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# A stochastic algorithm for modeling heat welded random carbon nanotube network

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ABSTRACT

Carbon nanotubes (CNTs) are one of the presents of nanotechnology being investigated due to their extraordinary mechanical, thermal and electrical properties. Carbon nanotube networks feed the idea that CNTs can be used as the building blocks of new advanced materials utilizing the superior characteristics of CNTs. In this way, nanoscale features of CNTs can be scaled up to even continuum proportions. In this study, 2-D and 3-D CNT network generation methods are introduced by which the geometrical parameters, such as CNT length, chirality, intersection angle and junctional density, can be controlled and a random CNT network is obtained. Then, molecular dynamics (MD) simulations are used to create covalent bonds between intersecting CNTs, which allow the investigation of the mechanical, thermal and electrical properties of random CNT networks.



## 1. Introduction

Carbon nanotubes (CNTs) are one of the extraordinary nanomaterials that are promising candidates for thermal, electrical and structural applications due to their unique properties [1,2]. Since their discovery by Iijima (1991), thousands of studies, so far, have been adopted to their exceptional high strength and unusual electrical and thermal properties, displaying the desirable nature of their multifunctional capability. In the last years, the usefulness of CNTs has been enormously extended by their use as CNT networks through which CNTs are self-intersected in two or three dimensional space [3–9]. Because of their strong electrical conductivity with high light transmittance, CNT networks are attractive alternatives to silicon based macroelectronic devices [10]. Initial studies on 2-D CNT networks deposited onto flexible and polymeric substrates have focused on their electronic and sensor properties [11–14]. Several CNT network based thin films have been proposed to obtain lightweight, unbreakable displays and other flexible electronic devices [15]. Bottom-up controlled production of such networks can also enable them to be used in applications such as sensors, filters, composites and electromechanical actuators [1]. However, CNT networks also display the charming mechanical abilities of individual CNTs to macro-mechanical applications such as nanocomposites, similar to CNTs that are used as reinforcing units in nanocomposites, CNT networks can also be employed in nanocomposites for both reinforcement and damage detection purposes [16–18]. In this study, a self-controlled algorithm for generating a 2- or 3-D CNT network consisting of randomly oriented and intersected CNTs has been introduced, and a heat welding method is applied on sample networks to obtain covalently bonded networks by molecular dynamics (MD) simulations.

Furthermore, a recently developed CNT aerogel [4] material that is nanostructured by self-intersecting CNTs forming a random network at the continuum scale is one of the important examples displaying an ultra-high stiffness-to-weight ratio material with conductive properties. Instead of having a reinforcement role in nanocomposites, this material enables the CNT network to constitute a bulk material on its own.

Unquestionably, the electrical, thermal or mechanical properties of CNT networks depend on the density of junctions between CNTs as well as junction properties and impurities throughout the network. It has been shown that electrical conductance and mechanical strength of the junctions may be enhanced by manipulating junction area, i.e. increasing the crossing area [19]. In this manner, the algorithm proposed in this study will provide complete control on the junction properties and all the other geometrical features (e.g. CNT length, diameter, CNT intersection angle), which will enable the exploration of CNT networks theoretically or numerically by using much more realistic models.

Generally, CNT networks can be formed randomly by depositing CNTs locally on the catalyzed substrates by chemical vapor deposition methods (CVD) or depositing from CNT embedded polymeric suspensions remotely by using spin coating, spray coating, or

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vacuum filtration techniques [20–23]. With deposition on the catalyzed substrates, alignment of long CNTs can be more evenly created with directional control, but growth temperatures are high and unwanted byproducts may also be accumulated [23]. Therefore, deposition from solutions with suspended CNTs is much more popular due to the ability to produce them at ambient conditions, despite the time-consuming steps such as purification and dispersion of CNTs.

Most studies investigating the properties of CNT networks are carried out experimentally following a corresponding production phase. For instance, the thermal stability of CNT networks produced by the dielectrophoresis method on microelectrodes has been studied experimentally to predict their uses in electrical applications [24]. Similarly, Wang et al. [19] investigated the electrical and mechanical properties of CNT networks with measurements taken in situ inside the transmission electron microscope. In another experimental study, CNT networks are used as reinforcing materials in composites and compared with composites reinforced by dispersed CNTs [16]. Thostenson and Chou studied the damage and strain sensing capabilities of CNT networks utilized in glass fiber–epoxy composites and have shown through tensile testing that conductive percolating CNT networks can detect the initiation and progression of damage [18]. Several other studies [25–29] exist in literature on the electrical, thermal or mechanical properties of CNT networks carrying out experimental investigations which, as the other aforementioned studies, inherently require equipment of manufacturing and testing. On the other hand, the majority of computational studies have focused on the investigation of individual CNTs and their nanocomposites [30–33]. Due to the lack of a proper method for the generation of random CNT networks and computational limitations, there is a limited number of studies that use numerical models and computational methods for the investigations of CNT networks. Moreover, existing studies [34–39] that employ numerical network models mostly use ordered networks which do not include the geometrical irregularities and other possible imperfections, such as bond rearrangements, at junctions. Moreover, in these models only one type of junctions are modeled. A recently published study [40] investigates the mechanical behavior of short single-walled CNT (SWCNT) aggregates composed of randomly dispersed non-intersected CNTs by molecular mechanics.

In this study, a quasi-random self-intersected CNT network generation algorithm that enables the control of behavior decisive parameters, such as CNT length scale, density of junctions and relative angular position, is presented and several example networks with different parameters mentioned above are generated. Following the generation of a CNT network in which the CNT units are so close together that heating to certain temperatures can yield a covalently bonded network, MD simulations are carried out to obtain bonded networks. As a result, parameter-controlled covalently bonded CNT network models can be further used within MD simulations to investigate mechanical, thermal and electrical properties of CNT networks and their co-operating systems (i.e. nanocomposites).

## 2. General concepts

The process of generating random networks can be introduced as a process of configuring the network items (in this case, randomly intersected CNTs) randomly in the design space. The placing of network items is carried out under constraints in a random nature. The algorithm explained here is also employed for the generation of random porous networks with spherical cells which is used to model microcellular carbon foams [41].

Before explaining the process of the algorithm, some important concepts that will often be used in the explanation should be defined. First of those, the **design space** is the volume where CNT items are placed randomly. In 2-D network applications, it is defined as a plane with a thickness that encapsulates the CNT units normal to the plane. The other definition, **library**, is just a conceptual library comprised of all the different types of items (CNTs) that will be placed into the design space. Regarding this definition, CNT units used in the random network can differ in length and chirality (diameter). As a result, distinct CNT items can be reserved in the library and chosen randomly before the placement. Another concept, **target**, which is important when putting the selected item into the design space is the item (CNT) on which other items are closely placed to create an interconnected network. Each target element has a number of **cross-linked CNTs** which will be welded on the target element. Finally, CNT items selected from the library can be placed into the design space only if they satisfy certain **design constraints** imposed on the welded network generation. Without satisfying the constraints, the CNT item remains as a **candidate** for the network and it is called a candidate CNT.

## 3. General algorithm

Network generation is a typical cyclic procedure. At each step of the loop, a new CNT item that satisfies the constraints is placed into the design space. At each step, a number of candidate CNTs are created consecutively and tested against the design constraints until a suitable candidate is found. There are 2 loops in this procedure. The first loop is for continuation of the generation of the CNT items while the second loop is the sub-loop for generation and validation of candidate CNTs. When one of the CNT candidates satisfies all the design constraints in the sub-loop, the generation process continues with the selection of CNT from the library in the main loop. The main procedure can be summarized in 5 steps.

(1) Taking from the library: a candidate CNT item is chosen randomly from the CNT library where multiple CNTs having different lengths and chiralities are stored. For each chirality, the diameters of corresponding CNTs are calculated.
(2) Rotation and translation: After randomly selecting CNTs from the library, they are randomly rotated and translated in the design space.
(3) Checking constraints: After rotation and translation, the design constraints are checked on the candidate CNT.
(4) Making a decision: If the constrains are satisfied, the candidate CNT is placed into the design space. Otherwise, the procedure is restarted from the beginning.
(5) Writing LAMMPS input data file: Atomic coordinates of each CNT item are written into different files instead of storing them in the memory. Using these files (atomic coordinates), the LAMMPS input data file is written to another file.

### 3.1. Line segment representation of CNTs

To save computational time, CNTs are represented by 3-D line segments passing through the central points of the CNTs cylindrical geometry as shown in Fig. 1.

Each line segment is represented by the coordinates of end points, which are the central points of the circles at the CNTs end. During the generation process, end point coordinates are also stored and written to a file. Rotation and translation operations are also applied to end points, while all distance and angle calculations are done via the line segments instead of atomic coordinates.

The random points on the line segments are generated via parametric line equations as shown in Eq. (1) where $\{x_0, y_0, z_0\}$ and $\{x_1, y_1, z_1\}$ are the tip coordinates and $t$ is the parameter