Carbon Nanotubes "Buckypaper" Radiation Studies for Medical Physics Application

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Abstract

Radiation dosimetry underpins safe and effective clinical applications of radiation. Many materials have been used to measure the radiation dose deposited in human tissue, their radiation response requiring the application of correction factors to account for various influencing factors, including sensitivity to dose and energy dependence. In regard to the latter, account needs to be taken of difference from the effective atomic number of human tissue, soft or calcified. Graphite ion chambers and semiconductor diode detectors have been used to make measurements in phantoms but these active devices represent a clear disadvantage when considered for in vivo dosimetry. In both circumstances, dosimeters with atomic number similar to human tissue are needed. Carbon nanotubes have properties that potentially meet the demand, requiring low voltage in active devices and an atomic number similar to adipose tissue. In this study, single-wall carbon nanotubes (SWCNTs) buckypaper has been used to measure the beta particle dose deposited from a strontium-90 source, the medium displaying thermoluminescence at potentially useful sensitivity. As an example, the samples show a clear response for a dose of 2 Gy. This finding suggests that carbon nanotubes can be used as a passive dosimeter specifically for the high levels of radiation exposures used in radiation therapy. Furthermore, the finding points towards further potential applications such as for space radiation measurements, not least because the medium satisfies a demand for light but strong materials of minimal capacitance.

Keywords: Thermoluminescence, Carbon Nanotubes, Radiation Therapy.

1.- INTRODUCTION

Since CNTs were first reported upon by Iijima on early 1990s (Iijima 1991), the properties of carbon nanotubes have been the subject of intensive investigation and have been considered for many potential applications, such as chemical sensors (Kong, Franklin et al. 2000, Moradi, Sebt et al. 2013, Penza, Rossi et al. 2010, Wongchoosuk, Wisitsoraat et al. 2010, Zhao, Buongiorno Nardelli et al. 2005), biosensors(Timur, Anik et al. 2007, Balasubramanian, Burghard 2006, Koehne, Li et al. 2004, Li, Ng et al. 2003, Wang 2005), field-emission displays(Talin, Dean et al. 2001, Saito 2010, Lee, Chung et al. 2001, Kuznetzov, Lee et al. 2010, Choi, Chung et al. 1999), memory storage(Lu, Dai 2006, Ganguly, Kan et al. 2005) and hydrogen storage (Yang, Cho et al. 2010, Ranjbar, Ismail et al. 2010, Liu, Chen et al. 2010). In particular, the specific structure of the atoms forming the carbon nanotubes give interesting physical and chemical properties such as electric and thermo conductivity. Carbon nanotubes, CNTs, can be described as a beehive sheet fabricated to form a cylinder, the two ends of the cylinder being open or closed with a semicircle lid. This cylinder-like form can be shielded by other cylinders with open ends to form a multi-wall carbon nanotubes structure, MWCNTs. Figure 1 displays single and multi-wall carbon nanotubes with open ends. The dimensions of the structure are of the range of nanometres, the diameters ranging from a few nm up to 30 nm and the length of the tubes going up to ~ 1 μm (Iijima 1991). These dimensions can vary depending on the method of growth and the procedure parameters (Nihei, Kawabata et al. 2003). In this study, carbon nanotube Buckypaper was made at the University of Surrey, to be irradiated by strontium-90, a source of beta source. The thermoluminescence signal was obtained as the primary indicator of dose absorption.

1.1.- Buckypaper Samples

To enable the carbon nanotubes to be used as a passive radiation dosimeter they first need to be produced in such a way that they can be handled and used safely. For this, buckypaper can be made from the starting carbon nanotubes powder. Examples of buckypaper are illustrated in figure 2. The dimensions of the buckypaper can be controlled, dependent on the application and the interest prompting dose measurements. Typically the thickness of the buckypaper can be varied from few micrometres up to 0.5 millimetres.

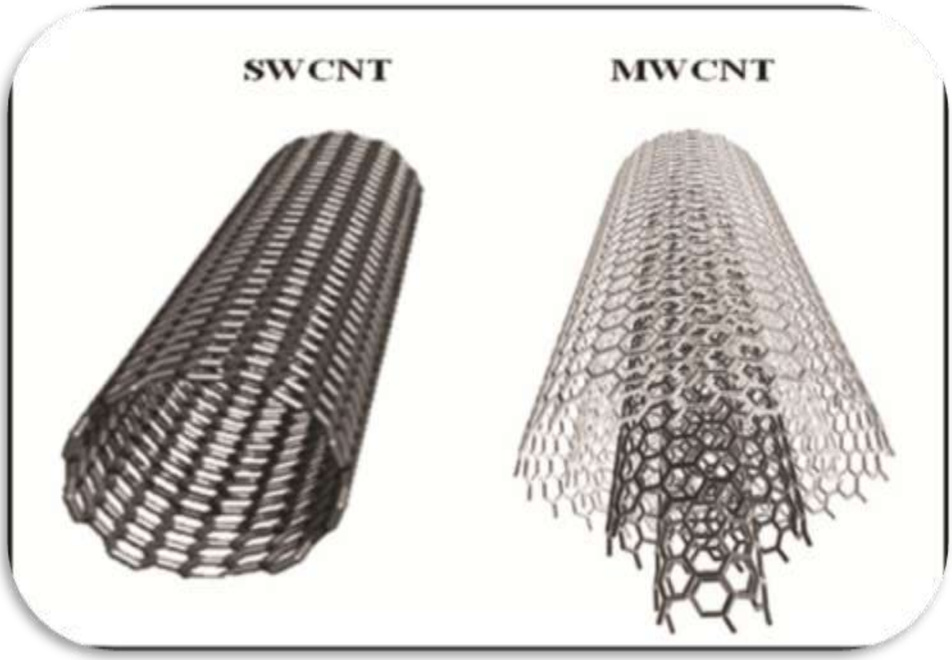

Figure 1: The structure of Single-Wall Carbon Nanotube, SWCNT, is displayed to the left-hand side while Multi-Wall Carbon Nanotubes structure is displayed to the right(Choudhary, Gupta 2011).

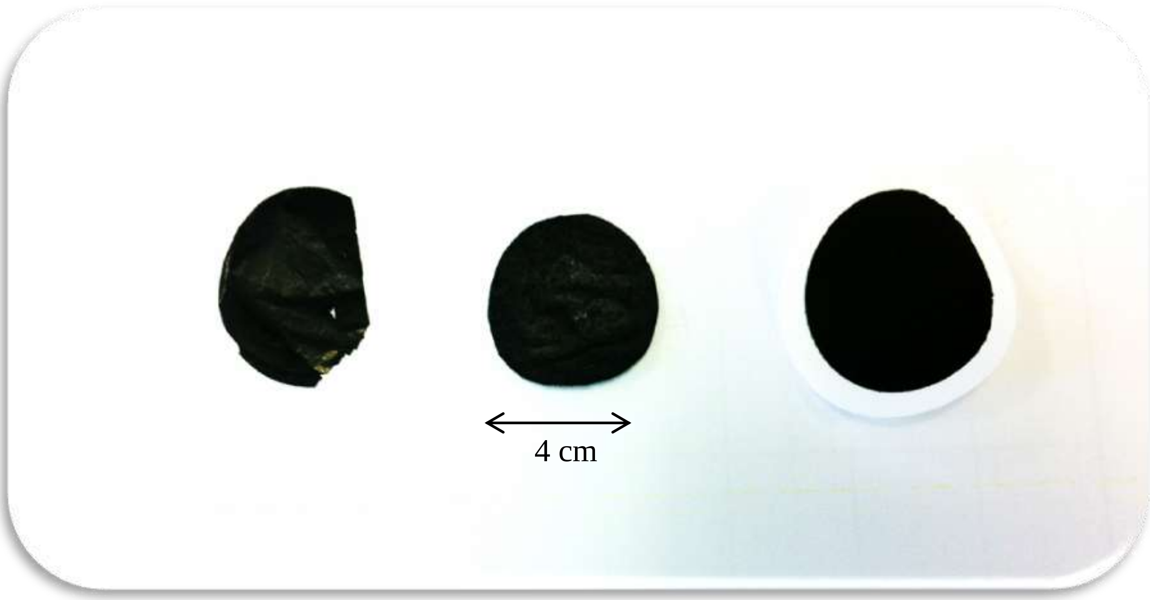

Figure 2: Samples of buckypaper of different thicknesses have been prepared and developed in the Nanomaterial and Structure Lab at University of Surrey.

Different qualities of single-wall carbon nanotubes, SWCNTs, were purchased from the company Unidym in the form of powder, Raw SWCNTs, Pure SWCNTs, and Super Pure SWCNTs. Triton X-100 has been used as a surfactant. A magnetic stirrer was used to maintain a suspension of the surfactant on the deionised water. The membrane filters, MCE MF-Millipore plain white, 0.22 µm pore size with diameter of 47 mm were purchased from Fisher Scientific Company to filter and accumulate the CNTs. A tip sonication, Branson–Sonifier 150, was used for dispersion of the mixture.

2.- METHODS OF PREPARATION OF THE SAMPLES

As there were different purifications of SWCNTs, various concentrations of the surfactant and SWCNTs were used for each quality. Different concentrations of CNTs require different quantities of the surfactant. In this study the ratio by weight was the reference in the calculations. The optimum surfactant to CNT ratio was found to be 5:1 to 10:1 by weight (Islam, Rojas et al. 2003). In this work 10:1 surfactant to CNT ratio was used to prepare all the samples and the amounts of CNT were from 0.03 g to 0.140 g depending on the purity of the CNTs.

2.1.- Strontium-90 Source for Beta Irradiation of CNT Samples

The various samples studied herein have been exposed to the strontium-90 source, as illustrated in figure 3. The source is retained within a plastic holder, contained within a leaded glass box to minimise the radiation hazard to users. The CNTs and TLD-100 (LiF(Tb) used as a well-characterised reference TLD medium) samples have been placed in an aluminium tray. The aluminium tray is designed to hold samples of size 5 × 5 mm. As we have different concentrations and quality of CNTs in the buckypaper, the edges of the tray are labelled by numbers and letters to help identify the samples.

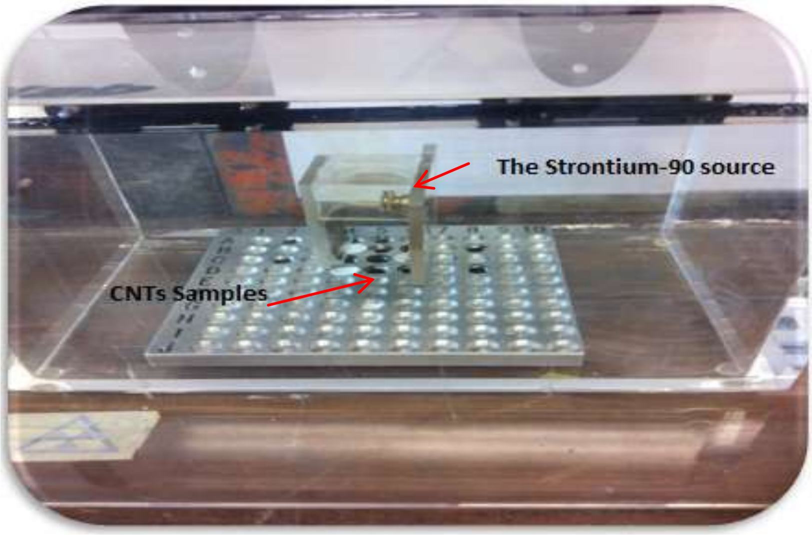

Figure 3: Set-up for the beta irradiation source, 90Sr, the buckypapers of size 5 × 5mm, and TLD-100 samples.

Buckypaper samples of various purities and concentration were irradiated in this experiment. Raw single-wall carbon nanotubes, pure single-wall carbon nanotubes, and super pure carbon nanotubes were used to form the various samples of buckypaper. The concentration of the CNTs in the dispersion used to form the buckypaper was 0.1, 0.1, and 0.05 gm 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. The three samples received a dose of 2 Gy.

3.- RESULTS AND DISCUSSION

3.1.- Raw Single-Wall Carbon Nanotubes

Figure 4 illustrates the thermoluminescence (TL) signal from the buckypaper prior to any treatment.

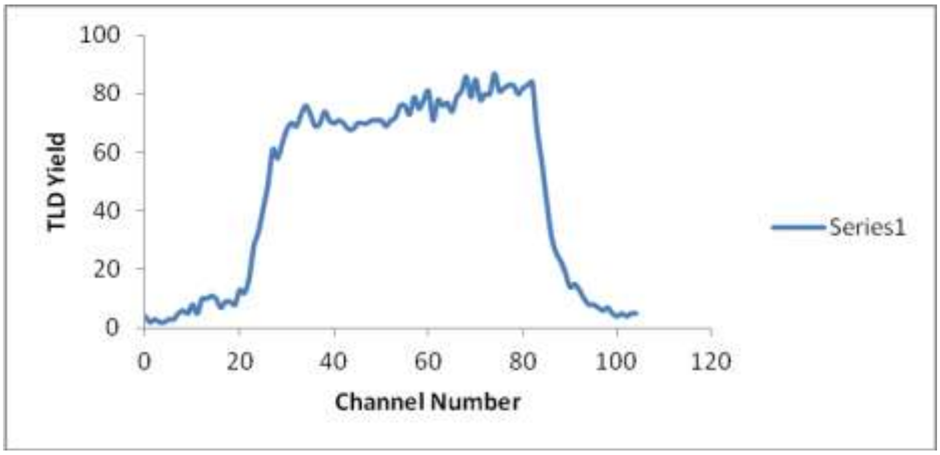

Figure 4: Thermoluminescence from buckypaper made of 0.1gm of Raw Single–Wall Carbon Nanotubes dispersed in 100ml of deionized water by the use of Triton X-100, the sample size 5x5mm.

This measurement was required to enable comparison between the TL yield from a fresh unirradiated sample that had not been subject to any heat treatment and the TL yield from a sample treated with heat during an annealing or readout process. The sample consists of buckypaper made of 0.1g of Raw Single–Wall Carbon Nanotubes dispersed in 100 ml of deionized water through the use of Triton X-100. The sample size was 5 × 5 mm. The signal profile has the character of multiple peaks which might be attributed to the random defects created during the accumulation of CNTs when the sample was prepared. This is supported by the fact that following annealing of the sample, the signal profile changes dramatically, the TL yield reducing by a factor of 18 (Figure 5, red line). The figure shows two graphs, one for the TL yield from an non-irradiated sample after annealing process takes place (red line), and the other the TL yield (represented by the blue line) from the sample following irradiation to a dose of 2 Gy, irradiated with a ^{90}Sr beta source. The TL signal from the irradiated sample shows a pronounced TL response compared with that without irradiation. The TL signal was improved by a factor of about 21 exceeding by far the TL yield from the unirradiated sample. The peak of this curve was found to be encompassed within the temperature range 180 to 250 °C.

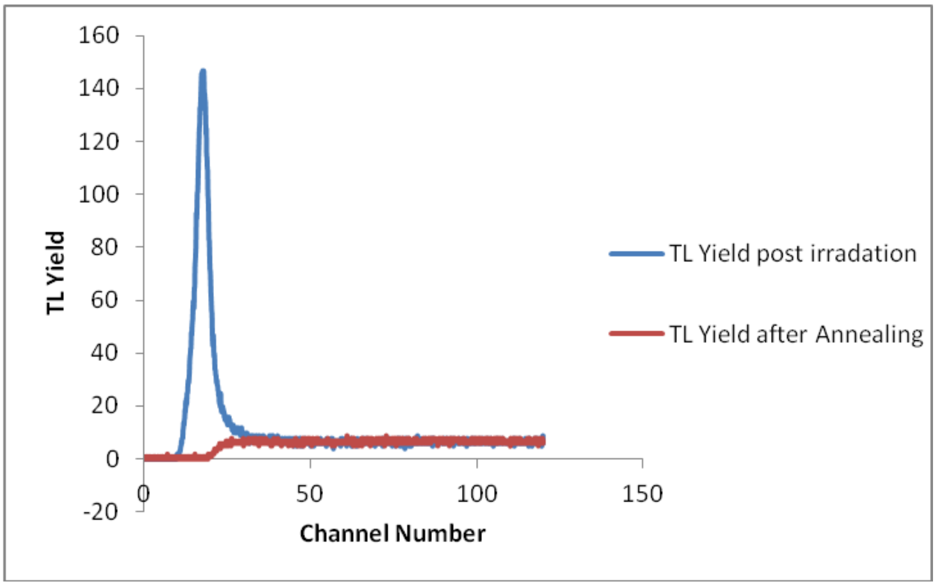

Figure 5: Blue line illustrates the thermoluminescence from the sample post-irradiation, while the red line illustrates the thermoluminescence from the sample unirradiated, post-annealing.

Another measurement was taken to measure any residual TL signal not released during the first readout procedure. Figure 6 shows two overlapping graphs, the blue line illustrating the first readout and the red line illustrating the second readout for the same raw SWCNT sample. From this result it is apparent that one heating cycle is sufficient to release practically all of the trapped electrons within the irradiated sample.

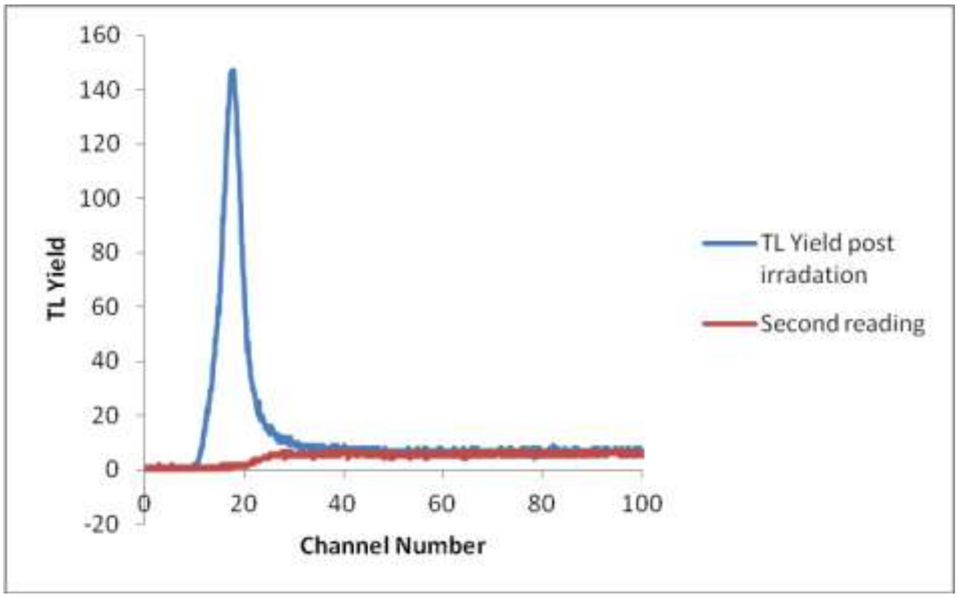

Figure 6: The blue line illustrates the first readout, and the red line illustrates the second readout for the same sample, raw SWCNTs after been irradiated to a dose of 2 Gy.

3.2.- Pure Single-Wall Carbon Nanotubes

Similar measurements were conducted when treating the other samples, pure SWCNTs and super SWCNTs. Figure 7 below illustrates the background reading for a fresh sample that has not been exposed to radiation or heat treatment. The results are similar to that obtained for the raw SWCNT sample for both measurements, pre and post annealing. The TL yield from the annealed sample was practically identical to that provided by the raw SWCNT sample. However, comparing the raw and pure SWCNT buckypaper sample, the magnitude of the TL signal is lower for the pure SWCNTs buckypaper sample than that from the raw SWCNT buckypaper sample, for both the fresh and irradiated samples. This reduction in TL signal is expected as the sample is purified of iron and other elemental components. The absence of such impurity has a clear effect in the TL signal as can be seen in Figures 7 and 8, comparing with the signal obtained from the raw SWCNT buckypaper sample shown in Figures 8 and 9.

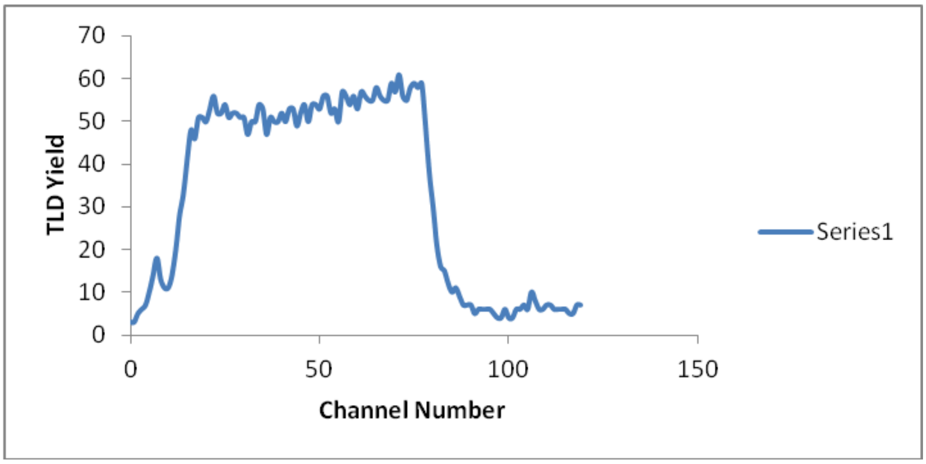

Figure 7: Thermoluminescence from Buckypaper made of 0.1 g of Pure Single–Wall Carbon Nanotubes dispersed in 100 ml of deionized water through the use of Triton X-100. The sample size was 5 × 5mm.

Reduction of TL yield of a factor of 1.33 was found for the pure SWCNT buckypaper in comparing with the TL Yield from the fresh samples in Figs. 8 and 10. Reduction of TL yield of a factor of 3.5 was obtained from the pure sample irradiated to a dose of 2 Gy using the same source and set up (Figure 8) compared with the yield from the raw SWCNT buckypaper sample (Figure 6). Figure 8 illustrates the first and second readings for the pure sample subsequent to irradiation delivering a dose of 2 Gy using the ^{90}Sr source. The blue line illustrates the first reading cycle and the red line illustrates the second read cycle, again supporting the previous sample results indicating little necessity for annealing when the sample is exposed to a dose in the range of several Gy.

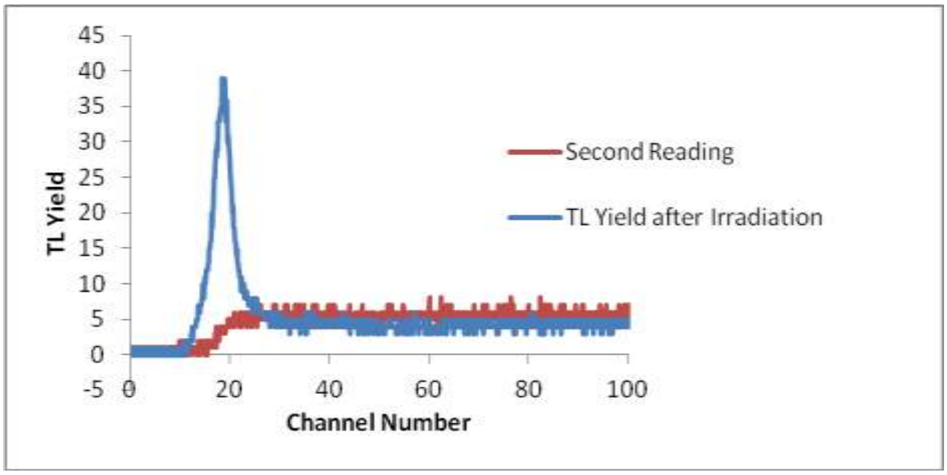

Figure 8: The blue line illustrates thermoluminescence from pure SWCNT buckypaper irradiated to a dose of 2 Gy using a beta source ^{90}Sr; the red line illustrates the signal obtained on second reading.

3.3.- Super-Pure Single-Wall Carbon Nanotubes

Figure 9 shows the TL yield for a fresh super-pure SWCNT buckypaper sample.

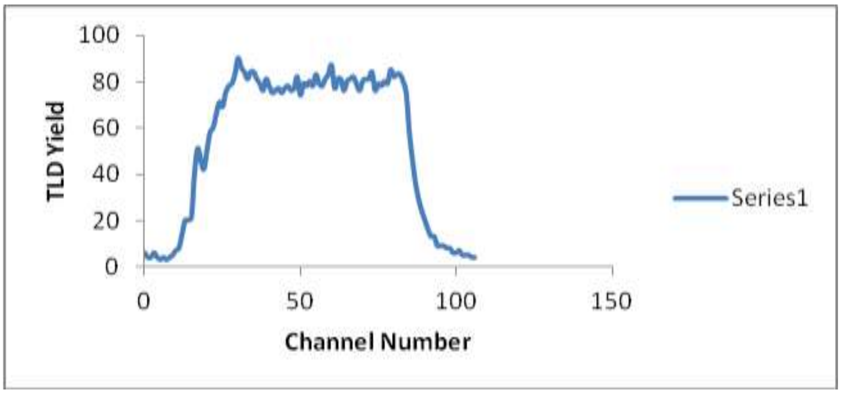

Figure 9: Thermoluminescence, before irradiation, from buckypaper made of 0.05g of Super-Pure Single–Wall Carbon Nanotubes dispersed in 100ml of deionized water through the use of Triton X-100. The sample size was 5 × 5mm.

The sample consists of buckypaper made of 0.05g of Super-Pure Single–Wall Carbon Nanotubes dispersed in 100ml of deionized water through the use of Triton X-100. The sample size was again 5 × 5 mm and again the sample was irradiated to a dose of 2 Gy using the ^{90}Sr beta source. The graph is multi-peaked for the non-irradiated sample as obtained from raw and pure SWCNT buckypapers. However, this sample provides a higher TL yield than the pure SWCNT sample, being almost similar to the TL yield obtained from the raw SWCNT sample. This was not expected as the sample is super purified from impurities. However, as nitrogen is used in the process of purification of the samples, this increase is thought to be due to contamination during that process, the nitrogen atoms potentially causing increase in the trap centres in the sample.

Figure 10 shows two overlapping graphs, the blue line illustrating the TL yield from the super-pure SWCNT sample after exposure to 2 Gy using the ^{90}Sr source, and the red line illustrating the second reading for the same sample. The TL yield from the super-pure sample is higher than the TL yield from the pure sample by factor of 2.2. However, by visual examination of the super pure sample we found that it has a rough surface compared to the raw and pure samples. This invites question about the kind of trapping centres or defects that have been created.

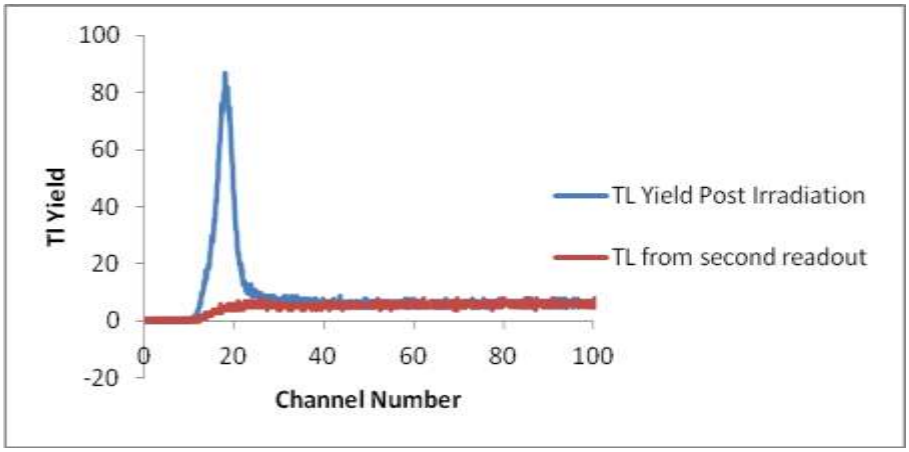

Figure 10: The blue line illustrates the TL yield from the super-pure SWCNT sample after exposure delivering a dose of 2 Gy using the ^{90}Sr source, and the red line illustrates the second reading for the same sample.

3.4.- TLD-100

Comparison was made of the response of SWCNTs buckypaper sample with TLD-100 formed from lithium fluoride. It is clear that the response of TLD-100 is very much higher compared to the response from the SWCNT samples; the graph in Figure 11 illustrates the TL yield from the TLD-100 sample. The peak in this graph is not shown do to the high sensitivity setting used for the TLD Reader, leading to clipping.

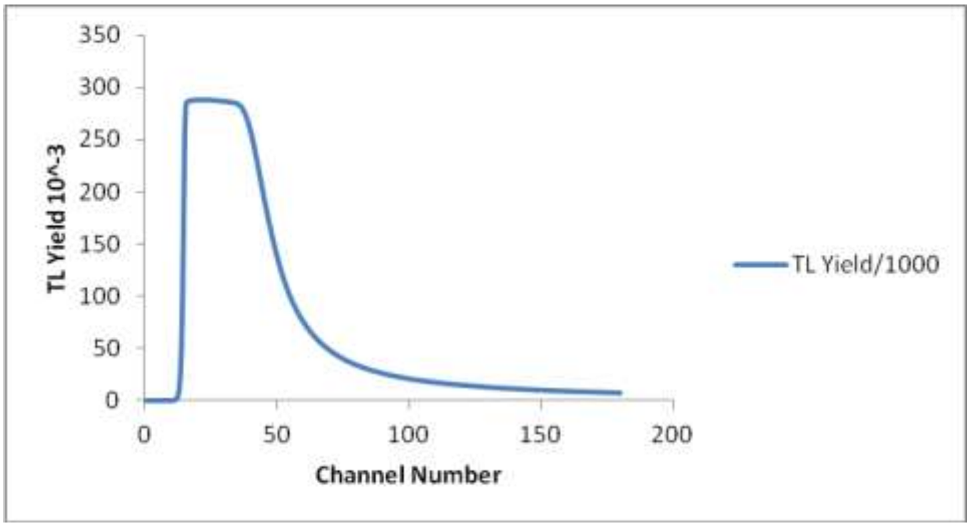

Figure 11: TL yield from TLD-100 irradiated with 2 Gy using the ^{90}Sr source, the TL scale is normalised by dividing by 1000.

While the low TL response of CNT compared against TLD-100 would seem to be disadvantageous, it does also suggest that CNTs can tolerate higher levels of radiation for high level radiation dosimetry as in radiation processing and sterilization.

4.- CONCLUSION

Three types of SWCNT buckypaper, each of different impurity, were prepared for irradiation using different sources. TLD-100 was used for comparison. The samples were irradiated using a ^{90}Sr source. The TL yield signal from the sample irradiated by the ^{90}Sr beta source indicates that the response of the buckypaper is very much less than the response obtained from TLD-100. The results suggest that CNTs can tolerate higher levels of radiation dose and can be used for elevated dose dosimetry as in radiation processing and sterilization or in space radiation medicine applications.

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