Nuclear Instruments and Methods in Physics Research A 791 (2015) 1–5

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Study on a novel dosimeter based on polyethylene–carbon nanotube composite

S. Malekie, F. Ziaie *

Radiation Application Research School, Nuclear Science & Technology Research Institute, PO Box 11365-3486, Tehran, Iran

ARTICLE INFO

Article history:
Received 1 February 2015
Received in revised form
12 April 2015
Accepted 13 April 2015
Available online 24 April 2015

Keywords:
Carbon nanotube
Percolation
Electrical conductivity
Composite
Dosimetry

ABSTRACT

In this research work, the electric current of polyethylene-carbon nanotube composite, in the electrical percolation threshold region, over the absorbed dose under the applied bias voltage was investigated via the finite element method. The investigated geometry was formed by a two-dimensional cross-sectional view of randomly orientated nanotubes as ellipses. The variable range hopping model developed by Mott and thermally activated hopping model was used to calculate the electrical conductivity of carbon nanotubes and polymer, respectively. Regarding the calorimetric approach, we considered the absorbed dose equal to heat capacity of polyethylene-carbon nanotube composite multiplied by temperature rise. Results showed that this kind of composite can be used for low dose rate applications for monitoring and radiation protection utilizations.

1. Introduction

Dosimetry and detection of ionizing radiation can be listed among the important investigation fields in the nuclear industry. Passive dosimeters, such as photographic films and thermoluminescent materials, are largely used for individual dosimetry of ionization radiation, but they do not provide a direct reading and need a further treatment after irradiation to yield any information on the dose level [1]. After the discovery of carbon nanotubes (CNT) by Iijima [2], it was possible to fabricate polymer-CNT composites. CNTs have been widely used because of their high electrical conductivity [3]. The nano-sized, high surface area and the high aspect ratio (AR) of CNTs, offer the great opportunities to enhance the electrical conductivity of the polymer-CNT composites even at a very low loading of CNTs in the polymer matrix [4]. The polymer-CNT composite is light; moreover, it is tissue equivalent, it has low cost and is easily processed. Accordingly, this kind of material has potential applications in the ionizing radiation dosimeter. Several investigations have been conducted into on these types of materials to be used as radiation sensors. The effects of length and critical density of CNTs on the electrical conductivity of a radiation sensor based on percolation theory were studied [5]. Other researchers investigated the interaction of radiation with functionalized CNTs that have been incorporated into various host materials, particularly polymeric ones [6].

The objective of this work was to evaluate the dose response of polyethylene-carbon nanotube (PE-CNT) composite close to electrical percolation threshold (EPT) so as to design a real time active dosimeter at a suitable voltage for low dose dosimetry and monitoring purposes.

2. Percolation media

The electrical conductivity of polymers is very low and mainly spanned in the range of 10^{-16}–10^{-12} S/m [7]. Adding CNTs to polymer matrix in a particular weight fraction entitled EPT, leads to a sudden several orders of magnitude increasing in electrical conductivity of the obtained nano-composite, due to the unique electrical characteristics of CNTs such as high electrical conductivity between 10^5 and 10^6 S/m [3]. Within a polymer matrix, a continuous channel may form by contacts between nanotubes in proximity leading to abrupt enhancements in the electrical conductivity [8]. In fact, at the point of EPT, spanning cluster is formed [9]. The EPT depends on the type, size, shape, surface area, and distribution of the filler particles [10]. The considerable difference between the electrical conductivity of these materials makes electrical percolation theory an ideal modeling tool to predict the electrical characteristics of the polymer-CNT composite in different weight fractions of inclusions [11]. The smart polymer-CNT composite can be investigated for dosimetric purposes of ionizing radiations. In this research work, the dose response of the PE-CNT composite in different CNT weight percentages terms was investigated. Since the optimized dose response and sensitivity of

* Corresponding author.
E-mail address: fziaie@aeoi.org.ir (F. Ziaie).

http://dx.doi.org/10.1016/j.nima.2015.04.031
0168-9002/

the under study dosimeter are highly dependent on CNTs weight fraction, so the electrical percolation theory is an appropriate approach to precise control of CNTs amount in the PE-CNT composite.

Polyethylene was chosen as a polymeric matrix because of its high breakdown voltage, semi-crystalline structure, and effective radiation resistance [12]. Single-wall carbon nanotube (SWCNT) has particular electrical properties, so it has the highest conductivity among any known fibers [6,13]. There are three main sources of electron conduction mechanisms in a percolating CNT network; namely: (i) the intrinsic conductance of the nanotubes, (ii) the direct contact conductance, and (iii) the conductance resulting from electron tunneling between sufficiently close nanotubes [14]. It is obvious that when CNTs are long, the tubes have a higher possibility to connect with each other, therefore, few nanotubes are required to form conductive paths within certain area [5]. To predict the electrical conductivity of a composite ($\sigma_{\rm com}$), two-exponent phenomenological percolation equation or general effective medium (GEM) equation was proposed [15]:

$$\frac{(1-\varphi)(\sigma_m{}^{\frac{1}{s}}-\sigma_{com}{}^{\frac{1}{s}})}{(\sigma_m{}^{\frac{1}{s}}+A\sigma_{com}{}^{\frac{1}{s}})}+\frac{\varphi(\sigma_{cnt}{}^{\frac{1}{t}}-\sigma_{com}{}^{\frac{1}{t}})}{(\sigma_{cnt}{}^{\frac{1}{t}}+A\sigma_{com}{}^{\frac{1}{t}})}=0 \tag{1}$$

where $\varphi$is the volume fraction of the inclusions,$A=(1-\varphi_c)/\varphi_c$, φ_C is the critical volume fraction, s and t are critical exponents, σ_m and σ_{cnt} represent the electrical conductivity of the matrix and CNTs, respectively. Eq. (1) yields the two limits of:

$$\sigma_{cnt}\to\infty \quad : \quad \sigma_{com}=\sigma_m\left(\frac{\varphi_c}{\varphi_c-\varphi}\right)^S \quad , \quad \varphi<\varphi_c \tag{2}$$

$$\sigma_m\to 0 \quad : \quad \sigma_{com}=\sigma_{cnt}\left(\frac{\varphi-\varphi_c}{1-\varphi_c}\right)^t \quad , \quad \varphi>\varphi_c \tag{3}$$

This theory applies throughout the nano, micro and macro-media [15]. The values of s and t directly depend on EPT value of the polymer nano-composites [16]. In preparation of a nano-composites, uniform distribution of CNTs within a matrix becomes predominantly preferable [8]. The relation to convert the volume fraction(φ_V)to the weight fraction (φ_W)is as [17]:

$$\varphi_W=\frac{1}{1+\frac{\rho_m}{\rho_{cnt}}\left(\frac{1-\varphi_V}{\varphi_V}\right)} \tag{4}$$

where ρ_m and ρ_{cnt} are the densities of the matrix and CNTs, respectively. For a uniform distribution and homogeneous system, it can be assumed that the volume fractions (in 3D) and surface fractions (in 2D) of inclusions are equivalent($\varphi_V\cong\varphi_S$).

3. Simulation methodology

3.1. Cross-sectional view of randomly oriented CNTs (CSROC)

To investigate the electrical characteristics of PE-CNT composite using the finite element method, considering a homogeneous dispersion of CNTs in a three dimensional system, it can be assumed that the volume fraction of CNTs is equivalent to their surface fraction in two dimensional status. Since the large numbers of CNTs are needed to form the percolating paths in 3D system, and because computer processing and memory usage are time consuming, the authors proposed a simplified two dimensional modified model.

According to Fig. 1, the cross section of an arbitrary CNT in 3D space can be exhibited as an ellipse. Depending on the polar angle, this cross section can be simulated with random numbers in a wide range of geometric shapes from a circle to a narrow ellipse. In fact, exerting a cut plane on 3D PE/CNT dielectric can be modeled simplistically by a 2D simulation, in which the cross-sections of randomly dispersed CNTs are described as ellipses depending on specified AR. The percolation behavior in PE/CNT nano-composites is attributed to the electrical conductivities in general, and electrical conductivity or electric current of PE-CNT composite can be predictable by the electrical percolation theory.

In this simulation, firstly, 2D media with size of 10 μm × 10 μm in which 200 ellipses are introduced randomly as designated. Depending on the angular orientation of CNTs and considering an AR=1000, the length of ellipses can vary from 2 nm to 2 μm randomly. Also, the overlap of two ellipses is not allowed according to the excluded area approach.

According to Fig. 1, to specify and locate an ellipse in a 2D system, four random numbers must be generated. These random numbers determine the center of ellipse (x,y), polar angle (θ) and azimuthal angle (φ). The polar angle determines the deviation of CNTs from z-axis, while the azimuthal angle represents the angle of semi-major axis of the elliptical cross sections related to CNTs with x-axis. Therefore, the coordinates of the elliptical cross-sections can be generated through the equations such as $x=[(L-2a)\times rand]+a$, $y=[(W-2a)\times rand]+a$, $\phi=\pi\times rand$, and $\theta=\pi/2\times rand$, where a and b are the ellipse semi-major and semi-minor axes, respectively($a=b/\cos(\theta)$, $b\le a\le AR$), L is the media length, and W is the media width (L=W=10 μm).

In this simulation b remains fix at 1 nm for all situations. The electrical potential was calculated through solving Laplace's equation numerically by the finite element method in defined boundary conditions with $\rho_{PE}=0.91\ \ g/{\rm cm}^3$and $\rho_{cnt}=1.5\ \ g/{\rm cm}^3$[7,18]. Fig. 2 depicts a schematic view of the model, consisting of randomly distributed CNTs embedded in a PE matrix forming a thin film composite sandwiched between the silver electrodes for

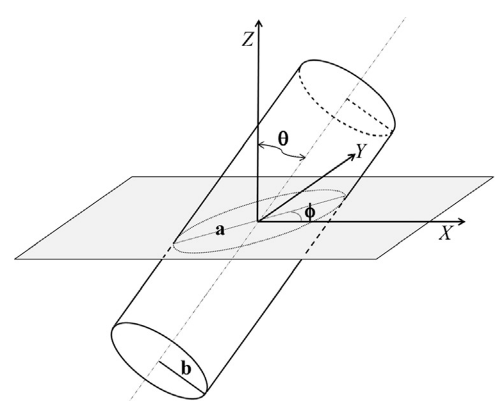

Fig. 1. Spatial representation of CNT with related elliptical cross section (CSROC model).

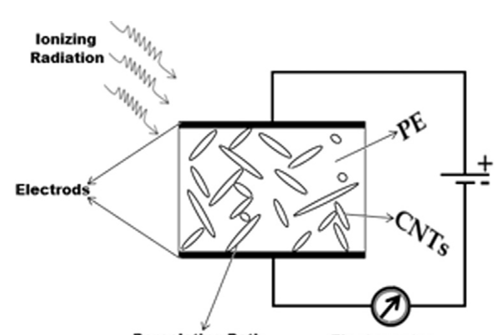

Fig. 2. Schematic diagram of 2D depicted CNTs in PE matrix as a composite dosimeter.