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Determination of variation of compositions of (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics on protective characteristics in gamma radiation shielding

Abstract. The work is devoted to the study of the effect of variation of the ratio of oxides in the composition of (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics on shielding characteristics when employed as materials to mitigate the adverse effects of gamma radiation with different energy. The primary incentive behind these investigations is to discover novel protective shielding materials that lack lead or its oxide forms yet possess high shielding efficiency. Additionally, the aim is to identify the most promising compositions for potential practical applications in shielding. During the studies of shielding characteristics, it was observed that partial replacement of zinc oxide with bismuth oxide leads to a pronounced increase in shielding for low-energy gamma rays, for which, according to the data obtained, an increase in the concentration of bismuth oxide leads to an increase in the linear and mass coefficient by more than 0.8 – 1.2 times. An analysis of the influence of variations in the ratio of zinc and bismuth oxides in the composition of glass-like ceramics on the shielding characteristics revealed that the replacement of zinc oxide with bismuth oxide, leading to an elevation in density of the order of 10 – 15 %, leads to a steep rise in the shielding efficiency for low-energy gamma rays (more than 120 %), and to a 15 – 17 % growth in shielding efficiency in the case of gamma-quanta shielding for which the main interaction mechanisms are the Compton effect and the formation of electron – positron pairs.

Key words: shielding materials, ionizing radiation, glass-like ceramics, absorption efficiency, oxide materials, gamma radiation.

Introduction

In recent years, attention towards the advancement of new shielding materials has been directed towards research exploring technological solutions aimed at creating composite vitreous ceramics or amorphous glasses, which are based on oxide components combining heavy, rare earth and light elements [1-4]. Simultaneously, significant focus is dedicated to investigating variations in the compositions of these oxides. Altering these compositions enables the creation of numerous shielding materials with promising potential in the field of shielding [5,6]. These studies are based on the principle of obtaining the densest ceramics or glasses, comparable in shielding efficiency to traditional material in the form of lead, but at the same time possessing transparency (for medical use in cases where direct visualization of exposure to ionizing radiation is necessary), non-toxicity, high strength parameters (hardness and crack resistance), as well as low cost [7-9]. Also, much attention in such studies is paid to the search for a simple method for producing protective materials, the creation of which eliminates the need to use high-tech processes. In most cases, the method of producing such protective materials is considered to be solid-phase chemical synthesis combined with thermal sintering, the conditions of which are selected to create an amorphous structure of ceramics or initiate glass transition processes [11-14]. Also, the use of this synthesis method makes it possible to expand the range of materials obtained by varying the molar or weight ratios of the components used for the synthesis of samples, which opens up great prospects in this area of research.

The aim of this study is to assess the efficacy of using (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glassy ceramics as shielding materials against gamma and electron radiation, while also investigating the impact of composition variations on shielding characteristics. The interest in this research area stems mainly from the broadening scope of protective ceramic and glassy ceramic materials utilized for ionizing radiation shielding. Moreover, there is potential for their application as transparent materials for direct visualization or observation of ionizing radiation objects. The use of aluminum and bismuth oxides is due to the possibility of enhancing the resistance of the synthesized samples to external factors, including mechanical influences. Thus, in [15], it was demonstrated that the addition of aluminum and bismuth oxides to the ZnO–Al_2O_3–Bi_2O_3–B_2O_3 composition leads to an increase in the Young's modulus and, therefore, the crack resistance of glasses under external influences. The use of bismuth oxide is also due to the possibility of stabilizing and compacting glasses during their thermal sintering since this oxide is one of the stabilizers for glass transition processes. Also, the replacement of zinc oxide with bismuth oxide leads to an increase in the density of the synthesized samples, due to differences in the density of the oxides (p_{ZnO} – 5.61 g/cm^3, p_{Bi2O3} – 8.9 g/cm^3).

Materials and research methods

The objects of study were (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics obtained by solid-phase synthesis. The manufacturing method included mechanical grinding of the original ZnO, Al_2O_3, WO_3 and Bi_2O_3 oxides in a given molar ratio. The ratio of zinc oxide and bismuth oxide varied in the x range from 0.05 to 0.25 M, which allowed the production of glass-like ceramics with different element ratios. The combination of the method of mechanochemical solid-phase synthesis with thermal annealing of ground mixtures made it possible to obtain a series of samples that have different shielding characteristics. The choice of this method is based on the possibility of obtaining ceramics and glasses with a controlled ratio of elements, as well as those that are highly resistant to external mechanical influences.

The oxides ZnO, Al_2O_3, WO_3 and Bi_2O_3 purchased from Sigma Aldrich (Sigma, USA) were chosen as the starting components; the chemical purity of the powders under study was 99.95%. Mechanical grinding was carried out in a planetary mill PULVERISETTE 6 (Fritsch, Berlin, Germany) at a grinding speed of about 400 rpm for 1 hour. The grinding time and the grinding speed were chosen to avoid the cold welding effect of grinding powders under the action of mechanical deformation caused by the action of grinding bodies. Grinding was carried out in a ratio of 1 to 3 by the mass of ground powder and grinding bodies in the form of tungsten carbide balls with a diameter of 10 mm. Grinding was carried out in a glass made of tungsten carbide, the hardness of which eliminates the effect of contamination of the grinded powders with impurities. Grinding resulted in powders that were uniform in composition and grain size, which were later used in thermal sintering. After grinding, the resulting mixtures were annealed in a Nabertherm LE 4/11/R6 muffle furnace (Nabertherm, Lilienthal, Germany) at a temperature of 1500 °C for 5 hours at a heating rate of 20 °C/min. Annealing was carried out in zirconium dioxide crucibles capable of withstanding temperatures of about 2000 °C. The samples were cooled together with the furnace for 24 hours after reaching the annealing time. The choice of cooling time is determined by the need to comply with the technical rules for operating muffle furnaces, according to which access to the atmosphere can be allowed until the heating elements have completely cooled.

The choice of these synthesis conditions made it possible to achieve the initialization of the amorphization processes of the studied samples during the glass transition process, which was confirmed by X-ray phase analysis data, according to which no significant reflections characteristic of ordered structures were recorded in the X-ray diffraction patterns.

At the same time, the variation was carried out by partially replacing zinc oxide with bismuth oxide, the choice of which is due to the possibility of plasticization during glass formation in the case of thermal melting of the initial components, as well as increasing the shielding efficiency due to the larger atomic mass, while maintaining the optical transparency due to zinc oxide. The range of substitution concentrations was from 0.05 to 0.25 M. At the same time, the substitution was carried out by varying zinc oxides (its decrease) and bismuth oxide (its increase), with a constant ratio of the molar fractions of aluminum oxide and tungsten oxide. Previously, in [16], it was shown that thermal annealing in the temperature range from 500 to 1100 °C leads to the formation of multiphase ceramics, the formation of which occurs due to phase formation processes associated with thermodynamic reactions of the synthesis of complex oxides. Moreover, it was determined a priori, considering the data presented in the work, that thermal annealing at a temperature of 1500°C leads to the formation of amorphous glass-like ceramics with fairly high strength parameters (hardness of 1000 – 1100 HV). Figure 1 reveals the results of X-ray diffraction of the studied samples of glassy ceramics, reflecting the amorphous nature of the obtained samples after thermal annealing under specified conditions.

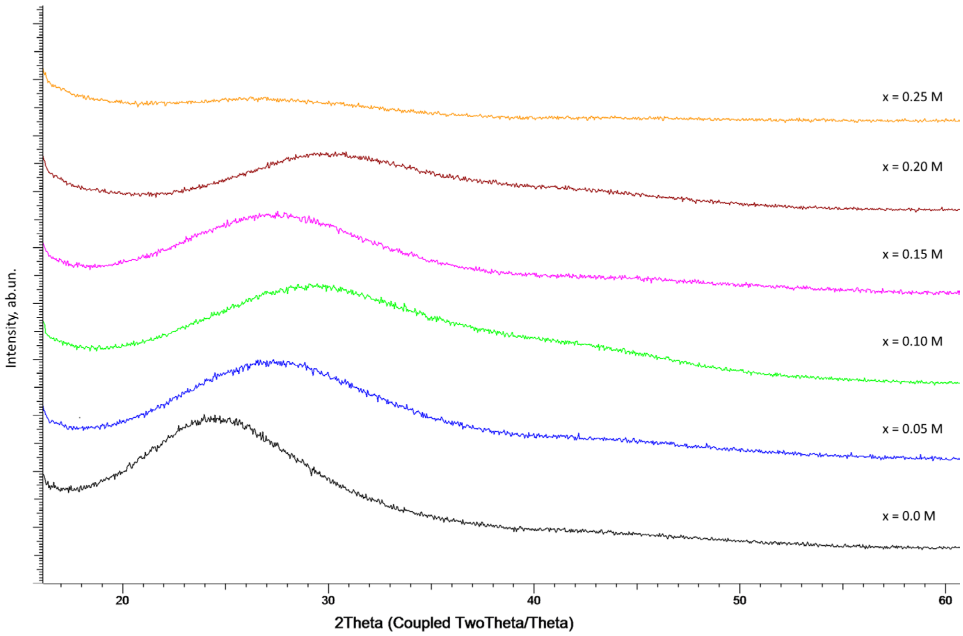


Figure 1 – X-ray diffraction results reflecting the amorphous nature of the studied samples of (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics

Determination of the shielding characteristics of the synthesized samples depending on variations in their composition was carried out by conducting serial experiments related to determining the intensity of gamma radiation before and after the use of a protective shield in the form of synthesized samples using three standard sources of ionizing radiation Co^{57} (130 keV), Cs^{137} (660 keV), Na^{22} (1270 keV). The determination of effectiveness was assessed by calculating the linear and mass attenuation coefficients (LAC and MAC), as well as the half-attenuation layer value (HVL), which is used to conduct a comparative analysis of the shielding effectiveness of different materials of different compositions. The shielding scheme was chosen in the form of a source placed in a lead container with a 10 mm hole, near which at a distance of 10 cm a protective shield made of synthesized glassy ceramics is placed, behind which a detector is placed to record the intensity of the transmitted ionizing radiation.

Simulation of the interaction processes of gamma radiation with composite ceramic compositions, varying in their molar composition, were simulated using the XCOM program code. A detailed description of the use of program code for modeling the processes of interaction of gamma radiation with materials is presented in works [17-19]. Figure 2 illustrates the variations in the MAC value for specific compositions, depending on the zinc and bismuth oxide ratio. The values, ranging from 0.001 to 10 MeV, encompass all three primary mechanisms of gamma radiation interaction with matter (Compton effect, photoelectric effect, and electron-positron pair formation). As evident from the provided data, altering the ratio of oxide components within the selected system does not result in discernible changes in the relationship between variations in the MAC value and gamma ray energy. The overall trend corresponds to the generally accepted theory of describing alterations in shielding efficiency. In the case of low energies of gamma rays (about 0.01 MeV), the shielding efficiency has maximum values, while an elevation in the gamma quanta energy results in an exponential decrease in the shielding efficiency, due to the dominance of the mechanisms of formation of electron-positron pairs, as well as secondary radiation in the form of gamma quanta of lower energies during the interaction of high-energy gamma quanta with the target structure.

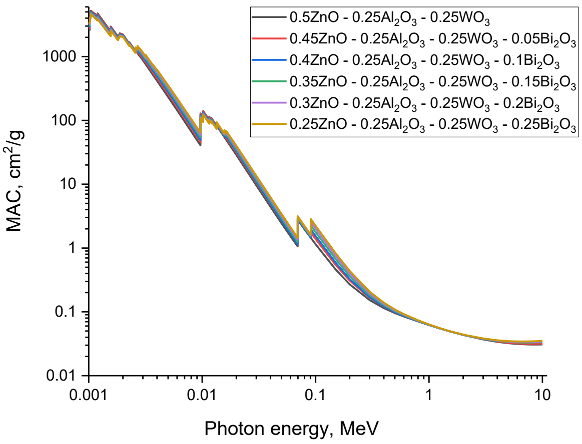
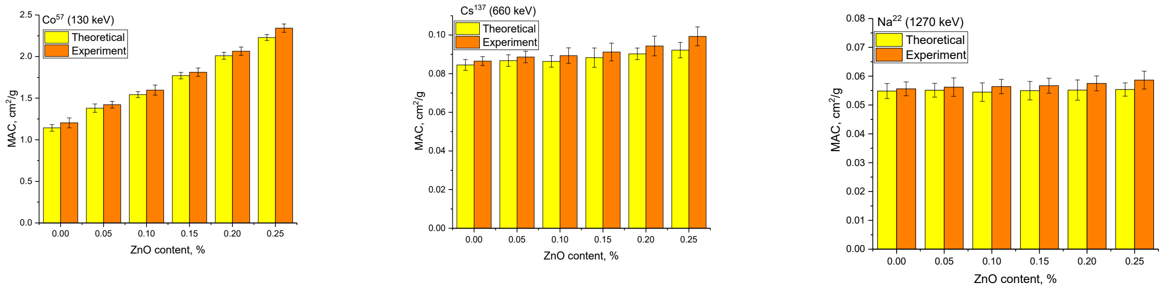


Figure 2 – Simulation results of the dependence of MAC on gamma radiation energy obtained using the XCOM code

It is worth to highlight that the partial replacement of zinc oxide with bismuth oxide causes changes in the shielding efficiency in the region of low energies of gamma rays (of the order of 0.3 – 0.6 MeV), for which the dominant interaction processes are photoelectric mechanisms, for which the dependence on the charge number of the composite material is most clearly visible.

Results and discussion

Figure 3 presents the results of a comparative analysis of MAC values obtained by modeling in the XCOM program code (values taken for specific gamma ray energies characteristic of those emitted by sources used in shielding experiments) and experimental values, obtained by shielding of gamma quanta emitted by various sources. According to the general assessment of the comparison of theoretical and experimental MAC values, there is a good agreement between the simulation results and experimental values, and the difference in values is no more than 3 – 5 %, which can be explained by several factors. Firstly, the higher MAC values obtained experimentally may be due to the fact that when modeling interaction processes, the objects under study were specified as a composite in the form of oxide compounds with different weight contributions. At the same time, according to the data of [16], during thermal sintering of these composites at temperatures above 900 °C, complex oxides are formed in the form of zinc tungstates ($ZnWO_4$) and bismuth (Bi_2WO_6), as well as spinel $ZnAl_2O_4$, the presence of which causes a change in density that is different from the density of composites consisting of oxides. In this case, at high annealing temperatures (1500 °C), according to X-ray diffraction data, the composition of the obtained samples is represented by amorphous structures, however, the possibility of the presence of fine grains in the form of complex tungstate compounds in the amorphous matrix cannot be excluded, since these compounds are thermally stable at fairly high temperatures. Secondly, the most pronounced differences are observed for samples in which the bismuth oxide content is on the order of 0.2 – 0.25 M, which indicates that changes in MAC values may also be due to differences in the charge number, the change of which is due to the high bismuth content in the composition.



a) b) c)

Figure 3 – Comparative analysis of MAC values calculated using the XCOM code and experimentally obtained values during shielding experiments

The alterations in the LAC value presented in Figure 4 depending on the energy of gamma quanta reflect the influence of variations in the ratio of components on the efficiency of attenuation of gamma radiation during shielding. The most pronounced changes are observed when shielding low-energy gamma quanta emitted by the Co^{57} source (130 keV). In this case, changing the ratio of the components of zinc and bismuth oxides leads to a more than twofold increase in the shielding efficiency, expressed by an increase in LAC from 6.2 cm^{-1} for samples 0.5ZnO – 0.25Al_2O_3 – 0.25WO_3 to 12.7 – 14.2 cm^{-1}, for samples in which the Bi_2O_3 concentration is 0.2 – 0.25 M. In this case, the shielding efficiency can be assessed at a fairly high level, taking into account the fact that the obtained values have a good correlation with the MAS data obtained using the simulation method. In the case of shielding of gamma rays with energies of 660 keV and 1270 keV, the differences in efficiency are reduced, however, the addition of bismuth oxide to the composition results in a rise in the shielding efficiency of the order of 20 – 25 % in comparison with the composition of 0.5ZnO – 0.25Al_2O_3 – 0.25WO_3 glass-like ceramics.

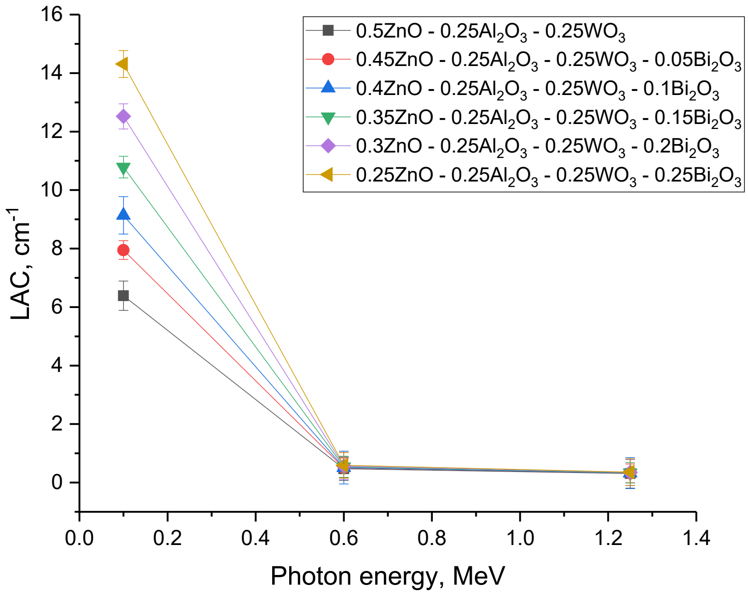


Figure 4 – Dependence of the LAC value for the studied (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics depending on the gamma quanta energy

Figure 5 demonstrates the results of a comparative analysis of the relationship between changes in the LAC value and the density of the samples under study, which is due to variations in the ratio of oxides in the composition of glass-like ceramics. The density of the samples, due to their X-ray amorphism, was determined by the Archimedes method, according to which it was found that partial replacement of zinc oxide with bismuth oxide results in compaction of glass-like ceramics, which is due to the fact that the density of bismuth oxide is higher than that of zinc oxide (data on the densities of zinc and bismuth oxides are given in the Introduction section when describing the formulation of the problem and the relevance of the study).

As can be seen from the data presented, the greatest contribution to the change in shielding efficiency is made by the change in the density of the samples under study on the shielding of low-energy gamma quanta with an energy of 130 keV. At the same time, a change in the density of the samples by 14.5 % (at a bismuth oxide concentration of 0.25 M) shielding efficiency, according to comparison of LAC values, is about 1.2 – 1.25 times in comparison with the results obtained for samples that do not contain bismuth oxide. In the case of gamma rays with energies of 600 keV and 1270 keV, the change in shielding efficiency is less pronounced depending on the density of the samples and is about 25 and 17 %, respectively, at a bismuth oxide concentration of 0.25 M.

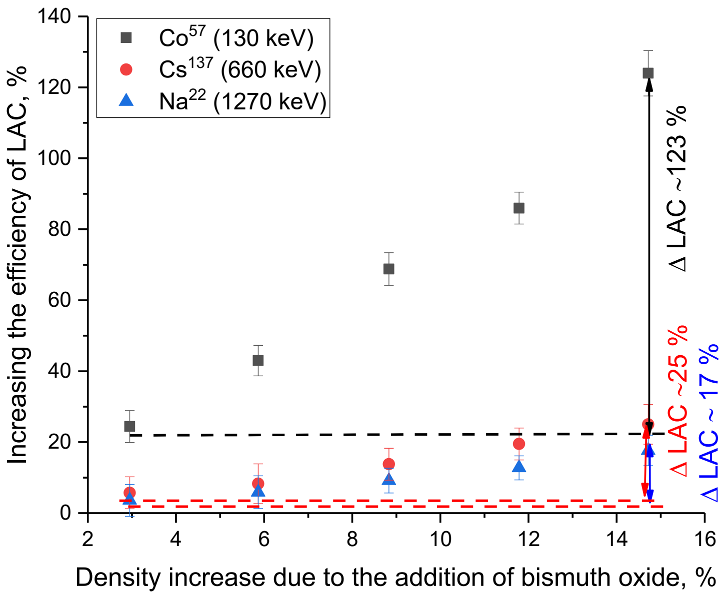


Figure 5 – Results of a comparative analysis of the dependence of LAC for synthesized ceramics depending on changes in density with increasing bismuth oxide content in the glass composition

A comparison of the shielding efficiency of synthesized samples with other types of samples, data on the shielding characteristics of which were taken from works [20 – 24], was carried out by comparing the HVL value obtained for gamma quanta with an energy of 660 keV. The choice of this energy of gamma quanta for comparison is due to the possibility of determining two types of processes of interaction of gamma quanta with materials (Compton effect and photoelectric effect), which are the most common interaction processes, with the exception of the processes of formation of electron – positron pairs, characteristic of high-energy gamma quanta with energies of more than 1.0 – 1.5 MeV. For comparison, Pb_3O_4-SiO_2-ZnO-WO_3 (LAC ~ 0.728 cm^{-1} to 0.856 cm^{-1} depending on WO_3 concentration) [20], Al_2O_3-PbO-B_2O_3-WO_3 glass [21], B_2O_3–$SrCO_3$–TeO_2–ZnO glass [22], as well as two types of commercial materials considered as shielding protective materials were chosen. The choice of ferrite and telluride glasses as commercial materials is due to their prospects for use as protective shielding materials, which are determined by their structural features, as well as several other unique properties, the combination of which allows us to consider them as one of the most promising materials for shielding. For example, in [23,24] it is shown that the combination of magnetic, structural, and antibacterial properties of ferrite nanoparticles and nanocrystals opens up great prospects for this class of materials when used as protective shields, which can also be used in the medical field, which uses sources of ionizing radiation. The results of the comparative analysis are presented in Figure 6.

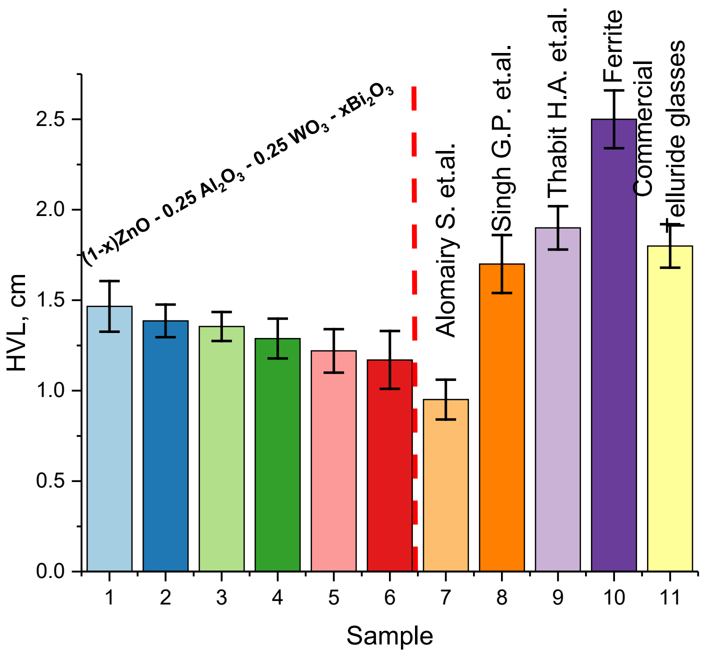


Figure 6 – Results of a comparative analysis of the HVL value of synthesized samples of (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics and commercial samples (HVL data taken for gamma quanta energy of 660 keV)

As can be seen from the presented data on changes in the HVL value depending on changes in the ratio of the components of zinc and bismuth oxides, an increase in the bismuth oxide content leads to a decrease in the HVL value, which indicates an increase in shielding efficiency, as well as the possibility of using less thick glass-like ceramics for protection, which makes it possible to reduce weight and overall dimensions. In comparison with commercial samples of ferrite and telluride glasses, the efficiency is more than 1.5 – 2 times, which indicates that the use of the proposed compositions of glass-like ceramics is more promising in comparison with these types of samples, as well as the compositions proposed in [21,22]. In comparison with samples of Pb_3O_4-SiO_2-ZnO-WO_3 glasses, the proposed compositions of glass-like ceramics are inferior in efficiency, however, the use of lead oxide in the proposed glass compositions from [20] imposes certain restrictions associated with the toxicity of lead (part of the oxide), which narrows the scope of application of these glasses, and high efficiency indicators are due to the presence of heavy elements in the form of lead and bismuth.

Conclusion

During comparative analysis of the MAC values obtained using the XCOM program code and the experimentally obtained values during shielding, it was found that the difference between the theoretical and experimental values is no more than 3 – 5 %, which indicates good convergence, as well as the possibility of using computational codes to simulate the shielding of complex ceramic compositions. At the same time, differences in values may be due to the effects of structural features that arise during the synthesis of (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics, leading to denser ceramics, with the possible formation of inclusions in the form of complex oxides, observed at lower sintering temperatures of these compositions.

A comparative analysis of shielding efficiency by comparing HVL values with the results of other studies, as well as with data obtained for commercial samples, showed that the use of (1-x)ZnO-0.25Al_2O_3-0.25WO_3-xBi_2O_3 glass-like ceramics is quite effective in comparison with the majority of the considered shielding material compositions having similar components, as well as with commercial samples. Moreover, a rise in the concentration of bismuth oxide in the composition results in a decline in the HVL value from 1.466 cm for samples not containing bismuth oxide to 1.7 – 1.2 cm for bismuth oxide concentrations equal to 0.2 – 0.25 M. Compared to commercially available materials, ferrite and telluride glasses, the shielding efficiency is more than 1.5 – 2 times.

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