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Polymer-based nuclear radiation shielding materials: state-of-the-art and emerging trends for engineering applications

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The continuous advancement of science and technology has led to the widespread application of nuclear technology across a diverse spectrum of fields, including scientific research, industrial processes, and particularly the medical domain for diagnostics and therapeutics. This proliferation, while beneficial, necessitates stringent protective measures for personnel who may be exposed to various forms of ionizing radiation, such as X-rays, γ-rays, and neutrons. In this context, polymer-based composite materials have emerged as a pivotal class of shielding solutions. These materials are typically fabricated by incorporating functional fillers—containing elements with high atomic numbers for photon attenuation or neutron-absorbing isotopes—into a continuous polymer matrix. This design strategy synergizes the processability and lightweight nature of polymers with the superior radiation shielding efficacy of the dispersed fillers, thereby offering robust and adaptable protection for individuals in nuclear-related occupations. This paper provides a comprehensive analysis of this material system. It begins by elucidating the fundamental attenuation and shielding mechanisms that govern the interaction of radiation with matter, establishing the theoretical foundation for material design. Subsequently, the paper offers a detailed review of the development history and recent research progress in polymer-based radiation shielding, tracing its evolution from conventional lead-impregnated rubbers to modern nanocomposites. The current research status of various material types is systematically summarized, highlighting technical achievements and innovative breakthroughs, such as the use of multi-layered structures or hybrid fillers. Furthermore, the paper analyzes the critical selection criteria for polymer matrices, considering factors like radiation resistance, thermal stability, and mechanical properties. Finally, it synthesizes the key challenges that remain to be addressed in current materials and provides a forward-looking perspective on future development directions, aiming to guide subsequent research and development efforts in this critical field.

KEYWORDS

nuclear radiation, polymer-based nuclear radiation shielding materials, nuclear radiation protection, polymer matrices, shielding mechanisms

1 Introduction

Human understanding of nuclear radiation originated at the end of the 19th century, and in the 1940s, nuclear technology was used in the military field and played a great role in ending World War II. After the 1950s, nuclear technology ushered in a rapid development phase in both civil and military fields, including civil nuclear power plant (Fuhrmann, 2009a; Fuhrmann, 2009b), irradiation modification and processing of industrial materials (Xu et al., 1999; Gao et al., 2006), medical and healthcare (Hung et al., 2023; Tekin et al., 2018), agricultural breeding (Havlickova et al., 2024; Pacher and Puchta, 2017), as well as oil field logging, archaeological fields (Fuhrmann, 2009a; Fuhrmann, 2009b; Smethurst et al., 2017; Festa et al., 2020), development and application of nuclear-powered carriers, nuclear-powered submarines, nuclear weapons, and related nuclear materials (Adamy, 2022; Press, 2022; Gillies and Haylock, 2022; Ibrahim, 2021; Townsend et al., 2022). Although nuclear technology has brought significant benefits and strategic deterrence to humanity in both civil and military fields, it can also cause irreversible damage to the health of related practitioners. For example, genetic mutations, immune system damage, skin damage, and acute radiation syndrome induced by nuclear radiation (Mendelsohn et al., 2016; Madan et al., 2015). To ensure the personal safety of operators in the radiation environment, it is essential to enhance the safety protection of practitioners.

In recent years, X/γ-ray and neutron shielding materials have become a research hotspot in the field of nuclear radiation protection. According to the shielding mechanism of X/γ-rays and neutrons and extensive related research work, the reduction of the nuclear radiation dose can be realized by the following three approaches (Cheon et al., 2018): (1) reducing the time of exposure to the nuclear radiation environment, (2) increasing the distance from the source of radiation, and (3) placing of shielding materials between the human body and the radiation source. X/γ-ray shielding materials should have high density and high atomic number (Z), which are the most important properties of X/γ-ray shielding materials (AbuAlRoos et al., 2019; Singh et al., 2014; Chang et al., 2015). Lead (Pb) and lead-containing materials are widely used in the field of individual nuclear radiation protection after being compounded with polymers owing to their low cost and satisfactory shielding capabilities (Ihsani et al., 2024; Rammah et al., 2020). However, Pb is highly biological toxicity and environmentally harmful (Ogawa et al., 2008; Hsiao et al., 2011), and it easily migrates out of the polymer matrix, so it has been gradually phased out in the field of individual nuclear radiation protection and replaced by other non-toxic or low-toxic high-atomic-number (high-Z) heavy-metal shielding elements, such as bismuth (Bi), tungsten (W), tantalum (Ta), and other metal monomers or compounds. Neutron shielding materials can be made from metals (e.g., cadmium (Cd), iron (Fe), gadolinium (Gd), europium (Eu)), nonmetals (water, boron (B)), concrete, polymers (natural rubber (NR), polyethylene (PE), and other materials with high hydrogen (H) content), and composites of the above materials, which have high macroscopic neutron absorption cross-sections (Fu et al., 2021). Currently, neutron shielding is mainly achieved using PE/B composites and B-reinforced concrete for fast neutron shielding (Jumpee et al., 2020; Tuna and Bayrak, 2017).

The aim of this review is to summarize the current research status and development trends of different types of polymer-based nuclear radiation shielding materials and analyze the development needs of polymer-based nuclear radiation shielding composites. When designing radiation shielding materials for defensive structures and individual protection, their protective properties against X/γ-rays, neutrons, radioactive fragments, and strong charged particles must be considered. The electromagnetic spectrum contains various radiations classified by wavelength (or frequency), as shown in Figure 1. These radiations can be broadly divided into ionizing and non-ionizing radiation, and this analysis focuses on ionizing radiation.

2 Criteria for the selection of materials for nuclear radiation protection

To select suitable nuclear radiation shielding materials, it is crucial to understand the type of nuclear radiation (particles or photons), the interaction mechanisms with shielding materials, and whether secondary radiation is generated. Interactions of energetic X/γ-rays with shielding materials.

2.1 Interaction of energetic X/γ-rays with shielding materials

The interactions of high-energy X/γ-rays with matter consists of the following three main forms: the photoelectric effect, Compton scattering, and the pair production effect (Zeng et al., 2023). When an incident photon interacts with matter through the three aforementioned interactions, there exists a certain relative probability. This probability is typically quantified by the atomic cross-section (σ), which represents the interaction probability between an incident photon and a single target atom per unit area. Specifically, σ_{ph} denotes the photoelectric effect cross-section, σ_c the Compton effect cross-section, and σ_p the pair production effect cross-section (Li et al., 2024). The magnitude of the cross-section is related to the energy of the incident photon and the properties of the target material.

2.1.1 Photoelectric effect

If the energy of the incident photon is higher than the binding energy of the electron in the shielding element, the electron is ejected from the atom by the photon and the remaining energy of the photon is converted into the kinetic energy of the electron (as shown in Figure 2). This process occurs mainly in the 10 keV–100 keV energy range and is enhanced as the atomic number of the shielding material increases. The interaction cross-section of the photoelectric effect (t) is given by the following Equation 1 (Rooyen, 2020; Jaeger et al., 2013; Bijanu et al., 2021; Jayakumar et al., 2023):

$$\sigma_{ph} \propto \frac{Z^4}{(h\nu)^3} \tag{1}$$

Where σ_{ph} is the interaction cross-section, Z is the atomic number of the shielding material, h is Planck's constant, and ν is the photon frequency, ∝ is positive correlation symbol.

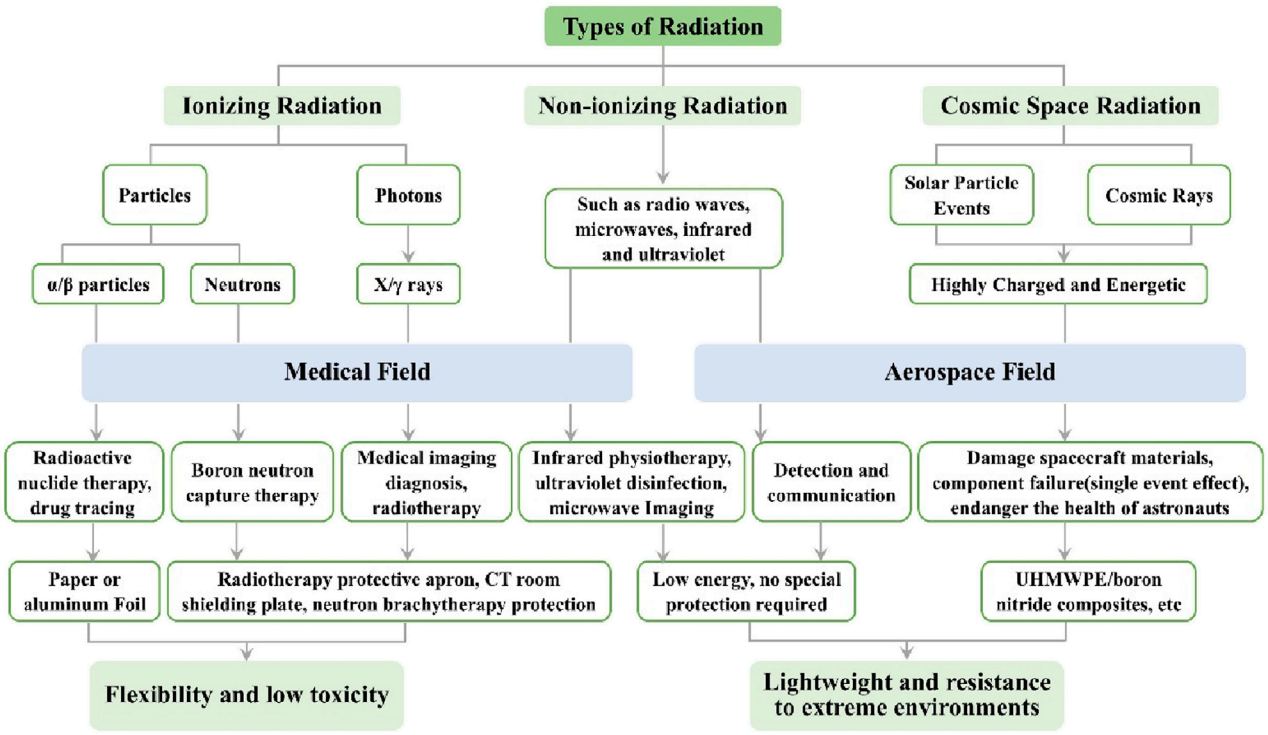

FIGURE 1
Types of radiation and corresponding countermeasures.

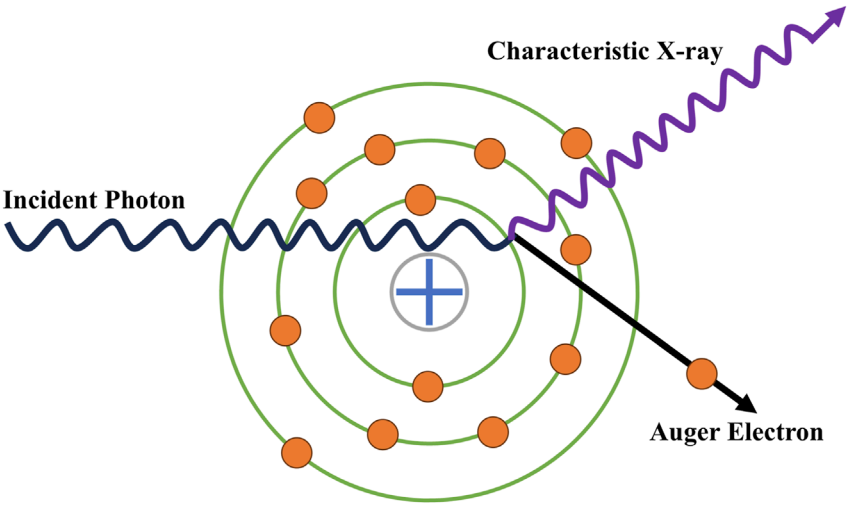

FIGURE 2
Schematic diagram of photoelectric effect.

2.1.2 Compton scattering

Incident photons undergo inelastic scattering with electrons in the elements of the shielding material (Figure 3), and the energy of the photons decreases and dissipates while part of the energy is transferred to the electrons (Rooyen, 2020). The cross-section of the Compton scattering interaction (σ_c) is given by the following Equation 2 (Rooyen, 2020; Jaeger et al., 2013; Bijanu et al., 2021; Jayakumar et al., 2023):

$$\sigma_c \propto Z \tag{2}$$

Where σ_c is the Compton scattering cross-section, and Z is the atomic number of the shielding material.

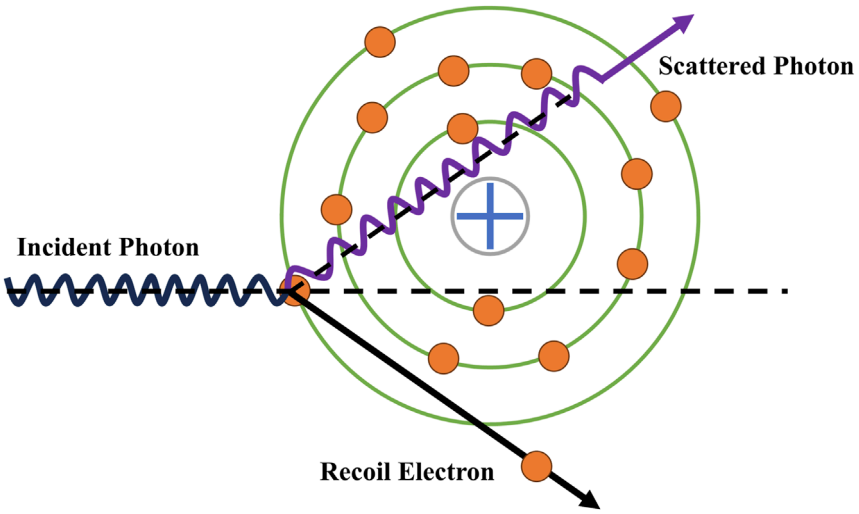

FIGURE 3
Schematic diagram of Compton scattering.

2.1.3 Pair production effect

If the incident photon energy is greater than twice the electron rest mass (0.511 MeV), i.e., the incident photon energy exceeds 1.02 MeV, the photon interacting with the nucleus produces an electron and a positron (Nambiar and Yeow, 2012), as shown in Figure 4. During electron pair generation, the incident photon interacting with the nucleus generates a particle pair: an electron and its antiparticle (positron). Excess energy is converted into the kinetic energy of the electron and positron, with the emitted electron and positron slowed down by Coulomb interaction (Rooyen, 2020). When the positron loses most of its energy, it recombines with its antiparticle (i.e., the electron) and two gamma photon beams with energies of at least 0.511 MeV are produced by annihilation. These photons travel in opposite directions, approximately 180° apart, to conserve linear momentum. The cross-section of the pair production effect interaction (σ_p) is given by the following Equation 3 (Jaeger et al., 2013; Bijanu et al., 2021; Jayakumar et al., 2023):

$$\sigma_p \propto Z^2 \tag{3}$$

2.2 Interactions between neutrons and shielding materials

Neutrons are neutral particles and have no charge of their own, so neither the electrons nor the positive charges around the nucleus alter the trajectory of the neutron, which only deviates from its original trajectory when it interacts with the nucleus. The interaction of neutrons with matter can be divided into two categories.

2.2.1 Scattering

Neutrons can undergo elastic or inelastic scattering. In elastic scattering, when the target nucleus interacts with a neutron, it emits a neutron, with no energy transfer during nuclear excitation. In elastic scattering, maximum energy loss occurs when colliding with another particle of equal mass. Thus, the lower the atomic number, the greater the energy loss in a single collision. When a low-energy neutron collides, part of its energy is transferred to the nucleus as kinetic energy, and total kinetic energy is conserve (Rinard, 1991). Therefore, low-atomic-number elements are more suitable for neutron shielding via elastic scattering. Hydrogen, the element with the lowest atomic number, has the highest neutron shielding efficiency. For heavier nuclides, however, energy loss from elastic collisions is practically negligible, and inelastic scattering predominates. In inelastic collisions, the nucleus is excited to higher energy levels and can decay to the ground state by emitting one or more γ-rays.

2.2.2 Absorption

During absorption, neutrons are completely absorbed, forming a compound nucleus. The formation of a compound nucleus can be understood as an interaction process between a neutron and a nucleus, which alters the nuclear internal structure and forms a new, relatively stable nucleus (i.e., a compound nucleus). This process involves the following main steps: (1) Neutron capture: the nucleus captures a neutron, absorbing the neutron's kinetic energy; the neutron itself is converted into a proton (or a neutron and a proton), releasing one or more γ particles (i.e., high-energy electromagnetic radiation). (2) Changes in nuclear internal structure: When a neutron is captured by the nucleus, it is converted into a proton, changing the number of protons and neutrons in the nucleus. This increases the atomic number, forming a new nucleus. (3) Energy release: After neutron capture, the newly formed compound nucleus is generally unstable and reaches a more stable state by emitting γ-rays or releasing energy via β-particles (electron or positron emission), etc. (4) Compound nucleus decay: A compound nucleus can decay over time, releasing energy and transforming into another atomic nucleus.

During this process, neutrons are completely absorbed, and the nuclear structure is altered, enabling energy transfer and

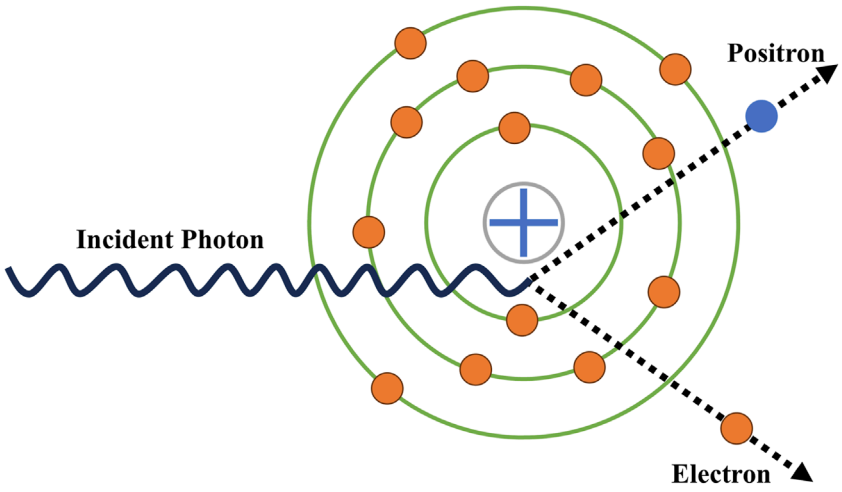

FIGURE 4
Schematic illustration of pair production effect.

transmission. When the energy of incident photons exceeds 10 MeV, photoneutrons are generated (Naseri and Mesbahi, 2010). Thus, a suitable shielding material must be selected based on the radiation intensity in the environment. For shielding against high-energy X/γ-rays, materials with high Z-values and high density are suitable, as elements with higher atomic numbers have larger interaction cross-sections (Equations 1–3). Neutrons, as uncharged particles, can easily penetrate denser materials and generate secondary neutrons and γ-rays when they interact with the nucleus. Therefore, neutron shielding typically uses low-Z materials (H, B, Li, Gd), which tend to have larger interaction cross-sections. However, these low-Z elements emit secondary radiation when they interact with the nucleus, generating γ-rays and neutrons. For example, Cd can emit γ-rays with energies up to 9 MeV after neutron capture (Hogan, 2011). Similarly, after neutron capture, B and H can emit secondary γ-rays with energies up to 2.79 MeV (L'annunziata, 2016) and 2.225 MeV (Podgoršak, 2016), respectively, while Li can emit fast neutrons with energies up to 16 MeV (Santoro et al., 2016). Therefore, materials containing B and H elements are preferred as conventional neutron shielding materials because they generate less high-energy secondary radiation after interaction, ultimately reducing the required thickness of shielding materials.

Nuclear radiation shielding materials used in various scenarios include Pb, concrete (light/heavy), lead glass, metal/polymer composites, and barite plates. Radiation shielding concrete (light/heavy), Pb and barite plates are mainly used for the construction of radiation shielding structures in nuclear power plants, radioisotope facilities, hospitals, sterilization facilities, etc. Concrete, in particular, has excellent shielding properties against neutrons and high-energy photons. Optically transparent Pb glass is often used for transparent windows in nuclear radiation shielding structures. Pb plates and other Pb composites were among the first materials used in nuclear radiation protection. Pb plates and other Pb composites were the first materials used to manufacture personal protective equipment, such as nuclear radiation protective suits and gloves, which are used to protect the vital organs of personnel working near nuclear radiation (Wang et al., 2021). However, shielding materials made from the above materials are bulky and heavy, suitable only for specific radiation protection scenarios, making them difficult to use in portable nuclear devices, manned spacecraft, and radiation protective suits worn by nuclear workers and medical personnel. Cosmic radiation mainly includes galactic cosmic radiation (GCR), solar particle events (SPEs) (Council and Board, 1997; Cucinotta et al., 2006), and secondary neutrons. Therefore, protection against cosmic rays is mainly provided by composite materials containing H and B with large interaction cross-sections, such as high-Z metal-polymer composites, which can effectively shield neutrons while attenuating high-energy photons. High-Z metals increase the probability of interaction between incident photons and matter with higher attenuation coefficient (AbuAlRoos et al., 2019); the probability of interaction between shielding materials and nuclear radiation photons is directly related to the individual interaction cross-section and atomic number. The interaction cross-section of photons with matter varies with the type of interaction (Equations 1–3) and the energy of the radiation. Since organic polymers contain a large amount of H, they are suitable for absorbing neutrons. Doping B and its compounds into polymers makes them highly effective in neutron shielding. Polymer composites can therefore provide good shielding for both ionized photons and neutrons. Based on this research context, high-Z metal-polymer composites for protection against nuclear radiation will be discussed later in this article.

3 High-Z metals and their compounds-polymer composites

Materials made by combining polymers with high-Z metals and their compounds are called metal-doped polymer composites. In these materials, metal compounds are uniformly dispersed in the polymer matrix via physical doping or chemical bonding interactions. Since the attenuation of nuclear radiation depends

TABLE 1 Densities and linear attenuation coefficients (μ) of common polymers.

Polymer	Chemical formula	Density (g/cm^3)	Linear attenuation coefficient at 122 keV (cm^{-1})
Polyamide (Nylon 6) (PA-6)	$[C_6H_{11}NO]_n$	1.13	0.1749 ± 0.0069
Polyacrylonitrile (PAN)	$[C_3H_3N]_n$	1.18	0.1854 ± 0.0079
Polyvinylidene chloride (PVDC)	$[C_2H_2Cl_2]_n$	1.63	0.2663 ± 0.0061
Polyaniline (PANI)	$[C_6H_4NH]_n$	1.36	0.2018 ± 0.0079
Polyethylene terephthalate (PET)	$[C_{10}H_8O_4]_n$	1.38	0.2030 ± 0.0038
Polyphenylene sulfide (PPS)	$[C_6H_4S]_n$	1.35	0.2155 ± 0.0088
Polypyrrole (PPy)	$[C_4H_3N]_n$	1.48	0.2181 ± 0.0085
Polytetrafluoroethylene (PTFE)	$[C_2F_4]_n$	2.20	0.3130 ± 0.0058

entirely on the shielding material, it is necessary to select suitable metals/metal compounds and a high-performance co-doped polymer matrix to produce composites with high attenuation coefficients, high shielding efficiency, and good mechanical properties.

3.1 Polymer materials for nuclear radiation shielding matrix

In the manufacture of polymer composites for nuclear radiation protection, the ideal polymer matrix must have good mechanical strength, thermal and chemical stability, non-toxicity, and excellent nuclear attenuation properties. Polymers currently used as matrices for polymer-based radiation shielding materials include polyurethane (PU), polyvinyl alcohol (PVA), low-density polyethylene (LDPE), high-density polyethylene (HDPE), silicone polymers, and ethylene-vinyl acetate (EVA) copolymers (Muthamma et al., 2019; Dubey et al., 2016; Eren Belgin and Aycik, 2015; Molina Higgins et al., 2019; Erol et al., 2016; Soylu et al., 2015; Husain et al., 2018). The densities of some polymer matrices commonly used in the preparation of polymer-based nuclear radiation shielding materials and the attenuation coefficients at 122 keV are presented in Table 1 (Stawarz et al., 2019).

As shown in Table 1, the attenuation coefficient of polymer materials increases with increasing density. Despite their lowest density, PE materials are widely used as matrix materials for radiation shielding of space probes against GCR and SPE interference, owing to their high H content with a high neutron interaction cross-section. In addition, PE materials have excellent formability and impact resistance, and a low coefficient of friction. Depending on the requirements of the application scenario, three types of PE materials are commercially available as substrates for nuclear radiation shielding materials: LDPE, HDPE, and ultra-high molecular weight polyethylene (UHMWPE). UHMWPE, in addition to all the excellent properties of HDPE, also has a lower coefficient of friction than HDPE (Mansouri et al., 2020). Silicone polymers are also used in a wide range of applications for matrix materials for nuclear radiation protection. Polydimethylsiloxane (PDMS), for example, is a widely used silicone polymer that is non-toxic, has good elasticity, and is easy to process and mold (Grassetti et al., 2015; Nambiar et al., 2013). Commercially available epoxy resins are also commonly used as matrices for nuclear radiation shielding materials, due to their low cost, low shrinkage, ease of processing and molding, and excellent resistance to weathering and corrosion (Nawab et al., 2013; Stawarz et al., 2019). Different polymers with the different physicochemical properties mentioned above can be used with various high-Z metals/metal composites to prepare diverse composite materials for nuclear radiation protection.

3.2 X/γ-ray shielding materials

3.2.1 Pb-based and Pb-compound X/γ-ray shielding materials

For almost a century, traditional flexible X/γ-ray shielding materials have been used with Pb or Pb compounds as shielding components. Since Pb and Pb compounds have high density, high interaction cross-sections and relatively low cost, millimeter-graded lead has a shielding efficiency of over 90% for X/γ-rays and good attenuation shielding for X/γ-rays. They are used as protective collars, gonad protectors, protective aprons, and protective gloves, etc., to protect operators in nuclear radiation environments (Singer, 2005; Bartal et al., 2018; Honigsberg et al., 2017). For example, Pb, lead oxide (PbO), lead dioxide (PbO_2), lead sesquioxide (Pb_2O_3), lead sulfide (PbS), lead silicate ($PbSiO_3$), lead nitrate ($Pb(NO_3)_2$), and lead chloride ($PbCl_2$) are some lead compounds with high density and sufficient mass attenuation coefficients (Rabie et al., 1979; Rudraswamy et al., 2010), which are dispersed as fillers in the polymer matrix for nuclear radiation shielding materials.

The combination of lead-containing compounds and a polymer matrix for γ-ray shielding materials was first reported in 1991 by Abdel-Aziz et al. (1991), who prepared γ-ray shielding materials by mixing PbO as a doped filler with styrene-butadiene rubber (SBR) as the polymer matrix. The linear attenuation coefficient of the composites was found to increase with the volume fraction

of PbO, reaching a saturation point at a PbO volume fraction of 5.6%. The tensile strength, elongation at break, and coefficient of volume expansion of the composites showed a downward trend with increasing PbO volume fraction. The shielding capacity of PbO-filled SBR for γ-rays (^{60}Co as radioactive source) reaches 63% of that of pure Pb. In 2009, Harish et al. (2009) doped PbO into isophthalic acid resin, achieved uniform dispersion of both via a high-speed physical stirring method, and then obtained resin-based nuclear radiation shielding composites via a room-temperature molding method. When the mass percentage of PbO is 30% or more, the composites have a good γ-ray attenuation effect, in particular, the mass attenuation coefficient of 0.0948 cm^2/g for γ-rays of 0.662 MeV for composites with a mass fraction of 50%. In 2013, Azman et al. (2013) used micrometer-sized PbO doped into epoxy resin to fabricate radiation shielding materials. Epoxy resins are thermosetting polymers with improved mechanical properties and thermal stability compared to thermoplastic polymers, and are therefore resistant to higher-energy X-rays. The mass attenuation coefficients of the 50 wt% PbO/epoxy and 50 wt% Pb_3O_4/epoxy composite samples were 0.0978 cm^2/g and 0.0974 cm^2/g, respectively, at γ-irradiation of 0.662 MeV. Wei and co-workers (Wei et al., 2021) used room-temperature-cured paraffin wax (Pn)@lead-tungstic acid (PWO) fillers dispersed in silicone rubber of phase change microcapsules (filling volume 60 phr) with different core-shell ratios to fabricate flexible thermally controlled radiation-shielding materials. The results showed that microcapsules with different core-shell ratios were uniformly dispersed in silicone rubber, and the particle size of the microcapsules decreased as the core-shell ratio decreased, which could further improve the mechanical properties and γ-ray shielding performance of silicone rubber/(Pn@PWO) composites. Under γ-ray irradiation at 105.3 keV, the mass attenuation coefficient of silicone rubber/(Pn@PWO-1:2) is 2.05 cm^2/g, 144.05% higher than that of pure silicone rubber (0.84 cm^2/g). Due to the doping of phase change microcapsules, silicone rubber/(Pn@PWO) can be stabilized via solid-liquid phase change processes under non-isothermal conditions for temperature control and thermal management purposes.

Despite Pb and its oxides exhibit excellent X/γ-ray shielding properties, Pb or Pb-containing compounds have several disadvantages. Firstly, Pb as an X/γ-ray shielding materials are inherently bulky and less flexible. For example, a lead apron with a lead equivalent of 0.5 mm weighs 5 kg and also presents problems such as low flexibility and poor air permeability (Ni et al., 2016; Zuguchi et al., 2008). To overcome the above-mentioned problems, Pb-based radiation shielding fibers with some flexibility and air permeability can be prepared by spinning lead and its oxides into staple fibers or by spinning them as functional particles with fiber-forming polymers. For example, Mirzaei et al. (2019) prepared X-ray protective fibers by adding lead and tin as protective components in a polypropylene (PP) spinning solution, and knitted them by melt-spinning technology, obtaining fabrics with excellent flexibility, breathability, and X/γ-ray shielding capability.

Secondly, the shielding efficiency of Pb or Pb-containing compounds has a weak absorption region within a specific energy band (Jamil et al., 2019). For example, the shielding efficiency of Pb and its compounds for X/γ-rays in the energy range of 50–88 keV is low, which is referred to as the weak absorption region of Pb. To mitigate the effect of the weak absorption region of Pb and its compounds, blend or modify Pb and its compounds with other heavy metal materials to form X/γ-ray shielding materials via synergistic effects (McCaffrey et al., 2007). Yaffe et al. (1991) prepared an apron for X-ray shielding by combining Pb with substances containing W and Ba elements. The introduction of W and Ba elements not only reduced the use of Pb, but also decreased the weight of the new X-ray shielding apron by approximately 30% while maintaining the same shielding efficiency. Lin et al. (2015) developed an X-ray protective vest by applying a polyurethane resin mixture containing silicon (Si), Pb, stannum (Sn), titanium dioxide (TiO_2), and other compounds as X-ray protection functional fabrics to an open mesh sandwich fabric (SAMF). The polyurethane resin was well bonded to the SAMF, effectively filling the gaps between the fabric and X-ray shielding particles, and the SAMF also effectively prevents X-ray protection functional particles from impairing the fabric's flexibility. The X-ray shielding efficiency of this double-layer fabric reaches 98.47%. Hashemi et al. (2018) found that graphene oxide (GO) modified with Pb_3O_4 can significantly enhance the X-ray shielding effect. The team selected X-ray energies of 40 keV, 60 keV, and 80 keV for testing and found that, for the same amount of doped shielding components, the shielding effectiveness of GO-doped Pb_3O_4 increased by 124.3%, 124.6%, and 103.6%, respectively, compared with undoped Pb_3O_4. However, the material's shielding effectiveness decreased with increasing X-ray tube voltage.

Finally, the biggest problem with X-ray shielding materials containing Pb is the biotoxicity, low stability and volatility of Pb, which can cause damage to the nervous system and other tissues and organs, with Pb being even more harmful to children's nervous systems (Specht et al., 2019; Frith et al., 2005; Frankel, 1994). Prolonged use of protective clothing containing Pb can lead to Pb poisoning, and the lead plates they contain can also break down after prolonged use, causing the protection to fail (Lakhwani et al., 2019).

3.2.2 Non-Pb heavy metal and compound X/γ-ray shielding materials

In recent years, people have gradually become aware of the biological toxicity and environmental hazards of lead-containing materials. Thus, in the field of nuclear radiation safety and personnel protection, other high-atomic-number heavy metals-such as tungsten (W), Bi, barium (Ba), antimony (Sb), cerium (Ce) and Gd-have been widely used in the shielding function of X/γ-ray shielding materials (McCaffrey et al., 2007). Simulation studies by Wang and Zou (2011) showed that the attenuation coefficient of W is significantly higher than that of Pb when the photon energy is greater than 500 keV, but the difference is not evident when the photon energy is less than 500 keV. Given the low toxicity of W and Bi—with W exhibiting better shielding effects than Pb for medium-energy sources (via the Compton effect) and Bi outperforming Pb for low-energy sources (via the photoelectric effect)—W and Bi are effective Pb replacements (AbuAlRoos et al., 2019). Yue et al. (2009) used W instead of Pb as the functional shielding component. They ball-milled and mixed styrene-butadiene-styrene copolymer (SEBS) with W powder, then hot-pressed the mixture at 120 °C to obtain samples 0.3 ± 0.01 cm. When 85% W was added to the SEBS matrix, the prepared composite had a maximum density of 4.4 g/cm^3 and the γ-ray attenuation at 9 MeV and 12 MeV was better than that of Pb with the same doping

level. Dong et al. (2012) modified WO_3 and cerium dioxide (CeO_2) via surface grafting and used epoxy resin as the substrate to prepare multilayer WO_3/CeO_2/epoxy resin radiation shielding materials. The linear attenuation coefficient of this material was calculated using EGSnrc software and compared with results from actual γ-ray tests. The simulation-calculated linear attenuation coefficients were found to be essentially consistent with the experimental results. A comparison of the shielding performance of the bilayer structure showed that the shielding performance of the multilayer material preceded by CeO_2 is better in the low energy region and the shielding performance of tungsten trioxide (WO_3) is better in the high energy region. Yu et al. (2012) prepared WO_3/E44 nanoscale and micrometer-scale samples, via a solidification molding method, and found that the WO_3/E44 nanoscale samples have better shielding properties in the γ-ray field, which was attributed to the nanoscale shielding functional components with larger γ-ray interaction area. Zhang (2015) modified carbon nanotubes with functional particles (WO_3, Sm_2O_3), then combined the modified carbon nanotubes with a modified epoxy resin matrix to obtain composites. They found that the material significantly improves shielding performance under low-energy γ-radiation conditions and exhibits better shielding performance than materials prepared by physically mixing functional particles, carbon nanotubes, and epoxy resin matrix. Kim et al. (2014) prepared PE/W composite particles by ball milling, in which W particles were well ground, encapsulated and dispersed in PE, and then W/PE nanoparticles dispersed in an ethylene propylene (EPM)/HDPE monomer matrix via melt blending technique. Nanoscale W composite polymers were shown to improve γ-ray shielding performance by 75% at 0.3 MeV compared to micrometer-scale W composites. Erol et al. (2016) reported a composite polymer using HDPE with microscopic-grade tungsten carbide (WC) and boron carbide (B_4C) as fillers for γ-ray and neutron protection. The samples were prepared by homogeneously mixing the aforementioned matrix materials and the functional shielding components and then forming them into sheets about 1 mm thick using a heat press. The sheet containing 50% WC, 20% B_4C, and 30% HDPE exhibited 4.32% and 0.81% higher attenuation factors than Pb of the same thickness at 662 keV and 364 keV, respectively. Moonkum et al. (2023) showed that barium sulfate ($BaSO_4$) and bismuth oxide (Bi_2O_3) powders aggregated and unevenly dispersed in silicone rubber by mixing $BaSO_4/Bi_2O_3$ powders with different amounts of filler (10, 30, 50, 70 wt%) with silicone rubber and curing the mold at room temperature when the amount of filler was more than 50 wt%. The X-ray absorption and linear attenuation coefficients of $BaSO_4$ and Bi_2O_3 composites increased with the amount of filler, and when the amount of filler was 70 wt%, the radiation absorption was 90.19% and 94.87%, respectively, at a tube voltage of 120 kVp. Dubey et al. (2016) prepared Bi_2O_3-PDMS composite materials using Bi_2O_3 as the shielding functional component and PDMS as the matrix material for shielding gamma rays emitted by ^{241}Am. The atomic numbers of Bi and Pb are close, and Bi and its oxides are less toxic, while PDMS has better heat resistance and mechanical and chemical stability. When 30 wt% of Bi_2O_3 was doped into PDMS, the shielding efficiency of the composite for γ rays emitted by ^{241}Am reached 70%. Soylu et al. (2015) prepared shielding composites with WC contents of 0%, 60%, and 70%, respectively, using EVA copolymer as the matrix material and WC as the functional shielding component. It was found that at a γ-ray energy of 662 keV, composites with 70 wt% WC had the best γ-ray shielding effect: the 1-mm thick sample exhibited a shielding efficiency of 86.9% ± 0.1%, while the 2 mm thick sample had a γ-ray shielding efficiency of 93.3% ± 0.2%. Eren Belgin and Aycik (2015) used PbO and WO_3 as shielding components doped into linear low-density polyethylene (LLDPE), and conducted a study on shielding performance against γ-rays. Four composites with different contents were prepared with mass fractions of 10, 15, 20, and 25% of the shielding functional components. It was found that PbO and WO_3 exhibited excellent γ-ray attenuation performance when their combined mass fraction in the composites was approximately 25%–30%. In the energy range from 60 keV to 1836 keV, the mass attenuation coefficients of composites with a mass fraction of 25%–30% of shielding functional components are higher than those of Pb/LLDPE composites with the same mass fraction. Wei et al. (2013) prepared three types of rubber-based radiation shielding materials, namely, active Bi_2O_3, bismuth tungstate (Bi_2WO_6) and Bi_2O_3/Bi_2WO_6 (7:3), respectively, and compared the γ-ray shielding performance of the three at a rubber composite thickness of 1 mm. The experimental results showed that Bi_2O_3/rubber exhibits better shielding performance when the doping level of the shielding functional component is less than 25%. When the doping level exceeds 25%, the Bi_2O_3/Bi_2WO_6/rubber ternary composite system shows better γ-ray shielding performance. The shielding efficiency of the ternary system can reach 24.94% at a doping level of 40%. Akman et al. (2020) added barium titanate ($BaTiO_3$) and calcium tungstate ($CaWO_4$), respectively, to the polymer matrix to obtain polymeric shielding materials. According to experimental and theoretical results, when $BaTiO_3$ and $CaWO_4$ each account for 20 wt% in the polymer matrix, the composites exhibit better shielding effects, and for the same mass fraction, the $CaWO_4$-doped sample shows better shielding performance than the $BaTiO_3$-doped sample.

Rare earth elements are also a class of materials with excellent X/γ-ray shielding ability, especially in the low and medium energy ranges, where their shielding effect surpasses that of heavy metals such as Bi, W, and Pb. An et al. (2006) prepared resin/nano-Ce composites via ion exchange and found that resin/nano Ce composites with the same content of shielding components have stronger X-ray shielding ability than conventional $Ce(NO_3)_3$ materials for X-ray radiation and can be used for X-ray shielding in the low and medium energy bands. Jia (2017) introduced La or Ce into barium tantalate ($Ba_5Ta_4O_{15}$) and prepared rubber matrix composites. With a constant molar amount of Ta elements, the composites achieved a shielding ratio of 38.66% at 16% Ce doping or 37.25% at 12% La doping. Gao et al. (2019) showed that SBR rubber molecules with high styrene content, which are composed of styrene and butadiene chain segments, have good radiation shielding properties, and the shielding effectiveness of lead-free SBR-based shielding materials is approximately 11.6% higher than that of lead-boron vinyl. Zhang (2021) investigated the shielding properties of silicone rubber-based composites with different filler contents (10%–70%) and fillers (Fe, $BaCO_3$, $BaSO_4$, Pb) at different energy levels (0.66 MeV, 1.173 MeV, 1.332 MeV) using γ-ray spectrometry and found that γ-ray shielding was the best for Pb/silicone rubber composites and Fe/silicone rubber composites were the next best. Although the shielding effect increases with the filler content, the corresponding composite is more difficult to prepare. Peyman et al.

(2018) prepared flexible lead-free γ-ray composites based on a room-temperature-vulcanized silicone rubber matrix, with 37.5% W and Bi_2O_3 as protective components. The linear attenuation coefficient, transmittance, half-value layer (HVL), and ten-value layer (TVL) of the composites were determined experimentally. Under ^{152}Eu radioactive source conditions in the 122–964 keV energy range, the composites were found to exhibit superior γ-ray shielding characteristics compared with commercial radiation shielding (76 wt% Pb). The radiation shielding composites show the lowest HVL and TVL values in the γ-ray energy bands and the best γ-ray shielding when they contain 18.75 wt% W and 18.75 wt% Bi, respectively. İrim et al. (2018) mixed hexagonal boron nitride nanoparticles (h-BN) and gadolinium oxide nanoparticles (Gd_2O_3) with HDPE to obtain HDPE-based composites. The ternary composites showed 52% higher γ-ray protection efficiency than pure HDPE.

Li J. S. et al. (2011) prepared Sm_2O_3/epoxy and samarium acrylate/epoxy radioprotective materials via surface treatment and graft polymerization, respectively. When comparing the mechanical properties of the two composites, it was found that the mechanical properties of the samarium polyacrylate/epoxy resin were better than those of the Sm_2O_3/epoxy resin. With ^{238}Pu as the radioactive source, the Sm_2O_3/epoxy resin (2 cm) exhibits shielding efficiencies of 71.64% and 25.44% under 79.9 keV and 167.6 keV radiation, respectively. For the 2 cm samarium polyacrylate/epoxy resin, the shielding efficiencies are 67.57% and 41.26% under 79.9 keV and 167.6 keV radiation, respectively. In terms of shielding efficiency, the shielding performance of the rare earth element Sm or the rare earth element Er is better than that of Pb in the irradiation of low-energy radiation, which can be used in medical emission sites or nuclear power workplaces (Li J. et al., 2011; 2010; Li, 2011). Dong et al. (2009) prepared $La_2(CO_3)_3$, $Ce_2(CO_3)_3$, $Pr_2(CO_3)_3$ and $Nd_2(CO_3)_3$/thermoplastic polyurethane (TPU) composites via mechanical blending. The results showed that at an X-ray emission tube voltage of 120 kV, the $Nd_2(CO_3)_3$/TPU composite exhibited the optimal X-ray protection performance when the mass ratio of lanthanide sulfate to TPU reached 4:1. Wu (2007) prepared rare earth/TPU composites using lanthanum oxide (La_2O_3), cerium oxide (CeO_2), praseodymium oxide (Pr_6O_{11}), neodymium oxide (Nd_2O_3), and samarium oxide (Sm_2O_3) as fillers and compared them with conventional PbO/rubber composites. The results show that with the same amount of filler, the radiation shielding performance of the composites prepared above is positively correlated with the atomic number of rare-earth elements, and all are better than the PbO/rubber radiation shielding materials at the same mass fraction. Wang et al. (2020) simulated the shielding performance of five rubber types and four lanthanide compounds using the Monte Carlo method. The results showed that SBR exhibited the highest energy deposition when rays passed through it, and La_2O_3 had the optimal γ-ray shielding effect. ^{137}Cs was chosen as the radioactive source, SBR as the matrix, and La_2O_3 as the filler, and the shielding effect was excellent when the mass ratio between filler and matrix was 7:3. In addition, Wang et al. compared the differences in shielding effectiveness between conventional metallic elements and rare earth elements and found that the shielding effectiveness of Cd, Ba, and La is similar, while Ta and Lu can substitute for each other (Wang, 2021). The results of Dai and Xiao (2016) showed that the mass attenuation coefficient of yttrium oxide (Y_2O_3)/germanium oxide (GeO_2)/epoxy resin-based shielding materials in the low energy range is comparable to that of Pb, so they can substitute for Pb in the low energy range. The performance of the polymeric materials against gamma radiation and X-ray radiation as reported in the literature is shown in Supplementary Appendix Tables 2, 3.

3.3 Neutron shielding materials

PE is a versatile polymer with high hydrogen content. Owing to its non-toxicity, excellent chemical stability, and superior mechanical properties, it is widely used as a material for neutron radiation protection (Guetersloh et al., 2006). The neutron shielding capabilities of polymeric materials as reported in the literature are displayed in Supplementary Appendix Table 4. In 2014, Sukegawa and Anayama (2014) from the US National Aeronautics and Space Administration (NASA) used PE as a matrix, doped and blended B_4C and FeW with PE, and prepared pedal neutron and γ-ray shielding materials. The results showed that these composites exhibit excellent neutron and γ-ray shielding properties. Baykara et al. (2020) added Gd_2O_3 and B to a polyimide (PI) matrix. B has a large cross section for neutron interaction, and Gd, which is a high atomic number metal, contributes to γ-ray shielding. The composites were prepared via injection molding with a thickness of 1 cm and a diameter of 2.5 cm. The density of the composites reached 1.21 g/cm^3 when the composites were doped with 11% h-BN and 1% Gd_2O_3. The composites containing 11% h-BN achieved the best neutron shielding at a thickness of 3 cm, absorbing 80%–99% of neutrons. The γ-ray shielding capacity of the composite material was increased by 50% compared to pure PI. The mass attenuation coefficient of pure PI was 0.067 cm^2/g, whereas that of the composite containing 11% h-BN and 3% Gd_2O_3 was 0.0865 cm^2/g at 4 MeV. However, the tensile strength, elongation at break, and impact strength decrease with increasing filler content in the composites. This is because, at high filler content, the filler in PI undergoes unavoidable agglomeration, leading to defects within the polymer matrix and a subsequent decrease in mechanical properties. In 2020, Sayyed et al. (2020) used UHMWPE and molybdenum dioxide (MoO_3) as fillers to prepare polymer composites for neutron shielding. Since MoO_3 has a high density, UHMWPE particles and MoO_3 nanoparticles were thoroughly mixed via ball milling. The mixture was then hot-pressed at 1,100 °C under a pressure of 20 MPa, and samples with different weight fractions (from 1% to 5%) of MoO_3 were prepared. The density of composites containing 5% MoO_3 was 1.0725 g/cm^3 and the total macroscopic cross-sectional area (TMCS) of composites containing 1%, 2%, 3%, 4%, and 5% MoO_3 was 0.0627, 0.0631, 0.0641, 0.0661 and 0.0674 cm^{-1}, respectively, which was higher than that of paraffin (0.0599 cm^{-1}).

3.4 Composite materials for space radiation protection applications

Space radiation includes SPEs, GCR, and neutrons produced by the interaction of SPEs and GCR with barrier materials (Thibeault et al., 2015). GCR contains high-energy heavy ions

and protons, with energy reaching hundreds of GeV, and has extremely strong penetrating power, which can cause the ionization of atoms in spacecraft materials. SPEs, on the other hand, suddenly generates high-intensity proton streams. Both can damage the structure of devices (such as single-particle effects) and threaten the health of astronauts. More importantly, when energetic particles (e.g., protons) from cosmic rays interact with atomic nuclei in Earth's atmosphere, the resulting neutron streams can damage semiconductor devices in electronic equipment, impairing their proper functioning. Neutron interactions with the nuclei of vehicle materials can alter material properties—such as embrittlement, swelling, or deformation—undermining the structural integrity and service life of the vehicle. For human transport vehicles like the International Space Station, cosmic ray neutrons threaten astronaut health, as neutron radiation can increase their risk of cancer and other radiation-related illnesses. It is therefore important to protect against neutrons generated by high-energy particle streams in cosmic space. Hydrogen has the highest charge-to-mass ratio and can fragment high-charge and high-energy ions, block SPEs protons, and slow down secondary neutrons. Therefore, hydrogen-containing materials are the preferred choice for shielding space radiation. Due to the high hydrogen content of PE and PP, these materials are widely used to shield ionizing radiation in space. Even NASA has reported that UHMWPE is used as a reference material for testing multifunctional composites (especially accelerator-based composites). For space applications, materials must not only be radio-transparent to this radiation but also be high-temperature stable, lightweight, and have high impact and mechanical strength. Winroth et al. (2020) developed two types of composites, one consisting of reinforced UHMWPE fiber and polybenzoxazole, and the other of two layers of carbon fiber cladding with a UHMWPE core layer. The UHMWPE-polybenzoxazole composites showed a 325% increase in specific strength and a 31% reduction in dose equivalent compared to aluminum (Al). The sandwich structure showed a 144% increase in tensile modulus and a 227% increase in flexural modulus compared to Al. For all sandwich composites, the radiation dose increased with increasing cladding volume (decrease in shielding effect), as the cladding volume contained no hydrogen. The equivalent dose of the sandwich composites increased by up to 11.9% compared to UHMWPE-polybenzoxazole composites. In addition, the dose equivalent of all sandwich composites was lower (better shielding performance) than that of Al.

To meet the synergistic requirements of radiation shielding and *in-situ* resource utilization (ISRU) in crewed space missions, Zaccardi et al. (2022) selected PE, a solid radiation shielding material with high hydrogen content, and basalt powder-based Martian regolith simulant (RG) to construct a research system, systematically verifying the space radiation shielding efficiency of PE/RG composites and the process feasibility of fused filament fabrication (FFF) 3D printing. The results showed that under the GCR and SPEs radiation fields on the Martian surface, the composite still exhibited effective shielding performance even under high RG loading. Specimens were prepared via 3D printing and casting processes, even when the RG content reached 60wt%, the impact strength of the molded PE/RG specimens was still superior to that of neat PE, with the optimal impact performance achieved at 10wt% RG loading. RG showed good compatibility with the PE matrix, differential scanning calorimetry (DSC) analysis indicated that RG could act as a heterogeneous nucleating agent, significantly improving the crystallinity of PE/RG composites and thus enhancing their impact properties. Further research explored filament extrusion and FFF processes for PE/RG systems with 5wt% and 10wt% RG loading, and specimens were successfully printed using a HDPE build plate. Tests demonstrated that the tensile strength of 3D-printed specimens was comparable to that of molded specimens for both neat PE and PE/RG composites with 5wt% RG loading, highlighting the effectiveness of the FFF process in preparing such composites and providing key technical support for integrating the material's radiation shielding capability with the utilization of *in-situ* resources like Martian regolith.

4 Effects of nuclear radiation on polymer composites and cost-effective substitution of shielding functional components

4.1 Effects of nuclear radiation on polymer composites

The effects of nuclear radiation on polymer composites are multidimensional and include changes in the physical, chemical, mechanical, thermal, electrical, and optical properties of the materials.

First, with respect to physical properties, nuclear radiation causes an increase in volume and a decrease in density of polymer composites. This is due to nuclear radiation-induced chemical reactions that produce gases trapped in the polymer material, forming bubbles and thereby increasing the polymer volume. At the same time, changes in the molecular structure of the polymer material can also lead to a decrease in the overall density of the composite; in terms of chemical properties, the degradation and cross-linking of polymer chains caused by nuclear radiation has the greatest effect (Bhattacharya, 2000). The degradation process involves radiation-induced breakdown of polymer chains, leading to a reduction in molecular weight and a subsequent decrease in the material's mechanical properties. The opposite is true for cross-linking, i.e., the radiation-induced formation of new chemical bonds between the polymer chains, which leads to an increase in molecular weight, which increases the hardness and strength of the material, but can also reduce its flexibility and processability. Changes in the mechanical properties of polymer matrix composites are an important aspect of exposure to radiation. Radiation degradation significantly reduces the tensile strength, compressive strength, and impact resistance of polymer composites. On the other hand, the crosslinking reactions can lead to a hardening of the material and increase its brittleness, which reduces its resistance to rupture (Nambiar and Yeow, 2012). In addition, the modulus of elasticity of the material can also change due to cross-linking reactions or fracture induced by radiation. In terms of thermal properties, radiation reduces the thermal stability of polymer composites, causing them to begin decomposing at lower temperatures. The thermal conductivity of polymer composites can also change due to changes in the internal structure of the material, which can affect the performance of the material at

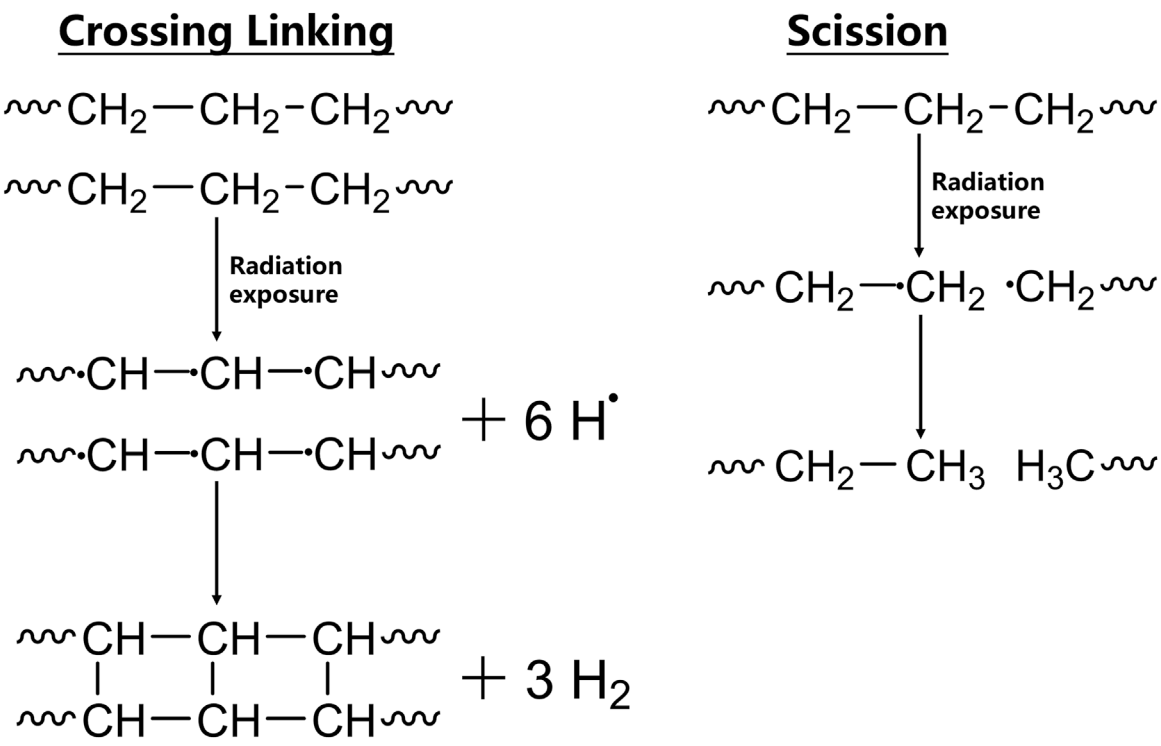

FIGURE 5
Mechanism of polymer chain crosslinking or scission under radiation conditions.

high temperatures; changes in electrical properties are especially important for polymer composites containing conductive fillers. Radiation can alter the material's conductivity and dielectric properties, which can be a serious issue for electronic devices and electrical insulation materials; in terms of optical properties, radiation can lead to a change in the color of the material and a decrease in transparency, which can affect the performance of the material in optical applications such as lenses and window materials; and finally, in the case of biodegradable polymer composites, radiation can affect the rate of biodegradation and thus the degradation behavior of the material.

The mechanism by which polymer chains crosslink or break under irradiation is illustrated in Figure 5. The stability of polymers under irradiation depends on the structure of molecular chain segments—specifically the presence of unsaturated bonds, aromatic rings, and their elemental composition—particularly with regard to neutron shielding (Clayton et al., 2006). The presence of unsaturated bonds, aromatic rings, and conjugated groups in polymer chains facilitates the radiation cross-linking reaction. Due to their low density, polymers cannot shield against the high energies of nuclear radiation. Thus, high-Z metals are introduced and doped into the polymer matrix to form polymer-based nuclear radiation shielding materials for attenuating external radiation. It has been found that, compared with pure polymers, the introduction of high-Z metals improves not only the radiation shielding properties of polymers, but also their mechanical properties and thermal stability (Hu et al., 2008; Iqbal et al., 2011; Park et al., 2001; Bhowmik and Benedictus, 2007).

The improvement in the overall properties of polymer composites is mainly attributed to the size of the filler, the type of interaction between the polymer and filler and its distribution in the polymer matrix (Ajayan et al., 2003; Harrison et al., 2008). The smaller the filler particle size, the lower the stress concentration in the polymer composite when subjected to external stresses and the greater the likelihood of maintaining the polymer's plasticity (Ajayan et al., 2003). Mahdi et al. (Sabri et al., 2019) compared polymer composites doped with micrometric and nanometric fillers to demonstrate the effect of filler size. Polymer composites doped with nanometric fillers showed better linear attenuation coefficients, mechanical strength, and formability compared to those doped with micrometric fillers. Soltani et al. (2016) investigated the effect of the size and percentage of B_4C added to HDPE for neutron shielding. By comparing experimental and simulation results, they concluded that reducing the particle size of the shielding filler significantly increases the probability of thermal neutron collisions with the shielding material, implying enhanced shielding capacity. Polymer clay nanocomposites also proved to be an economical and effective material for shielding against high-energy radiation. Numerous studies have shown that the addition of nano-clay materials (mainly montmorillonite) to a saturated polyester matrix can effectively improve the thermal stability and mechanical properties of the polyurethane matrix (Baran Inceoglu and Yilmazer, 2003; Bharadwaj et al., 2002; Chieruzzi et al., 2013; Dhakal et al., 2006; Jo et al., 2008; Kornmann et al., 1998; Şen et al., 2013; Tsai et al., 2010). The addition of nano-clay materials has also improved the wear resistance and flame retardancy of composites (Jawahar et al., 2006; Nazaré et al., 2006). Nanomaterials can act

as an effective absorber of defects produced during irradiation (Samaras et al., 2003; Chimi et al., 2001; 2006) and efficiently shield photon or particle radiation due to their high surface-to-volume ratio and large surface area. Therefore, depending on the type of filler and the application, different types of efficient and durable polymer composites can be designed, including (i) high-Z metal-based polymer composites and (ii) nanometal-based polymer composites. Numerous studies have shown that polymer composites doped with high-Z metals/metal compounds are effective in protecting against X-rays, γ-rays, neutrons, and the secondary radiation they produce.

4.2 Cost-effective alternatives for radiation-shielding particles

Traditional Pb is biotoxic and environmentally hazardous, and has gradually been replaced by high-Z elements such as W, Bi, Ta, and rare earths. However, W, Bi, and Ta are expensive and their use as Pb substitutes is not cost-effective. Researchers are therefore seeking more environmentally friendly, cheaper, and more nuclear radiation-resistant metals as alternatives. Iron (Fe) is one of the most abundant elements in the Earth's crust, is easily available and is environmentally friendly. Hematite has a high density and is already widely used in medicine, industry, and applied sciences. On this basis, several research teams have started investigating Fe as a filler material for radiation protection in polymers. In 2017, Toyen et al. (2018) prepared flexible lead-free radiation shielding materials with natural rubber as the matrix material and nanoparticles of iron oxide (II, III), tungsten oxide (III), and bismuth oxide (III) as reinforcing materials. Natural rubber (NR) provides flexibility, abrasion resistance and toughness to the composite, while the high-Z metal nanoparticles contribute to the shielding effect. The γ-ray attenuation performance of the composites prepared using Bi_2O_3 as the shielding component is better than that of the composites prepared using WO_3 and ferroferric oxide (Fe_3O_4) as the shielding component. The Bi atoms in Bi_2O_3 have higher atomic numbers than W and Fe, and thus interact more strongly with γ-rays, and their γ-ray attenuation efficiency is higher than that of the composites containing WO_3 or Fe_3O_4. For NR/Bi_2O_3 composites, the mass attenuation coefficients are 3.6 ± 0.3 and 7.0 ± 0.4 at γ-ray energies of 1.25 MeV and 662 keV, respectively. For NR/Fe_3O_4 composites, the mass attenuation coefficients are 1.8 ± 0.2 and 3.3 ± 0.4 at γ-ray energies of 1.25 MeV and 662 keV, respectively. The performance difference between NR/Fe_3O_4 and NR/Bi_2O_3 composites is essentially a multi-dimensional trade-off effect jointly regulated by the intrinsic properties (atomic number, density, surface energy) of high-Z shielding fillers and the filler-matrix interface interaction: Bi_2O_3 has a much larger atomic number of Bi (Z = 83) than Fe (Z = 26), so its interaction cross-section with γ-rays is larger. The mass attenuation coefficients of NR/Bi_2O_3 composites at γ-ray energies of 1.25 MeV and 662 keV are approximately twice those of NR/Fe_3O_4 composites, leading to significantly better γ-ray shielding efficiency. However, the raw material cost of Bi_2O_3 (about 57 USD/kg) is 2.5 times that of Fe_3O_4 (about 23 USD/kg), and the γ-ray attenuation efficiency per unit cost (123 cm^2·USD)) is lower than that of Fe_3O_4 (143 cm^2·USD)), resulting in insufficient economic viability for large-scale applications. From the perspective of mechanical properties, the density of Bi_2O_3 (8.9 g/cm^3) is 1.71 times that of Fe_3O_4 (5.2 g/cm^3). High-density Bi_2O_3 particles tend to form local stress concentration regions in the NR matrix; when the composite is subjected to bending or tensile loads, these stress concentration points significantly restrict the free sliding ability of NR polymer chain segments. Since the flexibility of polymer materials essentially depends on the mobility of chain segments, the introduction of Bi_2O_3 greatly reduces the flexibility of the composite. In contrast, the lower density of Fe_3O_4 can effectively reduce the stress concentration effect; combined with the good interface bonding brought by its uniform dispersion in the NR matrix, it not only retains the mobility of polymer chain segments in the NR matrix but also improves the mechanical properties of the composite through "particle-matrix" cooperative stress-bearing. Eventually, the flexibility, tensile strength, and impact strength of NR/Fe_3O_4 composites are all superior to those of NR/Bi_2O_3 composites. Although NR/Fe_3O_4 composites need to sacrifice approximately 50% of γ-ray shielding efficiency, they are more suitable for dynamic shielding scenarios that require frequent bending (such as protective gloves and protective clothing) due to their better flexibility and mechanical reliability. Therefore, the selection of the two types of composites should be comprehensively determined based on the priority of requirements for shielding efficiency, flexibility, cost, and mechanical properties in specific application scenarios. Turhan et al. (2020) incorporated hematite as a filler into an unsaturated polyester resin in order to study its γ-ray attenuation properties, and samples containing 100% hematite reached a maximum density of 1.55 g/cm^3. The mass attenuation coefficients of samples containing 25% and 100% hematite were 0.3256 ± 0.0067 and 0.5521 ± 0.0116 cm^2/g, respectively, at 59.5 keV. At 1,408 keV, the mass attenuation coefficient of the 100% hematite sample was 0.0556 ± 0.0011 cm^2/g, indicating that the mass attenuation coefficient decreases significantly with increasing energy. Thus, polymer composites based on iron compounds can also be effectively used for nuclear radiation shielding because iron is cheap, environmentally friendly, and one of the most abundant metals in the Earth's crust.

5 Outlook and conclusion

5.1 Outlook

A great deal of research has been conducted in the field of nuclear radiation protection to date, but several pressing issues remain unaddressed. These include the need for shielding materials to have broad-spectrum shielding capacity within a certain energy range, ensuring uniform distribution of shielding fillers in the polymer matrix, and improving the thermal stability of polymers at high temperatures and their chemical stability under extreme conditions. Currently, most studies are carried out by dispersing nano- or micro-sized fillers in the polymer matrix as a way of protecting polymer composites from nuclear radiation, but these micro-nano scale fillers often experience particle detachment or agglomeration over time or under high doping ratios. For example, a study by Livingstone and Varghese (2018) revealed the inhomogeneity of X-ray attenuation by commercial lead-equivalent aprons due to defects resulting from the uneven distribution

of high-Z fillers in the matrix, which could inadvertently harm the wearer.

Therefore, rather than simply dispersing nano- or micro-particles in polymers, it is more effective to form strong chemical bonds between polymer chains and high-Z micro- and nano-sized filler particles. This approach can solve the problems of uniform distribution and chemical stability of micro- and nano-sized filler particles through the effective cooperation of chemical bonds. Even if the effect of this method is unclear, it can at least point us toward further research into developing effective radiation shielding materials with better chemical stability, thermal stability, and uniform distribution properties.

Other issues that currently need to be addressed in the field of radiation protection are the size and weight of materials and toxicity, especially of shielding materials. Future research should focus on developing lighter, thinner, and less toxic materials for nuclear radiation protection. A new approach to solving these problems could be using recycled plastics or industrial polymer waste to produce polymer composites. This would not only help reduce costs, but also address environmental problems such as water, soil, and air pollution. Compared to producing new resins or polymers, recycling achieves significant energy savings (More et al., 2021). The development of such recycled polymer-based protective materials is highly aligned with the concept of circular economy and consistent with the low-carbon development logic of biopolymers. For instance, biobased materials like polylactic acid (PLA) discussed by Nanda et al. (2022) can significantly reduce carbon footprints; moreover, resource circulation and the sustainable characteristics of "renewable raw materials and low-carbon footprints" can be achieved through recycling. In radiation protection applications, recycled polymers including recycled high-density polyethylene (r-HDPE) and recycled polyethylene terephthalate (r-PET) can serve as matrix materials, and their protective functions can be realized via composite modification. Their raw materials can be derived from industrial or domestic plastic waste, and the unification of environmental value and functional value is achieved through the recycling and utilization of solid waste (Yang et al., 2021). Additionally, some countries have guided green development through environmental regulations to promote the replacement of traditional highly toxic shielding materials with low-toxicity and low-carbon recycled polymer protective materials, thereby amplifying the synergistic value of "solid waste consumption-energy conservation-economic growth" (Murshed et al., 2021). Thus, recycling is not only environmentally friendly but also economically viable. In addition, flexible polymer-based materials for protection against nuclear radiation should be developed in the direction of "Multifunctionality" and "Lightness with a broad spectrum". "Multifunctionality" means that the material simultaneously possesses nuclear radiation protection performance and one or more additional properties, such as sealing, UV protection, antibacterial activity, easy cleanability, and wear resistance; "Lightness with a broad spectrum" means that the material exhibits excellent nuclear radiation protection performance within a certain energy range and can simultaneously satisfy the comprehensive shielding function for X/γ rays of various energy bands and neutrons. In addition, the use of new materials such as nanomaterials, rare earth materials, graphene, and carbon nanotubes in polymer-based nuclear radiation shielding materials should also be focused on.

5.2 Conclusion

With the increasing use of harmful yet useful nuclear radiation in various industries, there is a growing demand for lightweight, high-quality, and flexible lead-free radiation shielding materials. A wide range of lead-free materials are currently being used, including radiation shielding glass, special concrete, and heavy metal composites. The choice of materials for radiation protection depends on the source, type and energy of the radiation, as well as the mechanism by which the material reacts to the radiation. The flexibility, light weight, non-toxicity, and corrosion resistance of polymer composites make them suitable for the manufacture of aprons, gloves and other wearable protective equipment. Due to the ease with which polymers can be processed, they can be used to coat and bond various high-Z metal compounds, which can be achieved via physical doping or chemical grafting. For high-energy photons (e.g., X/γ-rays), composites based on high-Z metals such as Bi, W, and Ce can provide effective shielding and good mechanical properties. However, researchers are working to develop cheaper materials and iron-based composites are an ideal alternative because they are economical; for neutron shielding, elements with high capture cross-sections, such as B, Li, and Gd, are chosen as shielding components. Since polymers contain large amounts of hydrogen (which has a neutron capture cross-section of up to 0.3 barns), they function effectively as neutron shielding materials. Both low and high Z-value metals are used in neutron shielding because neutron radiation also produces secondary γ-rays. Polymers with low Z-values are responsible for neutron attenuation, while metals with high Z-values are used as shielding against the secondary γ-rays produced by the interaction. In addition to the type of heavy metal, the size of the filler metal is also an important factor that determines the shielding efficiency. It has been shown that incorporating nanoscale heavy metal particles into the polymer matrix can provide a more uniform distribution and lower stress concentration, thus achieving better attenuation performance and mechanical properties compared to micrometer-sized heavy metal shielding particles.

This article focuses mainly on the research progress of polymer-based composites for nuclear radiation protection, detailing the technological advances and innovations in this field in recent years. It systematically presents the background to the use of polymer-based composite materials in nuclear radiation protection and highlights their important role in replacing traditional heavy metal-based shielding materials. Through in-depth discussions on the preparation technologies of polymer matrix composites, different types of fillers, radiation protection mechanisms, and material property optimization, the article showcases the recent advancements of polymer-based nuclear radiation shielding composites in both scientific research and practical applications. At the same time, the article also analyzes the problems and shortcomings of current research, such as the uniformity of filler distribution, thermal stability, chemical stability, etc., and provides

an outlook on the direction of future development, aiming to offer useful references for the research and application of nuclear radiation shielding materials. With the continuous advances in science and technology, it is believed that polymer-based nuclear radiation protection composites could play a more important role in the field of nuclear and radiological engineering.

Author contributions

X-PL: Conceptualization, Investigation, Supervision, Writing – original draft, Writing – review and editing. H-SY: Investigation, Methodology, Supervision, Writing – original draft. YZ: Supervision, Writing – original draft. BY: Writing – original draft, Writing – review and editing. JZ: Supervision, Writing – original draft, Writing – review and editing. LL: Supervision, Writing – original draft. HL: Supervision, Writing – review and editing. XL: Supervision, Writing – review and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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