Gamma Photon Exposure Buildup Factors for Some Spin Ice Compounds Using G-P Fitting Method

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Abstract: Exposure buildup factors (EBF) for some spin ice compounds such as Dysprosium Titanate (Dy₂Ti₂O₇), Dysprosium Stannate (Dy₂Sn₂O₇), Holmium Titanate (Ho₂Ti₂O₇) and Holmium Stannate (Ho₂Sn₂O₇) useful in nuclear engineering have been computed for photon energy range 0.015 to 15 MeV upto penetration depth of 40 mean free path. The EBF for these compounds were found to be the largest at photon energy 15 MeV excepts in low energy. The EBF for the compounds containing tin were found to be the largest as well as shown a peak at 30 keV photon energy.

Keywords: spin ice; gamma; exposure; shielding

1. INTRODUCTION

The intensity of gamma-ray beam through a medium follows Lambert’s Beer law (I=I₀ e⁻µt) for narrow, monochromatic gamma-ray for thin absorbing material, where I and I₀ are transmitted and initial photon densities, µ is linear attenuation coefficient and t is the thickness of medium. In case of broad beam, polychromatic gamma-ray entering into thick medium, the law is made applicable by introducing a correction factor called as “buildup factor B”. Now the modified equation is I= B×I₀ e⁻µt including the buildup factors, B. The B is 1 when all the above conditions are satisfied. The buildup is defined as the ratio of total value of specified radiation quantity at any point to the contribution to that value from radiation reaching to the point without having undergone a collision.
Dysprosium Titanate ($\text{Dy}_2\text{Ti}_2\text{O}_7$), Dysprosium Stannate ($\text{Dy}_2\text{Sn}_2\text{O}_7$), Holmium Titanate ($\text{Ho}_2\text{Ti}_2\text{O}_7$) and Holmium Stannate ($\text{Ho}_2\text{Sn}_2\text{O}_7$) are geometrically frustrated magnetic materials called as “Spin ice compounds” due to incompatibility between the lattice symmetry and spin-spin interactions [1]. Geometrical frustrations in the spin ice compound occur when the spins on its lattice are not capable of simultaneous minimizing their pair-wise energies. The long-range magnetic dipolar interactions are responsible for spin ice behavior in the Isingpyrochlore magnets [2]. The magnetic rare-earth ions in these materials are situated on the lattice of corner-sharing tetrahedral, where their spins are constrained by crystal field interactions to point either directly toward or directly away from the centers of the tetrahedral. The neutron scattering measurements of spin correlations show that the magnetic Coulomb force acts between the magnetic monopoles by fluctuating the magnetic monopoles between high and low density states close to the critical point [3,4].

The spin ice compounds have high neutron absorption cross section and hence these materials are useful for research and power reactors for controlling the nuclear fission processes. The $\text{Dy}_2\text{Ti}_2\text{O}_7$ and $\text{Dy}_2\text{TiO}_5$ are being used as a neutron absorber material in the form of control rods in VVER types Russian reactors [5,6]. On the other hand, $\text{Dy}_2\text{Ti}_2\text{O}_7$ exhibits n-type semiconductor properties for single crystal with band-gap of 2.4 eV [7]. The existence of magnetic monopoles in the spin ice compounds provides new era of material properties of condensed matter. It is interesting to note that the spin ice compound exhibit the magnetic monopole at low temperature which become thrust area in condensed matter physics. Therefore, it is interesting to study the buildup of gamma-ray in these materials whereas the photon interaction parameters for some spin ice compounds has been already reported [8]. Although various investigators studied exposure buildup factors for composite materials such as soils, fly-ash, building materials, glasses, detectors, etc elsewhere. However, exposure buildup factors for spin ice compounds are not found in the literature. This has encouraged us to study the exposure buildup factors for spin ice compounds. In view of above, the exposure buildup factors, EBF for some spin ice compounds such as $\text{Dy}_2\text{Ti}_2\text{O}_7$ (DTO), $\text{Dy}_2\text{Sn}_2\text{O}_7$ (DSO), $\text{Ho}_2\text{Ti}_2\text{O}_7$ (HTO) and $\text{Ho}_2\text{Sn}_2\text{O}_7$ (HSO) were calculated using G-P fitting method for photon energy range 0.015 to 15 MeV up to penetration depth 40 mfp. The calculation of buildup factor is explained below.

2. THEORETICAL BACKGROUND FOR EXPOSURE BUILDUP FACTORS

The compilation for buildup factors by various codes is reported by American Nuclear Society (ANS) [9]. The data in the report covers energy range 0.015-15
MeV up to penetration depth of 40 mean free path (mfp). The buildup factors in the ANS report [9] are for 23 elements (Z=4-92). Harima et al., [10] developed a fitting formula, called Geometric Progression (G-P) which gives buildup factors of the good agreement with the ANS [9]. Harima [11] reviewed extensively and reported the current gamma-ray buildup factors. Various researchers investigated gamma-ray buildup factors for different materials; soil [12], fly-ash [13], building materials [14], borosilicate glass [15], and neutron detectors [16] which shows that the G-P fitting is a useful method for estimation of the buildup factors. The EBF by American Nuclear Society [9], G-P fitting method and MCNP-5 [17] for water for photon energy range 0.015-15 MeV up to 40 mfp are standardized [13,15]. It was found that the calculation of buildup factor by present work and ANS [9] agrees within a few percentages. Therefore, G-P fitting method was chosen in the present investigation for calculation of buildup factor for spin ice compounds. The computational work of the EBF for spin ice compounds is done in three steps as;

1. Calculation of equivalent atomic number [18,19]
2. Calculation of the G-P fitting parameters [18,19]
3. Calculation of the exposure buildup factors [10,11]

The buildup of photons is mainly due to multiple scattering events by Compton scattering, so that equivalent atomic number, $Z_{eq}$ is derived from the Compton scattering interaction process. The $Z_{eq}$ for individual compound is estimated by the ratio of ($\mu/\rho$)$_{Compton}$ / ($\mu/\rho$)$_{Total}$ at a specific energy with the corresponding of an element at same energy. Thus first the Compton partial mass attenuation coefficient, ($\mu/\rho$)$_{Compton}$ and the total mass attenuation coefficients, ($\mu/\rho$)$_{Total}$ are obtained using WinXcom program [20]. The $Z_{eq}$ for a compound is calculated by logarithmic interpolation method [18,19] using formula as;

$$Z_{eq} = \frac{Z_1 (\log R_2 - \log R) + Z_2 (\log R - \log R_1)}{\log R_2 - \log R_1}$$  \hspace{1cm} (1)$$

where $Z_1$ and $Z_2$ are the atomic numbers for elements corresponding to the ratios $R_2$ and $R_1$ respectively and R is the ratio for the compound at a specific energy. The G-P fitting parameters are calculated in a similar fashion of logarithmic interpolation procedure for $Z_{eq}$.

Finally buildup factors are calculated using G-P fitting parameters (b, c, a, $X_k$ and d) in the following equations given below;

$$B(E,x) = 1 + \frac{(b-1)(K^x-1)}{K-1} \quad \text{for} \quad K \neq 1$$  \hspace{1cm} (2)$$
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Table 1: G-P fitting parameters for exposure buildup factors and equivalent atomic numbers for Dysprosium Titanate.

| E (MeV) | b     | c     | a     | $X_b$  | d      | $Z_{eq}$ |
|---------|-------|-------|-------|--------|--------|----------|
| 0.015   | 1.0038| 0.4726| 0.1412| 17.5594| -0.0853| 54.0117  |
| 0.02    | 1.0051| 0.4059| 0.1785| 16.1795| -0.0633| 54.6299  |
| 0.03    | 1.1389| 0.3922| 0.2006| 12.9133| -0.0586| 29.4912  |
| 0.04    | 1.2557| 0.3294| 0.2279| 15.1075| -0.0915| 29.8026  |
| 0.05    | 1.2701| 0.3222| 0.1883| 12.7991| -0.1215| 30.0687  |
| 0.06    | 2.5696| 0.4150| 0.0431| 8.8073 | -0.0879| 52.2446  |
| 0.08    | 1.9233| 0.1107| 0.3618| 15.8968| -0.0474| 53.2488  |
| 0.1     | 1.4373| 0.0465| 0.6729| 14.1635| -0.2353| 53.7892  |
| 0.15    | 1.2024| 0.2662| 0.3335| 13.8105| -0.1891| 54.5460  |
| 0.2     | 1.2126| 0.4639| 0.1883| 14.3167| -0.1009| 54.9626  |
| 0.3     | 1.3285| 0.5942| 0.1257| 13.9224| -0.0597| 55.4439  |
| 0.4     | 1.4359| 0.7260| 0.0832| 14.1235| -0.0491| 55.9198  |
| 0.5     | 1.5062| 0.8131| 0.0575| 14.0940| -0.0392| 55.8980  |
| 0.6     | 1.5487| 0.8743| 0.0388| 13.8378| -0.0297| 56.0205  |
| 0.8     | 1.5990| 0.9415| 0.0213| 13.7096| -0.0228| 56.1452  |
| 1       | 1.6109| 0.9799| 0.0126| 13.2603| -0.0216| 56.2063  |
| 1.5     | 1.5461| 1.0847| -0.0120| 13.8071| -0.0092| 55.1397  |
| 2       | 1.5550| 1.0858| -0.0097| 13.0930| -0.0135| 52.3293  |
| 3       | 1.5318| 1.0598| 0.0030| 12.9511| -0.0319| 47.7906  |
| 4       | 1.4884| 1.0287| 0.0160| 13.3932| -0.0438| 45.6202  |
| 5       | 1.5095| 0.9469| 0.0460| 13.6227| -0.0711| 44.4616  |
| 6       | 1.4927| 0.9239| 0.0575| 13.8438| -0.0807| 43.7796  |
| 8       | 1.5303| 0.8823| 0.0795| 14.1512| -0.0985| 42.9015  |
| 10      | 1.5021| 0.9687| 0.0584| 14.2084| -0.0780| 42.3980  |
| 15      | 1.5731| 1.1185| 0.0349| 14.1585| -0.0599| 42.0452  |

\[
B(E, x) = 1 + (b - 1)x \quad \text{for} \quad K = 1, \quad \text{(3)}
\]

where,

\[
K(E, x) = cx^a + d\frac{\tanh(x / X_b - 2) - \tanh(-2)}{1 - \tanh(-2)}, \quad \text{(4)}
\]

for penetration depth (X) \leq 40 mfp
| E (MeV) | b     | c     | a     | X_k   | d      | Z_{eq} |
|--------|-------|-------|-------|-------|--------|--------|
| 0.015  | 1.0037| 0.4780| 0.1387| 16.4327| -0.0765| 54.5644|
| 0.02   | 1.0054| 0.4071| 0.1775| 16.3207| -0.0628| 54.9871|
| 0.03   | 3.1580| 0.9567| 0.1267| 28.4222| -0.1869| 39.2444|
| 0.04   | 3.4746| 0.3233| 0.1123| 21.9350| -0.0424| 39.5706|
| 0.05   | 2.8139| 0.0836| -0.2359| 12.0947| 0.0273 | 39.8069|
| 0.06   | 2.2308| 0.5783| 0.1072| 12.2439| -0.1169| 54.3571|
| 0.08   | 1.9811| 0.1576| 0.1367| 16.1207| 0.0053 | 54.9555|
| 0.1    | 1.4526| 0.0430| 0.6674| 14.3022| -0.2031| 55.2681|
| 0.15   | 1.1984| 0.2518| 0.3466| 13.7769| -0.1972| 55.7035|
| 0.2    | 1.2035| 0.4571| 0.1916| 14.2598| -0.1026| 55.9456|
| 0.3    | 1.3180| 0.5869| 0.1284| 13.8802| -0.0610| 56.2361|
| 0.4    | 1.4252| 0.7190| 0.0852| 14.1216| -0.0499| 56.4081|
| 0.5    | 1.4959| 0.8074| 0.0589| 14.0974| -0.0396| 56.5187|
| 0.6    | 1.5400| 0.8688| 0.0401| 13.8213| -0.0301| 56.6006|
| 0.8    | 1.5921| 0.9368| 0.0224| 13.6786| -0.0232| 56.6786|
| 1      | 1.6051| 0.9761| 0.0135| 13.2471| -0.0220| 56.7216|
| 1.5    | 1.5421| 1.0784| -0.0105| 13.7239| -0.0100| 56.0583|
| 2      | 1.5541| 1.0670| -0.0045| 13.0872| -0.0175| 54.3747|
| 3      | 1.5284| 1.0448| 0.0080| 13.0977| -0.0362| 51.7961|
| 4      | 1.4736| 1.0305| 0.0174| 13.4513| -0.0460| 50.4516|
| 5      | 1.4911| 0.9564| 0.0460| 13.6863| -0.0728| 49.6781|
| 6      | 1.4806| 0.9319| 0.0588| 13.9112| -0.0838| 49.2358|
| 8      | 1.5259| 0.9077| 0.0766| 14.1585| -0.0983| 48.6869|
| 10     | 1.5036| 1.0201| 0.0501| 14.1857| -0.0724| 48.3721|
| 15     | 1.5879| 1.1877| 0.0280| 13.9642| -0.0573| 48.1449|

where x is the source-detector distance for the medium in terms of mfp and b, the value of the exposure buildup factor at 1 mfp, K (E, x) is the dose multiplicative factor, and b, c, a, X_k and d are computed G-P fitting parameters which depends on the attenuating medium and source energy. The equation (4) represents the dependency of K on x; a, c, d and X_k are dependent upon...
Table 3: G-P fitting parameters for exposure buildup factors and equivalent atomic numbers for Holmium Titanate.

| E (MeV) | Holmium Titanate |   |   |   |   |
|---------|------------------|--|--|--|--|--|
|         | b    | c    | a    | X_h | d    | Zeq |
| 0.015   | 1.0036 | 0.4806 | 0.1375 | 15.8837 | -0.0722 | 54.8357 |
| 0.02    | 1.0057 | 0.4088 | 0.1762 | 16.5083 | -0.0622 | 55.4650 |
| 0.03    | 1.2565 | 0.4251 | 0.1963 | 13.8163 | -0.0660 | 29.9850 |
| 0.04    | 1.3881 | 0.3290 | 0.2210 | 15.5150 | -0.0885 | 30.3112 |
| 0.05    | 1.3644 | 0.3076 | 0.1624 | 12.7561 | -0.1124 | 30.5883 |
| 0.06    | 2.4216 | 0.4864 | 0.0711 | 10.3089 | -0.1006 | 53.1574 |
| 0.08    | 1.9521 | 0.1341 | 0.2497 | 16.0083 | -0.0212 | 54.0915 |
| 0.1     | 1.4461 | 0.0445 | 0.6697 | 14.2439 | -0.2166 | 54.6413 |
| 0.15    | 1.1994 | 0.2553 | 0.3434 | 13.7851 | -0.1952 | 55.4176 |
| 0.2     | 1.2044 | 0.4578 | 0.1913 | 14.2652 | -0.1024 | 55.8517 |
| 0.3     | 1.3160 | 0.5855 | 0.1289 | 13.8722 | -0.0613 | 56.3876 |
| 0.4     | 1.4206 | 0.7160 | 0.0861 | 14.1208 | -0.0502 | 56.7026 |
| 0.5     | 1.4894 | 0.8038 | 0.0598 | 14.0995 | -0.0398 | 56.9151 |
| 0.6     | 1.5333 | 0.8646 | 0.0411 | 13.8087 | -0.0304 | 57.0463 |
| 0.8     | 1.5859 | 0.9326 | 0.0233 | 13.6505 | -0.0235 | 57.1653 |
| 1       | 1.5996 | 0.9724 | 0.0144 | 13.2345 | -0.0224 | 57.2159 |
| 1.5     | 1.5416 | 1.0776 | -0.0103 | 13.7136 | -0.0101 | 56.1731 |
| 2       | 1.5545 | 1.0767 | -0.0072 | 13.0902 | -0.0154 | 53.3056 |
| 3       | 1.5280 | 1.0590 | 0.0034 | 12.9681 | -0.0324 | 48.7426 |
| 4       | 1.4847 | 1.0299 | 0.0160 | 13.4005 | -0.0440 | 46.4614 |
| 5       | 1.5065 | 0.9484 | 0.0460 | 13.6330 | -0.0714 | 45.2662 |
| 6       | 1.4909 | 0.9251 | 0.0577 | 13.8539 | -0.0812 | 44.5589 |
| 8       | 1.5297 | 0.8858 | 0.0791 | 14.1522 | -0.0985 | 43.6622 |
| 10      | 1.5023 | 0.9755 | 0.0573 | 14.2054 | -0.0773 | 43.1490 |
| 15      | 1.5751 | 1.1276 | 0.0340 | 14.1329 | -0.0595 | 42.8001 |

attenuating medium and source energy, E. The $Z_{eq}$ and G-P fitting parameters for the spin ice compounds are given in Table 1 to 4.
Table 4: G-P fitting parameters for exposure buildup factors and equivalent atomic numbers for Holmium Stannate.

| E (MeV) | b     | c     | a     | $X_k$  | d      | $Z_{eq}$ |
|--------|-------|-------|-------|--------|--------|----------|
| 0.015  | 1.0036| 0.4789| 0.1383| 16.2521| -0.0751| 54.6535  |
| 0.02   | 1.0054| 0.4074| 0.1773| 16.3543| -0.0627| 55.0724  |
| 0.03   | 3.1554| 0.9560| 0.1268| 28.4026| -0.1868| 39.2103  |
| 0.04   | 3.4713| 0.3233| 0.1125| 21.9249| -0.0425| 39.5539  |
| 0.05   | 2.8114| 0.0840| -0.2352| 12.0958| 0.0271 | 39.7889  |
| 0.06   | 2.2176| 0.5847| 0.1097| 12.3777| -0.1180| 54.4411  |
| 0.08   | 1.9838| 0.1598| 0.1261| 16.1313| 0.0078 | 55.0371  |
| 0.1    | 1.4534| 0.0428| 0.6671| 14.3096| -0.2013| 55.3483  |
| 0.15   | 1.1981| 0.2508| 0.3475| 13.7746| -0.1977| 55.7821  |
| 0.2    | 1.2028| 0.4566| 0.1918| 14.2553| -0.1027| 56.0244  |
| 0.3    | 1.3169| 0.5862| 0.1287| 13.8759| -0.0612| 56.3170  |
| 0.4    | 1.4239| 0.7181| 0.0855| 14.1214| -0.0499| 56.4910  |
| 0.5    | 1.4945| 0.8066| 0.0591| 14.0979| -0.0397| 56.6030  |
| 0.6    | 1.5387| 0.8680| 0.0403| 13.8189| -0.0302| 56.6860  |
| 0.8    | 1.5910| 0.9360| 0.0225| 13.6736| -0.0232| 56.7650  |
| 1      | 1.6041| 0.9754| 0.0137| 13.2449| -0.0220| 56.8087  |
| 1.5    | 1.5417| 1.0778| -0.0104| 13.7159| -0.0101| 56.1471  |
| 2      | 1.5540| 1.0662| -0.0042| 13.0869| -0.0177| 54.4696  |
| 3      | 1.5287| 1.0440| 0.0083| 13.1039| -0.0364| 51.9010  |
| 4      | 1.4745| 1.0294| 0.0178| 13.4565| -0.0463| 50.5636  |
| 5      | 1.4907| 0.9566| 0.0460| 13.6876| -0.0728| 49.7889  |
| 6      | 1.4804| 0.9321| 0.0588| 13.9125| -0.0839| 49.3469  |
| 8      | 1.5258| 0.9081| 0.0766| 14.1586| -0.0983| 48.7986  |
| 10     | 1.5036| 1.0210| 0.0499| 14.1853| -0.0723| 48.4838  |
| 15     | 1.5881| 1.1889| 0.0278| 13.9609| -0.0573| 48.2556  |

3. RESULTS AND DISCUSSION

Variation of exposure buildup factor, EBF for the spin ice compounds with photon energy (0.015 to 15 MeV) for selected penetration depths 1, 5, 10, 20 and 40 mfp is shown in Fig. 1 (a-d). The EBF for the compounds are small in
low photon energy region which can be explained by photon interaction cross section dependency on photon energy and atomic number for the elements. The Z_{eq} for a compound plays the similar role as Z for an element.

The EBF for the compounds are the minimum in low-energy due to dominance of photoelectric effect where interaction cross section is proportional to Z_{eq}^{4-5}/E^{1.5}. The large values of Z_{eq} of the compounds reduce the buildup factors. With increase in photon energy, EBF increases due to multiple scattering as Compton scattering dominates. The pair production takes over the Compton scattering process for photon energy equal or above 1.022 MeV. In the pair production, interaction cross section is proportional to Z_{eq}^{2}, so low-Z_{eq} shows lowest EBF in the pair production region. This type of high buildup factors for the elements, compounds and mixtures have been reported [21-24].

It is also noted that the compounds containing tin (Dy\_2Sn\_2O\_7 and Ho\_2Sn\_2O\_7) show peaks at photon energy 30 keV (see Fig.1 (b) and (d)). To explain this, we have plotted the EBF for tin [9] at different penetration depths (1, 5, 10, 20 and 40 mfp) in Fig.2. From Fig.2, it is observed that the sharp peak in EBF at 40 mfp for tin (2.88×10^{13}) is at energy 30 keV analogous to Dy\_2Sn\_2O\_7 (3.34×10^{7}) and Ho\_2Sn\_2O\_7 (3.29×10^{7}). The height of peaks in the spin ice compounds changes due to other elements. Therefore, it is concluded that peaks in EBF in low-energy are due to presence of the tin element.

Figure 1 (a-d): Variation of exposure buildup factors for spin ice compounds with photon energy for penetration depths 1, 5, 10, 20 and 40 mfp.
Figure 2: Exposure buildup factors for tin at penetration depth 1, 5, 10, 20 and 40 mfp.

Figure 3: (a-d): Variation of exposure buildup factors for spin ice compounds with penetration depths for selected photon energies.

Variation of EBF for the spin ice compounds for penetration depth (0-40 mfp) is shown in Fig. 3 (a-d). It is to be noted that the EBF for the compounds increase with photon energy and penetration depth. However, in low-energies
the EBF are found to be constant. The EBF for the selected compounds are found to be unity at 0.015 MeV photon energy. As photon energy increases, the EBF increases and become linear at high energy (15 MeV). The EBF for the compounds containing tin are found to be the largest. This type of behavior of buildup factor for superconductors has been reported [24].

4. CONCLUSIONS

In the present study, we studied exposure buildup factors for some spin ice compounds Dysprosium Titanate, Dysprosium Stannate, Holmium Titanate and Holmium Stannate useful in nuclear engineering for photon energy range 0.015 to 15 MeV. The EBF for the compounds containing tin were found to be the largest as well as shown a peak at 30 keV photon energy. This study could be very useful for shielding evaluation of this and similar type of spin ice compounds.

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