NanoWorld Journal

https://doi.org/10.17756/nwj.2023-s2-061

Research Article

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Investigation of Structural and Radiation Shielding Parameters for Lead Pyrophosphate Glasses

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Received: July 25, 2023
Accepted: September 26, 2023
Published: September 29, 2023

Citation: Abadi S, Hana NA, Sayyed MI, Benkhouja K, Haddad M, et al. 2023. Investigation of Structural and Radiation Shielding Parameters for Lead Pyrophosphate Glasses. *NanoWorld J* 9(S2): S362-S366.

Published by United Scientific Group

Abstract

We have elaborated the Na_2O-(1-x)PbO-xCuO-P_2O_5, with (0 ≤ x ≤ 1), using a conventional melt quenching technique. The object of the present investigation is an analysis of the relationship between the structure and gamma ray shielding properties of the studied compounds. The Raman spectroscopic study showed that the substitution of Pb^{2+} by the Cu^{2+} ions leads to a large modification in the structure of $(P_2O_7)^4$ groups. Additionnaly, we have investigated the gamma ray shielding properties of elaborated glasses. Using the XCOM software, mass attenuation coefficients (μ/ρ), half value layers (HVL) and effective atomic numbers (Z_{eff}) were calculated. The values of μ/ρ and Z_{eff} were found to decrease with the increase in CuO content. The results show that Cu^{2+} ions affect the structure and gamma ray shielding properties of the glasses systems Na_2O-(1-x)PbO-xCuO-P_2O_5.

Keywords

Phosphate glasses, Pyrophosphate, Raman, Shielding properties

Introduction

The structure of the vitreous P_2O_5 compound was formed by a 3D network of corner sharing PO_4 tetrahedra. The hygroscopic property of this material reduced the domain of its use. The addition of the oxide to the vitreous network of P_2O_5 produces more stable materials. The xCuO-(1-x) P_2O_5 system, studied by Raman spectroscopy, showed that the CuO depolymerises the ultraphosphates into metaphosphates, polyphosphates and pyrophosphates while increasing the covalence character of the bond P-O, leading to the reticulation of the glass network via the creation of Cu-O-P cross-linkages [1]. The addition of CuO to 0.5ZnO-0.5 P_2O_5 products materials used against the gamma irradiation [2]. The no toxic character of Cu^{2+} ions, its paramagnetic property and its smaller ionic radius (0.73 A°) than that of Pb^{2+} (1.19 A°) are some reasons for the substitution of Pb^{2+} ions by Cu^{2+} ions in the Na_2O-(1-x)PbO-xCuO-P_2O_5 system glasses (0 ≤ x ≤ 1). The substitution of the PbO by the CuO enhances the resistance of the material against the devitrification, modified the local symmetry of the site and the nature of the exchange interactions between paramagnetic ions and decreases the transition temperature between two magnetic states to lower value [3]. The objective pursered by continuing the study the effect of the substitution of the PbO ions by the CuO ones on the structure of these materials and shielding properties against the radiation.

Materials and Method

Synthesis

The glasses Na_2O-(1-x)PbO-xCuO-P_2O_5 were elaborated using the starting materials Na_2CO_3, PbO, CuO, and $(NH_4)_2HPO_4$. The differents mixtures corresponding to the selected composition were homogeinized and ground in an agate mortar. The mixtures were placed in a platinum crucible and heated successively at 200 °C for 3 h andat 400 °C during 12 h to allow the decomposition of the sodium carbonate and diammonium hydrogen phosphate. All used materials are nanoscale materials.

The resulting materials were ground again and heated progressively to reach their meltingpoint. Finally, the melted materials were quenched on a metal platepreheated at 200 °C. The resulting glasses are blue and transparent and become darker as the copper content increases. The X-ray diffractograms of the elaborated Na_2O-(1-x)PbO-xCuO-P_2O_5 glasses are shown in figure 1. No peaks of crystallization were observed, which indicates the amorphous structure of all the elaborated materials.

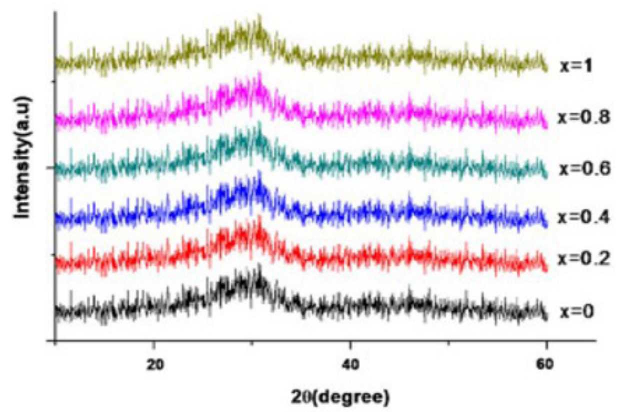

Figure 1: The XRD patterns of the Na_2O-(1-x)PbO-xCuO-P_2O_5 glass system [3].

Raman spectroscopy

Raman spectra were recorded using a Renishaw micro-Raman spectrometer (RM1000) equipped with a CCD detector, a 1800 g.mm^{-1} grating and an external Leica DMLM confocal microscope. The excitation source is a He-Ne laser (19 mW) operating at 632.8 nm.

XCOM program

XCOM is a computer program used to calculate the linear attenuation coefficient (μ) of X- and gamma-rays and the interaction of cross-sections of different types of materials in the energy range 1 kGeV - 100 GeV.

Results and Discussion

Spectroscopic study

The Raman spectra of Na_2O-(1-x)PbO-xCuO-P_2O_5 glass system (Figure 2) show that the replacement of Pb^{2+} ions by Cu^{2+} caused the modification in the structure of $(P_2O_7)^{4-}$ units.

The intensities of the all bands decrease and some of them

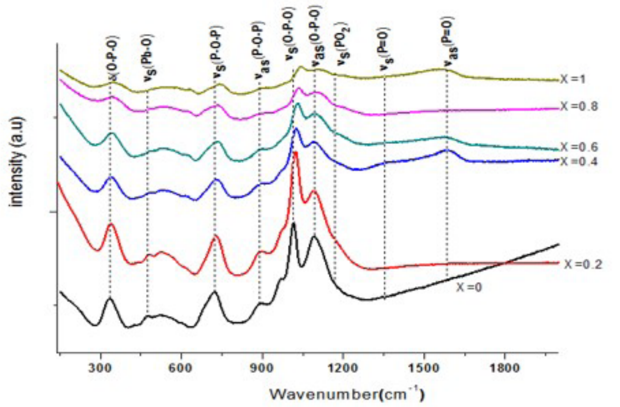

Figure 2: The Raman spectra of the Na_2O-(1-x)PbO-xCuO-P_2O_5 glass system.

move to the higher values of the wave number. The decrease in the intensities of the bands can be explained by an increase in the covalence character of O-P-O and P-O-P bonds. On the other hand by the polymerization of the pyrophosphate units to form the metaphosphate [4]. The shift of the absorption bands (P-O-P and O-P-O) to the higher frequencies indicates that the increase of CuO content in the glass leads to change the P-O-P and O-P-O bond's angles.

Shielding properties

The mass attenuation coefficient (μ/ρ) is considering one of the most essential quantities to investigate the interaction of photons with a specific glass sample. The μ/ρ values for the prepared Na_2O-(1-x)PbO-xCuO-P_2O_5 glasses can be calculated according to the Bragg law as in the following relation [5]:

$$(\mu/\rho)_{glass} = \sum_i w_i (\mu/\rho)_i \quad (1)$$

Where w_i is the fraction by weight of the element while the subscript i denotes the elements used in the glass preparation (i.e., Na, Pb, Cu, P, and O). The $(\mu/\rho)_i$ required for the analysis of the radiation shielding properties of the prepared glasses has been obtained using XCOM program [6]. The μ/ρ values for the Na_2O-(1-x)PbO-xCuO-P_2O_5 glasses as a function of photon energy is plotted as shown in figure 3.

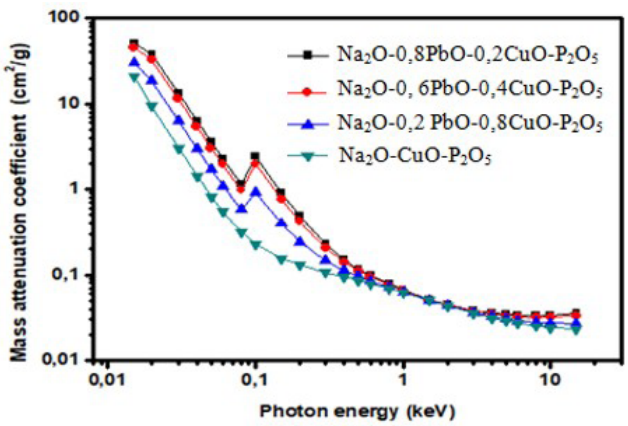

Figure 3: The mass attenuation coefficients (cm²/g) for the prepared Na_2O-(1-x)PbO-xCuO-P_2O_5 (0 ≤ x ≤ 1)glasses in the energy region of 0.015 - 15 MeV.

The present μ/ρ results follow the same trends as found by Dong et al. [7], who investigated the μ/ρ and some other related parameters of lithium zinc bismuth borate glasses in the energy range 0.015 - 10 MeV. On the other hand, the tendency of the μ/ρ curve presented in figure 3 overlaps with the results reported by Singh et al. [8], who studied the radiation shielding properties of different borate and silicate heavy metal oxide glasses.A general observation of figure 3 shows that the μ/ρ values depend on the composition of the glasses in addition to the energy of the photon. The variation of μ/ρ with the energy for all prepared glasses is nearly the same. The μ/ρ values seem to be relatively high in the low energy zone (in order of 20.8 - 52 cm^2/g at 0.015 MeV). They decrease sharply with further increase in the energy, and tend to be constant when the photon energy reaches about 3 MeV. This behavior of μ/ρ can be explained according to important radiation physics concepts. In brief, photons can interact with certain material depending on the atomic number of this material as well as the energy of the photon in three mechanisms: photoelectric absorption effect, compton scattering, and pair production [9]. In the low energy zone, the photoelectric absorption is the dominant process and the μ/ρ values vary with the photon energy as $1/E^{3.5}$ as well as with the atomic number as Z^4 in this process. In the moderate energy zone, compton scattering is the dominant mechanism of interaction for photons. The cross-section of compton scattering depends on the photon energy and atomic number as E^{-1} and Z, respectively. Therefore, one can easily observe that all the prepared glasses have almost the same μ/ρ values in this zone (i.e. for $0.8 < E < 3$ MeV).In the last energy zone ($E > 3$ MeV), the behavior of μ/ρ can be explained according to the predominance of pair production in this higher energy region [10].

Besides, discontinuities in μ/ρ values can be observed for the prepared glasses (Except for x = 1) at 0.08 MeV due to the Kedge absorption of Pb. The absence of the discontinuities in μ/ρ values for x = 1 sample (Pb-free) confirms this explanation.

From figure 3 there is a decreasing order of μ/ρ from x = 0.2 to x = 1. Na_2O-0,8PbO-0;2CuO-P_2O_5 glass sample has the highest μ/ρ while Na_2O-CuO-P_2O_5 sample has the lowest. This order could be attributed to varying weight percent of Pb (Pb is heavier than Cu) which was decreasing in order of x = 0.2, 0.4, 0.8, and 1. In addition, Pb has a higher effective atomic cross section than Cu. Since the Na_2O-0,8PbO-0;2CuO-P_2O_5 sample possesses the highest values of the μ/ρ, then we can conclude that this sample can attenuate more photons than the rest of the samples, thus has superior shielding properties.

The effective atomic number (Z_{eff}) is another important parameter used to evaluate the radiation shielding properties for the glass samples. This parameter represents the fraction of total number of electrons in a glass sample (or generally any medium) participates in photon - atom interaction number [11]. Hine et al. [12] has reported that the Z_{eff} for any material cannot be represented in a single number. Recently, this parameter has been widely used in many practical applications in different areas like radiation dosimeter, radiation protection in nuclear facilities, etc. [5].

For the Na_2O-(1-x)PbO-xCuO-P_2O_5 glass system, the following equation was used to calculate the Z_{eff}:

$$Z_{eff} = \frac{\sum_i f_i A_i \left(\frac{\mu}{\rho}\right)_i}{\sum_j f_j \frac{A_j}{Z_j} \left(\frac{\mu}{\rho}\right)_j} \quad (2)$$

Where f_i is the fractional abundance of the element i relative to the number of atoms, A_i is the atomic weight, and Z_i is the atomic number.

By using the calculated μ/ρ values and with the help of Equation 2, we computed the Z_{eff} for the prepared glasses and the results were plotted as a function of energy as shown in figure 4.

Apparently, the Z_{eff} has the maximum values in the low energy region (photoelectric region), while in the compton scattering region (Moderate energy region) the variation of Z_{eff} is insignificant and the Z_{eff} reaches minimum values in this zone. As can be seen from figure 4, a slight increase in Z_{eff} values occurs for high photon energies (Where the pair production is dominant). Additionally, the Z_{eff} for Na_2O-(1-x) PbO-xCuO-P_2O_5 glass samples were found in the range 16.39 - 41.99, 15.23 - 37.99, 12.68 - 20.45, and 11.44 - 16.27 for x = 0.2, 0.4, 0.8, and 1, respectively. Moreover, Na_2O-0,8PbO-0,2CuO-P_2O_5 (The sample that has the maximum amount of Pb) possesses the highest Z_{eff} values at the energy region under investigation. This shows that this glass sample is the superior gamma radiation attenuator among the prepared glasses.

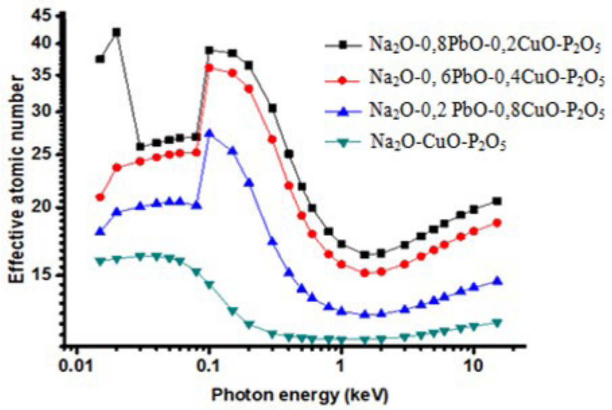

Figure 4: The effective atomic number for the prepared Na_2O-(1-x)PbO-xCuO-P_2O_5 ($0 \le x \le 1$)glasses in theenergy region of 0.015 - 15 MeV.

The half value layer is a quantity that characterizes the interaction between photons with the glass sample. It gives a good indication about shielding efficiency against radiation. It'sdefined as the thickness of a sample at which the transmitted intensity of the photon is exactly 50% the incident photon intensity [13]. The following relation was used to evaluate the HVL for the prepared glasses:

$$HVL = \frac{0.693}{\mu} \quad (3)$$

Where μ is the linear attenuation coefficient (Equal to μ/ρ multiply by the density of the sample).

The HVL as a function of photon energy for the prepared Na_2O-(1-x)PbO-xCuO-P_2O_5 (0 ≤ x ≤ 1)glasses is shown in figure 5.

Obviously, the HVL values for all glasses increase as the energy increases, and this trend is expected based on the μ/ρ in figure 3. Since the HVL related to the μ/ρ, the trend of HVL seen in figure 5 can be explained according to the three main interaction mechanisms for gamma ray in matter. It is well known that the longer HVL indicates more distance that the photon moves inside the glass sample. Therefore, shorter HVL values for certain glass samples are designed to get better radiation shielding materials. Perusal of the data presented in figure 5 shows that HVL decreases with increase in Pb content. So, we can see that Na_2O-0,8PbO-0,2CuO-P_2O_5 glass sample has the lowest HVL. This result confirms our assessment in the previous paragraph that this sample has the best radiation shielding performance among the prepared glasses in this work.

Mean free path (MFP) is a quantity that plays a crucial role in estimating the exponential attenuation of gamma radiation. The MFP is equal to the reciprocal of the linear attenuation coefficient (i.e., MFP = 1/μ) [14]. The shorter MFP for any glass sample means more photons interaction with this glass ample thus giving shorter distance between two interactions, hence possesses the better shielding properties. The MFP values for the prepared glasses as a function of photon energy are exhibited in figure 6.

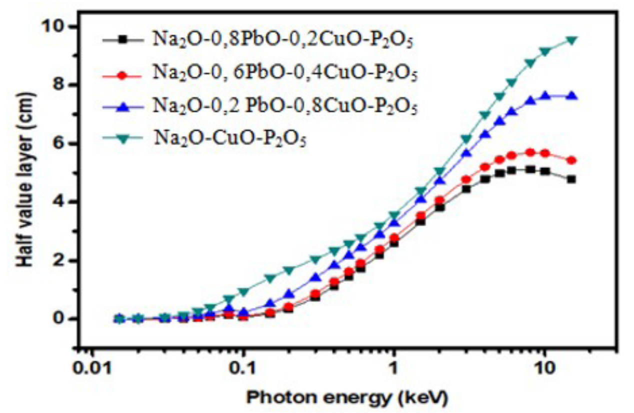

Figure 5: The half value layer (cm) for the prepared Na_2O-(1-x)PbO-xCuO-P_2O_5 (0 ≤ x ≤ 1)glasses in the energy region of 0.015 - 15 MeV.

Clearly, the variation of MFP with the photon energy is in similar trend with the HVL trend. From figure 6, there is an observable decreasing order in the MFP from sample Na_2O-CuO-P_2O_5 to sample Na_2O-0,8PbO-0,2CuO-P_2O_5. This emphasizes the fact that Na_2O-0,8PbO-0,2CuO-P_2O_5 showed the best glass sample useful in shielding photons at these energy range because it has the lowest HVL and MFP.

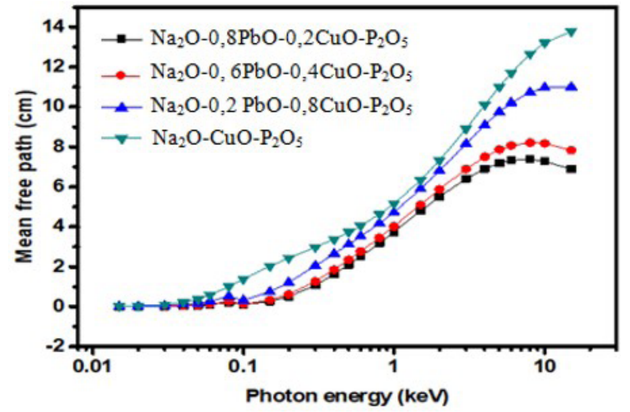

Figure 6: The mean free path (cm) for the prepared Na_2O-(1-x)PbO-xCuO-P_2O_5 (0 ≤ x ≤ 1)glasses in the energy region of 0.015 - 15 MeV.

Conclusion

The Raman spectra show that the structural of the phosphate groups is influenced by the substitution of the PbO by the CuO. The radiation shielding results revealed that Na_2O-0,8PbO-0,2CuO-P_2O_5 glass sample has the highest values of the μ/ρ and Z_{eff}, thus this sample can attenuate more photons than the rest of the samples, and therefore has superior shielding properties. This result confirms the structural change detected by Raman spectroscopic. These results make us think to study the application of these materials in the magnetic and electrical fields.

Acknowledgements

None.

Conflict of Interest

None.

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