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THE EFFECT OF MULTI-WALL CARBON NANOTUBES ADDITION ON THE SHIELDING PROPERTIES AGAINST GAMMA RADIATION†

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In this work, the effect of Multi-Wall Carbon Nanotubes (MWCNTs) addition on the materials shielding properties against Gamma radiation with an energy of 662 keV from a ^{137}Cs source is investigated. The linear attenuation coefficient of MWCNTs-based materials (gelatin-water mixture) with MWCNTs concentrations of 0%, 5%, and 10% is measured. To isolate the contribution of the MWCNTs unique structure to the shielding capabilities, samples with the same concentrations of activated carbon were fabricated and their linear attenuation coefficients were obtained. Also, the linear and the mass attenuation coefficients are obtained theoretically for the same concentrations using the XCOM program and compared with measured values. It is found that the addition of MWCNTs by 5% or 10% has increased the linear attenuation coefficient by around 5% when compared to the same concentrations of activated carbon. This increase in the shielding capabilities against gamma radiation can be related to the interaction of gamma radiation with the extraordinary geometry and structure of MWCNTs.

Keywords: *Radiation; Shielding; Attenuation coefficient; XCOM; Multi-wall carbon Nanotubes*

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1. INTRODUCTION

Carbon nanotubes (CNTs), because of their outstanding physical and geometrical properties are considered one of the key materials for many of the current and future technologies [1, 2, 3, 4]. So far, there is a large number of studies focusing on the electrical, thermal, and mechanical properties of CNTs-based composites targeting vast technological applications [5, 6]. However, when it comes to the interaction of gamma (γ) radiation with CNTs based composites there is a large number of studies that explore the effect of γ irradiation on the electrical and mechanical properties [7, 8]. Nevertheless, focusing on the shielding properties of CNTs-based composites against γ radiation has drawn less attention [9, 10]. One reason for this is related to the fact that shielding capability against radiation is mainly directly proportional to the atomic numbers of the elements in the used shielding material and their mass density [11]. However, engineering the shielding materials at the nano-scale level has proven to enhance the shielding properties in some applications. For example, in previous studies by Hassan et al. as in Ref.[12] and by El-Khatib et al. as in Ref.[13], aiming to fabricate better gamma radiation shields, materials (e.g., concrete, polymers) were doped with lead oxide nano-particles to enhance the shielding properties due to the existence of the lead (Pb) element. In fact, lead is known as one of the best elements to shield gamma radiation due to its large atomic number and its high mass density. In the X-ray range, in a work by Fujimori et al. [9], the shielding properties against X-ray radiation were enhanced when CNTs are used in comparison with other forms of carbon structures in highly oriented pyrolytic graphite (HOPG) and fullerenes (C_{60}). This enhancement in the shielding properties could not be explained by the known theories, which raises the need for an alternative theoretical model to explain such enhancement. In a study by Zhang et al.[14], the shielding capabilities of carbon nanotube (CNT)-based film materials against gamma-ray are investigated for energies from ^{241}Am and ^{137}Cs. The study shows that CNT films have higher shielding capabilities against gamma radiation when compared with carbon fiber-reinforced composites, owing to the interaction of gamma radiation with the outstanding cylindrical structure on the nanoscale. In another work by Viegas et al.[15], an enhancement of X-ray shielding in the functionalized graphene oxide-based nanocomposites when compared to MWCNTs is reported for energies between 6.9 to 22.1 keV. The study showed that the structure of carbon used in the composite is an essential parameter to consider when studying the shielding properties. Moreover, the work has pointed out that a new interaction mechanism between the graphene structure and the incident γ-ray could be responsible for such enhancement.

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Since most studies did not focus on the shielding against gamma radiation when CNTs are utilized at least in the bulk case, it's worth searching for potential contributions to the shielding capability against gamma radiation due to the unique geometrical and structural properties of CNTs. Recently, there is an increasing interest in the medical applications of CNTs for treating cancer by dielectric heating of infected tissues by the application of microwave radiation [16]. In special cases, if gamma radiation therapy is used during the existence of CNTs in the tissue, it is important to estimate the shielding level of CNTs to give the correct dose of γ radiation [17].

When studying the shielding capability of any material after the addition of CNTs, there are key parameters to consider in such study, e.g., the geometry of CNTs (number of walls, diameter, and length), alignment and orientation of CNTs, and their concentration in the hosting material. Another important parameter to consider is the energy of the gamma radiation E_γ used, where E_γ is related to the wavelength λ of γ-ray as $E_\gamma = hc/\lambda$ where h is the Planck's constant and c is the speed of light [11]. In the current study, we are considering the concentration of MWCNTs parameter only which can be investigated within the available resources.

To investigate the effect of MWCNTs addition on the radiation shielding properties, the linear attenuation coefficient (LAC) will be measured for a gelatin-water mixture that is doped with different concentrations of MWCNTs against γ-radiation from ^{137}Cs at 662 keV. Activated carbon is used in the current study as a control variable (in terms of structure) to isolate the effect of the unique structure of MWCNTs at such conditions. The chemical structure of activated carbon is very close to pure graphite structure in which carbon atoms are arranged as layers of connected hexagons [18]. To help understand the effect of MWCNTs addition on the shielding properties, the XCOM program will be used to calculate the mass attenuation coefficient μ values and the LAC values theoretically [19]. The importance of the XCOM program in the current study stems from the fact that the XCOM program does not account for the structure of the shielding materials in general and for the MWCNTs structure specifically. Such calculations can help isolate the contribution of the MWCNTs structure when compared with the experimental measurements of the shielding properties.

One more important goal for the current work is to verify the need for a new theory to explain the mechanism of interaction between the γ radiation and the MWCNTs at such an energy range. For the γ rays at an energy of 662 keV from the ^{137}Cs, the dominant mechanism of interaction is the Compton effect at which the γ rays scattered at different angles θ with less energy (depending on the angle) [20].

2. THEORY

For a radiation of a single energy γ radiation with an intensity of I_0 initially incident on the surface of a material that has a thickness of x, the intensity of the gamma radiation after traveling a distance x inside the material will drop according to the exponential attenuation law (Lambert-Beer law) as [21]:

$$I(x) = I_0 e^{-LACx} \tag{1}$$

Here LAC represents the linear attenuation coefficient in units of cm^{-1}. For a shielding material with a large LAC value, the intensity of γ-rays gets attenuated dramatically as it travels through the material [21]. The total mass attenuation coefficient μ for any shielding material is given as $\mu = LAC/\rho$, where ρ is the mass density of the sample in units of g/cm^3. For any chemical compound or a mixture of elements the μ value can be calculated theoretically using the following relation:

$$\mu = \sum_i w_i (LAC/\rho)_i \tag{2}$$

where the fractional weight of the i^{th} component is w_i. The value of μ can be obtained theoretically using the XCOM program that is provided by the National institute of standards NIST (USA) [19]. Also, one can find the Half Value Thickness (HVT), which is the thickness of the shielding material at which the radiation intensity drops to one-half of its initial value, it can be calculated according to the following equation:

$$HVT = \frac{\ln 2}{LAC} \tag{3}$$

The HVT value is a key parameter to consider when comparing the shielding capability of different radiation-absorbing materials [22].

3. EXPERIMENTAL SETUP

3.1. Fabrication of the samples

In the current study, a mixture of gelatin and water was prepared as a hosting material for the MWCNTs due to the simplicity of casting the samples and due to the good dispersion of the MWCNTs into the mixture after curing [16]. The host material mixture is fabricated out of gelatin (gelatin from bovine skin type B, Sigma-Aldrich) and distilled water, as demonstrated in a work by Altarawneh et al.[23]. A small amount of p-toluic acid and n-propanol were added to the distilled water and gelatin to enhance the bonding in the host material and to preserve it for longer periods of time [24]. Table 1 shows the percentage of the elements composing the host material used in this work. The MWCNTs (supplied by Cheap Tubes Co., with a length of 10-20 μm, purity $>$ 95 wt.% outer diameter 30-50 nm, and ash$<$1.5 wt.%) or the activated carbon (supplied by Acros Organics BVBA Janssen Pharmaceutical, with grains size $>$1 μm) of the desired concentrations were added to the distilled water initially and sonicated using an ultrasonic processor (UP100H- Hielscher Ultrasonics technology) for 30 minutes. Then, the gelatin and the rest of the components were added according to the procedure described in Altarawneh et al.[23]. The samples containing the MWCNTs or the activated carbon were fabricated with concentrations by weight of 0, 5%, and 10% for each of them.

Table 1. The percentage of the main components of the hosting material mixture

p-toluic acid	n-propanol	deionized water	gelatin
0.080%	3.44%	81.84%	14.64%

The resultant mixtures with different concentrations and different fillers types were molded in 10 cm long glass tubes with a diameter of 1.2 cm. The glass tubes containing the samples were sealed with thin plastic sheets and left to cure at room temperature for five days. The final form of the samples is flexible cylindrical rods that can be easily cut by a sharp blade. The mass density for each sample was measured and tabulated to be used in the rest of the study. For each of the fabricated samples, the LAC value was measured as demonstrated in the following subsection.

3.2. Experimental setup for studying Gamma radiation shielding capabilities

The setup used in this work to measure the LAC values is as demonstrated in Figure 1. The setup is composed of a Sodium Iodide (NaI) detector (manufactured by Canberra, model: 2007P) that is housed in a 4.0 *cm* thickness lead shield to block radiation from background sources.

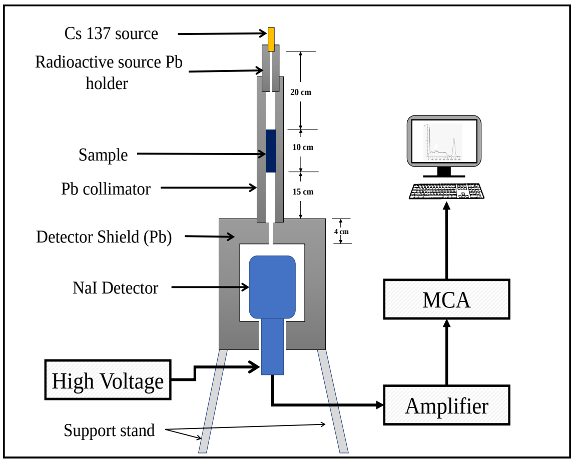

Figure 1. The schematic diagram of the setup used to measure the LAC values of each of the fabricated samples.

The NaI detector was connected to an operating high voltage source of 700V (manufactured by Canberra, model: 3102). The output signal was sent to an amplifier (manufactured by Ortec, model: 575A) where its output is connected to a Multi-Channel Analyzer (MCA). The data from the MCA was acquired and plotted on a computer monitor with the help of the MAESTRO Version 7.0 package (supplied by Ortec).

The fabricated samples were inserted in the custom-made lead housing with a 1.2 cm internal diameter, 40.0 cm length, and 2.0 cm thickness. The detector was placed at a large enough distance to ensure narrow beam geometry. The Cesium source (^{137}Cs) has a pen shape with an outer diameter of 1.0 cm (supplied by Amersham) that was inserted in a custom-made lead holder with a length of 8.0 cm. The sample thickness was adjusted by pushing the sample out of the glass tube and by cutting it with a sharp blade. Inserting a small amount of oil between the glass tube and the sample can help slide the sample in and out of the glass tube during the adjustment of the thickness.

The fabricated samples were irradiated by gamma radiation from the ^{137}Cs source where the intensity was collected for a time interval of 90 minutes. The values of the *LAC* were obtained by fitting Eq.1 using Igor Pro 9 data analysis software (provided by Wavemetrics). Suitable background subtraction was conducted for all collected intensity [25, 26].

3.3. Calculations of the mass attenuation coefficients using the XCOM program

In the theoretical part of this work, the XCOM program is used to calculate the mass attenuation coefficients of the fabricated samples at energies from 1 keV to 100 GeV. The XCOM program runs under the Windows operating system on the Google Chrome web browser [27]. The elements percentages in each sample were calculated initially and used in the XCOM program to calculate the mass attenuation coefficient for each sample. In Table 2, the net percentages of the elements (carbon, oxygen, hydrogen, and nitrogen) making up each of the fabricated samples were calculated.

Table 2. The elements percentage of the 0% host material, for the 5% additives of carbon, and for 10% additives of carbon either in the activated carbon form or the MWCNTs form.

% of carbon additives	% C	% H	% N	% O
0%	9.54	10.62	2.48	77.29
5%	13.82	10.11	2.36	73.63
10%	17.88	9.64	2.25	70.17

The output data generated by the XCOM program is tabulated and plotted for comparison. The theoretical mass attenuation coefficient values are found for the fabricated samples using the XCOM program and plotted as a function of energy as in Figure 2.

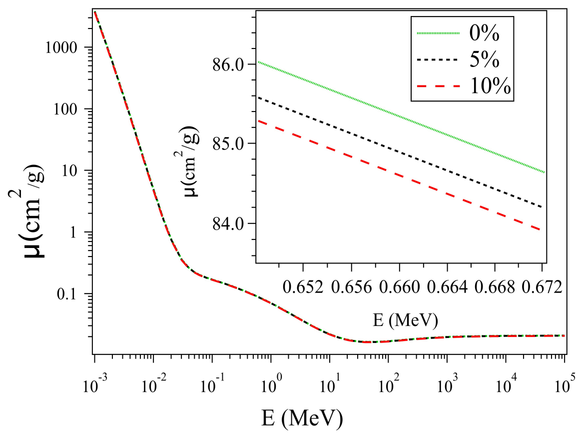

Figure 2. The mass attenuation coefficients as calculated by the XCOM program for the samples with 0%, 5%, and 10% of carbon additives. The inset shows the values of μ around the ^{137}Cs energy of 0.662 MeV.

4. RESULTS AND DISCUSSION

The gamma radiation intensity I from the ^{137}Cs source at 662 keV was collected using the setup described earlier for different thicknesses of gelatin-water mixtures with no additives (0%), with the addition of MWCNTs (5% and 10%), and with the addition of activated carbon (5% and 10%). After suitable background subtraction, the intensities I for each of the five samples were plotted as a function of the sample's thickness, and the values of the LAC were obtained by fitting Eq.1. As it can be seen in Table 3 and in Figure 3-a., there is a small decrease in the measured LAC values from 0.089 cm^{-1} down to 0.085 cm^{-1} and 0.087 cm^{-1} when the gelatin-water mixture was doped with 5 % and 10% of activated carbon respectively.

Table 3. The Measured and calculated parameters related to the attenuation of gamma radiation for 0%, 5%, and 10% concentrations of carbon nanotubes and activated carbon.

	Host Material	Activated carbon	MWCNT	Activated carbon	MWCNT
concentration	0	5%	5%	10%	10%
LAC (cm^{-1})	0.089 ± 0.001	0.085± 0.001	0.089 ± 0.001	0.087 ± 0.001	0.091± 0.001
HVT (cm)	7.79± 0.09	8.16± 0.09	7.79± 0.09	7.97± 0.09	7.62± 0.08
ρ (g/cm^3)	1.06 ± 0.01	1.08± 0.01	1.09 ± 0.01	1.10 ± 0.01	1.12± 0.01
μ(cm^2/g) exp	0.084±0.001	0.079±0.001	0.082±0.001	0.079±0.001	0.081±0.001
μ(cm^2/g) XCOM	0.0854	0.0850	0.0850	0.0846	0.0846

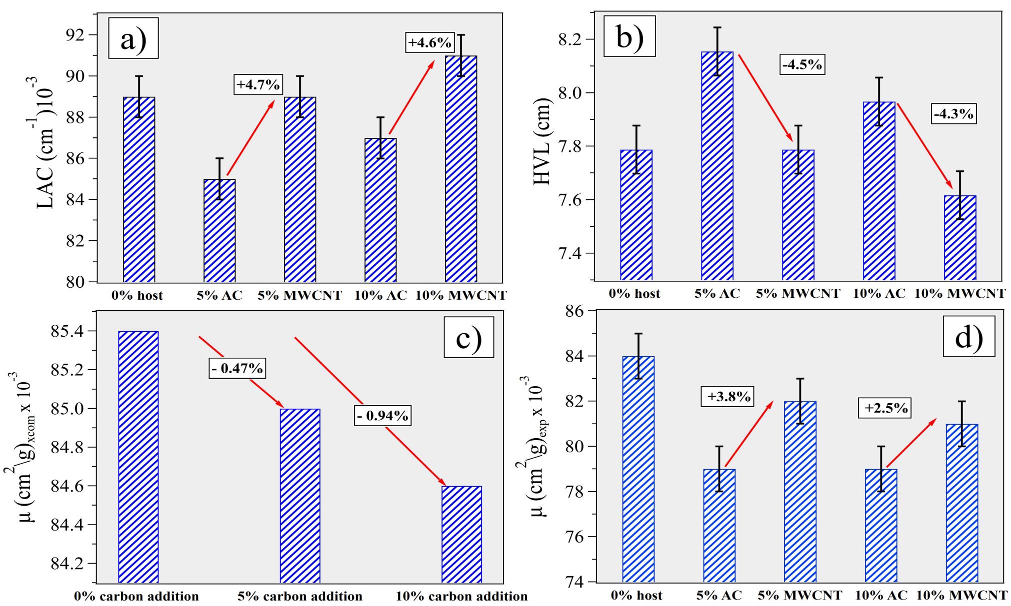

Figure 3. a) the LAC values for all the samples in this study, b) the HVT, c) the μ values as obtained by XCOM, and d) comparison between values of μ from the XCOM program and the experimental values for the different types and concentrations of carbon in the study.

In Figure 3-a, the decrease in the LAC values after the addition of activated carbon can be attributed to the fact that the LAC value is less for materials that are composed of elements of smaller atomic numbers. Particularly, increasing the concentration of carbon atoms in the sample results in reducing the concentrations of the rest of the elements in the sample like oxygen and nitrogen which each has slightly a larger atomic number (see Table 2 for the concentrations of the elements). However, the addition of MWCNTs to the gelatin-water mixture (by 5% and 10%) results in an increase of about $\approx$ 4.7% and 4.6% in the LAC values relative to the gelatin-water mixture with 5% and 10% of activated carbon respectively. Since the concentration of carbon (in the two forms, as activated carbon or MWCNTs) in the compared samples is the same, the extraordinary structure of the MWCNTs is the only factor that can be responsible for such an increase in the LAC values.

The HVT value of the gelatin-water mixture after the addition of 5% and 10% of MWCNTs decreases with 4.5% and 4.3 % respectively when compared with the addition of activated carbon as demonstrated in Figure 3-b. Such a decrease in the HVT is due to the extra shielding from the interaction between the γ radiation and the MWCNTs' cylindrical geometry. The increase in the HVT value after the addition of activated carbon is justified by the increase of carbon percentage in the mixture relative to oxygen and nitrogen.

The theoretical mass attenuation coefficients μ for the gelatin-water mixture with the addition of 0%, 5%, and 10% carbon obtained using the XCOM program were presented in figure 3-c. It's evident in figure 3-c that the gelatin-water mixture has less μ as the concentration of carbon is increased. However, the current experimental results show that the addition of carbon in the MWCNTs form has the opposite effect as demonstrated in Figure 3-d. Particularly, while the addition of the activated carbon (for both 5% and 10% concentrations) decreases the values of μ, the addition of MWCNTs increases the values of μ for the same concentrations. Based on this, it is clear that MWCNTs due to their unique structure add extra shielding strength to the gelatin-water mixture.

Since the MWCNTs geometry and structure appear to be the only factors enhancing the shielding properties, we should try to find a connection between the physical properties of the γ radiation and the main features of the MWCNTs structure and geometry. It's noteworthy that the wavelength of the γ-rays used in this study at an energy of 0.662 MeV is around 1.87pm, where the interlayer spacing between the walls of the MWCNTs is around 0.35 nm [28]. In this case, the ratio between the wavelength of γ-ray and interlayer spacing size is around 1:200. For such a small ratio between the wavelength and the interlayer spacing, one would expect less interaction between the γ-rays and the MWCNTs. The ratio between the wavelength of the X-ray and the interlayer spacing between the walls of MWCNTs can be calculated as 1:20 in Fujimori et al. work [9], where the used X-ray wavelength was 7.11 nm and the spacing between MWCNTs walls was 0.35 nm. The ratio in both cases can be correlated with the shielding capabilities where a stronger interaction is expected for the X-rays case. Based on the values of the two ratios above, it can be concluded as the ratio between the photon wavelength and the interlayer spacing is close to 1:1, the shielding capability against electromagnetic radiation would be stronger. This can be observed in the achieved shielding enhancement due to the addition of MWCNTs against the X-ray radiation (50% enhancement, as in Ref.[9]) compared to the shielding enhancement against the γ-rays shielding in the current work (5% enhancement) when MWCNTs are used.

5. CONCLUSIONS

In the current study, the *LAC* values for the gelatin-water mixture, for the gelatin-water mixture with activated carbon addition (5% and 10%), and for MWCNTs addition (5% and 10%) are investigated experimentally and theoretically. In contrast to the theoretical prediction of the XCOM program of a decrease in the LAC values, the addition of MWCNTs has increased the LAC values by 5% when compared to the addition of activated carbon. The main reason for such an increase is only explained by the interaction of γ radiation with the unique structure of MWCNTs when compared with the activated carbon. It is recommended to use the MWCNTs to enhance the shielding properties in the X-ray range rather than in the γ-ray range due to stronger interaction when the wavelength of the radiation is close to the interlayer spacing of the MWCNTs. In the medical fields, if MWCNTs are introduced to enhance heating effects in living tissues due to the application of microwaves radiation as many studies have proposed [17], γ radiation therapy can be used too on the tissue without the need to dramatically increase the radiation doses.

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ВПЛИВ ДОДАВАННЯ БАГАТОСТІННИХ ВУГЛЕЦЕВИХ НАНОТРУБОК НА ВЛАСТИВОСТІ ЕКРАНУВАННЯ ВІД ГАММА-ВИПРОМІНЮВАННЯ

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У цій роботі розглядається вплив додавання багатостінних вуглецевих нанотрубок (MWCNT) на екрануючі властивості матеріалів. Досліджено гамма-випромінювання з енергією 662 кеВ від джерела ^{137}Cs. Виміряно коефіцієнт лінійного загасання матеріалів на основі MWCNT (суміш желатин-вода) з концентрацією MWCNT 0%, 5% і 10%. Щоб виділити внесок унікальної структури MWCNT в екрануючі можливості, були виготовлені зразки з такою ж концентрацією активованого вугілля та отримані їхні лінійні коефіцієнти ослаблення. Крім того, лінійний і масовий коефіцієнти ослаблення отримані теоретично для однакових концентрацій за допомогою XCOM програми та порівняно із виміряними значеннями. Виявлено, що додавання MWCNTs на 5% або 10% збільшило лінійний коефіцієнт ослаблення приблизно на 5% у порівнянні з тими ж концентраціями активованого вугілля. Це збільшення можливостей екранування від гамма-випромінювання може бути пов'язане із взаємодією гамма-випромінювання з геометрією та структурою MWCNTs.

Ключові слова: *радіація; екранування; коефіцієнт ослаблення; XCOM; багатостінні вуглецеві нанотрубки*