constraints. Carbon based electrical conductors have attracted considerable attention, as potential replacements for copper, since they may offer improved specific conductance or higher ampacity. Carbon nanotube [1, 2] based conductors have been studied both experimentally and computationally, as a promising new cable technology. Their relatively low conductivity, as compared to copper, has encouraged the consideration of doped nanotubes or CNT-copper nanocomposites [3] as energy-efficient replacements in mass sensitive applications. Tables 1 [3, 4, 5, 6] and 2 [3, 4] compare published data on the electrical conductivity and the mass specific electrical conductivity of several doped CNT or CNT-based composites with the corresponding properties of copper.

Although experimental research on the development of CNT based electrical conductors has been productive, the difficulty of the conductor design problem motivates complimentary computational research. This paper describes a series of calculations performed to estimate the ballistic conductance properties of iodine doped carbon nanotubes, under consideration as a potential replacement for copper in ship and aircraft applications. Such applications are typically mass constrained, as opposed to volume constrained, hence specific conductivity (as opposed to conductivity) is of most interest. Since the fabrication of macroscale conductors will presumably require the systematic integration of nanotube bundles, the conductance properties of doped and undoped nanotube junctions are also investigated.

Table 1: Electrical conductivity

Material	$\sigma\ (S/cm)$
Cu	5.80×10^5
Cu-CNT composite	3.50×10^5
undoped CNT fiber	2.90×10^4
iodine doped CNT fiber	5.00×10^4
acid doped CNT fiber	3.89×10^4

Table 2: Mass specific electrical conductivity

Material	$\sigma/\rho\ (S - cm^2/g)$
Cu	6.47×10^4
Cu-CNT composite	8.15×10^4
iodine doped CNT fiber	3.57×10^4

2 CONDUCTANCE ANALYSIS

The conductance properties of doped and undoped nanotubes and nanotube junctions were computed using the open source codes SIESTA [7] and TransSIESTA. First Siesta is used to determine the electronic structure, then TransSiesta is used to compute the modeled system's electrical conductance. The electrical conductance (G) is calculated using the Landauer formula [8]

$$G = 2\frac{e^2}{h}\int\left(-\frac{\partial f\left(E\right)}{\partial E}\right)T(E)\,dE \tag{1}$$

where 'e' is the charge on an electron, 'h' is Planck's constant, and 'T' is the transmission

$$T(E) = \mathrm{Tr}\left[t\left(E\right)^{\dagger}t\left(E\right)\right] \tag{2}$$

with 'f' the Fermi-Dirac distribution function and 't' a matrix of transmission coefficients for waves propagating along the conductor. The latter are functions of

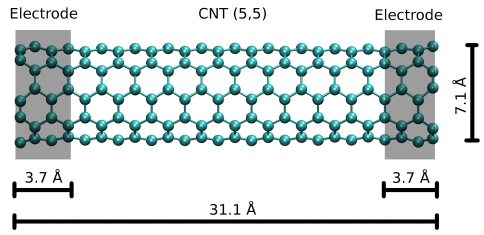

Figure 1: Nanotube model

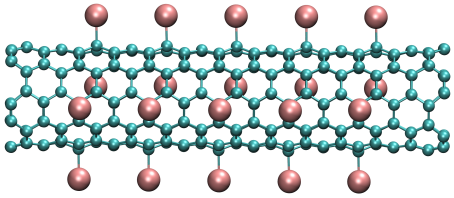

Figure 2: Iodine doped nanotube model

the wave energies E. To obtain the conductance at the ground state, let

$$-\frac{\partial f\,(E)}{\partial E} = \delta(E - E_f) \tag{3}$$

where E_f is the Fermi energy and 'δ' is the Dirac delta function, so that

$$G = G_0\,T(E_f), \quad G_0 = 2\frac{e^2}{h} \tag{4}$$

where G_0 is the standard quantum conductance unit. Note that for an ideal metallic carbon nanotube, $T(E_f) = 2$ and $G = 2G_0$.

The analysis presented here assumes ballistic transport: the mean free path (L_m) of an electron is assumed to be greater than length of the conductor (L). For single-walled CNT's at room temperature, L_m is estimated to fall within the range 10-4,000 nm [9, 10]. In the analyses which follow, $L = 4$ nm, the nanotube diameters (D) are 0.7 nm, and the maximum estimated upper limit for ballistic conduction is associated with an L/D ratio of approximately 5,000. Note that the analysis assumes zero temperature conditions, in the molecular sense, since the calculations are performed for fixed nuclei (Born-Oppenheimer approximation [11]).

3 MODEL CONFIGURATIONS

The analysis considered six model configurations:

- Single CNT's, doped and undoped
- Junctions of two CNT's, aligned and misaligned, doped and undoped

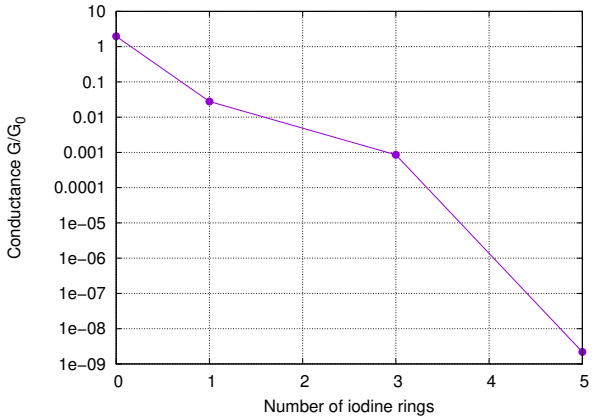

Figure 3: Nanotube conductance versus applied dopant

The term 'aligned' refers to the positions of neighboring atoms in disctinct nanotubes, and will be defined in a later section of the paper. All calculations were performed for metallic (5,5) single-walled carbon nanotubes, and the modeled dopant was iodine [12].

4 NANOTUBE CONDUCTANCE

The conductance was first calculated for isolated nanotubes (Figure 1), at two different lengths. In this case the conduction calculations were performed after relaxation of the system to an equilibrium state (all forces relaxed to within 0.04 eV per Å). Consistent with published experimental data, CNT's are ballistic conductors [13] and the nanotube conductance takes on the value $G = 2G_0$ ($T = 2$) at the Fermi Energy [14].

Next the conductance was computed for isolated nanotubes with various numbers of iodine atoms 'bonded' to the CNT sidewall (Figure 2). Conduction calculations were performed after relaxation of the system to an equilibrium state (all forces relaxed to within 0.04 eV per Å). The axial separation distance of the iodine atoms was 5.1 Å, and the length of the carbon-iodine bond was 2.2 Å. Note that published experimental research [15] has classified the iodine bonding as covalent, and that previous computational work [16] modeling various covalently bonded addends (e.g. F) to CNT's indicates that such doping reduces CNT conductance. Figure 3 shows the computed conductance, as a function of the number of iodine 'rings' bonded to the CNT sidewall. Consistent with previous research on covalent doping, iodine doping (in this case, in a ring configuration) sharply reduces CNT conductance.

5 NANOTUBE JUNCTION CONDUCTANCE

Next the conductance was computed for both doped and undoped nanotube junctions, arranged as indicated

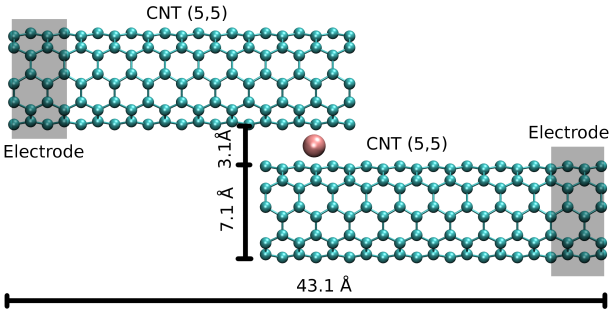

Figure 4: Nanotube junction model

in Figure 4. In this case, a geometric configuration was assumed and the conduction calculations were performed without relaxation of the system to an equilibrium state. The junction overlap was varied in increments of 9.9 Å, as shown in Figure 5, while the axial separation distance of the iodine atoms was fixed at 4.9 Å. Undoped configurations were obtained by simply removing the dopant atoms.

Two different junction alignments were analyzed. In the aligned case, the dopant atom and the adjacent carbon atoms formed a 'sandwich' substructure [17]. In the misaligned case, one nanotube was shifted axially. The two modeled configurations are depicted in Figure 6. Previous computational work has suggested that optimal doping treatments [17] and optimal overlap configurations [18] offer the possibility of constructing multijunction networks which exhibit the excellent conductance properties of single nanotubes. Figure 7 plots the results of the current calculations, indicating that a relative maximum in the conductance was observed only for the doped junction configurations (in the undoped cases, conductance increases monotonically with overlap). Note that the junction conductance is sensitive to alignment effects, in particular for doped junctions. At the best modeled combination of doping, alignment, and overlap, junction conductance is approximately eighty percent of that for a single ideal nanotube.

6 CONCLUSIONS

The following general conclusions are suggested: (1) the experimentally observed benefits of doping appear to be due to effects at the nanotube junctions, and (2) the effects of doping on metallic nanotubes may be negative. More analysis is clearly needed, including refined versions of the models analyzed to date. Future research will investigate the effects of other dopants, such as iodine monochloride [19], cobalt oxide [20], and acid treatments [5], on electrical conductivity. More complex molecular scale structures [21, 22, 23] constructed by combining CNT with boron, boron nitride, or graphene are also of interest.

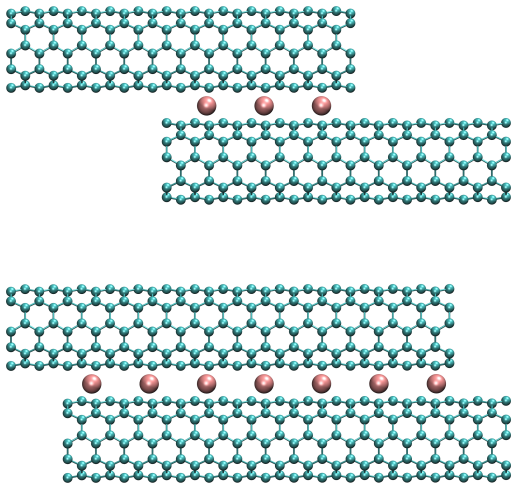

Figure 5: Junction model at two different overlaps

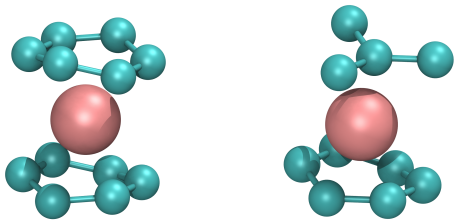

Figure 6: Aligned and misaligned configurations

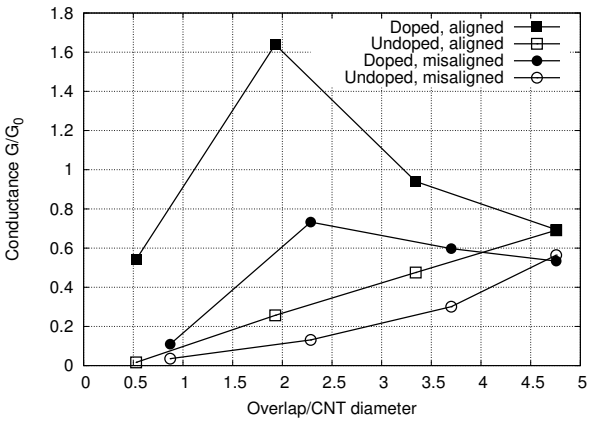

Figure 7: Junction conductance versus overlap

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