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CRediT author statement

V Grushko: Conceptualization, Formal analysis, Writing- Original draft preparation. O Beliuskina: Methodology, Writing- Reviewing and Editing. A Mamalis: Writing- Reviewing and Editing. V Lysakovskiy: Resources, Investigation, E Mitskevich: Sofrware, Investigation, A Kiriev: Investigation, Visualization. E Petrosyan: Methodology. R Chaplynskyi: Validation, Formal analysis. O Bezshyyko: Conceptualization, Data Curation. O Lysenko: Supervision.

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Energy conversion efficiency in betavoltaic cells based on the diamond Schottky diode with a thin drift layer

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Abstract

The HPHT diamond Schottky diode was assembled as a Metal/Intrinsic/p-doped structure betavoltaic cell (BC) with a very thin (1 μm) drift layer and tested under $5-30$ keV electron beam irradiation using a scanning electron microscope (SEM). The effect of the β-radiation energy and the backscattering of electrons on the energy conversion was studied. From the results obtained, it is shown that, the efficiency of the investigated BC increases from 1.01 to 3.75 % with the decrease of β-particle energy from 30 to 5 keV due to an increase of the electron beam absorption in a thin drift layer. Maximum efficiency is achieved when the electron beam energy is close to the average β-decay energy of ^{3}H. The BC maximum output power of the 1.6 μW was obtained at an electron beam energy of 15 keV, that matches the β-decay energy of ^{63}Ni. The total BC conversion efficiency at 15 keV electron-beam energy is about 3%. The calculations indicated that a preferable β-source for the diamond based BCs with a thin (1 μm) drift layer is ^{63}Ni.

Keywords: betavoltaic, diamond, energy conversion efficiency, thin drift layer, Schottky diode.

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1. Introduction

The concept of using p-i-n and p-i-m (Schottky) junctions using for the betavoltaic energy conversion was proposed in the 1950s (Rappaport (1954); Pfann & Van Roosbroeck (1954)). Recently, numerous betavoltaic cell (BC) devices based on Si, SiC and GaN, were developed (Bao et al. (2012); Zhang et al. (2018); Chandrashekhar et al. (2006); Qiao et al. (2008); Eiting et al. (2006)). Note that, the betavoltaic devices are widely used in the harsh radioactive environment, like power plant reactors, spent nuclear fuel storages etc., remote places such as space and mine-like underground, undersea and so on. Moreover, they are most notable for medical devices such as pacemakers.

Most commonly used radioactive sources for the industrial BCs are the ^{3}H, ^{63}Ni, ^{90}Sr, ^{90}Y, ^{147}Pm, ^{35}S, ^{33}P, ^{204}Tl, ^{85}Kr isotopes (San et al. (2013); Sun et al. (2005); Lu et al. (2011); Yao et al. (2012); Chen et al. (2011); Preiss et al. (1957); Meier et al. (2009); Eiting et al. (2006)). The typical output power of the tritium BC is about 120 nW. In the betavoltaic effect studies with ^{63}Ni an induced current of tens of nanoamps to few microamps was achieved (San et al. (2013); Lu et al. (2011); Yao et al. (2012); Chen et al. (2011)). A further development of long lifetime energy sources, based on β-isotopes to increase power and reduce size of BC, is extremely challenging.

Limitations of betavoltaic cells with a variety of beta sources had been studied extensively over past years. For example, Theirrattanakul & Prelas (2017) reported that the betavoltaic efficiency of nuclear batteries based on silicon carbide betavoltaic cells with ^{3}H, ^{63}Ni, ^{35}S, ^{147}Pm, ^{90}Sr and ^{90}Y thin planar beta sources decreases with increasing beta energies. The ^{3}H source had the highest absolute efficiency at 3.95% using the full spectrum energy distribution model. Oh et al. (2012) reported that the calculated conversion efficiencies were limited to a range of 0.013% to 2.02% for SiC-based betavoltaic cell with different beta sources. According to Zhang et al. (2018) the conversion efficiency was in a range of $3.74 - 4.58\%$ as the result of the SiC PIN betavoltaic cell simulation. Wu et al. (2011) presented results on theoretical study of silicon betavoltaic

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microbattery using ^{63}Ni as beta source with obtained conversion efficiency of 5%. Wu & Zhang (2019) introduced the simulation of conversion efficiency of multilayer BC with silicon p-n junction converters and ^{63}Ni sources. The total conversion efficiency of this BC limited to 3.3%. Murphy et al. (2019) reported that in the case of silicon diodes with three-dimensional features when coupled with ^{147}Pm oxide theoretical efficiencies of $2.9 - 5.8\%$ can be achieved. More information about the BC reported effectiveness can be found in the review of nuclear batteries by Prelas et al. (2014).

The reliability and safety of BC, especially for medical applications, mainly depend on the radiation hardness and the mechanical strength of the semiconducting material. Due to the high radiation tolerance, the high mechanical strength and chemical inertness diamond is an excellent material for betavoltaic devices. Examples of BCs using diamonds are presented by Delfaure et al. (2016), Bormashov et al. (2015), Tarelkin et al. (2016) and Zhao et al. (2017).

It is noted that, the small output power of the diamond BCs limits their use. The solution to this problem is to create multilayer power sources. In this case, the output power depends on the geometrical dimensions of the multilayer structure and, therefore, on the BC layer's thickness.

Recently, a multilayer BC based on a diamond Schottky diode with the the drift single layer cell size of 15 μm was presented (Bormashov et al. (2018)). A reduction of the thickness of the drift layer and, accordingly, a decrease in the size of a multilayer BC based on a diamond Schottky diode, constitute the best way to increase the power density of such a device. The aim of our research was to study a betavoltaic energy conversion efficiency in a diamond Schottky diode with a drift layer thickness of 1 μm.

2. Methods and Materials

2.1. The current generation in the diamond Schottky diode

The operation principle of the betavoltaic device is based on the electron-hole pair generation in a diamond Schottky diode induced by β-particles emitted

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60 from radioactive isotopes. The process of electron-hole pairs (EHP) generation can be described by the generation function: $g(x) \sim exp(-\alpha x)$ (Sachenko et al. (2015)), where, α is the linear electron absorption coefficient and x is the depth in diamond bulk. Note that, only part of the β-particle total kinetic energy is absorbed in the drift layer with thickness of $l = 1$ μm, see Figure 1(a). It is 65 clearly indicated that, the higher the penetrating power (i.e. longer stopping depth) of the β-radiation, the smaller this part will be. The electron-hole pairs induced by β-radiation in a drift layer are directly converted into an electric current of BC.

Due to its capability of efficiently converting the ionizing radiation into an 70 electric current, and, furthermore, its high radiation tolerance, thermal conductivity, mechanical strength and chemical inertness, diamond was chosen as the main component of the radioactive power source. The diamond has a wide band gap and high donor and acceptor ionization energies. At room temperature, as well as below it, the Fermi level is located close to donor or acceptor levels, de- 75 pending on the concentration of impurities which is predominant in the crystal (Collins (2002)). The electron-hole pairs formed inside the depletion region of the Schottky diode are separated by an internal electric field, thereby forming a radiation-induced current in the BC (Manasse et al. (1976)). The Schottky barrier height defines the maximum battery voltage. This value depends strongly 80 on the band gap structure of the semiconductor (Tung (2014)).

It is known, that a high concentration of a doping impurity in a semiconductor leads to a thinning of the depletion region and, thereby, reduces the number of the electron-hole pair generated in this region (Bormashov et al. (2015)). Therefore, in diamond Schottky diodes, the depletion region is typically broad- 85 ened to a value of $5 - 10$ μm at zero bias by the formation of an epitaxial drift layer on the surface of a doped semiconductor, into which a small amount of doping impurity diffuses. It is also known that, the charge collection efficiency, Q of BC may dramatically decrease when a diode is irradiated by high-energy electrons which have higher penetrating power, because of the longer stopping 90 depth (Bormashov et al. (2015)).

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2.2. Experimental setup

The schematic diagram of a diamond Schottky diode betavoltaic cell is shown in Figure 1 (a). The p^+ substrate of diamond diode was made from a boron-doped single crystal diamond grown by the temperature gradient method at high pressure - high temperature (HPHT) (Novikov et al. (2003)) with the boron contents of 10^{18} cm^{-3}. Subsequently, diamond crystal was cut as {001} planes and mechanically polished up to roughness of 2 nm RMS. The surface quality after polishing was controlled by the Scanning Tunnelling Microscope (STM) technique (Grushko et al. (2014); Lysenko et al. (2010)). Since a large number of structural defects significantly reduces the lifetime of non-equilibrium carriers in a semiconductor, worsening, therefore, the efficiency of betavoltaic energy conversion, much attention was paid to the quality of this boron-doped plate. Due to the large number of dislocations, stacking faults and twin boundaries in the {111} growth sectors of the HPHT diamond (Chepugov et al. (2013)), mainly {001} and {311} sectors were used as a substrate for the Schottky junction.

In order to avoid the formation of the ohmic contact between the metal and the crystal plate and to increase the number of the electron-hole pairs in the Schottky junction region the p^- epitaxial drift layer was deposited on the diamond plate by the CVD method (Zhao et al. (2017)). The quality of Schottky junction substrate surface significantly affects the quality of the diamond epitaxial CVD layer. In our case the thickness of the deposited CVD layer did not exceed 1 μm.

The presence of non-equilibrium carriers in the drift layer limits the penetration depth of the electric field in the semiconductor and, respectively, the depth of the depletion region (Bormashov et al. (2015)). The thickness of the drift layer determines the Schottky barrier height and limits the depth of the depletion region where the induced charge is collected. With an increase of the drift layer thickness from 100 to 500 nm, the height of the Schottky barrier increase from 1 to 1.8 eV (Zhao et al. (2017)). According to Bormashov et al. (2015) and Tarelkin et al. (2016), for a drift layer depth of about 10 μm and a zero offset the depth of the depletion region is about 5 μm.

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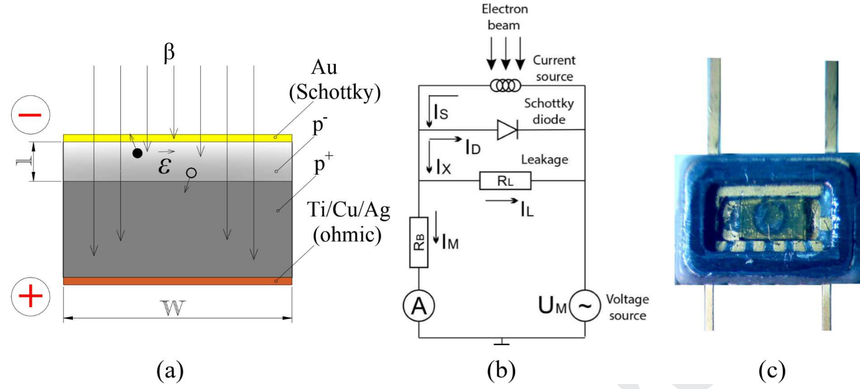

Figure 1: (a) The schematic diagram of a diamond Schottky diode betavoltaic cell. The solid marker denotes electrons and the open marker denotes holes, $\vec{\varepsilon}$ is the built-in electric field in the drift layer, l and W are the drift layer's depth and width, respectively. (b) The electric circuit for the betavoltaic energy conversion efficiency measurements. (c) The diamond betavoltaic cell prototype.

After the drift layer deposition, an Au layer with a thickness of 10 nm was deposited using the magnetron sputtering method in the Ar atmosphere. The Au layer thickness was controlled by STM. On the backside of the diamond p^+ plate, the Ti adhesive layer, the Cu conducting layer and, finally, a thin Ag layer were deposited by the same method. The sputtering of Ti provided the formation of a TiC interlayer on the diamond surface and the stable ohmic contact of the diamond plate with a Cu/Pt conducting layer.

The conversion of β-decay energy into electricity in a diamond Schottky BC was investigated using the Electron Beam Induced Current (EBIC) technique (Delfaure et al. (2016)). Figure 1 shows the electric circuit used for the estimation of the total efficiency and the prototype of a diamond Schottky diode BC based on the chip KD917A without cover wafer. The diamond diode p^+ plate is placed at the bottom of the chip, that creates an ohmic contact with a pair of upper electrodes of the chip. Two bottom electrodes provide a contact with the gold layer deposited on the top of the plate.

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To study the conversion of the β-decay energy into the electricity in a diamond Schottky diode, the BC dark current-voltage characteristics (I-V) in forward and reverse bias modes were measured. Results of these measurements
140 are shown in Figure 2. The forward and reverse branches of dark I-V characteristic (Figure 2) allow for drawing a conclusion about the good rectifying property of the diode connection. The leakage current of the diode was less than 100 pA at the reverse bias of about 100 V. At the forward bias greater than 1 V the dark current rises sharply. A typical rectification factor is of the order of
145 10^9 at the forward bias of about 2 V.

The typical betavoltaic curve obtained at the beam energy of 15 keV is shown in Figure 3. The open-circuit voltage of the diamond BC reaches the value of more than 1 V. The induced current of BC remains approximately constant at the applied reverse bias. As it can be seen in Figure 3(a), even at zero bias
150 almost all electron-hole pairs generated by the electron beam are separated by the internal potential of the diamond diode and the induced current reaches its saturation. Figure 3(b) shows values of the short-circuit current of the diode at different electron beam energies. From this Figure, it is indicated that, the maximum current of 1.72 μA corresponds to an electron beam energy of 15 keV.
155 At beam energies above 15 keV the short-circuit current decreases. As stated below, the number of generated electron-hole pairs in a thin drift layer decreases due to an increase of the penetration power of β-particles.

3. The total conversion efficiency

The total conversion efficiency η of the BC, as described in Olsen (1973b),
160 is:

$$\eta = \eta_\beta \eta_c \eta_s, \tag{1}$$

where:

$$\eta_\beta = \frac{N_\beta}{N_0}. \tag{2}$$

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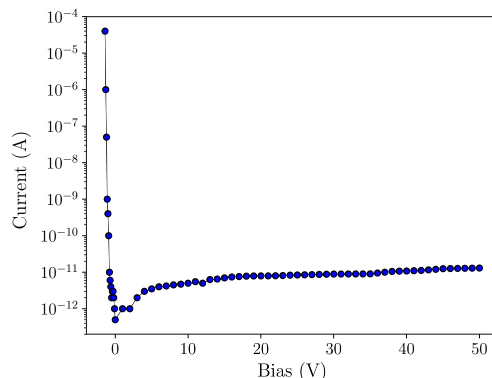

Figure 2: The dark current-voltage characteristic (I-V) of the diamond Schottky diode at forward and reverse biases. The positive voltage values correspond to the reverse bias and the negative values to the forward bias.

η_β is the irradiation efficiency of the diamond Schottky junction by the β-particles, defined as the ratio of the β-flux reaching the diamond surface to the total β-flux. Considering the EBIC experiment configuration, η_β can be assumed equal to 1.

$$\eta_c = (1 - r)Q, \tag{3}$$

η_c is the interaction efficiency of the incident β-particles with the diamond cell. r is the β-particle reflection coefficient from the diamond surface, and Q the charge collection efficiency (CCE) of the diamond, strongly depending on the β-particle absorption coefficient, minority carrier diffusion lengths, cell dimensions and surface recombination velocities (Olsen (1973a)). The value $(1-r)$, corresponds to the absorption probability of the β-particle in the diamond.

$$\eta_s = \frac{eV_{oc}}{\varepsilon} FF \times 100\%, \tag{4}$$

η_s is the efficiency of the semiconductor cell itself, e the elementary charge, V_{oc} the open-circuit voltage of the cell and FF the fill factor of the BC's current-

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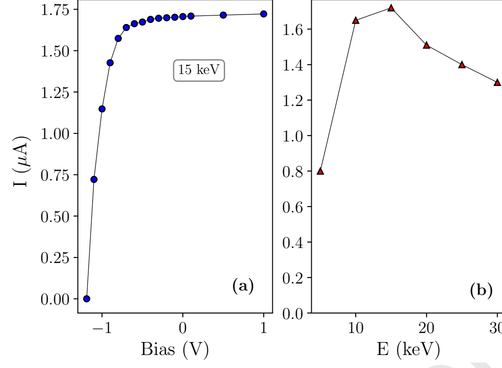

Figure 3: (a) Typical I-V betavoltaic curve of a diamond BC by 15 keV electron beam and (b) BC's short-circuit current as a function of electron beam energies.

voltage characteristic. At an open-circuit voltage of about 1.2 V and a short-circuit current of about 1.7 μA FF equals to 0.75.

$$\varepsilon = 2.8E_g + 0.5\ eV. \tag{5}$$

ε is the energy required to create one electron-hole pair and $E_g = 5.5\ eV$ is the depth of the diamond semiconductor band gap (Klein (1968)). Thus, the total efficiency η of BC in the EBIC measurements can be written as:

$$\eta = (1 - r)Q\frac{eV_{oc}}{\varepsilon}FF \times 100\%. \tag{6}$$

The reflection coefficient r can be calculated from the maximum possible short-circuit current $I_{sc_{max}}$ of the BC (Olsen (1973a))

$$I_{sc} = QI_{sc_{max}} = \frac{I_0}{\varepsilon}E(1 - r)Q, \tag{7}$$

I_0 is the current of β-particles incident on the semiconductor surface with an average energy E and I_{sc} the real device short-circuit current.

The total efficiency η can now be written as

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$$\eta = \frac{I_{sc} e V_{oc} FF}{I_0 E} \times 100\% = \frac{I_{sc} V_{oc} FF}{I_0 V_0} \times 100\% = \frac{P_{max}}{P_{in}} \times 100\%, \quad (8)$$

V_0 is the beam voltage, $P_{max} = I_{sc} V_{oc} FF$ is the maximum power delivered by
185 a cell and $P_{in} = I_0 V_0$ the injected power from the SEM.

Assuming that β-particles are absorbed in the semiconductor exponentially with an absorption coefficient α, according to the Lenard law (Casey & Kaiser (1967)):

$$\frac{I}{I_0} = e^{-\alpha x}, \quad (9)$$

$$\alpha = 1.9 \cdot 10^{11} \rho Z^{1/2} V_0^{-2}, \quad (10)$$

190 I/I_0 is the fraction of the electron flux transmitted, x the electron penetration depth in diamond bulk, ρ the density of the semiconductor, Z its atomic number, V_0 the electron beam accelerating voltage of SEM. Note that, for the diamond material with density of 3.52 g/cm^{-3} and $Z = 6$, the coefficient α decreases from $6.55 \cdot 10^4$ to $2.42 \cdot 10^3$ cm^{-1} due to the beam voltage increase from 5 to
195 30 kV. The beam current I_0 was measured by the Faraday cup; at the beam energy of 15 keV the value of the beam current I_0 was 3.8 nA.

The experimental value of the total efficiency η_e, obtained using eq. 8, the theoretical value of the total efficiency η_t for different α, calculated from eq. 6, and the absorption coefficient α (see eq. 10) as a function of beam energies are
200 presented in Figure 4.

For the theoretical analysis of the η_t dependence on the beam energy we used the expression for Q obtained by the method based on the solution of the diffusion equation for minority carriers in a semiconductor described in Olsen (1973a):

$$Q = \frac{\alpha L_n}{1 - (\alpha L_n)^2} \left[\frac{\alpha_n - \gamma_n e^{-\alpha l}}{\beta_n} + \alpha L_n \left(\frac{e^{-\alpha l}}{\beta_n} - 1 \right) \right], \quad (11)$$

205 L_n is the electron diffusion length in the drift layer, i.e. this is the average distance that an electron, induced by a β-radiation, can travel from the place

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of its appearance to the place where it recombines with the hole; it is about 10 μm in accordance with the reported data by Tarelkin et al. (2016). It is assumed that the Schottky junction is abrupt and has a thickness $l = l_d + l_t$, where $l_d = 1$ μm is the thickness of the drift layer and $l_t = 0.1$ μm the thickness of the transition p^-p^+ layer on the back face of the Schottky junction. From eq. 12

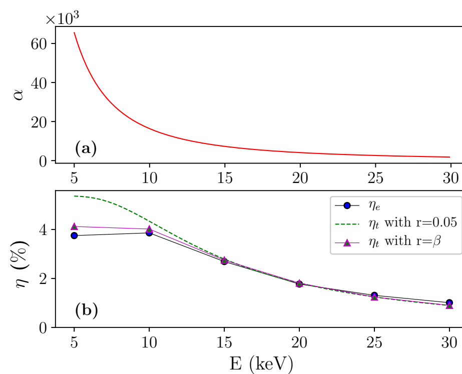

Figure 4: (a) Linear electron absorption coefficient, α; (b) Total efficiency of BC as a function of an incident election beam energy. η_e and η_t represent experimental and theoretical total BC efficiencies, respectively.

$$\alpha_n = \sinh\frac{l}{L_n} + \gamma_n \cosh\frac{l}{L_n},\ \beta_n = \cosh\frac{l}{L_n} + \sinh\frac{l}{L_n},\ \gamma_n = \frac{s_b\tau_e}{L_n}, \tag{12}$$

s_b and τ_e denote surface recombination velocity and the minor charge-carrier lifetime, respectively. We assume $s_b = 0$ and $\tau_e = 40$ ns (Almaviva et al. (2005)).

The reflection coefficient r in eq. 6 can be found assuming $r \sim \beta$, where β is the BC β-particles backscattered coefficient. The coefficient β was evaluated using the Monte Carlo simulations performed by the CASINO V2.5 software (Drouin et al. (2007)), see Figures 5 - 7). The BC β-particles backscattered coefficient and the beam energy loss in a 10 nm Au layer as the function of the β-radiation energy are shown in Figure 5.

Figure 4 (b) shows the BC theoretical total efficiency η_t for a constant value $\beta = 0.05$, with the β being evaluated using the Monte Carlo simulations. As

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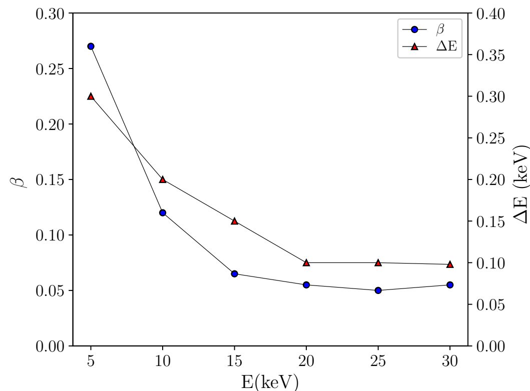

Figure 5: BC β-particles backscattered coefficient, β and beam energy loss, ΔE in a 10 nm Au layer as a function of the electron beam energy.

can be seen from this Figure, the theoretical curve for different values of β that calculated using CASINO modeling, is in a very good agreement with the experimental data. The maximum value of the total conversion efficiency of the BC, about 4 %, gradually decreases at beam energies above 10 keV to a value of about 1 % at 30 keV.

4. Discussion

The intensity of EHP generation (pair/(s·nm)), simulated by CASINO as a function of BC depth at different beam energies after subtracting energy losses in the gold layer, is presented in Figure 6. The area under the curve corresponds to the created charge subsequently converted into the output current. The highest value of the created charge was obtained at beam energies of $15 - 20$ keV, which agrees well with the experimentally observed maximum value of the short-circuit current of the BC $I_{sc} = 1.72\ \mu A$ at 15 keV beam energy, see Figure 3(b).

The decrease of the BC total efficiency at increasing beam energy, see Figure 4(b), is due to a decrease of the β-particle absorption in the thin drift layer. At beam energies of $15 - 20$ keV and more, the intensity of the electron-hole pairs generation decreases, see Figure 6, because a significant number of β-particles

240 stops far beyond the drift layer (stopping depth $1.5 - 2\mu$m, see Figure 7(b)). The significant lateral spread of β-particles (lateral projection is about 1.5 μm at the beam energy of 20 keV) leads to losses of β-particles at the edges of the drift layer. A further increase of irradiation energy leads to a further decrease of the area under the intensity curve and, as a result, of the observed value of 245 the BC output power.

The total efficiency slowly saturates at the beam energy of 5 keV, see Figure 4(b), apparently due to an increase of backscattering (from 6% at 15 keV to 30% at 5 keV) and the beam energy losses (from 0.8% at 15 keV to 6% at 5 keV) of the β-radiation in a thin layer of gold on the surface of the drift layer, see 250 Figure 5).

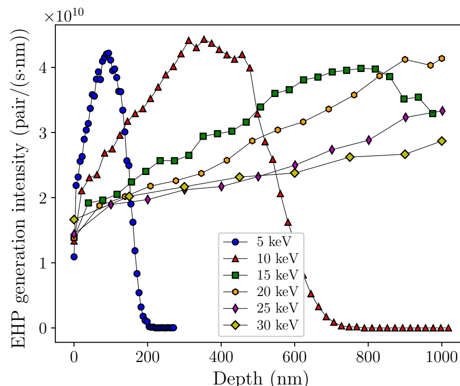

Figure 6: EHP generation intensity as a function of the penetration depth of β-particles in the drift layer of BC under different electron beam energies.

The decrease of the total conversion efficiency with the beam energy increase, see Figure 4(b), agrees well with the observed small value of the total efficiency for high-energy β-particles (Bormashov et al. (2015)). For example, according to Bormashov et al. (2015), for a mixed ^{90}Sr$-^{90}$Y source with an average β-particle 255 energy of about 1.1 MeV (penetration depth of about 2 mm and depletion region of 5 μm), the measured value of $\eta = 0.004\%$, due to the fact that only a small part of the β-particles kinetic energy is absorbed in the Schottky junction

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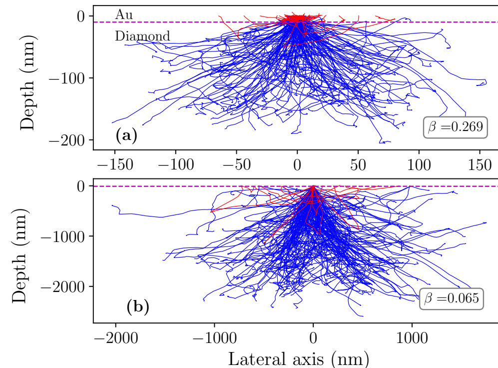

Figure 7: Trajectories of β-particles in the BC and the backscattered coefficient β at a beam energy of 5 keV (a) and 20 keV (b). Backscatter trajectories are marked in red.

depletion region and converted to electricity.

Backscattering of β-particles in the diamond material is small (up to several percent), due to a small atomic number of carbon, compared to other semiconductor materials (for example, Si 15% or GaN 25%) (Bormashov et al. (2015)), which helps to increase the total efficiency coefficient η.

5. Conclusions

From the theoretical and experimental studies reported above, the following concluding remarks may be drawn:

(a) The total conversion efficiency of the diamond Schottky diode with a thin (1 μm) drift layer increases from about 1% to 3.8% at the electron beam energy decrease from 30 keV to 5 keV, due to more efficient β-radiation absorption by a thin drift layer at low beam energies.

(b) The influence of β-particle backscattering in the BC metallic coating on the total conversion efficiency at low ($5-15$ keV) beam energies has to be taken into account.

(c) According to the Monte Carlo simulations performed with CASINO software the penetration of β-particles into the drift layer at energies greater than

275 15 keV exceeds 1 μm, which leads to a decrease in the BC output power and the efficiency of a β-decay energy conversion, since most of the trajectories of β-particles go beyond the drift layer in which the current of BC is generated.

(d) Lateral spread of β-particles at electron beam energies greater than 20 keV exceeds 1.5 μm, which can lead to a significant loss of the output power 280 and efficiency at the edges of the drift layer. This effect can be significant for micro betavoltaic cells with a small area of drift layer with W $<$ 5 μm.

(e) The β-radiation energy of 5 keV, at which the maximum energy conversion efficiency of about 3.8 % is reached, is close to the average β-decay energy of ^{3}H. Therefore, to obtain the highest efficiency of the β-decay energy conver- 285 sion into electrical energy ^{3}H is the most preferred source of β-radiation. But the output power with such source is just barely over 0.8 μW.

(f) The maximum output power (1.6 μW) of BC was measured at an electron beam energy of 15 keV, which is close to the β-decay energy of ^{63}Ni, at the same time the energy conversion efficiency was high (about 3 %). Thus, we can 290 conclude that ^{63} Ni is the most preferred β-source for the diamond betavoltaic cells based on a Schottky diode with a thin drift layer.

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Highlights

- The total conversion efficiency of the diamond Schottky diode with a thin (1µm) drift layer increases from about 1 to 3.8 % while the electron beam energy decrease from 30 keV to 5 keV due to the increase in the adsorption of beta particles by a thin drift layer.
- The most preferred beta source for the diamond Schottky diode with a thin drift layer is ^{63}Ni.
- The β-particles backscattered coefficient and the beam energy loss in the Schottky contact significantly affects the total conversion efficiency of the diamond Schottky diode at low (5 -15 keV) beam energies.
- The lateral electron spread exceeds 1.5 µm at beam energies greater than 20 keV, which can lead to a significant loss of the output power and efficiency at the edges of the drift layer. This effect can be significant for micro betavoltaic cells with a small (<25 µm^2) area of a drift layer.

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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