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Original Article

Study of a Betavoltaic Battery Using Electroplated Nickel-63 on Nickel Foil as a Power Source

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ABSTRACT

A betavoltaic battery was prepared using radioactive ^{63}Ni attached to a three-dimensional single trenched P–N absorber. The optimum thickness of a ^{63}Ni layer was determined to be approximately 2 μm, considering the minimum self-shielding effect of beta particles. Electroplating of radioactive ^{63}Ni on a nickel (Ni) foil was carried out at a current density of 20 mA/cm^2. The difference of the short-circuit currents (I_{sc}) between the pre- and post-deposition of ^{63}Ni (16.65 MBq) on the P–N junction was 5.03 nA, as obtained from the I–V characteristics. An improved design with a sandwich structure was provided for enhancing performance.

1. Introduction

Electricity generated by the decay of radioisotopes can provide a high energy density for several decades, although the device is located in a place inaccessible to harvestable energy [1,2]. The mechanism of a nuclear battery is the same as that of a P–N junction diode for solar cell application. A photovoltaic battery is operated by converting photons into electrical energy in the junction. In a betavoltaic battery, beta particles are collected and converted into electrical energy by a similar principle to that used in a photovoltaic battery. A very low current, of the order of nano- or microamperes, is generated in the devices [3]. If a radioisotope with a long half-life (over 100 years) is used, the lifetime of the power source is extended to as long as the half-life time of the radioisotope. Some special applications require long-lived compact power sources [4,5].

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These include space equipment, sensors in remote locations (space, underground, etc.), and implantable medical devices. The power source of the radioisotope shows that a larger amount of energy released per reaction than that release during the conversion of chemical energy into electricity [6,7]. A radioisotope battery is a novel solution for solving the power needs of these applications [8–10]. For the ^{63}Ni beta source that we used, the half-life is 100.2 years [2]. Hence, the power sources can extend a system's operating life by several decades or even by a century, during which time the system can gain behavior without worrying about the power turning off. The beta spectrum of ^{63}Ni is below the radiation damage threshold (approximately 200 keV for Si) of semiconductors such as Si and SiC [4]. In addition, ^{63}Ni is easier to handle than other beta particles such as ^{3}H, ^{90}Sr, and ^{147}Pm because of its low energy spectrum and solid-metal form. For this reason, it is suitable for the power source of betavoltaic batteries to be within the nano- to microwatt range [8–10]. There are several methods for forming nickel (Ni) deposits onto a substrate, such as electroplating, electroless plating, and chemical vapor deposition [11]. Among them, the electroplating process is most commonly used for Ni deposition when using ^{63}Ni as a power source for batteries. Radioactive thin-film-based power sources also have energy densities that are several orders of magnitude higher than chemical-reaction-based energy sources. This enables submillimeter-scale power sources, which is significant given the crucial role of the metrics of power and energy density in determining the usefulness of pervasive computing systems' applications with limited size.

The aim of this work is to optimize the method and conditions of electroplating to maximize the electrical efficiency of a betavoltaic battery using radioactive ^{63}Ni. In this study, a betavoltaic battery was fabricated using ^{63}Ni attached on a P–N junction semiconductor, and the I–V characteristics were measured using a probe station. The thickness-dependent self-shielding effect of the radioisotope layer was investigated [12]. In addition, a betavoltaic battery was newly designed and is suggested as a means of increasing the power output of the battery.

2. Materials and methods

To fabricate the P–N absorber, a new type of three-dimensional single trenched P–N absorber for easy trenching and doping was developed by the Electronic Telecommunications Research Institute. The P–N spacing was 50 μm. To measure the power output of a single cell, a ^{63}Ni beta source was deposited on the substrate, and the unsealed source was attached on a P–N junction using vacuum.

The process of electroplating using radioactive ^{63}Ni in a hot cell (Bank-2, HANARO research reactor in Korea Atomic Energy Research Institute) was carried out using a two-step process: preparation of an ionic solution including ^{63}Ni and electroplating on a substrate. Ni coatings were deposited by DC electroplating at a current density of 20 mA/cm^2. The deposition condition for ^{63}Ni is explained in the works of Uhm et al [13]. The basic composition of the chloride bath was 0.166M Ni and 0.4M boric acid (H_3BO_3). Ni metal powders were dissolved in a mixture of HCl and distilled water. Boric acid is used in Ni-plating solutions for buffering purposes. The pH of the bath was adjusted to 4.0 ± 0.2 by the addition of KOH. Dimension of the layer deposited as an anode on a P–N junction is 4 × 4 mm^2.

The thickness-dependent self-shielding effect of the radioisotope layer and the penetration depth of the beta particles in Si were studied using a Geant4 Monte Carlo code [14]. The I–V characteristics of a ^{63}Ni-attached semiconductor were investigated using a probe station of the Precision Source/Measure Unit, B2911A. Radioactivities were estimated by comparing the measurements of a liquid scintillation counter (Tri-Carb 2910 TR; PerkinEmer Co. Waltham, Massachusetts, USA) for the bath before and after electroplating. The standard solution for ^{63}Ni was 100.6 μCi/5 mL in 0.1N HCl.

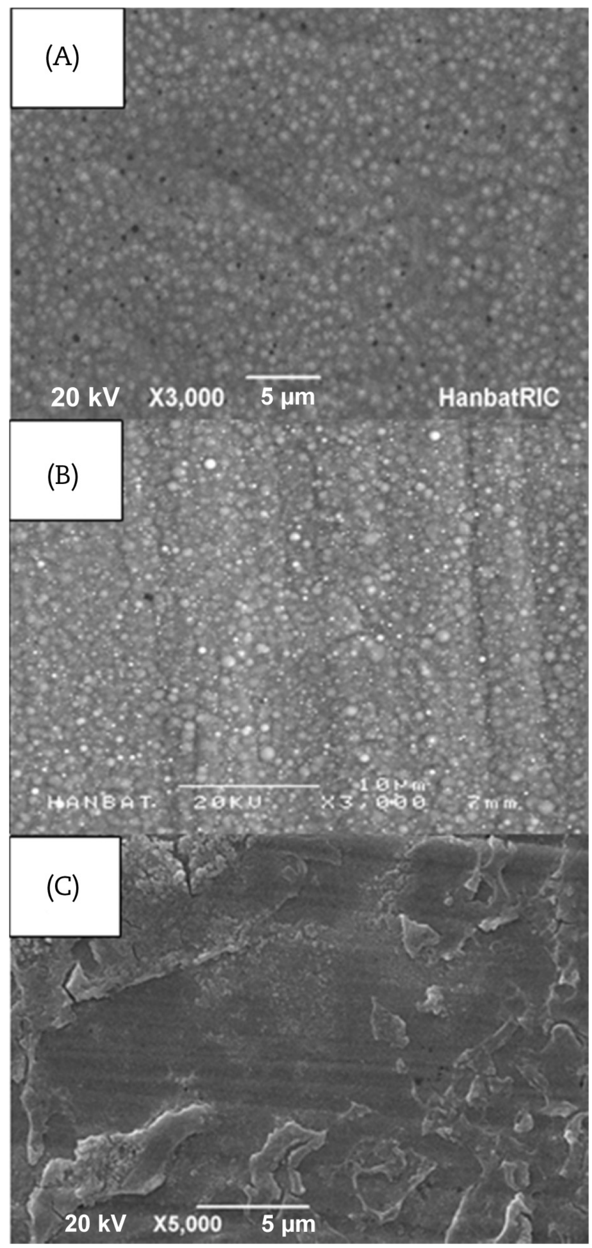

Fig. 1 – SEM images for electroplated Ni on Ni foil. Electrodeposition at a current density of (A) 10 mA/cm^2, (B) 20 mA/cm^2, and (C) 30 mA/cm^2.

3. Results

To evaluate the P–N junction prepared by the Electronic Telecommunications Research Institute, the electron beam-induced current technique has been employed to experimentally simulate beta emission from ^{63}Ni and to estimate the total device current [15]. The open-circuit voltage was found to be 0.29 V. The short-circuit current was 3.3 A. The power output was found to be 66.5 W/cm^2. From the e-beam illumination test, we confirmed the good operation of the P–N absorber.

A Ni-plating solution was prepared by dissolving metal particles, and the deposition conditions had been optimized in a previous study by studying the influence of the current density. In addition, the proposed prototype condition was applied to radioactive ^{63}Ni electroplating. The electroplating was carried out in a two-step process: preparation of an ionic solution including ^{63}Ni and coating on the substrate. The prototype of electroplating ^{63}Ni was carried out in a glove box in a hot cell (Bank-2, HANARO Reactor in Korea Atomic Energy Research Institute). The radioactivity of the ^{63}Ni electroplated on the foil with planar dimensions of 1×1 cm^2 was estimated to be about 92.5 MBq (2.5 mCi). The specific radioactivity was 15.91 GBq/g (0.43 Ci/g). Radioactivities of the plating bath before and after electrodeposition were measured using a liquid scintillation counter. In addition, we prepared electroplated ^{63}Ni on a foil with the same planar area (0.4×0.4 cm^2) as that of the P–N junction. Its radioactivity was 16.65 MBq (0.45 mCi). Morphologies of the coating layer were measured using natural Ni prepared in the same bath and same plating conditions as those of the radioactive sample. About 1 g of the starting material (stable isotope ^{62}Ni) was compacted and irradiated in a reactor pool. The irradiated powder (1 g) was dissolved in a chloride bath of about 85 mL. We established the conditions of dissolving and plating ^{63}Ni at a previous study [13].

Fig. 1 shows scanning electron microscopic (SEM) images at different magnifications for Ni electrodeposited on a Ni foil at current densities of 15 mA/cm^2, 20 mA/cm^2, and 30 mA/cm^2. The particles on the Ni sheet had a spherical shape, as shown in Figs. 1A–1C. As shown in Fig. 1C, electroplating of Ni was not possible at and above a current density of 30 mA/cm^2, because a large amount of salts in the solution were also

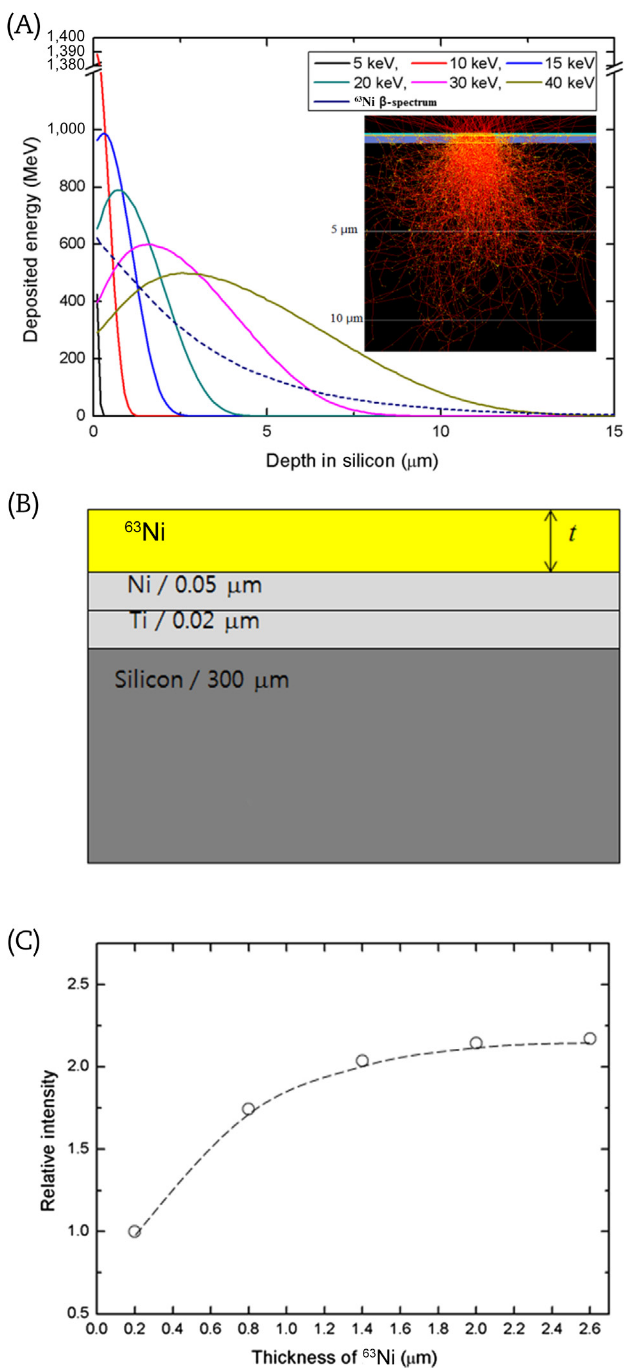

Fig. 2 – Energy deposition as a function of depth in silicon. (A) Depth energy deposition of beta particles in silicon and the trajectory of beta rays in the silicon. (B) A defined geometry for simulation. (C) the results of self-shielding as a function of thickness for the deposited layer of ^{63}Ni.

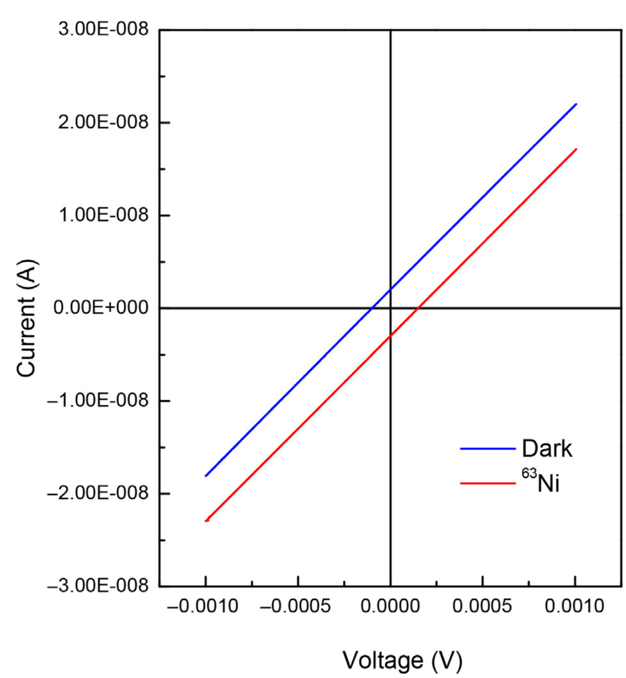

Fig. 3 – Planar RI battery performance characteristic of magnified *I*–*V* curves. Behaviors of diodes with and without a ^{63}Ni layer on the junction. RI, radioisotope.

deposited on the Ni substrate. The electroplating was carried out at a current density of 20 mA/cm^2. The proposed prototype for the synthesis could be applied to radioactive ^{63}Ni electroplating. In this study, the difference in the radioactivity of the plating bath before and after deposition was defined to be the radioactivity of a coating layer.

The principles used for the operation of a betavoltaic battery are very similar to those of a photovoltaic battery. Electron–hole pairs are created, collected in a junction, and then converted into a current [4, 5]. Each beta particle produces thousands of hole pairs as it inelastically scatters through the silicon substrate. The penetration depth of the particles in the silicon device was reported in a Katz–Penfold range equation [10]. This equation considers only the density of the materials and energy of the particles. In this study, we modeled energy deposition as a function of depth in the silicon substrate using the Monte Carlo code. The thickness-dependent self-shielding effect of the radioisotope layer was also investigated. Fig. 2A shows depth energy deposition of beta particles in silicon and the interaction of beta particles with silicon, Fig. 2B shows a defined geometry for observing self-shielding effect, and Fig. 2C shows the results of self-shielding as a function of thickness for the deposited layer of ^{63}Ni. In Fig. 2A, some beta particles reach up to a depth of 15 μm in a Si wafer. An illustration of the moving routes for the beta particles shows a deep penetration of the silicon substrate. The simulation and illustration were carried out using the Geant 4 code and OpenGL software, respectively. Both Ni and Ti layers between ^{63}Ni layer and Si wafer are seed layers with a defined geometry, as shown in Fig. 2B. These metal layers are required for direct coating of ^{63}Ni on a P–N junction. However, the seed layer strongly attenuates low-energy beta particles [15]. Absorption energies in the Si wafer were calculated, when the thickness of ^{63}Ni coating layers were 0.2 μm, 0.8 μm, 1.4 μm, 2.0 μm, and 2.6 μm. The essentially zero response for the total thickness of 2 μm of ^{63}Ni indicates the self-shielding effect, as shown in Fig. 2C. The thick thickness of the ^{63}Ni coating layers negatively affect performance. In contrast, the penetrating depth of the beta rays in the Si semiconductor is relatively deep.

We attached a prepared beta source on the P–N junction using vacuum. The I–V curves of both predeposition (dark) and deposited ^{63}Ni show almost the same values, as shown in Fig. 3. The difference between the predeposition and ^{63}Ni deposition can be obtained through magnification of the I–V curve [12]. The difference of the short-circuit current between the pre- and postdeposition of ^{63}Ni on the foil was found to be 5.03 nA. The short-circuit current was 0.16 mA. The power output was found to be 0.8 pW. The power density was 5 pW/cm^2. The power output of a single cell operated at a radioactivity of 0.45 mCi reached the same value as that of a single cell with a seed

(A)

(B)

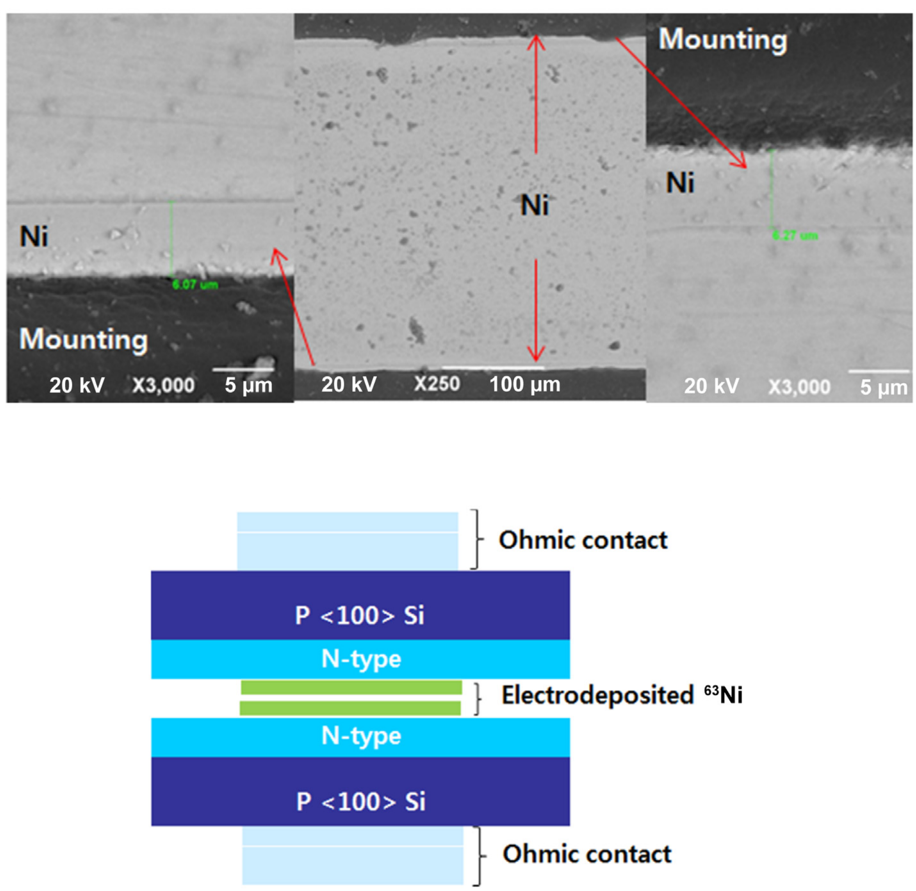

Fig. 4 – Deposition of Ni on the both sides of a Ni foil through electroplating and a new design of betavoltaic. (A) SEM image of the cross section of an electroplated Ni layer on double both sides of a Ni foil. (B) A schematic design for the cross-sectional of a pair cell with ^{63}Ni layers of an RI battery with a sandwich structure. RI, radioisotope; SEM, scanning electron microscopy.

layer (thickness of 500 Å) on the P–N junction operated at a radioactivity of 92.5 MBq [15]. The seed layer shows negative performance such as beta ray shielding in a cell [15]. In this study, we wanted to get rid of the seed layer on the P–N Junction. The fabrication process of a single cell could be simplified. The maximum power output of the betavoltaic battery prepared by Ulmen et al [12] reached 2.5 pW at a voltage of 0.4 mV operated at 148 MBq (4 mCi) of ^{63}Ni. Recently, the maximum performance was achieved at 5–500 nW of stack structure (package) prepared by Widetronix Co. (Ithaca, New York, USA) [16]. We obtained a relatively enhanced power output at a single cell, although the radioactivity was lower in value than those of other groups. The attaching method of a beta source and a P–N junction should be modified to attain high efficiency in future works. In this study, we manually attached ^{63}Ni foil on the P–N junction using vacuum. Some parts of the P–N junction were uncovered with a foil and exposed to air. If the ^{63}Ni foil is strongly fixed on a P–N junction during the fabrication process, the efficiency will increase.

4. Discussion

Beta particles of ^{63}Ni were deposited by electroplating on a Ni foil and attached on a trenched P–N absorber. Thickness of the ^{63}Ni layer was determined to be about 2 μm considering the self-shielding effect of beta rays. The optimum fabricating condition for the ^{63}Ni-coated foil was decided in this study. The difference in the short-circuit current between P–N junctions with and without radioactive ^{63}Ni (16.5 MBq) was found to be 5.03 nA. The power output was found to be 0.8 pW and the power density 5 pW/cm^2. A relatively enhanced power output was achieved from a very weak power source due to the P–N junction without a seed layer.

Although power output was enhanced, a very low current ranging from nano- to microamperes was generated in the devices. Adjusting the depletion region depth in a P–N junction is a key to getting higher performance. Once a study of the effect of this parameter on the performance of the planar P–N device is conducted, it will be possible to develop a battery with enhanced performance. However, a long time is needed to achieve this objective. In this study, a newly designed device with a sandwich structure is suggested to get a higher power output and more efficiency in the same single cell without any changes in the P–N junction. To fabricate a betavoltaic battery with a sandwich structure, ^{63}Ni should be coated on both sides of the substrate. Fig. 4A represents a scanning electron microscopic image of the Ni electroplated on both sides of the Ni foil. A schematic of the device is shown in Fig. 4B. The optimum integration of a pair junction will be designed in a further study. Considering that the location of the deposited energy is few microns deep within the substrate, the optimum thickness of the ^{63}Ni layer on both sides of the substrate between the upper and lower P–N junctions is of the order of several microns. This design allows a greater value of the output power per device. The addition of the opposite layer, such as the upper junction, plays the role of radiation shielding using the P–N junction itself. The electric power per unit cell with a pair junction is increased to twice the value of a unit cell with a single junction.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgments

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