Gamma ray buildup factors and fast neutron removal of Tb_2O_3 doped WO_3: Gd_2O_3: B_2O_3 glass systems

Kittisak Sriwongsa[1,2,*] Punsak Glumglomchit[3] and Jakrapong Kaewkhao[4,5]

[1]Lecturers responsible for Bachelor of Education Program in Physics, Faculty of Education, Silpakorn University, Nakhon Pathom, 73000, Thailand

[2]The demonstration school of Silpakorn University, Nakhon Pathom, 73000, Thailand

[3]Huahin Vitthayalai School, Hua-Hin, Prachuapkhirikhan, 77110, Thailand

[4]Physics Program, Faculty of Science and Technology, Nakhon Pathom Rajabhat University, Nakhon Pathom 73000, Thailand

[5]Centre of Excellence in Glass Technology and Materials Science (CEGM), Nakhon Pathom Rajabhat University, Nakhon Pathom 73000, Thailand

---

Abstract

Gamma ray buildup factors and fast neutron removal of Te_2O_3 doped WO_3: Gd_2O_3: B_2O_3 glass systems were estimated. The buildup factors values were determined for photon energy 0.015–15 MeV up to penetration depths of 40 mfp (mean free path) by the geometrical progression (GP) method. The EABF and EBF values were found dependent upon photon energy, penetration and Te_2O_3 content. In low and high photon energy ranges, the EABF and EBF values were minimum whereas maximum in intermediate energy range. The fast neutron removal cross sections were computed by the partial density method. The glass systems with 2.0 mol% Te_2O_3 is found to be superior gamma ray and neutron transparent shielding. The results indicate that this glass can be used as radiation shielding material

Keywords: Buildup factors, Neutron, Tellurite, Glass

บทนำ

The creating of radiation shielding medium is an interesting area in the field of radiation application in medical, agriculture, industries, nuclear reactors and accelerator technologies, and future generation fusion reactors. The materials such as concretes, lead and other high–Z materials are the suitable alternative to protect personnel from neutral radiation such as X–ray, gamma ray and neutron. The shielding against compound or mixture of gamma and neutron requires an adequate composition of low and high–Z elements [1,2]. The transmitted intensity of a gamma ray beam moves pass a medium follows Lambert's Beer law under three conditions: (i) mono–

---

* **ผู้ประสานงาน (Corresponding Author)**
e-mail: sriwongsa_k@silpakorn.edu

chromatic ray (ii) thin absorbing medium, and (iii) narrow beam geometry. In case, any one of these conditions is not met, this law is no longer valid. The law can be made valid by using a correction factor, called as "buildup factor". The concept of buildup factor was introduced in late 1950 by obtaining experimentally the buildup factor at 1.25 MeV photon for water up to 16 mfp (mean free path) [3,4].

The determination for buildup factors (BFs) by various codes were reported in ANSI/ANS-6.4.3–1991 by American Nuclear Society (ANSI/ANS–6.4.3, 1991) at energy range 0.015–15 MeV up to penetration depth of 40 mean free path (mfp). The BFs in report ANS–6.4.3 have 23 elements of atomic number, Z = 4–92. The Geometrical Progression (GP) which developed by Harima is gives BFs of the good agreement with the ANS–6.4.3. And then, the GP fitting formula is used to reported gamma ray buildup factors in many materials such as concretes, fly–ash materials, soils & ceramic, soils and fly–ash which shows that the GP fitting is a useful method for estimation of the exposure buildup factors [5,6].

In high density glass with high neutron cross section element, exposure buildup factor (EBF) values and fast neutron removal cross section are important parameters, which can explain behavior of gamma ray and neutron though this material. The main objective of this study is computing the Energy absorption buildup factor (EABF) and exposure buildup factor (EBF) values and fast neutron removal cross section of the high density WO_3-Gd_2O_3-B_2O_3-Tb_2O_3 glass systems and variation of the buildup factor values with photon energy, penetration depth and Tb_2O_3 molar composition. The EABF and EBF values of the glass systems were computed for photon energy 0.015–15 MeV up to a penetration depth of 40 mfp by using the GP method. The fast neutron removal cross sections of glass systems were computed. The study should be very useful for effectiveness of transparent shielding glass material for a wide energy range applications and design of shielding.

วัตถุประสงค์

1. To investigate the mass attenuation coefficients of glass systems in energy range of 1 keV–100 GeV.
2. To compute buildup factors for photon energy 0.015–15 MeV up to a penetration depth of 40 mfp by using the GP method.
3. To determine the fast neutron removal cross sections of glass systems.

Material and methods

The theoretical/method basis have been separated into following sub-sections:

วิธีการวิจัย

Radiation shielding parameters

The theoretical computation of mass attenuation coefficients (μ_m) for glass systems using WinXCom software program for photon at energies range 1 keV to 100 GeV by Eq. (1) [7]:

$$\mu_m = \sum_i w_i (\mu_m)_i \tag{1}$$

where wi and (μ_m)i are weight fraction and mass attenuation coefficient of element i, respectively.

Buildup factors

Computation of equivalent atomic numbers (Z_{eq})

Equivalent atomic numbers (Z_{eq}) is value to explain properties in terms of equivalent elements identical to atomic number for single element [8]. Z_{eq} values for chose material were applied by matching ratio R (μ_{Comp}/μ_{total}) of specific glass at same energy with according ratio of element. Compton partial attenuation coefficient (μ_{Comp}) and total attenuation coefficients (μ_{total}) values were applied by using WinXCom software program at energy from 0.015–15 MeV. Z_{eq} were computed by logarithmic interpolation formula (Eq. (2)) [9] and exhibited in Fig. 2.

$$Z_{eq} = \frac{Z_1(\log R_2 - \log R) + Z_2(\log R - \log R_1)}{(\log R_2 - \log R_1)} \tag{2}$$

Here Z_1 and Z_2 are atomic numbers of elements according to μ_{Comp}/μ_{total} ratios, R_1 and R_2, respectively, and R (μ_{Comp}/μ_{total}) is ratio for chose material at specific energy, which lies between ratios R_1 and R_2.

Computation of geometric progression (GP) fitting parameters

American National Standards ANSI/ANS–6.4.3 were reported buildup factor values for 23 elements (Z = 4–92), one compound and two mixtures (i.e., air and concrete) in energy range 0.015–15 MeV up to deep 40 mfp for used to standard reference database. This database was used to compute G-P fitting buildup factor coefficients values for glasses compounds according to formula [9,10]:

$$P = \frac{P_1(\log Z_2 - \log Z_{eq}) + P_2(\log Z_{eq} - \log Z_1)}{\log Z_2 - \log Z_1} \tag{3}$$

here P_1 and P_2 are GP fitting coefficients values which according to atomic numbers of Z_1 and Z_2 at same energy, respectively.

Computation of energy exposure and absorption buildup factors

At last, GP fitting parameters were used to compute energy absorption and exposure build-up factors at chosen incident photon energies in range 0.015 < E < 15.0 MeV up to 40 mfp deep penetration. This was determined by [9,10]:

$$B(E,x)=\begin{cases} 1+(b-1)x, K=1 \\ 1+\dfrac{b-1}{K-1}(K^{x}-1), 1<K\leq 40 \\ otherwise, K\geq 40 \end{cases} \quad (4)$$

K(E,x) is photon dose multiplication factor is function of tangent hyperbolic which is expressed by following equation [9,10]:

$$K(E,x)=cx^{a}+d\frac{\tanh(x/X_{k}-1)-\tanh(-2)}{1-\tanh(-2)}, x\leq 40 \quad (5)$$

here E is incident photon energy, x is deep penetration in terms of mean free path (mfp) and a, b, c, d, X_k are GP fitting parameters.

Fast neutron removal cross section

A method for computation of the attenuation of fast neutrons by application of effective removal cross section was developed. The effective removal cross section of a compounds and homogenous mixtures is possibly determined from the value of Σ_R (cm^{-1}) or Σ_R/Σ (cm^2/g) of the elements in the mixtures or compounds by mixture rule. Difference in application of mixture or compound for neutron interaction differs as weight fraction is replaced by partial density and mass attenuation coefficient by neutron removal cross section [11,12].

$$\sum_{R/\rho}=\sum_{i}w_{i}(\sum_{R}/\rho)_{i} \quad (6)$$

and

$$\sum_{R}=\sum_{i}\rho_{i}(\sum_{R}/\rho)_{i} \quad (7)$$

where ρi and $(\Sigma_R/\rho)i$ are the partial density and the fast neutron mass removal cross section of the *i* th constituent, respectively. The $\Sigma_R/_\rho$ values of elements of the glasses were taken from Kaplan and Chilton.

ผลการวิจัย Results and discussion

The chemical composition of the glass systems is listed in Table1. The variation of mass attenuation coefficient with photon energy for the glass systems has been shown in Fig. 1. The variation in the Energy absorption buildup factor (EABF) and exposure buildup factor (EBF) values of the glass systems with photon energy are shown in the Figs. 3 and 4 (a–e) for different penetration depths (1, 5, 10, 15, 20, 25, 30, 35 and 40 mfp). The EABF and EBF values for the glass systems as a function of penetration depths at photon energies 0.015, 0.15, 1.5 and 15 MeV are shown in Figs. 6 and 7 (a–d) and the fast neutron removal cross sections of the glass systems for neutron are shown in Fig. 8.

Table 1 Chemical compositions of glass systems in mol%.

Code	Mole fraction				Chemical Formula
	WO_3	Gd_2O_3	B_2O_3	Tb_2O_3	
Tb0	42.5	27.5	30.0	0.0	$42.5WO_3$: $27.5Gd_2O_3$: $30B_2O_3$
Tb1	42.5	27.5	29.9	0.1	$42.5WO_3$: $27.5Gd_2O_3$: $29.9B_2O_3$: $0.1Tb_2O_3$
Tb2	42.5	27.5	29.5	0.5	$42.5WO_3$: $27.5Gd_2O_3$: $29.5B_2O_3$: $0.5Tb_2O_3$
Tb3	42.5	27.5	29.0	1.0	$42.5WO_3$: $27.5Gd_2O_3$: $29.0B_2O_3$: $1.0Tb_2O_3$
Tb4	42.5	27.5	28.0	2.0	$42.5WO_3$: $27.5Gd_2O_3$: $28.0B_2O_3$: $2.0Tb_2O_3$

Mass attenuation coefficient of glass systems

The mass attenuation coefficient, μ_m of glass systems is shown in Fig. 1. It is shown that the μ_m values of glass systems are highest in photoelectric absorption region, reduces gradually and become lowest in Compton scattering region. After that, it again starts increasing and become nearly constant at around photon energy of 100 MeV. The graphs of μ_m value for glass systems was discontinuous in photoelectric absorption region because of absorption edge of elements as show in Table 2. These variations can be discussed by photon energy and Z dependency of interaction cross section of the elements.

Table 2 The photon energies (in ◊ 10–3 MeV) of absorption edges for elements.

Element	K	L1	L2	L3	M1	M2	M3	M4	M5
Gd	50.24	8.376	7.930	7.243	1.881	1.688	1.544	1.217	1.185
Tb	52.00	8.708	8.252	7.514	1.968	1.768	1.611	1.275	1.241
W	69.53	12.10	11.54	10.21	2.820	2.575	2.281	1.872	1.809

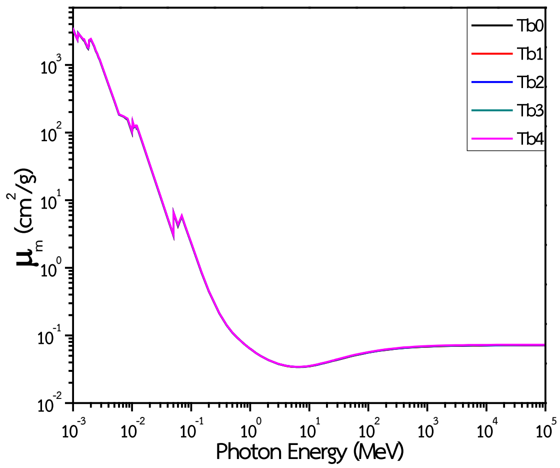


Fig. 1 The mass attenuation coefficients of glass the glass systems for photon energy of 1 keV – 100 GeV.

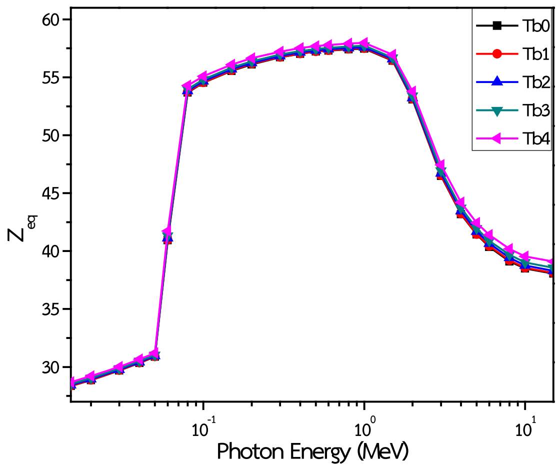


Fig. 2 The equivalent atomic numbers of systems.

Variation of EABF and EBF with photon energy

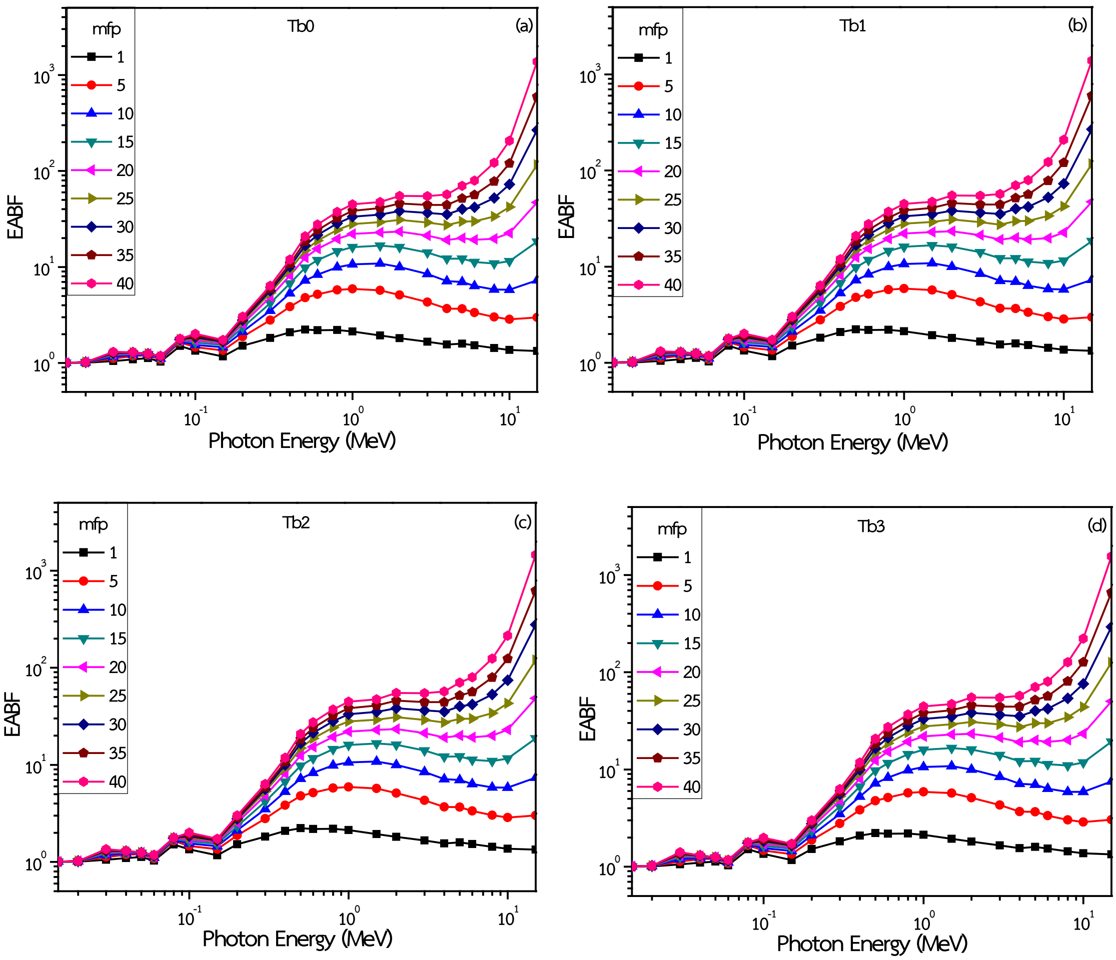
Fig. 2 gives equivalent atomic numbers (Z_{eq}) of shielding radiation materials at energy range from 0.015–15 MeV. This figure is exhibit Z_{eq} dependence of Tb_2O_3 content which Z_{eq} increase with increasing Tb_2O_3 content.

The variations of EABF and EBF, with photon energy, for the glass systems and at different penetration depths, have been shown in Figs. 3 and 4 (a–e), respectively. Form the figures, it is seen that with increasing the photon energy, the EABF and EBF values, for glass systems, increase up to a maximum value at intermediate energies range. This can be discussed in the fundamental of dominance of different partial photon interaction processes in different energy ranges. However, in the low energy range, photoelectric absorption is the main photon interaction process, so maximum number of photons will be absorbed and consequently, EABF and EBF values are reduced. Same as, in the high energy range, another photon absorption process, that is pair production, is the main one. In the intermediate energy range, Compton scattering is the main process of photon interaction that only help in degradation of photon energy because of scattering and fails to completely remove the photon. So, more the life time of the photon is long, more the probability of photon to get away the material is important. This process results in the EABF and EBF values increase.

At high energy and large penetration depth, graph can observe a sharp peak in EABF and EBF. This is because of buildup of secondary gamma photons build by electron–positron annihilation in material because of to multiple scattering incidences. Truly, the increase in penetration depth of the mediums leading to increase the thickness of the interacting medium which in turn results in increasing the scattering events in the interacting medium, in particular for the medium with the highest equivalent atomic number. Hence it results in large EABF and EBF values.

Clearly from Fig. 3 (a–e) there is a sharp peak at 0.08 MeV energy that may be because of edge absorption of Tb.

It can observe that with increasing Tb_2O_3 content, the variation of EABF and EBF is same and the only difference is in the scale of EABF and EBF. Also, it's obvious differences between EABF and EBF values were observed as the Tb_2O_3 content changes. It is observed that these differences decreased the Tb2O3 content increases.



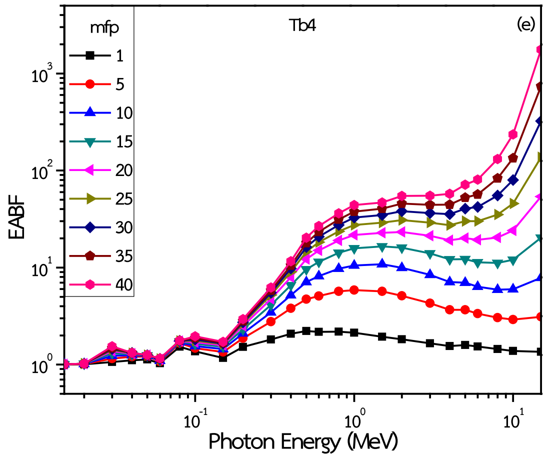
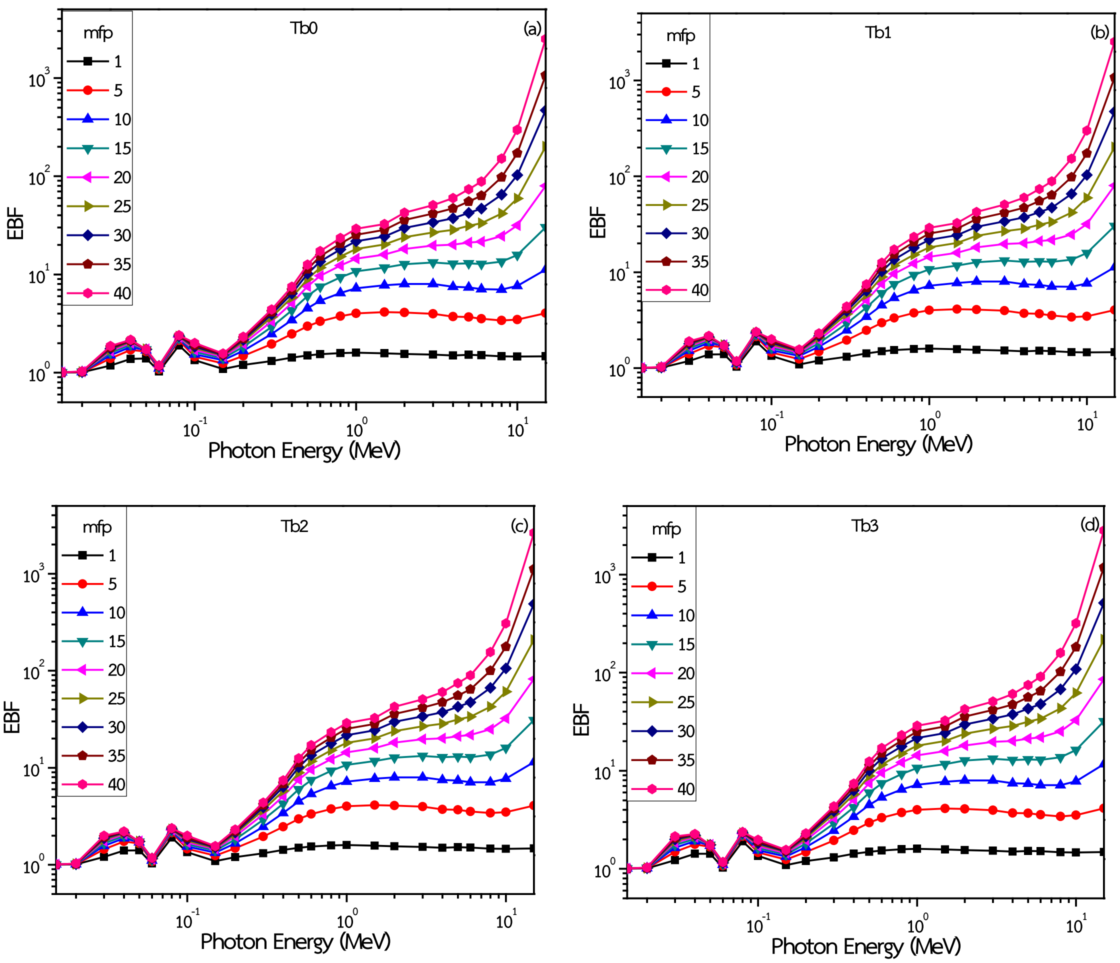


Fig. 3 (a–e) The EABF of glass systems with photon energy for different Tb_2O_3 content.



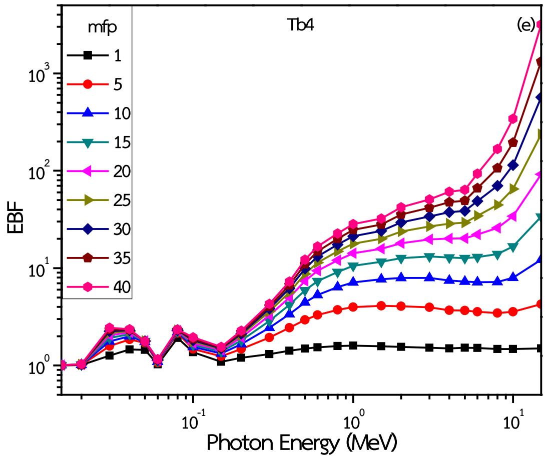


Fig. 4 (a–e) The EBF of glass systems with photon energy for different Tb_2O_3 content.

Variation of EABF and EBF with penetration depths

Figs. 5 and 6 show an increase of EABF and EBF values with penetration depths for the glass systems, at specific the photon energies (0.015, 0.15, 1.5 and 15 MeV). At the lowest energy (Figs. 5 and 6 a), the EABF and EBF values are roughly constant above 5 mfp. The EABF and EBF values according to the chemical composition except at photon energy 0.15 and 1.5 MeV, as shown in Figs. 5 and 6 (c).

It found that for the glass systems with low equivalent atomic numbers (Z_{eq}) (Tb0), the EABF and EBF values are small in the photon energy 0.015 and 15 MeV. However, for the glass systems with higher equivalent atomic numbers (Tb4), the EABF and EBF values are relatively small.

In contrast, at fixed photon energy 0.15 and 1.5 MeV (Figs. 5 and 6 (b–c)), EABF and EBF values are nearly free of chemical composition of glass systems. It may be because of the reason that in this range, the Compton scattering process whose cross section varies linearly with Z_{eq} of the glass systems is the main interacting process, therefore no significant variation of EABF and EBF were observed at this energy.

In high energy range ($E \geq 1.5$ MeV), the trend of EABF and EBF values started increase with increasing Z_{eq} (Figs. 5 and 6 (c–d)). This can be discussed on the fundamental of pair production which the main interaction in this energy range. The cross section of pair production process varies with equivalent atomic number as $(Z_{eq})^2$. So, the glass systems with higher Z_{eq} (Tb4) has higher probability to undergo pair production. The result of this process, electron and positron must be produced with different kinetic energies depending on the photon energy. The electron–positron pair suffers several numbers of collisions when it moves pass the glass systems and hence loses its energy. Since, the penetration depth of the glass systems is adequately large for the electron–positron pair to lose its energy with the glass systems and comes to rest. After that, this electron and positron can undergo annihilation process, which results in the creation of two new secondary gamma rays of 0.511 MeV. These secondary rays have energies in the main range of Compton scattering process. Therefore, these photons will reduce energy in multiple collisions before completely absorbed with in the glass systems by photoelectric absorption. So, the probability of photons to move pass the larger dimensions of glass systems increases, resulting in higher values for EABF and EBF.

At last, for a better radiation shielding material, a high value of μ_m and low value of buildup factor are requested. The μ_m values in the investigated glass systems increase as the content of Tb_2O_3 is increased. The EABF and EBF values increase as the content of Tb_2O_3 is increased in low and high photon energies. Obviously, the higher values of Tb_2O_3 content in the glass systems will improve the radiation shielding properties in terms of μm and buildup factor. Hence, it can conclude that glass systems with 2.0 mol% Tb_2O_3 appears as best gamma ray shielding glass due to higher values for μ_m all energies range and lower values of EABF and EBF in intermediate energies.

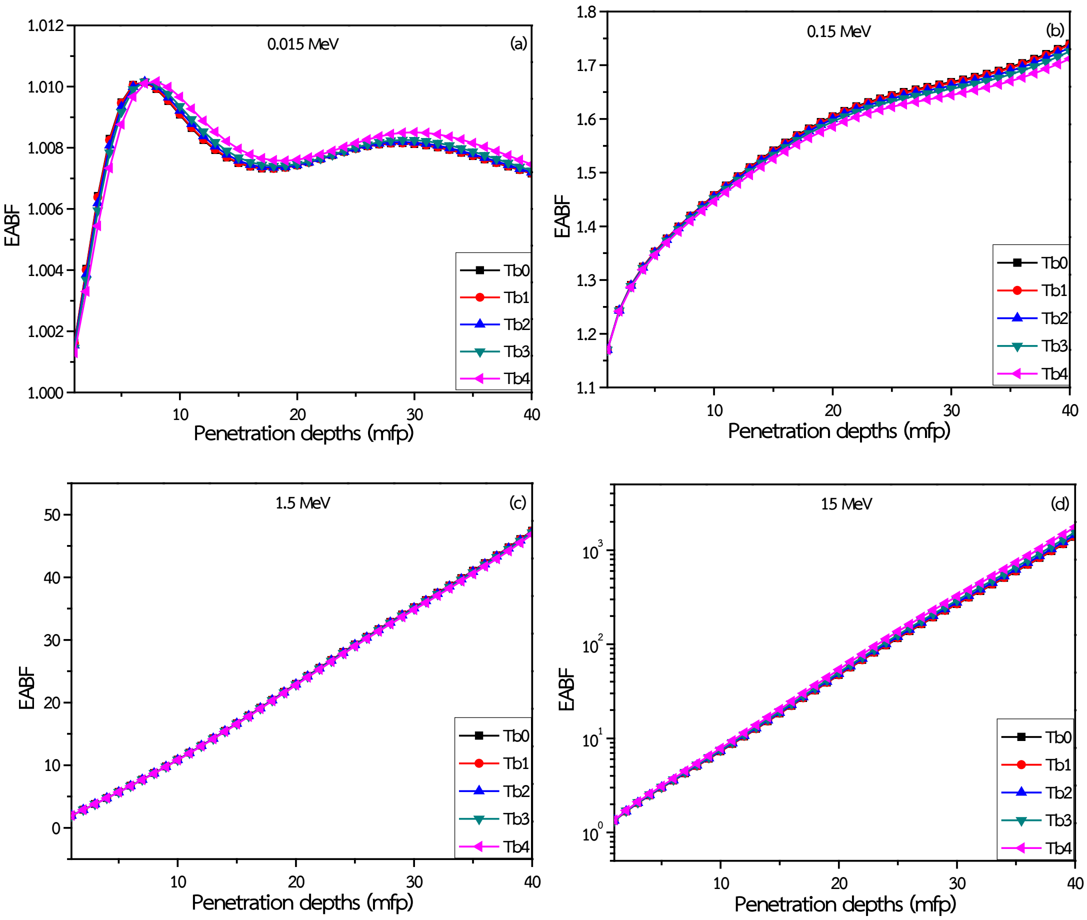


Fig. 5 (a–d) The EABF of glass systems with penetration depths at photon energies (0.015, 0.15, 1.5 and 15 MeV).

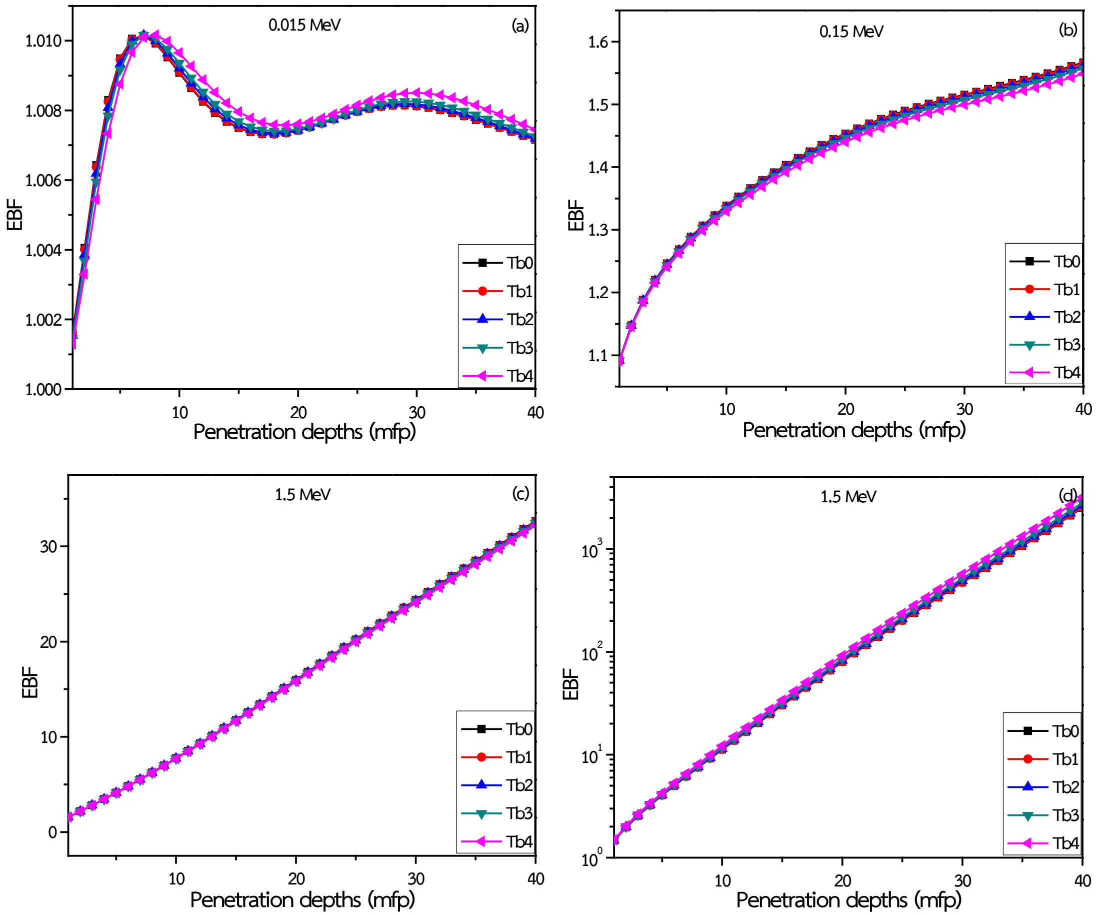


Fig. 6 (a–d) The EABF of glass systems with penetration depths at photon energies (0.015, 0.15, 1.5 and 15 MeV).

Fast neutron removal cross section

The fast neutron removal cross section Σ_R (cm^{-1}) of the glass systems with 0, 0.1, 0.5, 1.0 and 2.0 mol% Tb_2O_3 is shown in Fig. 6. It found that Σ_R of glass systems for Tb_2O_3 at 2.0 mol% had the highest value. Highest value of removal cross section of 2.0 mol% of Tb_2O_3 was observed because of highest elemental composition of low–Z, B_2O_3. Therefore, it is concluded that low–Z elemental composition contributes a vital role in neutron shielding properties.

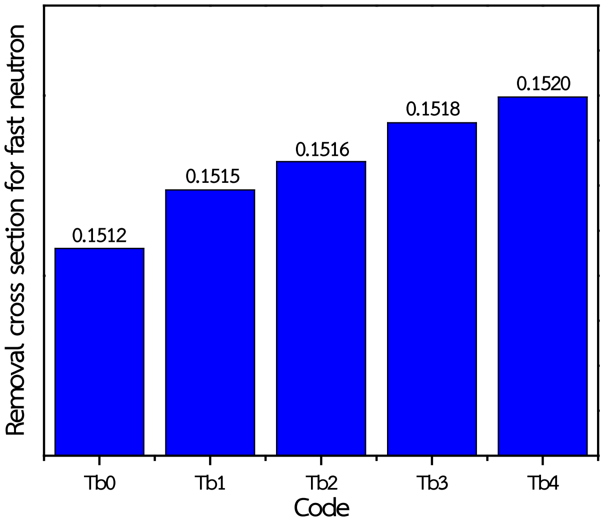


Fig. 7 The fast neutron removal cross section for glass systems.

Conclusions

In this work, the shielding properties for 42.5WO_3: 27.5Gd_2O_3: (30–x)B_2O_3: xTb_2O_3 glass systems were investigate with different molar composition of Tb_2O_3. The mass attenuation coefficient was computed for total photon interaction in the energy range of 1 keV to 100 GeV.

It was found that the mass attenuation coefficient increases with increasing Te_2O_3 concentration. Moreover, by Geometric Progression method (G-P), energy absorption buildup factors and exposure buildup factors were determined for photon energy in the range 0.015–15 MeV, up to penetration depths of 40 mfp (mean free path). It was found that EABF and EBF possess maximum values in the intermediate energy range, where Compton scattering is the main photon interaction process. The EABF and EBF depend strongly on the chemical composition of the glass systems in the lower and higher energy range, become nearly independent at the intermediate energy range. For glass systems with 2.0 mol% of Tb_2O_3, the EABF and EBF values are found the lowest in low–to–intermediate energy, thus it is having superior gamma ray shielding properties. The obtained results have scientific values to develop excellent shielding properties of materials and provide references to synthesize new materials for gamma rays shielding applications.

Acknowledgments

The authors would like to thank Nakhon Pathom Rajabhat University to support for glass formal and software. This work is partially supported by National Research Council of Thailand (NRCT) and Silpakorn University.

References

Preet Kaur, Devinder Singh, Tejbir Singh. (2016). Heavy metal oxide glasses as gamma rays shielding material. *Nuclear Engineering and Design,* 307, 364–376.

Vishwanath P.Singh, N.M. Badiger, N. Chanthima, J. Kaewkhao. (2014). Evaluation of gamma-ray exposure buildup factors and neutron shielding for bismuth borosilicate glasses. *Radiation Physics and Chemistry,* 98, 14–21.

Kulwinder Singh Mann, Jyoti Singla, Vipan Kumar, Gurdeep Singh Sidhu. (2012). Investigations of mass attenuation coefficients and exposure buildup factors of some low–Z building materials. *Annals of Nuclear Energy,* 43, 157–166.

M.I. Sayyed, H. Elhouichet. (2017). Variation of energy absorption and exposure buildup factors with incident photon energy and penetration depth for boro–tellurite (B2O3–TeO2) glasses. *Radiation Physics and Chemistry,* 130, 335–342.

M. Kurudirek, D. Sardari, N. Khaledi, C. Çakır, K.S. Mann. (2013). Investigation of X–and gamma ray photons buildup in some neutron shielding materials using GP fitting approximation. *Annals of Nuclear Energy,* 53, 485–491.

Vishwanath P. Singh, N.M. Badiger. (2014). Gamma ray and neutron shielding properties of some alloy materials. *Annals of Nuclear Energy,* 64, 301–310.

Cherdsak Bootjomchai, Jintana Laopaiboon, Chadet Yenchai, Raewat Laopaiboon. (2012). Gamma–ray shielding and structural properties of barium–bismuth–borosilicate glasses. *Radiation Physics and Chemistry,* 81, 85–790.

Vishwanath P. Singh, N.M. Badiger. (2014). Comprehensive study on energy absorption buildup factors and exposure buildup factors for photon energy 0.015 to 15MeV up to 40 mfp penetration depth for gel dosimeters. *Radiation Physics and Chemistry,* 103, 234–242.

M.I. Sayyed., Saleem I. Qashou., Z.Y. Khattari. (2017). Radiation shielding competence of newly developed TeO_2–WO_3 glasses. *Journal of Alloys and Compounds. 696,* 632–638.

Preet Kaur, Devinder Singh, Tejbir Singh. (2018). Gamma rays shielding and sensing application of some rare earth doped lead-alumino-phosphate glasses. *Radiation Physics and Chemistry,* 144, 336–343.

Murat Kurudirek. (2017). Heavy metal borate glasses: Potential use for radiation shielding. *Journal of Alloys and Compounds,* 727, 1227–1236.

Berna Oto, Nergiz Yıldız, Turgay Korkut, Esra Kavaz. (2015). Neutron shielding qualities and gamma ray buildup factors of concretes containing limonite ore. *Nuclear Engineering and Design,* 293, 166–175.

คณะผู้เขียน/ ผู้เขียน

รศ.ดร. จักรพงษ์ แก้วขาว

คณะวิทยาศาสตร์และเทคโนโลยี มหาวิทยาลัยราชภัฏนครปฐม

คณะวิทยาศาสตร์และเทคโนโลยี มหาวิทยาลัยราชภัฏนครปฐม

เลขที่ 85 ถนนมาลัยแมน อำเภอเมือง จังหวัดนครปฐม รหัสไปรษณีย์ 73000

e-mail: mink110@hotmail.com

อ.ดร. กิตติศักดิ์ ศรีวงค์ษา

คณะศึกษาศาสตร์ มหาวิทยาลัยศิลปากร

คณะศึกษาศาสตร์ มหาวิทยาลัยศิลปากร

เลขที่ 6 ถนนราชมรรคาใน อำเภอเมือง จังหวัดนครปฐม รหัสไปรษณีย์ 73000

e-mail: sriwongsa_k@silpakorn.edu

นายพันศักดิ์ กล่ำกล่อมจิตร์

โรงเรียนหัวหินวิทยาลัย หัวหิน ประจวบคีรีขันธ์

โรงเรียนหัวหินวิทยาลัย

เลขที่ 240 ถนน เพชรเกษม ตำบลหัวหิน อำเภอหัวหิน จังหวัดประจวบคีรีขันธ์ รหัสไปรษณีย์ 77110

e-mail: punsakk@gmail.com