radiation

Review

# Views on Radiation Shielding Efficiency of Polymeric Composites/Nanocomposites and Multi-Layered Materials: Current State and Advancements

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**Simple Summary:** Highly energetic radiations such as X-rays, gamma rays, cosmic rays, neutrons, protons, etc. in outer space directly affect space crafts. To attain light weight and durability, advanced multi-functional materials (composites and nanocomposites) have been used instead of metallic shields. This article aims to review essential radiation shielding materials for aerospace structural applications. Advanced polymeric composite and nanocomposites have been developed with superior radiation shielding properties, practically applicable for the spacecraft. In this regard, various polymer and nanofiller combinations have been developed to shield the space equipment/structures from harmful radiation. In addition, these materials must be proficient at bearing the mechanical and extreme temperature effects under severe environmental conditions. Multi-layered shields of polymeric nanocomposites have also been developed as efficient radiation protection shields. Despite the advantageous properties, challenges have been identified for the design and application of polymeric nanocomposites and multi-layered materials in highly effective radiation shields. Hence, focused future research efforts are indispensable for high performance radiation shielding materials/technology in aerospace equipment and structures.

**Abstract:** This article highlights advancements in polymeric composite/nanocomposites processes and applications for improved radiation shielding and high-rate attenuation for the spacecraft. Energetic particles, mostly electrons and protons, can annihilate or cause space craft hardware failures. The standard practice in space electronics is the utilization of aluminum as radiation safeguard and structural enclosure. In space, the materials must be lightweight and capable of withstanding extreme temperature/mechanical loads under harsh environments, so the research has focused on advanced multi-functional materials. In this regard, low-Z materials have been found effective in shielding particle radiation, but their structural properties were not sufficient for the desired space applications. As a solution, polymeric composites or nanocomposites have been produced having enhanced material properties and enough radiation shielding (gamma, cosmic, X-rays, protons, neutrons, etc.) properties along with reduced weight. Advantageously, the polymeric composites or nanocomposites can be layered to form multi-layered shields. Hence, polymer composites/nanocomposites offer promising alternatives to developing materials for efficiently attenuating photon or particle radiation. The latest technology developments for micro/nano reinforced polymer composites/nanocomposites have also been surveyed here for the radiation shielding of space crafts and aerospace structures. Moreover, the motive behind this state-of-the-art overview is to put forward recommendations

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for high performance design/applications of reinforced nanocomposites towards future radiation shielding technology in the spacecraft.

**Keywords:** polymer; composite; nanocomposite; low-Z material; multi-layered; radiation shielding

## 1. Introduction

Energy-rich particle radiation from solar particle events and galactic cosmic rays (GCR) is one of the main concerns for extraterrestrial activity [1,2]. About 80% of the GCR consist of extremely energetic protons which may damage electronic equipment and astronauts through atom displacement and energy deposition in the irradiated materials [3,4]. Currently, most spaceflight missions are still in the Low Earth Orbit (LEO) [5]. In space missions, charged particle radiations, especially hazardous cosmic radiations, have energies up to tens of MeV, which may cause serious damage to the spacecrafts' structural material and electronic components and also induce carcinogenic effects in man-based missions [6]. In order to protect the spacecraft and astronauts from harmful effects of ionized particle radiation, efficient lightweight shielding materials have received tremendous attention for successful space trips [7]. In this regard, the International Commission of Radiation Protection (ICRP) has devised a radiation protection strategy including minimizing operating time, maximizing the distance from the source and protective shielding materials [8].

In the past few years, polymer composites have gained attention as potential light weight candidates to replace metal protection materials for radiation shielding [9]. Since, the shielding material (once installed) is difficult to repair in the upper atmosphere, designs must meet harsh environment criteria such as heat resistance and structural integrity [10]. According to theoretical calculations, materials with larger charge-to-mass ratios (Z/A) will more effectively absorb energy from incident-charged particles through Coulomb interactions [11]. High-Z materials such as aluminum alloys may attain shielding performance comparable to low-Z materials. Crucially, high-Z materials have higher chances of nuclei fragmenting under intense particle radiation and producing secondary radiations such as neutrons, protons, electrons, x-rays and $\gamma$-rays. Moreover, in high-Z metal shielding, the secondary radiation caused by initial incident particles may also cause sensitive electronics malfunctioning and biological effects to living systems [12].

In polymer-based radiation shields, two different types of materials have been used i.e., composite/nanocomposite and multi-layered materials. Inclusion of filler or nanofiller has enhanced the shielding capability of materials. The shielding performance of the composite/nanocomposite materials has been found to be superior towards different kinds of radiation, compared with neat polymeric materials [13]. Here, polymer matrices filled with metal or carbon nanofillers have led to remarkable results due to exceptional radiation shielding properties. Similarly, the shielding effect of the multi-layered composite/nanocomposite was better than that of singly layered composite/nanocomposite materials. In multi-layered structure configuration, the shields have greater probability of scattering and absorbing the incoming radiation [14]. However, obtaining a homogeneous combination of constituent multi-layered materials can be challenging and may lead to variable shielding capabilities [15,16]. Multi-layered shielding also offers a solution to pinhole issues by incorporating different layers with controlled porosity [17]. Concisely, this review thoroughly addresses various emergent polymer composite/nanocomposite shielding materials and multi-layered structures for radiation protection in the upper Earth atmosphere towards harmful ionized particle radiations. Hereafter, the main aim of this review is to survey various important composites and multi-layered materials in space craft shielding applications. In this regard, advancement in these materials/nanomaterials have been explored for the related technologies and foremost challenges regarding the applications in aero spacecraft. Literature reports on radiation shielding polymeric nanocomposites were found between 2010–2022. However, significant research reports on radiation shielding polymer composites/nanocomposites were observed in the years 2000–2010, therefore some important studies were indispensable for inclusion in this article. Hence, we attempted to include almost all possible important literature to portray the main progress in this field during the last two decades. Future developments in the field of novel radiation shielding composites/nanocomposites are not possible for researchers before obtaining preceding knowledge from the reported field literature.

## 2. Polymers Composites/Nanocomposites: Fundamentals and Radiation Shielding

For years, polymers have been well known materials in several advanced application fields [18]. Polymers are actually multi-functional materials which can be easily molded for the desired applications. However, several aspects need to be considered in this field, because a single polymer cannot meet the requirements for technical use. Therefore, polymer composites have attracted worldwide attention. Polymer composites are materials having a polymer matrix with reinforced compounds, both components having significant properties. Consequently, polymeric composites have been modified by functional fillers and reinforcing agents. The choice of fillers or additives mainly depends on the end use because their incorporation in the host polymer may affect the matrix properties. Compared with traditional materials, the resulting composite materials may have multiple advantages (strength, stiffness, modulus, dimensional stability, thermal stability, processability, formability, etc.) for the desired practical end use [19]. Composite materials are generally composed of at least two different phases to achieve combined properties. The composites can be classified in three subclasses: particles, fibers and structural composites (Figure 1).

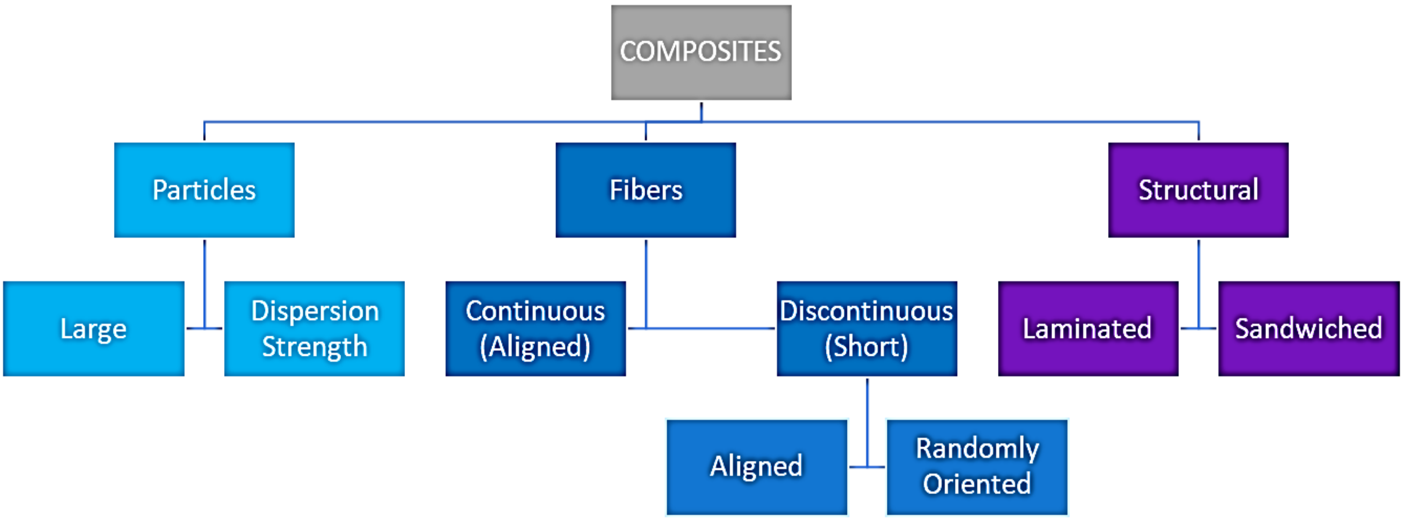


**Figure 1.** General classification of polymer composites.

Composite materials have been investigated for their low density, heat resistance, thermal conductivity, flame resistance, wear, toughness, strength, durability and high radiation shielding, features. A very important use of polymers and polymeric composites has been observed in the aerospace and automotive industry. Table 1 depicts the comparison between some important properties of polymer composites vs. conventional material used in aero spacecraft. In this regard, research on nanocomposites has become increasingly important in developing new high-performance materials. A nanocomposite is typically a mixture of bulk matrix and nano dimensional solid phase material. The polymer derived nanocomposites have advantages in forming ability and dimensional variations. An imperative use of polymers, composites and polymeric nanocomposites has been found in the radiation shielding. Accordingly, noteworthy research has been focused towards designing lightweight, inexpensive and flexible shielding materials for radiation protection [20]. Polymer composite or nanocomposite-based materials have been applied in various industries such as aerospace, satellites, nuclear reactors, etc. (Figure 2). Incidentally, the polymer composites have become striking candidates for developing materials with capability of attenuating photons or particle radiations.

**Table 1.** Assessment of polymer composite based shielding materials vs. conventional metal material.

| Industries | Materials | Standard Material | Mass Density | Thermal Conductivity & Material Strength | Formability | Toxicity |
|---|---|---|---|---|---|---|
| Aerospace | Neat polymers such as epoxy, polyethylene, polyether-imide, poly sulfone, etc. | Aluminum or metal alloys | Low | Equivalent or bit low | High | Very low |
| Aerospace/automobile /transportation | Polymer plus micro/nanofillers such as graphite, carbon fiber, carbon nanotube, nano-clay, etc. | Aluminum or metal alloys | Low | Equivalent or low | High | Low |

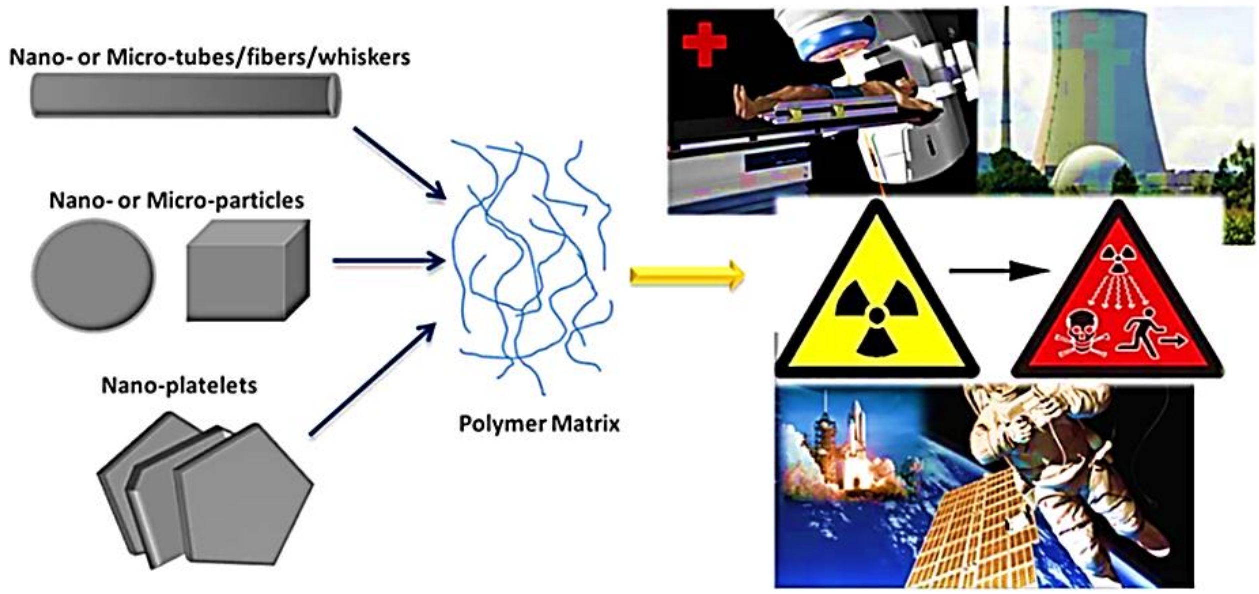


**Figure 2.** Polymeric composite/nanocomposite materials for radiation protection [20]. Reproduced with permission from ACS.

Consequently, various polymers and filler combinations have been used as radiation protection materials. Low-Z polymers, such as polyethylene and poly-methyl meth-acrylate, have inferior mechanical strength, electrical conductivity and thermal conductivity compared to aluminum alloys. In order to discover lightweight alternatives to aluminum alloys with acceptable strength and high performance against radiation, multiple research efforts have examined various forms of polymer-based composites [21]. Nano/micro-sized fillers can be dispersed in the polymer matrix to enhance mechanical strength, electrical properties, thermal properties, electromagnetic interference shielding, photon radiation shielding and neutron attenuation characteristics. The radiation shielding effectiveness of polymer composites or nanocomposites against charged particles, however, was shown to be hardly affected by the inclusion of the fillers.

Nambiar et al. [22] have synthesized lead-free radiation protection nanomaterials. They developed poly-dimethyl siloxane (PDMS) nanocomposites by adding different weight percentages of bismuth oxide ($Bi_2O_3$) nano powder. The materials were recommended for good X-ray shielding. Figure 3 shows the experimental set-up for direct and scattered X-rays. The X-ray tube-voltages were set between 40 to 150 kV. The set-up consists of an X-ray source, ion chamber and X-ray detector. The lead box was placed to filter the scattered photons. The exposure time was between 100 ms and 250 ms. Figure 4 shows the effect of bismuth oxide concentration on the attenuation of PDMS. The materials were studied under a primary X-ray beam. The samples of equal thickness were tested. Increase in nanofiller loading was found to increase the attenuation. The nanocomposites with 44.44 wt.% nanofiller were highly effective for X-ray attenuation.

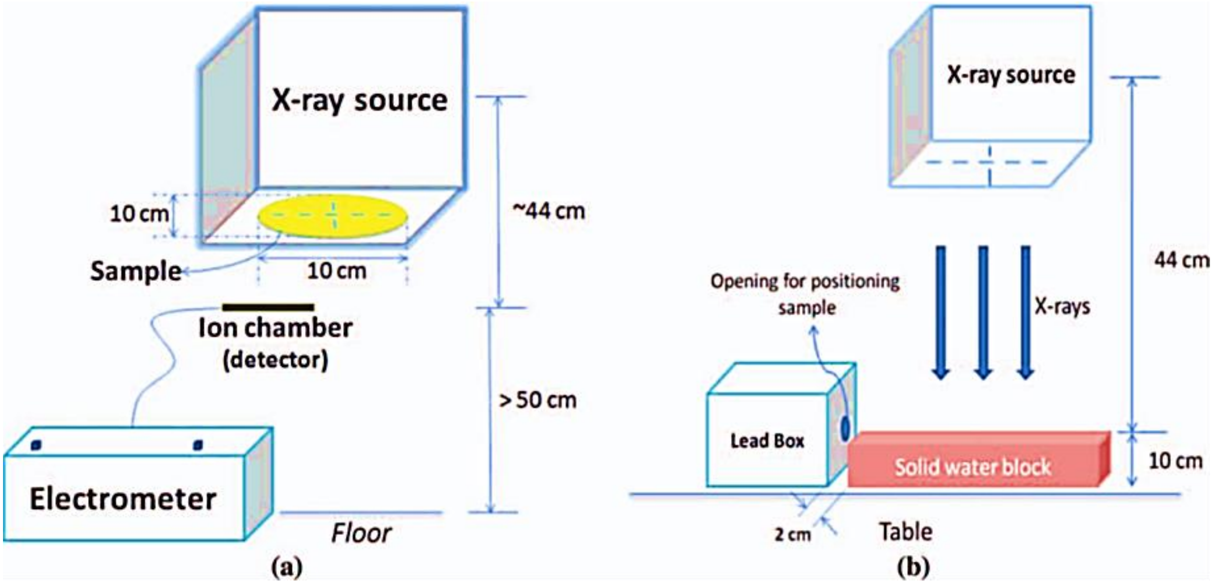


**Figure 3.** (**a**) Experimental set-up for direct X-rays. (**b**) Experimental set-up for scattered X-rays [22]. Reproduced with permission from Wiley.

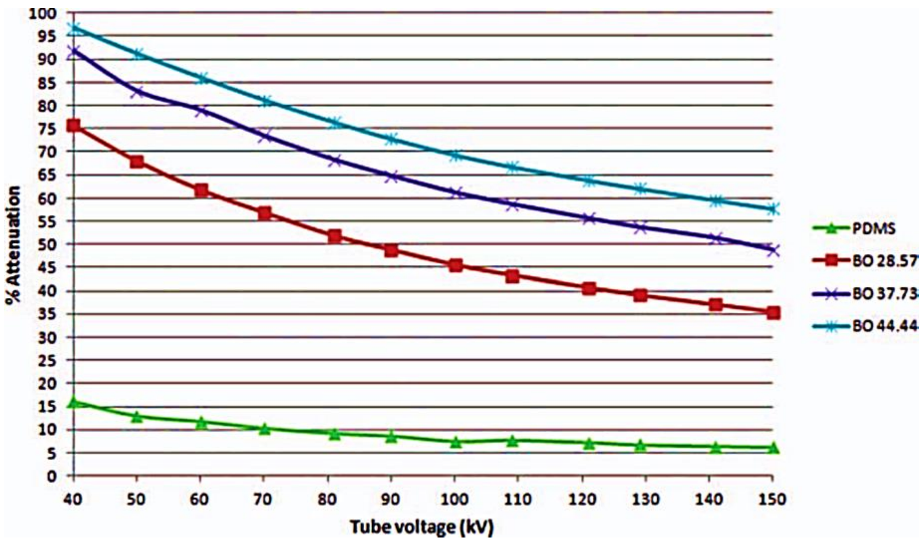


**Figure 4.** Percentage attenuation for different thicknesses of poly-dimethyl siloxane (PDMS) and bismuth oxide (BO) nanocomposites using primary X-ray beam [22]. Reproduced with permission from Wiley.

Ambika et al. [23] fabricated unsaturated polyester resin reinforced with the $Bi_2O_3$ nanoparticles. The study disclosed that the gamma ray shielding properties were enhanced with increasing $Bi_2O_3$ concentration in the matrix. Yurt Lambrecht et al. [24] developed ethylene vinyl acetate and tungsten powder based nanocomposite. It was observed that the metal nanoparticles significantly enhanced the gamma ray shielding property of the polymer matrices. The mechanical hardness of the nanocomposites was also raised with metal nanoparticle addition. Harrison et al. [25] formed polyethylene/boron nitride composites. The 2 wt.% boron nitride loaded polyethylene composite revealed enhanced radiation shielding than the neat polyethylene matrix. The polyethylene/boron nitride composites were operative in the absorption of slow neutrons as well as for space radiations. Gwaily et al. [26] loaded natural rubber and boron carbide composites. The boron carbide loading was useful for thermal neutron radiation shielding. Epoxy resin has been used as an important matrix for composites and nanocomposites due to good dimensional stability, low shrinkage and excellent adhesion to several reinforcements. In this respect, Adeli et al. [27] fabricated epoxy/boron carbide composites for neutron absorption applications. Chang et al. [28] prepared epoxy/tungsten nanoparticle composites. Including different tungsten powder contents enhanced the neutron and x-ray/gamma protection, in addition to the mechanical and thermal stability. Numerous studies have been conducted on the use of fillers/nanofillers [29,30] and polymers [31–35] in the production of radiation shields.

Thus, polymers have been widely exploited for producing radiation shields due to light weight, low price and flexibility [36,37]. However, polymer chains may experience crosslinking, chain scission, or degradation, when exposed to various radiation sources with varied frequency rays [38]. Inclusion of fillers to polymers usually slows down the deterioration of chains and lessen the radiation's harmful effects. In order to build functional structures from polymer and filler for intended end-use, it is essential to take into account the atomic structure of the filler. The radiation shielding effectiveness of a filler depends on its atomic number. High atomic number fillers are typically utilized for gamma radiation protection. For neutron radiation shielding, however, it is better to utilize fillers with a low atomic number. The compatibility of the polymer-filler has a considerable impact on the radiation shielding effectiveness of the manufactured composite material [39]. Furthermore, it is impossible to overlook the radiation harm brought on by secondary neutrons. Secondary neutrons are more challenging to protect than other ionizing radiation sources due to their electrical neutrality. Large neutron capture cross-sections are necessary for effective neutron shielding [40]. The boron isotope 10B is a commonly used element and its compounds have shown effectiveness in the fabrication of neutron-shielding nanocomposite. However, the second shield layer may increase the total weight of the shield. For galactic cosmic rays (HZE), passive shields have been used [41]. Since it is technically impossible to completely stop high energy nuclei by a shielding material, one practical solution is to absorb relatively low energy particles and then fragment the HZE particles into lighter particles. The research using particle accelerators has focused on the shielding capabilities of materials such as aluminum and polyethylene [42]. The best element for protecting heavy nuclei and protons is hydrogen. However, due to the chemical instability of atomic hydrogen, hydrogen-rich shielding materials such as polyethylene materials have been applied in the walls and water-rich towels of the international space station (ISS) [43,44].

## 3. Epoxy Composites/Nanocomposites for Radiation Shielding

Engineering composites have been fabricated using various fiber fillers such as carbon fibers, glass fibers, etc. [45]. Polymeric matrices such as epoxy, phenolic, polyester, etc., have been filled with layers of fibers, especially for spacecraft [46,47]. Epoxy/layered fiber-based composites have been used to form a high performance structure [48–50]. There have been several attempts to improve the shielding properties of elemental particle filled epoxy matrix composites or nanocomposites [51]. Researchers have repeatedly tested the effects of particle loading on radiation shielding through reducing particle size, using a doping matrix with various particle concentrations, exposing composite specimens to various radiation sources, analyzing microstructural changes under various radiation doses and testing composite failure mechanisms. Al-Sarray et al. [52] examined the ability of epoxy-based composite panels to shield against radiation. The linear attenuation coefficient of the constructed epoxy composites was assessed using $Co_{60}$ and $Cs_{137}$ radioactive sources with various barite concentrations up to 50 wt.%. With an increase in barite content, the radiation shielding effectiveness was found to increase. Moreover, the impact of lead oxide and barium oxide loading on the radiation shielding effectiveness of epoxy composites have been investigated [53]. The addition of barium oxide improved the gamma radiation performance of epoxy/lead oxide composites; nonetheless, the barium oxide contents need to be added at a rate twice than that of lead oxide. Additionally, as compared to concrete, steel and gadolinium oxide/epoxy composites, epoxy resin with 40 wt.% barium oxide revealed excellent radiation shielding capability [54]. Li et al. [55] dispersed micro- and nano-gadolinium oxide particles in the epoxy matrix and evaluated the mechanical and radiation shielding quality of the resulting composite. Due to the dominant photoelectric action of the gadolinium element, addition of gadolinium oxide improved the radiation shielding properties. In comparison to epoxy/micro-gadolinium oxide composites, epoxy/nano-gadolinium oxide composites showed greater flexural modulus and flexural strength along with the improved X-ray and gamma shielding properties. More et al. [56] established the radiation shielding characteristics of epoxy/metal chloride composites. The attenuation characteristics of the epoxy composites were increased by doping the resin with greater wt.% of metal chloride. Alavian et al. [57] reported on the mechanical qualities, structural traits and gamma shielding effectiveness of the epoxy composites. Inorganic nanoparticles including lead, zinc, zinc oxide, titanium and titanium oxide were doped into the epoxy resin to enhance the shielding properties. The 59.54 and 662 keV $\gamma$-rays emitted from $^{137}$Cs point sources were detected using 2″ × 2″ NaI(Tl) detector. Figure 5 shows the experimental setup of both narrow and broad beam geometries used. An important step was to subtract the background spectrum. Figure 6 designates the background measured $^{137}$Cs and background subtracted $^{137}$Cs spectra. In this study, the $\gamma$-ray spectra were taken via a 2″ × 2″ NaI(Tl) scintillator (energy resolution of 8% at 662 keV). The detector was surrounded by a lead shield to prevent the background radiation from reaching the detector. In the first step, background spectrum was subtracted from the measured spectrum. The spectrum (black line) after background substation was considered as real recorded spectrum. The spectrum without background subtraction (blue lines) shows considerable difference in peak count due to the presence of background intensities. After background subtraction, difference in the intensity of peak count of $\gamma$-rays reaching the detector can be observed, which was taken as the real spectrum. The shielding effectiveness of epoxy nanocomposites was improved by increasing the nanoparticle loading. Epoxy nanocomposites with 25 wt.% lead oxide have shown improved shielding capabilities compared with other nanocomposites.

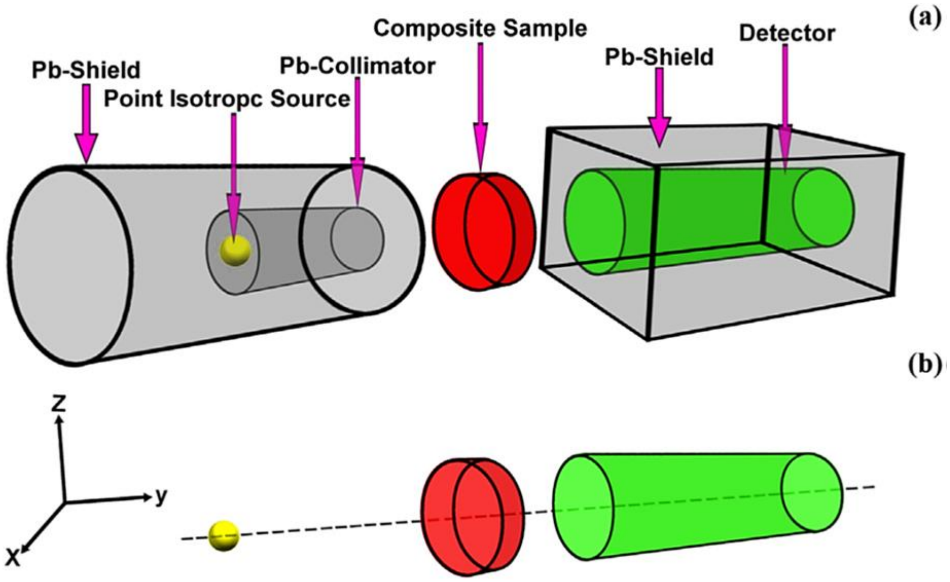


**Figure 5.** Schematic view of (**a**) narrow and (**b**) broad beam experimental setup [57]. Reproduced with permission from Elsevier.

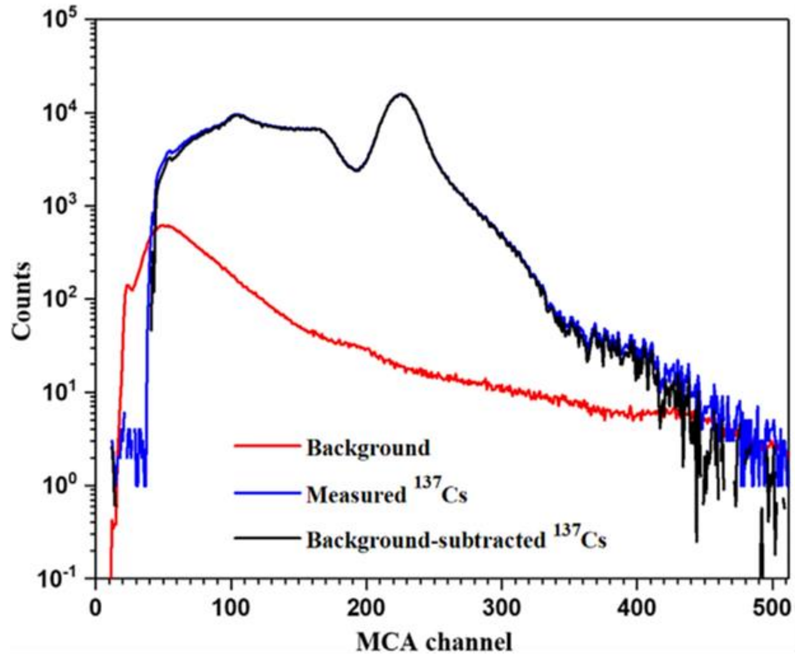


**Figure 6.** Typical background, measured $^{137}Cs$ and background-subtracted (real) $^{137}Cs$ recorded using 2″ × 2″ NaI(Tl) scintillator [57]. Reproduced with permission from Elsevier.

Saiyad et al. [58] studied the degradation of epoxy resin composites using high frequency radiation. The composite materials were prepared by adding lead, boron nitride and graphite to the epoxy resin. The epoxy composites were exposed to radiation from Am-Be neutron sources. The maximum shielding effectiveness was noted in the epoxy/graphite composites. Moreover, the linear absorption coefficients of composites were dependent on the filler dispersion. Azman et al. [59] used epoxy/nano tungsten oxide composites in radiography equipment. The nanomaterials were tested for up to 120 keV X-ray voltages and found to have high radiation shielding.

Recently, Aldhuhaibata et al. [60] prepared epoxy/$Al_2O_3$ nanocomposites for gamma radiation shielding. Epoxy nanocomposites with 6 and 15 wt.% nanofiller contents were exposed to various energies of gamma rays (0.662–1.333 MeV). For gamma ray detection, the mass attenuation coefficient and mean atomic number were measured. Epoxy nanocomposites were found effective for improved gamma radiation shielding. Among recent attempts, La et al. [61] designed lightweight epoxy/$Gd_2O_3$ epoxy nanocomposites. The material was used to replace the traditional lead-based material for X-ray protection. The 16 mm thick nanocomposite shield was applied to attain > 99% X-ray protection. The X-ray attenuation efficiency per unit area mass of the nanocomposite was high, i.e., $\sim$0.88 $cm^2/g$. Recently, Karabul et al. [62] designed epoxy nanocomposites filled with $Bi_2O_3$ and $WO_3$ nanofillers. The effect of the inclusion of $Bi_2O_3$ and $WO_3$ nanoparticles was studied on the shielding properties of the epoxy matrix. The NaI(Tl) scintillation detector was used to detect the radiation from $^{137}Cs$ and $^{60}Co$ sources. The nanocomposites were suggested as useful for radiation imaging machines such as roentgen. Prabhu et al. [63] formed epoxy and $Bi_2O_3$ decorated graphene oxide based nanocomposite. The nanofiller was added in 5–15 wt.% contents. Epoxy/$Bi_2O_3$-graphene oxide nanocomposite was used for X-ray and $\gamma$-ray shielding in a wide energy range of 30–1332 keV. Cha et al. [64] filled MWCNT nanofiller in the epoxy matrix. The materials were found useful in replacing pristine epoxy polymers for effective cosmic radiation shielding in space missions.

In addition to other radiation, epoxy based nanocomposites have been developed for electromagnetic interference (EMI) shielding [65]. Nanni et al. [66] used carbon nanofibers to enhance the dielectric properties of the epoxy nanocomposite. The fine nanofiller dispersion and low shield thickness were found to enhance the electromagnetic radiation absorbing properties. The absorbing performance was studied in the range of 8–20 GHz. Consequently, the carbon nanofibers increased the permittivity of epoxy resin. Inclusion of low carbon nanofiber contents (1 wt.%) at 19.8 GHz yielded better electromagnetic radiation absorption and permittivity properties with shield thickness of 2mm. Gupta et al. [67] used reduced graphene oxide/zinc oxide coated wearable cotton fabrics to study the EMI shielding effectiveness in X-band (8.2–12.4 GHz). The uniform coating of reduced graphene oxide/zinc oxide was formed on cotton fabrics using the spraying technique. The EMI shielding effectiveness was found to increase with the nanofiller loading. Inclusion of 7 wt.% nanofiller resulted in a fairly high EMI shielding effectiveness of 54.7 dB. The reduced graphene oxide/zinc oxide loading coating was found to block the pores of the cotton fabric, so enhancing the absorption of the electromagnetic waves. Thus, a high EMI radiation absorption of ~90% was attained. Song et al. [68] prepared EMI shielding nanocomposite based on poly-dimethyl siloxane and three-dimensional hierarchical reduced graphene oxide foams decorated with ZnO nanowires. Inclusion of 3 wt.% nanofiller loading produced nanocomposite with a thickness of 4.8 mm. The poly-dimethyl siloxane/reduced graphene oxide/ZnO nanowires nanocomposite revealed EMI shielding effectiveness of 27.8 dB at 9.57 GHz. The material was suggested as useful for efficient lightweight EMI shielding materials for aerospace. Recently, Liang et al. [69] designed EMI shielding materials based on epoxy, silver platelets and reduced graphene oxide foam. The epoxy matrix with 0.44 vol.% reduced graphene oxide, and 0.94 vol.% silver platelets resulted in high EMI shielding effectiveness of 58 dB in the X-band. The corresponding electrical conductivity was also high at ~45.3 $Sm^{-1}$. Thus, various filler and nanofillers have been filled in the epoxy matrix and shielding effectiveness of the materials was observed towards different types of radiation for space applications.

## 4. Modification of Epoxy Resin with Fibers/Fillers for Radiation Shielding

Studies on radiation shields have recently concentrated on failure mechanisms and reducing the shield weight [70]. In this regard, researchers have studied numerous polymeric matrices with different fillers/nanofillers to fabricate effective alternate shields. Kim [71] noted the effect of tungsten particle size and dispersibility on radiation shielding of high-density polyethylene (HDPE). Here, three different types of tungsten-loaded shields of equal thickness and size were prepared [72]. The shields offered high shielding efficiency against low dosage exposure. HDPE sheets with high nanoparticle contents were found to be more radiation-resistant [73]. Shreef and Abdulzahara [74] investigated the fabrication and radiation shielding capability of nano gadolinium oxide in poly-methyl meth-acrylate (PMMA) nanocomposites. The 10–40 wt.% gadolinium oxide was used to make composite shields. Using cobalt ($Co_{60}$) and cesium ($Cs_{137}$) radiation sources, the shielding effectiveness of composites and thickness were evaluated. According to the results, increasing nanoparticle concentration in the epoxy composites and thickness enhanced the attenuation coefficient. Zheng et al. [75]. fabricated 1:1 glass epoxy/fabric composites. The materials were exposed to radiation from a $Co_{60}$ source. The exposure to gamma rays had no effect on S-glass fiber and degraded the epoxy resin. The color of the composite changed from yellow to brown as the gamma radiation exposure dose increased. Moreover, the tensile strength of the composites decreased with radiation exposure. Li et al. [76] developed mechanically robust radiation shielding epoxy/basalt fiber composite. The composites loaded with erbium oxide were fabricated using the prepreg autoclave technique. The shielding effectiveness of the composites towards X and gamma radiations were tested. The epoxy/basalt fiber/erbium oxide composites revealed low photon energies of 31–80 keV and high mass attenuation coefficient, compared with the pure Al. Moreover, epoxy/fiber composites have been tested for improved fracture toughness, flexural characteristics, impact resistance and thermal stability by several research groups [77,78]. Saleem et al. [79] studied the radiation shielding of epoxy composites loaded with lead nanoparticles along with the glass or carbon fiber. The lead nanoparticles were found to enhance the composite shielding properties and mass attenuation coefficients. Inclusion of 50 wt.% lead nanoparticles yielded mass attenuation coefficients of 0.2145 $cm^2/gm$ and 0.2152 $cm^2/gm$, respectively, for carbon and glass fiber reinforced epoxy composites. Hoffman and Skidmore [80] investigated the effects of gamma radiation on epoxy/carbon fiber composites. The front and back surfaces of woven carbon textiles were treated with epoxy/hardener (2:1). There were no notable changes in the mechanical resistance of composites after radiation. However, the gamma radiation significantly altered the thermal characteristics, spectroscopic analyses and hardness value of neat epoxy samples. Zhong et al. [81] reported ultra-high-molecular-weight polyethylene (UHMPE)/nano-epoxy composites for cosmic radiation shielding. The combination of continuous fibers such as UHMWPE and/or graphite nanofibers produced multi-functional hybrid systems with excellent structural characteristics, cost-effectiveness and radiation shielding performance. In another study, Mani et al. [82] created UHMWPE/epoxy composites with gadolinium and boron nanoparticles and found these materials efficient for neutron shielding, Recently, Kim et al. [83] proposed polyethylene and tungsten nanoparticle decorated boron nitride nanosheets derived nanocomposite. Compared with neat polyethylene, higher yield strength and through-plane conductivity of 16.4% and 35% were attained, respectively. The nanocomposites had high radiation shielding ability for neutron and gamma radiations, i.e., 4.80 $cm^2/g$ and 0.093 $cm^2/g$, respectively. Zegaoui et al. [84] investigated the impact of hybridization on the mechanical, thermal and radiation shielding effectiveness of epoxy resin/silane-treated glass and basalt fibers composites. By incorporating hybrid fibers into a bicomponent matrix system, the resin was hybridized to develop mechanical and thermal qualities as well as excellent shielding properties. In epoxy and other polymer matrices, bismuth nanoparticles along with fibers have also been found to effectively block X-ray radiation, relative to the microparticles at similar loading levels [85]. Durability studies have also been carried out on radiation shields [86]. Alajerami et. al. [87] used melt-quenching technique to form bismuth-borate glasses with different concentrations of cadmium oxide (CdO). The CdO was loaded in 0–15 mol.%. The gamma ray and neutron shielding properties of bismuth-borate glasses/CdO were analyzed. The durability of the bismuth-borate glasses/CdO was evaluated by immersing the glasses in distilled water. The difference in weight was measured after specific time periods (2, 7 and 14 days). The composite durability decreased gradually from $7.523 \times 10^{-7}$ to $1.415 \times 10^{-7}$ $g.cm^{-2}.d^{-1}$ due to an increase in CdO contents. The CdO addition actually reduced the hygroscopic nature of the bismuth-borate glass and so reduced the water-resistance of the prepared glasses. Mhareb et al. [88] prepared boro-tellurite glass with SrO, $MoO_3$, $Al_2O_3$ and $TeO_2$ fillers using the melt quench method. Inclusion of filler enhanced the Poisson's ratio from 0.406 to 0.420, whereas the packing density was reduced from 0.675 to 0.573 due to the dispersion of nanofiller particles. The nanofiller addition revealed high durability and radiation absorption efficiency.

Bel et al. [89] reported the space irradiation effect on the beta attenuation of polymethyl meth-acrylate/colemanite (PMMA/CMT) nanocomposite using the installed beta exposed facility. Investigation of space radiation effects by using PMMA/CMT demonstrated the enhancement in beta shielding performance with the CMT addition. The use of CMT induced high beta shielding in PMMA matrix, due to calcium and boron atoms in its structure. Figure 7 shows the Experiment Handrail Attachment Mechanism (ExHAM) at the International Space Station (ISS) using the PMMA/CMT samples module. The samples module was thrown by Falcon-9 rocket and the vehicle launch was achieved. Figure 8 shows the effect of increase in thickness on the drop-off beta intensity of the PMMA/CMT shield. The 15 wt.% CMT addition considerably decreased the beta intensity. The difference in beta shielding of neat PMMA and PMMA/CMT composite was analyzed. The CMT was quite operative in reducing beta penetration into the composite.

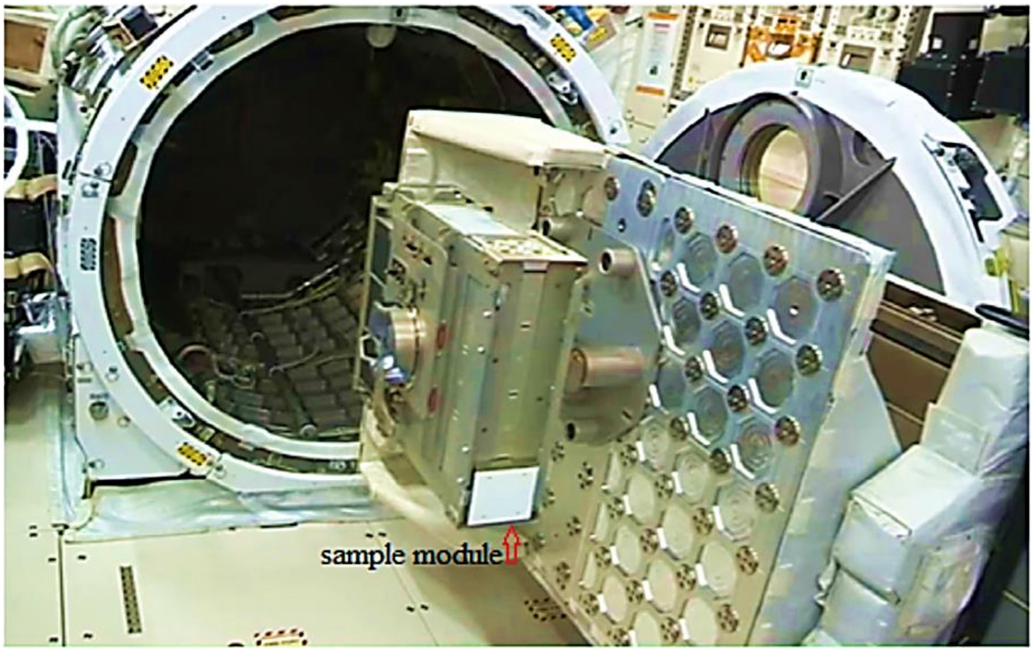


**Figure 7.** The attachment of the sample module to ExHAM in the ISS. ExHAM = Experiment Handrail Attachment Mechanism; ISS = International Space Station [89]. Reproduced with permission from Wiley.

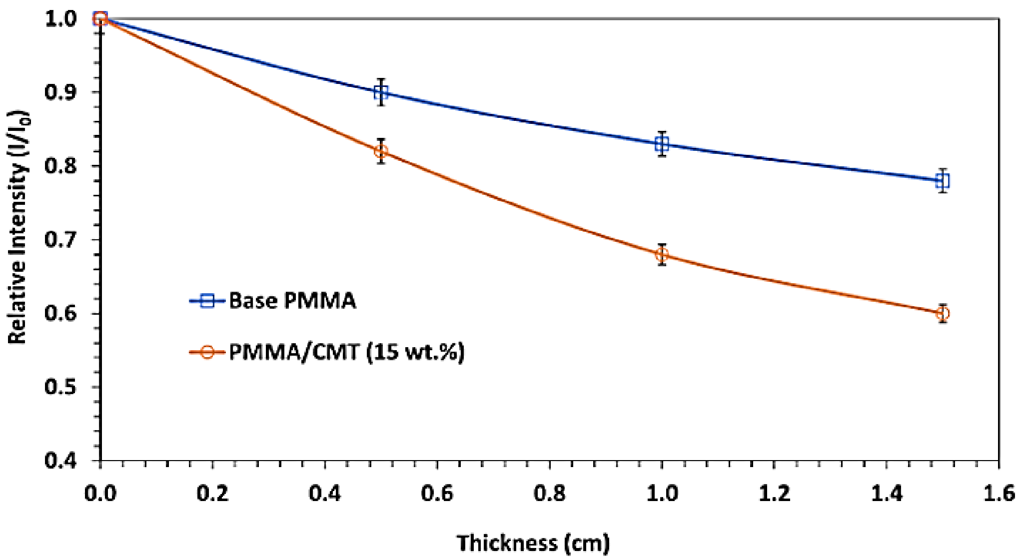


**Figure 8.** Comparison of beta attenuation of base PMMA and PMMA/colemanite composite at different thickness. PMMA = poly-methyl meth-acrylate; CMT = colemanite [89]. Reproduced with permission from Wiley.

## 5. Multi-Layered Radiation Shielding of Polymer Composites/Nanocomposites

Unlike single-layer shielding materials, the stacking in multi-layered shielding has been found to be more effective, but may involve complex phenomenon [90,91]. A monoenergetic radiation flux passing through the first layer may result in an additional flux with various energies due to absorption and scattering processes. Hence, it is difficult to determine the cross sections, linear attenuation and mass attenuation coefficient for the multilayer shielding materials. Consequently, simulations and experimental studies have been used to assess the secondary radiation produced in the multilayer shields through buildup factor calculations. The buildup factor actually depends on the type of radiation, energy, shielding material and geometry involved [92]. The buildup factor was also found to be significantly affected by the material arrangement and thickness of each layer. It has been observed that the buildup factor usually increases with thickness, but this effect depends on the variation in material sequence.

Several studies have been observed in the literature relating the buildup factor of the multi-layered material to the radiation shielding performance. Abbas [93] deduced that lead-water gamma shielding had greater buildup than water-lead (the material order was changed) shield at 1 and 2 MeV. Al-Arif and Kakil [94] demonstrated that there was no significant difference in the buildup factor when the order of material was changed. However, buildup factor revealed dependence on the atomic number and the photon energy. At low energy of radiation, the buildup factor increased with the decrease in atomic number. On the other hand, at high energy, the buildup factor increased with the rise in atomic number. Mann et al. [95] reported a double layered shield in a low-Z/high-Z orientation, at fixed energy. A lower buildup factor was observed for the double layered shield, as compared to the single layered material [96]. Shin and Hirayama [91] also found that the buildup factor generated by the multi-layered configurations was lower than the single layer materials. Using two complementary metal-elastomer bilayers reduced the overall shield weight and caused higher attenuation, relative to pure lead [97]. According to Mann et al. [95], bi-layer shielding was more effective than single layer shielding in gamma ray protection.

Recent studies depicted that the bilayers of iron-containing material performed better than the single layer of concrete in neutron shielding [98]. To design the optimal multi-layered materials, a genetic algorithm (GA) metaheuristic was used by several research groups including Hu et al. [99], Kim and Moon [100] and Cai et al. [101]. The string of integers was used to optimize the multi-layered shield for radiation protection. However, no general agreement was observed for the effects of changing the layer arrangement on the shielding performance.

For neutron shielding, use of hydrogen containing material was found to be a better technique [78]. The thickness of the multi-layers and their order were used to modify the value of neutron buildup factors. Whetstone and Kearfott [102] found that thin alternating multi-layered steel and polyethylene designs had a similar transmitted neutron spectrum. They found no significant change in shielding performance of different layers, regardless of the layer stacking order.

In the case of X-ray shielding, McCaffrey et al. [97] observed that the low-Z upstream/high-Z downstream yield up to five times more attenuation at 50 kVp, than that of the reverse order material (high-Z/low-Z). However, this difference disappeared at 150 kVp. Kim et al. [103] disclosed that the high-Z/low-Z order was better than the opposite low-Z/high-Z order arrangement for 50 kVp X-rays. Türkaslan [104] fabricated multi-layered graphene oxide coated polyester/cotton nanocomposite based flexible fabrics for X-ray shielding. The graphene oxide coated polyester/cotton fabric was formed by layer-by-layer technique. The material was tested as an effective shielding material to replace the conservative lead-based shielding material. Figure 9 shows the setup for the X-ray irradiation. Single layered fabric or double layered fabric samples were positioned under acrylic step-wedge and then the digital images were verified. Figure 10 shows the scanning electron microscopy (SEM) images of graphene oxide (GO) multi-layered coated fabrics. The GO nanoparticles can be seen on the layered fiber surface. With the increase in layering, the GO particle density was found to be enhanced. The multi-layered films were studied as the cationic suspension in the poly(diallyldimethyl-ammonium chloride). Thus, the choice of a multi-layered material can be tailored for the particular radiation type and desired application. Table 2 shows a simple comparison of the specifications of various materials used for radiation shielding.

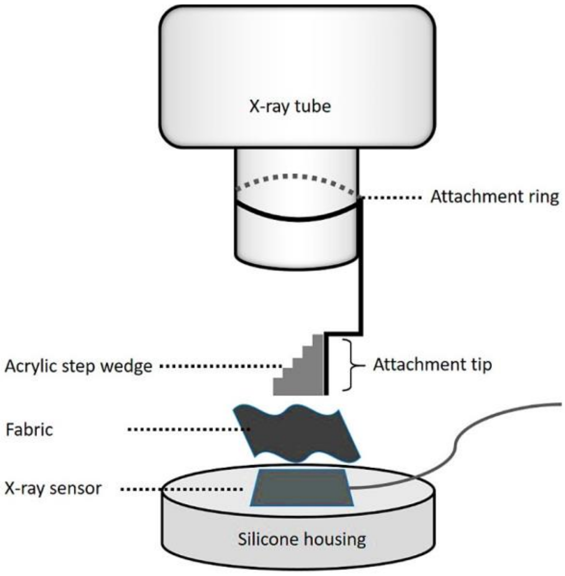


**Figure 9.** Schematic of X-ray irradiation setup [104]. Reproduced with permission from MDPI.

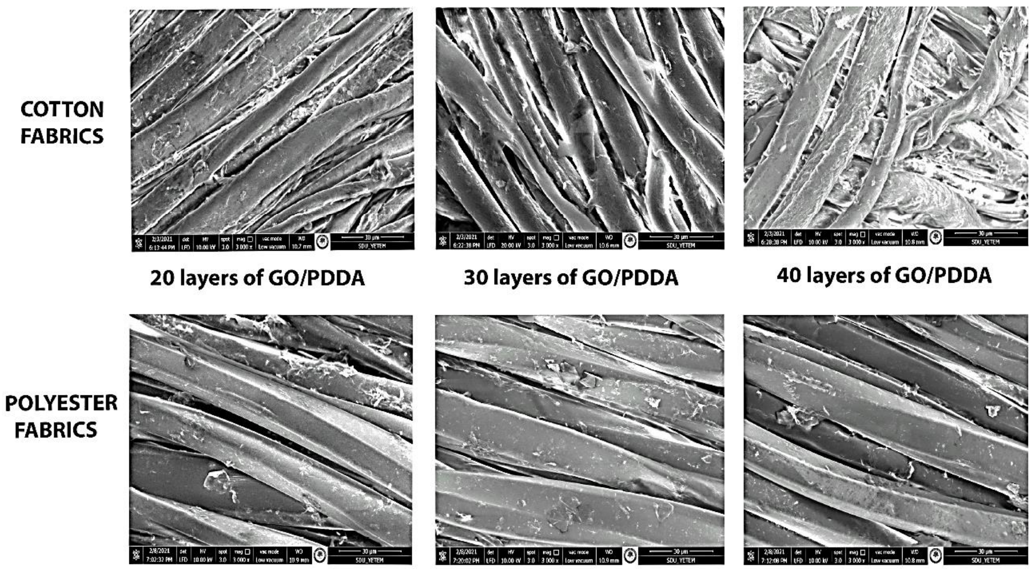


**Figure 10.** SEM micrographs of graphene oxide multilayered coated polyester/cotton fabrics [104]. PDDA = poly(diallyldimethyl-ammonium chloride). Reproduced with permission from MDPI.

**Table 2.** Properties of various composite materials used for shielding.

| Materials Utilized | Shielding Performance | Radiation | Ref. |
|---|---|---|---|
| Carbon Fabric/Polyether ether ketone | 17% higher than Aluminum (Al) (13 and 31 g/cm$^2$) | For heavy and light ions | [105] |
| Carbon fiber reinforced plastic and SiC composite plastic | Dose reduction 1.9 times compared with Al | - | [106] |
| Multilayered shield of composite/tantalum/composite | Radiation shielding efficiency equivalent to Al but 25% mass reduction | - | [6] |
| Carbon fabric coated with heavy metals | Flexible shield better performance than lead and Al shields | - | [11] |

**Table 2.** *Cont.*

| Materials Utilized | Shielding Performance | Radiation | Ref. |
|---|---|---|---|
| Natural materials (Boron, Aluminum, lead, etc.) mixed with cement | Hard shield better than lead and Al | Gamma & Neutrons | [107] |
| Tungsten composite and polyethylene terephthalate fiber fabrics containing $BaSO_4$ | Radiation shielding equivalent to 0.018 mm Pb and 0.03 mm Pb | Cosmic radiations | [20] |
| Poly(dimethyl sulfoxide) (PDMS)/multi-walled carbon nanotube nanocomposite | Lighter than pure Al and PDMS shields with comparable shielding effect | High Energy Protons | [108] |
| Graphene oxide coated nanocomposite flexible fabric | Promising for X-ray shielding | X-Rays | [104] |
| Graphene oxide paper | Study of chemical changes with 500 KeV proton irradiation | Low energy protons | [109] |
| Poly-methyl meth-acrylate/multi-walled carbon nanotube nanocomposite | 18–19% lighter in weight than Al and generated up to 5% fewer secondary neutrons | For stopping protons & secondary neutrons | [17] |
| Carbon fiber reinforced composites and polymers | Light weight, thermally stable | For atomic oxygen and UV protection | [22] |
| Ultra-high-molecular-weight polyethylene fiber reinforcement and hydrogen-rich poly-benzoxazine matrix | 44.6% dose reduction as compared to Al (Simulated results) | Galactic cosmic rays | [28] |
| High density polyethylene/BN and high density polyethylene/$B_4C$ composites | Superior shielding property for neutron shielding | Neutron shielding | [110] |
| Polyhedral oligomeric silsesquioxane coated 3D-graphene infused polyimide | High material durability of up to 10 years and low corrosion rate | For electrostatic discharge and atomic oxygen | [40] |
| Graphite and polyethylene composites | Effect of layered arrangement on radiation shielding | For Galactic Cosmic Ray Particles in high earth orbit | [29] |
| Poly-methyl meth-acrylate modification by colemanite (CMT) | CMT addition improve the shielding of beta particles | For electron shielding | [30] |
| Bismuth nanoparticles reinforced in polymer matrix | Bismuth nanoparticle addition produced lightweight shields as compared to micron particles-based materials having same shielding effect | For X ray shielding | [31] |
| Aluminum Bronze and molybdenum layers with a copper carrier | Shield can with stand 53–72% more ionizing dose than the conventional materials | For protons and electrons | [54] |
| Poly-methyl meth-acrylate/multi-walled carbon nanotube/Bismuth oxide-based nanocomposite | Promising material combination for electron shielding | For electron shielding | [60] |
| Epoxy/carbon nanofibers | Electromagnetic radiation absorption at 19.8 GHz and 2mm thickness | Electromagnetic interference shielding | [66] |
| Cotton fabrics/graphene oxide/zinc oxide | Electromagnetic interference (EMI) shielding effectiveness of 54.7 dB; EMI radiation absorption of ~90% | Electromagnetic interference shielding | [67] |
| Poly-dimethyl siloxane/reduced graphene oxide/ZnO nanowires | EMI shielding effectiveness of 27.8 dB at 9.57 GHz; shield thickness of 4.8 mm | Electromagnetic interference shielding | [68] |

## 6. Conclusions

In this review, the continuously developing interest for radiological protective materials has been surveyed. A material's ability to block radiation depends on its physical characteristics as well as the radiation source, composition, energy, duration of exposure, secondary radiations and thickness. The polymeric materials have also been found as potential options for mixed neutron-gamma ray shielding due to their advanced characteristics.

According to research reports, for radiation shields, inclusion of inorganic metal oxide nanoparticles has been found to reinforce in large amounts, such as 10–40 wt.%, gadolinium oxide in PMMA caused high radiation shielding. Similarly, 50 wt.% lead nanoparticles caused high radiation attenuation properties. The epoxy with S-glass fiber or basalt fiber filler maintained high tensile strength of the composite upon radiation exposure. These materials performed as efficient shields for neutron, X-ray and gamma radiations shielding without deteriorating the epoxy matrix. On the other hand, in epoxy/carbon fiber composites, mechanical properties were sustained but thermal characteristics were altered upon gamma radiation exposure. Therefore, carbon fibers were found less effective at maintaining the simultaneous radiation shielding and physical features, relative to S-glass fiber or basalt fiber filler in epoxy matrix. The best durability and stability performance was observed for borate glasses with CdO, $MoO_3$, $Al_2O_3$ and $TeO_2$ fillers (small amounts) in enhancing the absorption efficiency for radiation. Next, the poly-methyl methacrylate/colemanite nanocomposite with 15 wt.% nanofiller revealed high beta attenuation performance, when used in the Falcon-9 rocket. The multi-layered materials also possess better radiation shielding performance than single layered materials. Consequently, the multi-layered configurations yielded higher buildup factor, relative to single layer materials. Multi-layered polyester/cotton coated with graphene oxide has also been used as effective X-ray shield.

Numerous researchers have reported that polymers and composites with microscopic components (fillers) have improved radiation shielding properties. This innovative literature review highlights the fact that low atomic number polymeric materials were insufficient in attenuating/absorbing very intense ionizing radiation, such as gamma rays. Therefore, the use of different materials such as high atomic number elements (other than lead), metal oxides, graphitic nanofibers, etc., with polymers has been proposed as one of the emerging solutions to enhance radiation shielding efficiency. Consequently, polymeric composites or nanocomposites have been found as efficient lightweight gamma radiation shields, compared with pure lead. The ideal shield for protecting electrons may consist of a high-Z layer positioned in the middle of two low-Z layers. A multi-layered shield may efficiently reduce the dose for trapped electrons by >60%, compared with the single layered material shield. In several technical applications such as satellite structure, the outer and inner low-Z layers have already been used, but the high-Z component needs to be added. Thus, the ideal shield for protons can be a single layered low-Z material and high-Z can only be added as a middle layer. The evaluation of novel multi-layered designs may provide new research opportunities in shielding space materials. However, appropriate shield selection can be improved through primitive understanding of the fundamentals of interactions of ionizing radiation with matter and attenuation properties. Thus, advancements in this field may lead to the development of several high-performance composites/nanocomposites for spacecraft and other aerospace related shielding applications.

Polymeric nanocomposites with low nanofiller contents have benefits in better attenuation of cosmic, gamma, X-rays, neutron and other radiation, compared with the filler particles in traditional composites. Subsequently, the presence of nanoparticles with high surface-to-volume ratio has increased the probability of interactions between the radiation and the nanocomposites. Similarly, nanofillers have high electron density and can be uniformly dispersed in the polymer matrix, so enhancing the attenuation mechanism. Despite the advantages of the polymeric nanocomposites and related multi-layered materials for radiation shielding, there are several challenges in using these materials for high performance technical applications. An important concern of the radiation shielding materials studied here is to enhance the durability of materials by decreasing corrosion and other degradation effects. For this purpose, radiation shield designs must be improved by employing modified carbon and metal oxide nanoparticles. In addition, polymeric nanocomposite and multi-layered materials have shown superior radiation shielding properties in addition to better thermo-electric properties, corrosion resistance and mechanical stability characteristics. Nevertheless, numerous environmental issues concerning the use of such materials may arise, such as the rising pollution levels due to nanocomposite (polymer/nanofiller) residues which may be gathered in nature/landfills. Therefore, strategies must be developed towards using recyclable or sustainable materials and processes to maintain an eco-friendly environment. Hence, the researchers should be driven toward the use of sustainable materials, to reduce the possible negative impact of solid wastes on the environment.

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