

## Dimension, Strength, and Chemical and Thermal Stability of a Single C–C Bond in Carbon Nanotubes

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For single-walled carbon nanotubes (SWCNTs), given the known product of the Young's modulus  $Y$  and the wall (bond) thickness  $t$  ( $Yt \cong 0.3685$  TPa·nm) and the known temperature of tip-end melting ( $T_m = 1593$  K), as well as their functional dependence on atomic coordination and bonding energy, the dimension and strength of a C–C bond in a SWCNT have been determined. The uniqueness of the solution reveals that the C–C bond (or the wall of the SWCNT) is  $\sim 0.142$  nm thick and  $\sim 0.125$  nm long (an 18.5% contraction of diamond bond length, 0.154 nm), with a 68% increase in binding energy, with respect to the bulk graphite values. Therefore, the measured  $Yt$  and  $T_m$  values essentially represent the true SWCNT situation.

Since the discovery of carbon nanotubes (CNTs),<sup>1</sup> there has been ever-increasing interest in this new form of carbon, not only because of the novel structures and the properties but also because of the potentially important applications, such as use in atomic-force microscopy (AFM) microscope tips,<sup>2</sup> field emitters,<sup>3</sup> electronic devices,<sup>4</sup> hydrogen storage,<sup>5</sup> and chemical sensors.<sup>6</sup> Overwhelming efforts have been exerted, primarily in regard to CNT growth, characterization, and functioning, whereas insight into the origin behind the unusual behavior of the CNT is highly desirable.

The Young's modulus ( $Y$ ) of the CNTs has been a confusing issue for a long time; it has been reported to vary over a range of 0.5–5.5 TPa, compared with that of the bulk graphite ( $Y = 1.02$  TPa). Actually, one can only measure the *product* of the Young's modulus  $Y$  and the wall thickness  $t$  ( $Yt$ ), rather than the individual parameters. This is the reason why the reported  $Y$  values vary with the presumption of the single-wall (C–C bond) thickness ( $t_1$ ).<sup>7–9</sup> If one assumes the equilibrium interlayer spacing of graphite sheets,  $t_1 = 0.34$  nm, to represent the bond thickness, the derived  $Y_1$  value is  $\sim 1.1$  TPa.<sup>10,11</sup> If  $t_1 = 0.066$  nm, which is close to the radius of a free C atom (0.0771–0.0914 nm), the  $Y_1$  value is derived as 5.5 TPa.<sup>9</sup> Although the wall thickness and the scattered  $Y$  values need yet to be certain, the product  $Yt$  surprisingly approaches a constant value ( $0.3685 \pm 0.0055$  TPa·nm). For the single-walled carbon nanotubes (SWCNTs), under the presumed  $t$  value, the  $Y_1$  value varies slightly with the tube diameter and the tube helicity, because of the curvature-induced strain.<sup>12</sup> However, for multiwalled carbon nanotubes (MWCNTs), two typical trends of the change in  $Y$  have been observed: (i)  $Y$  is almost independent of the tube diameters,<sup>10</sup> and (ii)  $Y$  increases as the number of walls are reduced.<sup>13</sup>

In contrast, atoms at the tip end of a SWCNT could melt or coalesce first and then the tube body follows at temperature much lower than the melting point of the bulk graphite ( $T_m =$

3800 K). The coalescent temperature of the CNT increases as the number of walls ( $\lambda$ ) increases. Coalescence of the SWCNTs happens at 1073 K under energetic (1.25 MeV) electron beam irradiation and the coalescence starts at vacancies via a zipperlike mechanism.<sup>14</sup> The STM tip end that is made of CNTs starts to melt at 1593 K under ultrahigh vacuum.<sup>15</sup> Annealing at 1670–1770 K under medium-high vacuum in flowing argon and N<sub>2</sub> atmospheres shows that 60% of the SWCNTs coalesce with their neighbors.<sup>16</sup> Heating under an argon flow in the temperature range of 1873–2273 K results in a progressive destruction of the SWCNT bundle, followed by coalescence of the entire bundle.<sup>17</sup> SWCNTs transform at temperatures of 2473 K or higher to MWCNTs with an external diameter of several nanometers, and the Fe–C impurity bonds can be completely removed at 2523 K.<sup>18</sup> Recently, it has been found that an ordinary camera flash<sup>19</sup> could burn the SWCNT under ambient conditions, showing the higher chemical reactivity for oxidation of the SWCNT. These results show the consistently lower thermal and chemical stability of the CNT. In particular, the tip end of the SWCNTs melts first and then is followed by the wall of the SWCNT. MWCNTs are more thermally stable than the SWCNTs, and the stability of the MWCNTs increases as the  $\lambda$  values increase.<sup>8,20</sup>

The mechanisms behind the  $Y$  enhancement and the  $T_m$  suppression of the CNTs and the certainty in the wall thickness of the SWCNTs are still puzzling, although the atoms that surround the defects or are located at the tip ends or at the surface are expected to have unusual, yet unclear, roles in dominating the mechanical and thermal behavior of the CNTs.<sup>14,21</sup> Here, we show that, with the known identities and their functional dependence on the atomic coordination, the bond length, and the bond strength, the dimension and strength of a single C–C bond can be uniquely determined by solving a group of simple equations, to advance a consistent understanding of the mechanical strength and of the chemical and thermal stability of the CNTs.

The striking difference between the bulk graphite and a SWCNT is that the effective atomic coordination number (CN, or  $z_i$ , where  $i = 1$  for a SWCNT) of a C atom reduces from a

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CN of 12 to a CN of 3 upon SWCNT formation. For an atom that is surrounding a defect or is located at the tip end of an open edge, the CN is 2. The bulk effective atomic CN is always 12, regardless of the nature of the bond or the crystal configuration (comparing the covalent bond length in graphite (0.142 nm) with that in diamond (0.154 nm), the effective atomic CN in graphite is found to be  $\sim 5.5$ , according to current iteration). According to Goldschmidt, Pauling, and Feibelman,<sup>22</sup> the reduction in CN causes the remaining bonds of the lower-coordinated atom to contract spontaneously. The spontaneous bond contraction is associated with a magnitude increase of the bond energy. This bond order–length–strength (BOLS) correlation<sup>23</sup> can be formulated as

$$\begin{cases} c_i(z_i) = \frac{d_i}{d_0} = \frac{2}{1 + \exp[(12 - z_i)/(8z_i)]} \\ E_i = c_i(z_i)^{-m} E_0 \end{cases} \quad (1)$$

$E_0$  and  $d_0$  are the corresponding bulk values of bond energy and bond length, respectively, and  $m$  is an adjustable parameter. For pure metals of gold, copper, silver, and tin,  $m \approx 1$ ;<sup>24</sup> for covalent silicon and ionic CdS and CdSe,  $m \approx 4$ .<sup>23</sup> The BOLS correlation suggests that the reduced CN ( $z_i$ ), the shortened bond-length ( $d_i$ ), and the enhanced bond-energy ( $E_i$ ) contribute not only to the cohesive energy ( $E_{\text{coh}} = z_i E_i$ ) of a single atom of concern but also to the binding energy density ( $E_b = n_i E_i$ ) in the relaxed region. The variable  $n_i$  is the bond number per unit volume.  $E_{\text{coh}}$  defines the thermal stability,<sup>25</sup> and  $E_b$  is related to the mechanical strength.<sup>26</sup> The functional dependence of  $Y_i$  and  $T_{m,i}$  on the atomic coordination and bond energy at the equilibrium atomic separation are given as<sup>25,26</sup>

$$\begin{cases} Y_i = v \frac{\partial P}{\partial v} \Big|_{r=d_i} = v \frac{\partial^2 u(r)}{\partial v^2} \Big|_{r=d_i} \propto n_i E_i = d_i^{-2} E_i \\ T_{m,i} \propto E_{\text{coh}} = z_i E_i \end{cases} \quad (2)$$

where  $P = -\partial u(r)/\partial v|_{r=d_i}$ , being the first-order differentiation of the interatomic potential at equilibrium distance, is the bond stress;  $v$  is the atomic volume, and  $u(r)$  is the interatomic potential energy. For a SWCNT,  $n_1 \propto d_1^{-2}$  is the bond number per unit area, which is independent of the wall thickness. Obviously, no other argument could change the  $Y$  value unless the bond length were shortened and/or the single bond energy were increased. At the melting point, the bonds of the lower-coordinated atom will be thermally loosened and the atom will coalesce with its neighbors. Therefore, correlation between the measured  $(Yt)_1$  value (0.3685 TPa·nm) and the  $T_{m,1}(z=2)$  value (1593 K) for a SWCNT with the corresponding graphite bulk values ( $T_{m,b} = 3800$  K,  $Y_b = 1.02$  TPa) satisfies the relation

$$\begin{cases} \frac{T_{m,i}(2)}{T_{m,b}} = z_{i,b} c_i(2)^{-m} & (\text{tip end}) \\ \frac{T_{m,i}(3)}{T_{m,i}(2)} = \frac{3}{2} \left( \frac{c_i(3)}{c_i(2)} \right)^{-m} & (\text{wall-tip relation}) \\ \frac{(Yt)_i}{Y_b t_i} = c_i(3)^{-(2+m)} & (\text{CNT-wall}) \end{cases} \quad (3)$$

$z_{i,b}$  is the CN of a C atom at the tip end ( $z=2$ ) or in the tube wall ( $z=3$ ), divided by the ideal bulk value of  $z=12$ . The value of  $c_i(z)$  is given in eq 1. These simple relations give immediate solutions of  $m = 2.5585$ ,  $t_1 = 0.142$  nm, and the tube-wall melting point ( $T_{m,1}(3) = 1605$  K). Furthermore, the activation energy for chemical reaction is also a portion of the atomic cohesive energy. Therefore, the chemical stability of the lower-coordinated atoms is less than the bulk values, which may explain why the CNT could be burnt using an ordinary camera flash under ambient conditions.

The accuracy of the numerical solutions is subject to the input of  $T_{m,1}(2)$  and  $Yt$ . Errors in measurement or structural defects of the CNT may affect the accuracy of the solutions; however, they never determine the nature of the observations. Varying any of the input (given as bold figures in Table 1) leads to solutions that are physically forbidden. For example, replacing the quoted  $Yt$  value with  $Y = 0.8$  TPa (disregarding the thickness  $t_1$ ) gives  $m = -3.2$ . The corresponding  $E_i/E_0$  value ( $E_i/E_0 = c_i(3)^{-m} = 0.52$ ), and the value  $T_{m,1}(3) = z_{1,b} \times c_i(3)^{-m} \times T_{m,b} = 493$  K, are unacceptable, because the  $T_{m,1}(3)$  value is much lower than any reported values. Assuming the SWCNT tip-end melting point to be  $T_m(2) \geq 1620$  K gives  $m \geq 2.605$  and  $T_m(3) \leq 1620$  K, which is not the case of observation: the tip end melts first. If a value of  $m = 4$  is assumed for the covalent or ionic solids,  $T_{m,1}(2) = 2674$  K and  $T_{m,1}(3) = 2153$  K, which are much higher than the measured values. Therefore, the solution with the quoted  $Yt$  and  $T_{m,1}(2)$  values is unique and, hence, the quoted  $Yt$  and  $T_{m,1}(2)$  are essentially true for the SWCNT.

For nanobeams (solid nanorods and MWCNTs), the relative change of a measurable quantity (denoted as  $Q$ ), which is dependent on size and shape, of a nanosystem with dimension  $D$  can be quantized with shell structures as<sup>26</sup>

$$\begin{cases} \frac{\Delta Q(D)}{Q(\infty)} = \sum_{i \leq 3} \gamma_i \frac{\Delta q_i(d_i, z_i, E_i)}{q(d_i, z_i, E_i)} \\ \gamma_i = \frac{(D_{\text{out},i}^2 - D_{\text{in},i}^2)_M + (D_{\text{out},i}^2 - D_{\text{in},i}^2)_m}{D_M^2 - D_m^2} \propto \frac{1}{\lambda} \end{cases} \quad (4)$$

where  $q_i$ , which is the density of quantity  $Q$  on an atomic scale, is functionally dependent on the bond length  $d_i$ , the effective

**TABLE 1: Comparison of the Calculation Results with Various Input Parameters,<sup>a</sup> Proving that the Obtained Solution is Unique and the Quoted Data Represent the True Values**

$Yt$ (TPa·nm) or $Y$ (TPa)	$(Yt)_1 = \mathbf{0.3685}$ , $Y = 2.595$	$Y = \mathbf{0.8}$	$(Yt)_1 = \mathbf{0.3685}$	
tip-end melting point, $T_{m,1}(z=2)$ (K)	<b>1593</b>		2153	$\geq \mathbf{1620}$
tube-wall melting point, $T_{m,1}(z=3)$ (K)	1605	493	2674	$\leq \mathbf{1620}$
$m$	2.5585	<b>2.5585</b>	<b>4</b>	2.6050
bond thickness, $t_1$ (nm)	0.142			
bond length, $d_1$ (nm)	0.125			
bond energy, $E_i/E_0$	1.68	0.53		
remarks	acceptable	forbidden	forbidden	forbidden

<sup>a</sup> Input parameters are shown in bold typeface.

coordination  $z_i$ , and the bond energy  $E_i$ . The difference of  $\Delta q_i$  between the surface region and the bulk initiates the change of the value  $Q$ ; the surface-to-volume ratio,  $\gamma_i$ , of a nanosolid dominates the trends of  $Q$  change. The index value  $i$  is counted from vacuum sides to the center solid of the nanosolid, up to a value of 3, because no CN imperfection is expected for  $i > 3$ . The weighting factor,  $\gamma_i$ , is the volume ratio of the  $i$ th atomic layer to the entire NT with an outer diameter  $D_M$  ( $D_M = 2kd$ ) and an inner hollow diameter  $D_m$  ( $D_m = 2(k - \lambda)d$ , where  $kd$  is the radius of the tube and  $\lambda d$  is the wall thickness for MWCNT.  $D_{out,i}$  and  $D_{in,i}$  correspond to the outer and inner radius of the  $i$ th atomic layer ( $D_{out,i} - D_{in,i} = d_i$ ). The relation  $\gamma_s = \sum_{i \leq 3} \gamma_i$  decreases in an inverse fashion ( $\lambda^{-1}$ ), from unity to being infinitely small, when  $\lambda$  grows from unity to infinity. Therefore, it is not surprising that, for a solid rod or a MWCNT with  $k \leq \lambda$ , the overall  $\Delta Q(D)/Q$  value varies with the inverse radius ( $1/kd$ ) and the  $\Delta Q(D)/Q$  value differs from the corresponding bulk value ( $\Delta Q(D)/Q = 0$ ). For a hollow MWCNT with constant  $\lambda$ , the  $\Delta Q(D)/Q$  should not vary with the diameter of the MWCNT.

These predictions agree with the observed trends in the  $Y$  enhancement<sup>7–13</sup> and  $T_m$  suppression<sup>15–21</sup> of the nanobeams. The direct evidence for the  $\lambda$  dependence is that the  $Y$  value was calculated to vary over a range of 4.7–1.04 TPa when the wall number  $\lambda$  increases from unity to infinity.<sup>13</sup> AFM measurements<sup>10</sup> of the  $D$  dependence of the Young's modulus of SiC nanorods and carbon MWCNTs reveal that the MWCNTs are about twice as stiff as the SiC nanorods and that the strengths of the SiC nanorods are substantially greater than those found for large SiC structures (600 GPa). The Young's modulus is 610 and 660 GPa for SiC rods with diameters of 23.0 and 21.5 nm, respectively. For MWCNTs, the modulus is  $1.28 \pm 0.59$  TPa, with no apparent dependence on the diameter of the nanotubes. The broad range of measured values should be more attributable to the scattered number of walls of the nanotubes than to the error in measurement.

Using a coagulation-based CNT spinning technique, Dalton et al.<sup>27</sup> recently spun surfactant-dispersed SWCNTs from a rotating bath of aqueous poly(vinyl alcohol) to produce nanotube gel fibers that they then converted to solid nanotube composite fibers. The resulting CNT rope (100 m long, 50  $\mu\text{m}$  thick) had a tensile strength of 1.8 GPa and an energy-to-break value of 570 J/g. The fibers, which are suitable for weaving into electronic cloth, are 4 times tougher than spider silk and 17 times tougher than the Kevlar fibers that are used in bulletproof vests. The fibers also have twice the stiffness and strength and have a toughness that is 20 times greater than that of the same weight of steel wire. As artificial muscle, NT fibers are  $\sim 100$  times stronger than natural muscle with the same diameter.<sup>28</sup> This discovery concurs with the BOLS prediction, which indicates that the CN-imperfection-induced bond contraction and the associated bond strength dictate the higher mechanical strength and the lower activation energy of atomic dislocations.<sup>26</sup>

Thus, the known  $Y$  value and the known temperature of tip-end melting for a SWCNT, and their functional correlation with the bonding identities, have enabled the dimension and energy of a single C–C bond in SWCNT to be uniquely quantified, which, in turn, deepens our insight into the fascinating properties of CNTs. The C–C bond of the SWCNT contracts by  $\sim 18.5\%$ , with a concurrent energy increase of  $\sim 68\%$ . The effective thickness of the C–C bond is  $\sim 0.142$  nm, which is the diameter of a C atom, rather than the graphite sheet separation (0.33 nm) or the radius of a free C atom (0.066 nm). The melting point of the tube wall is slightly greater (by  $\sim 12$  K) than that of the tube end. The unique solution clarifies that the quoted values

essentially represent the true situations of a SWCNT in which the Young's modulus is 2.5 times greater and the melting point is 0.42 times that of bulk graphite. Predictions of the wall-thickness dependence agree well with the insofar-observed trends in  $T_m$  suppression and  $Y$  enhancement of the nanobeams. The findings provide consistent insight into the unusual thermal, chemical, and mechanical behaviors of nanobeams, as well as an effective approach toward bonding identities that are beyond the scope of direct measurement.

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