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Electron-hole pairs generation rate estimation irradiated by isotope Nickel-63 in silicone using GEANT4

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Abstract. To optimize parameters of beta-electrical converter of isotope Nickel-63 radiation, model of the distribution of EHP generation rate in semiconductor must be derived. By using Monte-Carlo methods in GEANT4 system with ultra-low energy electron physics models this distribution in silicon calculated and approximated with Gauss function. Maximal efficient isotope layer thickness and maximal energy efficiency of EHP generation were estimated.

Introduction

New generation micropower devices and microelectromechanical systems need compact long-lifetime power supplies. Betavoltaic effect radioactive energy to electric energy transformation is one of perspective ways for meeting such requirements. Betavoltaic semiconductor converter of 63Ni isotope radiation is considered to be a perspective technology by multiple scientific researches in this area. 63Ni isotope emits beta-particles having the energy level below structural damage energy for most semiconductors, does not emit gamma-particles and has half-decay time at about 100 years.

Isotope undergoes radioactive decay

$$^{63}_{28}\mathrm{Ni} \rightarrow {}^{63}_{29}\mathrm{Cu} + \mathrm{e}^-,$$

the result of which is emission of electron (β-particle) having average energy 17.3 keV with a random direction. During travel through the space-charge region of p-i-n junction such particle causes the generation of electron-hole pair (EHP) which is being separated by the built-in electric field. Separated charge carriers travel to the opposite directions: holes into the p-region, electrons into the n-region. As the result, difference of potentials is induced on p and n region leads, the same way as it occurs inside photoelectric converter. If connectors are attached to external resistance, the current flows through the circuit and generated energy can be used to power the load.

The main advantages of radioisotope-based power sources are long service life (determined by half-decay time), high specific energy, high energy density, wide range of operational environment conditions (determined by semiconductor) and high reliability. It is possible to create flexible hybrid voltage or current source incorporating the charge accumulation for continuous operation or pulse-mode operation.

It is necessary to perform the modelling of all energetic processes – primary beta-electrons generation, electron-hole pairs generation, particles diffusion and drift inside space-charge region, recombination and others. It is possible to use COMSOL Multiphysics for modelling some of listed processes, but it doesn?t incorporate the model of betavoltaic particles generation. For this reason Monte-Carlo method modelling using GEANT4 software was performed to estimate the distribution of EHP generation rate in semiconductor.

Problem definition

Different scientific research teams use different approach to estimate the the distribution of electron-hole pairs generation rate.

Betavoltaic effect modelling was performed in monograph [9]. It was proposed, that energy carriers generation rate is proportional to the distribution of energy generation rate through the depth of semiconductor. Generation rate for every particle energy (see fig. 1(a)) is estimated using table data from [5]. Energy generation rate was approximated using Gauss function

$$J(z) = A \exp\left(-\frac{(z-z_0)^2}{2\sigma}\right), \tag{1}$$

where parameters z_0 and σ were extrapolated to the low-energy range.

Charge carriers generation rate dependency on electron penetration depth for ^{63}Ni isotope with radioactivity 20 mKi is shown on figure 1(b). Charge carriers generation rate was calculated as

$$G = \int_0^{67\text{keV}} A(E) \cdot \Omega(E) \cdot J(E)\, \mathrm{d}E,$$

where $A(E)$ - amplitude of absorption function, $\Omega(E)$ - energy spectrum ^{63}Ni isotope, $J(E)$ - energy generation function, estimated using equation (1). From graph of generation rate dependency on electron penetration depth one can estimate that maximum charge carriers generation rate located at the depth about 6 um.

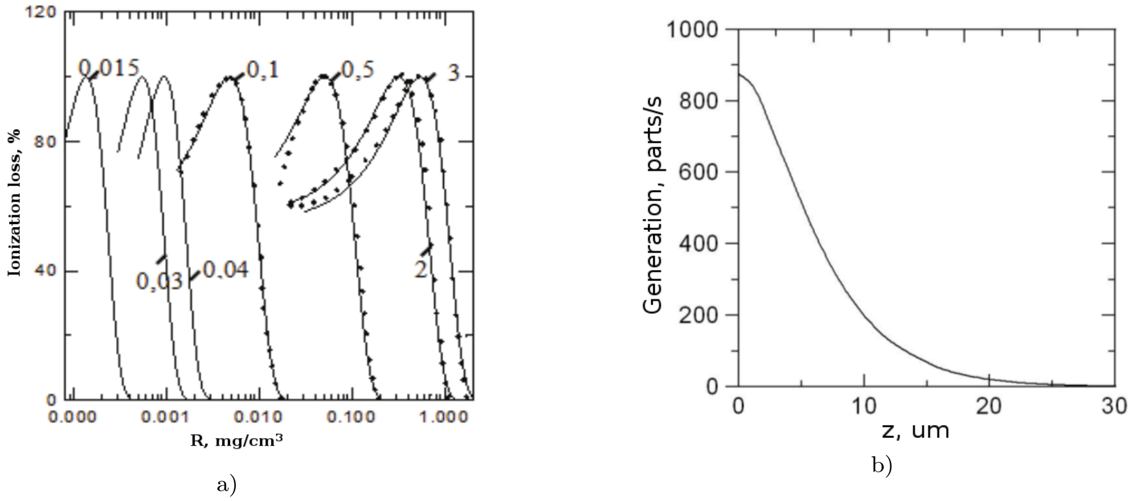


Fig. 1. a) Energy generation rate distribution by the depth of semiconductors for electrons having different energy. Numbers 0.015 - 3 corresponds to the radiation energy in MeV.
b) Charge carriers generation rate dependency of electrons penetration depth when using ^{63}Ni, per one beta-particle. Graphs taken from [9].

The article [6] studies beta-particles energy loss in silicone. Calculation is performed using Monte-Carlo method. Plot graph of ionization energy losses for isotope ^{63}Ni having mass density 1mg/cm^2 is shown on figure. The most of the energy loss takes place in near-surface layer of silicone, energy losses distribution is close to exponential. Total energy ionization losses is up to 80% at silicone thickness 5 um and near 100% at 20 um.

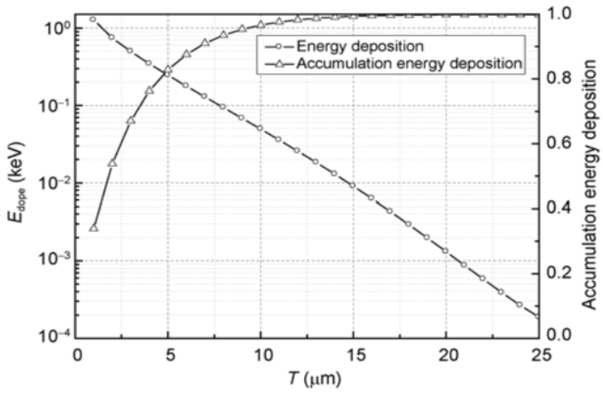


Fig. 2. Dependence of ionization losses of electron penetration depth when using ^{63}Ni isotope (from [6]).

In the article [7] interaction of betaelectron radiation with silicone is calculated using Monte-Carlo method in CASINO software. The dependency of electron energy losses of depth of penetration is shown on fig 3. Authors observed that the energy loss curve for 10 keV particles has maximum at 400 nm, therefore p-n junction depth should be at least 500 nm to provide efficient operation.

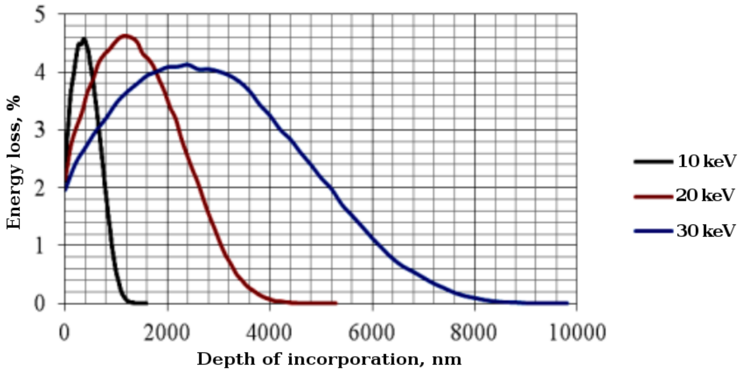


Fig. 3. Dependence of radiation energy ionization losses from electron penetration depth (from [7]).

The comparison of researches listed above shows that energy generation rate curves differ, which may be caused by simplification of electron-matter interaction models.

Current article propose to use the electron-matter interaction model, which is defined for low levels of energy. For instance, model proposed in [2] describe the ionization and electron transition into conduction zone for energy range from 16.6 Ev to 50 kEv. The calculated interaction cross-sections are used to plot the tracks of beta-electrons for isotope ^{63}Ni in silicone.

The primary goal of current research is to build the model of electron-hole pairs generation rate in silicone. The selected geometrical schematic of this problem is shown on figure 4(a).

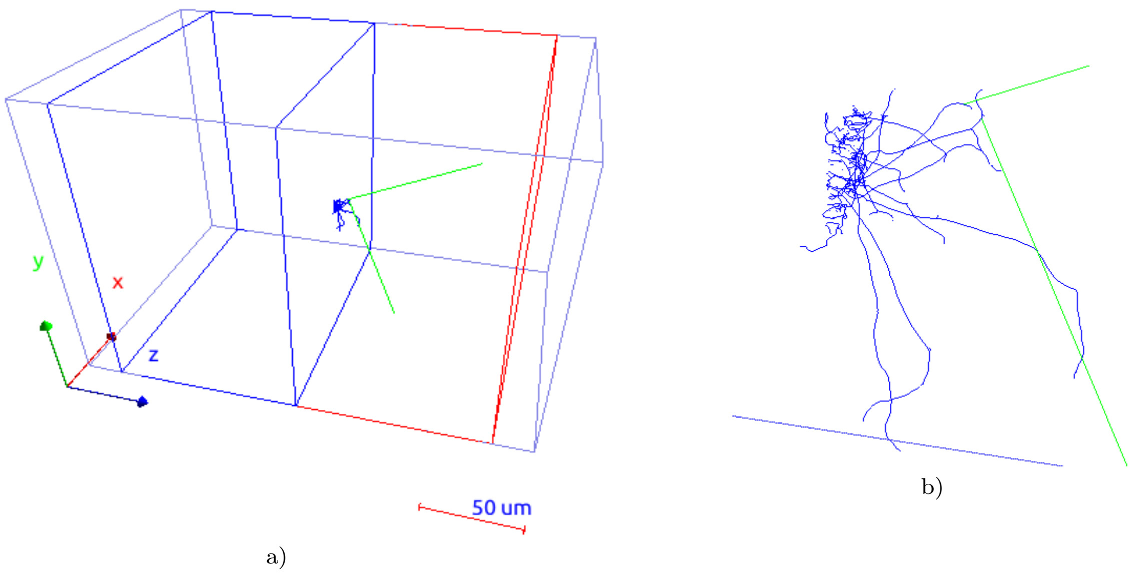


Fig. 4. a) geometry of the problem; b) the tracks of beta-electrons, emitted by ^{63}Ni isotope (100 tracks).

The object is a cuboid consisting of two equal parts - isotope and silicone. Both parts are 400 um thick, which is equal to standard silicone wafer thickness. Particle tracks starting from the rectangle shaped area 6x6 um, placed at the plane between isotope and semiconductor. Figure 4(b) shows the resulting beta-electrons tracks in silicone.

Fast electrons generation takes place in the isotope layer. The spectrum of beta-radiation corresponds to ^{63}Ni isotope (see fig. 5), spectrum is taken from article[1]. One can see that spectrum, approximated with exponential function $a\exp(-b \cdot E)$ differs from measured curve (criterion $R^2 = 0,991$). Despite this fact, exponential model was used in current research to increase computation speed, even though GEANT4 supports table-defined distributions.

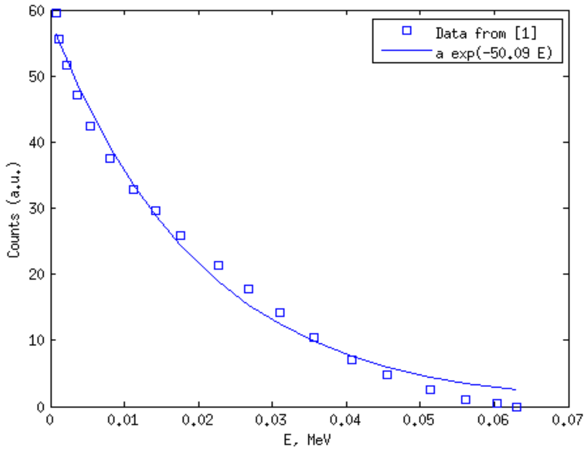


Fig. 5. ^{63}Ni radioactivity spectrum approximation.

Calculations listed below do not take into account the built-in electric field of p-i-n junction space-charge region, despite the fact that it should influence the distribution of generation rate. Currently there is no dopant concentration distribution data for beta-voltaic converter being developed, for this reason space-charge region electric field estimation is not possible.

Calculation sequence:

1. Tracks calculation in GEANT4, corresponding to the software-defined experiment schematic;
2. Saving tracks data (nodal points coordinates, the amount and type of energy loss) as CSV-files;
3. Reading of generated CSV-files into memory, saving to float data files;
4. Separation the data of every track from whole data array, taking in consideration only ionization losses; secondary electrons are filtered out from tracks data, they are taken into account as separate particle tracks);
5. Ionization energy losses (the number of electron-hole pairs generated) is calculated for every elementary volume of silicone.
   Ionization energy losses calculation is performed using the following algorithm. For every track, the segment table of energy losses is filled, including starting and ending points of segment. The electron-hole pair energy in silicone is estimated as $E_{e-h} = 3,6$ eV.
   Using this estimation, the number and coordinates of generated electron-hole pairs is calculated, considering the probability of generation to be constant during the whole track segment. The space of semiconductor is divided into elementary volumes and for each one the quantity of generated pair is calculated.
6. Calculation of electron-hole pairs generation rate field in semiconductor.
   Estimated field of energy generation rate is normalized relative to total initial energy of beta-radiation, representative segment of field is separated and approximated using Gauss function [8]:

$$C(x,y) = \frac{N}{\sqrt{2\pi} \cdot R_P} \exp\left(-\frac{(y-R_P)^2}{2\,\Delta R_P{}^2}\right) \cdot \left(\operatorname{erf}\frac{x+a}{\sqrt{2}\,\Delta R_X} - \operatorname{erf}\frac{x-a}{\sqrt{2}\,\Delta R_X}\right), \tag{2}$$

   where y – position on axis, perpendicular to semiconductor surface, um; x – position on axis, parallel to surface; N – injection doze, electron/cm^2; R_P – projective range, um; ΔR_P – standard deviation of projective range, um; a – beta-emitter width coefficient; ΔR_X – standard deviation of lateral range, um.
   First part of this equation was used to approximate one-dimensional (normal to surface, pointed into the depth of semiconductor) generation rate distribution (2):

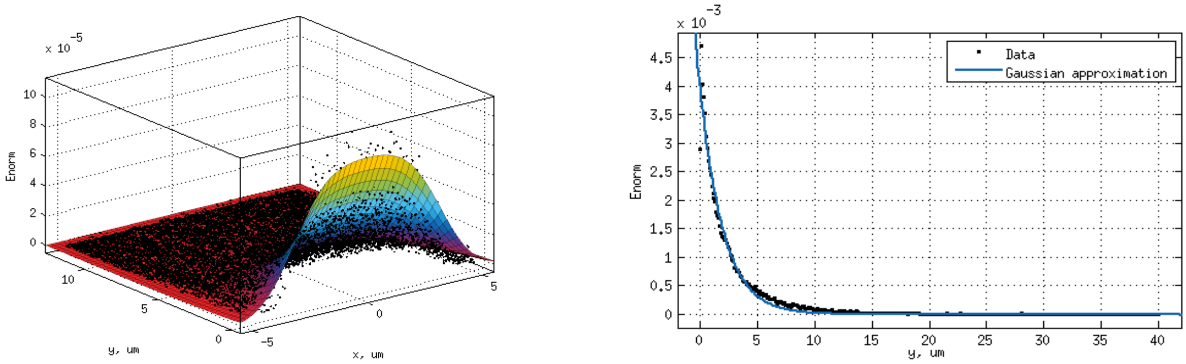
$$C(y) = \frac{N}{\sqrt{2\pi} \cdot R_P} \exp\left(-\frac{(y-R_P)^2}{2\,\Delta R_P{}^2}\right). \tag{3}$$

GEANT4 source code used for track estimation is uploaded to GitHub[6]. Number of tracks used for simulation was 20000, to increase the simulation precision. Calculation was separated into 4 threads to minimize the simulation time.

[6] https://github.com/alex-khoroshko

Results and discussion

The field of EHP generation rate in silicon after radiation influence of 3 um ^{63}Ni layer and it's Gauss function approximation (2) is presented on figure 6. One-dimensional distribution (fig. 6, b) approximated with function $a \cdot \exp(-(y-b)^2/c^2)$, where $a = 7,184e+04$, $b1 = -68,74$, $c = 16,81$. One can see that the beta-particle flux generate EHP generally in depthward direction to silicon, to sideways the radiation disperse weaker. Most of EHP generated in subsurface layer, at depths not more 20 um.



a) b)

Fig. 6. The distribution of EHP generation rate in the depth of silicone when radiated by ^{63}Ni isotope (isotope thickness is 3 um, run 80000 tracks), a) two-dimensional, b) one-dimensional

To estimate of isotope layer thickness influence to EHP generation rate, spatial radiation source was replaced by plain surface (square 6x6 um, parallel to silicone surface). Radiating surface displaced from silicone surface to isotope region. All graphs are normalized relatively to initial particles radiation energy. Gauss function coefficient values are listed in the table 1.

Table 1. Coefficients of approximating function.

	2D, eq. (2)					1D, eq. (3)		
Δy, um	N	R_P	ΔR_P	a	ΔR_X	N	R_P	ΔR_P
0	-2.2646e+08	-161.3	48.518	3.004	0.24918	-12153	-67.2	6.5909
0.25	-1.4792e+08	-142.4	42.93	3.119	0.8358	-1284.7	-56.89	6.3953
0.5	-1.0004e+08	-147.1	44.331	3.194	1.0677	-245.45	-49.63	6.2145
1	-6.2004e+07	-127.9	38.299	3.095	1.2763	-38.072	-42.19	5.9699
1.5	-2.666e+07	-137.5	41.473	3.169	1.4093	-3.4767	-35.5	5.9447
2	-1.3394e+07	-141.7	42.729	3.639	1.7918	-1.4363	-30	5.5299
3	-1.126e+06	-131.7	41.11	3.12	1.8413	-0.36347	-26.15	5.246

Figure 7 shows the distribution of EHP generation rate along the normal to surface of silicone. The comparison of obtained distributions and distributions at figures 1 b) and 2, shows that distributions have same curve shapes, values match closely to each other.

Figure 8 shows the graph of total EHP energy depending on the distance of nickel particle-generating layer. Total emitted energy equals to 1, thus the graph corresponds to the energy efficiency of EHP generation. One can see that no more than 18% of initial energy is spent on EHP generation. This curve is approximated by the exponent $a \cdot \exp(-\Delta y/T_y)$, where $a = 0,1878$, $T_y = 0,7987$ um. The layer having thickness of $3 \cdot T_y = 2,4$ um generates 95% of useful energy, further increment of thickness does not increase useful energy generation. Using this factor, one can estimate economically justified isotope thickness. Total generation efficiency for semi-infinite isotope layer can be estimated as

$$\eta = \int_L a \cdot \exp\left(-\frac{\Delta y}{T_y}\right) \mathrm{d}l = aT_y = 0,15.$$

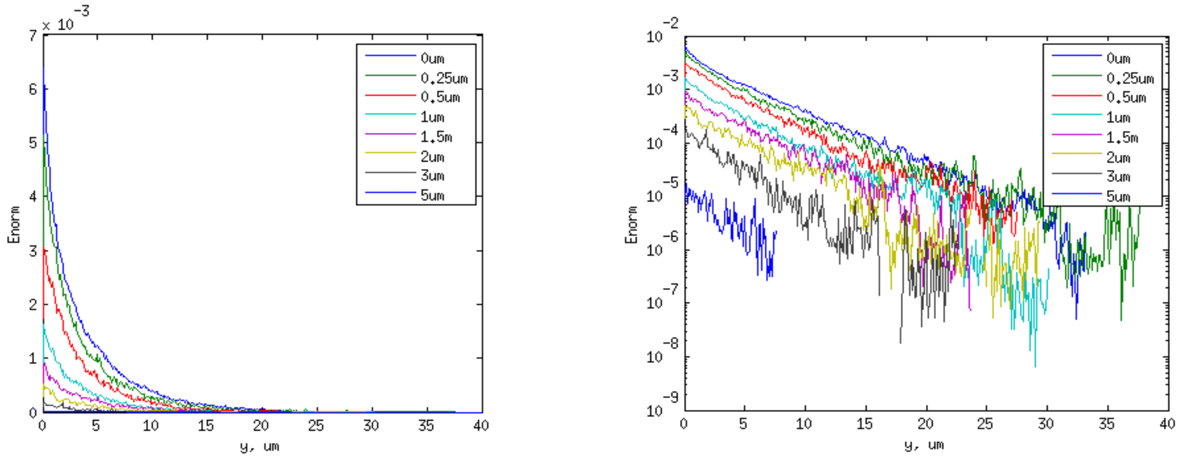


Fig. 7. The distribution of EHP generation rate with radiation source with different displacement.

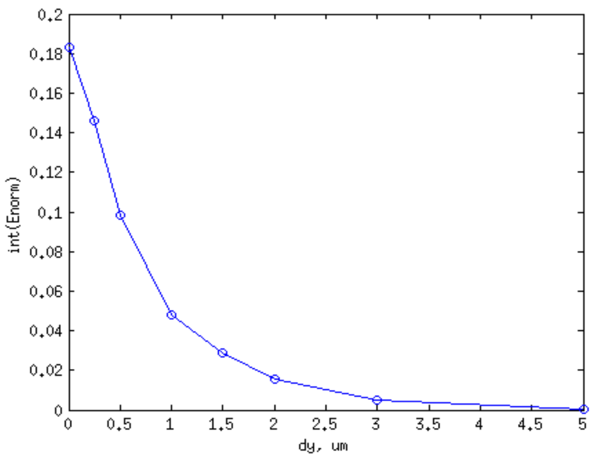


Fig. 8. Total energy

Conclusion

As the result of modelling of EHP generation in silicone induced by ^{63}Ni isotope radiation in GEANT4 with ultra-low energy models, maximal efficient isotope layer thickness was estimated - 2, 4 um. Maximal energy efficiency of EHP generation is about 15%. The fields of EHP generation rate with different thickness of ^{63}Ni isotope were estimated, so that it is possible to optimize parameters of silicone semiconductor structure. Calculated Gauss function approximation parameters can be used to define the fields for numeric modelling of beta-electrical converters, in software such as Comsol Multiphisics.

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