

Optimization of the Alpha Energy Deposited in Radioluminescence Thin Film for Alphaphotovoltaic Application

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Abstract. Activated zinc sulfide (ZnS) is a semiconductor material which able to emit photon in the form of visible light when expose to external energy. The capability of activated ZnS, mainly doped with silver (Ag) and copper (Cu), to convert radiation become light to make it potentially applicable as the radioluminescent thin film for alphaphotovoltaic-type nuclear battery. One of the important specifications of the radioluminescence layer that influences the fluorescence efficiency is the thickness. This work presents a study on the thickness optimization for ZnS:Ag:Cu as the radioluminescent film for alpha particles using Monte Carlo model. Simulation to study alpha particles' energy deposited by using Stopping and Range of Ions in Matter/TRansport of Ions in Matter (SRIM/TRIM) code. The model examined the transport of 5.485 MeV alpha particles emitted by ²⁴¹Am to determine the best thickness based on energy deposition depth. Based on TRIM module simulation, the optimal thickness for radioluminescence film is approximately 19-22 μm . Most energy from 5.485 MeV alpha particles is deposited in 18.92 μm depth activated zinc sulfide. The results from SRIM/TRIM model then compare with analytical calculation using Bragg-Kleman rule. The alpha particles stop at 22 μm from the SRIM/TRIM simulation while using Bragg-Kleman formula the alpha particles stop at 23.51 μm .

Keywords: Alphaphotovoltaic, Radioluminescence, SRIM/TRIM, ZnS:Ag:Cu

Abbreviations: Stopping and Range of Ions in Matter (SRIM), TRansport of Ions in Matter (TRIM), zinc sulfide (ZnS)

INTRODUCTION

Zinc sulfide (ZnS) is a direct gap II-VI intrinsic semiconductor material which abundant and environmental friendly. ZnS material has a wide band gap of 3.66 eV, good chemical stability, high light transmittance and a low dispersion in the visible and infrared region. An extensive research effort about ZnS material has developed rapidly due to its potential utilization for numerous fields such as in cathode ray tube, semiconductor light-emitting diode, and bio light-emitting diode. In addition, ZnS material is suitable for use as the radioluminescence layer in an alphaphotovoltaic indirect-conversion nuclear battery (Russo et al., 2017).

Nuclear battery has potential for use as the substitute of conventional battery due to radioisotopes possess theoretical energy densities 1000 times greater than chemical battery, longer operation time, and durability in extreme environments. Nuclear battery with power scale from μW until mW offers promising alternative as power supply for sensor and low power electronic devices. Nuclear battery can supply sufficient and long-lived power for various applications such as mobile sensor platforms, military and remote electronic devices.

Radioluminescence layer in an alphaphotovoltaic indirect-conversion nuclear battery absorbs alpha particles emitted from the radioisotope source and convert the radiation in the form of visible light. The visible light is absorbed and converted into electricity

by photovoltaic which is placed adjacent to the radioluminescence layer. The characteristic of the radioluminescence layer plays important role to absorb the energy from radiation and transport the emitted luminescence produce by the radioluminescence layer. Appropriate thickness of the radioluminescence layer is needed to optimize radiation absorption into luminescence. Design proper structure of phosphor or radioluminescence layer may increase the luminescence efficiency both production luminescence and transmission the emitted luminescence in the material (Xu et al., 2014).

Xu et al. (2014) have researched about the effect of phosphor layer structure to radioluminescence intensity. The research is examined phosphor layer with different thickness and measured the radioluminescence intensity by using Cary Eclipse fluorescence spectrophotometer (Agilent Technologies, USA) with 4.93 mCi/cm^2 ⁶³Ni and 2.88 mCi/cm^2 ¹⁴⁷Pm beta particle sources. Based on the radioluminescence measurements, it is apparent that radioluminescence intensity has correlation with the phosphor structure. The phosphor structure has influence on RL intensity in two factors: absorption of the radiation and transmission of the luminescence in the phosphor layer. If the phosphor layer is thicker than the penetration depth of the radiation, the negative effect of luminescence self-absorption in the phosphor layer was bigger than the increasing of energy deposited. On the contrary, only a few energy from alpha particles will be deposited in the phosphor layer if the layer is thinner

than the penetration depth. Thus, research about the optimal layer thickness for indirect-conversion nuclear battery is important to do.

Stopping and Range of Ions in Matter/TRansport of Ions in Matter (SRIM/TRIM) code were used to determine optimum thickness based on energy deposition depth by alpha particles in the ZnS material. The model examined the radiation of 5.485 MeV alpha particles emitted by Americium-241 (^{241}Am). Energy deposition in 22 μm thickness of ZnS:Ag:Cu was calculated for slab geometry using SRIM/TRIM. These geometry is the most possible geometry deployed for alphaphotovoltaic indirect-conversion nuclear battery (Oh, 2011).

The results from software simulation are compared to theoretical calculation using Bragg-Kleman rule. Formula from the Bragg-Kleman rule can be found in Knoll (1989). This formula provides information about the range of particles in a material without energy loss data. Bragg-Kleman rule calculated the range of certain particles in a material using range information of the particles in another material. Bragg-Kleman rule is shown as follows:

$$\frac{R_1}{R_2} = \frac{\rho_2}{\rho_1} \sqrt{\frac{A_1}{A_2}} \quad (1)$$

Where ρ is the density of the material, A is the atomic weight or effective atomic weight if it is a compound material, R is the particles' range. Theoretical calculation about particles' range in a certain material using Bragg-Kleman law exceeds two-step process:

1. Determine the range in air;

$$R (\text{mm}) = (0,05T + 2,85) T^{\frac{3}{2}} \quad 4 < T \leq 15 \text{ MeV} \quad (2)$$

Symbol R denoting particles' range in the air, whereas T shows particles' energy emitted by the source.

2. The data obtained from process number 1 is used to determine the range in the material by using Bragg-Kleman rule.

MATERIALS AND METHODS

This research is conducted by using theoretical computation and analytical calculation. Initial literature study is done to find characteristic data of alpha radiation source Americium-241 (^{241}Am) and information about radioluminescence material ZnS:Ag:Cu. The selection of ^{241}Am as isotope to use in a nuclear battery based on its long half-life, high purity and energy density, short endpoint energy. Research about nuclear battery is focused on alpha and beta radiation due to their relatively short range, thus their application would not require large-scale configuration.

Simulation and Analytical Calculation

Sets of personal computer with Windows 10 operating system and Core i3-2328M 2.20 GHz is used to run software SRIM 2013. Actually, there are few codes suitable for modeling alpha particle interactions. The code chosen in this study which treat alpha particle interaction with matter is SRIM/TRIM 2013. SRIM/TRIM 2013 demonstrates mono-directional and mono-energetic alpha particles interact with matter in only the slab geometry.

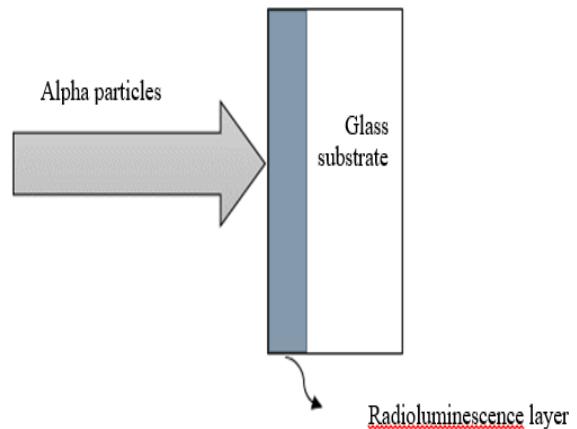


Figure 1. Slab model used for SRIM/TRIM simulation. A mono-directional and mono-energetic alpha beam impinges on the slab target.

Results obtained from SRIM/TRIM simulation are compared to analytical calculation using Bragg-Kleman rule. Alpha particle range in ZnS material is calculated using Equation (1) and (2).

RESULTS AND DISCUSSION

SRIM/TRIM Simulation

Simulation is done using 99,999 ions for increasing accuracy of the calculation. The accuracy of calculation is determined by the number of ions followed. Typically, a calculation for 1000 ions will give better than 10% accuracy (SRIM, 2010). Thus, simulation with 99,999 ions can achieve the accuracy for more than 90%.

Figure 2 illustrates the tracks of alpha particles within the slab and demonstrates their relatively nearly linear paths. When a charged particle penetrates in matter, in this case is alpha particle which has positive charged, a charged particle will interact with the electrons and nuclei present in the material through the electromagnetic force. For particles with energy more than 1 MeV, the charge particles will mostly interact with the electrons present in the material. However there are some of the ions collide with the nucleus which make the direction change. Those interactions are showed in Figure 2, the scattered ions are the ions collide with the nucleus in the ZnS material.

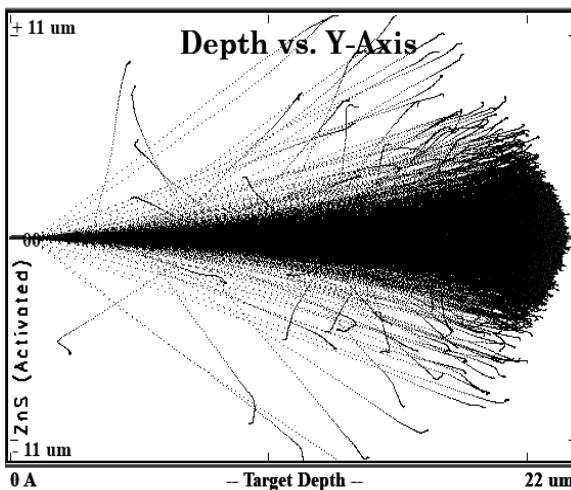


Figure 2. Illustration the tracks of Am-241 alpha particles within the ZnS slab using SRIM/TRIM.

Figure 3 shows the alpha particle's Bragg curve in the ZnS target. The SRIM/TRIM result from the ZnS slab model shows that the peak of the Bragg curve occurs at around 18.92 μm. The energy loss maximum of charged particles near the end of the range called as "Bragg peak". Beyond the Bragg peak, the energy deposition drops sharply. This is because alpha particles have lost almost all of their energy when they traveled at the range 18.92 μm. As the particle penetrates in the medium, its energy loss per unit length will change. The energy loss of a particle as a function of its distance of penetration is shown in Table 1. The Bragg curve shows that alpha particles have lost all of their energy in the ZnS material at around 22 μm.

Energy deposition depth of alpha particles is shown in Table 1. The percentage of energy deposited can be calculated by adding up all the energy loss from alpha particles when penetrates in matter. In addition, Table 1 shows that the maximum energy deposition occurs between 18 to 19 μm from the source. SRIM/TRIM predicts the percentage of energy deposited in this layer is ~ 7.24 % for slab configuration.

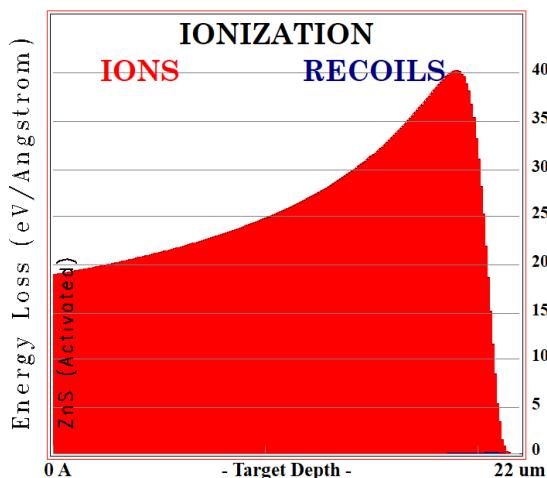


Figure 3. The alpha particle's Bragg curves at in ZnS target using SRIM/TRIM.

When a charged particle penetrates in matter, it will loss its energy by transferring a small amount of energy to a large number of electrons along its trajectory. Based on Table 1, alpha radiation deposited 99.74% of their energy into the atom target. The main interaction which cause the alpha particles loss their energy is excitation and electron ionization. The type of energy loss due to interaction of the charged particle with electrons is often called as "energy loss due to ionization". When a charged particle has sufficient energy to excite or ionize atoms in the medium, it will deposit several eV of its energy until it lost all the kinetic energy and come to rest. The term "energy loss due to ionization" stated all the energy loss from both ionization interaction and electron excitation, since many atoms are only brought to an excite state.

Alpha particles mostly deposited all of their energy between 18 until 19 μm in the ZnS target. The percentage of energy deposited in this layer is ~ 7.24 % which is the Bragg peak of the curve. Beyond this distance, the energy deposited drops sharply from ~6.69% at the range 19 until 20 μm; 2,56% at the range 20 until 21 μm; 0,04% at the range 21 until 22 μm. Based on SRIM/TRIM simulation we can conclude that theoretical thickness needed for radioluminescence layer can optimally convert all the radiation energy from ²⁴¹Am is approximately 19 until 22 μm.

Table 1. SRIM/TRIM calculations for predicting energy deposition in ZnS target for slab model.

Range (μm)	Energy (keV)	% deposited
0-1	189,8	3,46
1-2	193,9	3,54
2-3	198,3	3,62
3-4	203,2	3,70
4-5	208,5	3,80
5-6	214,2	3,91
6-7	220,4	4,02
7-8	227,4	4,15
8-9	234,9	4,28
9-10	243,3	4,44
10-11	252,5	4,60
11-12	263,3	4,80
12-13	275,2	5,02
13-14	289,2	5,27
14-15	305,4	5,57
15-16	324,5	5,92
16-17	347,1	6,33
17-18	373,1	6,80
18-19	397,1	7,24
19-20	366,9	6,69
20-21	140,6	2,56
21-22	2,3	0,04
Total	5471	99,74

Theoretical Calculation

The range of alpha particles in air with normal temperature and pressure in the energy range $4 < E < 15$ MeV is calculated by using an empirical equation:

$R \text{ (mm)} = (0.05T + 2.85)T^{3/2}$ $4 < T \leq 15 \text{ MeV}$
 Alpha particles emitted from ^{241}Am decay process have energy of 5.485 MeV.

$$R \text{ (mm)} = (0.05 \times 5.485 + 2.85) 5.485^{3/2}$$

$$R \text{ (mm)} = 40,134 \text{ mm}$$

Bragg-Kleman rule to compute the range of alpha particles in any material is using the following equation:

$$R_1 / R_2 = (\rho_1 / \rho_2) (A_1 / A_2)^{1/2}$$

Because ZnS is in the form of compound material, so that atomic weight A_i is calculated using effective atomic weight A_{ef} :

$$\sqrt{A_{ef}} = \left(\sum_{i=1}^n \frac{w_i}{\sqrt{A_i}} \right)$$

Air is consisted by 20% oxygen and 80% nitrogen, so that for air:

$$w_O = \frac{0.2 \times M_O}{0.2 \times M_O + 0.8 \times M_N} = \frac{0.2 \times 16}{0.2 \times 16 + 0.8 \times 14} = 0.222$$

$$w_N = 1 - 0.222 = 0.778$$

$$\sqrt{A_{ef}} = \left(\frac{w_O}{\sqrt{A_O}} + \frac{w_N}{\sqrt{A_N}} \right)^{-1} = \left(\frac{0.222}{\sqrt{16}} + \frac{0.778}{\sqrt{14}} \right)^{-1} = 3.796$$

$$A_{ef,air} = (3.796)^2 = 14.41$$

For ZnS material is calculated by using the following formula:

$$w_{Zn} = \frac{M_{Zn}}{M_{Zn} + M_S} = \frac{65}{65 + 32} = 0.67$$

$$w_N = 1 - 0.67 = 0.33$$

$$\sqrt{A_{ef}} = \left(\frac{w_{Zn}}{\sqrt{A_{Zn}}} + \frac{w_S}{\sqrt{A_S}} \right)^{-1} = \left(\frac{0.67}{\sqrt{65}} + \frac{0.33}{\sqrt{32}} \right)^{-1} = 7.07$$

$$A_{ef,ZnS} = (7.07)^2 = 49.99$$

Bragg-Kleman rule:

$$R_{ZnS} = (\rho_{ZnS} / \rho_{udara}) (A_{ZnS} / A_{udara})^{1/2} = (0.00129 / 4.1) \times (49.99 / 14.41)^{1/2} = 5.86 \times 10^{-4}$$

$$R_{ZnS} = 5.86 \times 10^{-4} \times R_{udara}$$

$$R_{ZnS} = 5.86 \times 10^{-4} \times 40.134 = 23.51 \text{ } \mu\text{m}$$

The range of alpha particles have energy of 5.485 MeV in ZnS material is 23.51 μm .

CONCLUSIONS

Based on the study it can concluded the SRIM/TRIM result from the ZnS slab model shows that the peak of the Bragg curve occurs at around 18.92 μm . The Bragg curve shows that alpha particles have lost all of their energy in the ZnS material at around 22 μm . Alpha radiation deposited 99.74% of their energy into the atom target. The main interaction which cause the alpha particles loss their energy is excitation and electron ionization.

Alpha particles mostly deposited all of their energy between 18 until 19 μm in the ZnS target. The percentage of energy deposited in this layer is $\sim 7.24\%$ which is the Bragg peak of the curve. Beyond this distance, the energy deposited drops sharply from $\sim 6.69\%$ at the range 19 until 20 μm ; 2.56% at the range 20 until 21 μm ; 0.04% at the range 21 until 22 μm . Based on SRIM/TRIM simulation we can conclude that theoretical thickness needed for radioluminescence layer can optimally convert all the radiation energy from ^{241}Am is approximately 19 until 22 μm . The alpha particles stop at 22 μm from the SRIM/TRIM simulation while using Bragg-Kleman formula the alpha particles stop at 23.51 μm .

ACKNOWLEDGEMENTS

Author would like to gratefully thank PSTA BATAN Yogyakarta and PT Ensterna for the opportunity of doing research in the institution.

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