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Lower limits of detection in using carbon nanotubes as thermoluminescent dosimeters of beta radiation

Abdulaziz Alanazi^{a,b}, Izabela Jurewicz^a, Amani I. Alalawi^c, Amjad Alyahyawi^{a,d}, Abdullah Alsubaie^{a,e}, Steven Hinder^f, Jorge Bañuls-Ciscar^f, Mohammed Alkhorayef^{a,g}, D.A. Bradley^{a,h}

^a Centre for Nuclear and Radiation Physics, Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, UK

^b Medical Physics Department, Cancer Centre, Prince Mohammed Medical City, P.O. Box 2254, Sakaka Aljouf 42421, Saudi Arabia

^c Physics Department, Faculty of Applied Sciences, Umm AL-Qura University, Makkah, P.O. Box 175, Saudi Arabia

^d Department of Diagnostic Radiology, University of Hail, Hail, Saudi Arabia

^e Physics Department, Taif University, Al-Taif, PO Box 888, Saudi Arabia

^f The Surface Analysis Laboratory, Department of Mechanical Engineering Sciences, University of Surrey, Guildford, Surrey GU2 7XH, UK

^g Department of Radiological Sciences, King Saud University, P.O. Box 10219, Riyadh 11433, Saudi Arabia

^h Sunway University, Institute for Health Care Development, Jalan Universiti, 46150 PJ, Malaysia

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ABSTRACT

World-wide, on-going intensive research is being seen in adaptation of carbon nanotubes (CNTs) for a wide variety of applications, particular interest herein being in the thermoluminescent (TL) properties of CNTs and their sensitivity towards energetic radiations. Using beta radiation delivering dose levels of a few Gy it has been observed in previous study that strain and impurity defects in CNTs give rise to significant TL yields, providing an initial measure of the extent to which electron trapping centres exist in various qualities of CNT, from superpure to raw. This in turn points to the possibility that there may be considerable advantage in using such media for radiation dosimetry applications, including for *in vivo* dosimetry. CNTs also have an effective atomic number similar to that of adipose tissue, making them suitable for soft tissue dosimetry. In present investigations various single-wall carbon nanotubes (SWCNT) samples in the form of buckypaper have been irradiated to doses in the range 35-1.3 Gy, use being made of a 90 Sr beta source, the response of the CNTs buckypaper samples that the raw SWCNT buckypaper offer the greatest sensitivity, detecting doses down to some few tens of mGy.

1. Introduction

Since the discovery of CNTs by Iijima (Iijima, 1991), the potential for their applications has encouraged a large numbers of studies, both across the underpinning sciences as well as across the technologies that enjoy associations with these, investigations of CNT being in terms of properties and their utilisation. In regard to the advantageous properties of CNTs, among these one can mention their unusually large electrical- and thermal-conductivities (Hone et al., 2000) as well as their very large tensile strengths (Yu et al., 2000). Concerning the latter aspect, it is to be further appreciated that the microscopic network of thin samples formed from the CNTs in the creation of buckypaper (Fig. 1) gives rise to considerable flexibility, allowing large damage-free shape change. These particular properties point in part to the possibility of CNTs being candidate media for thermoluminescence dosimetry of energetic radiations, not least for *in vivo* applications. A further fundamental aspect for CNTs to be potential TLD media is the need for trapping centres, forming one particular focus of present investigation. Among the alternative forms of dosimetry are also included active devices such as semiconductor diodes and graphite ion chambers, these representing electrical risks, particularly for *in vivo* work, also passive TLD systems based on phosphors, such as LiF, that due to their inherent hygroscopic nature also have limited prospects for *in vivo* applications. CNTs also have an effective atomic number similar to that of adipose tissue, making it suitable for soft tissue dosimetry.

In previous work by this group (Alanazi et al., 2016) the thermoluminescence signal was measured from samples of Single-Wall

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E-mail address: a.alanazi@surrey.ac.uk (A. Alanazi).

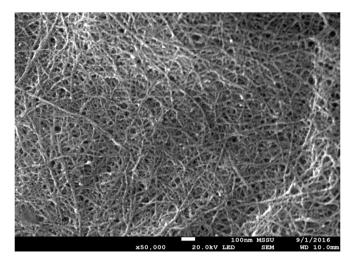


Fig. 1. Microscopic view of the buckypaper sample made of single-wall CNTs. Obtained using accelerating potential energy of 20 kV, and magnification power of 50,000. The scale is shown in the form of the 100 nm long white bar, provided in the lower part of the figure.

Carbon Nanotubes (SWCNTs) produced in the form of buckypaper, the samples being of varying purity. These aspects have been discussed in detail in the cited work. The samples (raw SWCNTs, pure SWCNTs, and super-pure SWCNTs) were irradiated by a ⁹⁰Sr source to obtain various doses. In present work we have conducted further investigations of the materials, seeking to obtain a measure of the lower limit of detection (LLD) for the particular samples and their associated production methods. The doses used were in the range of 35 mGy to 1.3 Gy, the response of the CNT buckypaper against doses showing a trend towards linearity.

2. Methods of sample preparation

The buckypaper samples formed from SWCNTs were of three types: raw SWCNTs, pure SWCNTs and super-pure SWCNTs. The three different qualities of SWCNTs were purchased from Unidym (Sunnyvale, CA, USA), delivered in the form of powder. The method of buckypaper preparation was as discussed in the previously cited work (Alanazi et al., 2016). The surfactant Triton X-100 was used to disperse the SWCNTs. With deionised water a magnetic stirrer was used to maintain a suspension of the surfactant. Membrane filters (0.22 µm pore size and of diameter 47 mm; MCE MF-Millipore plain white) were purchased from Thermo Fisher Scientific (Waltham, MA, USA), filtering and accumulating the CNTs. High frequency ultrasound was used for dispersion of the mixture, use being made of a Branson tip sonication system (Danbury, CT, USA) Sonifier 150. The optimum surfactant to CNT ratio was found to be in the range 5:1 to 10:1 by weight (Islam et al., 2003). In this and previous work, a surfactant to CNT ratio of 10:1 was used to prepare the various samples. The concentration of the single-wall carbon nanotubes in the dispersion that was used to form the buckypaper were 0.1, 0.1, and 0.05 g for raw, pure, and super pure single-wall carbon nanotubes respectively. These quantities were dispersed in 100 ml of deionized water through the use of Triton X-100 as previously mentioned.

Surface roughness and variability in the buckypaper samples as shown in Fig. 2 indicate considerable variability in the amount of CNTs deposited in the surface of buckypaper. Normalization for surface roughness can be expected to lead to reduction in the variability in response to dose.

2.1. Strontium-90 source for beta irradiation of CNT samples

Exposure of the CNTs samples was carried out in the radiation

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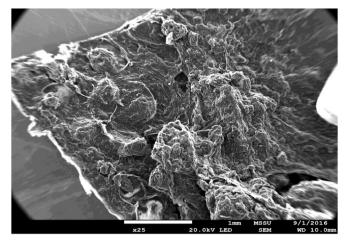


Fig. 2. Microscope image of one of the buckypaper super-pure single-wall CNT samples, the image being obtained using an acceleration potential 20 kV and magnification 25×, the surface roughness being highlighted. A 1 mm long scale is provided in the lower part of the figure.

laboratory, University of Surrey. A $^{90}\mathrm{Sr}$ beta source of approximate activity 95 MBq was used to irradiate the various CNTs samples, raw, pure, and super-pure. In order to minimise exposure to the user, the source was located within a box shielded by lead. The samples were positioned in a rotating disk that can accommodate 26 individual samples within it, each labelled to identify the individual samples. The set-up is as shown in Fig. 3. The different qualities of single-wall carbon nanotubes have been exposed to the beta-ray emissions to provide doses of 0.02, 0.035, 0.627, 0.836, and 1.245 Gy.

3. Results and discussion

3.1. Glow curve of the CNTs samples

Fig. 4 above shows the glow curves obtained for the irradiated CNT buckypaper samples for doses within the quoted range, the TL yields being observed to be encompassed within the temperature range 180–250 °C. The absence of noticeable differences in shape or distribution of TL yield with dose is indicative of an essentially identical trap centre activation energy distribution over the particular dose range. The temperatures have been made to ramp from room temperature to the maximum value of some 397 °C at a ramp rate at 6.5 °C per second, leading to an ability to discern above-background glow curves for doses down to 35 mGy, extending to 1.3 Gy, allowing the samples to be considered to be of utility for radiotherapy dosimetric applications. Thus said, we have also observed a reproducible hyper-response at 0.2 Gy of the order of four times greater than that obtained at any other dose within the dose range. The more general trend, in response to the



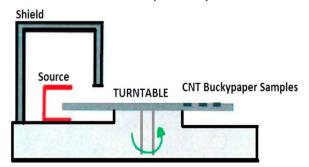


Fig. 3. The diagram shows the beta source irradiation set-up used to expose the CNT buckypaper samples.

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