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Determination of the Shielding Properties for Propose Material using In-Situ Object Calibration Software (ISOCS)

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Abstract
In this study, gamma ray shielding properties such asmass attenuation coefficient (μ_m), half value layer (HVL), tenth value layer (TVL) and mean free path (MFP) for glasses composition $xTiO_2$–$20Na_2B_4O_7$–$30PbO$–$20SiO_2$–(30-x) Bi_2O_3 ($0\leq x\leq 30$ mol%), are prepared by melt quenching technique. The proposed materials have been estimated at three different gamma- ray energies (661.7 Kev from ^{137}Cs, 1173 and 1332 Kev from ^{60}Co)by using In-Situ Object Calibration Software (ISOCS), for serving as competitive candidates for radiation shielding. This method provides a user interface with a procedure to describe the measurement geometry, physical parameters, and computational algorithms to quantify radioactivity in samples.Therefore,in many situations, we found in this study ISOCS calculations are preferred because of its cost in which includes money, effort, and time is lower than the experimental method. Also, it is found that ISOCS calculations are a very good method for the calculation of the previous gamma ray shielding properties at different photon energies.

Keyword: *Titanite oxide, (ISCOS), Heavy metal oxide glasses, γ-ray attenuation, Massattenuationcoefficient, Thickness.*

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I. Introduction

There is no Non Destructive Assay (NDA) technique which is capable to verify as wide range of Nuclear Materials (NMs) types with different matrices', chemical and physical forms, isotopic compositions and container shapes as ISOCS technique. Gamma ray sources and radioactive materials in several sectors including nuclear power plants, nuclear reactors, nuclear medicine and industry have harmful effects on humans and it is essential to provide shield against gamma radiations. Photon interactions with matter are very different from those of charged particles, when X-rays or Gamma rays interact with matter, some are absorbed, some pass through without interactions, and some are scattered as low energy photons. The attenuation of Gamma ray beam by an absorber material is usually characterized as occurring under "Good Geometry" conditions where every photon that interacts is either absorbed or scattered out of the primary beam such that those reach the receptor have kept all of their original energy [1,2].

Obviously, the consolidation of Bi_2O_3 in various glass systems had drown observable speculations. These glass systems are characterized by the small value of the glass transitions temperatures, the high third – order nonlinearities and the special optical transparencyinfrared. Recently borate (B_2O_3) glasses have been focused of interest due to their glasses forming ability, structure, and variety of application [3]. Borate glasses possess scientific interest because of the occurrence of boron anomaly, and are incorporated into various glass system as a flux material to attain materials of high technological application [3]. It is well known that addition of heavy metal oxides, (HMO), especially to borate- based glass have spread wide applications in the field of glass ceramics, layers for optical and electronic devices, thermal and mechanical sensors, reflecting window, etc [4].

Now a day heavy metal glasses are proving to be conventional shielding material like concrete [5]. The commonly used heavy metal in glasses is lead. But due to its toxic nature its use in heavy metal glasses is discouraged. Borate and silicate are the most commonly used glass formers because of their easy availability and low cost. Also, these bismuth bases glasses have considerable technological applications due to their density, high refractive index, of these glasses make them of great importance on many technical applications and low melting point [6].

On the other hand, titanium has gained importance recently for its broad range of applications in the biomedical field and in the glass and glass- ceramics. TiO_2 is one of the first oxides to be investigated for the antitumor properties and hence it is widely used as a biomaterial for several dental and orthopedic clinical purpose. It is accepted that TiO_2 can be inserted as dopants to act as a good nucleating agent, so its consolidation in a glass system is valuable in some specified glasses for electrical and optical uses. The electronic configuration of titanium ions arises out of the empty orunfilled d- shell of the ions in glasses that resulted in the existence of trivalent (Ti^{3+}) and tetravalent (Ti^{4+}) valence states. Accordingly, Alkali Borate and alkali silicate glasses favor the colorless high tetravalent (Ti^{4+}) ions. In spite of the bismuth borate glasses are non-toxic to environment and can provide alternate to lead free shielding materials. Also,these have advantage to conventional shielding materials in terms of their better shielding properties. These glasses are transparent to visible light and have low melting points. Further high density values and high refractive index of these glasses make them of great importance for many technical applications. However, from the literature survey, there are numerous articles concerning heavy metal oxides (HMO) included in the network of borosilicate glasses and to the best of our knowledge, few interested articles with titanium additions in this complicated network presented [7].

One set of composite material that has attracted attention in theradiation shielding industry is heavy metal oxide (HMO) glasses. Heavymetal oxide glasses are used in gamma-ray radiation shielding because of their high effective atomic number and high densities when compared to other glasses. Among HMO glasses, bismuth-borate glasses areparticularly important because they are a class of low melting pointlead-free glasses [8]. The low melting point glasses are expected to playa major role in the design of materials to mitigate radiation skyshine orsimilar radiation leakage due to limitations and defects in the radiationshield [9,10]

Gamma-rays are attenuated by processes which are functions of atomic number and mass (that is they all involve interactions near the nucleus or interactions with the electrons around the nucleus). Therefore, gamma radiation is best absorbed by atoms with heavy nuclei; the heavier the nucleus, the better the absorption [11]. Nowadays, some research work has been done with different types of glasses as anew gamma-ray shielding material. Gamma-ray shielding properties of various glasses have been studied theoretically and experimentally at different energies in literature [12-20,21]. Examples are bismuth-borated glasses [14], ZnO-PbO-B_2O_3 glasses [13], CaO-SrO-B_2O_3 glasses [21], PbO-B_2O_3 glass system, Bi_2O_3-PbO-B_2O_3 glass systems [13], and PbO-BaOB_2O_3 glass system [20], etc. It has been reported that compared with some standard radiation shielding concretes, these glasses have shown better performance in terms of their volume required for shield design with added advantage of being transparent to visible light [20].

TheIn-Situ Object Calibration Software(ISOCS) [22-25] software brings the possibility to establish absolute efficiency curve for desired energy range based on numerical simulation with the use of known or estimated geometry and chemical composition of measured items. Also, the software provides variety of geometry which covers wide range of possible item shapes such as cylinders, pipes, boxes as well as more complex geometries.Venkataraman et al [24] have been determined the full energy peak efficiencies of a germanium detector in the 45 keV - 7 MeV energy range for practically any source matrix and geometry. The mathematical techniques used in ISOCS and Laboratory Source Less Calibration Software (Lab SOCS) have undergone significant improvements and enhancements. They concluded that results are presented highlighting the improved performance achieved using the methodology [24].

Such as ISOCS make use of knowledge of the measurement configuration to establish calibration parameters. Such measurements approach has been used for a variety of purposes at Oak Ridge National Laboratory (ORNL) including LLW/ transuranic (TRU) sorting of a wide variety of radionuclides and waste streams in 55 gallons and over pack drums. A model- based calibration approach with variable geometry is now being developed for ORNL TRU waste [25,26].

The ISOCS modeling software allows performing absolute efficiency calibration for items of arbitrary containershape and wall material, matrix chemical composition, material full-height, uranium or plutonium weight fraction insidethe matrix and even nuclear material/matrix with nonhomogeneous distribution. Furthermore, in a number of cases,some key parameters such as matrix density and U/Pu weightfraction can be determined along with analysis of nuclearmaterial mass and isotopic composition. These capabilitiesprovide a verification solution suitable for a majority of caseswhere the quantitative and isotopic analysis should be performed [27]. Model-based methods have been used to calibrate nondestructive systems to characterize wastes contaminated withplutonium, uranium, and other radioactive isotopes.

The Objective of this work is to characterize the role of TiO_2 in HMO – alkali Borosilicate glasses $Na_2B_4O_7$-PbO-SiO_2-$Bi_2O_3$to estimate the attenuation properties by using In-Situ Object Calibration Software (ISOCS) Techniqueto quantify radioactivity in the samples under study.

II. Experimental Procedures

In order to prepare glass samples with chemical formula xTiO_2- 20 $Na2B_4O_7$-30 PbO -20 SiO_2-(30-x) Bi_2O_3 (0≤ x ≤ 30 mol%), convenient amounts of analytically pure grade chemicals of PbO, TiO_2, SiO_2, Bi_2O_3 and $Na_2B_4O_7.10H_2O$ were completely unsettled in an agate mortar. The mixture was melted in a ceramic at 1050 ^oC for ~ 1h to form a liquid without any bubbles. In order to accomplish homogeneity, the melt was rotated in the crucible several times. The molten was then cast into a brass mold and then annealed at 400^oC. The glass samples were ground and polished optically to have suitable dimensions with non- parallelism of the two opposite side faces less than 0.01^o.

Mathematical techniques as In Situ Object Calibration Software (ISOCS) and LabSOCS which have undergone significant improvementsandenhancements. Inthis method, the detector response wascharacterized by creating a set of fine spatial efficiency gridsat 15 energies in the 0.045–7MeV range. The spatial grids are created in (r, θ) space about the detector, with the radiusr varies from 0 to 500 m**,** and the angle θ varying from 0to180^o. The ISOCS technique was introduced into the Agency's (Egyptian Nuclear Radiological Regulatory Authority (ENRRA) activities just a few years ago, and this methodology is being expanded to more and more NDA measurement techniques.

The ISOCS calibration method is conventional tool for calibrated detector efficiency as function of energy, for wide variety of source geometries and activity distributions. The ISOCS method consists of a characterization of the detector, user input of source geometry data, and the ISOCS software, which uses these to produce the efficiency calibration. A point source geometry is one in which a source is placed far enough away from the detector, so that the source can be considered to be essentially a point. A radioactive source of volume 1 cc, and filled with water, was modeled using each of the 11 ISOCS templates. The source to detector distance in each case was 5 meters as shown in Fig. (1). Since the source is essentially point, the efficiencies calculated by ISOCS in all the 11 cases should be very close to each other, if not identical. The ISOCS software is complex ensemble of computer codes that use elegant mathematical techniques to compute the efficiencies. It contains a series of the mathematical models that can simulate a wide variety of common sample shapes (boxes, cylinders, spheres, pipes, stacked discs, marinelli beakers, etc.). These models allow easy input of appropriate parameters necessary for efficiency computation. The ISOCS software divides each source region into a large number of voxels (1024). A point location is defined within each voxel. The point location inside a voxel is determined in a quasi- random fashion. At a given user specific energy, the detector efficiency is calculated for each voxel. The attenuation due to absorbers within the source and also in the intervening space between the source and detector is taken into account.

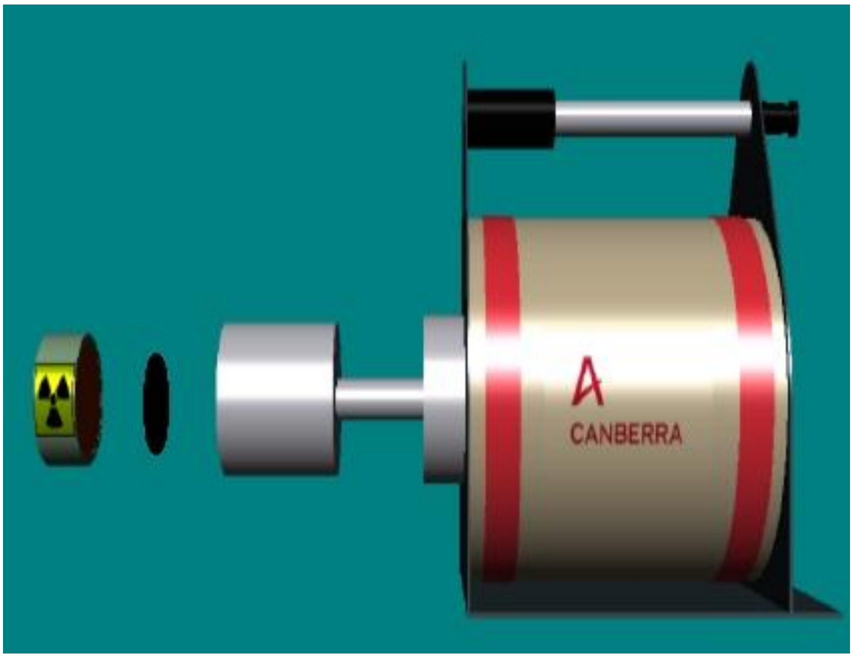

Figure 1.Detector and sample set up in ISOCS

Five samples in spheres form with different ratios have been used in the proposed method. the specification of assayed samples which were used are listed in Table (1).Each sample was measured in such a way that its axis of symmetry is coincidence with the extended axis of symmetry of the detector. The distance between the sample and the Al cap of the detector is 15 cm.Specific gamma ray's energy at 661.7, 1173 and

1332 keV, which is a signature for ^{137}Cs and ^{60} Co isotope respectively were measured. the ISOCS technique is used to estimate Mass attenuation coefficient *(μ_m)*, half value layer (HVL)and mean free path (MFP) for the verified samples.

Table 1. The nominal composition of the xTiO_2-20$Na_2B_4O_7$-30PbO-20SiO_2-(30-x) Bi_2O_3 glass system.

Sample code	Bi_2O_3	$Na_2B_4O_7$	PbO	SiO_2	TiO_2
T1	30	20	30	20	0
T2	20	20	30	20	10
T3	10	20	30	20	20
T4	5	20	30	20	25
T5	0	20	30	20	30

III. Results and Discussion

The linear attenuation coefficient can be determined using Beer-Lambert's law with thefollowing equation as [28];

$I = I_0 e^{-\mu x}$,(1)

where, I_0 and I are the intensities of the primary (emitted from theradioactive source) and passing through the absorber, µis the linearattenuation coefficient and x is the absorber thickness.

Many radiation attenuation parameters such as μ/ρ, HVL, TVL,MFP and Zeff can be determined by using the linear attenuationcoefficient. The μ/ρcan be obtained mathematically by dividing thelinear attenuation coefficient to mass density of the absorber. The μ/ρ is one of the most important parameters used in radiation research literature since it characterizes well the interaction probabilityof a photon with the shielding material. A photon may, fundamentally, experience absorption (photoelectric effect, pair production) andscattering (Rayleigh and Compton) interactions with the atoms of themedium.The μ/ρ is used to assess the ability of the glass to attenuate the photon. It gives an indication about the interaction of the photons with the atoms of the glass. Furthermore, μ/ρ is used to compute some other quantities such as electron density (N_{eff}), linear attenuation coefficient (μ), …etc. Also,the mass attenuation coefficient (μ/ρ) is the key quantity to estimate the shielding effectiveness of a given material. It gives an indication about the ability of the sample to attenuate gamma radiation[32].

Therequired absorber thickness which reduces the initial radiationintensity to 50% after passing through the absorber is called halfvalue layer (HVL) and it is obtained as [29];

$$HVL = 0.693 / \mu. \quad (2)$$

The required absorber thickness which lowers the initial radiation intensity to 10% after passing through the absorber is knownas tenth value layer (TVL) as [30];

$$TVL= 2.303 / \mu. \quad (3)$$

The required absorber thickness which reduces the initial radiation intensity to 36.8% after passing through the absorber iscalled mean free path (MFP) as [31];

$$MFP = 1/ \mu. \quad (4)$$

If a material has low HVL, TVL and MFP values, this indicates thatthis material is a good shielding material.

Figures (2 and 3) show the results obtained for the µ/ρ and µfrom ISCOS simulations were plotted versus content TiO_2 at different photon energies. It is clear from the figure that the attenuation factor such as µ/ρ and µ, is very large at the low energy and decreases rapidly with the raise of the energyand increased TiO_2 content.The comparatively high values in the attenuation factor at 661.7 keV can be attributed to photoelectric phenomenon as discussed early by El-Sayed et al. [33]. In this phenomenon, the µ/ρ has a high dependence on the atomic number, and the glasses in the present system contain heavy elements such as Bi which increase the chances of this phenomenon to happen.The sharp drop in µ/ρvalues can beexplained as follows: it is known that at low photon energy the photoelectric absorption (PE) is the most predominant photon-matter interaction mode, and therefore, the occurrence probability of the PEmode is usually governed by two important experimental parametersnamely; the element atomic numberand the incident photonenergy.It is clear from Figure (2) that, the linear attenuation coefficient (µ) decreases linearly with increasing energy for all types of samples, this is due to the different photon absorption mechanism for different photon energies [34,35]

Several interactions occur between the low energy photons with the present glasses while as the energy raises the chances of the interactions are reducingthe µ/ρvalues at lower energies and decreases sharply until 1173 keV thenstay almost constant until 1332 keV [Figure (3)]. The maximumvalues of µ/ρfor the five samples are found at 662 keV while the minimumvalues are observed at1173 and 1332 keV. This trend in the µ/ρis explainedby the interaction processes of photons at the low, medium andhigh energy part of the spectrum where photoelectric effect,Compton scattering and pair production process dominates, respectively [36]. It is also clear from the figure that the K-absorption edgeappears when the bismuth element added to glass systems,the values of (µ/ρ) for glasses decrease with the photon energy increases.

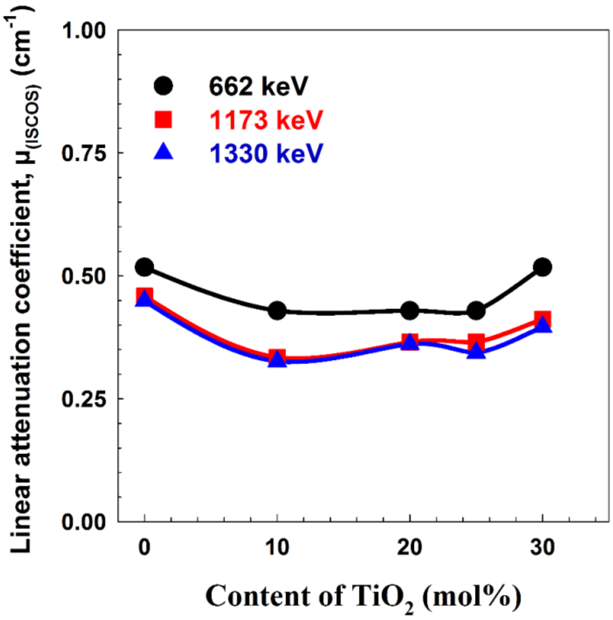

Figure 2. The variation of the linear attenuation coefficient with content of TiO_2 at different photon energies

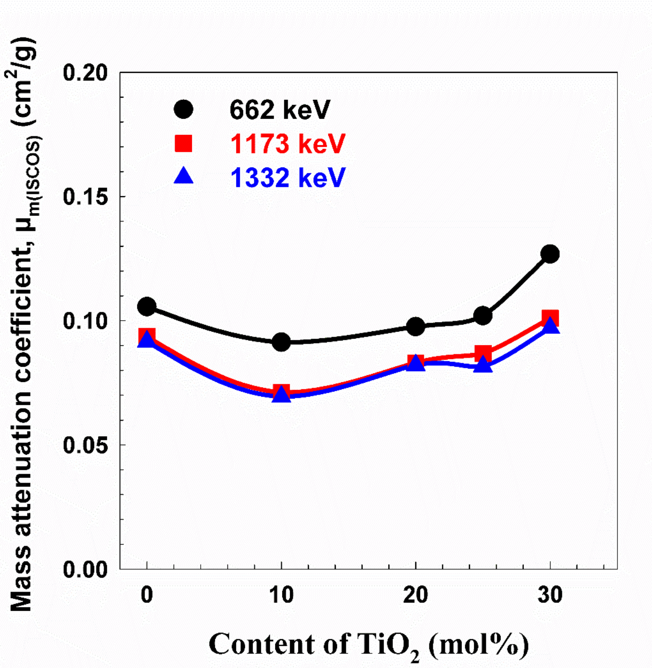

Figure 3.The variation of the mass attenuation coefficient with content of TiO_2 at different photon energies.

Half value layer (HVL) is another parameter derived from the μ/ρvalues and characterizes the shielding capability of the materials.The lesser value of HVL meansbetter shielding performance against the gamma rays, more photons can be attenuated with a material which has lower HVL values. Figure (4) shows the variation of the HVL with content of TiO_2 at different photon energies. It is clear from the figure that the additionof bismuth oxide affects the HVL, namely HVL decreases with the insertion of Bi_2O_3 on the expense of TiO_2. This is related directly to therelation between the density of the sample and the shielding potentialityof the sample. This result implies thatthe likelihood of photon-glass interaction is very high if the energy of thephoton is low, while this likelihood is small for high energy photons**.**By increasing the TiO_2 to be 30 mol% content, the HVL is became lower value which can be attributed to the high density of this sample.This result implies thatthe likelihood of photon-glass interaction is very high if the energy of thephoton is low, while this likelihood is small for high energy photons.

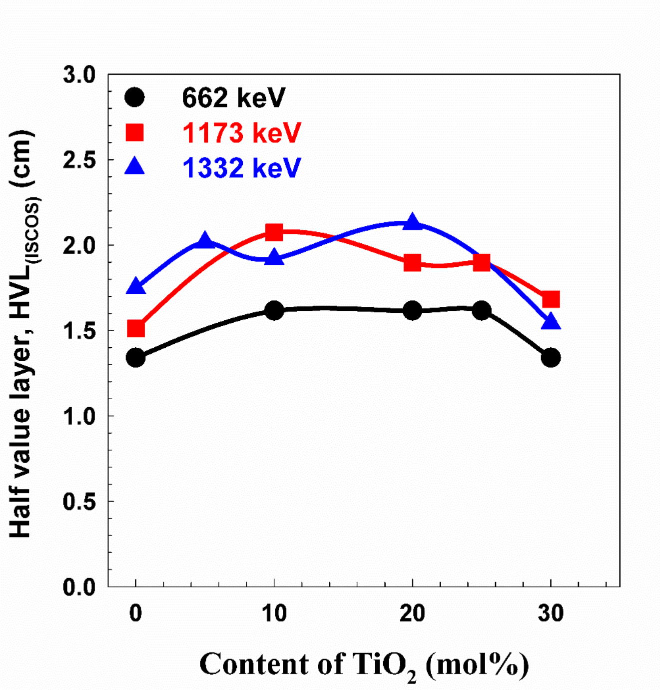

Figure 4.The variation of the half value layer with content of TiO_2 at different photon energies.

The tenth value layer (TVL) is another interesting parameter used todecide the attenuation behavior of certain sample, and gives directinformation about the dimension or the thickness of the specimen thatcan block most of the incoming photons (i.e., 90% of the initial value).Practically, glass specimen with small TVL is required and one can reduce this thickness by using heavy metal oxides like Bi_2O_3 with anappropriate amount or high density elements. Figure (5) shows the variation of the tenth value layer (TVL) with content of TiO_2 at different photon energies. It is observed that with increased the TiO_2 content the tenth value layer is increased slightly at low energy, but for high energies, it is increased with increasing the contents of TiO_2up to 15mol% and then decreased to become almost similar to 0 mol% of TiO_2.

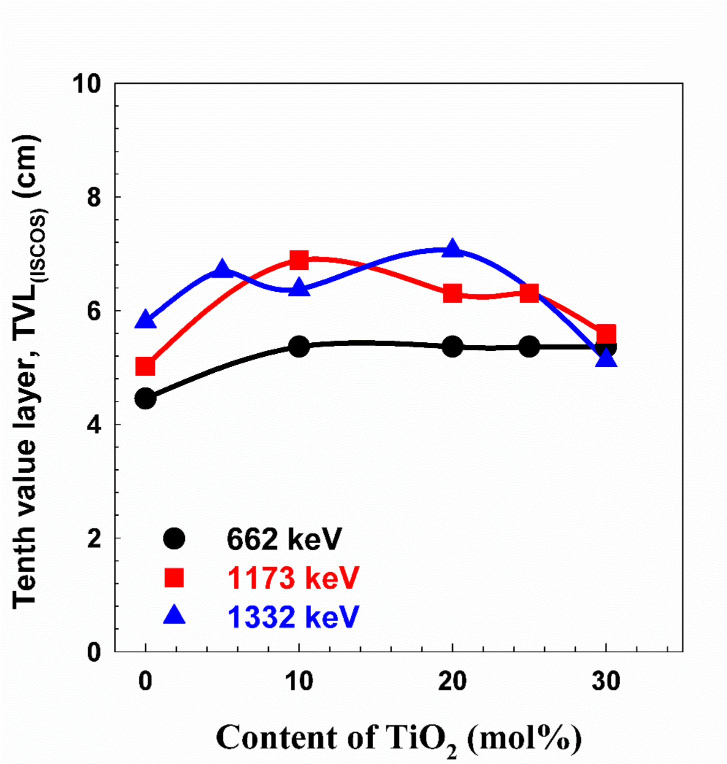

Figure 5.The variation of the tenth value layer with content of TiO_2 at different photon energies.

Also, the thickness/penetration depth of interacting material is measured in the units of mean free path, where one mean free path(MFP) represents the average traveled distance between two successive interactions of photons, which results in decreasing theintensity of incident photon beam by the factor of 1/e. It is equalto the reciprocal of linear attenuation coefficient according to equation (4).Since, the linearattenuation coefficient is an energy dependent parameter; therefore, the mean free path also varies depending on the incident photon energy. Figure (6) shows the variation of the mean free path (MFP) with content of TiO_2 at different photon energies. It is clear from the figure that the MFP is decreases with raise the photon energy at increase TiO_2 content, but at low energy we found semi decrease in the MFP with increases TiO_2 content.

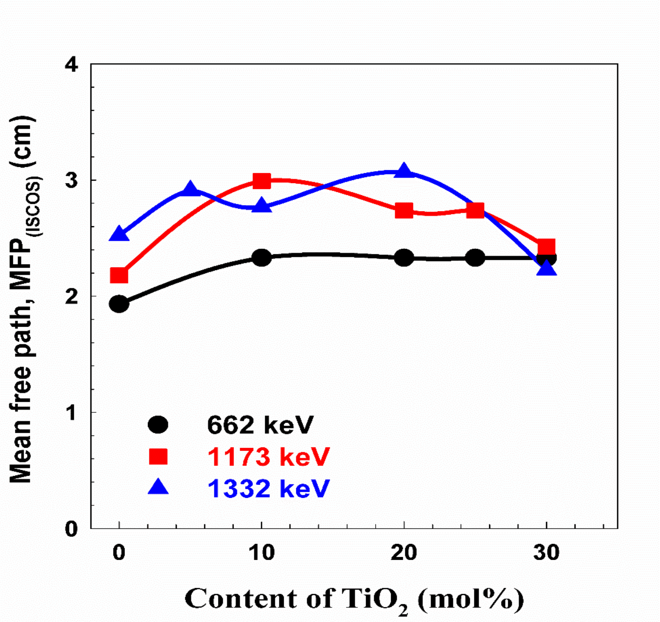

Figure 6. The variation of the mean free path with content of TiO_2 at different photon energies.

IV. Conclusions

In this study, Mass attenuation coefficient *(μ_m)*, half value layer (HVL), tenth value layer (TVL) and mean free path (MFP) for glasses composition $xTiO_2$–20 $Na_2B_4O_7$–30 PbO–20 SiO_2–(30-*x*) Bi_2O_3 ($0 \le x \le 30$ mol%), have been calculated theoretically at 662 kev from^{137}Csand1173 &1332 Kev from ^{60}Co photon energies by using ISOCS software. it is found that ISOCS calculations are a very good method for the calculation of the previous gamma ray shielding properties at different photon energies. Also, we found that ISOCS calculations are preferred because of its cost in which includes money, effort, and time is lower than the experimental method.

Mass attenuation coefficient, mean free path, half value layer, tenth value layer values of theinvestigated glass systems are strongly energy dependent parameters. These parameters exhibit differenttrends in variation byenergy due to that fundamental photon interaction processes (i.e.photoelectric, scattering and pair production) are dominant throughdifferent energy regions.

A material tobe used as a gamma-ray radiation shielding material must havelow values of HVL and MFP. Therefore, it is indicated thatBi_2O_3–TiO_2 glass systems, which show low values of HVL and Hence it is proposed that the prepared glass samples can be promising nominees as non – conventional alternate gamma – ray shielding materials.

NDA method based on gamma ray spectrometry is used to determination of Shielding Properties for different materials in various shapes by In-Situ Object Calibration Software (ISOCS).The proposed method may be extended to different materials to verify and determined material Shielding Properties with different matrices, chemical and physical forms and isotopic compositions. This technique can be applied in the case of experimental tools not available and give very acceptable results.

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