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Review Article

Gamma Radiation Effect on Carbon Nanotubes

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ABSTRACT

An extensive review of the gamma radiation effect on carbon nanotubes is given in this study. The interaction of carbon nanotubes with different doses and energies gamma radiation has been studied in recent years. Carbon nanotubes are desirable materials in technological applications because of their extra features such as good thermal properties, ultra-light structures, different conductivity properties, durability, and superior heat resistance. For these reasons, they are used extensively in device construction. These devices are used extensively in environments exposed to radiation such as medicine, aviation, nuclear reactors, nuclear waste storage. Knowing the response of carbon nanotube materials to radiation is very important for the stability of the devices made. As can be seen from the literature review, the interaction of the material with gamma radiation is quite variable according to the type of material, the purity, and the atomic lattice of material, dose and energy of the applied radiation and the environment (water, air, etc.) subjected to.

Keywords: *Gamma radiation effects, Carbon nanotubes, Defect formation, Structure healing, Device stability*

Karbon Nanotüpler Üzerine Gama Radyasyonu Etkisi

ÖZET

Bu çalışmada, karbon nanotüpler üzerindeki gama ışınımının etkisi kapsamlı bir şekilde derlenmiştir. Karbon nanotüplerin farklı dozlarda ve enerjilerde gama radyasyonu ile etkileşimi son yıllarda yoğun olarak incelenmiştir. Karbon nanotüpler, iyi termal özellikler, ultra hafif yapılar, farklı iletkenlik özellikleri, dayanıklılık ve üstün ısı direnci gibi ekstra özelliklerinden dolayı teknolojik uygulamalarda arzu edilen malzemelerdir. Bu nedenlerden dolayı, teknolojik cihaz yapımında yaygın olarak kullanılırlar. Bu cihazlar, tıp, havacılık, nükleer reaktörler, nükleer atık depoları gibi radyasyona maruz kalan ortamlarda kullanılmaktadır. Karbon nanotüp malzemelerinin radyasyona tepkisini bilmek, yapılan cihazların kararlılığı için çok önemlidir. Literatür taramasından görülebileceği gibi, malzemenin gama radyasyonu ile etkileşimi, malzemenin türüne, malzemenin saflığına ve atom örgüsüne, uygulanan radyasyonun dozuna, enerjisine ve uygulama ortamına (su, hava vb.) göre oldukça değişkendir.

Anahtar Kelimeler: *Gama radyasyonu etkileri, Karbon nanotüpleri, Kusur oluşumu, Yapı iyileşmesi, Cihaz kararlılığı*

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I. INTRODUCTION

Today carbon is a very popular research subject because of its versatile uses. It has many various features which can be used in different applications depending on how the carbon atoms are arranged [1]. In the design of carbon atoms, the interaction of carbon nanotubes with different radiation sources is quite important. When the studies examined carbon nanotubes, which interact with the radiation; the quality parameters, electrical characteristics, morphology and crystalline structure of carbon nanotubes the significant changes were detected [2-10]. Important physical and chemical properties such as thermal and electrical conductivity in carbon-based materials are subjected to significant changes with the change of formation of atoms [3]. These changes are quite crucial for the carbon nanotubes in areas exposed to the radiation. Aerospace work and medical applications have a high level of radiation exposure. Due to their sensitive, lightweight, rugged and compact properties, carbon nanotubes are also used in radiation detectors developed to observe and characterize radiation environments [11] Sensors are used in the control of radiation-emitting environments as well as in all technological applications. Carbon nanotubes are used extensively in the construction of a small and robust sensors due to their large surface area, high surface volume ratios, and extraordinary electrical and chemical properties [12]. They can be used in several areas of biomedical engineering [13]. The construction of a small, high-function device is one of the main goals of today's scientific world in all kinds of technological fields we can think of. Such studies are available in many fields such as medicine, aviation, nuclear energy, computer, nuclear scanning and detection, bio-engineering applications, bio-physics applications, pharmacy, in-body micro-robotic studies. Carbon nanotubes have desirable properties in technological applications because of their properties such as being very light, having excellent electrical and mechanical properties, exhibiting a very large surface area in small volume, thermal durability. Stability is the most important and vital point for all devices operating in radiation environments. This is an indispensable necessity for human and environmental health. This study focuses on the existing literature on the interaction of gamma radiation with carbon nanotube structures. Gamma rays are formed due to the change in the energy levels of an atomic nucleus. This electromagnetic wave has the highest energy. They occur as a result of nuclear reactions. They have ionizing properties. Super-nova bursts, radioactive distortions in space, neutron stars, black holes, and active stars are the source of gamma light. Gamma rays are a penetrating x-ray type. It is used extensively in imaging and follow-up processes in medical applications [14]. Due to the natural sources of gamma radiation are also intense, their interaction with these structures is important. It is possible to monitor the changes caused by different kinds of radiation on carbon nanotubes with today's physical and chemical analysis methods. The most commonly used spectroscopic and microscopic techniques for the investigation of carbon-based structures are Raman spectroscopy, X-Ray Diffraction (XRD), Field-Emission Transmission Electron Microscope (FETEM), Fourier Transform Infrared Spectroscopy (FTIR), Transmission Electron Microscopy (TEM), Scanning Electron Microscope (SEM), X-Ray Photoelectron Spectroscopy (XPS), X-ray Auger-electron spectroscopy (XAES). When the current literature is examined, it is observed that the interaction of all types of carbon-based structures and gamma rays is examined intensively.

Graphite and multi-walled carbon nanotubes (MWCNTs) irradiated by gamma-rays were examined X-ray diffraction, Raman spectroscopy, and transmission electron microscopy, It was observed that gamma-irradiation improve the structural arrangement of graphite. The resulting effect is altering with the amount of irradiation dose. 200 kGy dose is most effective. When the irradiation dose exceeds 200 kGy the defect dynamics change and the nanotubes are annealed. Due to defects occurring in the carbon atom lattice, gamma-irradiation deteriorates nanotubes graphitization degree. After gamma-irradiation, Structural differences between graphite and MWCNTs are increasing. I_D/I_G ratio is a quality parameter for the carbon structures. The rate is determined from the measurements obtained from Raman spectroscopy. MWCNTs damaged with gamma-irradiation examined with XRD (results in Figure 1), and Raman spectroscopy (results in Figure 2). These figures reveal the occurring defect in the structure of the MWCNTs. At 200 kGy, they were destroyed most severely. FETEM images (in Figure 3) presented these deformations intuitively [15].

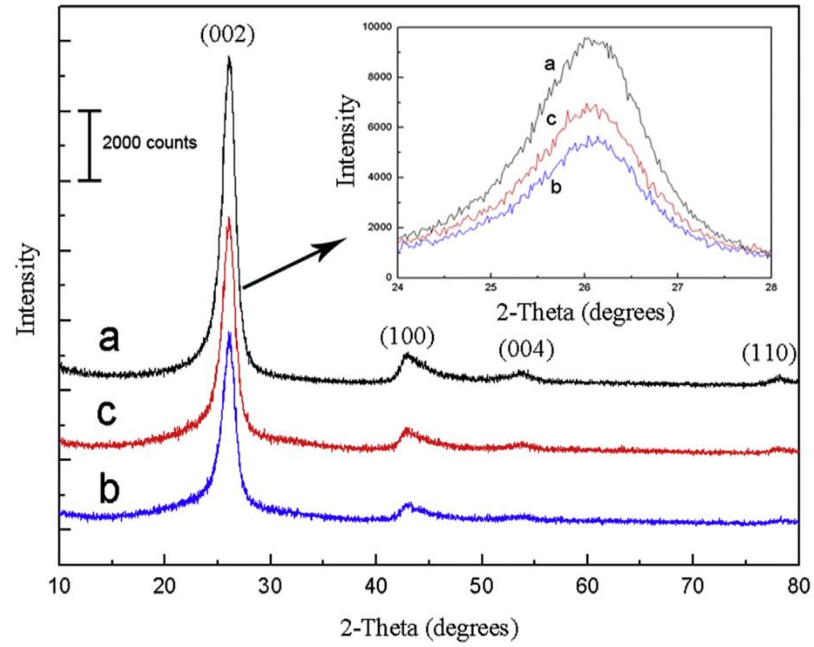

Figure 1. *XRD graphic for (a) virgin MWCNTs, (b) irradiated MWCNTs with 200 kGy dose and (c) irradiated MWCNTs with 2 MGy dose. The inset graphic represent the interval from 24 to 28 (2θ) [15].*

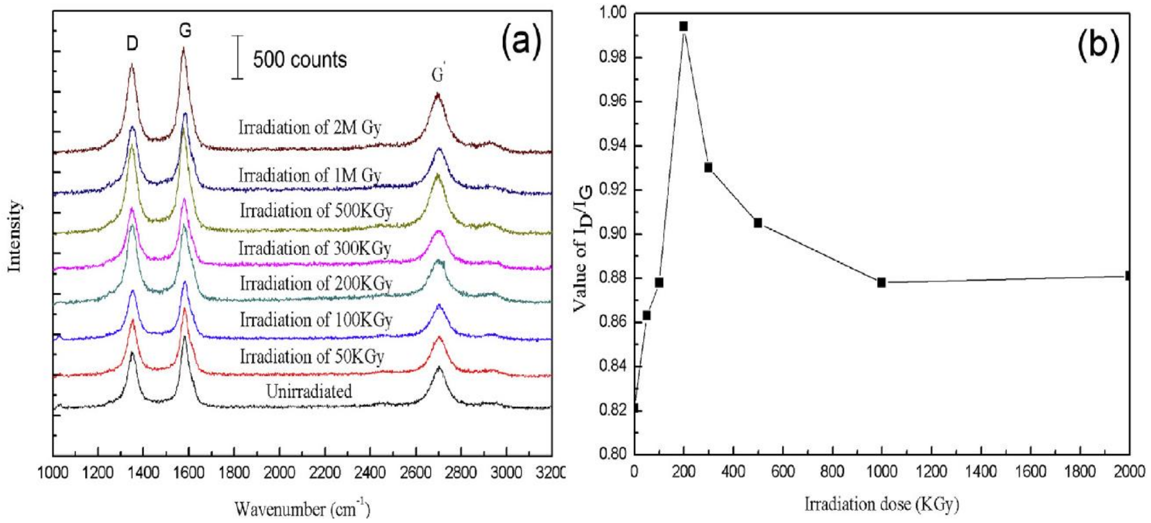

Figure 2. *Raman spectroscopy obtained from virgin and gamma-irradiated MWCNTs (a) varying I_D/I_G ratio depending on radiation dose (b) [15].*

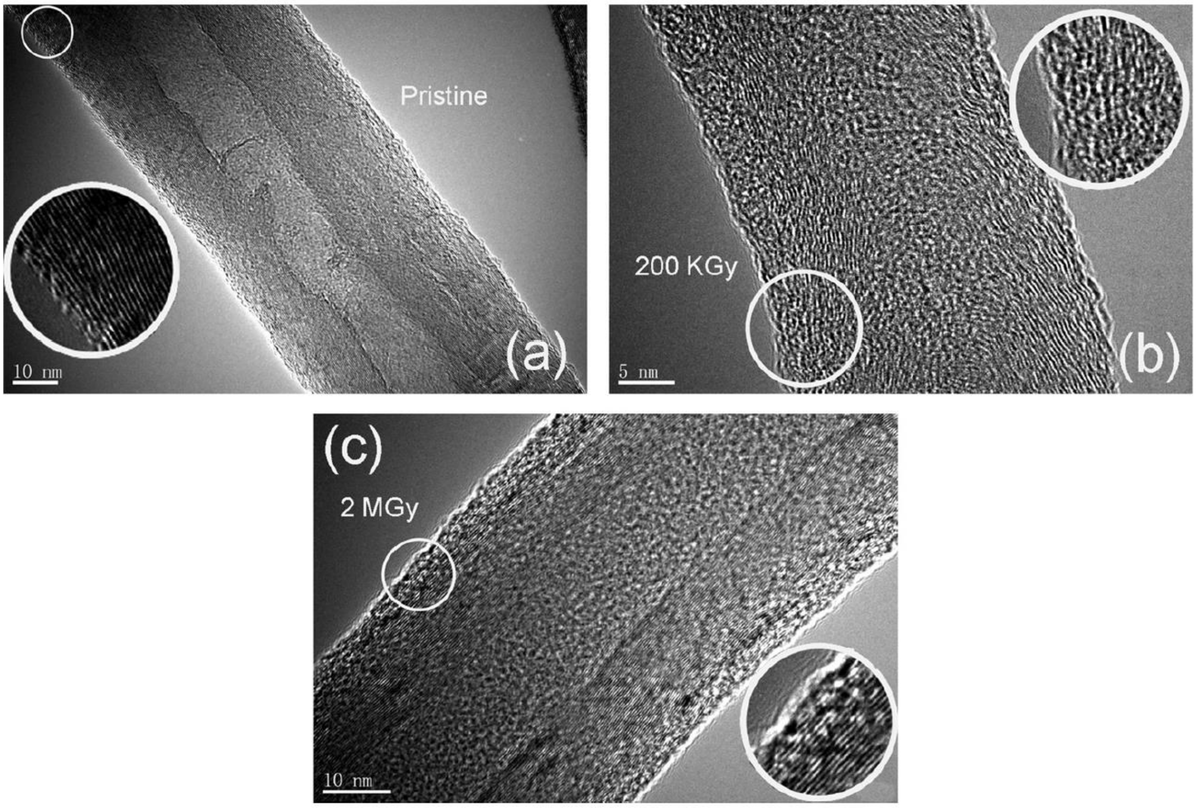

Figure 3. *FETEM images for (a) pristine and various dose irradiated MWCNTs: (b) 200 kGy; (c) 2 MGy [15].*

MWCNTs irradiated at lower doses gave different responses to gamma-rays. Raman, XPS, SEM, TEM results of non-irradiated samples and the samples were subjected to 10 kGy and 20 kGy gamma doses were examined. Defects and amorphous formations were observed in the sample irradiated with 20 kGy, whereas structural improvement and defect reduction were observed in the sample irradiated with 10 kGy. The I_D/I_G ratio obtained from Raman spectroscopy is considered as the quality parameter of carbon structures (in Table 1). This ratio is low, which indicates an increase in the quality of the building, and an increase indicates a defect formation. When the Compton profile of the sample exposed to 10 kGy gamma ray was examined, it was found that there was a slight change in the Compton profile distribution compared to the non-irradiated sample. It is thought that this change does not have an effect on the electronic properties of the structure. It is a remarkable result in terms of the stability of the material [16, 17].

Table 1. *D-band, G-band and I_D/I_G ratios values for virgin and the irradiated with gamma-rays in MWCNTs [16].*

MWCNT Samples	D-band (cm^{-1})	G-band (cm^{-1})	I_D/I_G ratios
virgin	1539.5	1571.3	0.92
Irradiated at 10 kGy	1343.5	1570.6	0.88
Irradiated at 20 kGy	1347.6	1580.9	0.94

Carbon-based structures are used to develop a new generation of lightweight materials needed in space electronics. By injecting CNTs into macroscopic fibers or sheets, many problems that these materials can create in technological applications are solved. Since aviation and space research are areas with intense radiation presence, the response of gamma radiation to these CNTs yarns structures having sensitive and vital application areas has been investigated. The strength and elasticity modulus of CNTs yarns exposed to 100 kGy gamma radiation increased slightly compared to the virgin sample. The same increase was observed in Young's modulus. The positive answers of the material are remarkable [18].

Single-walled carbon nanotubes, another form of carbon-based structures, are used in industrial applications. The interaction of this form with radiation has been studied. The effect of gamma irradiation on single-walled carbon nanotube (SWCNT) was carried out. Gamma irradiation was performed on nanotubes in air, water, and aqueous ammonia. After irradiation, the samples were evaluated by using Thermogravimetric analysis (TGA), Fourier transforms infrared spectroscopy (FTIR), elemental analysis (EA) and Raman spectroscopy to observe the changes in the SWCNT mesh. Looking at the results of TGA measurements, with 100 kGy doses it can be seen the highest percentage of introduced groups in SWCNTs structure. The attachment of hydroxyl, carboxyl and nitrile functional groups to the SWCNT sidewalls can be observed by looking at FTIR spectroscopy results expressed in Figure 4. EA results confirm the presence of these groups. Hydroxyl and carboxyl groups' formations were observed in all medium after irradiation. But nitrile functional groups were only identified in irradiated SWCNTs in aqueous ammonia. As can be seen from Figure 5 and 6, Raman spectroscopy results illustrated a correlation between irradiation dose and defect degree in the carbon nanotube structure. When administered the dose of 100 kGy to nanotubes, Raman I_D/I_G ratio was three times higher than for the pristine ones. After 100 kGy doses, nanotube length decreased by 50%. Atomic force microscopy demonstrated this decrease. Through the transmission electron microscopes results expressed in Figure 7, it can be seen remarkable changes in the morphology and structure of irradiated SWCNTs [6].

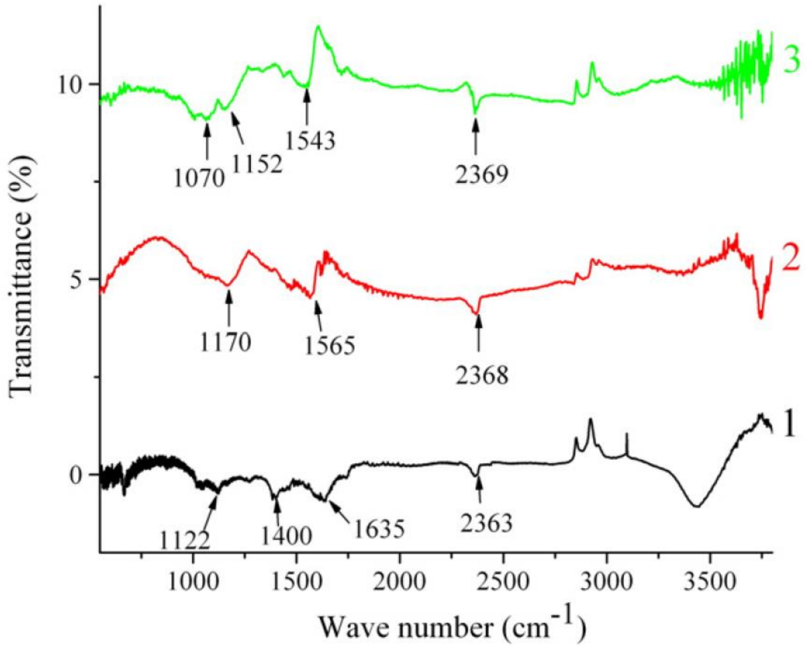

Figure 4. *FTIR spectra obtained from gamma irradiated SWCNTs in aqueous ammonia with doses of 25 kGy 1), 50 kGy 2) and 100 kGy 3) [6].*

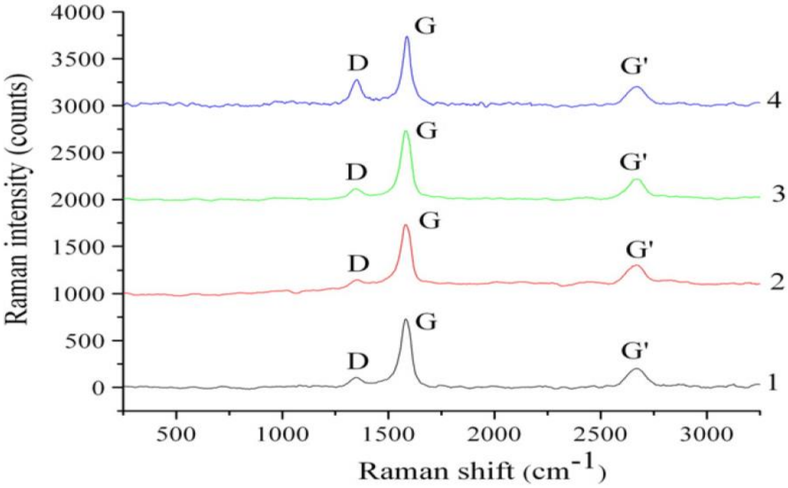

Figure 5. *Virgin sample' Raman spectra (1) and irradiated nanotubes in water at doses of 25 (2), 50 (3) and 100 kGy (4) [6].*

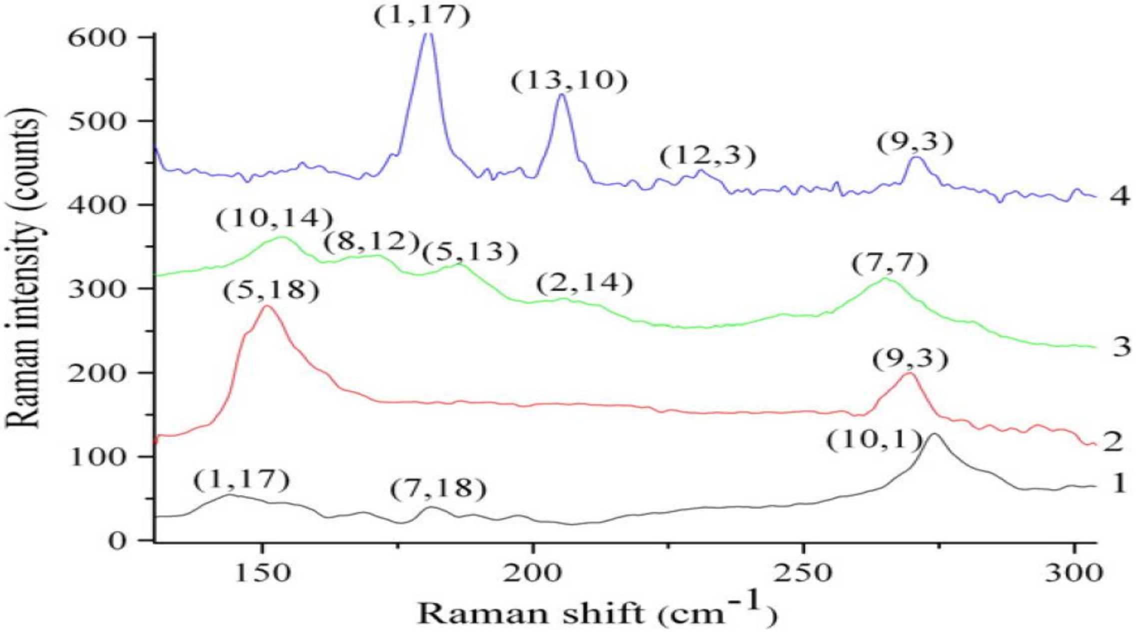

Figure 6. *Raman spectra for virgin SWCNTs (1), γ-air-SWCNTs (2), γ-water-SWCNTs (3) and γ-ammonia-SWCNTs (4) at 50 kGy [6].*

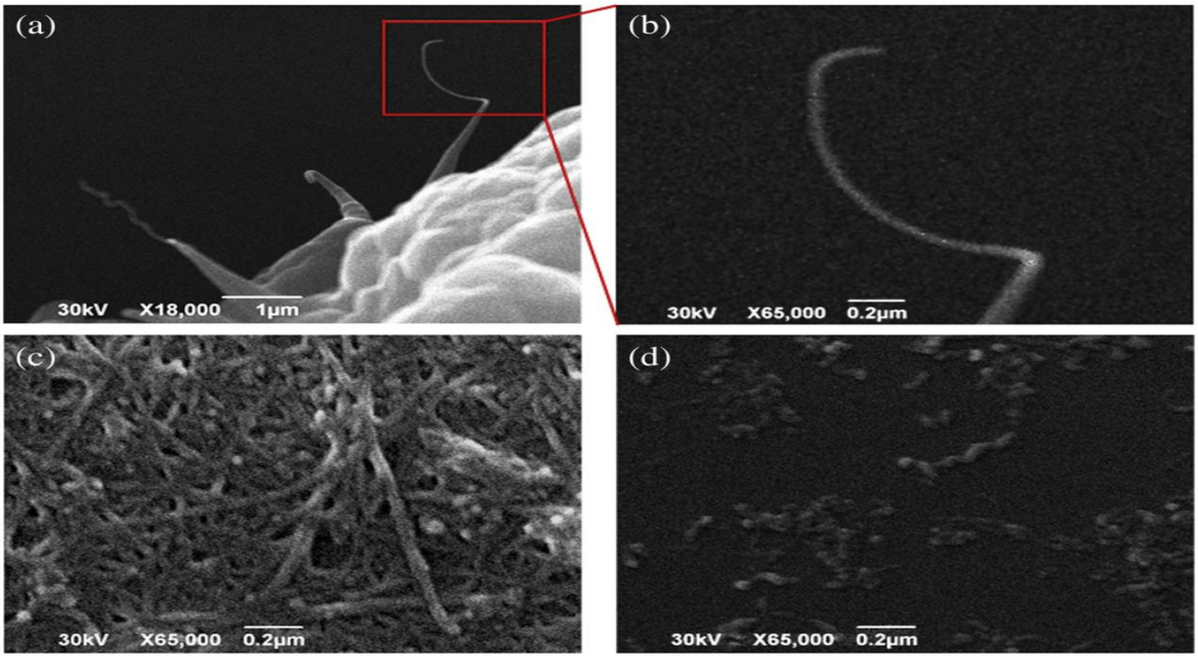

Figure 7. *Irradiated SWCNTs' SEM images in water environment with dose of 25 (a), part highlighted in Figure 7 (a), irradiated SWCNTs with 50 (c) and 100 kGy (d) [6].*

The hydrogen storage process is being studied in the scientific world as a popular subject. Carbon nanotubes are known to be used in these processes because of their large surface area. Hydrogen storage capacities of MWCNTs subjected to gamma radiation were also investigated. MWCNTs were irradiated with gamma rays of 25, 50, 100, 150 and 200 kGy from Co-60 source. At 25 and 50 kGy dose values, the structure improved and the defect rate decreased. At the dose values of 100 and 150 kGy, defect formation occurred with amorphous structures. At a dose of 200 kGy, the structure was completely deformed. As a result of the analysis, it was seen that the hydrogen storage capacity of the samples exposed to 150 kGy doses increased. This increase was at 100°C. No hydrogenation was observed in irradiated samples at low doses. It can be concluded that defect formation at a certain rate provides benefits in hydrogen uptake. Raman results obtained from experiments presented in Figure 8 [19].

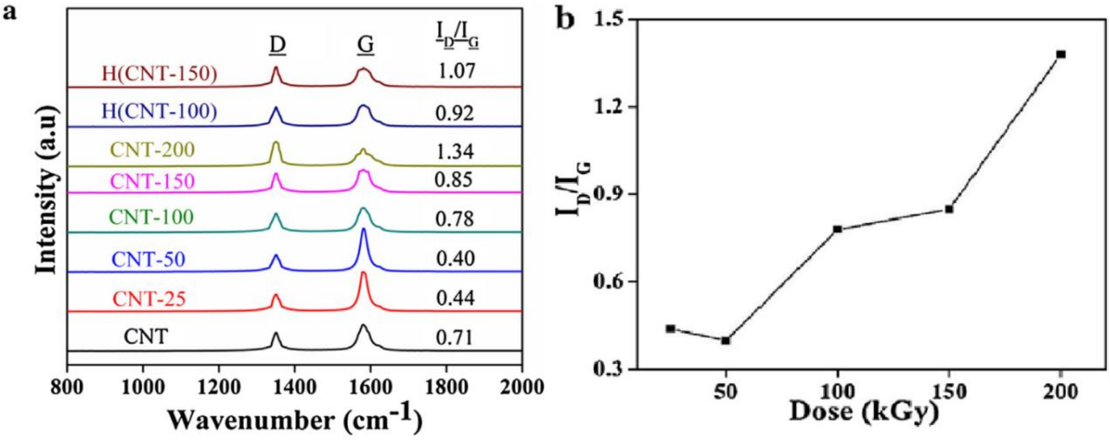

Figure 8. *(a) Raman spectra for virgin, γ-irradiated CNTs with different doses, and γ-irradiated CNTs after hydrogenation, (b) The variation in ID/IG values with different irradiation doses [19].*

The electrical properties of carbon structures and their response to gamma radiation are important in the study of sensitive and vital instruments made by using these materials. It has been investigated how the electrical properties of network transistors using CNTs have been changed by irradiation with gamma rays from a Co-60 source. It was determined that the structure of CNT network channels did not change up to 50 kGy doses. However, charge traps were formed near the CNT/SiO2 bushing interfaces. As a result, it was found that CNT-network transistors are suitable to be used as radiation detectors at dose values over 30 kGy in an air environment. As can be seen in Figure 9, it is seen that the D/G ratios determining the structural quality status of carbon-based materials are close to the non-irradiated sample. This shows that there are not very serious deformations in the structures [20].

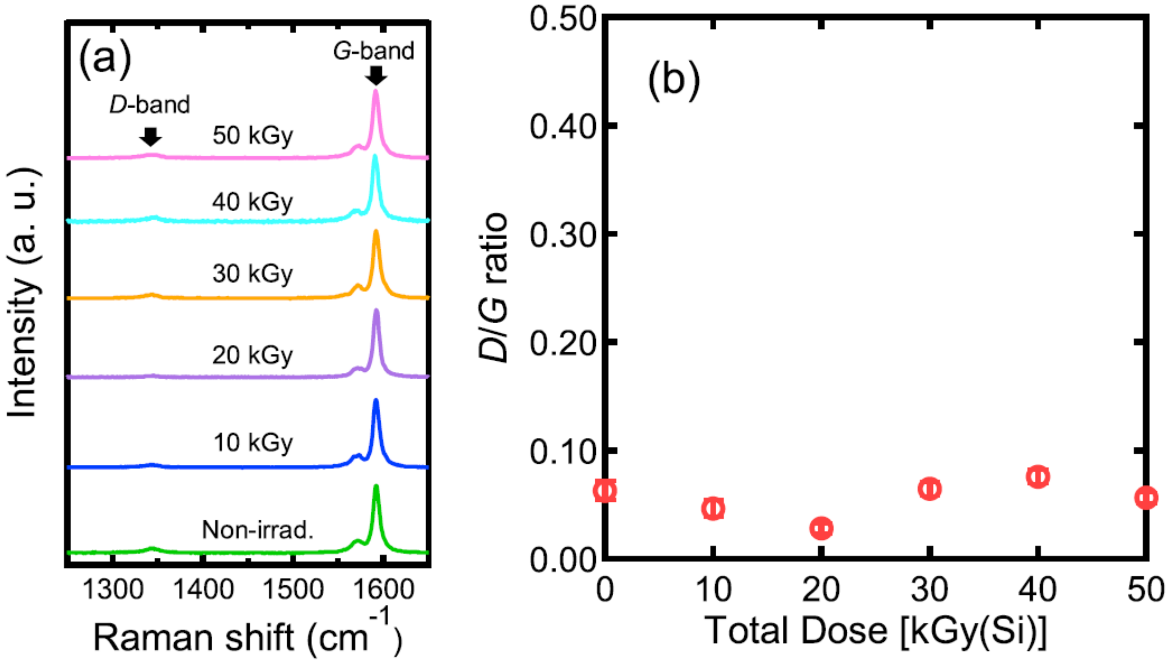

Figure 9. *(a) Micro-Raman spectra belonging the carbon nanotube (CNT) network between the source and drain electrodes. (b) (D/G) ratios for virgin and irradiated samples. The dose change is 0, 10, 20, 30, 40, and 50 kGy [20].*

In some gamma dose values, it was determined that the quality parameters developed after the irradiation of MWCNTs. In irradiated samples at 10 kGy and 100 kGy dose values, it was observed that the defect rates decreased and the graphitization increased compared to the non-irradiated sample [16, 21]

Palladium nanoparticles production and supporting the process with multi-walled carbon nanotubes (MWCNT) can be made through gamma irradiation. It was observed that water gamma radiolysis produced Pd metallic particles. These particles could be used as nucleation seeds. Before production, the water-isopropanol solution was prepared with a ratio 2:1. This salutation mixed with surfactant sodium dodecyl sulfate (SDS) and palladium chloride. As an ion scavenger was used to balance the reaction. During this processing, by adding SDS as a stabilizer, metal nanoparticle coalescence was managed. Nanoparticle distribution sizes on (CNT) were investigated with different radiation doses and surfactant concentrations. Morphological, chemical and structural characterizations of nanostructure were sighted by using SEM, STEM, and XPS. With doses interval 10 and 40 kGy, nanoparticles were obtained in size 5-30 nm. The smaller were produced with increasing doses [22].

On magnetically oriented carbon nanotubes, it was performed polarized infrared reflectivity measurements through conventional light sources as well as synchrotron radiation. In the far-infrared spectral zone, a strong anisotropy was found indicating a preferred orientation of metallic tubes. For semiconducting tubes, less effect was seen. A strong polarization was detected in metallic nanotubes in the range of free-carrier absorption. Due to semiconducting tubes, much less polarization was detected in the inter-band transition region [23].

Epoxy resins based on diglycidyl ether of bisphenol A (DGEBA) with the cationic initiator in the form of iodonium salt were exposed to gamma-rays, in order to initiate the curing process. In this study considering the on-line thermal effects, initiator concentration influence, dose rate, monomers' chemical structure, and carbon nanotubes presence was determined. With decreasing dose rates, the radiation during induction time increased. Through the same process, the exact opposite, the concentration of the initiator and oxirane groups were reduced. Looking at the results of SEM, it was understood that carbon nanotubes were uniformly distributed over the matrix and closely surrounded due to the macromolecules. With the application of gamma rays, mechanical properties' quality of nanocomposites increased [24].

The electrical and mechanical properties of single-walled carbon nanotubes (SWCNTs) that have been functionalized and exposed to gamma radiation have been investigated. Functionalized SWCTs papers using thionyl chloride ($SOCl_2$) were examined. As a result of this investigation, it was found that the electrical conductivity and Young modulus of the samples exposed to 170 kGy gamma dose increased. There has been an increase in the hardness of SWCT papers. As seen from the infrared absorption spectra, radical changes occurred, especially in the electronic structure. The changes in the young modulus and electrical conductivity are shown in Figures 10 and 11 [25].

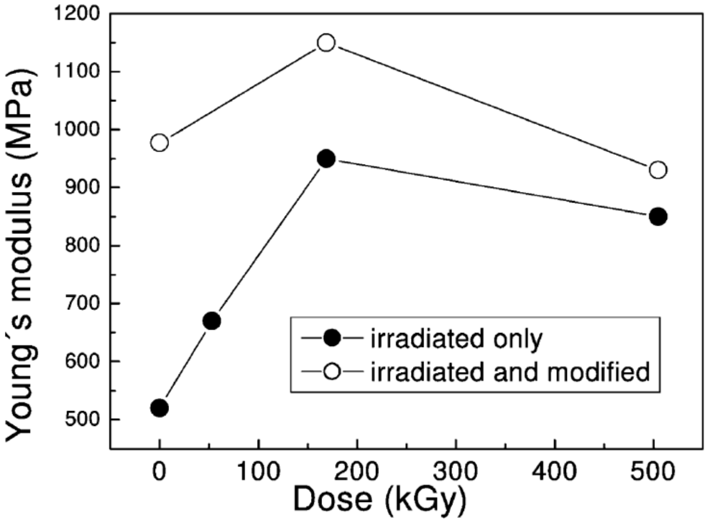

Figure 10. *Young modulus changes for doses of gamma-irradiation (●) and then functionalized (○) NT-paper [25].*

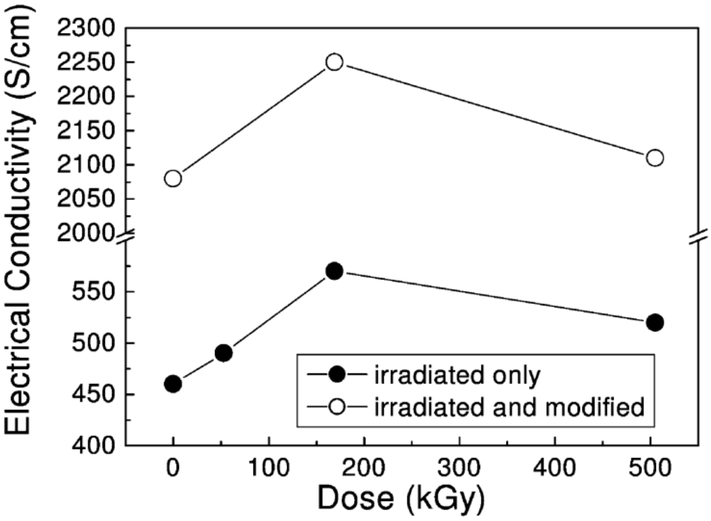

Figure 11. *Electrical conductivity change for doses of gamma-irradiation (●) and then functionalized (○) NT-paper [25].*

The response of low-wall gamma radiation by single-walled carbon nanotube doped field-effect transistors (SWCNT-FETs) was investigated. The research was carried out with sound spectroscopy. As a result of the analysis, it was seen that the transmission capacities and noise levels of SWCNT-FETs did not change significantly after irradiation. Irradiations $1x10^6$ and $2x10^6$ rad. doses. Active radiation exposure at low doses has been observed to improve the transport and sound properties of the structure [26].

Graphene is another carbon-based structure with a wide technological application area. The interaction of this structure with gamma radiation gives interesting results. The effect of gamma radiation on the structure and composition of low-layered graphene material was investigated. Fully functionalized graphene oxide and graphene nano-ribbons were exposed to gamma-rays in the air. Dose values of 60, 90 and 150 kGy were administered. The results of XRD, FTIR, thermogravimetric analysis (TGA), Raman spectroscopy and XPS were obtained. The irradiation did not change the XRD parameters of the structure, i.e. the distance between the crystalline layers, the crystal constant. FTIR and TGA results showed slight differences. However, major changes were observed in Raman spectroscopy and XPS analysis. Raman spectroscopy results can be seen from Figure 12. This shows that the formation of flaws in the structure and formation of functional groups have changed significantly with irradiation. These values are more dominant than the change in the reduction process of graphene oxide structures. The carbon lattice of the graphene has been severely altered, but the oxygen deposition was also less effective. In light of these results, although there are small changes in the chemical composition of the structure as a result of irradiation, it can be said that there are serious changes in carbon mesh. This predominant change in the knitting structure is also seen in Raman graphs [27].

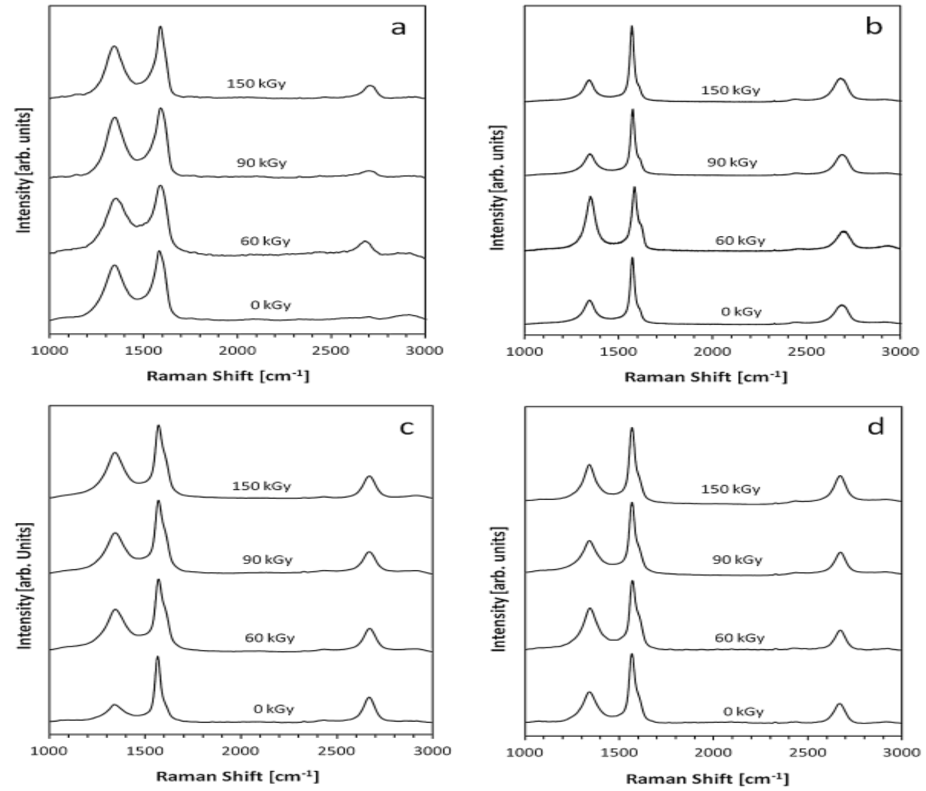

Figure 12. Raman spectra (532 nm) of: (a) graphene oxide, (b) Reduced graphene oxide, (c) Graphene oxide nanoribbons, and (d) resulting reduced suspension, after different γ-irradiation treatments [27].

The interaction of carbon nanotube strands with gamma radiation in the air was investigated. Irradiation rehabilitated the tensile strength and yarns modulus. Sonic wave tests also show that irradiation increases the sound transmission rate and dynamic modulus of carbon nanotube strands. The formation of carboxyl groups in the irradiation structure is singularized. This result was obtained by XPS analysis. The increase of oxygen and carbon nanotubes interacted with the strength of the structure. These are important and remarkable inferences in technological applications. The process of preparation of the material and the changes in its parameters can be seen in Figures 13, 14, 15 and 16 [28].

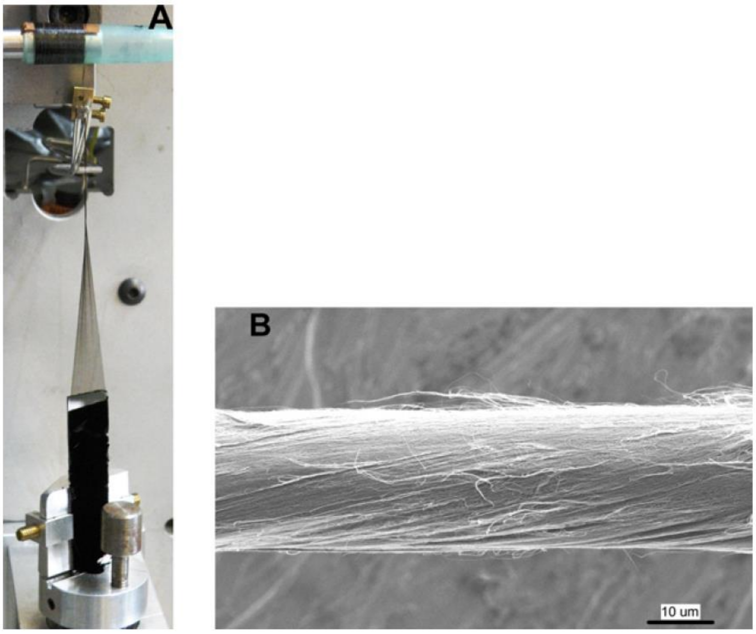

Figure 13. *The yarn spinning system of carbon nanotube and carbon nanotube yarn spun SEM image [28].*

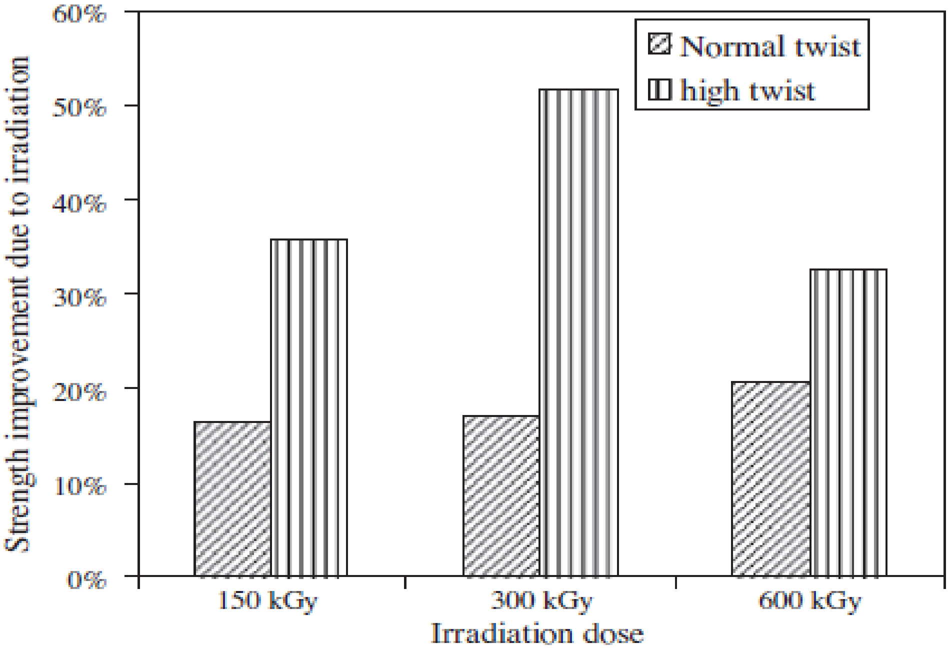

Figure 14. *Gamma-irradiation influence for CNT yarn twist on strength improvement [28].*

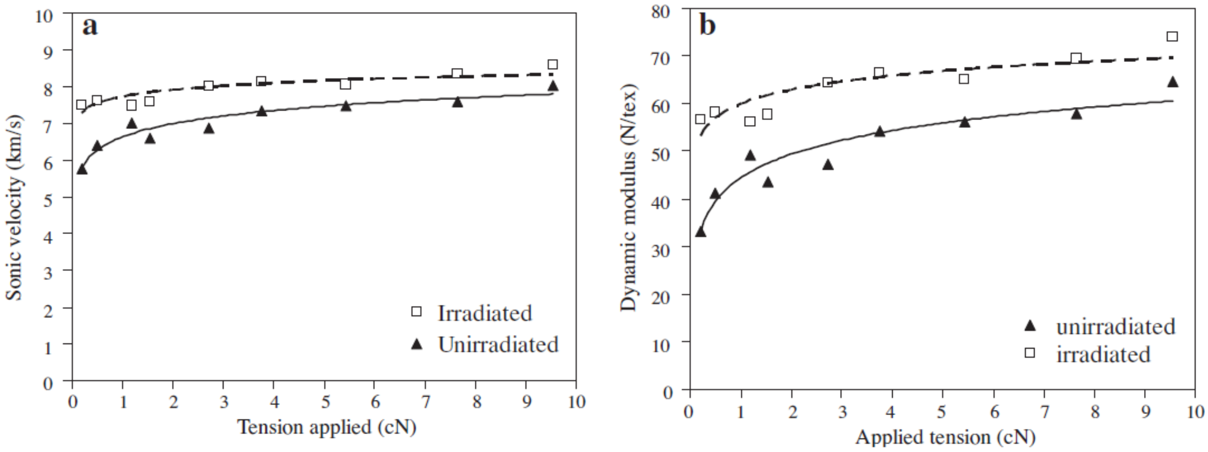

Figure 15. *Gamma-irradiation effect on sonic velocity (a) and dynamic modulus (b) of CNT yarns. Gamma-irradiation dosage: 600 kGy at 4.2 kGy/h [28].*

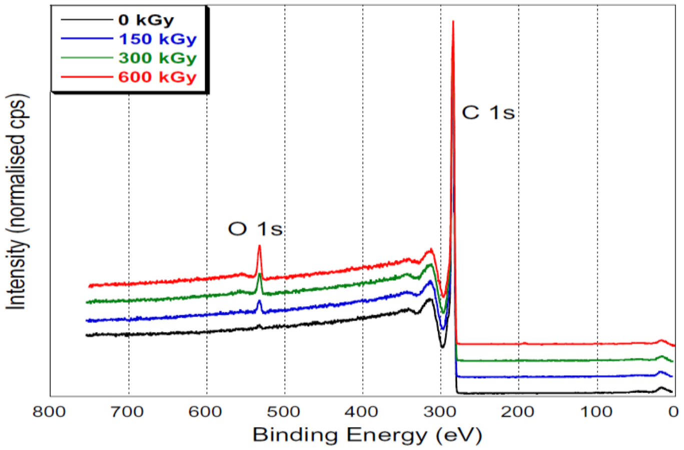

Figure 16. *Irradiated and un-irradiated CNT forests' XPS survey spectra. All spectra were normalized to the C 1s peak height, then offset by 500 cps [28].*

It was determined that different carbon-based structures having different atom and layer sequence forms react differently to gamma radiation. Graphite and the SWCNT samples were irradiated with gamma rays having 1.3 MeV energy. As a result of this irradiation, defects were observed in the structure of both materials. Radial breathing mode decreased in the SWCNT structure. Although the G band representing graphitization does not change in graphite structure, it increased in the SWCNT structure. This situation is considered as a situation to be explained and the situation is explained by softening the selection rule Q = 0. Because this cannot be elucidated by the defect-induced double resonance mechanism. Irradiation defects in SWCNT structure prevented the synchronous movement of atoms in nanotube lattice. Changes are clearly observed in Raman spectra [29].

It is possible to change the electrical, mechanical and optical properties of the materials by injecting carbon nanotubes. In this way, the desired new materials in technological applications can be developed. In composite materials, such additions are frequently made. Properties of carbon nanotube (CNT) reinforced polycaprolactone (PLC) based composites prepared by compression molding are discussed. The addition of 0.2% CNT increased the tensile strength (TS) (in Figure 17) of the PLC films by 131%. Again with the addition of CNT, the tensile modulus (TM) (in Figure 18) and elongation at break (Eb) of the PLC were significantly improved. Daily water vapor permeability rate decreased from 1.51 g.mm/m^2 to 1.08 g.mm/m^2 with the contribution of CNT. Oxygen conduction rate (OTR) decreased with CNT doping. However, the carbon dioxide permeation rate (CO2TR) increased. The effect of gamma irradiation was investigated on both PLC film and CNT doped PLC-based composite structures. The TS value of the PLC film structure irradiated with a gamma dose of 10 kGy increased by 75%. The TS value of the copolymers with 0.2% CNT added reached 41 MPa with a 15 kGy dose application. Another important application is the barrier feature of PLC film structure. In this context, the barrier properties of virgin PLC film, 10 kGy gamma-irradiated PLC film and 0.2% CNT doped PLC films were investigated. Water irradiation (WVP) of PLC film and composite decreased after irradiation. Water vapor blocking property improved. OTR and CO2TR values decreased by irradiation for PLC film and composite structure. As a result of surface and interfacial analysis of composites made with an electron microscope, it was observed that CNT addition in 0.05-0.5% range improved the mechanical properties of PLC films. CNTs deposited at the interfaces contribute to the development of barrier and mechanical properties of PLC films. CNT was not detected on the surface. Significant findings were obtained for packaging and isolation under atmospheric conditions [30].

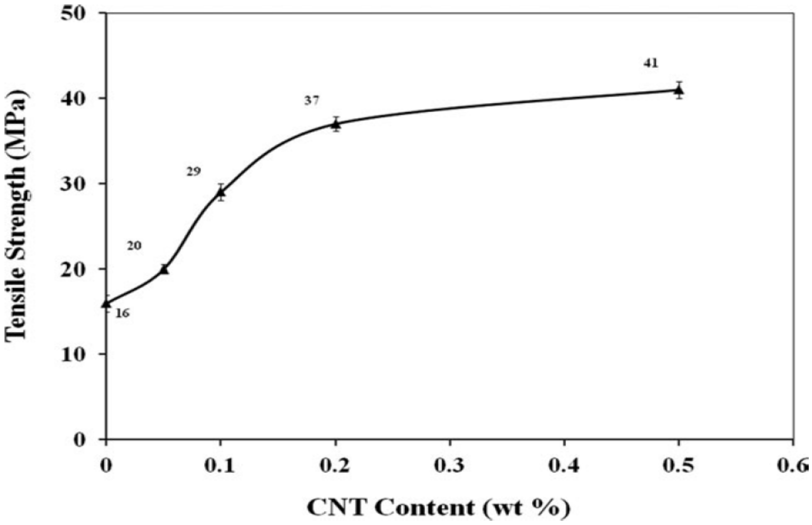

Figure 17. *TS values for the composite films [30].*

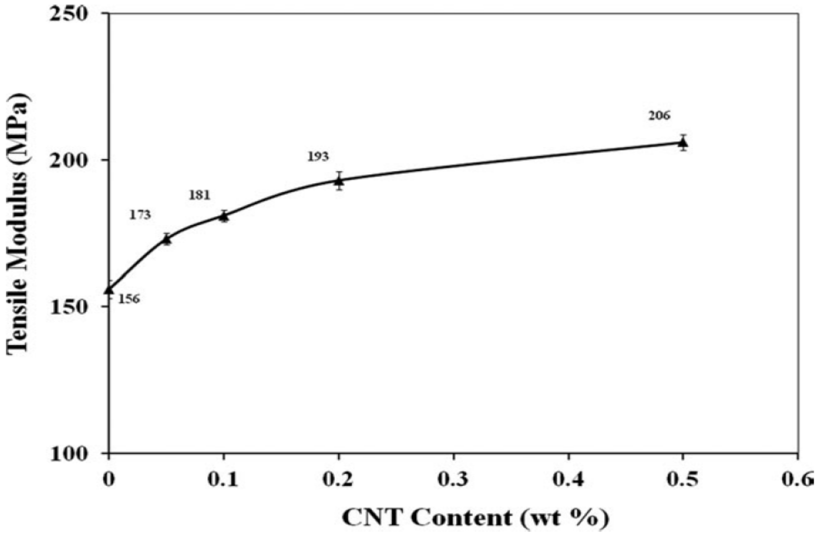

Figure 18. *TM values for the composite films [30].*

II. CONCLUSION

The response of carbon nanotubes to gamma radiation varies depending on many variables. These variables are the type of carbon nanotube (graphite, graphene oxide, MWCNTs, SWCNTs, Double-walled carbon nanotubes (DWCNTs), etc.), the dose and energy of the gamma radiation applied, the duration of application, the environment where the radiation is applied (water, air, etc.). In the interaction of the structure with gamma radiation, defect formation was determined in certain dose values and defect reduction was determined in certain dose values. Gamma irradiation was used to achieve the desired properties of carbon structures. It is observed that the structure maintains its stability at low doses and important deformations occur at high doses. Dense deformation and combustion of the structure above the dose value of 150 kGy were observed. It was observed that the strength and elongation elasticity of the structure increased by irradiation in polymer structures injected with carbon nanotube. It was possible to observe the desired and undesirable changes in electrical and mechanical properties. When the interaction of carbon-based structures with radiation is under control, it may also be possible to produce materials having the desired properties. Again, it is understood that they can be used in radiation-sensitive environments under certain conditions since the electrical distribution of the structure does not change much despite the structural change. When the interaction of carbon-based structures with radiation is under control, it may also be possible to produce materials having the desired properties. Again, it is understood that it can be used in radiation-sensitive environments under certain conditions since the electrical distribution of the structure does not change much despite the structural change. It can be concluded that the stability of carbon-based materials can be used in precision device construction, except in extreme conditions.

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