

Research Article

The Gamma Ray, Thermal and Fast Neutrons Shielding Properties of Gd₂O₃-SiO₂-Y₂O₃-CaO-B₂O₃ Glass Series Using FLUKA Simulation Code

Kittisak Sriwongsa^{1,2*}, Papungkorn Tantiamnuay², Pongrawee Thepchai², Punsak Glumglomchit³, Punda Papatron³, Patnaree Yangyuen³, Sunantasak Ravangvong⁴, Chumphon Khobkham⁵, Chalermpon Mutuwong⁶ and Cherdsak Bootjomchai⁶

Received: 7 January 2023

Revised: 6 March 2023

Accepted: 13 March 2023

ABSTRACT

The γ -ray, thermal and fast neutrons shielding properties of Gd₂O₃-SiO₂-Y₂O₃-CaO-B₂O₃ glass series have been studied. The mass attenuation coefficients (μ_m) for γ -ray of this glass series have been obtained at an energy range 59.6-1332 keV using Phy-X software and FLUKA Monte Carlo code simulation. The obtained results were found to be in good agreement. The 50Gd₂O₃-20SiO₂-5Y₂O₃-10CaO-15B₂O₃ glass sample showed the highest μ_m value with the lowest half value layer (HVL) and mean free path (MFP) value compared with the other ones in this glass series. As for the case of thermal and fast neutron shielding properties, the FLUKA Monte Carlo code and partial density method were used for stimulation, respectively. It was found that 50Gd₂O₃-20SiO₂-5Y₂O₃-10CaO-15B₂O₃ glass sample showed the mass attenuation coefficients (μ_m) for thermal neutrons decreased with increasing Gd₂O₃ content for fast neutrons, and the removal cross section (Σ_R) showed the highest value while half value layer (HVL) and the relaxation lengths showed the lowest value. These results indicated that this glass series has high Gd₂O₃ content possesses excellent γ -ray and fast neutron shielding properties, while the 20SiO₂-5Y₂O₃-10CaO-65B₂O₃ glass sample possesses superb thermal neutrons shielding.

Keywords: FLUKA, Gamma-ray shielding, Thermal neutrons, Fast neutrons, Glass

¹ Lecturer responsible for the Bachelor of Education Program in Physics, Faculty of Education, Silpakorn University, Nakhon Pathom, Thailand

² The demonstration school of Silpakorn University, Nakhon Pathom, Thailand

³ Huahinvitthayalai School, Hua-Hin, Prachuap Khiri Khan, Thailand

⁴ Division of Science and Technology, Faculty of Science and Technology, Phetchaburi Rajabhat University, Phetchaburi, Thailand

⁵ Faculty of Engineering, Thonburi University, Bangkok, Thailand

⁶ Department of physics, Faculty of Science, Ubon Ratchathani University, Ubon Ratchathani, Thailand

* Corresponding author, email: sriwongsa_k@silpakorn.edu

Introduction

Gamma (γ) ray is ionizing radiation. It poses a threat to both the environment and human health. For this reason, γ -ray shielding researchers are interested in investigating new materials for the development of γ -ray shielding materials [1]. Glass is a great alternative material due to its opaque concrete and the toxicity of lead (Pb) as a radiation shielding material [2,3]. The significant factors are the high degree of transparency of the glasses, lack of health hazards, resistance to corrosion, and environmental friendliness. Concrete frequently has issues including being opaque to visible light, having cracks, and losing water when exposed to radiation from nuclear reactors for an extended period of time. Pb-based glass is a fantastic option since it has superior and stable chemical and physical characteristics [4–6]. However, since Pb and the toxicity of its compounds are site-specific, therefore it is forbidden to use it as radiation shielding.

Because heavy rare earth oxides can be employed as an effective γ -ray absorber, heavy rare earth oxides glasses are currently very promising for γ -ray shielding applications. One of the heavy rare earth ions that offers the platform to co-dope with other rare earth for their effective energy transfer properties included in glasses as activators is gadolinium oxide (Gd_2O_3 , density = 7.41 g/cm^3). Moreover, Gd_2O_3 will not develop a color center in the glass matrix and is therefore chosen to be added to glass structures. The Gd_2O_3 's high atomic number and high density can increase the probability that photon energy will interact with glasses [7–9]. The study also discovered that the effectiveness of shielding against neutrons and γ -ray can be improved by the addition of Gd_2O_3 [10,11]. Furthermore, gadolinium has a high atomic number ($Z = 64$), high density (density = 7.90 g/cm^3), high absorption cross-section for thermal neutron (49700b), high refractive index, and high stopping power [12].

FLUKA (FLUktuirende KAskade) is the Monte Carlo computing code that simulates the transport of particles and their interactions with matter [13]. About 60 distinct particles constitute FLUKA, which can be employed for a variety of tasks like radiation therapy, design, and shielding [14–17]. Collaboration between CERN (European Council for Nuclear Research) and INFN (Istituto Nazionale di Fisica Nucleare) was used to create FLUKA. It has two modes of operation: a biased mode and an entirely analog mode. Other Monte Carlo codes do not offer this feature. To the best of our knowledge, no author has used FLUKA to study various shielding parameters of composite glasses, despite the fact that certain writers [18,19] have reported the mass attenuation coefficients of soil samples and concretes using the FLUKA Monte Carlo code. The fundamental benefit of the simulation technique is the generation of theoretical data for the experiment's flexible geometry and the number of primary radiations, which can be used to assess the feasibility of specific experimental measurements.

The $xGd_2O_3-20SiO_2-5Y_2O_3-10CaO-(65-x)B_2O_3$ glass system, where $x = 0, 10, 20, 30, 40$ and 50 mol\% were analyzed for the γ -ray shielding properties using FLUKA Monte Carlo code. The characteristics that needed to be examined were the mass attenuation coefficient (μ_m), half value layer (HVL) and mean free path (MFP). In this work, ^{241}Am , ^{22}Na , ^{133}Ba , ^{137}Cs , and ^{60}Co are the radiation sources to simulate a radiation transmission event. The thermal and fast neutrons shielding properties

were also simulated using the FLUKA Monte Carlo code and partial density method, respectively, to cover applications in practice.

Materials and Methods

Glasses description

The theoretical glass series of this work is in formula $x\text{Gd}_2\text{O}_3-20\text{SiO}_2-5\text{Y}_2\text{O}_3-10\text{CaO}-(65-x)\text{B}_2\text{O}_3$, which x increased from 0-50 mol% in 10 mol% increments, and they have been displayed in Table 1.

Table 1 The chemical composition for glass series (mol%).

Elements	Glass code (mol%)					
	S1	S2	S3	S4	S5	S6
Gd ₂ O ₃	0	10	20	30	40	50
SiO ₂	20	20	20	20	20	20
Y ₂ O ₃	5	5	5	5	5	5
CaO	10	10	10	10	10	10
B ₂ O ₃	65	55	45	35	25	15

Physical parameter

The density (ρ ; g/cm³) of the glass series is an important quantity for the evaluation of the other values and can be calculated using equation (1) [20]:

$$\rho = (0.53) \frac{\left(\sum M_i x_i\right)}{\left(\sum V_i x_i\right)} \quad (1)$$

where M_i , V_i and x_i are molecular weight, packing density factor, and molar fraction of i^{th} component, respectively.

Gamma-ray interaction

The mass attenuation coefficients (μ_m) express the probability of interaction of γ -ray with the interacting medium. The μ_m of the selected glass samples were derived from Phy-X software for γ -ray energy range 356-1332 keV using the following equation (2) [21,22]:

$$\mu_m = \sum_i w_i (\mu_m)_i \quad (2)$$

where $(\mu_m)_i$ is the mass attenuation coefficient for the i^{th} element, and w_i is the weight fraction of the i^{th} element in the mixture.

Half value layer (HVL) is defined as the thickness of the sample that reduces the γ -ray intensity to half of its initial intensity and can be calculated from the linear attenuation coefficient (μ) by using equation (3) [21,22]:

$$HVL = \frac{0.693}{\mu} \tag{3}$$

Mean free path (MFP) is an average distance between two successive interactions of γ -ray in medium and can be simulated from equation (4) [21,22]:

$$MFP = \frac{1}{\mu} \tag{4}$$

FLUKA simulation

FLUKA (FLUktuierende KAskade) is favors used in particle-nuclear sciences. It is mainly in high energy experimental physics and engineering shielding. In this work, the FLUKA Monte Carlo code version 2011.2x-4 with schematic set up for simulate γ -ray and thermal neutrons are exhibited in Figures 1 and 2. The simulation for μ_m of material samples can be estimated using equation (5) [22,23]:

$$I = I_0 e^{-\mu x} \tag{5}$$

where I and I_0 are γ -ray and thermal neutrons intensity passed through without and with shielding material, respectively, and x is thickness of sample.

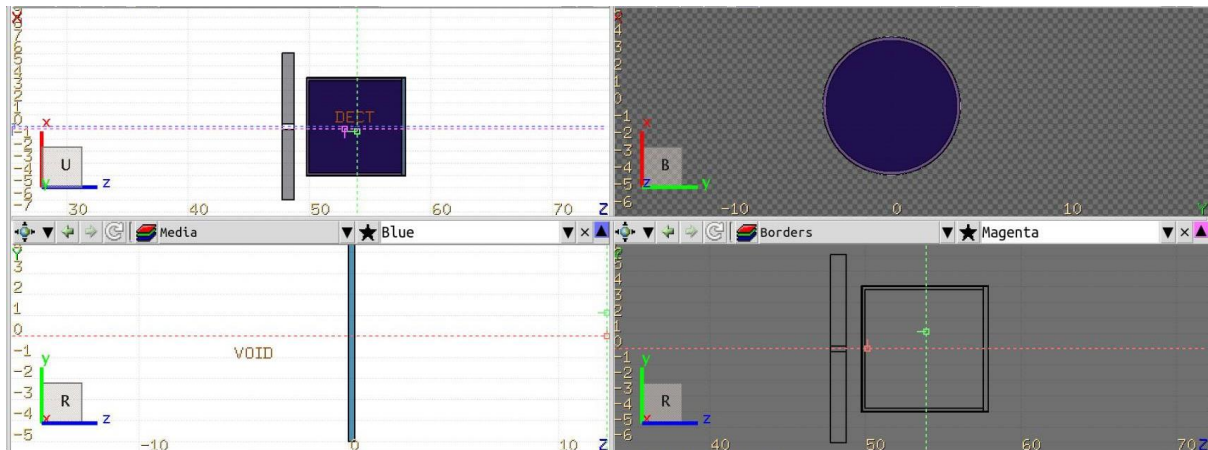


Figure 1 The schematic set up to simulate γ -ray.

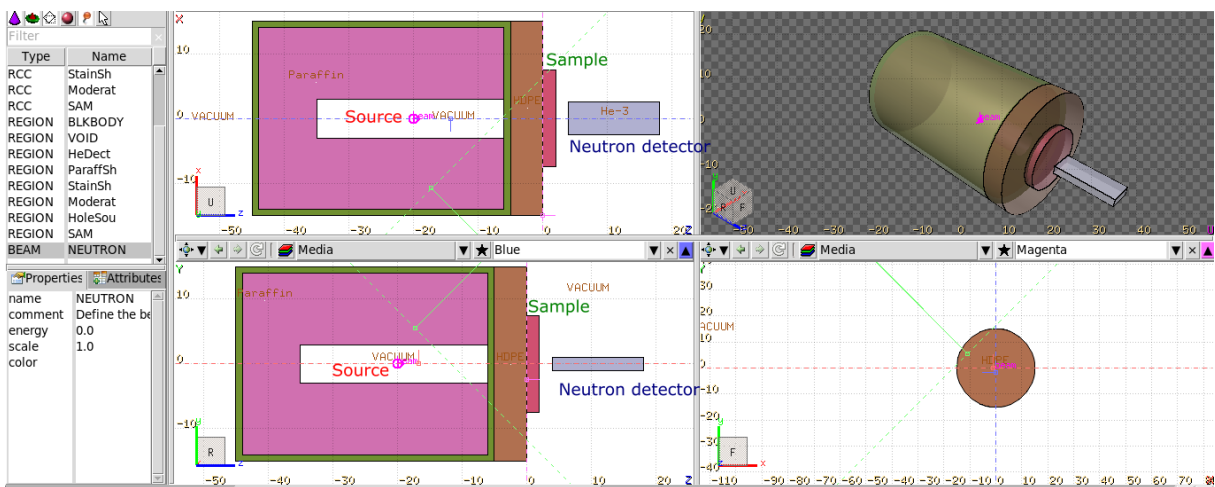


Figure 2 The schematic set up to simulate thermal neutrons.

Fast neutrons shielding properties

Microscopic cross section indicated the possibility of each type of the interaction between neutrons and medium consisting of transmission, absorption and scattering of neutrons. Those interactions are processes occurring through the microscopic cross section of the reaction [24,25].

The fast neutrons ($E > 0.1$ MeV) removal cross section (Σ_R) has been determined by applying relationship in equation (6):

$$\Sigma_R = \sum_i W_i (\Sigma_{R/\rho})_i \tag{6}$$

where $W_i = \rho w_i$ refers to partial density, w_i and ρ refer to a fraction by weight of element i and total density of the medium, respectively. In addition, Σ_R is the basic parameter using for the calculation of half value layer (HVL) and the relaxation lengths [25-27].

Results and Discussion

Physical parameter

The density is an important physical quantity to explain the changing formation of glass. As exhibited in Figure 3, the density of the glass series increases with increasing Gd_2O_3 content. This event is due to the replacement of Gd_2O_3 with higher molecular mass and density than B_2O_3 . That indicated the presence of Gd^{3+} ions in glass structure which adjusted the borate glass network by converting more $[BO_3]^{3-}$ triangles to BO_4^{4-} tetrahedral [28,29].

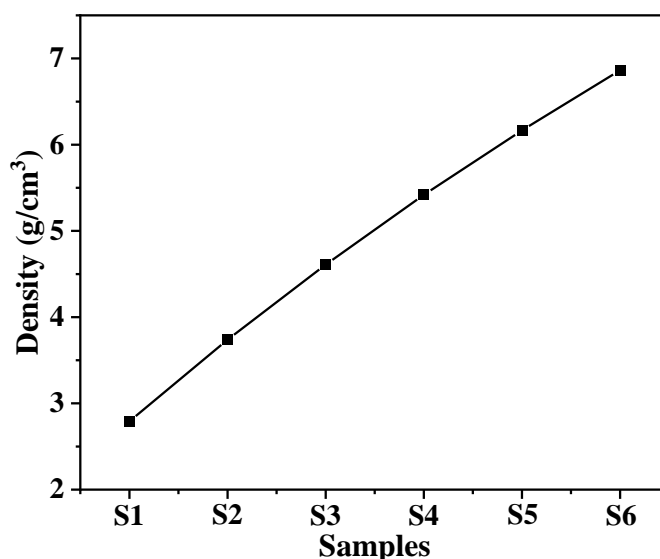


Figure 3 The density of glass series.

Gamma ray interaction

The mass attenuation coefficient (μ_m) for glass series at γ -ray energy is in the range 59.6-1332 keV at sample thickness 0.1-0.3 cm. The ^{241}Am (59.6 keV), ^{22}Na (511 and 1275 keV), ^{133}Ba (356 keV), ^{137}Cs (662 keV), and ^{60}Co (1173 and 1332 keV) are the radiation sources to simulate a radiation transmission event as exhibited in Figure 4. It can be seen that for the γ -ray energy range 59.6–356

keV, the value of μ_m increased with increasing Gd_2O_3 content, but above 356 keV these glass series properties were not good shielding anymore. From the results in Figure 4, the value of μ_m at energy range 600–1332 keV showed the same tendency of increasing μ_m value with these glasses series. These results were due to the replacement of the lower atomic number, B ($Z = 4$), with the higher atomic number, Gd ($Z = 64$), also, according to the main of photoelectric effect (PE) process at low energy ranges where the PE probability was large [28].

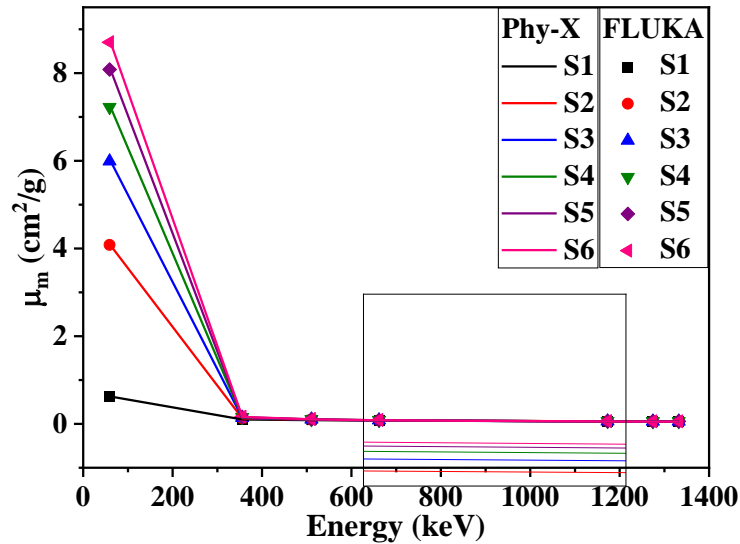


Figure 4 The mass attenuation coefficient for glass series at γ -ray energy range 59.6-1332 keV.

The HVL and MFP were plotted as shown in Figures 5 and 6. It was found that both values increased with increasing γ -ray energy, but S6 sample showed the lowest values of the HVL and MFP. It can be concluded that high density and high Z elements would be good γ -ray shielding [23,28].

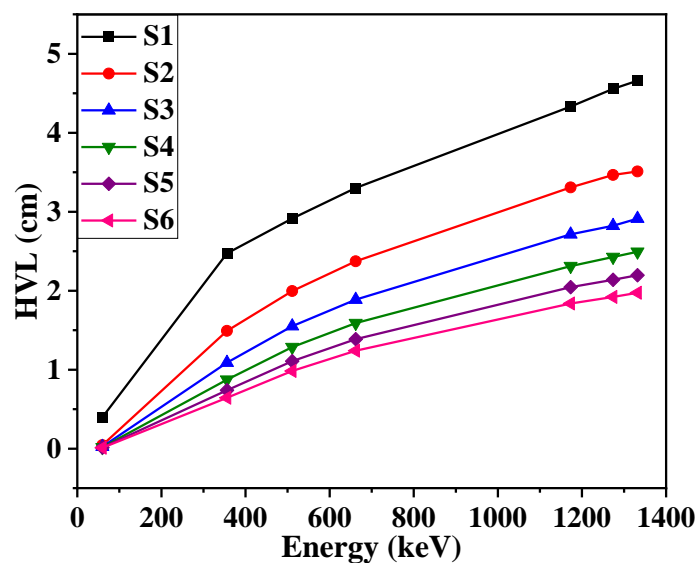


Figure 5 The HVL for glass series at γ -ray energy range 59.6-1332 keV.

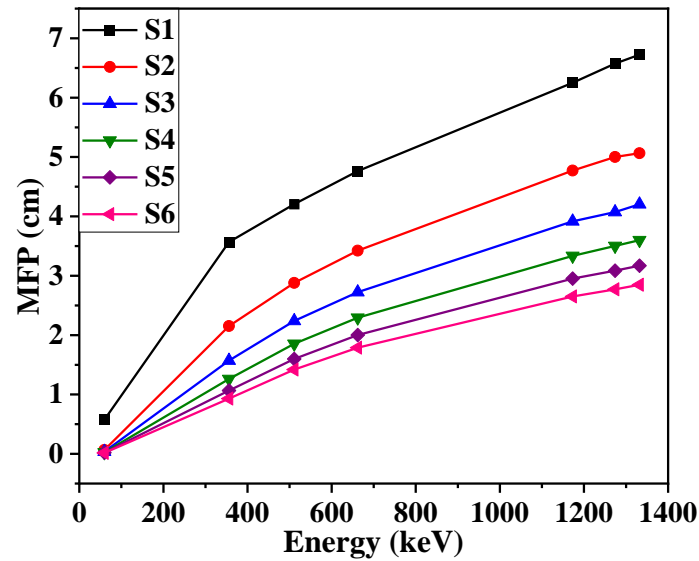


Figure 6 The MFP for glass series at γ -ray energy range 59.6-1332 keV.

Thermal neutrons shielding properties

The ^{241}Am -Be was used for the neutron source which was contained in a sealed thick paraffin cylinder and stacked 5 cm-HDPE sheets as a neutron moderator. He-3 was used for the neutron detector and all samples were placed between the neutron detector and the HDPE sheet [30]. This source emits neutrons at an energy range 1-12 MeV as a continuous spectrum because of (α , n) reaction with an average energy of 4.9 MeV as displayed in Figure 7 [31]. The thermal neutron passing through high density polyethylene (HDPE) for simulation use can be shown in Figure 8. It was found that thermal neutron had peak at energy of 0.024 eV.

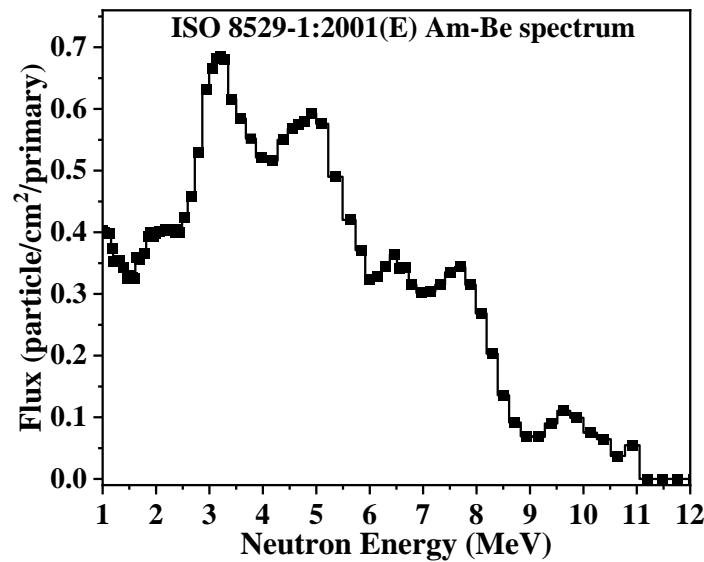


Figure 7 The Am-Be ISO spectrum.

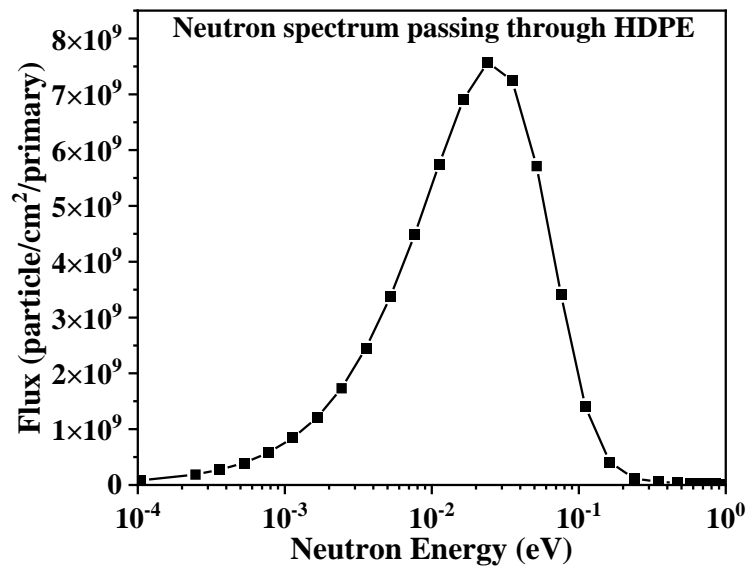


Figure 8 The thermal neutron spectrum passing through HDPE.

Figure 9 (a, b and c) shows the 2D graphs of the dose equivalent of thermal neutron. It was found that dose equivalent of thermal neutron did not pass through sample as exhibited in Figure 9 (a) better than the passed through the sample as exhibited in Figure 9 (b and c) and the dose equivalent of thermal neutron passed through the S1 more than S6 glass sample [32]. It was found that thermal neutrons were passing through S1 more than S6.

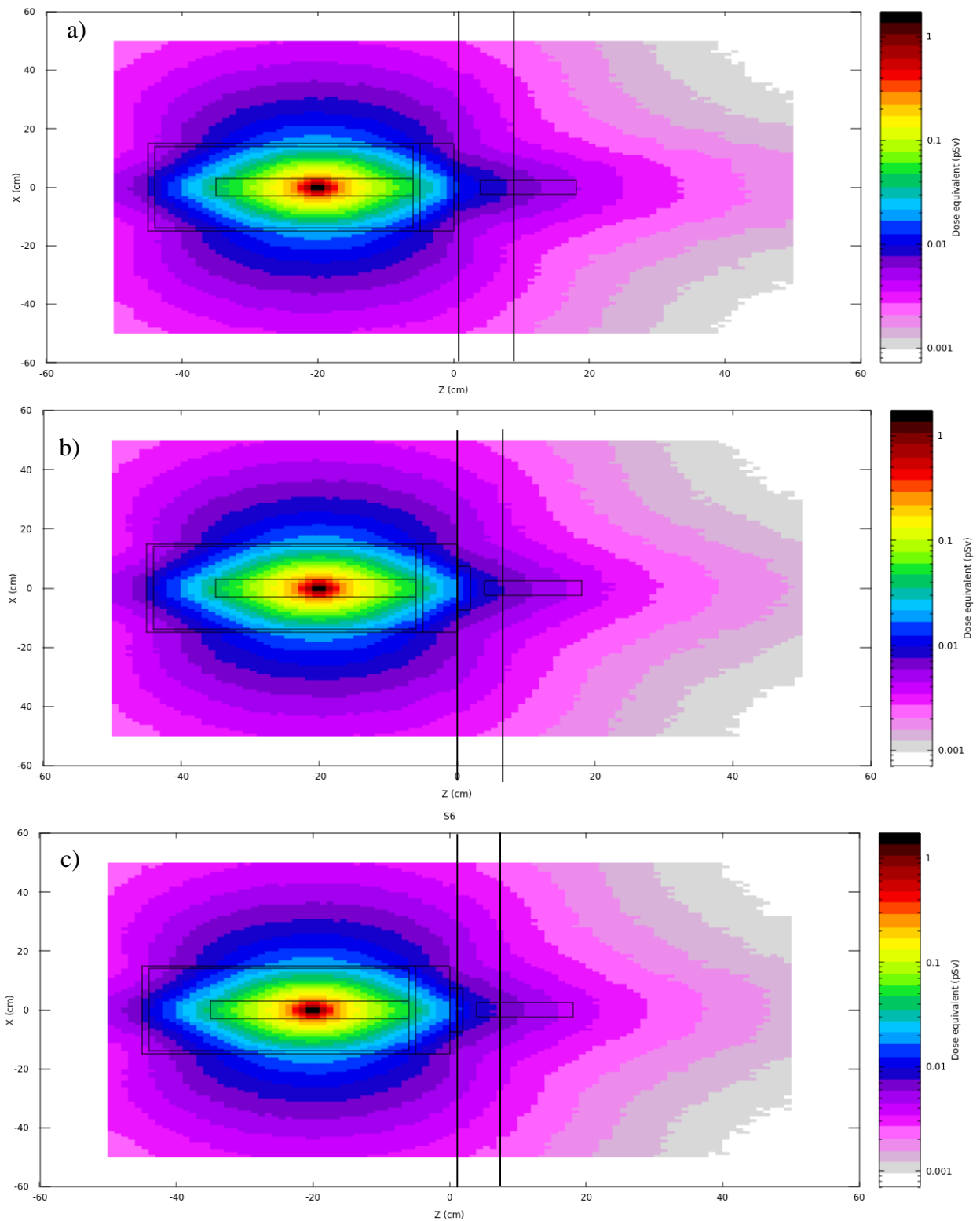


Figure 9 Typical dose equivalent of thermal neutron for a) I_0 , b) and c) passing through S1 and S6.

The relationship between the mass attenuation coefficient (μ_m) for thermal neutron and Gd_2O_3 content at thickness 2 cm is shown in Figure 10. The trend of μ_m value decreased with increasing Gd_2O_3 concentration. It indicated that thermal neutron shielding was being attenuated at high Gd_2O_3 content.

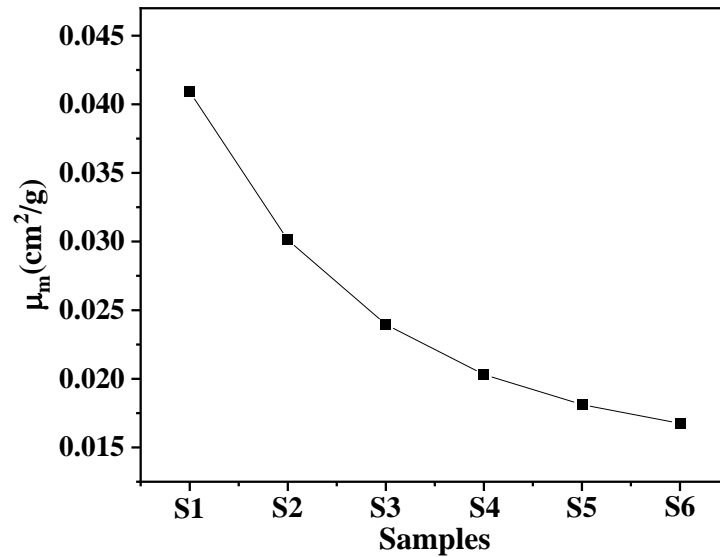


Figure 10 The mass attenuation coefficient of thermal neutron for glass series.

Fast neutrons shielding properties

The fast neutrons shielding properties for glass series were discussed on fast neutrons removal cross sections (Σ_R), HVL and the relaxation lengths as shown in Figures 11-13, respectively. It was found that Σ_R increased while HVL and the relaxation lengths decreased with increasing Gd_2O_3 content which increased the density of the glass series resulting in better fast neutrons shielding. These mean that the good fast neutrons shielding should be the heavy elements of high atomic number [27,33-36].

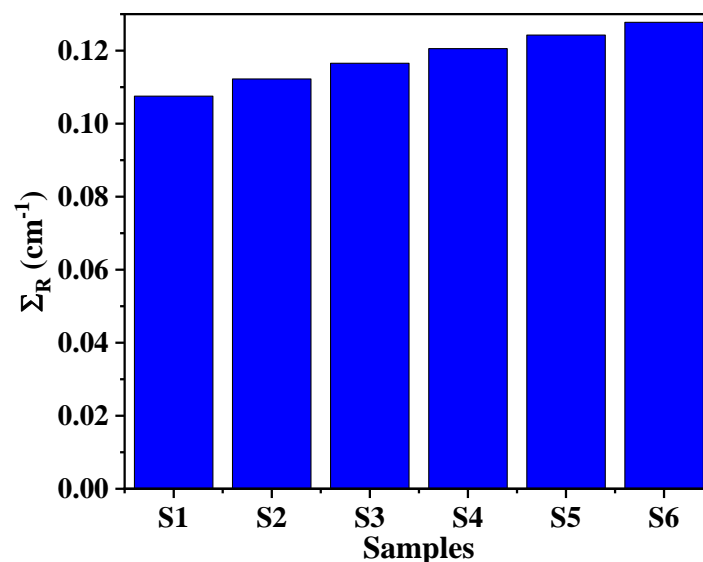


Figure 11 The removal cross section (Σ_R) for fast neutrons of glass series.

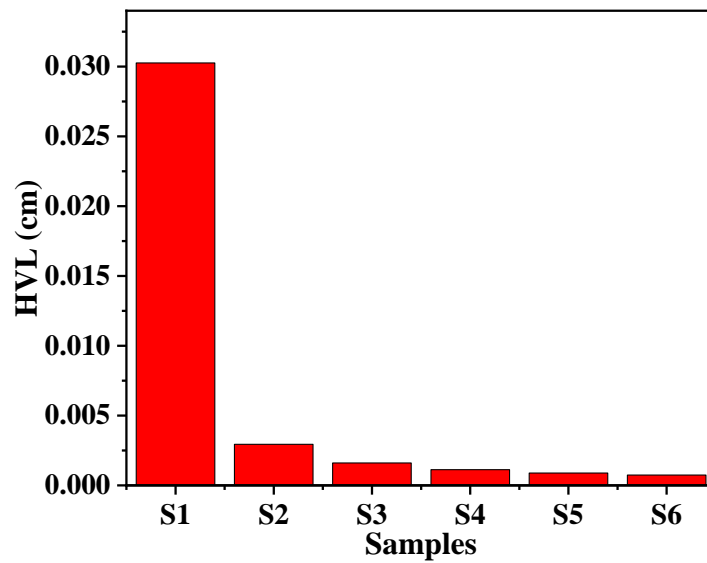


Figure 12 The HVL for fast neutrons of glass series.

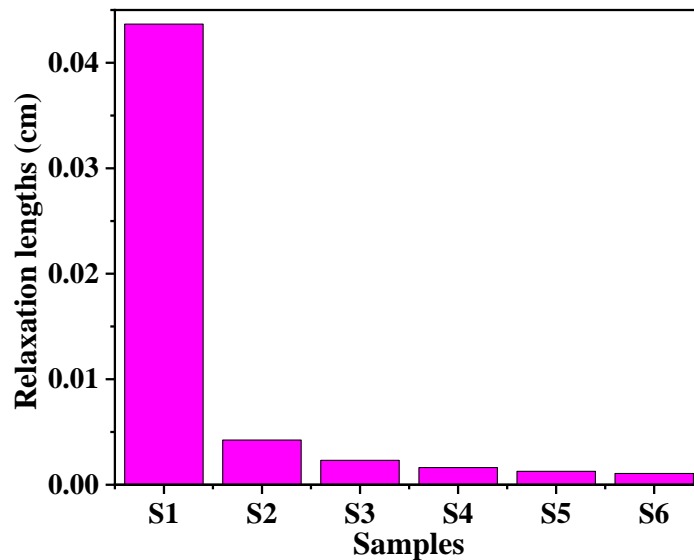


Figure 13 The relaxation lengths for fast neutrons of glass series.

Conclusions

From the available results of the study shielding properties for γ -ray, thermal and fast neutrons of the glass series. The results showed that the addition of Gd_2O_3 into $Gd_2O_3-SiO_2-Y_2O_3-CaO-B_2O_3$ glass series made the μ_m values increased while both HVL and MFP values decreased. These results indicated that the glass sample that contained 50 Gd_2O_3 mol% was the excellent shielding γ -ray. For the thermal and fast neutron shielding properties, it can be seen that the 50 Gd_2O_3 mol% glass sample showed the lowest value for thermal neutrons while the highest value for \sum_R and the lowest value for HVL and the relaxation lengths for fast neutrons were observed. These results indicated that this glass series which high Gd_2O_3 content possessed excellent γ -ray and fast neutrons shielding properties, while 20 $SiO_2-5Y_2O_3-10CaO-65B_2O_3$ glass sample possessed superb thermal neutrons shielding.

Acknowledgements

This research is financially supported by Thailand Science Research and Innovation (TSRI) National Science, Research and Innovation Fund (NSRF) (Fiscal Year 2022).

References

1. Graupner A, Eide DM, Instanes C, Andersen JM, Brede DA, Dertinger SD, et al. Gamma radiation at a human relevant low dose rate is genotoxic in mice. *Sci Rep.* 2016;6(1):32977.
2. Dogra M, Singh KJ, Kaur K. Investigation of γ ray shielding, structural and dissolution rate studies of alkali based bismuth borate glass systems with MoO_3 added. *Radiochim Acta.* 2019;107(3):261-9.
3. Al-Buriahi MS, Singh VP. Comparison of shielding properties of various marble concretes using GEANT4 simulation and experimental data. *J Aust Ceram Soc.* 2020;56:1127-33.
4. Issa SAM, Tekin HO, Erguzel TT, Susoy G. The effective contribution of PbO on nuclear shielding properties of $x\text{PbO}-(100-x)\text{P}_2\text{O}_5$ glass system: a broad range investigation. *Appl Phys A.* 2019;125:640.
5. Issa SAM, Mostafa AMA, Dong M, Singh VP, Tekin HO. Determining the gamma-ray parameters for $\text{BaO-ZnO-B}_2\text{O}_3$ glasses using MCNP5 code: a comparison study. *Radiat Eff Defects Solids.* 2018;173(5-6):510-25.
6. Sayyed MI, Lakshminarayana G. Structural, thermal, optical features and shielding parameters investigations of optical glasses for gamma radiation shielding and defense applications. *J Non Cryst Solids.* 2018;487:53-9.
7. Al-Hadeethi Y, Sayyed MI. Effect of Gd_2O_3 on the radiation shielding characteristics of $\text{Sb}_2\text{O}_3\text{-PbO-B}_2\text{O}_3\text{-Gd}_2\text{O}_3$ glass system. *Ceram Int.* 2020;46(9):13768-73.
8. Kaewjaeng S, Wantana N, Kothan S, Rajaramakrishna R, Kim HJ, Limsuwan P, et al. Effect of Gd_2O_3 on the radiation shielding, physical, optical and luminescence behaviors of $\text{Gd}_2\text{O}_3\text{-La}_2\text{O}_3\text{-ZnO-B}_2\text{O}_3\text{-Dy}_2\text{O}_3$ glasses. *Radiat Phys Chem.* 2021;185:109500.
9. Kaewnuam E, Wantana N, Tanusilp S, Kurosaki K, Limkitjaroenporn P, Kaewkhao J. The influence of Gd_2O_3 on shielding, thermal and luminescence properties of $\text{WO}_3\text{-Gd}_2\text{O}_3\text{-B}_2\text{O}_3$ glass for radiation shielding and detection material. *Radiat Phys Chem.* 2022;190:109805.
10. Al-Buriahi MS, TonguÇ B, Perişanoğlu U, Kavaz E. The impact of Gd_2O_3 on nuclear safety proficiencies of $\text{TeO}_2\text{-ZnO-Nb}_2\text{O}_5$ glasses: a GEANT4 Monte Carlo study. *Ceram Int.* 2020; 46(15):23347-56.
11. Intachai N, Wantana N, Kaewjaeng S, Chaiphaksa W, Cheewasukhanont W, Htun KT, et al. Effect of Gd_2O_3 on radiation shielding, physical and optical properties of sodium borosilicate glass system. *Radiat Phys Chem.* 2022;199:110361.
12. Saleh A. Comparative shielding features for X/Gamma-rays, fast and thermal neutrons of some gadolinium silicoborate glasses. *Prog Nucl Energy.* 2022;154:104482.

13. Ferrari A, Sala PR, Fasso A, Ranft J. FLUKA: a multi-particle transport code (Program version 2005), CERN Yellow Reports: Monographs; 2005.
14. Battistoni G, Boehlen T, Cerutti F, Chin PW, Esposito LS, Fassò A, et al. Overview of the FLUKA code. *Ann Nucl Energy*. 2015;82:10-8.
15. Ballarini F, Battistoni G, Brugger M, Campanella M, Carboni M, Cerutti F, et al. The physics of the FLUKA code: recent developments. *Adv Space Res*. 2007;40(9):1339-49.
16. Böhlen TT, Cerutti F, Chin MPW, Fassò A, Ferrari A, Ortega PG, et al. The FLUKA Code: developments and challenges for high energy and medical applications. *Nucl Data Sheets*. 2014;120:211-4.
17. Collamati F. *An intraoperative beta-probe for cancer surgery*. Switzerland: Springer International Publishing; 2016.
18. Medhat M., Demir N, Tarim UA, Gurler O. Calculation of gamma-ray mass attenuation coefficients of some Egyptian soil samples using Monte Carlo methods. *Radiat Eff Defects Solids*. 2014;169(8):706-14.
19. Demir N, Tarim UA, Popovici MA, Demirci ZN, Gurler O, Akkurt I. Investigation of mass attenuation coefficients of water, concrete and bakelite at different energies using the FLUKA Monte Carlo code. *J Radioanal Nucl Chem*. 2013;298:1303-7.
20. Kurtulus R, Kavas T, Mahmoud KA, Sayyed MI. A comprehensive examination of zinc borovanadate glass reinforced with Ag₂O in physical, optical, mechanical, and radiation shielding aspects. *Appl Phys A*. 2021;127(2):127-9.
21. Issa SAM, Ali AM, Susoy G, Tekin HO, Saddeek YB, Elsaman R, et al. Mechanical, physical and gamma ray shielding properties of xPbO-(50-x)-MoO₃-50V₂O₅ (25 ≤ x ≤ 45 mol %) glass system. *Ceram Int*. 2020;46(12):20251-63.
22. Issa SAM, Kumar A, Sayyed MI, Dong MG, Elmahroug Y. Mechanical and gamma-ray shielding properties of TeO₂-ZnO-NiO glasses. *Mater Chem Phys*. 2018;212:12-20.
23. Almuqrin AH, Sayyed MI, Kumar A, El-bashir BO, Akkurt I. Optical, mechanical properties and gamma ray shielding behavior of TeO₂-Bi₂O₃-PbO-MgO-B₂O₃ glasses using FLUKA simulation code. *Opt Mater*. 2021;113:110900.
24. Rammah YS, El-Agawany FI, Gamal A, Olarinoye IO, Ahmed EM, Abouhaswa AS. Responsibility of Bi₂O₃ content in photon, alpha, proton, fast and thermal neutron shielding capacity and elastic moduli of ZnO/B₂O₃/Bi₂O₃ glasses. *J Inorg Organomet Polym Mater*. 2021;31:3505-24.
25. Lakshminarayana G, Tekin HO, Dong MG, Al-Buriahi MS, Lee DE, Yoon J, et al. Comparative assessment of fast and thermal neutrons and gamma radiation protection qualities combined with mechanical factors of different borate-based glass systems. *Results Phys*. 2022;37:105527.
26. Lakshminarayana G, Issa SAM, Saddeek YB, Tekin HO, Al-Buriahi MS, Dong MG, et al. Analysis of physical and mechanical traits and nuclear radiation transmission aspects of Gallium (III) trioxide constituting Bi₂O₃-B₂O₃ glasses. *Results Phys*. 2021;30:104899.

27. Sahadath MH, Biswas R, Huq F, Mollah AS. Calculation of the neutron shielding properties of locally developed Ilmenite-Magnetite (I-M) concrete. *Radioprotection*. 2017;50(2):203-7.
28. Sayyed MI, Issa SAM, Tekin HO, Saddeek YB. Comparative study of gamma-ray shielding and elastic properties of BaO-Bi₂O₃-B₂O₃ and ZnO-Bi₂O₃-B₂O₃ glass systems. *Mater Chem Phys*. 2018;217:11-22.
29. Ersundu Mç, Ersundu AE, Gedikođlu N, Őakar E, Büyükyıldız M, Kurudirek M. Physical, mechanical and gamma-ray shielding properties of highly transparent ZnO-MoO₃-TeO₂ glasses. *J Non Cryst Solids*. 2019;524:119648.
30. Tiamduangtawan P, Wimolmala E, Meesat R, Saenboonruang K. Effects of Sm₂O₃ and Gd₂O₃ in poly(vinyl alcohol) hydrogels for potential use as self-healing thermal neutron shielding materials. *Radiat Phys Chem*. 2020;172:108818.
31. Basu P, Sarangapani R, Venkatraman B. Compact shielding design for 740 GBq ²⁴¹Am-Be neutron source transport container. *Radiat Phys Chem*. 2020;170:108670.
32. Ferrulli F, Silari M, Thomsen F, Zorloni G. A thermal neutron source for the CERN radiation Calibration Laboratory. *Appl Radiat Isot*. 2021;178:109977.
33. Perisanoglu U, El-Agawany FI, Kavaz E, Al-Buriahi M, Rammah YS. Surveying of Na₂O₃-BaO-PbO-Nb₂O₅-SiO₂-Al₂O₃ glass-ceramics system in terms of alpha, proton, neutron and gamma protection features by utilizing GEANT4 simulation codes. *Ceram Int*. 2020;46(3):3190.
34. Issa SAM, Rashad M, Zakaly HMM, Tekin HO, Abouhaswa AS. Nb₂O₅-Li₂O-Bi₂O₃-B₂O₃ novel glassy system: evaluation of optical, mechanical, and gamma shielding parameters. *J Mater Sci: Mater Electron*. 2020;31(24):1-18.
35. Sriwongsa K, Sirimongkolchaikul J, Sukrasorn C, Bussaparoek T, Kanunghet S, Phansuea T, et al. Radiation and fast neutron shielding properties of Nickel-Based superalloys: Inconel 600, 718 and 725 superalloys. *Integr Ferroelectr*. 2022;224:120-33.