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Study on the shielding performance of bismuth oxide as a spent fuel dry storage container based on Monte Carlo simulation

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ABSTRACT

For traditional spent fuel shielding materials, due to physical and chemical defects and cost constraints, they have been unable to meet the needs. Therefore, this paper carries out the first discussion on the application and performance of bismuth in neutron shielding by establishing Monte Carlo simulation on the neutron flux model of shielded spent fuel. Firstly, functional fillers such as bismuth oxide, lead oxide, boron oxide, gadolinium oxide and tungsten oxide are added to the matrices to compare the shielding rates of aluminum alloy matrix and silicone rubber matrix. The shielding rate of silicone rubber mixture is higher than aluminum alloy mixture, reaching more than 56%. The optimal addition proportion of bismuth oxide and lead oxide is 30%, and the neutron radiation protection efficiency reaches 60%. Then, the mass attenuation coefficients of bismuth oxide, lead oxide, boron oxide, gadolinium oxide and tungsten oxide in silicone rubber matrix are simulated with the change of functional fillers proportion and neutron energy. This simulation result shows that the mixture with functional fillers has good shielding performance for low energy neutrons, but poor shielding effect for high energy neutrons. Finally, in order to further evaluate the possibility of replacing lead oxide with bismuth oxide as shielding material, the half-value layers and various properties of bismuth oxide and lead oxide are compared. The results show that the shielding properties of bismuth oxide and lead oxide are basically the same, and the mechanical properties, heat resistance, radiation resistance and environmental protection of bismuth oxide are better than that of lead oxide. Therefore, in the case of neutron source strengths in the range of 0.01–6 MeV and secondary gamma rays produced below 2.5 MeV, bismuth can replace lead in neutron shielding applications.

1. Introduction

The intermediate storage of spent fuel serves to mitigate the problems caused by radioactivity and provides ample time for the development of reprocessing technology and policy formulation [[1,2]]. Recently, spent fuel dry storage has been in increasing demand by other nuclear states due to its advantages such as easy installation, scalability, transportability, economy, safety and public acceptance [3]. Spent fuel contains a lot of high-energy and high-hot neutrons, so shielding spent fuel is a very important work. There are two main types of neutrons shielding materials for spent fuel: one is neutron absorbing materials, such as boron and cadmium, and the other is neutron scattering materials, such as lead and concrete.

With the rapid development of the nuclear energy industry, traditional neutron shielding materials are no longer sufficient. To ensure rapid development, it is crucial to master advanced neutron shielding technology [[4,5]]. Traditional neutron shielding materials include boron-containing concrete, lead-boron polyethylene, tungsten boron polyethylene, boron-containing stainless steel, B_4C/Al composites, epoxy resin and so on. However, these traditional materials have certain limitations in terms of shielding performance, heat resistance, radiation resistance and mechanical properties [[6–9]]. Wang. P [10] and Okuno.

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K [11] proved that lead-boron polyethylene is light in weight, small in size and has good neutron and gamma shielding effects, but it is not suitable for engineering materials due to its low melting point, poor radiation resistance and poor mechanical properties. Divya, M. et al. proposed that due to the low solubility of boron in stainless steel, excessive boron addition would lead to the precipitation of boride (Fe, Cr)$_2$B, and the ductility and toughness of boron steel would be greatly reduced, making the preparation of boron steel with high boron content very challenging [12]. Park B et al. [13] and Kumar N et al. [14] researched that B_4C/Al alloy has good thermal neutron absorption properties, and its ductility and wear resistance can be improved by adding appropriate enhancers. Although fine uniform distribution of reinforcer is a necessary condition for optimal enhancement, it is difficult to achieve uniform dispersion [[15,16]].

Therefore, it is urgent to develop new neutron shielding materials that meet a series of engineering requirements such as high temperature resistance, radiation resistance and good mechanical properties. Abolfazl Zare Mehrjardi et al. proved that Bi_2O_3 can effectively attenuate X-rays, and with the increase of Bi_2O_3 content, X-ray attenuation also increases [17]. Mohamed Elsaf et al. found that the addition of bismuth oxide improves the linear attenuation coefficient (LAC) parameter. At 0.662 MeV, the performance of LAC is improved by 6 times when the content of bismuth oxide is increased by 30% [18]. El-Sharkawy R M et al. demonstrated that *R*–PVC nanocomposite samples containing 35.0 wt% Bi_2O_3 NPs showed the highest photon mass attenuation coefficient at 121 keV. The addition of Bi_2O_3 NPs to *R*-PVC facilitates the production of lead-free and sustainable polymer nanocomposites with improved gamma rays shielding properties [19]. Gülmen M et al. have found that Bismuth-based glass has good radiation protection effect in Bi_2O_3–B_2O_3–BaO glass system, and can be used as a substitute for traditional materials [20]. Bismuth oxide is the preferred doping material that can be used to study gamma shielding, with a high density (8.9 g/cm^3), non-toxic, and its gamma shielding ability is almost equal to that of lead [21]. A Ratep et al. showed that adding Bi_2O_3 to glass can increase the radiation absorption capacity, and the linear attenuation coefficient of glass has been increased by 26% at 0.015 MeV gamma ray energy [22].

However, there is no literature report on the structure and properties of neutron shielding materials containing bismuth oxide in the dry storage environment of spent fuel. Therefore, in this paper, bismuth oxide is used as the research object of neutron shielding filler, and it is compared with four commonly used neutron shielding fillers: lead oxide, tungsten oxide, gadolinium oxide and boron oxide. The aim is to study the shielding properties of bismuth oxide materials in dry storage of spent fuel and the potential application of bismuth oxide instead of lead oxide as neutron shielding filler.

2. Principle and method

2.1. Model construction

Monte Carlo method can not only provide a theoretical basis for sample design and preparation, but also save cost and time. Therefore, this paper uses Monte Carlo method to establish a model for shielding neutron flux of spent fuel. The simulation results are studied by changing the different nuclides (atomic number and relative atomic mass) and the mass ratio of nuclides. It mainly includes the setting of the source term of the spent fuel assembly and the geometric position relationship between the source term and the shielding material.

Setting of spent fuel assembly source terms: In this paper, the typical PWR nuclear power plant deep burnup spent fuel assembly is taken as an example, and neutron spectra in different energy regions is shown in Fig. 1 [23]. The neutron source strength is significantly higher in the 0.01–6 MeV energy range than in other energy regions. Therefore, the neutron source is set to a surface source of 0.01–6 MeV. The shielding materials are placed 1 cm above the surface source.

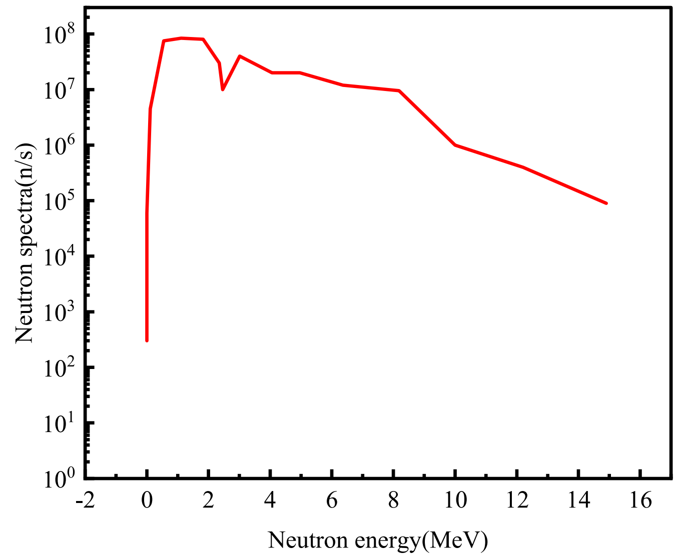

Fig. 1. Neutron spectra in different energy regions of deep burnup spent fuel assemblies in typical PWR nuclear power plants.

2.2. Shield thickness

The special structure of the spent fuel dry storage container reduces the neutron radiation dose from spent fuel decay and allows the container to remain stable for a longer period of time. Therefore, the shield thickness is crucial to the shielding effect and stability of the container in the neutron radiation field. In actual spent fuel storage containers, the protective layer of the shielding material is typically between 8 cm and 30 cm thick. When the thickness of the shield body increases, the shielding rate also increases. However, when the thickness increases to a certain extent, the growth rate of neutron shielding rate gradually slows down, and finally maintains a certain level [[24–26]]. This is because the energy of the neutron has gradually decreased, and the neutron interaction cross section becomes smaller. The shielding rate does not increase without limit as the thickness of the shield increases. According to Zhu Li's research [27], a 10 cm thick tungsten-nickel alloy can reduce fast neutron energy to 2 MeV, resulting in a 91.35% shielding rate for fast neutrons. Han Ruiyi's research shows that when the thickness of fast neutron shield body reaches 10 cm, the shielding rate of tungsten shield reaches 0.9, which is the best shielding effect [28]. Considering the weight and economy of the shield body itself in practical applications, the shield thickness has been uniformly set at 10 cm in this paper.

2.3. Evaluating shielding performance parameters

In spent fuel shielding, according to the situation of radiation source passing through the material, radiation shielding materials with special structure and specific shielding properties are used to protect human body and equipment [[29,30]]. When it comes to neutron shielding, some neutrons are absorbed by the material, while others are transmitted. The proportion of transmitted neutrons in relation to the total number of neutrons is known as the neutron transmittance of the material [31]. The neutron transmittance ($T_{(E)}$) is defined as:

$$T(E) = N(E_t) / N(E_0) \tag{1}$$

where $N(E_t)$ and $N(E_0)$ are the number of neutrons that penetrate the sample and the number of neutrons in the source neutron. The neutron transmittance can reflect the shielding performance of a shielding material with a certain thickness to a certain neutron field. This provides a reference for screening neutron shielding materials or selecting engineering materials.

The mass attenuation coefficients of various shielding materials are calculated by simulating the transmission of neutrons at different energies. Formula (2) describes the attenuation mass coefficient when neutrons pass through a medium of a certain thickness [32]:

$$N = N_0 \cdot e^{-(\mu/\rho)\rho \cdot x} \quad (2)$$

where N and N_0 are the dose after passing through the shielding material and the dose before passing through the shielding material; μ is the linear attenuation coefficient of the medium; ρ and x are the density and thickness of the shielding material, respectively. For a compound or mixture, the mass attenuation coefficient follows the Bragg additivity rule.

The radiation protection efficiency can evaluate the shielding performance of the shielding material against neutron radiation, which can be expressed in terms of shielding properties:

$$RPE = 1 - \frac{N}{N_0} \cdot 100\% \quad (3)$$

where N and N_0 are the dose after passing through the shielding material and the dose before passing through the shielding material. The better shielding efficiency of the material indicates greater shielding of the incident particles and therefore better radiation protection. The larger the RPE value, the better the protective effect of the material, and vice versa.

The absorption capacity of a substance for gamma rays can be expressed as the 'half-value layer'. The relationship between $d_{1/2}$ and μ is [33]:

$$d_{1/2} = \frac{\ln 2}{\mu} = \frac{0.693}{\mu} \quad (4)$$

3. Results and discussion

3.1. Comparison of shielding performance between aluminum alloy and silicone rubber matrix

Among neutron shielding materials, silicone rubber and aluminum alloy, as commonly used polymeric materials in the field of radiation protection, have the advantages of good shielding, chemical stability. In addition, silicone rubber has light weight and small size; aluminium alloy has good corrosion resistance, heat resistance and low density. It is stronger and easier to meet the mechanical properties of the material.

Aluminum alloy and silicone rubber are selected as the matrix, and boron oxide, lead oxide, gadolinium oxide, bismuth oxide and tungsten oxide are doped into the matrix materials, which can not only reduce the weight of the shield, but also improve the shielding performance. The neutron shielding properties of various oxide shielding fillers in aluminum alloy and silicone rubber are compared.

Fig. 2 shows the neutron radiation protection efficiency (RPE) of five oxide functional fillers with different proportions in aluminum alloy and silicone rubber. The RPE when the proportion is 0 corresponds to the shielding rate of the pure matrix material. The results show that the neutron radiation protection efficiency of silicone rubber composite is higher than that of aluminum alloy composite. Furthermore, the radiation protection efficiency of the silicone rubber polymer remains above 56% regardless of the change in shielding filler proportion. Compared with the aluminum alloy matrix, the silicone rubber matrix contains hydrogen element, which makes it easier to moderated neutrons, effectively attenuate high-energy neutrons, and weaken gamma radiation. Since there are no neutrons in the hydrogen nucleus, hydrogen also offers the additional benefit of no secondary neutron radiation. Therefore, the neutron radiation protection efficiency of silicone rubber matrix is better than that of aluminum alloy.

In the aluminum alloy matrix, neutron shielding properties of five kinds of oxide functional fillers increase with the increase of filler

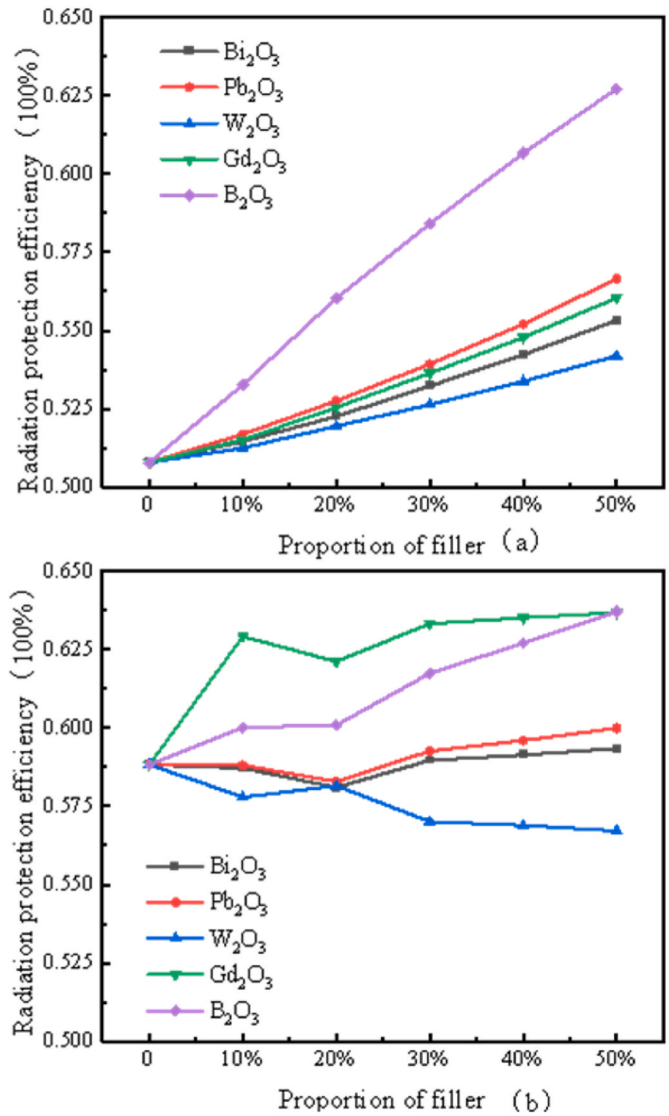

Fig. 2. Neutron radiation protection efficiency of functional fillers with different proportions in the matrix (a) Radiation protection efficiency of functional fillers with different proportions/aluminum alloy (b) Radiation protection efficiency of functional fillers with different proportions/silicone rubber.

proportion. Boron oxide has a significantly higher radiation protection efficiency than other oxides due to its high thermal neutron absorption cross-section. With the increase of boron in the matrix, the radiation protection efficiency is gradually increased, and the shielding performance is obviously improved. The differences of radiation protection efficiency among lead oxide, gadolinium oxide, bismuth oxide and tungsten oxide are not obvious, and their shielding efficiency increases with the increase of filler proportion. When the filler proportion reaches 30%, the radiation protection efficiency can reach more than 52.5%.

In the silicone rubber matrix, the radiation protection efficiency of gadolinium oxide is higher than that of boron oxide. This is due to the limited ability of silicone rubber itself to shield fast neutrons, and the shield performance mainly relies on the addition of oxide functional fillers to shield fast neutrons. For fast neutrons, the inelastic scattering of neutrons, which is mainly achieved by heavy elements, replaces elastic scattering. Gd has a higher atomic number than B, so the neutron moderating effect of Gd is better when the neutron energy is increased. Although boron has a high absorption cross section for thermal neutrons, it has a relatively small absorption cross section in the fast neutron region. Before the proportion of bismuth oxide, lead oxide and tungsten oxide does not reach 20%, the neutron shielding performance of the composite decreases slightly with the increase of the proportion. The slope of the shielding rate curve increases rapidly when the amount of filler reaches 20%. This indicates that when the filler content is low, there are many blank micro-regions without effective shielding elements in the composite material, and these blank micro-regions will form a "channel" of neutrons transmission, resulting in a low overall shielding properties of the composite material [34]. When the amount of filler increases to a certain extent, the filler network structure can be formed in the polymer matrix, resulting in a significant reduction of the blank micro-region that can be transmitted by neutrons, so that the shielding properties of the composite materials are rapidly increased. Due to the large atomic numbers of lead, bismuth and tungsten, they mainly have inelastic collisions with neutrons, and for neutrons with lower energy, they cannot reach the threshold of inelastic collisions and cannot participate in the reaction. With the increase of lead, bismuth and tungsten filler elements, the hydrogen element in the composite decreases, and the cross section of neutron elastic collision decreases. Therefore, with the increase of the proporition of lead oxide and bismuth oxide, the neutron shielding performance of silicone rubber mixture can be slightly improved. When the proportions of bismuth oxide are 30% and 50%, radiation protection efficiency is 60.4% and 60.6%, which is only increased by 0.2%; when the proportions of lead oxide are 30% and 50%, its radiation protection efficiency is 60.7% and 61.3%, which is only increased by 0.2%. Therefore, when the proportion of bismuth oxide and lead oxide is 30%, the shielding effect is better and the economic benefit is optimal. With the increase of the proporition of tungsten oxide, the neutron radiation protection efficiency of the silicone rubber mixture not only does not increase, but also decreases.

3.2. Mass attenuation coefficient of different materials

In order to compare the effect of different filler ratios on the shielding effect, the neutron transmission calculation model is established by adding 10%, 30%, 50% bismuth oxide, lead oxide, tungsten oxide, gadolinium oxide and lead boron oxide on silicone rubber matrix, respectively. The mass attenuation coefficients of different shielding materials at 0.1 MeV, 0.5 MeV, 1.0 MeV, 2.0 MeV, 3.0 MeV, 4.0 MeV, 5.0 MeV and 6.0 MeV neutron energies are calculated.

Fig. 3 shows the curve of the mass attenuation coefficient of various shielding materials with the neutron energy. As can be seen from Fig. 3, the mass attenuation coefficient increases with the increase of functional fillers content, mainly due to the large atomic number of the functional fillers, and the addition of them will increase the neutron absorption cross section of the material. Moreover, adding functional fillers increases the scattering cross section along the neutron propagation path [[35,36]]. In the low energy region, the mass attenuation coefficient changes obviously with the increase of the proportion. When the neutron energy is greater than 3 MeV, the change of the proportion of functional fillers has little effect on the mass attenuation coefficient. It is worth noting that the proportion of functional fillers added has a greater effect on low-energy neutrons.

With the increase of neutron energy, the mass attenuation coefficient of each shielding material tends to decrease, and the shielding performance of low-energy neutrons is better, while the shielding effect of high-energy neutrons is lower. When the neutron energy is less than 2 MeV, the mass attenuation coefficient of the composite containing tungsten oxide is the lowest. This is because gadolinium and boron have high thermal neutron absorption cross sections, and the addition of gadolinium oxide and boron oxide makes neutrons interact more with the shielding material to achieve a good shielding effect. The relative atomic number of lead and bismuth is larger than that of tungsten, and the scattering cross section of lead oxide complex and bismuth oxide complex is larger. When the energy is greater than 2 MeV, the mass attenuation coefficients of bismuth oxide and lead oxide are the lowest. With the increase of neutron energy, the inelastic scattering ratio between neutrons and matter increases, but the elastic scattering ratio decreases. This results in a lower effective absorption cross section for neutrons in the material. Therefore, high-energy neutrons have a greater ability to penetrate the material and are less likely to be absorbed, which results in a lower neutron mass attenuation coefficient.

3.3. Effect of shielding materials on the secondary gamma ray spectrum

High energy neutrons are slowed down and captured with the shielding material, and at the same time the energy is reduced, secondary gamma rays are also released [37]. Although the shielding material often contains high-Z materials which are effective in absorbing

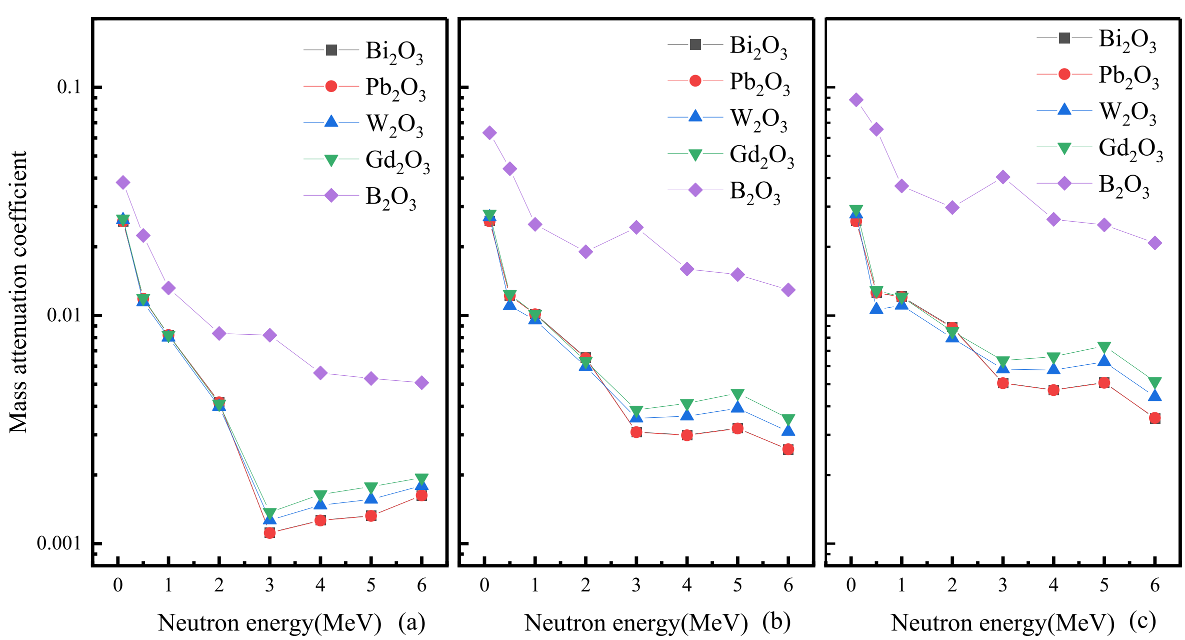

Fig. 3. The mass attenuation coefficient of different functional fillers in the silicone rubber matrix (a)add 10% functional fillers (b)add 30% functional fillers (c)add 50% functional fillers.

gamma rays, such as tungsten, lead, etc., some gamma rays are still able to penetrate the shielding [38]. This poses a threat to both equipment and personnel. Therefore, it is important to consider the impact of secondary gamma rays during the simulation process. To determine the effect of shielding materials on secondary gamma rays, a calculation model for gamma ray transmission of silicone rubber polymer with 30% bismuth oxide, lead oxide, tungsten oxide, gadolinium oxide and boron oxide is established. The number of gamma rays shot of different shielding materials at 0.01 MeV, 0.1 MeV, 0.5 MeV, 1 MeV, 1.5 MeV, 2 MeV, 2.5 MeV, 3 MeV, 3.5 MeV, 4 MeV, 4.5 MeV, 5 MeV, 5.5 MeV and 6.0 MeV γ energies are calculated. The secondary gamma ray energy spectrum is shown in Fig. 4.

Fig. 4 shows that, when the gamma-ray energy ranges from 0.01 MeV to 1.5 MeV, the gamma-ray transmission amount of gadolinium oxide is low. When the gamma-ray energy is 1.5 MeV–2.5 MeV, the amount of gamma-ray transmission by gadolinium oxide increases sharply. When the gamma-ray energy is 2.5 Mev–6 MeV, the amount of gamma-ray transmission by gadolinium oxide remains relatively stable. Gadolinium oxide has a good shielding effect on gamma rays below 1.5 MeV, and a poor shielding effect on gamma rays above 1.5 MeV. In the 0–2.5 MeV range, bismuth oxide and lead oxide transmit less gamma rays than boron oxide and tungsten oxide. This is due to the higher atomic numbers of lead and bismuth, which make them more likely to undergo photoelectric and Compton scattering effects with gamma photons, resulting in energy attenuation and effective shielding. Therefore, for photon energies below 2.5 MeV, bismuth and lead composites provide better shielding than boron and tungsten composites. When the photon energy exceeds 2.5 MeV, the three effects jointly affect the shielding of the photon, and the composite exhibits a similar amount of transmitted gamma rays. In the measured energy region, the secondary gamma ray spectra of bismuth oxide and lead oxide are almost identical, and they have similar abilities to shield secondary gamma rays. The addition of bismuth oxide and lead oxide shielding has a more obvious attenuation effect on low energy gamma rays.

3.4. Comparison of shielding properties between bismuth mixture and lead mixture

In order to objectively analyze the shielding effect of the composite from the required thickness of the shielding material, the half-value layers of bismuth oxide composite, lead oxide composite and pure aluminum alloy at 0.1 MeV, 0.5 MeV, 1 MeV, 3 MeV, 5 MeV radioactive energy are compared [39].

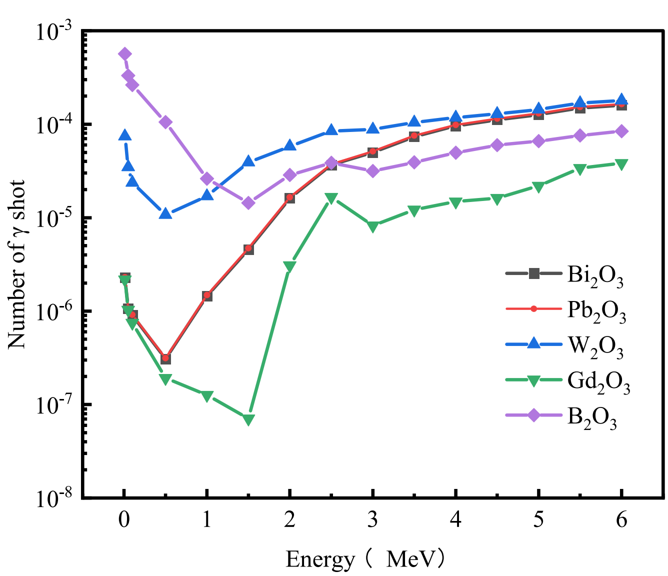

Fig. 4. Secondary gamma ray spectrum of silicone rubber matrix with 30% oxide added.

As shown in Fig. 5, the half-value layers of the composites decrease with the increase of the proportion of bismuth oxide and lead oxide. With the increase of photon energy, the half-value layer of bismuth oxide composite and lead oxide composite increases. It shows that the shielding performance of the composites for low energy photons is good, but the shielding performance for high energy photons is poor. When the photon energy is less than 1 MeV, the photoelectric effect and Compton effect mainly occur. The atomic number of the material determines the shielding effect of low energy photons. Since the atomic number of lead is 82 and the atomic number of bismuth is 83, the linear attenuation coefficient of the bismuth composite is almost equal to that of the lead bismuth composite. When the photon energy is between 1MeV–3MeV, three effects affect the shielding of the photon together. When the photon energy is greater than 3 MeV, the cross section of the interaction decreases, only the electron pair effect occurs with the target atom, and the two 0.511 MeV photons produced are shielded by the photoelectric effect and the Compton effect. Under the same irradiation conditions, the same proportion of bismuth oxide and lead oxide have similar half-value layers. That is, the bismuth-containing composite and the lead-containing composite have the same gamma-ray shielding effect.

There is little difference in the ability of bismuth-containing and lead-containing materials to shield neutrons and secondary gamma rays produced by neutrons. In addition, the mechanical properties, heat and radiation resistance of lead oxide and bismuth oxide are compared. Ekwipoo Kalkornsuranee [40] and Donruedee Toyen [41] showed that the modulus and hardness of bismuth oxide composites were higher than those of lead oxide composites when bismuth oxide and lead oxide were added to the matrix. When appropriate Bi_2O_3 is added to the composite, it can act as a co-activator, increasing the cross-linking density, resulting in an increase in tensile strength and elongation at break [42,43]. And bismuth oxide is generally harder than lead oxide, which makes it exhibit better wear resistance in some applications [44]. Both lead oxide and bismuth oxide are heat-resistant materials, which can resist chemical reactions with other substances at high temperatures and maintain the stability of their structures and properties. Lead oxide has a melting point of about 880 °C and bismuth oxide has a melting point of about 271 °C. In the application of dry storage of spent fuel, the surface temperature of the storage cylinder is 95.5 °C [45], and the melting point of lead oxide and bismuth oxide is higher than the surface temperature of the storage cylinder. So both lead oxide and bismuth oxide can be used for shielding of spent fuel. In the radiation environment, especially in the high dose of radiation environment, the radiation resistance of lead oxide is not very good, and its performance may be affected. Due to its crystal structure and electronic structure, bismuth oxide can effectively resist ionization damage caused by radiation [46]. Especially in high dose rate and rapid radiation environment, bismuth oxide shows excellent radiation resistance [[47,48]]. In general, bismuth oxide has better radiation resistance than lead oxide, which makes it more favorable for applications in high radiation environments.

What's more, lead is a toxic heavy metal, which also has problems such as heavy weight, toxicity to human body and environment in the process of use. Bismuth is a recently discovered 'green metal element' with low toxicity compared to other heavy metals. Many bismuth compounds are even less toxic than our daily consumption of salt [[49–51]]. In addition, pure bismuth is stable at room temperature, making mining and smelting relatively easy. Bismuth has global reserves slightly lower than silver, with China holding the largest reserves of bismuth resources worldwide. As a result, the price of bismuth is not high [52].

In summary, the advantages of using bismuth-containing mixtures instead of lead-containing mixtures are mainly reflected in the following three aspects: (i) Bismuth-containing mixture and lead-containing mixture have similar densities, which can be replaced in equal proportion in the material, reducing the toxicity and pollution of the material while improving its practicality. (ii) Bismuth-containing mixtures have a

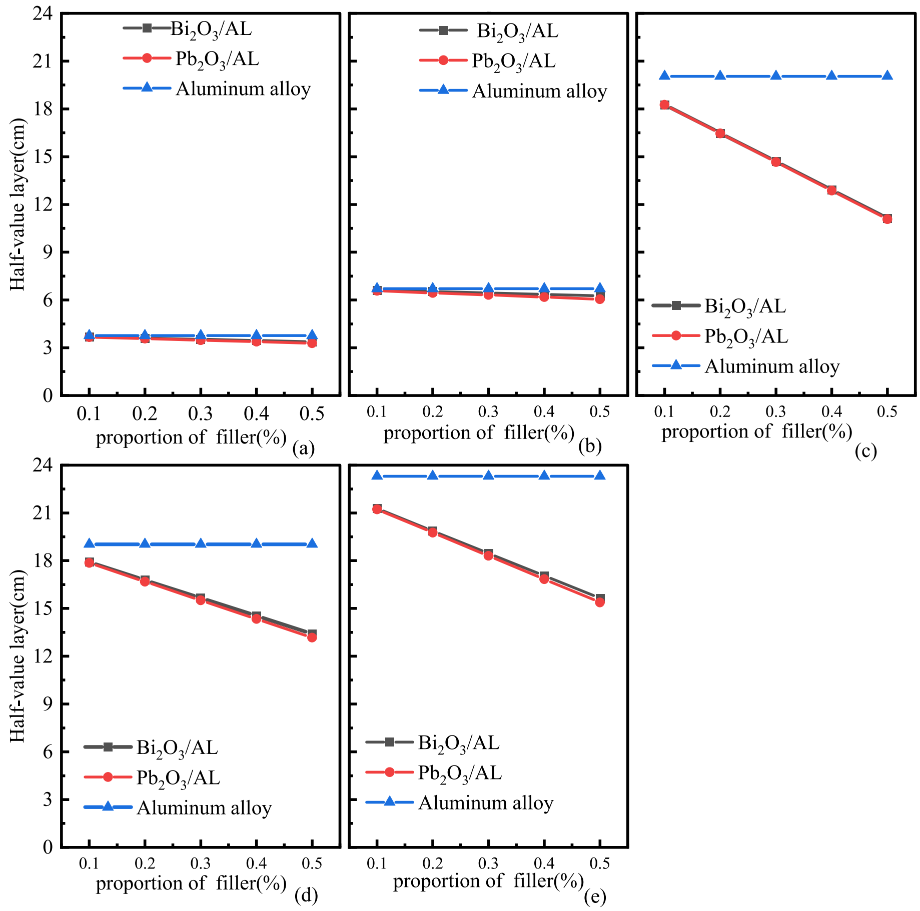

Fig. 5. The half-value layer values of different materials at 0.1 MeV(a), 0.5 MeV(b), 1 MeV(c), 3 MeV(d), 5Mev(e) gamma energies.

large storage capacity and are cost-effective. (iii) Bismuth-containing mixtures are environmentally friendly and almost harmless to humans. Bismuth can serve as an ideal substitute for lead as neutron shielding materials.

4. Conclusion

In this paper, various shielding materials are compared and analyzed to ensure the safety of shielding radioactive substances in spent fuel. In particular, the shielding effects and properties of bismuth oxide and lead oxide are compared, and the possibility of bismuth replacing lead as a shielding material for spent fuel is discussed.

The neutron flux model of shielding spent fuel is established by Monte Carlo method. The functional fillers such as bismuth oxide, lead oxide, boron oxide, gadolinium oxide and tungsten oxide are added to the matrices to compare the shielding rates of aluminum alloy matrix and silicone rubber matrix. The results indicate that the neutron shielding performance of the silicone rubber mixture is superior to that of the aluminum alloy mixture. Additionally, the radiation protection efficiency of the silicone rubber mixture remains above 56% regardless of changes in the shielding filler proportion. In the silicone rubber matrix, bismuth oxide and lead oxide have the same radiation protection efficiency. With the increase of the proportion of lead oxide and bismuth oxide, the neutron shielding performance of silicone rubber mixture can be slightly improved. The optimum proportion of bismuth oxide and lead oxide is 30%, and the radiation protection efficiency can reach 60%. With the increase of the proporition of tungsten oxide, the neutron radiation protection efficiency of the silicone rubber mixture not only does not increase, but also decreases.

Because the addition of functional fillers increases the neutron absorption cross section and the scattering cross section along the neutron propagation path, the mass attenuation coefficient of these five composites increases with the increase of functional fillers. The proportion of functional fillers has a greater effect on low energy neutrons. When the neutron energy is less than 2 MeV, the mass attenuation coefficient of bismuth oxide composite and lead oxide composite is greater than that of tungsten oxide composite; when the neutron energy is greater than 2 MeV, the mass attenuation coefficient of bismuth oxide composite and lead oxide composite is smaller than that of tungsten oxide composite.

On the other hand, Monte Carlo simulations are performed to evaluate the secondary gamma rays shielding performance. The results show that the five oxide shielding fillers have effective shielding for low energy photons, but inadequate shielding for high energy photons. And the shielding ability of bismuth oxide and lead oxide for secondary gamma rays is basically the same. For photon energies below 2.5 MeV, bismuth and lead composites provide better shielding than boron and tungsten composites.

To explore the possibility of replacing lead oxide with bismuth oxide as a shielding filler for spent fuel, the shielding effects of the composite materials are analyzed from the required thickness of the shielding material, and the half-value layer of the composite material containing bismuth oxide and lead oxide is compared. In the measured energy range, the bismuth oxide composite and the lead oxide composite have similar half-value layers. And with the increase of photon energy, the half-value layer of bismuth oxide composite and lead oxide composite increases. The results show that the shielding performance of low energy photons is good, while the shielding performance of high energy photons is poor.

In summary, bismuth oxide and lead oxide have similar capabilities in shielding neutrons and secondary gamma rays. However, bismuth oxide is better than lead oxide in mechanical properties, heat resistance, radiation resistance and environmental protection. Therefore, in the case of neutron source strengths in the range of 0.01–6 MeV and secondary gamma rays produced below 2.5 MeV, a bismuth-containing mixture can meet the basic requirements as a neutron shielding material and serve as an ideal substitute for lead.

Author contributions

All authors contributed to the study conception and design. The algorithm proposal and verification, raw data, and analysis were performed by Guo-Qiang Zeng, Shuang Qi, Peng Cheng, Sheng Lv, Fei Li, Qing-Ao Qin. The first draft of the manuscript was written by Guo-Qiang Zeng, Shuang Qi, Peng Cheng and Fei Li, and all authors commented on the previous versions of the manuscript. All the authors have read and approved the final version of the manuscript.

Data Availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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