Computing the compressive strength of carbon nanotube/cement composite

Hasan Ghasemzadeh1,*, Esmat Akbari Jalalabad2

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Abstract

In this study compressive strength of carbon nanotube (CNT)/cement composite is computed by analytical method. For this purpose representative elementary volume (REV) as an indicator element of composite is chosen and analyzed by elasticity relationships and Von Mises’ criterion applied to it. It is assumed that carbon nanotubes are distributed uniformly in the cement and there is perfect bonding in the interface of cement and nanotube. At first for simplicity of computations, carbon nanotubes (CNTs) are assumed to have unidirectional orientation in the cement matrix. In following, the relations are generalized to consider random distribution of nanotubes in cement, and a new factor suggested for random orientation of fibers in the CNT/cement composite. The results of analytical method are compared with experimental results.

Keywords: Compressive strength, Carbon nanotube (CNT), Cement matrix, Random orientation of fibers, Analytical solution.

1. Introduction

Since invented by Iijima [1] in 1991, carbon nanotubes have been widely used in fields such as electronic materials, biological technology, chemistry and multifunctional composites. Nowadays carbon nanotube (CNT) is known to be the material of the 21st century and is currently receiving a lot of interests due to its extremely high mechanical properties; seamless tubes of graphite sheets with nano-sized diameter, a very high theoretical strength about 100 times more than that of steel and a specific gravity only one sixth that of the latter [2]. Moreover, carbon nanotubes have an elastic strain capacity of 12%, 60 times that of steel [3]. Their Young’s modulus is up to 1 TPa and tensile strength is reported to be as high as 200 GPa [4].

Cement is a commonly used construction material due to its low cost and high compressive strength and enhancement of its performance has been an important issue for the research community. Having the best performance among carbon materials, it is expected that carbon nanotubes can remarkably enhance the performance of cement composites. Experimental researches have been done about this idea and it is confirmed that adding carbon nanotubes to cement matrix can improve its mechanical behavior [5-7].

Experimental approach for determining properties of composites containing fibers especially carbon nanotubes, needs using of complex experimental methods and expensive laboratorial equipments. Theoretical approach can lower the cost of predicting properties of these composites. So, in recent years, it has been a great concern of researchers and they have attained the good results. Halpin et al. [8] computed the mechanical properties of polymer composite reinforced with fibers by analytical method. The tensile and flexural strength of cement composite reinforced with fibers can be obtained by rule of mixtures [9]. Young’s modulus of carbon nanotube reinforced concrete composite was computed by Rouainia et al. [10]. The effect of cyclic loading on punching strength of flat slabs strengthened with Carbon Fiber Reinforced Polymer (CFRP) sheets was studied by Esfahani [11]. Also, the impact of short random fiber inclusion on consolidation settlement, swelling, hydraulic conductivity, shrinkage limit and the development of desiccation cracks in compacted clays was studied by Abdi et al. [12]. The combining of fibers for a cementitious matrix was investigated by Oucief et al. [13] and they identified fiber combinations that demonstrate maximum synergy in terms of flexural toughness. An experimental study was presented by Eshghi et al. [14] on seismic repair of damaged square reinforced concrete columns with poor lap splices, 90-degree
hooks and widely spaced transverse bars in plastic hinge regions according to ACI detailing (Pre.1971) and (318-02) using Glass Fiber-Reinforced Polymer (GFRP) and evaluated seismic performance and ductility of the repaired columns in terms of the hysteretic response and compared with those of the original columns.

Yet there are a few works on predicting the mechanical properties of cement composite reinforced with nanoparticles by analytical method. In this paper an analytical relationship is presented to predict compressive strength of CNTs/cement composites and compared with experimental results.

2. Determining the compressive strength of cement composite reinforced with unidirectionally oriented carbon nanotube

2.1. Averaging in the Representative Elementary Volume

As shown in Figure 1a, nanotubes are assumed unidirectionally and uniformly distributed in the cement matrix and they are straight. Representative Elementary Volume (REV) as an indicator element of composite is selected (Fig. 1b) and described as follow:

Each fiber of radius \( r_a \) having elastic constants \( k_e, m_p, E_f \) and \( \nu_f \) is imagined to be embedded in a cylinder of radius \( b \) having the elastic constants \( k_m, m_m, E_m \) and \( \nu_m \) where \( k \) is plane strain bulk modulus, \( m \) is shear modulus at \( \pi/4 \) from principal axes, \( E \) is modulus of elasticity and \( \nu \) is Poisson’s ratio. Subscripts \( f \) and \( m \) denote fiber and matrix materials, respectively.

The REV cylinder is surrounded by an unbounded homogeneous medium which is macroscopically indistinguishable from the composite. Suppose that a uniform transverse radial stress is applied at infinity and that strain in \( z \) direction, \( \varepsilon_z \), is kept equal to zero by a necessary longitudinal stress \( \sigma_z \). For the above mentioned composite material, the stresses in the composite are expressed as follow:

\[
\begin{align*}
u_m &= \frac{n}{2(1+\nu_m)} , \quad l = 2k\nu_m .
\end{align*}
\]

With substituting equation2 and relationship 4 in the equation 3, at \( r = b \) we have:

\[
\sigma_z = 2(\overline{K} - m\overline{B})
\]

Bar sign denotes composite properties.

In the above relation, parameters \( m, k, \overline{B}, \overline{T} \) are properties of composite computed by Eq. 6 [16,17], and the constants \( A, B \) obtained in the next section.

\[
\begin{align*}
\overline{k} &= \frac{k_e(k_f + m_m)(1-\varphi) + k_m(k_m + m_f)\varphi}{(k_f + m_m)(1-\varphi) + (k_m + m_f)\varphi} \\
\overline{T} &= \frac{\varphi l_f(k_m + m_m) + (1-\varphi)l_m(k_f + m_m)}{\varphi(k_m + m_m) + (1-\varphi)(k_f + m_f)} \\
\overline{B} &= m_f \frac{2m_m k_m (k_m + m_m) + 2(1-\varphi)m_f m_m + (1-\varphi)k_m (m_f + m_m)}{2m_m (k_m + m_m) + 2(1-\varphi)m_f m_m + (1-\varphi)k_m (m_f + m_m)}
\end{align*}
\]

where \( \varphi \) is volume fraction of nanotube in the CNT/cement composite.

2.1.1. Computation of \( A, B \)

To determine \( A, B \), cross section of REV has been shown in figure 2. In this paper, it is assumed that there is a perfect bonding at interfaces of nanotube and cement matrix, and there is no sliding at interfaces. Boundary conditions for this case are:

1) \( u_r \) and \( \sigma_r \) are continuous at \( r = a \) and \( r = b \).
2) \( \sigma_z \) at infinity \( (r = \infty) \) is equal to compressive strength of CNT/cement composite with unidirectionally oriented fibers \( \varepsilon \).

In the core (nanotube; \( r \leq a \)), it is assumed that the strain is constant so we have \( B = B_f = 0 \) and \( A = A_f \). In the shell (cement region of REV) as an indicator element of composite is selected (Fig. 1b) and described as follow:

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\overline{T} &= \frac{\varphi l_f(k_m + m_m) + (1-\varphi)l_m(k_f + m_m)}{\varphi(k_m + m_m) + (1-\varphi)(k_f + m_f)} \\
\overline{B} &= m_f \frac{2m_m k_m (k_m + m_m) + 2(1-\varphi)m_f m_m + (1-\varphi)k_m (m_f + m_m)}{2m_m (k_m + m_m) + 2(1-\varphi)m_f m_m + (1-\varphi)k_m (m_f + m_m)}
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Bar sign denotes composite properties.

In the above relation, parameters \( m, k, \overline{B}, \overline{T} \) are properties of composite computed by Eq. 6 [16,17], and the constants \( A, B \) obtained in the next section.

\[
\begin{align*}
\overline{k} &= \frac{k_e(k_f + m_m)(1-\varphi) + k_m(k_m + m_f)\varphi}{(k_f + m_m)(1-\varphi) + (k_m + m_f)\varphi} \\
\overline{T} &= \frac{\varphi l_f(k_m + m_m) + (1-\varphi)l_m(k_f + m_m)}{\varphi(k_m + m_m) + (1-\varphi)(k_f + m_f)} \\
\overline{B} &= m_f \frac{2m_m k_m (k_m + m_m) + 2(1-\varphi)m_f m_m + (1-\varphi)k_m (m_f + m_m)}{2m_m (k_m + m_m) + 2(1-\varphi)m_f m_m + (1-\varphi)k_m (m_f + m_m)}
\end{align*}
\]

where \( \varphi \) is volume fraction of nanotube in the CNT/cement composite.
matrix; \( a \leq r \leq b \) \( A=A_m \) and \( B=B_m \). In the other parts of the body \( (r \geq b) \), where properties match with the composite, we have \( A=A \) \( B=B \).

By using the above mentioned notes, the boundary conditions are applied:

\[ u_r \text{ is continuous at } r=a \text{ so from equation 1 we have:} \]
\[ A_r + B_r = A_m + B_m \]
\[ (7) \]

\[ \sigma_r \text{ is continuous at } r=a \text{ so from equation 5:} \]
\[ k_r A_r = k_m A_m - \frac{m B_m}{a^2} \]
\[ (8) \]

\[ u_r \text{ is continuous at } r=b \text{ so from equation 1:} \]
\[ A_r + B_r = A_m + B_m \]
\[ (9) \]

\[ \sigma_r \text{ is continuous at } r=b \text{ so from equation 5:} \]
\[ k_r A_r = k_m A_m - \frac{m B_m}{b^2} \]
\[ (10) \]

From condition (2) and equation 5, can be determined as:

To determine \( \sigma_c \) in this state, according to figure 3, boundary conditions are:

1. \( u_r \) and \( \sigma_r \) are continuous at \( r = a \)
2. \( \sigma_r \) at \( r = b \) is \( \sigma_c \)

where \( \sigma_c \) is radial compressive strength of REV.

\[ 2(k_m A_m - \frac{m B_m}{b^2}) = \sigma_c \]
\[ (16) \]

From equations 7, 8 and 16 the values of \( A_r, A_m \) and \( B_m \) are calculated by mathematical operations as follows:

\[ A_r = \frac{\sigma_r (m_k + k_f)}{2(k_r + m_k) - m_k \sigma_r (k_r - k_f)} \]
\[ (17) \]

\[ A_m = \frac{\sigma_r}{2[k_m - m_k \sigma_r (k_r + k_m)]} \]
\[ (18) \]

\[ \sigma_r = \sigma_c \]
\[ \text{Fig. 3. Loading of REV} \]

2.1.2. Von Mises’ yield criterion

Here, for prediction failure of composite, von Mises’ yield criterion is applied to REV. Von Mises’ criterion in the general form is as follows [18,15]:

\[ (\sigma_r - \sigma_\theta)^2 + (\sigma_\theta - \sigma_z)^2 + (\sigma_z - \sigma_r)^2 = 6K^2 \]
\[ (13) \]

\[ \sigma_r, \sigma_\theta, \sigma_z \] are radial, tangential and longitudinal stresses respectively determined in equation 5 and \( K \) is constant value that will be discussed in the next section. Here, it was assumed in the REV that \( \varepsilon_c = \theta \) (plane strain condition), therefore \( \sigma_c = \nu(\sigma_r + \sigma_\theta) \) where \( \nu = \frac{1}{2} \) is Poisson’s ratio and by substituting this relation in the equation 13 we have:

\[ (1+\nu^2 - \nu)(\sigma_r + \sigma_\theta)^2 - 3\sigma_c \sigma_\theta = 3K^2 \]
\[ (14) \]
Note that $B_f = 0$ and $a^2/b^2 = \phi$ that can be proved simply.

Now radial compressive stress at $r = a(\sigma_{rf})$ can be computed by equation (17) and (15b):

$$\sigma_{rf} = 2k_f A_f \rightarrow \sigma_{rf} = \frac{\sigma_c(m_a + m_m)}{k_e(k_f (m_a + m_m))}(1 - \xi)$$

(20)

where \(\xi = \frac{m_a \phi(k_m - k_f)}{k_f (m_a + m_m)}\) .

\(\sigma_c\) corresponding to \(\sigma_r\) at $r = b$, is obtained by equations (18, 19 and 15c as follows:

$$\sigma_c = \left[ \frac{k_m \sigma_c (k_f + m_m) + (k_m - k_f) m_f \phi \sigma_c}{k_m (k_f + m_m) - (k_m - k_f) m_f \phi \sigma_c} \right]$$

(22)

By substituting of \(\sigma_c\) (Eq. 21) and \(\sigma_\theta\) (Eq. 22) into equation 14, the constant value $K^2$ is obtained as:

$$K^2 = \frac{(1 + \varphi \tau - \varphi)(1 + h)^2 - 3h}{\sigma_c^2}$$

(23)

where $h = \frac{1 + \xi}{1 - \xi}$.

### 2.2. Computing the compressive strength of CNT/cement composite

Knowing all parameters in the von Mises' criterion and substituting Eq.5, Eq.6, Eq.11, Eq.12 and Eq.23 into Eq.13, and by mathematical operations, Eq.24 is obtained for compressive strength of cement composite reinforced with unidirectional orientation of nanotubes ($S$):

$$S = \sqrt{3} \frac{K}{D}$$

(24)

$$K = \sqrt{\frac{(1 + \varphi \tau - \varphi)(1 + h)^2 - 3h}{\sigma_c^2}}$$

$$h = \frac{1 + \xi}{1 - \xi}$$

$$\xi = \frac{m_a \phi(k_m - k_f)}{k_m (k_f + m_m)}$$

$$D = \frac{\frac{3}{\varphi(1 - \frac{k_f}{k_m})(\frac{m_f}{k_m} + 1) + \frac{m_f}{k_m} + \frac{m_m}{k_m} - \frac{k_f}{k_m} - \frac{m_f}{k_m}} + \left( \frac{r}{k_m} - 1 \right)^2}{\left[ \varphi(1 - \frac{k_f}{k_m})(\frac{m_f}{k_m} + 1) - \frac{k_f}{k_m} + \frac{m_f}{k_m} - \frac{k_f}{k_m} - \frac{m_f}{k_m} \right]}$$

where

$$m_j = \frac{(E_j)}{2(1 + \nu_j)} \quad k_j = \frac{(E_j)}{2(1 + \nu_j)(1 - 2\nu_j)} \quad l_j = 2\nu_j k_j,$$

$$m_o = \frac{E_o}{2(1 + \nu_o)} \quad k_o = \frac{E_o}{2(1 + \nu_o)(1 - 2\nu_o)} \quad l_o = 2\nu_o k_o$$

In the above equation it’s assumed that $(\nu_{zz}) = (\nu_{yy}) = (\nu_{yy}) = \nu_i$.

### 3. Computing the compressive strength of CNT/cement composite with random orientation of carbon nanotubes

Although it was assumed that nanotubes have been oriented unidirectionally in the cement matrix, in fact there is commonly no control on orientation of nanotubes in cement in the laboratory and they are oriented randomly in composite, as shown in figure 4. Therefore equation 24 must be generalized to a composite with random orientation of fibers. Cox et al. [19] studied the effect of fiber orientation on mechanical properties of fiber reinforced composite. The distribution function $f(\theta)$ is assumed for fiber orientation in the composite and effective mechanical properties of fibers are computed in terms of their position angle in the composite $(0 \leq \theta < 90)$. It was found that the random orientation of fibers reduces the effect of fiber reinforcing with respect to unidirectional orientation. Therefore an orientation factor $\alpha < 1$ is used considering random orientation in the comparison with unidirectional orientation.

To take into account of random distribution of nanotubes, factor $\alpha$ must be multiplied to mechanical properties of CNTs. Factor $\alpha$ for the evaluation of tensile strength of polymer/CNT composites [20] and evaluation of tensile/flexural strength of cement composite reinforced with fibers [9] has been obtained as follows: when the fiber length is greater or has the same order of magnitude of the specimen thickness, the fibers are assumed to be randomly oriented in two dimensions and the orientation factor is equal to $1/3 = 0.33$ and when the fiber length is much smaller than the specimen thickness, the fibers are assumed to be randomly oriented in three dimensions and the orientation factor $\alpha = 1/6 = 0.167$ is used. Since the length of nanotubes is much
shorter than the thickness of specimen, three dimensional CNT distribution is assumed for CNT/cement composite. In this paper this method is used for evaluation the compressive strength of CNT/cement composite. It is assumed that the effect of variations the young's modulus of carbon nanotube with orientation (θ) on compressive strength of CNT/cement composite is negligible and orientation factor α is multiplied only to critical pressure for buckling of nanotube. This is due to two reasons: 1) It was found that the equation 24 is much more sensitive to critical pressure than young's modulus of nanotube, as will be presented in the section 5. The same behavior can be seen in the halpin-tsai equation used to determine tensile strength of CNT/polymer composites \[20\] or in the rule of mixtures equation used to determine tensile/flexural strength of fiber reinforced cement composites \[9\] that there isn't the term of young's modulus nowise. 2) The relation between young's modulus and applied stress to carbon nanotube in the radial direction has not been ever known; i.e. this isn't obvious that if critical pressure becomes α times, how much the young's modulus can change.

Also Poisson's ratio of carbon nanotube is assumed to be nearly constant \(\nu_f=0.16\)\[21\]. So to take into account of random distribution, orientation factor α is multiplied only to critical pressure for buckling of carbon nanotube.

Here orientation factor α=0.13 is suggested by verifying the analytical method and experimental data. This factor is slightly smaller than α=0.167 suggested for computing tensile strength of composites. This is probably due to buckling of carbon nanotubes when CNT/cement composite is under pressure. According to above mentioned notes, equation 24 is valid for compressive strength of CNT/cement composite with random orientation of fibers except equation21 that is modified as follows:

\[
\sigma_c = \sigma_c \frac{k_f(k_f + m_\rho)[1 - \xi]}{k_f(m_\rho + k_\rho)} \tag{25}
\]

where \(\xi = \frac{m_\rho \rho(g_{k_f} - k_f)}{k_f \rho(k_f + m_\rho)}\).

To verify analytical method with experimental results, 3 tests are presented here.

### 4. Experimental data

#### 4.1. Test1

Li et al. \[6\] modified carbon nanotubes by using H\(_2\)SO\(_4\) and HNO\(_3\) mixture solution producing high bonding strength in the interface of nanotube and cement matrix. Surface-modified carbon nanotubes were added in the amount of 0.5% by weight of cement to cement matrix. Surface-modified carbon nanotubes were added in the amount of 0.5% by weight of cement to cement matrix. In tables 1 and 2 Properties of carbon nanotubes and cement matrix are shown respectively. The ratio of sand/cement/water was 1.5/1/0.45. CNT/cement samples were prepared and compressive strength tests conducted. The results are shown in table 3.

#### 4.2. Test2

Chaipanich et al. \[26\] added carbon nanotubes of 0.5% and 1% by weight of cement in a fly ash cement system. The ratio of sand/cement/water was 3/1/0.5. Nanotubes had the diameter of less than 100 nm and were dispersed in the cement using ultrasonic. Properties of cement matrix and proportions of different mixes are shown in table 4 and table 5 respectively. Compressive strength tests were conducted and the results are shown in table 6.
4.3. Test 3

Nochaiya et al. [27] added carbon nanotubes of 0.5% and 1% by weight of cement in a Portland cement matrix. Carbon nanotubes were dispersed using ultrasonic in the cement. The ratio of sand/cement/water was 2.5/1/0.5. Properties of Portland cement matrix and compressive strength of samples are shown in tables 7 and 8 respectively.

5. Results and discussion

To compare the present analytical method with experimental results, existent data in the tests are substituted in the equations 24 and 25 with terms of $\varepsilon$=0.13. The results are shown in table 9. The minimum error value is -3.3 % in the test 3 and the maximum error value is -9.94% in the test 1.

According to Equations 24 and 25, the effect of critical pressure for buckling of carbon nanotube ($\sigma_f$) on compressive strength of CNT/cement composite for different radial elastic moduli of CNT is shown in figure 5. This relation is linear and with increasing the critical pressure, the compressive strength of composite increases. Also this figure shows that in small radial elastic modulus of CNT the reinforcing effect of that in the cement matrix is increased, maybe since there is small difference between the values of elastic modulus of CNT and cement matrix in this case; therefore bonding strength of CNT/cement increases. Moreover in lower elastic moduli of carbon nanotube, with increasing the critical pressure, the compressive strength of composite increases rapidly.

Effect of the amount of carbon nanotube on compressive strength of CNT/cement composite for different compressive strengths of cement matrix ($S_m$) is shown in figure 6. With increasing the amount of CNT, the reinforcing content is increased and therefore compressive strength of composite increases slowly.

There is the volume fraction parameter of nanotube ($\phi$) in the relationships, whereas weight percent of that ($w_j$) is used in the laboratories. The conversion relation of these two parameters is as follows that can be proved simply:

$$\phi = \frac{1}{\rho_m} \left( \frac{\rho_f}{w_f} \right) + 1$$

where $\rho_m$ and $\rho_f$ are the density of cement matrix and carbon nanotube respectively.

In figure 7 variations of the compressive strength of CNT/cement composite is shown with respect to radial elastic modulus of nanotube for different critical pressures for buckling of CNT ($\sigma_f$). As it is seen, with increasing the radial elastic modulus of carbon nanotube, compressive strength of composite declined in nonlinear mode. This shows that reinforcing effect of CNTs in the composite reduces with increasing the radial elastic modulus of them. As stated before,

Table 7. Properties of Portland cement matrix

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compressive strength</td>
<td>52.6 MPa</td>
</tr>
<tr>
<td>Elastic modulus</td>
<td>29352 MPa</td>
</tr>
<tr>
<td>Density</td>
<td>2.19 g/cm³</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>0.18</td>
</tr>
</tbody>
</table>

Table 8. Strength of mixes after 28 days curing [27]

<table>
<thead>
<tr>
<th>Mix</th>
<th>Compressive strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PC</td>
<td>52.6</td>
</tr>
<tr>
<td>PC: 0.5% CNTs</td>
<td>58</td>
</tr>
<tr>
<td>PC: 1% CNTs</td>
<td>61.3</td>
</tr>
</tbody>
</table>

Table 9. Compressive strength of CNT/Cement composite (MPa)

<table>
<thead>
<tr>
<th>Test</th>
<th>CNTs</th>
<th>Experimental</th>
<th>Analytical</th>
<th>Error</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>%</td>
<td>data</td>
<td>method</td>
<td>%</td>
</tr>
<tr>
<td>Test 1</td>
<td>0.5%</td>
<td>62.13</td>
<td>55.954</td>
<td>-9.94</td>
</tr>
<tr>
<td>Test 2</td>
<td>0.5%</td>
<td>51</td>
<td>55.3987</td>
<td>+8.62</td>
</tr>
<tr>
<td>Test 2</td>
<td>1%</td>
<td>51.8</td>
<td>55.3988</td>
<td>+6.94</td>
</tr>
<tr>
<td>Test 3</td>
<td>0.5%</td>
<td>58</td>
<td>56.0672</td>
<td>-3.33</td>
</tr>
<tr>
<td>Test 3</td>
<td>1%</td>
<td>61.3</td>
<td>56.0673</td>
<td>-8.53</td>
</tr>
</tbody>
</table>

Note: PC = Portland cement
it is probably because of less bonding strength between carbon nanotube and cement matrix in higher radial elastic modulus of CNTs. But by comparison of figures 5 and 7, it’s apparent that the effect of radial elastic modulus of CNT on compressive strength of composite is very small in comparison with the effect of critical pressure for buckling of carbon nanotube, afore mentioned in the section 3.

6. Conclusion

In this paper an analytical relationship was presented to compute compressive strength of CNT/cement composite based on properties of its components i.e. carbon nanotube and cement matrix. Here, it’s assumed that carbon nanotube is transversely isotropic with continuum structure and distributed uniformly in the cement matrix. Considering orientation effect of carbon nanotubes in the CNT/cement composite, the relationship was improved according to laboratory tests by an orientation factor (\(\alpha\)).The effect of important factors on compressive strength of CNT/cement composite was obtained. In order to increase the performance of CNTs in CNT/cement composites, carbon nanotubes having lower radial young’s modulus and more critical pressure of buckling should be used.

References