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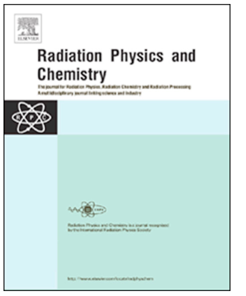

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Shams A.M. Issa: Investigation, Methodology, Writing – original draft, Visualization,

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Heavy metal oxide added glassy portable containers for nuclear waste management applications: *In comparison with reinforced concrete containers*

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Abstract

This study aimed to investigate the protective properties of Bi_2O_3 heavy metal oxide-doped glassy portable containers and the effect of reinforcement amount on these properties using the MCNPX (version 2.6.0) general-purpose Monte Carlo code. Accordingly, ^{60}Co and ^{137}Cs radioisotopes were defined as point isotropic radioactive sources to be transported with the newly designed containers. Four containers with different heavy metal oxide additives varying between 5% and 20% were designed and the deposited energy (MeV/g) values in the air were calculated for both ^{60}Co and ^{137}Cs radioisotopes. According to the findings of the first phase of the investigation, the sample (S4) with a 20% Bi_2O_3 additive ratio showed the highest protective properties and the least amount of deposited energy amount in the air. In the second and benchmarking phase of the investigation, we compared the amount of deposited energy in the air for the superior S4 glass container and a concrete container with a high amount of bitumen additive. The findings demonstrated that the S4 portable glass container with a 20% Bi_2O_3 reinforcement provided significantly lower deposited energy in the air and therefore greater nuclear safety than the concrete container. Heavy metal oxide-doped glass may be considered a viable choice for nuclear waste management and transportation operations due to its nuclear safety properties and superior physical, optical, and mechanical capabilities in comparison with concrete.

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Keywords: *Nuclear safety; Bi_2O_3 glasses; container; MCNPX; Monte Carlo simulation*

1. Introduction

Numerous facilities, buildings, and containers for the extended storage of nuclear waste substances are designed and developed using concrete(Ghouleh and Shao, 2018; Kurniawan et al., 2022). Reinforced concrete serves a variety of functions, including structural support, confinement, and environmental protection. Concrete is used for a wide variety of construction purposes, from surface structures to shallow subsurface vaults and deep subterranean repositories. Concrete structures in these facilities must meet additional requirements than those in conventional civil engineering applications, as decreased functionality or degradation of the material can be caused by both the contents (e.g., intense heat, exposure to radiation, and radionuclides from waste types) and external influences (e.g., chemical and physical attack), with embedded steel corrosion, leaching, increased temperature and irradiation being some of the primary risks (Gu et al., 2011; Han et al., 2020; Turick and Berry, 2016; Vupputuri et al., 2015). Additionally, depending on the application, the anticipated service life of these structures may vary from tens to hundreds or even thousands of years. During this time, reinforced concrete is required to act as a physical and chemical barrier between the waste and the surrounding environment (Arfa et al., 2022; International Atomic Energy Agency., 2005; Wisnubroto et al., 2021). Leaching and cracking will become more important in the long run, since water will serve as a transport medium for radionuclides if the other constructed barriers fail. Almost all chemical and physical processes that affect the durability of concrete buildings are influenced by transport mechanisms inside pores and fractures, as well as the presence of a fluid. The impermeability of concrete is consequently critical for the long-term durability of radioactive waste facilities. The permeability varies depending on the component quantities, degree of cement hydration, cement fineness, aggregate grade, and moisture level. Management of the factors which lead to an increase in the permeability or cracking of concrete is therefore critical for the long-term viability of radioactive waste management systems. Due to the risks associated with nuclear waste and comparable products during transportation and storage, researchers have developed a variety of materials which are more resistant to degradation. In recent years, researchers have prioritized the examination of heavy metal oxide-added glass composites and other types of glass as potential containers for the transportation of radioactive sources and other nuclear safety applications (Ojovan and Lee, 2011; Zakaly et al., 2021). Existing studies have determined and compared the radiation shielding properties of various types of glass materials with different kinds of heavy metal oxide (HMO) reinforcement across broad gamma energy ranges(ALMisned et al., 2021; Javaherdashti, 2009; Kassab et al., 2022; Kim and Yi, 2017; Kurudirek, 2017; Mostafa et al., 2020; Othman et al., 2019; Zakaly et al., 2022). While investigating the shielding properties of individual materials is critical for nuclear radiation applications, the design of these materials for practical applications and the subsequent situation in terms of environmental radiation and worker radiation safety should be investigated through experimental or advanced simulation methods. The purpose of this work was to construct various Bi_2O_3 heavy metal oxide doped glass materials(Abouhaswa et al., 2021a, 2021b; Rammah et al., 2021; H O Tekin et al., 2022; Huseyin Ozan Tekin et al., 2022) as nuclear containers and to explore the impact of increasing the quantity of heavy metal oxide on the amount of energy deposited in the air. The nuclear safety properties of the newly designed Bi_2O_3 reinforced glass container against Cobalt (^{60}Co) and Cesium (^{137}Cs) radioisotopes were investigated and compared to the reinforced concrete container (Reda and Saleh, 2021) using general-purpose MCNPX (version 2.6.0) (computer code Collection, 2002) Monte Carlo code. Also, By Using the MCNPX (version 2.6.0), the purpose of this study was to investigate the protective properties of

Bi_2O_3 heavy metal oxide-doped glassy portable containers and the effect of reinforcement amount on these properties. Specifically, the study wanted to look at how the amount of reinforcement affected these properties. In light of this, the radioisotopes 60Co and 137Cs were categorised as point isotropic radioactive sources, and they were designated to be carried using the newly constructed containers. The deposited energy values (MeV/g) in the air were determined for both 60Co and 137Cs radioisotopes, and four distinct containers were created with different heavy metal oxide additions ranging from 5 percent to 20 percent. The results obtained from the current investigation may provide significant scientific data to provide direction for the development and design of alternative containers for nuclear waste management.

2. Materials and Methods

In this study, four different portable heavy metal oxide reinforced glass (Barebita et al., 2020) containers were modeled using the MCNPX Monte Carlo code. Elemental compositions and material densities of the modeled containers are given in Table 1. In the composition variation from the S1 sample to the S4 sample, the most dominant change occurred in the Bi2O3 heavy metal oxide contribution from 5% to 20%. As a consequence of this net change of 15%, glass densities rose directly, with the density of the S1 sample increasing from 3.1589 g/cm^3 to 4.0108 g/cm^3 in the S4 sample. All simulations were performed using Lenovo® ThinkStation-P620/30E0008QUS Workstation-1x AMD-Ryzen, Threadripper PRO Hexadeca-core (16Core) 3955WX 3.90GHz-32 GB DDR4 SDRAM RAM.

2.1 Design of portable glass container in MCNPX code

Preparing the INPUT file in line with the code hierarchy should be regarded as the fundamental stage for studies using MCNPX code. In the MCNPX INPUT file, there are three main definition sections, namely CELL, SURFACE, and DATA cards. First, the boundaries of cellular structures concerning geometric surfaces are outlined in this study. Figure 1 depicts the model's portable glass container in two dimensions. As demonstrated, a point isotropic source geometry is placed on the central point of the simulation world (i.e., 0,0,0). A new cellular structure for the portable glass containers was created using the chemical compositions of the S1, S2, S3, and S4 glass, given in Table 1. Finally, in the input file, an air cell was defined that surrounded the glass container. Since the primary goal of this investigation was to measure the total energy deposited in the air, the air cell has been covered by an outer attenuator material. This was accomplished by accumulating the energy released into the air via radioactive decay of administrated ^{60}Co and ^{137}Cs radioisotopes housed inside the container, and then simulating the emission of that energy into four different containers.

2.2 Measurements of deposited energy amounts

INPUT files were prepared with two different radioisotopes for four different containers, therefore, a total of eight different INPUT files were created. Afterwards, these created INPUT files were run one by one for 10^8 particle numbers and the output files were saved, respectively. Figure 2 shows the MCNPX visual editor's three-dimensional rendering of the modeled portable glass container. The radioisotope is fully incorporated into the cylindrical container, and its top and bottom sections are covered. F6 TALLY MESH, a key output recording component of the MCNP code, is used in this study. Using F6 TALLY MESH, one can obtain total energy deposition in a cell as MeV/g or jerks/g. Three F6 TALLY MESH definitions were inserted into three distinct cell volumes in this research. Cells 10, 11, and 12 shown in Figure 1 and the air surrounding the portable glass container, shown in Figure 2, are the three separate cellular volumes. Each time a simulation cycle is completed, the energy deposition amounts in these three cells are extracted from the output file and summed. Thus, the sum of the energy deposition in the three cellular volumes of air surrounding the container was provided for each simulation cycle. As a standard definition in the eight different INPUT files, the F6 TALLIES were defined as below.

-F6:P 10
-F16:P 11
-F26:P 12

Each row represents a single F6 TALLY MESH, which has been constructed to record solely the energy deposits induced by photons (i.e., P) in cells 10, 11, and 12.

3. Results and Discussions

In this study, four portable glass containers with different elemental compositions were designed and their nuclear radiation protection potentials were comprehensively investigated. Two separate radioisotopes were used for each container to calculate the energy deposition in the air around the designed portable glass containers. To begin, the ^{60}Co radioisotope was placed into the designed glass containers, and the deposited energy (MeV/g) into the surrounding air cell was measured for each of the four containers. Figure 3 shows the variation of energy deposition amount in the air as a function of increasing Bi_2O_3 contribution in the glass samples for the ^{60}Co radioisotope. As demonstrated, the quantity of energy deposited in the air has changed in inverse proportion to the increasing amount of Bi_2O_3 in the elemental composition of the glass. This is supported by findings in the literature, where it has been reported that increasing quantities of Bi_2O_3 in glass materials improves their gamma-ray attenuation capabilities(Kurtulus et al., 2021; Lakshminarayana et al., 2021b, 2021a; Mahmoud et al., 2021). In this simulation, increasing these shielding properties in the positive direction had a direct effect on the amount of energy deposited in the air surrounding the container, with the S4 container (20% Bi_2O_3) demonstrating minimum

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energy deposition in the air for the ^{60}Co radioisotope (See Table 2). ^{137}Cs was used as the isotropic point radiation source in the next phase of the simulation, again, placed inside the designed containers S1, S2, S3 and S4 (see Figure 2). As before, the gamma-ray energy released by the point source and its influence on the surrounding air cell was computed for four samples. Figure 4 depicts the variation of energy deposition amount in the air as a function of increasing Bi_2O_3 contribution in the glass samples for ^{137}Cs radioisotope. A similar trend in the decrement was reported also for ^{137}Cs radioisotope. Despite the fact that the four glass samples were equal in terms of their elemental compositions, the quantity of energy deposited in the air significantly varied for both radioisotopes. This is illustrated in Figure 5, comparing the amounts of energy emitted through four different containers and deposited in surrounding air for both ^{60}Co and ^{137}Cs radioisotopes. It is clear to see that the overall quantity of energy deposited in the air varies for the two radioisotopes. This is explained by the gamma-ray energy specific to each radioisotope - the ^{60}Co radioisotope emits gamma rays with energies of 1.17 MeV and 1.33 MeV, while the ^{137}Cs radioisotope emits gamma rays with energies of 0.662 MeV (Lee et al., 2021; Murphy and Kamen, 2019; Zakaly et al., 2019). Meanwhile, color mapping was performed on the total amount of energy deposited in the air by ^{60}Co and ^{137}Cs radioisotopes, and the energy change trend on the scale from S1 to S4 is displayed in Figure 5. Figure 5 demonstrates that the maximum amount of energy deposited in the air decreases as the quantity of Bi_2O_3 in the material increases, i.e. for columns S1 to S4 (i.e., from left to right), while the minimum amount of energy deposited in the air increases. This situation demonstrates the direct influence of the elemental composition on the amount of energy deposited in the air, ranging from S1 to S4. As a result, one may say that the maximum quantity of blue zone for the S4 sample, which gives the greatest level of radiation protection, provides the maximum level of protection for this type of portable glass container. In the final

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phase of this study, the obtained results were compared for another container structure and the results were presented in Figure 7 and Figure 8 as a function of the amount of deposited energy (MeV/g) in the air. In a previous study, Reda and Saleh (Reda and Saleh, 2021) investigated the gamma radiation shielding efficiency of the cement-bitumen portable container using ^{60}Co and ^{137}Cs radioisotopes. Their findings indicated that a Cement 50% + Bitumen 50% mixture of cement and bitumen provided the highest shielding effectiveness. They have, however, explored the attenuation coefficients of the container models. We created the Cement 50% + Bitumen 50% mixture as a container under the same conditions as the newly designed glass containers presented in this investigation. As a result, we repeated the analyses for measuring the deposited energy in the air for ^{60}Co and ^{137}Cs radioisotopes in order to provide a comparison phase between the S4 sample and the Cement 50% + Bitumen 50% sample. Our results showed that the S4 sample has better protection efficiency than Cement 50% + Bitumen 50% container for both the ^{60}Co and ^{137}Cs radioisotopes (Figures 7 and 8).

4. Conclusion

The widespread application of nuclear technology in a variety of fields, from industry to health, has kept a broad range of technological investments in this field, from devices used in this field to protective equipment, in a state of continuous development and resulted in the establishment of its own research and development branches. Transportation, storage, and preservation of nuclear materials are some of these fields of study. While protection features are critical for the long-term preservation of nuclear waste, improving existing protection features through the implementation of innovative approaches in this area is critical in terms of both environmental and sustainable costs. In recent years, glass materials have emerged as the leading model for radiation shielding applications due to their desirable physical and structural attributes, and they continue to do so.

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The primary objective of this research was to contribute some practical results to the literature by expanding the possibility of employing glass materials in nuclear applications. This simulation found that the addition of Bi_2O_3 heavy metal oxide may be employed effectively to enhance the gamma-ray shielding characteristics of glass samples. This has the consequence of significantly reducing the quantity of deposited energy in the air, which is the primary topic of this research. Reducing the amount of energy deposited in the air to minimal levels is imperative for public health and environmental health issues. The comparison phase of this study provides evidence of the superiority of the heavy metal oxide-added, high-density glass (S4) over a 50% concrete + 50% bitumen container. This may provide motivation for the further development of the glass composition presented here as the S4 sample. Glass is highly durable, mostly transparent, simple to produce, non-toxic, and flexible in its use. This condition may serve as a powerful incentive for the scientific community to continue developing current features. Further work may be carried out to investigate the mechanical and thermal characteristics of the material suggested here.

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Table 1: Samples code, elemental weight fraction, density, and molar volume of (0.25-x) Bi_2O_3 -xB_2O_3 -0.75 (50%P_2O_5 -50%V_2O_5):x=0.05, 0.10, 0.15, and 0.20 glasses.

Code	Elemental weight fraction (wt.%)					Density (g/cm^3) [31]	Molar volume (cm^3/mol) [31]
	B	O	P	V	Bi		
Sample 1	0.027254	0.453788	0.146422	0.240816	0.13172	3.1589	50.2263
Sample 2	0.018171	0.403401	0.130164	0.214076	0.234189	3.5016	50.9738
Sample 3	0.010903	0.363085	0.117155	0.192681	0.316176	3.6909	53.7240
Sample 4	0.004956	0.330095	0.10651	0.175174	0.383264	4.0108	54.3781

Table 2: Total energy depositions (MeV/g) in the air for ^{60}Co and ^{137}Cs isotopes

	^{60}**Co**	^{137}**Cs**
Sample 1	0.00010055	0.00005738
Sample 2	0.00009977	0.00005591
Sample 3	0.00009926	0.00005493
Sample 4	0.00009889	0.00005419

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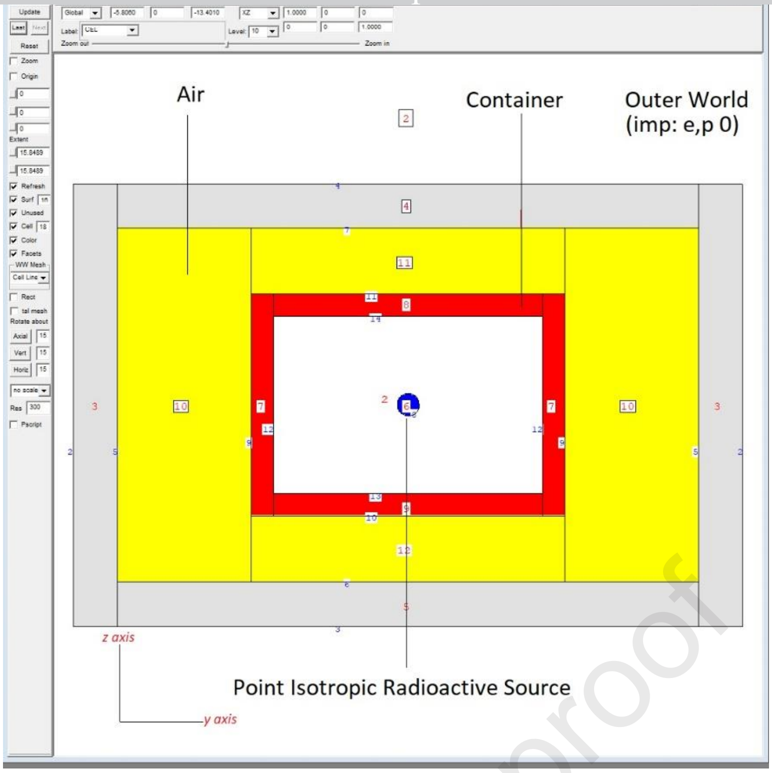

Figure 1: 2-D view of modelled glassy nuclear container (obtained from MCNPX Visual Editor**)**

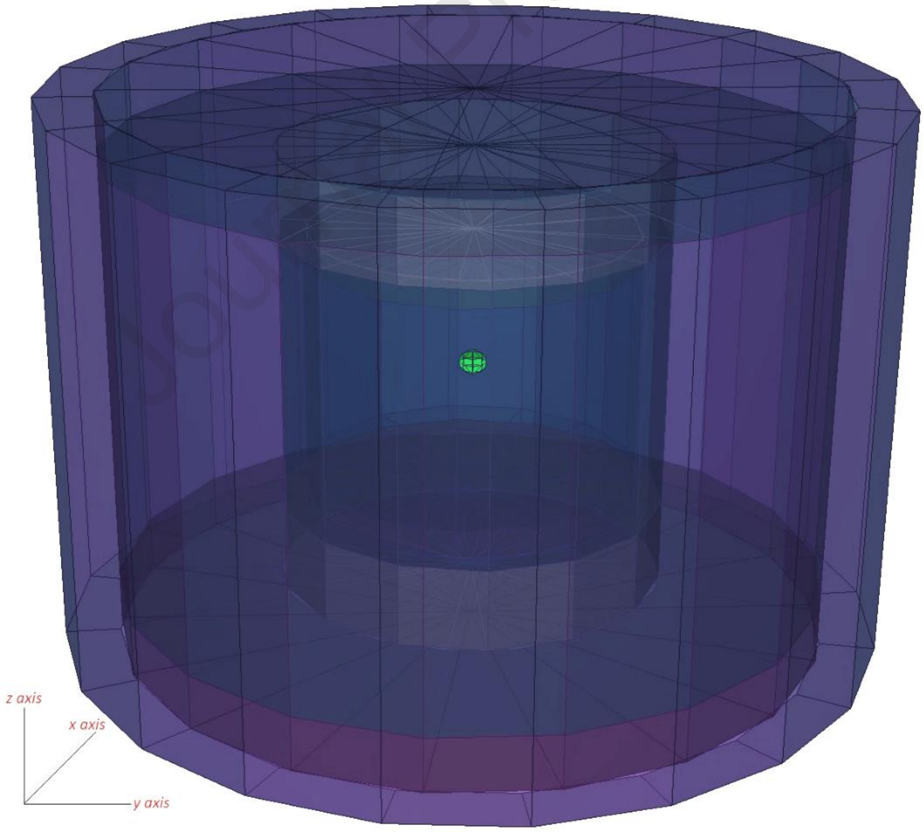

Figure 2: 3-D view of modelled glassy nuclear container (obtained from MCNPX Visual Editor**)**

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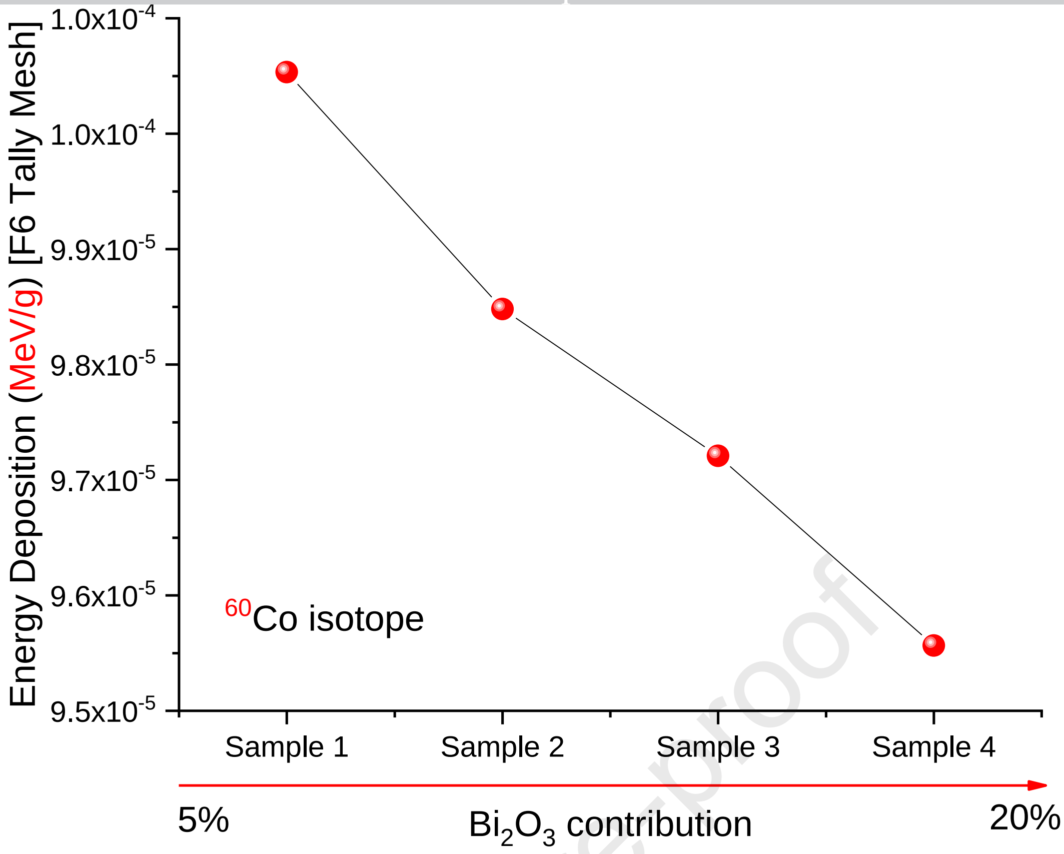

Figure 3: Variation of energy deposition in the air emitted from ^{60}Co isotope as a function of increasing Bi_2O_3 reinforcement in the container structure

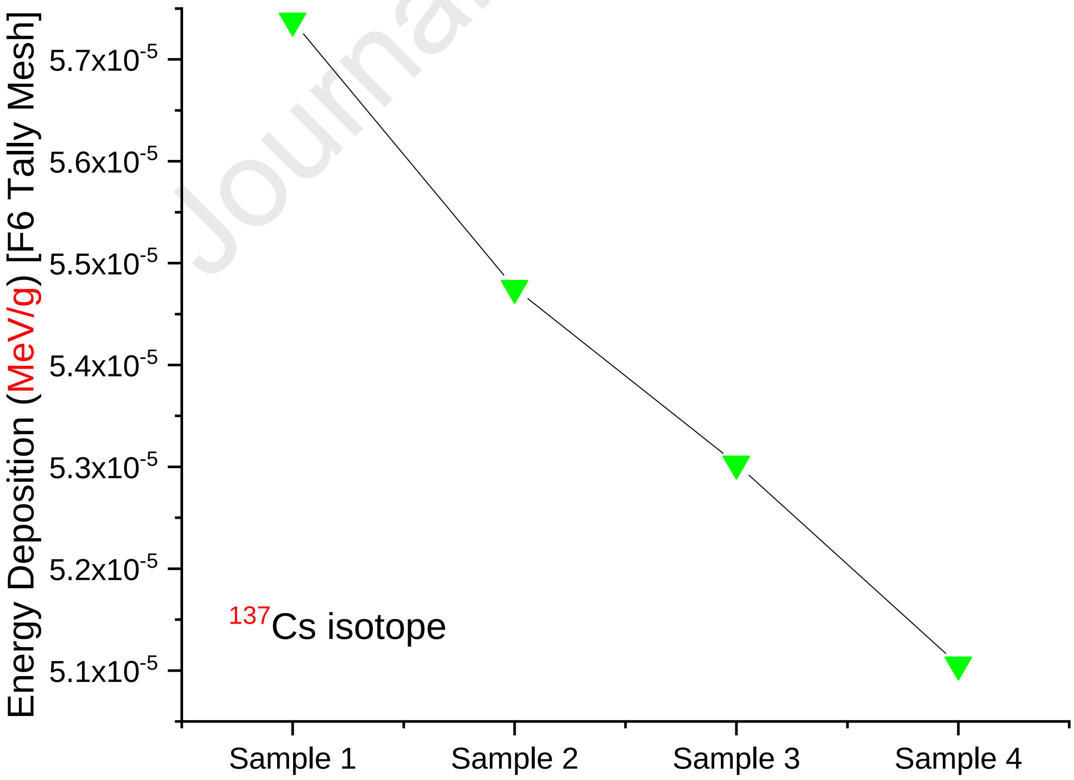

Figure 4: Variation of energy deposition in the air emitted from ^{137}Cs isotope as a function of increasing Bi_2O_3 reinforcement in the container structure

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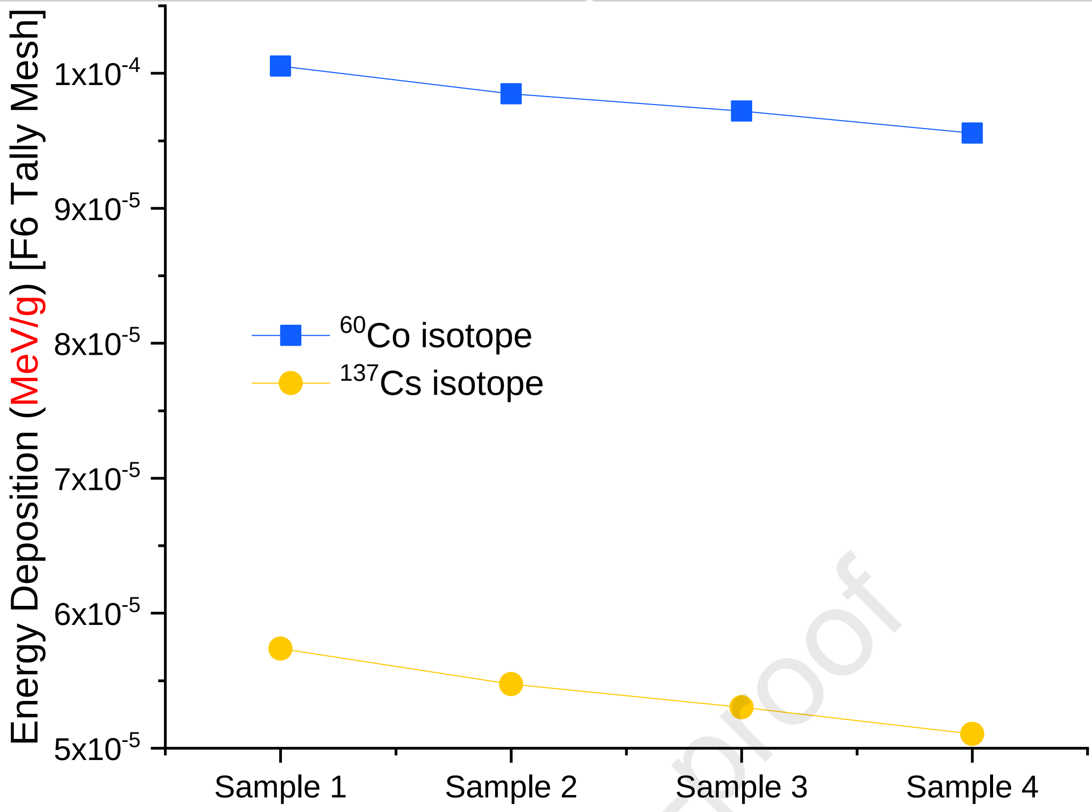

Figure 5: Comaprison of energy deposition in the air for ^{60}Co and ^{137}Cs isotopes

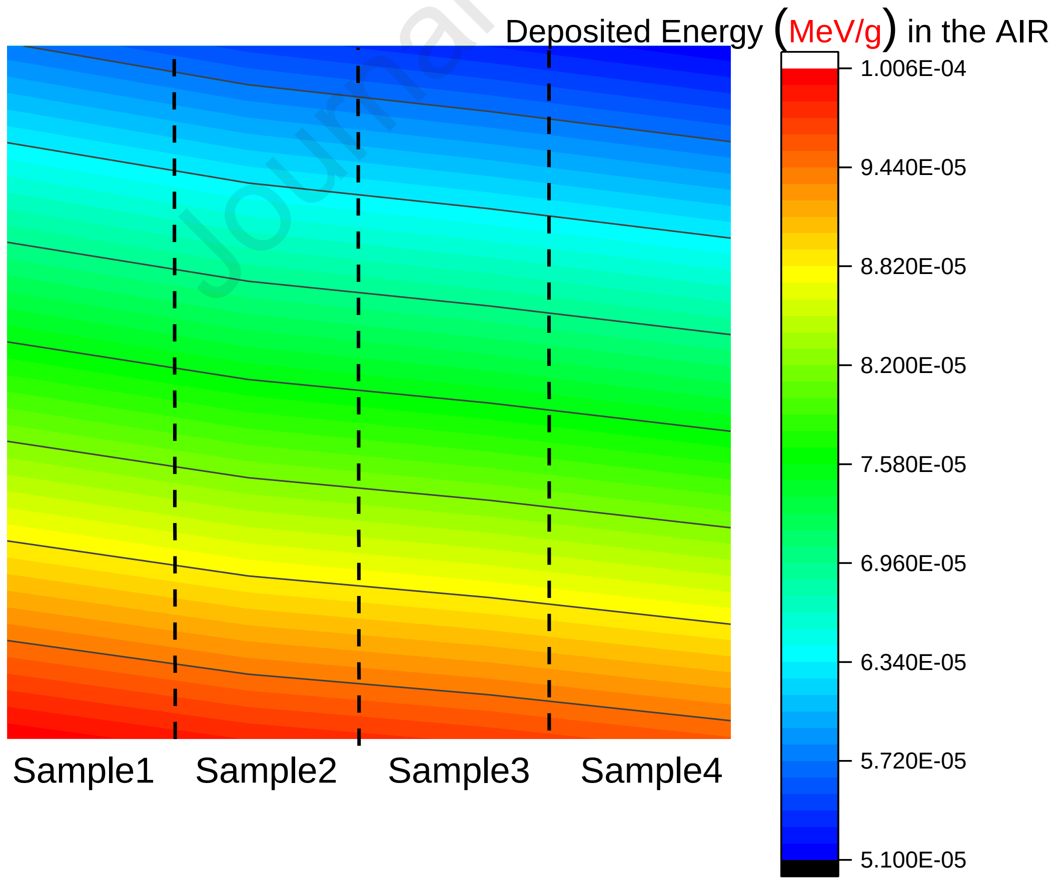

Figure 6: Distribution of energy in the air for designed glassy nuclear containers

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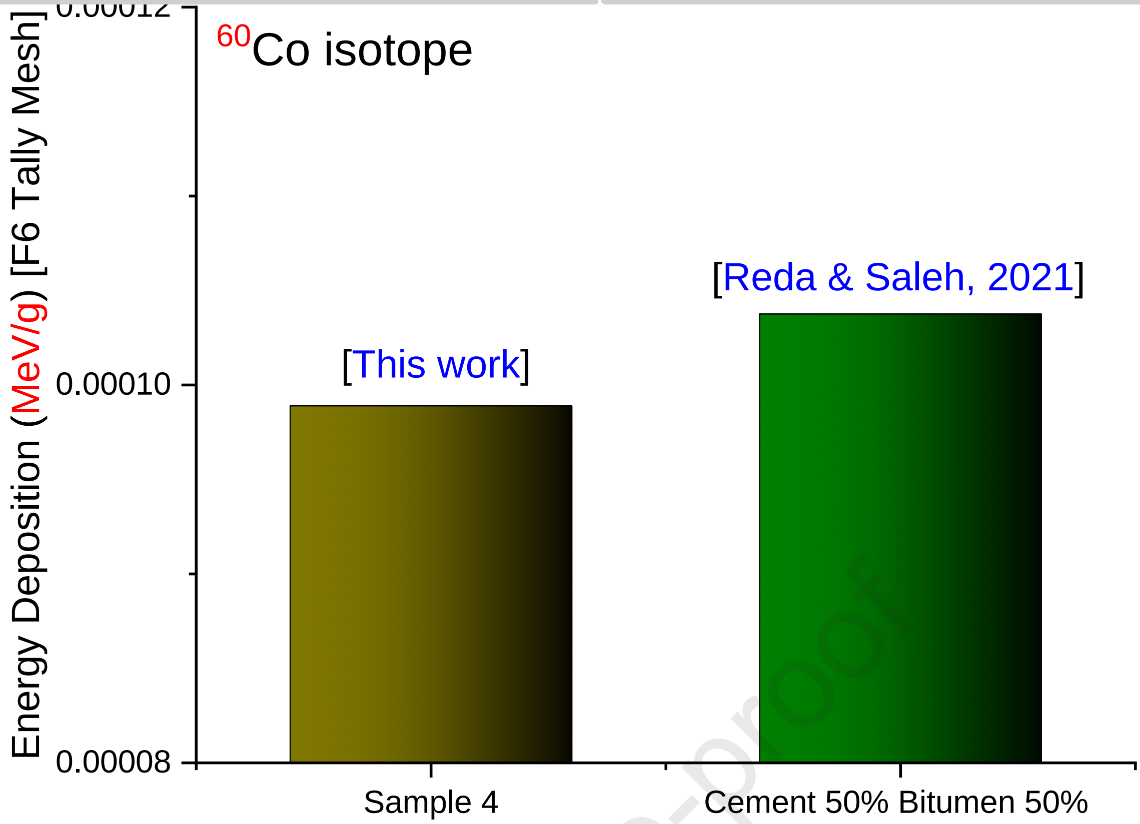

Figure 7: Comparison of energy deposition in the air emitted from ^{60}Co isotope

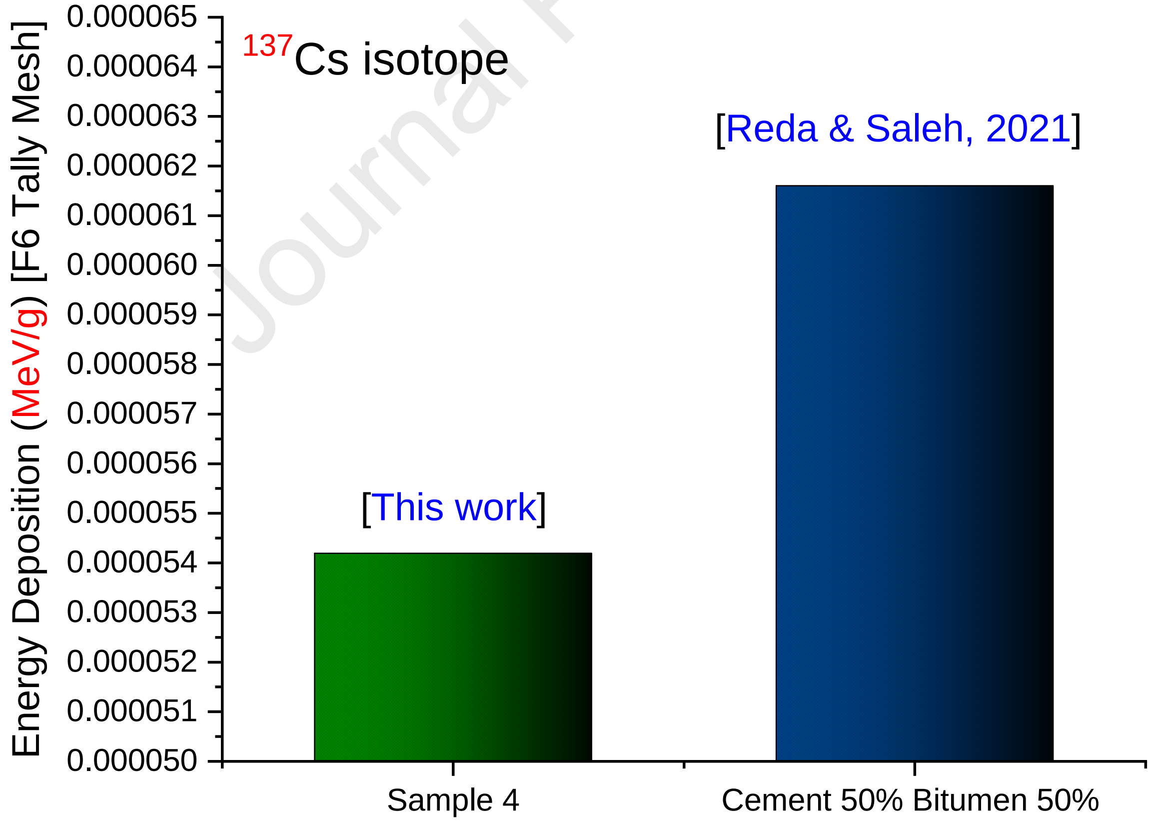

Figure 8: Comparison of energy deposition in the air emitted from ^{137}Cs isotope

Highlights

- Heavy metal oxide-doped glassy portable containers were designed
- MCNPX (version 2.6.0) general purpose Monte Carlo code was used
- ^{60}Co and ^{137}Cs radioisotopes were used as point isotropic sources
- Deposited energy (MeV/g) amount has been measured
- S4 container with a 20% Bi_2O_3 reinforcement provided significantly lower deposited energy in the air

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Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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