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Original Article

A nuclear battery based on silicon p-i-n structures with electroplating ^{63}Ni layer

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ABSTRACT

The paper presents the electrical performance measurements of a prototype nuclear battery and two types of betavoltaic cells. The electrical performance was assessed by measuring current-voltage properties (I–V) and determining the short-circuit current and the open-circuit voltage. With ^{63}Ni as an irradiation source, the open-circuit voltage and the short-circuit current were determined as 1 V and 64 nA, respectively. The prototype consisted of 10 betavoltaic cells that were prepared using radioactive ^{63}Ni. Electroplating of the radioactive ^{63}Ni on an ohmic contact (Ti–Ni) was carried out at a current density of 20 mA/cm^2. Two types of betavoltaic cells were studied: with an external ^{63}Ni source and a ^{63}Ni-covered source. Under irradiation of the ^{63}Ni source with an activity of 10 mCi, the open-circuit voltage V_{oc} of the fabricated cells reached 151 mV and 109 mV; the short-circuit current density J_{sc} was measured to be 72.9 nA/cm^2 and 64.6 nA/cm^2, respectively. The betavoltaic cells had the fill factor of 55% and 50%, respectively.

1. Introduction

The attention of experts of different scientific disciplines is drawn to a problem of using energy from radioactive decay. The betavoltaic effect is a promising technique to convert radioactive energy into electrical power. Efficiency and output power of betavoltaic cell has constantly increased and discovery of new practical applications [1–3].

The betavoltaic batteries generate insignificant electric power (nW-μW) and can be applied as power sources for microcircuits and sensors with low-power consumption as well as devices working in a pulse mode. The betavoltaic cells that use kinetic energy from the decay of a radioactive isotope to generate electricity have been studied for over 50 years [4,5]. The first converters based on Si had only 0.2% efficiency, and they degraded quickly under irradiation [5]. The most notable application of betavoltaic devices is found in pacemakers whose power source consists of a silicon converter and a ^{147}Pm isotope [6]. With the development of deep etching technology and the technology of high-quality creation of an i-layer on GaN, considerable success in the research of semiconductor elements of various constructions has been achieved [7–11].

The results of many studies and designs of betavoltaic cells have been usefully summarized in several publications [12,13]. Detailed theoretical and practical recommendations about the design of betavoltaic batteries and the description of their electrophysical characteristics have been also discussed [13–15]. Our comprehensive analysis of these papers [16–19] shows that an external source of beta particles is used, and this source is mounted on the semiconductor surface. However, betavoltaic cells with a radioactive deposition layer on the convertor's surface has not been adequately studied.

Only one research paper [20] indicates that the ^{63}Ni layer is deposited on the surface of metallic contacts of the p-n junction. The open-circuit voltage is found to be 0.8 mV, whereas the short-circuit current is 11 nA for an activity of 4 mCi ^{63}Ni. The received parameters are lower than those of an external source with the same activity [8]. The open circuit voltage is found to be 45 mV; whereas the short-circuit current is 18 nA.

The reason for low values of the open-circuit voltage is the pollution of the semiconductor surface and the inclusion of electrolyte in the ^{63}Ni layer, which appears during the deposition of the

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radioisotope. The surface charges formed by uncontrollable impurities increase the surface component of a reverse current that leads to a decrease of the open-circuit voltage and the output power. In this regard, our research aims to improve the method and the conditions of electroplating of ^{63}Ni on the surface of a semiconductor device.

In this work, we also develop a prototype of a nuclear battery that consists of 10 single betavoltaic cells. We also describe the initial steps of the design and the fabrication of a single cell and the nuclear battery and characterize its performance.

2. Materials and methods

2.1. Single cell

Silicon was selected as a semiconductor material for betavoltaic cells because it is both cheap and available. We used silicon wafers (P {100} 20 Ω·cm) with a thickness of about 470 μm to fabricate betavoltaic cells. The wafers were placed in a tube furnace at 950 °C for 60 min to create a heavily doped n^+ layer for the ohmic contact and n^+ ring by diffusing phosphorus. The heavily doped p^+ layer was manufactured with Boron implantation and was used to create the second ohmic contact. The implantation energy and the implantation dose were 60 keV and 500 μC/cm^2, respectively. The p-n junction was manufactured with Boron implantation and getter thermal annealing. The implantation was carried out through a film of SiO_2 with a thickness of 20 nm. The energy and the dose were 10 keV and 10 μC/cm^2, respectively. After implantation, getter thermal annealing at 900 °C was performed with slow cooling of 1 °C/min for 300 min.

There were two types of electrodes used in the betavoltaic cells: a frame electrode and a square electrode. The frame electrode was adopted to reduce the electrode reflection area. The ohmic contact was produced by Al deposition and annealing at 475 °C for 1 min. The square electrode was adapted to electroplating the ^{63}Ni layer. A metal layer of Ti/Ni (200/200 Å) was evaporated to form the p^+ ohmic contact. Titanium was used as an adhesive layer. After photolithography, the samples were annealed at 300 °C for 60 s in N_2 ambient. The size of single cells was chosen as 11x11 mm^2 to simplify the creation of an assemblage. The cross-sectional structures of the betavoltaic cells are shown in Fig. 1.

The radioisotope ^{63}Ni was deposited onto a metal surface of the betavoltaic cells by the reduction reaction from a water solution $(NH_4)_2C_6H_6O_7 + (NH_4)_2SO_4 + NH_3 \cdot H_2O + N_2H_4 \cdot HCl + {}^{63}NiCl_2$. The ^{63}Ni layer was deposited by AC electroplating at a current density of 20 mA/cm^2 and a temperature of 25 ^{0}C. The pH of the electrolyte was adjusted to 8.5–9.0.

The activity of the electroplated layer ^{63}Ni on the betavoltaic cells was established as the bath exhaustion ratio for ^{63}Ni radionuclide (98.7%), and was 10 mCi (370 MBq). The thickness-dependent self-shielding effect of the radioisotope layer and the penetration depth of the beta particles in Si were studied by Tariq R. Alam in Ref. [21]. Therefore, the thickness of the layer was established about 1.5 μm, and the area was 1x1 cm^2. The next step consisted of electroplating of a seal layer of stable Ni with a thickness of about 150 nm. The betavoltaic cells are shown in Fig. 2.

2.2. Prototype of the nuclear battery

While measuring the characteristics of every cell with the ^{63}Ni-covered source, we discovered a deviation in the distribution of the open-circuit voltage. The samples with close values of the open-

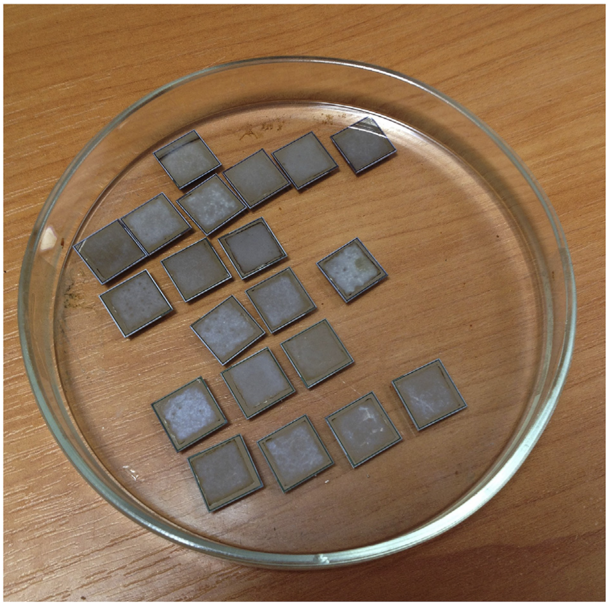


Fig. 2. Photograph of the fabricated betavoltaic cells.

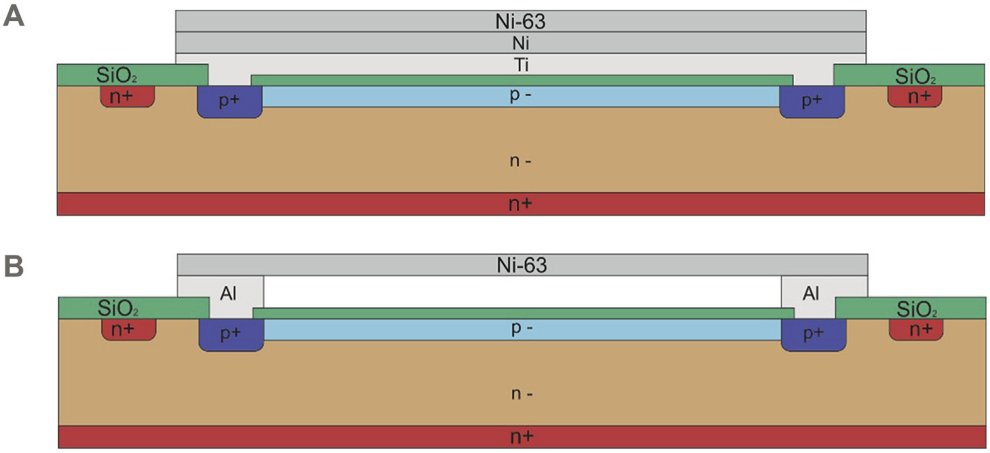


Fig. 1. Cross-sectional views of the betavoltaic cells (a) with a square electrode and (b) with a frame electrode.

circuit voltage ($\approx$100 mV) were selected for further assemblage. The distribution of the short-circuit current in all the samples did not differ much ($\approx$65 nA). We developed a prototype of a nuclear battery with a volume of 0.57 cm^3 that consisted of 10 single cells with the ^{63}Ni-covered source.

The serial assemblage of the elements was carried out in a special cassette using the conductive paste. The cassette provided a necessary orientation of the cells as well as necessary pressing to each other. The resulting assemblage was mounted in a case (Fig. 3). The sealing of the prototype of the nuclear battery was carried out by pouring radiation-resistant epoxide compound.

3. Results and discussion

I–V characteristics of the betavoltaic cells were investigated using a semiconductor analyzer Agilent 1500B. An external ^{63}Ni source with a specific activity of 10 mCi and dimensions of 10 × 10 × 0.1 mm was used for the samples with the frame electrode. The source was put in the center of the cell to close the junction surface. The measurements were made at room temperature in "dark" conditions (without any photon irradiation). The I–V characteristics of the sample with the external ^{63}Ni source and the ^{63}Ni-covered source are shown in Fig. 4.

Such parameters as J_{sc} and V_{oc} were determined to be 72.9 nA/cm^2, 64.6 nA/cm^2, 151 mV, and 109 mV, respectively, by re-plotting the absolute current density versus the bias voltage variation as shown in Fig. 4. Maximum bias voltages (V_m) 108 mV and 72 mV, and maximum current densities (J_m) 57 nA/cm^2 and 49 nA/cm^2 at peak values of the power density (P_{max}) 6.14 nW/cm^2 and 3.53 nW/cm^2 were determined. Fill factors (FF) were determined as 55.4% and 50.1% through the following relation:

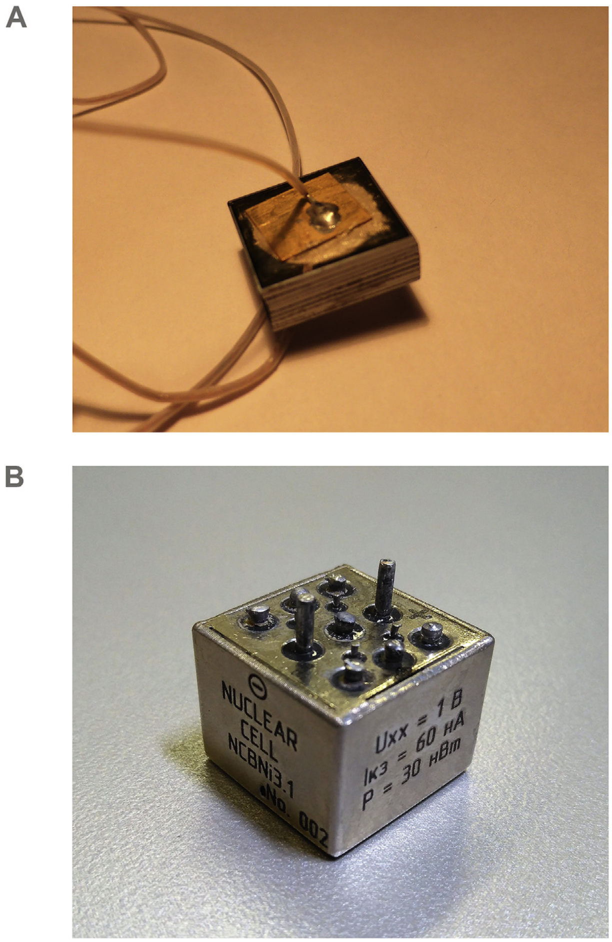


Fig. 3. (a) Photograph of the serial assemblage of the betavoltaic cells and (b) photograph of the prototype of the nuclear battery.

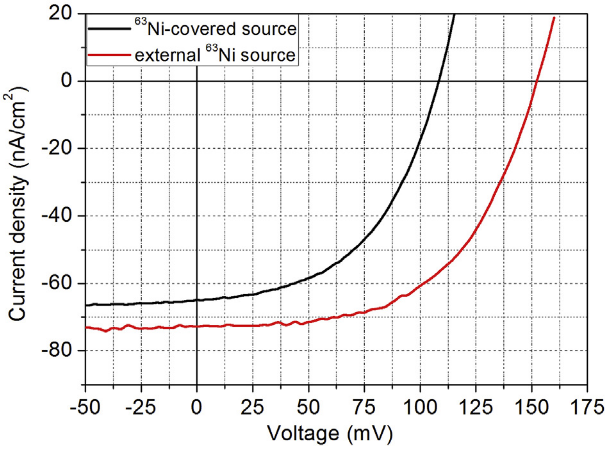


Fig. 4. The I–V characteristics for the betavoltaic cells with external ^{63}Ni source (red line), (b) with ^{63}Ni-covered source (black line). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

$$FF = \frac{V_m \cdot J_m}{V_{oc} \cdot J_{sc}} \cdot 100\% \tag{1}$$

The total conversion efficiency of the radiation energy into electrical power is a key parameter for the performance of betavoltaic cells. The conversion efficiency η can be defined as:

$$\eta_{total} = \frac{P_{\max}}{P_{source}} = \frac{FF \cdot V_{oc} \cdot J_{sc}}{3.7 \cdot 10^{10} \cdot A \cdot E_{avg} \cdot q}, \tag{2}$$

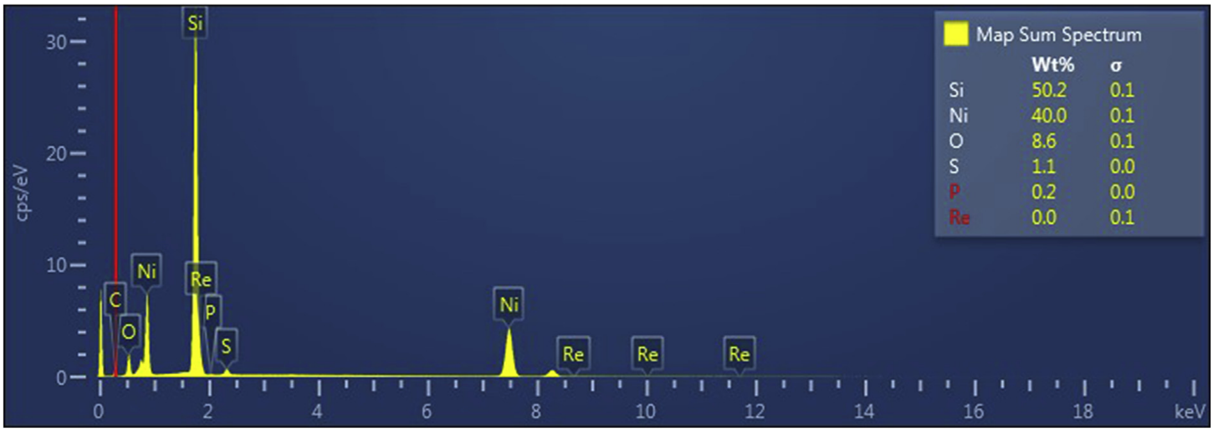
Where P_{source} is the radiation power of the source, A is the specific source activity, E_{avg} is the average beta energy of the isotope (17.1 keV for ^{63}Ni), and q is the electron charge (1.6 10^{-19} C). For the developed betavoltaic cells, the total conversion efficiencies are 0.6% and 0.35%, respectively (see Table 1). Considering the self-absorption, directional and backscattering losses, we established the converter's efficiency limit to be 3.5% and 2.0%, respectively. We assumed that the collection efficiency of EHPs was 100%, the self-absorption loss for 1.5 μm Ni layer was 60% [21], the directional loss was 50%, and the backscattering coefficient was 16%.

The short current density of the betavoltaic cell with the external ^{63}Ni source is 1.13 times higher than that of the betavoltaic cell with the ^{63}Ni-covered source. The presence of the Ti–Ni layers with a high charge number (Z) led to an increase of the backscattered β-particles and a loss of the kinetic energy that was wasted on the penetration through these layers. The open circuit of the betavoltaic cell with the external ^{63}Ni source is 1.38 times higher than that of the betavoltaic cell with the ^{63}Ni-covered source. Our general analysis of the elemental composition of the betavoltaic cell surface with the ^{63}Ni-covered source indicates the presence of contamination (see Fig. 5). Ions of Sulphur lead to an induction of charges in the semiconductor converter, which lead to the formation of surface leakage channels. Therefore, the leakage current on the surface is increased, which leads to a decrease in the open-circuit voltage and the output power.

Another key parameter is a specific maximum power, which can be defined as:

Table 1
Summary of the betavoltaic cells characteristics under^{63}Ni irradiation.

Type of cell	V_{oc}, mV	J_{sc}, nA/cm^2	P_{max}, nW/cm^2	R_{load}, MOhm	FF, %	η_{total}, %	$P_{specific}$, nW/g
external^{63}Ni source	151	72.9	6.14	1.89	55.4	0.6	27.7
^{63}Ni-covered source	109	64.6	3.53	1.47	50.1	0.35	26.4



	Wt%	σ
Si	50.2	0.1
Ni	40.0	0.1
O	8.6	0.1
S	1.1	0.0
P	0.2	0.0
Re	0.0	0.1

Fig. 5. General analysis of the elemental composition of betavoltaic cell surface metallized with ^{63}Ni.

$$P_{specific} = \frac{P_{\max}}{m} \tag{3}$$

Where m is the mass of the elements. It was defined by the geometrical sizes and the density of the coating Ni, which was accepted equal specific density of Ni (8.9 g/cm^3). The specific maximum power for both elements was at the same level. When using a silicon wafer with a thickness of less than 400 μm, the ratio of specific maximum power begins to increase and reaches a value of 1.45 at a thickness of 200 μm. In the calculation, the thickness of the external ^{63}Ni source was taken 100 μm (see Fig. 6).

The maximum output power was achieved within the optimal resistance range of 1.5–2 MOhm. Consequently, with real devices, it is necessary to ensure coordination of the output of the nuclear battery with the input load resistance. The I–V and P–V characteristics of the prototype of the nuclear battery are shown in Fig. 7.

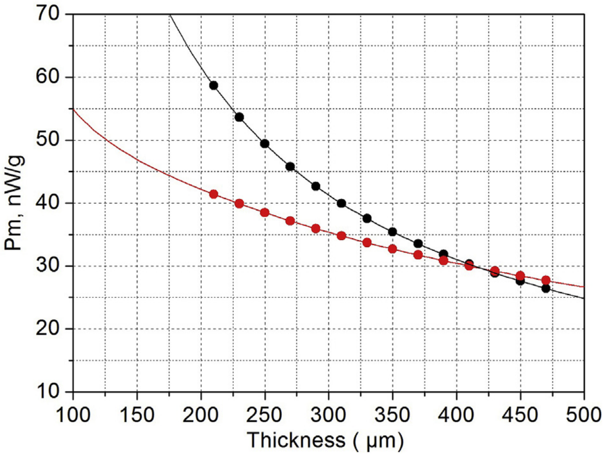


Fig. 6. The dependence of the specific maximum power on the thickness of the silicon substrate (red line with external ^{63}Ni source), (black line with ^{63}Ni-covered source). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The open-circuit voltage and the short-circuit current were 1.0 V and 64 nA, respectively. The maximum output power of the fabricated nuclear battery was about 32 nW, which corresponds to the power density of 24 nW/g. Notably, the total power produced in the ^{63}Ni source itself was about 10.1 μW, which gives the total efficiency of only 0.32%.

4. Conclusions

Our research described two types of the betavoltaic cells based on p-n junction. These betavoltaic cells are designed using Si and ^{63}Ni as a semiconductor material and a radiation source, respectively. We estimated the output characteristics under irradiation of an external solid plate ^{63}Ni source and a^{63}Ni-covered source. Under the irradiation with an activity of 10 mCi, the betavoltaic cells exhibit the short-circuit current density of 72.9 nA/cm^2 and 64.6 nA/cm^2; the open-circuit voltage of 151 mV and 109 mV; the total efficiency of 0.6% and 0.35%.

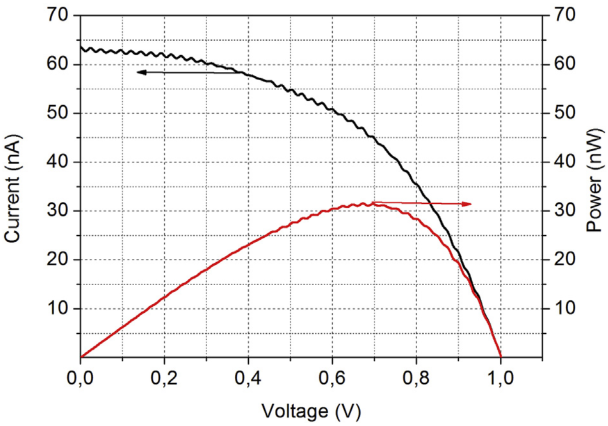


Fig. 7. Current-voltage (black line) and power-voltage (red line) curves for the prototype of the nuclear battery. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Due to high ^{63}Ni source self-absorption, an increase of the source enrichment and the reduction of the converter thickness are a more proper strategy to create a more powerful battery (W/g) than just an increase of the amount of a radioactive material. In addition, a decrease of the semiconductor converter thickness by less 400 μm using mechanical polishing or plasma etching can improve the specific power of the betavoltaic cell with a^{63}Ni-covered source.

In our research, we developed a prototype of a nuclear battery that consists of 10 single betavoltaic cells with the ^{63}Ni-covered source. Under irradiation of the ^{63}Ni source with the total activity of 100 mCi, the output power density power density of 24 nW/g was obtained. Due to intrinsic ^{63}Ni source self-absorption, the total efficiency of the nuclear battery was only 0.32%.

Changes to the prototype of a nuclear battery will depend on the application. For example, for independent power supply and data collection from sensors located in hard-to-reach places, the preliminary requirements are supply voltage 5 V and load current 25 mA. The application will be justified when the connecting with the power supply system is complicated and the period of updating the information is quite large (>200 s).

Another example is a backup power supply, which is necessary to save system variables and intermediate calculations at the time of failure. Based on the power consumption of the IC (BH62UV4000, BS616LV1010, CY62138EV30 and etc.) it is possible to formulate preliminary requirements for nuclear batteries to protect RAM blocks (1–16 KB). supply voltage is 3.3 V, and load current is 2.5 mA.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2019.06.003.

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