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Numerical Simulation of CNT Incorporated Cement

B. S. Sindu**,** Saptarshi Sasmal , Smitha Gopinath

Abstract**—**Cement, the most widely used construction material is very brittle and characterized by low tensile strength and strain capacity. Macro to nano fibers are added to cement to provide tensile strength and ductility to it. Carbon Nanotube (CNT), one of the nanofibers, has proven to be a promising reinforcing material in the cement composites because of its outstanding mechanical properties and its ability to close cracks at the nano level. The experimental investigations for CNT reinforced cement is costly, time consuming and involves huge number of trials. Mathematical modeling of CNT reinforced cement can be done effectively and efficiently to arrive at the mechanical properties and to reduce the number of trials in the experiments. Hence, an attempt is made to numerically study the effective mechanical properties of CNT reinforced cement numerically using Representative Volume Element (RVE) method. The enhancement in its mechanical properties for different percentage of CNTs is studied in detail.

Keywords**—**Carbon Nanotubes, Cement composites, Representative Volume Element, Numerical simulation

I. INTRODUCTION

IJIMA (1991) discovered the Carbon Nanotubes (CNTs) while studying the carbon produced by arc evaporation of graphite in the helium atmosphere using High Resolution Transmission Electron Microscopy (HRTEM). CNTs exhibit extraordinary strength with moduli of elasticity on the order of TPa and tensile strength in the range of GPa. The enhancement in the mechanical properties of polymeric and ceramic composites

In the last decade, Carbon Single Walled Nanotubes (SWNTs) and Multi Walled Nanotubes (MWNTs) have triggered intense research effort in the computational investigation of their mechanical properties, both to explain observations and to obtain information not accessible from experiments. Modeling and simulations can help in the understanding, analysis and design of nanomaterials. At the nanoscale, analytical models are difficult to establish, while experiments are expensive to conduct. On the other hand, modeling of nanocomposites can be done effectively and efficiently. Some initial guidelines to the experimental work can be obtained readily by performing modeling and simulation work which will help to reduce the scope, cost and time for the experiments. The main issues in simulations are the proper selection of the mathematical models or theories for the problems considered. Meaningful computer simulations are very much dependent on the accuracy of the mathematical models for the materials under investigation.

There are two main classes of theoretical methods which include atomistic methods and continuum mechanics based methods. Atomistic based methods are computationally complex because of the large number of atoms required to be handled whereas the continuum mechanics based methods needs all the assumptions to be validated from atomistic based methods.

The effective material properties of CNT based composites was evaluated from three 3-D nanoscale RVE based on continuum mechanics and Finite Element (FE) modeling by Liu and Chen [2]. Formulas to derive the material properties for cylindrical RVE were obtained from theory of elasticity. The results were compared with extended rule of mixtures and experimental investigations.

Chan and Andrawes [3] addressed the uncertainties involved in using CNT as reinforcing material using the developed FE model. This method can be used in choosing the optimal mechanical and interfacial parameters for better strength and ductility of CNT - cement composites. The effect of parameters like interfacial bond strength, allowable slip, residual bond stress, Young's modulus of CNT and aspect ratio on the Young's modulus and ductility of CNT – cement composites were studied using the developed model. Chan and Andrawes [4] used the nanoscale representative pull out model to determine the interfacial bond behavior of single CNT embedded in cement. Microscale smeared reinforcement model was used to study the flexural behavior of CNT-cement composite beam. Experiments were also conducted on CNT-cement composites to validate the numerical studies.

The effective material properties of CNT based composites were evaluated from square RVE based on continuum mechanics and FE method by Joshi et al. [5]. The input for RVE and FE model was derived from theory of elasticity. The results obtained from FE model was compared with that of rule of mixtures. Studies were carried out on composites with different CNT lengths and diameter. It was observed from the studies that addition of CNTs of 3.6% increased the stiffness of the composite by 33%.

The compressive strength of CNT-cement composite was studied by Ghasemzadeh et al. [6]. RVE was chosen as an indicator of CNT-cement composite and was analyzed using relations from theory of elasticity. von Mises yield criterion was applied in the study. It was assumed that a perfect bonding was present between CNT and cement. Expressions to determine the compressive strength of CNT-cement composite for uniform and random distribution of CNT were given.

Cement is weak in tension and hence reinforcing material like CNT is added to improve its tensile strength. Experimentally determining the mechanical properties of CNT reinforced cement requires huge amount of expertise and it is also time consuming. Analytical work can be carried out in order to give initial guidelines to the

Sindu B S, Quick Hire Scientist Trainee, with CSIR-Structural Engineering Research Centre/ CSIR Campus, Taramani, Chennai, India (phone: 9944992690; e-mail:sindu.lsn@gmail.com, sindu@serc.res.in).

Dr. Saptarshi Sasmal, Senior Scientist, with CSIR-Structural Engineering Research Centre/ CSIR Campus, Taramani, Chennai, India (e-mail:saptarshi@serc.res.in).

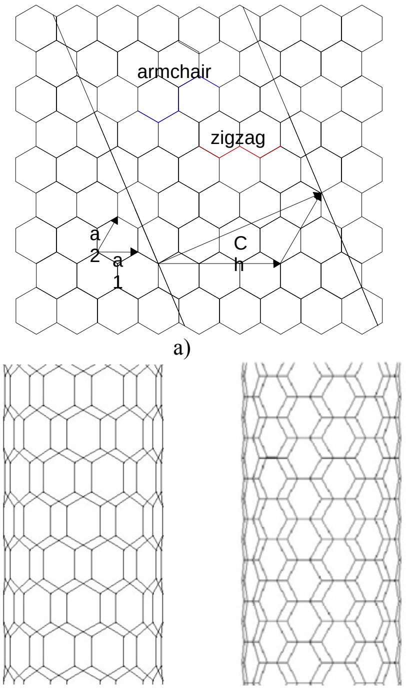
Smitha Gopinath, Scientist with CSIR-Structural Engineering Research Centre/ CSIR Campus, Taramani, Chennai, India (e-mail:smithag@serc.res.in).

experimental work. Hence analytical study is carried out on the tensile behavior of CNT reinforced cement for varying percentage of CNTs. In this paper, an overview of types of CNTs, its numerical simulation to determine the Young's modulus, the potential results for using CNT as a reinforcing material in cement and the challenges involved in it, modeling of CNT reinforced cement with different percentage of CNTs subjected to axial tension and evaluation of its effective material property is discussed.

II. CARBON NANOTUBES

A. *Structure of CNT*

Carbon nanotubes can be specified by two different parameters, viz., its length and chirality. Chirality of a carbon nanotube is specified by the multipliers of two base vectors in a chiral vector. A chiral vector is the one along which the graphene sheet can be rolled to form a carbon nanotube. There are two common types of nanotubes based on the way the graphene sheet is rolled to form CNT; they are zigzag and armchair. If the chirality of nanotube is (n,0), it is called zigzag nanotube and if it is (n,n), it is called armchair nanotube. Fig 1 a) shows the graphene sheet and the chiral vector. Fig 1 b) and c) shows zigzag and armchair CNT formed from rolling of graphene sheet.



a)

b) c)

Fig. 1 a) Zigzag and Armchair formed from graphene sheet
b) Zigzag CNT c) Armchair CNT

B. *Numerical Simulation of CNT*

A 3-D Finite Element Model has been created in ANSYS in which nodes were placed in the location of carbon atoms and the covalent bonds between the carbon atoms were replaced by three dimensional BEAM 4 elements. BEAM 4 element has six degrees of freedom and is defined by two nodes as well as its cross-sectional area, two moments of inertia, two dimensions and material properties. Jalalahmadi and Naghdabadi [8] used Morse potential in which the parameters were changed to fit the Brenner potential and obtained the effective bending and stretching stiffness of the bond as 854nN/nm and 0.9nN.nm/rad^2 respectively. Using these values and assuming circular cross-section, the diameter and the Young's modulus of the beam element can be found as 0.1296 nm and 9.382TPa respectively. Since the model is in linear regime, the Young's modulus of the nanotube can be obtained as

$$E = \frac{2U}{AL\varepsilon^2} \tag{1}$$

Where, ε is the strain, E is the Young's modulus, L is the length of the nanotube and A is the area which is equal to πDt in which D is the diameter and t is the thickness of the tube.

A Finite Element model of SWNT of particular diameter and length with the properties as described above has been generated in ANSYS software. It is fixed at one end and a tensile strain is applied on the other end. The model generated in ANSYS is shown in Fig.2. The total strain energy, U of the SWNT is calculated by summing up the strain energies of each beam element of the generated model and the Young's modulus is calculated using the above expression.

The Young's modulus of the SWNT obtained from Finite Element Modelling as mentioned above agrees well with the results obtained by Jalalahmadi and Naghdabadi [8]. A parametric study has been carried out to investigate the effect of change in radius of the nanotube on the Young's modulus for two different chiral tubes, viz., zigzag (n, 0) and armchair (n, n) was studied. The radius of the nanotube was varied from 0.3nm to 3nm keeping the aspect ratio constant equal to 6 in both the cases. The Young's modulus of the nanotube was plotted with respect to the tube radius for two different chiral tubes which is shown in Fig. 3. It is observed from the plot that the Young's modulus of the nanotube depends on the chirality of the tube. The Young's modulus of the zigzag nanotube is higher than that of armchair nanotube of same radius. It is also seen that the Young's modulus of nanotube for both the chiral tubes varied largely for tubes of radius less than 0.8nm. For nanotubes of radius greater than 0.8nm, the Young's modulus remained constant. This shows that Young's modulus of nanotubes of radius greater than 0.8nm does not depend on the tube radius and its chirality. For detailed discussions on the numerical simulation of CNT refer [9].

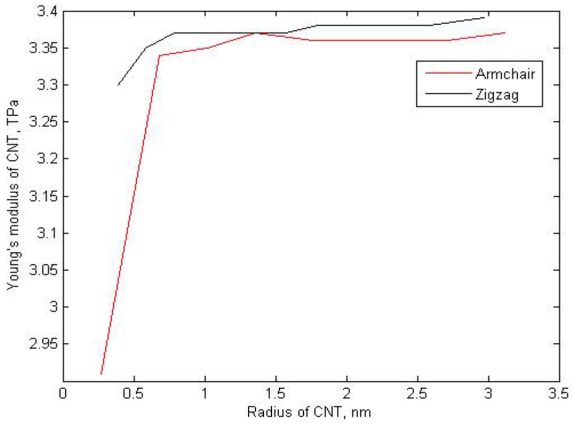


Fig. 2 Young's modulus vs radius of CNT

III. CNT Reinforced Cement – Challenges And Potential Results

While CNTs/CNFs have been extensively studied in polymeric composites, their use in cement has, to date, remained limited. Most research efforts have focused on CNTs compared to CNFs and has been performed on cement pastes. One of the main challenges is the proper dispersion of CNTs/CNFs into cement paste, partly due to their high hydrophobicity and partly due to their strong self attraction caused by high van der Waals forces. This attraction causes the CNTs to be more prone to agglomeration (bundling). This attraction causes them to rapidly settle out of suspension if not properly treated with surfactants and mixed with an ultrasonic mixer. (An ultrasonic mixer is a device that uses a high frequency driver to transmit acoustical energy throughout a liquid medium.) The energy in the shock waves is extremely high, significantly accelerates chemical reactions, and breaks the clumps and agglomerations of particles.

Although previous research has shown successful results in dispersing both CNTs and CNFs within aqueous solutions ([9], [10], [11]), few of these techniques can be applied to cementitious materials because of the retarded (i.e., delayed) hydration of the cement paste caused by large quantities of surfactants (Cwirzen [12]). For dispersing CNTs in aqueous solutions, Wang et al. [13] used a microwave to functionalize SWCNTs in a solution of nitric and sulfuric acid.

Li et al. [14] studied the mechanical behavior of cement composites incorporating MWNT treated by H_2SO_4 and HNO_3. The amount of CNT added was 0.5% by weight of cement. The composite was cured for 28 days and the compressive strength and flexural strength tests conducted on the specimens showed an increase in 19% and 25% of the respective strengths and also possessed the crack bridging properties. The study on porosity and pore volume using Mercury intrusion porosimetry showed that the use of CNT has decreased the porosity of cement by 64% and also has decreased its total pore volume. Microfibers may delay the nucleation and growth of cracks at the microscale, whereas nanoreinforcements will further delay the nucleation and growth of cracks at the nanoscale and stop their propagation to the microlevel. Hence, these nanofilaments, may prove to be superior alternatives or complements to traditional fibers and promising candidates for the next generation of high-performance and multifunctional cement-based materials and structures.

An efficient load transfer from the cement matrix to the nanotubes is required to take advantage of the very high Young's modulus and strength of carbon nanotubes in the composites. However, the strength of the interface between the nanotubes and the matrix is not well understood, and there is no well-developed experimental method for its direct measurement. The modeling and simulation of carbon nanotube reinforced cement can provide initial guidelines for the development of a material to help reduce the scope, cost, and time for the real experiments. Therefore, theoretically and computationally predicting the mechanical behavior of carbon nanotube reinforced cement will be of crucial importance before they are used in real structural applications.

IV. Modeling Of CNT Reinforced Cement

A. *General*

Nanocomposites extend from nano to micro or even to macro scales which is difficult to address using MD method and hence some other simulation techniques should be used separately or with MD to address these kinds of problems. Continuum mechanics approach can be used for this purpose. But while using continuum mechanics approach, every attempt should be made to check if all the assumptions of continuum mechanics hold good for nanomaterials. Continuum mechanics approach can be used to study the global responses like overall deformation and the load transfer mechanism. It cannot be used study the local effects like stresses at the interface.

In continuum mechanics method, CNT can be modeled as 1-d model (beam model) or 2-d model (shell model). However, since the surrounding matrix is modeled as 3-d model (solid model), CNT is also modeled as 3-d model in order to make compatibility of the degrees of freedom of CNT with that of the matrix at the interface.

CNTs can be of different types – MWNTs and SWNTs which in turn can be armchair or zigzag type. When it is dispersed in a matrix, it can be unidirectional or randomly oriented. CNTs are of different sizes also. All these factors make modeling CNT in a matrix complicated. To overcome these complications, the concept of Representative Volume Element (RVE) which is being used in modeling of fiber reinforced concrete was used to model CNT in a matrix by Liu and Chen [2].

In this study the concept of RVE is used to study the behavior of CNT reinforced cement. RVE is the simplest repeating unit in a composite in which a single CNT is surrounded by a matrix material with properly applied boundary conditions and interfacial properties which will represent the whole volume. This RVE model can be used to study the interaction of CNT and the cement matrix like the load-transfer behavior and to evaluate the effective material property of the composite material.

Liu and Chen [2] proposed three different RVEs to model CNT based composites – Circular (cylindrical) RVE, Square RVE and Hexagonal RVE as shown in Fig. 4. Circular RVE can be used to model the when CNTs are embedded in a carbon fiber or when CNTs has different diameters. Square RVE and hexagonal RVE can be used when CNT is arranged in a square array or hexagonal array respectively.

Circular RVE was used to evaluate the effective material properties of CNT composite under three different loading conditions. The analytical expressions based on strength of materials approach was derived evaluate the effective material property of the CNT based composite in the CNT direction. It was found that the results obtained by numerical methods showed higher stiffness of the composite compared to that obtained by rule of mixtures. The errors may be due to neglecting some material which is not covered by cylindrical cells. Hence, Liu and Chen [15] used square RVE to overcome the shortcomings of circular RVE.

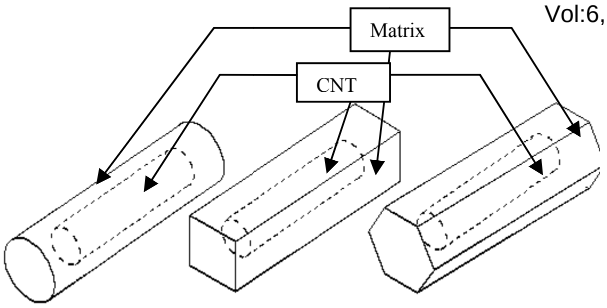


Fig. 3 Types of RVEs

B. *Evaluation of Effective Material Properties for Square RVE*

If the CNT is long, a segment of the composite can be modeled using RVE where the CNT runs along the entire length of the cement matrix. Fig.5 shows the RVE from the whole composite.

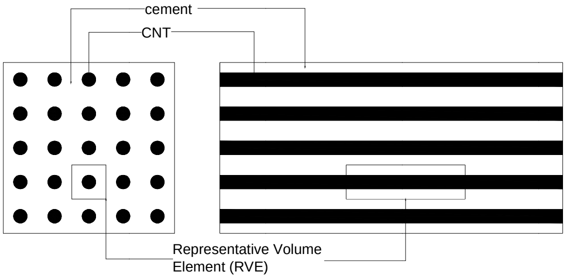


Fig. 4 Representation of RVE in a composite

The cross sectional area of square RVE is 2a x 2a and the length of RVE is L. In this square RVE, CNT of outer radius, r_o and inner radius r_i is embedded along the entire length of the composite. For a transversely isotropic material, the stress-strain relationship can be written as follows:

$$\begin{Bmatrix} \varepsilon_x \\ \varepsilon_y \\ \varepsilon_z \end{Bmatrix} = \begin{bmatrix} \frac{1}{E_x} & -\frac{\nu_{xy}}{E_x} & -\frac{\nu_{zx}}{E_z} \\ -\frac{\nu_{xy}}{E_x} & \frac{1}{E_x} & -\frac{\nu_{zx}}{E_z} \\ -\frac{\nu_{zx}}{E_z} & -\frac{\nu_{zx}}{E_z} & \frac{1}{E_z} \end{bmatrix} \begin{Bmatrix} \sigma_x \\ \sigma_y \\ \sigma_z \end{Bmatrix} \tag{2}$$

RVE is fixed at one end and subjected to an axial stretch ΔL at the other end as shown in Fig. 6

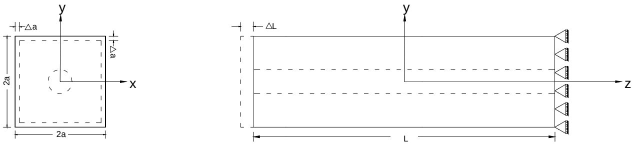


Fig. 5 RVE subjected to Boundary Conditions

The material constants, E_z and ν_{zx} can be obtained as follows by solving the equations obtained from the loading case as shown in Fig. 6.

$$E_z = \sigma_{z,avg} \frac{L}{\Delta L} \tag{3}$$

Where the average stress is obtained from

$$\sigma_{z,avg} = \frac{1}{A}\int_A \sigma_z\left(x, y, \frac{L}{2}\right) dxdy \tag{4}$$

where A is the area of the end surface.

and $$\nu_{zx} = -\left(\frac{\Delta a}{a}\right) / \left(\frac{\Delta L}{L}\right) \tag{5}$$

C. *Numerical modeling of RVE*

Finite Element Method has been used as a tool to characterize the mechanical properties of CNT reinforced cement. One quarter model has been developed for computational efficiency and symmetric boundary condition has been applied to represent the behavior of the full model. The mechanical properties of CNT reinforced cement are checked when the RVE is loaded in longitudinal direction.

The dimensions of the RVE are as follows:

Width and height of the cement matrix, 2a	: 95 nm
Length of the matrix, L	: 238 nm
Outer diameter of CNT, r_o	: 10 nm
Thickness of CNT	: 0.1296 nm
Inner diameter of CNT, r_i	: 9.7408 nm

Different models having same matrix dimensions and different number of CNTs representing different percentage of CNT by weight of cement have been developed. It is assumed that the CNT is uniformly dispersed in the matrix and hence models are developed with symmetric and uniform distribution of CNT.

The details of the models developed are shown in Table I.

TABLE I
DETAILS OF THE MODELS DEVELOPED

S. No.	**Number of CNTs**	**Percentage of CNT by weight of cement (%)**
1	4	0.093
2	9	0.22
3	16	0.42
4	25	0.72
5	36	1.2

The Fig. 7 shows a quarter model of RVE of CNT reinforced cement with 4 CNTs to study the properties along the longitudinal direction and Fig. 8 shows the cross-section of the quarter model of RVE of CNT reinforced cement with 36 CNTs.

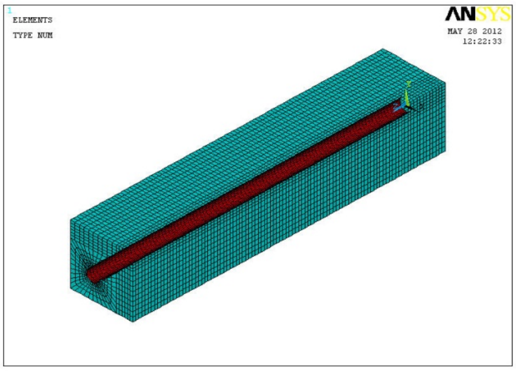


Fig. 7 Quarter model of RVE of CNT reinforced cement with 4 CNTs

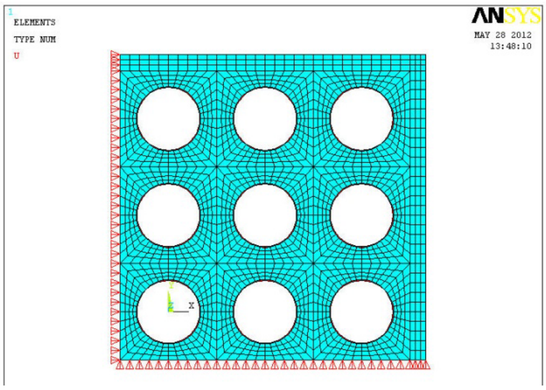


Fig. 8 Cross section of Quarter model of CNT reinforced cement with 36 CNTs

Both CNT and cement are modeled using 3-D solid elements. Solid 45 element of ANSYS is used to model the CNT with Young's modulus as 3.3GPa (average value obtained from previous study) and Poisson's ratio as 0.3.

Since cement cracks under tension and crushes under compression, the element capable of capturing this behavior should be used in numerical analysis. The element should be capable of handling non-linear material property. Solid 65 element in ANSYS is used for this purpose. It is a 8-noded element with three translational degrees of freedom in each node. It can be defined by nonlinear isotropic material property.

The assumptions of this element are as follows

- The material is assumed to be initially isotropic
- At each integration point, cracking is permitted in three orthogonal directions
- If cracking occurs through an integration point it is modeled through an adjustment of material properties which effectively treats cracking as a smeared band of cracks rather than discrete cracks

The stress-strain curve of cement given as input is from the experimental investigations conducted by Kuo et al. [16] on cement paste with water/cement ratio of 0.45 as shown in Fig. 9. The curve corresponding to $c_1 = 0$ represents the stress-strain curve for cement paste. The stress-strain value of this curve is given as input for cement.

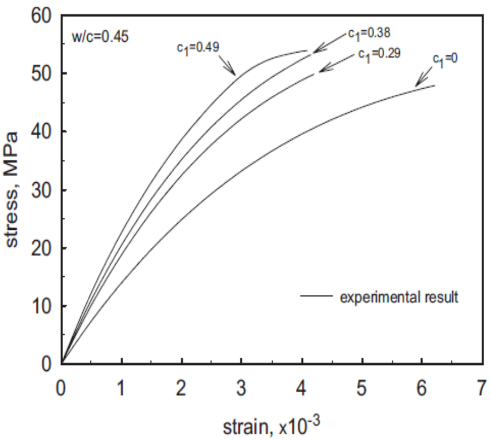
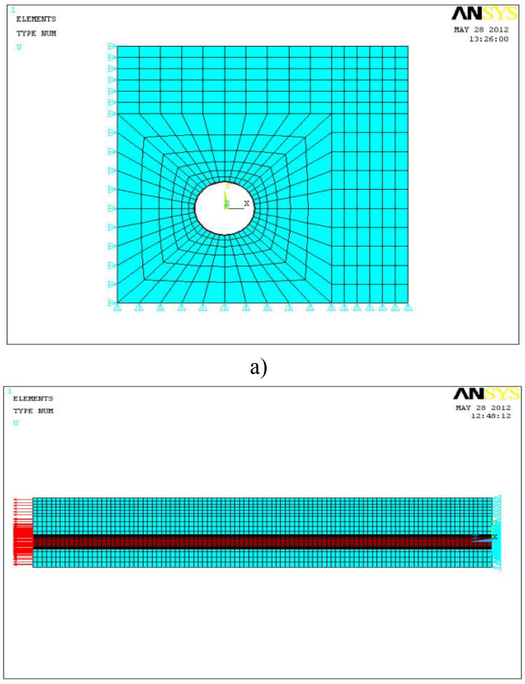


Fig. 9 Stress-Strain curve of cement

Rate independent plasticity criteria and William-Warnke failure theory are used suitably as defined in ANSYS to describe the failure theory of the cement. Rate independent plasticity is defined by the occurrence of irreversible strain in the material when a constant level of stress is reached. The plastic strains are assumed to occur instantaneously, i.e., independent of time. Associative flow rule is used to determine the direction of the plastic straining. Isotropic hardening rule is used to describe the change in the yield surface of the material with progressive cracking. In order to study the mechanical properties along the longitudinal direction, one end of the RVE is arrested to move along longitudinal direction and an uniform pressure is applied along the other end. Fig.10 a) shows the cross-section of the quarter model of RVE of CNT reinforced cement with 4 CNTs with symmetric boundary conditions and Fig. 10 b) shows the longitudinal view of the quarter model of RVE of CNT reinforced cement with 4 CNTs with displacement arrested along Z-direction along one end and pressure (tension) applied on the other end.



a)

b)

Fig. 10 Quarter model of RVE with applied Boundary Conditions subjected to pressure (tension) a) Cross-sectional view and b) Longitudinal view

A parametric study is carried out on the strength and stiffness along the longitudinal direction of RVE with different percentage of CNT.

V. Results And Discussions

The RVE is subjected to incremental tensile pressure along the longitudinal direction and the applied pressure versus end displacement is plotted for varying percentage of CNTs by weight of cement. The plot is shown in Fig. 11.

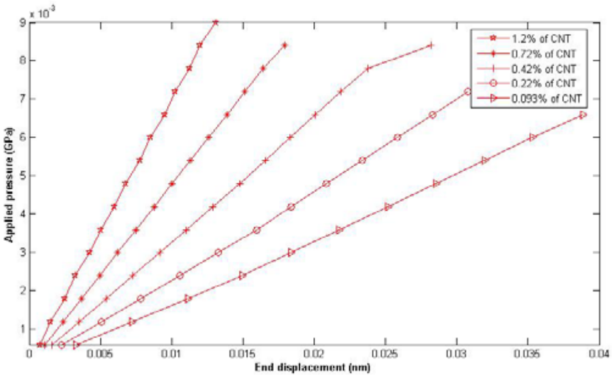


Fig. 11 Applied Pressure (tension) vs End displacement for different percentage of CNT

The Young's modulus along the longitudinal direction, E_Z is also calculated using the formula derived by strength of materials approach (Liu and Chen, 2004). Table 2 shows the values of Young's modulus of CNT reinforced cement for different percentage of CNTs.

TABLE II
YOUNG'S MODULUS OF CNT REINFORCED CEMENT

Percentage of CNT by weight of cement (%)	Young's modulus of CNT reinforced cement (GPa)
0.093	20.34
0.22	23.72
0.42	40.73
0.72	63.34
1.2	92.28

From Fig. 11 and Table II, it is seen that as the percentage of CNT increases, the ultimate strength and the stiffness of the composite increases.

VI. SUMMARY

In this paper, an overview of different types of CNTs and their mechanical properties are discussed. The numerical modeling of CNT to determine its mechanical properties has been discussed and the change in Young's modulus of CNT with respect to chirality and diameter has been reported. The promising results of the enhanced mechanical properties of cement reinforced with CNT and the challenges involved in reinforcing CNT in cement has been reviewed. It is also attempted to numerically investigate the mechanical properties of CNT reinforced cement. A parametric study is carried out on the pressure-displacement relationship of CNT reinforced cement subjected to axial tension with different percentage of CNTs incorporated into it. It is found that the rate of increase in the stiffness of CNT reinforced cement is prominent only upto certain percentage of CNT and later it slows down. Hence this study shows the possibility to arrive at the optimum percentage of CNT to be incorporated in the cement.

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