The investigation of self-absorption effect in cylindrical Ni-63/4H-SiC betavoltaic

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Abstract. The limiting efficiency of betavoltaic battery can be affected by the self-absorption of radioisotope source. Moreover, this effect can be more detrimental for a low energy beta emitter such as Ni-63. The cylindrical design is known to have a benefit over rectangular one in response to the self-absorption effect. In this study, the model of betavoltaic was built under MCNPX which also capable to do many particle simulation based on Monte Carlo method. The interaction of beta particle and its average energy deposition in 4H-SiC semiconductor as the radiation-electricity converter was calculated, along with photonic emission due to the bremsstrahlung event. Basically, the average energy deposition shows a deterioration along with the addition of source mass thickness. However, an optimum peak was observed at about 0.1424 mg/cm² source mass thickness in the N⁺ substrate average beta energy deposition. The cylindrical surface magnification also give a positive influence to the energy deposition. In addition, the absorbed dose in each layers of semiconductor was also investigated, and the results were consistent to the semi-empirical equation of self-absorption theory.

1. Introduction

Nuclear battery is a device that generate electricity using radioactive decay. Instead of using the fission chain reaction like nuclear reactor, nuclear battery converts the radioactive decay emissions to electricity by implementing a specific mechanism. One of the simplest form of nuclear battery is coupling a beta emitting source with a photovoltaic, which is widely known as betavoltaic. The process is nearly similar to photovoltaic, but it employs a radionuclide which undergoes a beta decay as the main source of ion pair generation. The unique feature of betavoltaic battery is a reasonably high energy density in a small package and the longevity. Hence, it is an attractive power source to be used particularly for a device that required a long-term micro electricity power, such as MEMS, sensors, and explorations.

Ni-63 has been extensively studied as it is one of the most suitable radioisotope source due to its pure beta emission and the longevity of its half-life (approximately 100 year) [1]. Soft beta sources such as Ni-63 can neglect the energy loss caused by radiation, which has a maximum energy that is equal to 0.067 MeV and a deposition energy approximately equal to the energy loss by excitation and ionization. However, due to its low beta energy emission and the difficulty of beta particle to penetrate inside the material, it is known to be highly influenced by the self-absorption effect. The majority of this effect can occur in material with high density, according to the approximation of beta particle maximum penetration depth by Bethe’s formula. The design of beta source is a critical aspect since the overall efficiency of a betavoltaic at least consists of three main factors which is given by [2]:

\[ \eta = \frac{E_{\text{out}}}{E_{\text{in}}} \]
\[ \eta = \eta_{\beta} \eta_{\beta-el} \eta_{el} \]  

where \( \eta_{\beta} \) is the beta source emission efficiency, \( \eta_{\beta-el} \) is the beta-electricity conversion efficiency and \( \eta_{el} \) is the charge collection efficiency.

There have been several studies conducted in the past to analyze the scattering events in the betavoltaic battery. The Monte Carlo method is a well-known approach, a powerful tool to simulate the transport of any given particle in a material [3–5]. The Monte Carlo N-Particle code (MCNP) is often used to calculate the deposition energy of beta particle by analyzing the interaction process as soon as the particle is emitted from radioisotopes in semiconductor materials. Many previous simulation studies of betavoltaic battery have done a quite well result by implementing the MCNP to simulate beta particle interaction inside the material and further calculate the electrical performance using the semiconductor equation related to its structural design [6–9]. As previously studied by Kim et al., the cylindrical shape of Ni-63 beta source is able to emit a better fluence rather than the rectangular one [4]. However the silicon is known to have a lesser density compared to silicon carbide (SiC), which is the best candidate of energy conversion material due to its feature such as a wide band-gap, radiation resistant, thermal stability, etc. Hence, the beta particle penetration and its energy deposition should be different from previously demonstrated in silicon.

This study aims to investigate the effect of Ni-63 source geometrical variation with cylindrical shape to the beta energy deposited and the dose absorbed in each layers of semiconductor cell as it could directly influence the electrical performance. The design of SiC semiconductor layers was chosen from the previous experiment from Guo et al. [10]. Furthermore, we also calculated the photon energy generated by radiative collision which is responsible for additional scattering events in the cell. The beta particle interaction was simulated using MCNPX, which is emitted from the source corresponds to the beta spectra of Ni-63.

2. Methods of simulation

There are two major processes which cause the degradation of kinetic energy to the beta particle along its penetration: (1) the electron-electron collision, and (2) the electron-nuclei collision. Compared to the electron-nuclei collision, the electron-electron collision is more favored since the probability of entering the nuclei is very low according to the scattering theory. The ratio between the orbital collision and radiative collision can be approximated using this equation[12]:

\[ \frac{S_{rad}}{S_{col}} \approx \frac{ZE}{800} \]

where \( S_{rad} \) and \( S_{col} \) is the radiative and orbital collision stopping power, \( Z \) is the atomic number of the absorber and \( T \) is the kinetic energy of incident electron (in MeV). For a low-energy electron like the beta particle emitted from Ni-63, which has the average kinetic energy about 0.017 MeV and the maximum of 0.067 MeV, the electron-electron collision becomes a dominant mechanism of energy loss. However, some of the energy loss by bremsstrahlung mechanism could generate several amounts of electromagnetic radiation. The radiation yields from bremsstrahlung emission for a monoenergetic electrons until its energy is depleted can be approximated with this formula:

\[ Y \approx \frac{6 \times 10^{-4}ZE}{1 + 6 \times 10^{-4}ZE} \]

For an incident beta particle with kinetic energy of 0.067 MeV and penetrates to the SiC, only a portion of 0.16% from its energy is converted to bremsstrahlung radiation until it completely stops. Still, an approximate of 100 eV photonic emission is also responsible for additional electron-hole pair (EHP) generation when it interacts with the atomic orbital.
Since the radioactive source emits the particle isotropically, there will be a variation of beta fluence and power distribution due to the surface interface. The power distribution over the surface parameter of a think layer source is given by [2]:

\[
\frac{dP_B}{dS} = \frac{A_m P_{sp}}{2 \mu_m} \int z \left[ 1 - \exp\left(\frac{\mu_m P_{sp} D_L}{z}\right)\right] dz
\]

where \( A_m \) is the specific activity of source in Bq/g, \( P_{sp} \) is the beta spectra distribution, \( \mu_m \) is the mass absorption coefficient, \( D_L \) is the thickness of source containing the radioactive material, and \( z \) is the space integration parameters. According to Fermi’s theory of beta decay, the beta emission energy extends from zero to a maximum value. The beta spectra of Ni-63 decay can be seen in Fig. 1, which is used to define the source in MCNPX.

![Figure 1](image)

**Figure 1.** The beta spectra of Ni-63 decays to Cu-63.

The self-absorption effect of Ni-63 can be directly investigated by calculating the beta energy deposited inside each layers of semiconductor used in betavoltaic. MCNPX allows the user to simulate the interaction of beta particle as it travels the medium for a given number of history and takes the average values of energy deposition related to its trajectory. The tally function \( F_6 \) can be used to determine the energy deposition of a specific particle averaged in a cell, which is given by [13]:

\[
E_D = \int_{s_i}^{s_{i+1}} \left( -\frac{dE}{ds} \right)_{col} ds
\]

However, many reports never mentioned the design of its beta source, such as the geometrical aspects or the radioactive container material. It might cause a difference between the radioactivity and the actual particle fluence to the cell, particularly in a low beta emitter since the self-absorption is significant.

Some of the betavoltaic studies are mainly focused on the search of an efficient semiconductor material to convert the radiation energy to electricity. When charged energetic particles like beta radiation are exposed directly to the semiconductor crystals, eventually it forms the vacancies and interstitials. Moreover, a continuous irradiation could lead to a faster degradation, thus it is necessary to have not only an efficient converter cell, but also a radiation hardness. Many approaches like introducing a radioluminescence material or a scintillator wave guide have also been done to minimize a direct radiation exposure, although it is not cost effective for a mass production. Besides, the geometrical configuration or chemically engineered source by combining the radioactive material with non-radioactive material to have a desired phase could also be done to minimize the form factor.
Figure 2. The model of 4H-SiC semiconductor based on N+ substrate with different doping concentration to form N-epilayer and P+ epilayer.

The SiC is a wide band-gap semiconductor and it features a higher radiation tolerance than Si which has been studied by Ohshima et al. [14]. It is considered to be the best among semiconductors to be used in harsh environment, such as space exploration or the accelerator facilities. The variety of structural layers can be formed as a p-n junction [15], p-i-n diode[16], schottky barrier[17], etc. Hence, we tried to simulate the p-i-n junction betavoltaic cell using previous design from Guo et al. and study the energy deposition inside each layers of semiconductor both from the beta and photon emission point of view. Figure 2 shows the model of cylindrical betavoltaic structure with three different layers: (a) N+ substrate, (b) N-epilayer, and (c) P+ epilayer. Previous study for a p-n junction Si betavoltaic with Ni-63 source from Kim et al. [4] has shown that cylindrical shape could minimize the self-absorption effect, thus emitting a better particle fluence to the cell. However, the study does not include how much the energy deposited and the dose absorbed in each cells, since it only provides the information of the incident particle fluence when reaching the semiconductor layers. The physical properties of betavoltaic cell and the environment used for MCNPX simulation are listed in Table 1. Since the level of doping concentration from each layers is much lower than the SiC concentration in the volume, thus the atomic fraction of dopants can be neglected.

Table 1. The physical properties of the betavoltaic elements with the environment used in the material description in MCNPX.

| Materials | Density (g/cm³) | Composition         |
|-----------|----------------|---------------------|
| Ni-63     | 8.902          | 100% Ni-63 Isotope  |
| SiC       | 3.21           | 50% Natural Si      |
|           |                | 50% Natural C       |
| Ti        | 4.54           | 100% Natural Ti     |
| Ni        | 8.902          | 100% Natural Ni     |
| Al        | 2.698          | 100% Natural Al     |
| Au        | 19.32          | 100% Natural Au     |
| Air (dry) | 0.0012         | 0.015% Natural C    |
|           |                | 78.443% Natural N   |
|           |                | 21.075% Natural O   |
|           |                | 0.467% Natural Ar   |
3. Results and discussions
The Monte Carlo simulation was executed according to the betavoltaic model in Figure 2. The number of particle simulated was set to 50 million particles for each repetition to ensure the statistical error can still be tolerated, especially when calculating the low energy photon emission. From our simulation, the highest statistical error was obtained from the photonic energy deposition about 1.7%, which is still an acceptable results.

3.1. Beta energy deposition
Using tally F6 in MCNP could give the estimation of energy deposition for a specific particle averaged in a cell, including electron and photon. Subsequently, the averaged energy deposition in the respective cell is multiplied with the cell mass. Generally, the profile of energy deposition in each cell is degraded along with the addition of source thickness. The curves indicates that the relationship of source thickness and the energy deposition follows an exponential decay curve. It is proven that the self-absorption or self-scattering effect occurred during the simulation process and brought a negative impact to the energy deposition, since many of the beta particle is weakened by the source itself before entering the semiconductor. The area magnification does not give a significant influence to the energy deposition, although we can notice a slight improvement at the N+ substrate.

Figure 3. The average beta energy deposition inside the 4H-SiC PIN semiconductor: (a) N+ substrate, (b) N-epilayer, (c) P+ epilayer with different surface interface area.

However, the unique profile appears at the N+ substrate beta energy deposition, which shows a peak near 0.1424 mg/cm² source mass thickness. This indicates that the beta energy could not deposited energy effectively to further depth when the source is remarkably thin or thick. If the source is too thin,
the particle fluence emitted from the source is low, thus some of the beta particles were already scattered back or their trajectory is deflected by the bremsstrahlung process. As a consequence, only a few beta particle could reach the N+ substrate. Meanwhile, the self-absorption in the source is taking over if the source has a huge thickness. It is also indicate that the distribution angle of emitted particle is also influenced with the source thickness.

As the beta particle interacts with nuclei, the braking radiation would be produce in response to the loss of its kinetic energy. The photon produced by internal scheme is also responsible for generating additional ion-pairs inside semiconductor. Figure 4 (a)-(c) shows the average energy deposition for photon in different layers of semiconductor, which are generally increased as the addition of source thickness. The Ni-63 source not only emits beta particles but also generates the inevitable braking radiation. The energy fluctuation can be seen from each layer, but N and P+ epilayers took much of it as they both are thin layers compared to the bulk of N+ substrate. According to the Bethe’s approximation of electron maximum penetration range, the beta particle emitted from Ni-63 could only travel about 1.54 μm inside the SiC at its average energy and 20.76 μm at its maximum energy. Hence, many electrons have deposited all of their energy before entering the substrate and produced bremsstrahlung radiation mainly at the upper layers.

![Figure 4](image)

**Figure 4.** The average photon energy deposition inside the 4H-SiC PIN semiconductor: (a) N+ substrate, (b) N-epilayer, (c) P+ epilayer with different surface interface area.

The bremsstrahlung radiation could also be produced by the secondary electron [18]. The area magnification also increases the average photon energy deposited in each layer, which can be noticed from Figure 4. The larger geometrical structure of betavoltaic battery means that it could produce more photon generation. The average photon energy deposition can be improved up to 139.6%, 22.886%, and 39.21% for N+ substrate, N-epilayer, and P+ epilayer, respectively.
3.2. Absorbed dose

From previous results, we have proofs that a large piece of radioactive material does not mean that it can deposite all of the emission energy to semiconductor. The self-absorption as a response to the geometrical aspect reduces the linearity of particle fluence effectiveness to generate EHPs in SiC. Basically, the energy deposition is proportional to the absorbed dose, which is the measure of how the incident ionizing radiation could effectively deposite its energy per unit mass. The absorbed dose could also be the measurement to predict the ionizing radiation damage to the semiconductor crystal over time. The radioactivity of beta source can be calculated by looking at its radioisotope specific activity. For Ni-63, the specific activity is about 2.097 GBq/g, thus the activity of source with thickness of 0.02 to 1.00 μm are varied from 0.25 to 12.55 mCi, which is sufficient to generate nA to μA current in the practical of betavoltaic battery.

![Graphs showing absorbed dose vs mass thickness](image1.png)

(a) (b) (c)

Figure 5. The absorbed dose of electron inside the 4H-SiC PIN semiconductor: (a) N+ substrate, (b) N-epilayer, (c) P+ epilayer with different surface interface area.

In this study, we focused on the simulation that was invariant to the time scale, thus neglecting the decay activity of beta source. In other words, we could tell that the source defined in a condition when there is no conversion of Ni-63 to Cu-63 according to the decay scheme of Ni-63, although in practical it is difficult to have a piece of radioactive material with 100% purity of radioactive element at the beginning. However, a solid Ni-63 has a density of 8.902 g/cm³, which is not too different with Cu-63 at about 8.960 g/cm³ [19]. This would not cause a big difference to the maximum penetration range, since the nuclear radius is almost the same to each other which is proportional to the collision or reaction cross section.

The area magnification effect to the absorbed dose is almost undetected in all parts of active cell, including the N+ substrate as shown in Figure 5 (a)-(c). Basically, the shape of these curves are
proportional to the self-absorption theoretical approach by Oldano-Pasquareli. The geometrical differentiation takes part of the distribution angle, which is measured by the counting geometric efficiency as given by this equation [3]:

\[ f_G = G_0 + (G_1 - G_0)\phi \]  

(6)

where \( G_0 \) and \( G_1 \) are both the constants from the series expansion of counting geometric function, which describe the distribution angle of beta particle that are emitted from the source. The slope function of \( \phi \) is defined by the error integral which depends on the geometrical structure of the source, given by:

\[ \phi = 2 \int_0^\infty e^{-\mu's}dx \]  

(7)

where \( s \) is the source thickness from a definite perspective, and \( \mu^* \) is the absorption coefficient for beta particle derived from the empirical formula, which can be described as:

\[ \mu^* = 0.0488 E_{max}^{-1.41} \]  

(8)

From the Ni-63 we have \( E_{max} = 0.067 \) MeV for a given beta particle, thus we obtained \( \mu^* = 2.206 \) cm\(^2\)/mg. By using the information from Equation (7) and (8), we could fit the absorbed dose data with Equation (6) to analyze the geometrical coefficients which is related to \( f_G \).

**Table 2.** The results of data fitting with counting geometric semi-empirical equation to investigate the distribution of emitted particles due to the scattering effect.

| Material       | \( G_0 \)       | \( G_1 \)       | \( R^2 \)       |
|----------------|-----------------|-----------------|-----------------|
| N+ substrate   | 4.79729E-5      | 0.0049          | 0.99961         |
| N epilayer     | 0.05584         | 0.8615          | 0.99257         |
| P+ epilayer    | 0.21283         | 1.61124         | 0.97695         |

As we evaluate the geometric coefficients from three different parts of semiconductor, we obtained a variety of \( G_0 \) and \( G_1 \) values. At the N+ substrate, the geometric coefficients tend to have the lowest value compared to the others, which indicates that beta particle comes with a directed trajectory and has a more uniform angular distribution. This also confirms that only the beta particle with sufficient energy and almost ‘straight’ trajectory could possibly reach the N+ substrate, since the particle not only interacts with the SiC atoms in the semiconductor, but also the Ni-63 itself. The P+ epilayer has a large value of geometric coefficient, and it is clear that the upper layer has a direct contact with Ni-63 source, thus it received a wider angular distribution than other parts of semiconductor.

4. Conclusions

We have simulated a cylindrical Ni-63/4H-SiC betavoltaic based on PIN semiconductor by using MCNPX to calculate the beta particle interaction and the energy deposition. From the simulation results, the self-absorption dealt a negative impact to the energy deposition of beta particle inside the semiconductor layers. Basically, the addition of mass thickness resulted in a deterioration of beta energy deposition. A peak of optimum source mass thickness related to the maximum energy deposition at the lowest substrate, however, is obtained around 0.1424 mg/cm\(^2\). In general, the magnification of surface area could enhance the overall energy deposition, both the beta particle and the photon. As we increased the source mass thickness, the photonic energy deposition attained an amplification, contrary to the electron. As we analyze the absorbed dose in the semiconductor, the curves show an indication of
saturation if we add the thickness further. This study is consistent with the theoretical study of self-absorption for the electrons. Furthermore, the absorbed dose data is fitted with Oldano-Pasquarelli model. According to the calculation of geometrical coefficients, we conclude that the beta particle emitted from Ni-63 should have a ‘straight’ trajectory when they reach the lowest parts of the semiconductor, thus a narrow angular distribution is achieved.

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